

INSTITUTE OF MACROMOLECULAR
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INTERNATIONAL UNION OF PURE
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CZECH CHEMICAL SOCIETY

EUROPEAN POLYMER FEDERATION

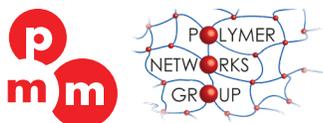


17 - 21 JUNE 2018 PRAGUE



POLYMER NETWORKS AND GELS 2018

82nd PRAGUE MEETING ON MACROMOLECULES
24th POLYMER NETWORKS GROUP MEETING



BOOK OF ABSTRACTS AND PROGRAMME

INSTITUTE OF MACROMOLECULAR CHEMISTRY
CZECH ACADEMY OF SCIENCES

INTERNATIONAL UNION OF PURE AND APPLIED CHEMISTRY

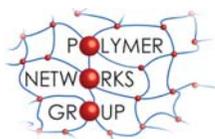
CZECH CHEMICAL SOCIETY

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POLYMER NETWORKS AND GELS 2018

17 – 21 June 2018, Prague

82nd PRAGUE MEETING ON MACROMOLECULES
24th POLYMER NETWORKS GROUP MEETING



Book of Abstracts and Programme



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**82nd PRAGUE MEETING ON MACROMOLECULES
24th POLYMER NETWORKS GROUP MEETING**

under the auspices of the
International Union of Pure and Applied Chemistry,
Czech Chemical Society and
European Polymer Federation

**Organized by the Institute of Macromolecular Chemistry,
Czech Academy of Sciences**

Miroslava Dušková-Smrčková
Conference Chair

Karel Dušek
Honorary Chair

Jiří Kotek
Director of the Institute

Daniela Illnerová, Marie Rodová, Marcela Havelková
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Ferenc Horkay (*USA*)

Eva Malmström Jonsson (*Sweden*)

Oguz Okay (*Turkey*)

Costas Patrickios (*Cyprus*)

Olga Philippova (*Russia*)

Marco Sangermano (*Italy*)

Mitsuhiro Shibayama (*Japan*)

Julius Vancso (*The Netherlands*)

Brigitte Voit (*Germany*)

Chi Wu (*Hong Kong*)

Miklos Zrínyi (*Hungary*)

Polymer Networks Group Meetings

- 1975 Strasbourg, F - Freiburg, FRG
- 1977 London, UK
- 1978 Breisach, near Freiburg, FRG
- 1979 Jablonna, PL
- 1980 Karlovy Vary, CS
- 1982 Strasbourg, F
- 1984 Manchester, UK
- 1986 Elsinore, DK
- 1988 Freiburg, FRG
- 1990 Jerusalem, IL
- 1992 San Diego, USA
- 1994 Prague, CZ
- 1996 Doorn, NL
- 1998 Trondheim, NO
- 2000 Krakow, PL
- 2002 Autrans, F
- 2004 Bethesda, USA
- 2006 Sheffield, UK
- 2008 Larnaca, CY
- 2010 Goslar, DE
- 2012 Jackson Hole, WY, USA
- 2014 Tokyo, JP
- 2016 Stockholm, SE
- 2018 Prague, CZ

Further information on Polymer Networks Group and its activities:
<http://www.polymernetworksgroup.org/>

Polymer Networks Group Committee

Mitsuhiro Shibayama (*The University of Tokyo, Japan*) - Chairman

Costas S. Patrickios (*University of Cyprus, Nicosia, Cyprus*) -
Vice-Chairman

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Miklós Zrínyi (*Semmelweis University, Budapest, Hungary*)

Christopher N. Bowman (*University of Colorado at Boulder, CO,
USA*)

Uday Maitra (*Indian Institute of Science, Bangalore, India*)

We thank the sponsors of the Conference

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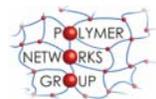


MERCK



Prague, June 2018

POLYMER NETWORKS AND GELS 2018



Dear Colleagues,

Welcome to POLYMER NETWORKS AND GELS 2018, the 82nd *Prague Meeting on Macromolecules* organized by the Institute of Macromolecular Chemistry, Czech Academy of Sciences (IMC Prague). It happens to be at the same time the 24th *Polymer Networks Group Meeting*.

We all are almost permanently connected through webs of instant library sources, e-mails, professional and social virtual networks. Yet, we still desire to travel to certain places, to attend meetings leaving our labs and offices behind. Why do we sit and listen to number of talks filling couple of intensive days? The answer is simple. Because we are humans and we can not only communicate through media, but we also urgently need to interact person to person. It is this contact that establishes and develops close relationships and makes it easy to explain our motivations for our hard work. It is when we can really learn from each other. Let me call these interactions *crosslinking* where the conference event is a major multifunctional micronetwork that will, hopefully, add and strengthen the network of international community gathered around the great topic. Perhaps, it will help community's members to know each other a little better, and, last but not least, will induce further crosslinking.

Medieval Prague is known to have been a center of alchemists thriving to transform various kinds of matter into precious gold and to discover the *elixir of wisdom*. While the futile transformation efforts were without success in those non-atomic ages to dissatisfaction of emperors (but several useful "materials" were discovered instead), the quest for the *elixir* was not entirely fruitless. It has transformed into what we call today a *networking* spirit spanning multitude of cultures and endeavors brought to Prague

from medieval Europe and melting into unique atmosphere of exploration and shared ideas, skills and experience. If a new idea sparks in your mind during these four days and you cannot wait to be back in your lab to pursue it, then this meeting is a success and the power of the *elixir* prevails!

The IMC Prague and its staff are dedicated to do their best to provide grounds for this double networking event where social networking penetrates polymer networks. Our most ambition is to make friendly environment in which the scientific exchange and open talks between participants is the most natural thing. This June, the IMC is doing so for the eighty second time and this time it happens to be on NETWORKS AND GELS again, as the first time in 1967!

On behalf of the POLYMER NETWORKS AND GELS 2018 organizers,

Miroslava Dušková-Smrčková and Karel Dušek
Conference Chairs

Much has happened since Professor Otto Wichterle launched the first session of Polymer Gels conference in 1967. Intriguing questions on these exciting materials still remain today...

CZECHOSLOVAK
ACADEMY
OF SCIENCES

CZECHOSLOVAK
CHEMICAL
SOCIETY

INSTITUTE OF MACROMOLECULAR CHEMISTRY

O. WICHTERLE

Head of the Institute
Chairman, Group of Macromolecular
Science, Czechoslovak Chemical Society

B. SEDLÁČEK

Chairman, Scientific Committee
of the Institute
Head of the Physics Division

MICROSIMPOSIUM ON MACROMOLECULES:

P O L Y M E R G E L S
AND CONCENTRATED
S O L U T I O N S

P R O G R A M M E
A B S T R A C T S

P R A G U E , S E P T . 1 1 - 1 4 , 1 9 6 7

MAIN LECTURES

- I G. REHAGE (Clausthal-Zellerfeld):
Diffusion, Osmosis and Phase Separation in
Macromolecular Systems (Monday, Sept.11, 10.00)
Chairman: R.Koningsveld; Deputy Chairman: B.Sedláček
- II W. HEITZ, W. KERN (Mainz):
Principles of Gel Chromatography and Possibilities
of its Development (Monday, Sept.11, 15.00)
Chairman: O.Wichterle; Deputy Chairman: J.Hnídek
- III S. YA. FRENKEL (Leningrad):
Fluctuating Networks in Polymer Solutions (Tuesday,
Sept.12, 15.00)
Chairman: R.H.Marchessault; Deputy Chairman:
M.Bohdanecký
- IV W. PRINS (Delft):
Intermolecular Effects in Non-Crystalline Polymer
Systems (Wednesday, Sept.13, 15.00)
Chairman: M.Gordon; Deputy Chairman: K.Dušek
- V D. PATTERSON (Montreal):
Thermodynamics of Non-Dilute Polymer Solutions
(Thursday, Sept.14, 15.00)
Chairman: A.Silberberg; Deputy Chairman: J.Pouchlý

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GENERAL INFORMATION

Venue

All sessions are held at the Institute of Macromolecular Chemistry, Czech Academy of Sciences, Heyrovského nám. 2, Prague 6, Czech Republic.

Language

The official language of the conference is English.

Registration

On-site registration takes place on Sunday, 17 June 2018, from 16:00 to 19:00 at the Institute of Macromolecular Chemistry and will continue the next day from 8:00 to 16:00.

Please be aware that only persons wearing name badges received upon registration are entitled to enter the lecture rooms at the Institute.

PRESENTATIONS

The conference presentations consist of main, keynote, and invited lectures and oral communications. There are also two poster sessions held on Monday, 18 June and on Tuesday, 19 June 2018.

Time allocated for lectures including questions and discussion:

Main lectures: 35 minutes

Keynote lectures: 20 minutes

Invited lectures: 20 minutes

Oral communications: 15 minutes

Presentation files upload

We recommend that you upload your presentation at least one session ahead of your lecture.

IT administrator Mr Jiří Kaprálek will help you to upload and check your presentation. Please use your last name as the presentation file name.

Recordings

Audio/video recording of the lectures or copying of lecture files is not allowed without the prior consent of the speaker. The organizers reserve the right to capture photographs for documentation during the meeting.

Poster presentations

Materials for fixing posters will be provided on-site. Posters can be mounted at the beginning of each poster session and should be removed thereafter. The area reserved for mounting a poster is 200 cm high and 100 cm wide.

Poster sessions

- **Poster session I: Monday, 18 June 2018, 17:20 – 19:00**
Posters P-01 – P-36
- **Poster session II: Tuesday, 19 June 2018, 17:10 – 19:00**
Posters P-37 – P-75

PNG 2018 YOUNG INVESTIGATOR AWARD

(sponsored by Sumitomo Bakelite Co., Ltd.)

The award is aimed to recognize outstanding achievement of young investigators in the field of polymer networks and gels, and to encourage young scientists and engineers to enter this field.

SPECIAL POSTER CONTEST

The scientific committee will evaluate student posters from both poster sessions. Winners will be announced at the Conference dinner and awarded by the chair of the conference.

REFRESHMENTS AND LUNCHES

There will be coffee breaks each day. Lunch buffets will be open from Monday through Thursday in the dining hall of the Institute during the lunch breaks. The cost of refreshments and lunches is included in the conference fee as well as the welcome buffet and conference dinner.

SOCIAL EVENTS

The costs of the following social events are included in the conference fee. Accompanying persons can pay for the Wednesday afternoon programme and conference dinner at the registration desk.

Sunday, 17 June 2018

16:00 – 19:00 Registration with Welcome Buffet

Participants of the conference will register and receive the conference materials. The Welcome Buffet will be an excellent occasion for meeting with participants of the 82nd PMM conference.

Wednesday, 20 June 2018

Guided Tours in Prague

Guided tours will depart from the conference venue at 14:00 and will end in the city by 17:00.

1. Off the Beaten Track Tour - Castle Town (Hradčany)

Chance to see beautiful palaces and surprisingly picturesque corners in one tour

Almost every visitor to Prague knows the Prague Castle, but not everybody knows about Hradčany (until 1784 it was an independent "Castle Town"), a very interesting area surrounding the Castle. The tour will take you through the complex of the Strahov Monastery founded in the 12th century. Then you will see a famous Prague Loreto (which hosts an exact copy of the Italian "Santa Casa" chapel) and pass by imposing noble palaces next to the small servants' cottages. Finally, you can enjoy the relaxing part of the tour - the Castle moat's peaceful secluded area. Through the Royal Garden we will reach the nearest tram stop and continue to Náměstí Míru.

2. Tour of the Energy Centre of the City - Charles Bridge and Old Town

The oldest area of Prague – for the real history lovers

The Charles Bridge is a unique historical sight renowned worldwide. It is undoubtedly one of Prague's main cultural symbols, but it's also considered to be the energy centre of the city. Let's cross it and learn more about its history and mystery. Afterwards you will visit the Old Town (the oldest of four historical towns of Prague). The centre and main attraction of it is the Old Town Square with the Astronomical clock, but it also covers the Jewish Town (former Jewish ghetto). After visiting all these sights we will take a metro and continue to Náměstí Míru.

3. Gardens and Parks Tour - Lesser Town

The most scenic area of Prague with its specific genius loci

This tour will take you through the gardens and parks of the Lesser Town, the second oldest historical town of Prague (founded in the 13th century). We will start at the spacious baroque Wallenstein garden which belongs to Wallenstein palace, nowadays seat of the Czech Senate. The tour will continue through the Vojan Gardens, former convent utilitarian garden. As the highlight of the tour we will delight in the splendid Vrtba Garden, which is situated on the slope of Petřín Hill. After enjoying the breathtaking views, we will also visit the Church of Our Lady Victorious with the miraculous "Infant Jesus of Prague". From there we will take a tram to Náměstí Míru.

4. Prague in Legends

Fantasy or reality? Believe or not, it is up to you...

Prague is a very old city that enchants all of its visitors, mostly due to its beautiful architecture and preserved historical centre. However, would you like to visit its hidden corners, tiny narrow streets, and secret passages? Doing so reveals more than the "ordinary" beauty of the city – it unfolds the city's magical power. Join this tour and learn about Prague's mysteries, fairy tales, and legends. Visit places where scary ghosts may appear and miraculous locations where people's wishes come true... If we don't get lost or bewitched, we will continue by metro to Náměstí Míru.

5. Must See Places Tour - Prague Castle Grounds

Well known and hidden places of the biggest castle in the world

The Prague Castle promises a breathtaking journey through one thousand years of Czech history. Four powerful European dynasties ruled Bohemia from here. It is a place where a variety of architectural styles mingle together. Let's explore three castle courtyards and then enter the magnificent St Vítus Cathedral, where you can admire the works of art by old medieval masters as well as those in modern Art Nouveau style. Besides the royal area, we will see also the Golden Lane where the common people used to live. Through the historical vineyard we will walk to the nearest metro station and continue to Náměstí Míru.

6. 20th Century History Tour – Old and New Town

Most of you remember living in the 20th century, but what was it like in Prague?

Prague is a very old city that has witnessed a variety of events during its history. This tour will focus specifically on the events which happened in the 20th century. In 1918 Prague became the capital of the new Czechoslovak republic. The happy times didn't last long as 21 years later, in 1939, it was occupied by the Nazi Germans. In 1945 it celebrated its liberation by the Soviet Army, but the liberty lasted only until 1948, when Czechoslovakia became a communist country. In 1968 the same Soviet Army occupied Prague with tanks. The communist regime ended with the Velvet Revolution in 1989. Let's visit all the places that recall these events. We will start at the Old Town Square and continue to the streets of the New Town, focusing on the vibrant Národní Street. We will finish at the Wenceslas Square and from there we will take a metro to Náměstí Míru.

17:00 – 20:00 Sounds of Science

National House, Náměstí Míru 9

Prague Philharmonia + panel discussion with three French Nobelists in chemistry and physics.

20:00 Conference dinner at the Kaiserstein Palace

Malostranské náměstí 37/23

(one tram stop from Malostranská Metro station line A)

You are invited to discover the discreet charm of Baroque artwork in Prague's Lesser Quarter, the Kaiserstein Palace. Originally built in 1654, the palace is now protected as a historical landmark by UNESCO. The palace has had many famous dwellers over the years, such as opera diva Ema Destinnová and the scientist Joachim Barrande.

PRAGUE TRANSPORT

Public transport

A ticket for 24 Czech Crowns (CZK) is valid for a 30-min journey. A ticket for 32 CZK is valid for a 90-min journey by all means of public transport (trams, buses, metro and local trains) for an unlimited number of transfers. The 24-hour ticket costs 110 CZK. Tickets are sold at the airport, at railway and metro stations, at newspaper stands, at hotel reception desks, etc. The Metro runs from 5:00 till midnight.

Ticket machines accepting contactless payment cards are at the tram stop and in Metro station near the conference venue.

Airport and railway stations

The Václav Havel Airport Prague is situated 9 km from the Institute, with a comfortable connection by bus No. 191 to/from the “Sídliště Petřiny” stop next to the Institute. International trains arrive at and depart mainly from the station Praha Hlavní nádraží (Metro red line C). **The Institute is situated near the Metro station “Petřiny”, green line A.**

EMERGENCY PHONE NUMBERS

General emergency: 112

Medical ambulance service: 155

Police: 158

20. 6. 2018, 17:00

Národní dům na Vinohradech
Náměstí Míru 9, Prague

The French institute in Prague presents

SOUNDS OF SCIENCE

General public lectures by 3 French Nobel Prize awardees:

Jean-Marie Lehn, Nobel Prize Chemistry 1987

Serge Haroche, Nobel Prize Physics 2012

Jean-Pierre Sauvage, Nobel Prize Chemistry 2016

+

Science-related musical program by PKF - Prague Philharmonia

+

Moderated by Daniel Stach, Česká televize

Event in English with Czech translation upon request

kultura.ifp.cz

Organizers



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PROF. JEAN-MARIE LEHN AS OUR ESTEEMED GUEST

The organizers are extremely happy to announce a **plenary lecture by the Nobel Laureate, Professor Jean-Marie Lehn.**

Our esteemed guest will deliver his lecture on **Thursday, 21 June**, during the afternoon session.



Jean-Marie Lehn is a French chemist. He received the Nobel Prize in Chemistry together with Donald Cram and Charles Pedersen in 1987 for his synthesis of cryptands.

Lehn was an early innovator in the field of supramolecular chemistry, i.e., the chemistry of host-guest molecular assemblies created by intermolecular interactions, and continues to innovate in this field. His group has published in excess of 900 peer-reviewed articles in chemistry literature.

Jean-Marie Lehn also studied music, saying that it became his major interest after science. He has continued to play the organ throughout his professional career as a scientist.

(Wikipedia)

CONFERENCE PROGRAMME

Sunday, 17 June 2018

16:00 – 19:00 **Registration and welcome buffet**

Monday, 18 June 2018

8:30 – 9:00

OPENING

Jiří Kotek (Director of the Institute)

Miroslava Dušková-Smrčková (Conference Chair)

Polymer Networks Group

Mitsuhiro Shibayama (Chair of PNG)

100 years M. Gordon and branching theory

Karel Dušek (Honorary Chair)

International Union of Pure and Applied Chemistry

Chi Wu (Representative of IUPAC)

LECTURE SESSION 1

Chaired by: Mitsuhiro Shibayama (Japan)

9:00 – 9:35

Main lecture ML-01

Jian Ping Gong (*Japan*)

Autonomous mechanical remodelling of double network hydrogels

9:35 – 9:55

Invited lecture IL-01

Richard Gerald Weiss (*USA*)

Molecular and polymeric hydro- and organo-gels.
Examples showing the conceptual links with structurally simple gelators

9:55 – 10:15

Coffee break

LECTURE SESSION 2

Chaired by: Jian Ping Gong (Japan)

- 10:15 – 10:35 Keynote lecture KL-01
Yoshihito Osada (*Japan*)
Intelligent gels---Emergent motions of gel machine---
- 10:35 – 10:55 Invited lecture IL-02
Olga E. Philippova (*Russian Federation*)
Smart self-assembled networks
- 10:55 – 11:30 Main lecture ML-04
Jean-François Gérard (*France*)
Bismaleimide-based networks: Reaction mechanisms and toughening strategies

11:30 – 11:40 *Short break*

LECTURE SESSION 3

Chaired by: Olga E. Philippova (Russian Federation)

- 11:40 – 11:55 Oral communication O-03
Tomoki Yasui (*Japan*)
Ionic liquid-based gels with specific inorganic/organic double network
- 11:55 – 12:10 Oral communication O-04
Christine M. Papadakis (*Germany*)
pH responsiveness of hydrogels formed by telechelic polyampholytes
- 12:10 – 12:25 Oral communication O-05
Ralf Weberskirch (*Germany*)
A modular hydrogel platform with tunable mechanical and biological properties

12:25 – 12:40 Oral communication O-06
Takayuki Nonoyama (*Japan*)
Hydrogel possessing thermo reversible robustizing

12:40 – 14:00 *Lunch*

LECTURE SESSION 4A

*Chaired by: Miroslava Dušková-Smrčková (Czech Republic) and
Costas Patrickios (Cyprus)*

14:00 – 14:20 Invited lecture IL-03
Costas Patrickios (*Cyprus*)
Discontinuous volume phase transitions in regular
amphiphilic block polymer conetworks

14:20 – 14:40 Invited lecture IL-04
Kell Mortensen (*Denmark*)
Structural studies of four-armed star-block
copolymer conetwork

14:40 – 15:00 Invited lecture IL-05
Béla Iván (*Hungary*)
Amphiphilic polymer conetworks and gels with
bicontinuous nanophasic morphologies in broad
composition ranges and their nanohybrids: From
nanoreactors to intelligent drug release

15:00 – 15:15 Oral communication O-07
Jun Fu (*People's Republic of China*)
Tough and multi-responsive polymer hydrogels
crosslinked by block copolymer micelles

15:15 – 15:30 Oral communication O-08
Jinhwan Yoon (*Republic of Korea*)
Programmable volume phase transition of the
hydrogels for smart soft matter devices

15:30 – 15:50 *Coffee break*

LECTURE SESSION 5A

Chaired by: Costas Patrickios (Cyprus)

- 15:50 – 16:05 Oral communication O-09
Eva Pinho (*Portugal*)
Antimicrobial composite wound dressing
- 16:05 – 16:20 Oral communication O-10
Bradley David Olsen (*USA*)
Predictions and limitations of a purely topological method for calculating polymer network connectivity
- 16:20 – 16:35 Oral communication O-11
Kristóf Molnár (*Hungary*)
How to prepare gel fibers
- 16:35 – 16:50 Oral communication O-12
Tetsuharu Narita (*France*)
Non-linear rheological properties of rigid polymer network made of a giant polysaccharide
- 16:50 – 17:05 Oral communication O-13
Tomoyuki Koga (*Japan*)
Fast pH-responsive supramolecular hydrogel from peptide-polymer hybrid
- 17:05 – 17:20 Oral communication O-14
Eiji Kamio (*Japan*)
High-strength gels composed of an ionic liquid and inorganic/organic hybrid networks formed via one-pot/one-step process

17:20 – 19:00 **Poster Session I: Posters P-01 – P-36**

LECTURE SESSION 4B

Chaired by: Karel Jeřábek (Czech Republic)

- 14:00 – 14:20 Invited lecture IL-06
Piero Baglioni (*Italy*)
Polymer hydrogel networks and complex fluid for the conservation of modern and contemporary art
- 14:20 – 14:40 Invited lecture IL-07
Evgeny Karpushkin (*Russian Federation*)
Polymer networks: Several application cases
- 14:40 – 14:55 Oral communication O-15
Dmitry V. Pergushov (*Russian Federation*)
Microgel-polyelectrolyte complexes: Toward stimuli-sensitive containers for capacious uptake and triggered release of multi-functional payloads
- 14:55 – 15:10 Oral communication O-16
Takehiko Gotoh (*Japan*)
Hydro-gel-metallurgy
- 15:10 – 15:25 Oral communication O-17
Klaus Opwis (*Germany*)
„Textile mining“ - recovery of noble metals from industrial process waters by the use of textile-fixed polyelectrolytes

15:25 – 15:50

Coffee break

LECTURE SESSION 5B

Chaired by: Lenka Hanyková (Czech Republic)

- 15:50 – 16:05 Oral communication O-18
Sang Youl Kim (*Republic of Korea*)
Synthesis of micro-hydrogel particles consisting of hyperbranched polyamidoamine for the capturing of heavy metal ions and CO₂

- 16:05 – 16:20 Oral communication O-19
Lukas Arens (*Germany*)
Various polyelectrolyte hydrogel architectures -
synthesis, characterization and their application for
salt water desalination
- 16:20 – 16:35 Oral communication O-20
Hayal Bulbul Sonmez (*Turkey*)
Polydimethylsiloxane hybrid polymers as sorbent for
the removal of oil/organic solvent from the
environment
- 16:35 – 16:50 Oral communication O-21
Elena Buratti (*Italy*)
Stimuli-responsive thin films prepared from
PNIPAM/PAAc based microgels
- 16:50 – 17:05 Oral communication O-22
Insu Jeon (*Republic of Korea*)
A hydrogel device for oil/water separation
- 17:20 – 19:00 **Poster Session I: Posters P-01 – P-36**

Tuesday, 19 June 2018

LECTURE SESSION 6

Chaired by: Yoshihito Osada (Japan)

- 8:30 – 9:05 Main lecture ML-02
Oguz Okay (*Turkey*)
Toughness improvement of semicrystalline
hydrogels
- 9:05 – 9:25 Keynote lecture KL-02
Chi Wu (*People's Republic of China*)
Volume phase transition? Continuous or
discontinuous?

9:25 – 9:45 Keynote lecture KL-03
Karel Dušek (*Czech Republic*)
Mixing contribution to equilibrium swelling of
polymer networks

9:45 – 10:05 *Coffee break*

LECTURE SESSION 7

Chaired by: Dirk Kuckling (Germany)

10:05 – 10:20 Oral communication O-23
Carlos G. Lopez (*Germany*)
The swelling of PNIPAM microgels: Effect of charge
and solvent quality

10:20 – 10:35 Oral communication O-24
Lenka Hanyková (*Czech Republic*)
Phase transition in hydrogels of thermoresponsive
interpenetrating polymer networks

10:35 – 10:50 Oral communication O-25
Alba Marcellan (*France*)
Responsive toughening in phase-separated gels

10:50 – 11:05 Oral communication O-26
Miriam Khodeir (*Belgium*)
Redox responsive hydrogels for drug delivery
applications

11:05 – 11:20 Oral communication O-27
Arne Ilseeng (*Norway*)
Predicting the onset of buckling during transient
swelling of hydrogels using FEM

11:20 – 11:30 *Short break*

LECTURE SESSION 8

Chaired by: Oguz Okay (Turkey)

- 11:30 – 11:50 Keynote lecture KL-04
Vladimir I. Lozinsky (*Russian Federation*)
Peculiar features of cryotropic gel-formation and cryostructuring of polymer systems
- 11:50 – 12:05 Oral communication O-28
Tomáš Sedláčik (*Japan*)
Double network cryogels
- 12:05 – 12:20 Oral communication O-29
Xavier P. Morelle (*France*)
Mechanics and fracture of tough hydrogels below water-freezing temperatures

12:20 – 14:00 *Lunch*

LECTURE SESSION 9A

Chaired by: Richard Gerald Weiss (USA)

- 14:00 – 14:20 Invited lecture IL-08
Julius Gyula Vancso (*Netherlands*)
Redox-active poly(ferrocenylsilane)s as actuators and memory hydrogels
- 14:20 – 14:40 Invited lecture IL-09
Alex Li (*USA*)
Photoswitching polymer nanoparticles impart novel bioimaging methods
- 14:40 – 15:00 Keynote lecture KL-05
Miklos Zrinyi (*Hungary*)
Colloid particles that make polymer smart

- 15:00 – 15:15 Oral communication O-30
Dirk Kuckling (*Germany*)
Synthesis of functional smart hybrid materials
- 15:15 – 15:30 Oral communication O-31
Anne-Charlotte Le Gulluche (*France*)
Dynamics of adsorbed layer in hybrid hydrogels

15:30 – 15:55 *Coffee break*

LECTURE SESSION 10A

Chaired by: Alex Li (USA)

- 15:55 – 16:10 Oral communication O-32
David Rochette (*Germany*)
Kinetically controlled crosslinking of metallo-supramolecular networks driven by the Belousov-Zhabotinsky reaction
- 16:10 – 16:25 Oral communication O-33
Shintaro Nakagawa (*Japan*)
Effects of crosslinking on the edge morphology of patterned polymer brushes
- 16:25 – 16:40 Oral communication O-34
Martha Franziska Koziol (*Germany*)
Microrheology on weakly associated PEG chains in semi-dilute solution
- 16:40 – 16:55 Oral communication O-35
Mohamad Hmadeh (*Lebanon*)
Controlled synthesis of ZIF-8, ZIF-67 and their mixed metal derivatives by a reaction diffusion process in agar gel

16:55 – 17:10 Oral communication O-36
Peter Kasak (*Qatar*)
Tunable properties of betaine-based materials

17:10 – 19:00 **Poster Session II: Posters P-37 – P-75**

LECTURE SESSION 9B

Chaired by: Peter Kořovan (Czech Republic)

- 14:00 – 14:20 Invited lecture IL-10
Aleksey Drozdov (*Denmark*)
Modeling the mechanical response of double-network gels under cyclic deformation
- 14:20 – 14:35 Oral communication O-37
Kengo Nishi (*Germany*)
Experimental observation of two features unexpected from the classical theories of rubber elasticity
- 14:35 – 14:50 Oral communication O-38
Toni Müller (*Germany*)
Shear deformation of entangled and unentangled polymer networks: A Monte-Carlo-Study
- 14:50 – 15:05 Oral communication O-39
Michael Lang (*Germany*)
On the elasticity of phantom networks with cyclic and linear defects
- 15:05 – 15:20 Oral communication O-40
Ivan Kryven (*Netherlands*)
New developments in random graphs

15:20 – 15:35 Oral communication O-41
Verena Schamboeck (*Netherlands*)
The effect of Euclidean space on graph-like models
of hyperbranched polymer networks

15:35 – 15:55 *Coffee break*

LECTURE SESSION 10B

Chaired by: Aleksey Drozdov (Denmark)

15:55 – 16:10 Oral communication O-42
Yuliia Orlova (*Netherlands*)
Modeling of ethyl linoleate polymer networks via
automated reaction mechanism

16:10 – 16:25 Oral communication O-43
Oleg Rud (*Czech Republic*)
Thermodynamic model for a reversible desalination
cycle using weak polyelectrolyte hydrogels

16:25 – 16:40 Oral communication O-44
Peter Kořovan (*Czech Republic*)
Self-consistent field model of weak polyelectrolyte
gels

16:40 – 16:55 Oral communication O-45
Diego Estupiñán (*Germany*)
Quantification of ligation points in photochemically
linked polymer networks

16:55 – 17:10 Oral communication O-46
Xiang Li (*Japan*)
A model physical gel crosslinked by double
stranded DNA

17:10 – 19:00 **Poster Session II: Posters P-37 – P-75**

Wednesday, 20 June 2018

LECTURE SESSION 11

Chaired by: Miklos Zrinyi (Hungary)

- 8:30 – 9:05 Main lecture ML-03
Ferenc Horkay (USA)
Hierarchical structure and function of cartilage matrix
- 9:05 – 9:25 Invited lecture IL-11
Dongsheng Liu (People's Republic of China)
DNA supramolecular hydrogels
- 9:25 – 9:45 Invited lecture IL-12
David Díaz Díaz (Germany)
Self-healing alginate gels that do not fail on stretching to 16000%

9:45 – 10:05

Coffee break

LECTURE SESSION 12A

Chaired by: Ferenc Horkay (USA)

- 10:05 – 10:20 Oral communication O-47
Milena Lama (France)
Mechanical properties of tissue-like hydrogels produced by injection of spray-dried collagen
- 10:20 – 10:35 Oral communication O-48
Sora Lee (Republic of Korea)
Immunomodulatory effects of schizophyllan on RAW264.7 cell in three-dimensional culture matrix

- 10:35 – 10:50 Oral communication O-49
Olatz Guaresti Larrea (*Spain*)
Design of thiol-modified chitosan hydrogels with different maleimide-based cross-linkers
- 10:50 – 11:05 Oral communication O-50
Iñigo Díez-García (*Spain*)
Waterborne poly(urethane-urea) synthesized from biobased polyol and triblock copolymers containing hydrophilic block
- 11:05 – 11:20 Oral communication O-51
David Juriga (*Hungary*)
Tooth derived stem cell cultivation on poly(aspartamide) based hydrogels

11:20 – 11:30 *Short break*

LECTURE SESSION 13A

Chaired by: David Díaz Díaz (Germany)

- 11:30 – 11:45 Oral communication O-52
Václav Petrák (*Czech Republic*)
Modification of hydrogels by a femtosecond laser:
From mechanism to ophthalmic applications
- 11:45 – 12:00 Oral communication O-53
Joerg C. Tiller (*Germany*)
Polymer networks for enzyme-induced mineralization
- 12:00 – 12:15 Oral communication O-54
Kamil Maciol (*Germany*)
Design of novel epoxide monomers for pH-sensitive poly(ethylene glycol) hydrogels via acid-labile and crosslinkable allyl side groups

12:15 – 12:30 Oral communication O-55
Larisa V. Sigolaeva (*Russian Federation*)
Rational design of advanced electrochemical
enzyme biosensors via surface functionalization by
stimuli-sensitive microgels

12:30 – 13:40

Lunch

LECTURE SESSION 12B

Chaired by: Jean-François Gérard (France)

10:15 – 10:25 Invited lecture IL-13
Brigitte Voit (*Germany*)
Multifunctional, responsive hydrogels reaction
compartments and in microfluidic application

10:25 – 10:45 Invited lecture IL-14
Marco Sangermano (*Italy*)
Polymeric capsules produced via miniemulsion or
aerosol cationic photopolymerization

10:45 – 11:00 Oral communication O-56
Kinga Pielichowska (*Poland*)
Polyoxymethylene/functionalized hydroxyapatite
nanocomposites with improved thermal stability

11:00 – 11:15 Oral communication O-57
Jessica Link (*France*)
Dynamics and structure of PVDF - solvent gels
filled with silica

11:15 – 11:30

Short break

LECTURE SESSION 13B

Chaired by: Marco Sangermano (Italy)

- 11:30 – 11:45 Oral communication O-58
Evgenia Vaganova (*Israel*)
What can be learned from polymerization in a two-component pyridine-based system
- 11:45 – 12:00 Oral communication O-59
Benjamin Le Droumaguet (*France*)
Metallic nanoparticles immobilized at the pore surface of polymeric materials: Towards versatile and efficient catalytic systems
- 12:00 – 12:15 Oral communication O-60
Karel Jeřábek (*Czech Republic*)
Mesoporous polymers created by microsyrnesis and their application
- 12:15 – 12:30 Oral communication O-61
Patrice Woisel (*France*)
« Colored » multi-stimuli responsive hydrogels

12:30 – 13:40

Lunch

14:00 – 17:00 **Social programme (Guided tours in Prague)**

17:00 – 20:00 **Sounds of Science**

Prague Philharmonia + panel discussion with three Nobelists in chemistry and physics

20:00 – 22:00 **Conference dinner**

Thursday, 21 June 2018

LECTURE SESSION 14

Chaired by: Miroslava Dušková-Smrčková (Czech Republic)

- 8:30 – 9:50 Invited lecture IL-15
Timothy Sirk (*USA*)
Relating mechanics to chain-level architecture in glassy crosslinked polymers
- 9:50 – 9:10 Invited lecture IL-16
Peter Krajnc (*Slovenia*)
Fixed volume hydrogels from emulsion templated polyacrylate networks
- 9:10 – 9:25 Oral communication O-01
Juliette Slooman (*France*)
Fracture of interpenetrated networks: From bond scission to macroscopic fracture
- 9:25 – 9:40 Oral communication O-02
Isabell Tunn (*Germany*)
Inspired by nature: Reinforcing coiled coil hydrogel building blocks with histidine-metal coordination

9:40 – 10:05

Coffee break

LECTURE SESSION 15

Chaired by: Peter Krajnc (Slovenia)

- 10:05 – 10:20 Oral communication O-62
Christian Krumm (*Germany*)
APCNs based on crystallizable poly(2-oxazolines)

- 10:20 – 10:35 Oral communication O-63
Erol-Dan Licsandru (*France*)
 Integration of industrial by-products in bio-based resins and composites
- 10:35 – 10:50 Oral communication O-64
Antonio María Borrero López (*Spain*)
 Influence of processing variables on the rheological properties of one-step processed castor oil/lignin-based gel-like polyurethanes
- 10:50 – 11:05 Oral communication O-65
Esperanza Cortés Triviño (*Spain*)
 Rheological behaviour of epoxide-functionalized cellulose pulp gel-like dispersions in castor oil
- 11:05 – 11:20 Oral communication O-66
Igor Krupa (*Qatar*)
 Hybrid silica hydrogel matrices for an immobilization of glucose sensitive proteins

11:20 – 11:40

Short break

LECTURE SESSION 16

Chaired by: Julius Gyula Vancso (Netherlands)

- 11:40 – 11:55 Oral communication O-67
Youngjong Kang (*Republic of Korea*)
 Perovskite nanoparticle composite films by size-exclusive mass flow lithography
- 11:55 – 12:10 Oral communication O-68
Federica Cavalli (*Germany*)
 A novel strategy for precision network formation based on *para*-Fluoro-Thiol ligation

12:10 – 12:25 Oral communication O-69
Hideyuki Otsuka (*Japan*)
Thermally healable and reprocessable polymer networks based on dynamic covalent chemistry of bis(hindered amino)disulfides

12:25 – 14:00

Lunch

LECTURE SESSION 17

*Chaired by: Miroslava Dušková-Smrčková and Karel Dušek
(Czech Republic)*

14:00 – 15:00 Nobel lecture
Jean-Marie Lehn (*France*)
DYNAMERS: Towards adaptive dynamic polymers and gels

15:00 – 15:35 Main lecture ML-05
Mitsuhiro Shibayama (*Japan*)
Phenolic resins—recent progress of structure, properties, and dynamics investigations—

15:35 – 15:45 **Closing of the Meeting, Farewell drink**

LIST OF POSTERS

- P-01 Z. Yang, Z. Dong** (*People's Republic of China*)
EOR of the combination flooding systems consisting of polymer microspheres and nonionic surfactant for Bohai oilfield
- P-02 M. Lin, Z. Dong, J. Zhang** (*People's Republic of China*)
Polymerizable microsphere-included high mechanical strength of composite hydrogel composed of acrylamide
- P-03 S. Sridhar, Y. Li, S. Wang, B. Xu** (*United Kingdom*)
An emerging thermo-electric generator driven by trampolining elastic gels
- P-04 A. I. Barabanova, V. S. Molchanov, O. E. Philippova, A. R. Khokhlov** (*Russian Federation*)
Synthesis of magnetic nanocomposites with tunable epoxy matrix
- P-05 H. S. Lim, W. J. Oh, J. S. Won, S. G. Lee** (*Republic of Korea*)
Manufacture and characterization of piezoelectric composites using PAR/PVDF sheath-core fiber
- P-06 M. Dauletbekova, G. Toletay, S. Kabdrakhmanova, S. Kudaibergenov** (*Kazakhstan*)
Hydrogenation of *p*-nitrobenzoic acid by gold and palladium nanoparticles immobilized within macroporous amphoteric cryogels in aqueous solution
- P-07 K. Kaniewska, W. Hyk, Z. Stojek, M. Karbarz** (*Poland*)
Transport properties of the hydrogel thin film electrodeposited on conducting surface
- P-08 M. Karbarz, M. Mackiewicz, K. Marcisz, Z. Stojek** (*Poland*)
Modification of gold electrode with monolayer of environmentally sensitive microgels

- P-09 Y. Han, J. Hu** (*Hong Kong*)
Skin collagen fiber/polyurethane biocomposite with water-induced shape memory ability
- P-10 C. Yun, H. Go, E. Han** (*Republic of Korea*)
The roll of organometalic sol-gel films for stretchable transparent electrodes
- P-11 K. Nishi, F. C. MacKintosh, C. F. Schmidt** (*Germany*)
Dynamics of semi-flexible filament in viscoelastic media: Microrheology using semiflexible polymer
- P-12 J. Sawada, T. Takata** (*Japan*)
A vinylic rotaxane cross-linker toughening network polymers via radical polymerization of vinyl monomers
- P-13 G. Toleutay, A. Shakhvorostov, M. Dauletbekova, A. Mukhan, S. Kabdrakhmanova, S. Kudaibergenov** (*Kazakhstan*)
“Quenched” polyampholyte hydrogels based on (3-acrylamidopropyl)trimethyl ammonium chloride and sodium salt of 2-acrylamido-2-methyl-1-propanesulfonic acid
- P-14 E. Šestáková, L. Hanyková, I. Krakovský** (*Czech Republic*)
Temperature-sensitive double network hydrogels
- P-15 N. Pettinelli, L. Barral, R. Bouza, Y. Farrag, B. Montero, M. Rico** (*Spain*)
Hybrid hydrogel based on κ -carrageenan and methacrylates for biomedical applications
- P-16 W. G. Jo, H. R. Lee, J. W. Jeong, J. S. Won, S. G. Lee** (*Republic of Korea*)
Study on the cure behaviors of the modified epoxy resins and mechanical properties of its carbon fiber composites
- P-17 E. Ito, D. Suto, M. Baba, H. Maruyama, K. Yamamoto** (*Japan*)
Surface structure analysis of silicone hydrogel by neutron reflectivity measurement

- P-18 I. Calina**, M. Demeter, C. Vancea, M. Micutz, T. Staicu, M. Albu Kaya (*Romania*)
E-beam cross-linking of collagen-co-poly(vinylpyrrolidone)-poly(ethylene oxide) triblock superabsorbant hydrogels
- P-19 R. Kiyama**, T. Nonoyama, T. Nakajima, T. Kurokawa, J. P. Gong (*Japan*)
Direct observation of single polymer strand based on double network strategy
- P-20 A. Tarasov, M. Rodin**, L. Romanova, V. Komratova (*Russian Federation*)
Influence of oligodiol content and chain length on properties of beta-cyclodextrin/oligodiol cross-linked polyurethanes
- P-21 T. Kureha**, X. Li, M. Shibayama (*Japan*)
Volume transition of poly(oligo ethylene glycol methacrylate)-based hydrogels
- P-22 J. John**, S. Varughese, A. P. Deshpande (*India*)
Role of distinct microstructures on the rheological behavior of pectin-Ca gels
- P-23 D. A. Martín**, T. S. Grigera, **V. I. Marconi** (*Argentina*)
Speeding up the the study of dipolar systems gelation
- P-24 I. V. Blagodatskikh**, V. E. Tikhonov, **O. V. Vyshivannaya**, V. A. Postnikov, A. R. Khokhlov (*Russian Federation*)
Design of a reactive polyvinylalcohol gel and rigid macroporous sorbents for biomedical application
- P-25 J. Ewald**, H. Pohlit, M. Worm, H. Frey (*Germany*)
Degradable PEG-hydrogels
- P-26 A. I. Cocarta**, J. Širc, R. Hobzová, K. Švojgr, P. Pochop (*Czech Republic*)
Methacrylate-based hydrogels for trans-scleral administration of topotecan and vincristine in retinoblastoma therapy

- P-27 J. H. (Jin Hyoung) Kim**, D. H. Jo, T. G. Lee, J. H. (Jeong Hun) Kim (*Republic of Korea*)
Bio-inspired corona formation mimicking the ocular environment for controlled *in vivo* therapeutic application of polymer particles
- P-28 J. H. (Jeong Hun) Kim**, B. J. Lee, J. H. (Jin Hyoung) Kim (*Republic of Korea*)
Polymer-DNA conjugates with amplified VEGF aptamers inhibit retinal vascular hyperpermeability
- P-29 K. Vránová**, V. Petrák, Z. Mics, M. Reidingerová, V. Stoy (*Czech Republic*)
Novel hydrogels for post-operatively adjustable intraocular lenses
- P-30 A. Klavina**, B. Maurina, I. Martinsone (*Latvia*)
Carboxymethyl cellulose gel systems with spropel extract
- P-31 L. Kománková**, M. Pařízek, H. Hlídková, M. Hrubý, M. Vetrík (*Czech Republic*)
Three-dimensional carbon-based polymer scaffold for bone tissue engineering
- P-32 G. Dura**, H. Waller, D. T. Peters, J. H. Lakey, D. A. Fulton (*United Kingdom*)
Novel biosynthetic Caf1 - based hydrogels with potential as cell scaffolds
- P-33 Z. Sadakbayeva**, M. Dušková-Smrčková, K. Dušek (*Czech Republic*)
Experimental evaluation of theoretical model of IPN hydrogel elasticity
- P-34 A. Kazakov**, P. Košovan, A. I. Cocarta, J. Širc (*Czech Republic*)
Modeling solute diffusion through a hydrogel

- P-35 E. Sipos**, M. Zrinyi (*Hungary*)
Mechanical properties of randomly oriented electrospun fibre texture
- P-36 M. Lang**, C. Schuster, R. Dockhorn, M. Wengenmayr, J.-U. Sommer (*Germany*)
Feringa type engines in polymer model systems: Folding, coiling, molecular stirling engines, and active gels
- P-37 T. Müller**, M. Lang, J.-U. Sommer (*Germany*)
The elasticity of real polymer networks without entanglements
- P-38 C. G. Lopez**, W. Richtering (*Germany*)
Flory-Rehner and scaling descriptions of thermoresponsive PNIPAM gels
- P-39 D. Kwon**, Y. Jochi, Y. Takeoka, T. Seki, K. Satoh, M. Kamigaito (*Japan*)
Precise synthesis and thermal properties of homogeneous copolymer gels with different monomer sequence
- P-40 H. R. Lee**, W. K. Cho, H. S. Lim, J. S. Won, S. G. Lee (*Republic of Korea*)
Crystallization behaviors of glass fiber/polyamide 6 composites
- P-41 T. Narita**, G. Ducouret, M. Kawai, T. Mitsumata, M. K. Okajima, T. Kaneko (*France*)
High frequency dynamics of a liquid crystalline, cyanobacterial, sulfated polysaccharide studied by DLS/DWS microrheology
- P-42 M. M. Villar-Chavero**, J. C. Domínguez, M. V. Alonso, M. Oliet, F. Rodriguez (*Spain*)
Complex viscosity modeling of ionogels reinforced with chitosan

- P-43 L. Arens**, K. Schlag, F. Weißenfeld, I. Wagner, M. Wilhelm
(*Germany*)
Osmotic engine - energy recovery from salt gradients via polyelectrolyte hydrogels
- P-44 M. Tomic**, B. T. Stokke (*Norway*)
Spatiotemporal features of responsive hydrogel materials characterized by quantitative phase contrast microscopy
- P-45 J. Štorkán**, T. Vampola, M. Dušková-Smrčková, Z. Sadakbayeva, K. Dušek (*Czech Republic*)
Swelling and deformation responses of porous hydrogel simulated with finite element method
- P-46 J. Zavřel**, T. Vampola, M. Dušková-Smrčková, D. Kubies, Z. Sadakbayeva (*Czech Republic*)
Modelling of porous hydrogel topology in 3D: When and how much the pores communicate?
- P-47 L. Nová**, P. Košovan, F. Uhlík (*Czech Republic*)
Swelling of weak polyelectrolyte gels in the presence of hydrophobic counterions
- P-48 M. Mackiewicz**, J. Romanski, Z. Stojek, M. Karbarz (*Poland*)
Degradable, thermo-, pH- and redox- sensitive hydrogel microcapsules for burst or sustained release of the drugs
- P-49 S. Nishimura**, S. Matsubara, N. Higashi, T. Koga (*Japan*)
Thermo-responsive hydrogel from amino acid-derived triblock polymers via flower-like micelle formation
- P-50 S.-H. Jung**, A. Pich (*Germany*)
Degradable supramolecular colloidal gels
- P-51 Z. Osváth**, T. Tóth, B. Iván (*Hungary*)
Thermoresponsive poly(n-isopropylacrylamide) and its 3-(trimethoxysilyl)propyl methacrylate based copolymers, hybrid networks and gels

- P-52** J. E. Martín Alfonso, E. Číková, M. Omastová, **C. Valencia**, J. M. Franco (*Spain*)
Electrospun polyvinylpyrrolidone/essential oil composites fibers
- P-53** **O. Linker**, J. Blankenburg, K. Maciol, H. Frey (*Germany*)
Carboxylic acid functional poly(ethylene glycol) for non-covalent crosslinked hydrogels
- P-54** **O. V. Vyshivannaya**, I. R. Nasimova, E. Yu. Kozhunova (*Russian Federation*)
Thermo- and pH-sensitive interpenetrating network microgels for coatings
- P-55** **L. Alfheid**, M. Geoghegan, N. Williams, W. Seddon (*Saudi Arabia*)
Double-network hydrogels improve pH-switchable adhesion
- P-56** Y. Nagasawa, Y. Amano, **Y. Seida**, T. Gotoh, E. Furuya (*Japan*)
Recovery of CO₂ using temperature-responsive amine gel slurry
- P-57** **P. Berg**, D. Simon, F. Obst, C. Pilger, H. Hachmeister, T. Huser, D. Appelhans, D. Kuckling (*Germany*)
Polymer networks in continuous flow reactors for organocatalytic reactions
- P-58** **L. Raksaksri**, S. Chuayjuljit, P. Chaiwutthinan, A. Boonmahitthisud (*Thailand*)
Preparation and properties of ENR-40/nanosilica nanocomposites via safe sulfur curing system
- P-59** **L. N. Khatiwada**, J. Pflieger, J. Peter, D. Chmelíková, D. Kubies, E. Chánová, M. Dušková-Smrčková (*Czech Republic*)
Water uptake by ultrathin crosslinked films studied by QCM

- P-60** A. Tenorio-Alfonso, M. C. Sánchez, **J. M. Franco** (*Spain*)
Adhesion and rheological assessment of bio-sourced polyurethane networks from cellulose acetate/vegetable oil mixtures
- P-61** **A. M. Borrero-López**, C. Valencia, A. Blánquez, M. Hernández, J. M. Franco (*Spain*)
NCO-functionalized wheat and barley straws-based oleogels: Influence of solid-state fermentation
- P-62** **J. Li**, D. Kuckling (*Germany*)
Dually crosslinking supramolecular polymer towards smart gel act as molecular sensors
- P-63** **B. Erdem**, M. Erdem, Z. G. Gungor (*Turkey*)
Investigation of Acid Orange 8 adsorption onto vinyl imidazole-based copolymeric hydrogels
- P-64** **M. Erdem**, T. G. Sayer, F. Gundogdu, B. Erdem (*Turkey*)
Modification of poly(N-isopropylacrylamide): Synthesis and characterization of novel thermosensitive copolymeric hydrogels
- P-65** **L. Debertrand**, J. Zhao, T. Narita, C. Creton (*France*)
A mechanical study of tough hydrogels with chemical and physical crosslinks
- P-66** **K. Marcisz**, M. Mackiewicz, J. Romanski, Z. Stojek, M. Karbarz (*Poland*)
Synthesis and characteristic of electroswitchable microgel
- P-67** **F. Jung**, P. A. Panteli, C. Tsitsilianis, C. S. Patrickios, C. M. Papadakis (*Germany*)
Light and X-ray scattering study of a temperature and pH responsive physical hydrogel
- P-68** **J. Ryoo**, C. S. Ki (*Republic of Korea*)
Photopolymerized double network hydrogel of hyaluronic acid and silk fibroin

- P-69 R. K. Raya**, J. Škvarla, K. Procházka, M. Štěpánek
(*Czech Republic*)
Thermoresponsive behavior of poly(N-isopropylacrylamide)s with dodecyl and carboxyl terminal groups in aqueous solution: pH-dependent cloud point temperature
- P-70 S. Kato**, K. Ishizuki, R. Goseki, D. Aoki, H. Otsuka (*Japan*)
Freezing-induced mechanoluminescence of cross-linked polymer gels that contains a light-emitting mechanochromophore
- P-71 T. Kosuge**, M. J. Robb, A. J. Halmes, V. M. Lau, D. Aoki, J. S. Moore, H. Otsuka (*Japan*)
Polymer-inorganic composite networks showing multicolor mechanochromism with dual mechanochromophores
- P-72 G. Dalkas**, A. Matheson, H. Vass, A. Gromov, V. Koutsos, P. Clegg, S.R. Euston (*United Kingdom*)
The structure and physical properties of mixed sterol organogels
- P-73 Y. Zhang**, L. Zhao, J. Liao, T. Wang, W. Sun, **Z. Tong**
(*People's Republic of China*)
Shape memory, spontaneous actuation, and programmable actions of hydrogels
- P-74 S. K. Park**, B. J. Park, M. J. Choi, E. J. Shin, S. Park
(*Republic of Korea*)
Poly(dimethylsiloxane)-based crosslinked film with high dielectric constant and low modulus
- P-75 Y. Zhuo**, V. Håkonsen, Z. He, S. Xiao, J. He, Z. Zhang
(*Norway*)
Enhancing the mechanical durability of icephobic surfaces by introducing autonomous self-healing function

MAIN LECTURES

ML-01

AUTONOMOUS MECHANICAL REMODELLING OF DOUBLE NETWORK HYDROGELS

T. Matsuda^a, T. Nakajima^{b,c}, J. P. Gong^{b,c}

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^b*Faculty of Advanced Life Science, Hokkaido University*

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Recently, a number of approaches have been developed to create hydrogels with excellent mechanical performance. Among them, double network hydrogels (DN gels) which consist of a brittle polyelectrolyte first network and a flexible neutral second network stand out because they exhibit high toughness as well as high Young's Modulus (Gong et al., *Adv. Mater.* 2003). These properties give DN gels great potential in load-bearing applications. The high toughness of DN gels comes from the internal fracture of covalent bonds in the brittle first network which dissipates significant energy. In this work, we developed a strategy to autonomously remodeling the double network hydrogels by mechanical stimuli.

ML-02

TOUGHNESS IMPROVEMENT OF SEMICRYSTALLINE HYDROGELS

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Owing to their similarities to biological tissues, hydrogels as soft and smart materials have important functions in a variety of biological and biomedical applications. Although hydrogels are traditionally brittle and exhibit a low modulus of elasticity in the range of kPa, significant progress has been achieved in the past 15 years in the design of mechanically strong and tough hydrogels. In this talk, I will present two strategies to improve the mechanical properties of supramolecular semicrystalline hydrogels.¹⁻³ We design a highly entangled physical network formed via lamellar crystals consisting of aligned side alkyl chains. The network bases on poly(N,N-dimethylacrylamide) (PDMA), which is a very useful hydrophilic biocompatible polymer with associative properties. The interconnected lamellar crystals forming a layered hydrogel structure are created by incorporating n-octadecyl acrylate (C18A) with side chain length of 18 carbons into the PDMA backbone. Addition of a small amount of weak hydrophobe such as lauryl methacrylate (C12M) in the gelation system, or prestretching of the hydrogel above the melting temperature of its crystalline domains results in 10-fold increase of toughness without losing their high modulus and high strength. Small- and wide-angle X-ray scattering measurements and mechanical tests reveal that the significant toughness improvement is due to the formation of more ordered crystalline domains and appearance of active tie molecules under external force interconnecting the lamellar clusters.

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ML-03

HIERARCHICAL STRUCTURE AND FUNCTION OF CARTILAGE MATRIX

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This work focuses on developing a quantitative understanding of how the main polymeric components (aggrecan, hyaluronic acid, collagen) of extracellular matrix determine the biological functions of cartilage. In particular, we investigate the structure and dynamic properties of model systems using osmotic pressure measurements and scattering measurements (SANS, SAXS, DLS, etc.). We identify important physical properties that distinguish these stiff polymers from other highly charged polyelectrolytes. These differences are important because such rigid structures are typical in living systems. Osmotic pressure measurements made on aggrecan solutions bring evidence of self-assembly of the bottlebrush shaped aggrecan molecules into microgel-like assemblies. It is demonstrated that in near-physiological salt solutions aggrecan assemblies display remarkable insensitivity to changes in the ionic environment, notably to multivalent cations. This insensitivity of the structure and dynamics of aggrecan assemblies to calcium ions is contrary to the behavior of linear polyelectrolytes. In the latter, multivalent cations generate short-range attractive multipolar interactions that favor the development of contacts between adjacent chains. In the aggrecan solution, replacing sodium counter-ions with calcium ions, i.e., changing the electrostatic interactions, does not alter the structure of the polyanion. The structural stability of aggrecan assemblies is critical for the load-bearing properties of cartilage. Our findings suggest that these “microgel particles” may act as an ion reservoir mediating calcium metabolism in cartilage and bone. The present results are consistent with the role of aggrecan as an essential structural component in the load bearing function of cartilage and as an ion-exchange matrix in bone mineralization.

ML-04

BISMALEIMIDE-BASED NETWORKS: REACTION MECHANISMS AND TOUGHENING STRATEGIES

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Three different types of bismaleimide systems were considered to design highly crosslinked networks: *i/* homopolymerization of bismaleimides to form very brittle networks; *ii/* copolymerization of bismaleimides and amines via Michael's reaction leading to less brittle networks as the crosslink density is lower; *iii/* copolymerization of bismaleimides and bisallyles which display similar aptitude to be processed as epoxies. Thus, the research focused on the reaction mechanisms of 1,1'-(methyl-di-4,1-phenyl)bismaleimide (BMI) and 2,2'-diallylbisphenol A (DBA) systems. The copolymerization analyzed by NMR on model reactants was found to proceed first from a Alder-ene type reaction of the two monomers leading the growth of linear chains and secondly to a crosslinking reaction from the homopolymerization of maleimide groups and/or addition reactions (Diels-Alder's or Wagner-Jauregg's) and recombinations involving maleimide functions and the adduct from the Alder-ene reaction. The chemorheological behavior of the reactive systems performed in isothermal conditions evidenced that the crosslinking mechanism involves radical character. As a consequence, such reactive bismaleimide/bisallyle systems proceed very differently than epoxy resins which evolve step growth polymerization mechanism. Thus, the resulting networks are heterogeneous in terms of crosslinking density as the construction of the networks proceeds issues via microgels formation. Such topological heterogeneities can be limited by introducing radical inhibitors. In order to improve fracture behavior of such networks, high T_g thermoplastics such as polyethersulfones (PES) and polyetherimides (PEI) were considered to design polymer blends morphologies from a reaction-induced phase separation process (which was identified as a spinodal decomposition occurring during curing).

ML-05

PHENOLIC RESINS—RECENT PROGRESS OF STRUCTURE, PROPERTIES, AND DYNAMICS INVESTIGATIONS—

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For more than a hundred years, phenolic resins, the first human-invented plastics, have been widely used since its discovery in 1907. Phenolic resins have excellent physical properties such as mechanical strength, electrical insulation, heat resistance, and solvent resistance.^{1,2} These properties result from their highly cross-linked network structures, which comprise phenolic and methylene units. In these structures, three methylenes can connect to a phenolic ring such that one can be at the para (*p*) and two can be at the ortho (*o*) positions that are adjacent to the hydroxyl group of the phenolic ring. To further improve these properties, the understanding of the relation between the network structure and physical properties is one of the most important issues. However, this relation has not been completely understood experimentally because a highly cured resin is insoluble and infusible. We have been tackling to these problems and investigating the structures and properties of phenolic resins in terms of scattering techniques, such as small angle neutron/X-ray scattering,^{3,4} quasi-elastic neutron scattering, dynamic light scattering, NMR spectroscopy, and molecular dynamics simulations.^{5,6} The structure–property relationship of the cured resin was characterized by atomistic molecular dynamics simulation for a structural model consistent with experimental results in terms of scattering function.

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NOTES

KEYNOTE LECTURES

KL-01

INTELLIGENT GELS---EMERGENT MOTIONS OF GEL MACHINE---

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One of the ultimate goals for gel science is to create hydrogels with “**Emergent Functions**” which biological soft tissues possess and eventually to replace them. In this connection, we have developed double-network gels (**DN gels**) which have similar mechanical toughness as biological tissue (elastic modulus 0.3 MPa, fracture stress 10 MPa and fracture energy up to 10^3 J m^{-2})¹ and have surface friction lower than that of synovial cartilage ($\mu < 10^{-4}$).²

A decade ago we also reported ATP fueled gel machines reconstructed from chemically cross-linked actin gel which moved along the myosin (muscle protein) gel by coupling to ATP hydrolysis (**Nano-Biomachine**).³ The point is this artificial bio-machine can move with increased velocity and increased power (energy) than those of the native proteins. This means the covalently bound protein gels are able to exert **Emergent Function** cooperatively synchronizing and integrating between component actin and myosin fibers. This situation is totally different from that of chemomechanical systems made of synthetic polymer gels.^{4,5} The behavior and mechanism will be discussed in terms of hierarchical structure of actin-myosin hydrogels.⁶

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KL-02

VOLUME PHASE TRANSITION? CONTINUOUS OR DISCONTINUOUS?

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The “volume phase transition” is a term that has been extensively used in the last 30 years in the gel community. The original concept was dated back to a paper published by Karl Dusek in 1968. Unfortunately, most of people, who are using this term or citing this paper, have not really read and understand it. In his original paper, the volume phase transition was predicted for a uniform gel network; namely, all the subchains have an identical length. However, real gels never satisfy such a condition. In parallel, a monodispersed or narrowly distributed polymer sample has well-defined phase transition temperatures at different concentrations, but a polydispersed polymer sample should not have a well-defined phase transition temperature because different chain lengths should lead to different phase transition temperatures. Therefore, real gels should not have a so-called discontinuous volume phase transition but a gradual transition because its subchain lengths are broadly distributed. People often argued that we did observe the discontinuous or sharp volume change. In this lecture, we will explain why we should not call such an observed sharp volume change as “volume phase transition” because it is due to other reasons, not that predicted by Karl Dusek.

KL-03

MIXING CONTRIBUTION TO EQUILIBRIUM SWELLING OF POLYMER NETWORKS

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Thousands of papers on swelling of cross-linked polymeric structures published in the last few decades are based on assumptions of additivity of mixing and elasticity contributions. The originally assumed independence of these two contributions has been challenged by experiments and newer theories and transformed into a well determined interdependence. Nevertheless, the original reduction of the effect of mixing of polymer segments with solvent molecules to that of mixing linear polymer of infinite molecular weight remains a base for interpretation of major experiments. This leads to “wild” empirical values of the interaction parameter χ and its dependences on polymer concentration and temperature. Moreover, a combination of volume determined models of mixing with elasticity of volumeless ghost (phantom) network is a very unequal marriage with dubious outcome.

Modern theoretical approaches view swelling as coexistence of various volume filling clusters of polymer segments and solvent molecules which is characteristic for polar and especially hydrogen bonded systems (hydrogels). The interacting species are viewed as volume occupying objects with several numbers and types of interacting surface sites (patches), the interactions being affected by states of topological and spatial neighbors (branching theory, Ising lattice). Contributions by covalent crosslinks are often marginal and affect very little phase transitions and mechanical instabilities of the system (cf., e.g., PNIPAM). The behavior is dominated by physical interactions and transiency of crosslinks; it is related to such new fields of interest as dynamers or self-healing systems.

Although the empirical dependences of χ on polymer concentration and temperature have no predictive power, they can be utilized for analysis of phase behaviors and fine tuning of phase transitions in real systems.

PECULIAR FEATURES OF CRYOTROPIC GEL-FORMATION AND CRYOSTRUCTURING OF POLYMER SYSTEMS

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Polymeric *cryogels* and *cryostructurates* are the supramolecular matrices, the formation of which occurs in the non-deeply frozen systems.^{1,2} When the buildup of the 3D polymeric network and its simultaneous cross-linking *via* chemical or physical bonds take place exactly in the frozen sample, the final, i.e. after defrosting, matrices are the cryogels. If the feed solution of the polymeric precursors is initially frozen, and the formed solvent crystals are further removed *via* sublimation or cryoextraction techniques, it results in a macroporous cryostructurate, which can be then fixed chemically using suitable cross-linking methods.

Cryotropic gelation processes possess a set of specific features, when the main observed effects are as follows: (i) an apparent decrease in the critical concentration of gelation upon the preparation of cryogels as compared with gelling of the same precursors a liquid medium; (ii) an increase in the gelation rates over certain range of cryogenic temperatures compared with the rates of the traditional gelation at temperatures above the freezing point (the temperature dependences of the parameters capable of reflecting the efficiency of gelation are, as a rule, of bell-shaped character); (iii) all cryogels are the macroporous gel matrices, since the crystals of frozen solvent perform as porogens, and the gel-formation itself occurs in between their particles in the space of so-called unfrozen liquid microphase, where the solutes are concentrated.

A fortunate combination of good physicochemical properties of diverse polymeric cryogels and cryostructurates with their specific porous morphology makes these materials useful for application in various applied areas, especially as gel matrices of biomedical and biotechnological interests.

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KL-05

COLLOID PARTICLES THAT MAKE POLYMERS SMART

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Electric-, and magnetic field sensitive polymer gels and elastomers are soft smart materials whose elastic- and thermodynamic properties are strong function of the field strength imposed upon them. Colloidal (nano-) particles with special electric and magnetic properties are built into flexible polymer matrix. Different preparation methods were applied including in situ fabrication of nanoparticles template in hydrogel matrix. The particles couple the shape of the gel (or elastomer) to the external fields. Shape distortion occurs instantaneously and disappears abruptly when electric- or magnetic field is applied or removed, respectively. Giant deformational effect, field controlled elastic modulus, non-homogeneous deformation and quick response to magnetic- and electric field open new opportunities for using such materials for various applications. The development of smart polymer composites that show spinning in static uniform field will also be presented. Electrospinning often referred as Quincke rotation (old colloidal phenomenon) is the spinning of non-conducting objects immersed in dielectric liquid and subjected to a strong homogenous DC electric field. It is shown that micro-sized rotors of polymer disk, hollow cylinder and gear wheel acts like colloidal electric motor with tuneable angular frequency.

NOTES

INVITED LECTURES

IL-01

MOLECULAR AND POLYMERIC HYDRO- AND ORGANO- GELS. EXAMPLES SHOWING THE CONCEPTUAL LINKS WITH STRUCTURALLY SIMPLE GELATORS

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Results from studies of some structurally simple molecular and polymeric gelators developed at Georgetown and the properties of their organo- and hydro-gels in a variety of liquids will be presented. Different modes by which sol/solution phases can be transformed into their corresponding gels will be described as well. Dynamic and structural aspects of the gels at different distance and time scales will be discussed. Results from oscillatory and extensional rheology will be employed to assess some of the dynamic properties, including their thixotropy and ability to be self-healing and self-standing. Analyses of X-ray diffraction, neutron scattering, and optical microscopy experiments will be used to describe the self-assembled fibrillar networks of the molecular gels and the crosslinked networks of the polymeric gels.

Some examples of the realized and potential application of the gels for oil spill recovery and conservation of objects of cultural heritage will be presented as well: (1) the use of a small well that has been developed to determine the influence of wave action on the viscoelastic properties of gel layers on water;¹ (2) how surfaces of paintings, frescos, and sculptures can be treated to remove different types of unwanted coatings;² and (3) how halogenated pollutants in water supplies can be removed by iron zero nanoparticles embedded in gel layers.³ In each case, the methods used to analyze the systems before and after treatment will be discussed.

We thank the US National Science Foundation for its support of this research through Grants CHE-1147353 and -1502856.

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IL-02

SMART SELF-ASSEMBLED NETWORKS

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Several approaches to construct self-assembled networks based on supramolecular and polymer chains and/or inorganic nanoparticles are proposed.¹⁻⁴ It is shown that the mixed networks demonstrate a synergistic increase in viscosity as compared to pure component solutions. At the same time, the addition of n-alkanes leads to the drop of viscosity by up to 5 orders of magnitude thus inducing the transition of a gellike system to a fluid with low viscosity. SANS data evidence that this process is accompanied by the breaking of the supramolecular chains with the formation of microemulsion droplets, which results in the disruption of the whole network. Thus, in the prepared smart gels, the polymer component strengthens the network, whereas the supramolecular component imparts a high responsiveness to hydrocarbons, which makes such materials promising for the various applications in oil recovery.

Addition of magnetite nanoparticles acting as additional multifunctional physical cross-links increases the viscosity, the plateau modulus, and the relaxation time of the network, and also imparts to it magneto-responsive properties. A combination of smart properties inherent to both the matrix and the particles makes such systems very competitive with other magnetic soft matter materials for various applications.

Acknowledgment: *The work was financially supported by Russian Science Foundation (project № 17-13-01535).*

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IL-03

DISCONTINUOUS VOLUME PHASE TRANSITIONS IN REGULAR AMPHIPHILIC BLOCK POLYMER CONETWORKS

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This year marks the 50th anniversary of Dušek's^{1,2} theoretical prediction of the discontinuous volume phase transition in polymer gels, and the 40th anniversary of its experimental confirmation by Tanaka.^{3,4} Although this discontinuous transition may also take place within uncharged polymer gels,⁵ it definitely takes place when the gel is even slightly charged and when the solvent is not too good for the polymer. Solvent quality, temperature, pH, salt concentration, light irradiation, electric field, biochemical stimuli, and stress were subsequently shown to be able to trigger this volume transition in gels; furthermore, multiple discontinuous transitions were observed in certain polyampholytic gel systems.⁶

In most of the above experimental systems, gel structure was poorly controlled, with the length of the chains between cross-linking points being broadly distributed and, in the case of copolymer gels, the two or more types of monomer repeating units being randomly distributed. This presentation will focus on a better-defined polymer gel system of amphiphilic polymer conetworks comprising end-linked amphiphilic ionizable ABA triblock copolymers.⁷ We will show, computationally, that, as the degree of ionization and the copolymer composition change, morphological transitions among unimers, spheres, cylinders and lamellae take place, accompanied by discontinuous changes in the aqueous degrees of swelling, the asymmetry ratios (in the case of the anisotropic morphologies) and the oil solubilization capacity.^{8,9}

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STRUCTURAL STUDIES OF FOUR-ARMED STAR-BLOCK COPOLYMER CONETWORK

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We present a structural study of a network of 4-armed PEO–PPO block copolymer stars. The molecules consists of a 1:1 blend of cross-linked tetra amine and tetra-N-hydroxysuccinimide terminated star molecules. The structural properties have characteristics known from simple linear PEO–PPO type of copolymers: at low temperatures, the measured structure is dominated by the characteristics of individual molecules, while at higher temperatures hydrophobic effects of the PPO domains cause self-assembly. The structure of the crosslinked 1:1 TAT–TNT system is organized into two-dimensional network sheets. At low temperatures, the system is rather disordered, while upon raising temperatures, PPO self-assembly causes organization across neighboring sheets, resulting in hexagonally ordered cylinder-assemblies perpendicular to the sheets.¹ The cross-linked network is elastic and can be stretched to more than 50%. Upon stretching, there is apparently no effect on the characteristic structural properties, within the small-angle length scale. The structural basis for the elastic modulus is thus still unclear.

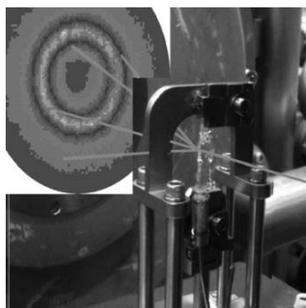


Figure 1. SANS scattering experiment on a 30% aqueous crosslinked TAT-TNT network sample mounted for in-situ stretching experiments.

At low temperatures, the system is rather disordered, while upon raising temperatures, PPO self-assembly causes organization across neighboring sheets, resulting in hexagonally ordered cylinder-assemblies perpendicular to the sheets.¹ The cross-linked network is elastic and can be stretched to more than 50%. Upon stretching,

there is apparently no effect on the characteristic structural properties, within the small-angle length scale. The structural basis for the elastic modulus is thus still unclear.

Acknowledgement: Funding from the Independent Research Fund Denmark and DANSCATT and SANS beam time from PSI (CH) are acknowledged.

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AMPHIPHILIC POLYMER CONETWORKS AND GELS WITH BICONTINUOUS NANOPHASIC MORPHOLOGIES IN BROAD COMPOSITION RANGES AND THEIR NANOHYBRIDS: FROM NANOREACTORS TO INTELLIGENT DRUG RELEASE

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Amphiphilic conetworks (APCNs),¹⁻⁸ composed of covalently bonded otherwise immiscible hydrophilic and hydrophobic polymer chains, belong to a new class of rapidly emerging nanostructured materials. Unique bicontinuous (cocontinuous) nanophase separated morphology exists in APCNs in a broad composition window. Various specialty new intelligent (smart, responsive) gels and organic-inorganic nanohybrids by applying one of the nanophases as nanoreactor have been recently obtained. The resulting novel materials have a variety of high added-value potential applications from nanocatalysis and photonics to biomaterials etc.

Acknowledgement: Support of this research by the National Research, Development and Innovation Office (K112094 and NN116252) and the National Development Agency (KTIA-AIK-12-1-2012-0014) is acknowledged.

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IL-06

POLYMER HYDROGEL NETWORKS AND COMPLEX FLUID FOR THE CONSERVATION OF MODERN AND CONTEMPORARY ART

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We pioneered the application of soft matter and materials science to several fields, one of the most exotic is the conservation of cultural heritage. Art Conservation poses a formidable and exciting challenge to Colloid and Interface Scientists in two respects. First, the majority of the most performing and environmentally-safe cleaning and consolidation agents for artworks are soft matter systems. Second, the interaction of these agents with the artifact involves an exceptionally complicated range of interfacial interactions. Recently we proposed an innovative cleaning system, namely a semi-interpenetrated polymer network (SIPN), where a covalently cross-linked poly(hydroxyethyl methacrylate), pHEMA, or poly(vinylalcohol), PVA, network is interpenetrated by linear chains of poly(vinylpyrrolidone), PVP. The chemical gels, simply loaded with water, were designed to safely remove surface dirt from water-sensitive artifacts. Modified SIPN can be designed to confine complex cleaning fluids, able to remove aged varnishes. These fluids are 4 or 5-components water-based nanostructured systems, where solvents are partially dispersed as nano-sized droplets in a continuous aqueous phase, with the help of surfactants. The mechanical behavior of the gel was optimized by varying both the cross-linking density and the polymer concentration. In this lecture, I will review the most meaningful achievements in this field, focusing on the application of micelles and o/w microemulsions confined into semi-interpenetrated hydrogels. These systems have been used on modern and contemporary artifacts as Picasso, Lichtenstein and Pollock. Finally, I will summarize the main perspectives that this field can disclose for Chemists and Conservators communities.

IL-07

POLYMER NETWORKS: SEVERAL APPLICATION CASES

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Formation of network assemblies is inherent to many types of polymer materials (including concentrated solutions, gels, blends, etc.), due to long-chain structure of macromolecules. Therefore, the study of fundamental features and properties of polymer networks has remained among key fields of macromolecular chemistry and physics over decades. Yet it is not often straightforward to use these fundamental ideas in the applied research.

In this contribution, several cases from recent studies in our group will be discussed, illustrating the bridge between fundamental and applied research in the field of polymer networks. The examples will demonstrate the application of polymer networks in the development of selective ion-conductive membranes used in redox-flow batteries, efficient gel-polymer electrolyte for metal-ion batteries, and manufacturing of several types of fibroin-based scaffolds for tissue engineering.

The presented research was partially supported by RFBR (project 16-33-60185_mol_a_dk).

REDOX-ACTIVE POLY(FERROCENYLSILANE)S AS ACTUATORS AND MEMORY HYDROGELS

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Stimuli-responsive polymer materials, capable of displaying large changes in response to minute external triggers, have attracted great interest as actuators for microfluidic control, drug delivery, artificial muscles, etc. Poly(ferrocenylsilane)s (PFSs) belong to a new class of redox-active materials which are composed of alternating ferrocene and silane units in the main chain and combine a high density of redox centers with excellent processability and redox characteristics.¹ Here we present a PFS-based hydrogel actuator, prepared by a one-step thiol-Michael addition reaction, which is capable of generating a mechanical motion. This relatively tough hydrogel displayed a reversible color change and expansion-contraction upon oxidation and reduction under physiological salt conditions, even under high stress close to its rupture strain, which makes it suitable for artificial muscles in biomimetic systems. By using a thiol-Michael addition cross-linking chemistry, reversible redox-switchable shrinking and swelling was accomplished by utilizing the PFS-PEG-based hydrogel using also thermal stimuli.

In addition, tributylphosphonium and alkanesulfonate anions were also introduced to the side group of PFS, which yielded a poly(ionic liquid) with redox-controlled LCST-type behavior. The LCST transition occurs in a wide temperature range depending on polymer concentration.² The aqueous solution also exhibits a redox-switchable LCST behavior. Hydrogels from these polymers were prepared by cross-linking, which exhibit non-ergodic memory behavior.

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2. Zhang, K., Feng, X., Ye, C., Hempenius, M.A., Vancso, G.J. Hydrogels with a Memory: Dual-Responsive, Organometallic Poly(ionic liquid)s with Hysteretic Volume-Phase Transition (2017) *Journal of the American Chemical Society*, 139 (29), pp. 10029-10035.

IL-09

PHOTOSWITCHING POLYMER NANOPARTICLES IMPART NOVEL BIOIMAGING METHODS

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Nanoparticles have evolved to be the carriers of multiple technologies, which garnished unprecedented power in molecular imaging. For example in bioimaging, a single fluorescent probe is usually lost in the ocean of myriad other molecules in a living cell, thus unambiguously measuring its signal becomes a daunting task because the signal is typically flooded with interferences and noises generated by the matrix. When photoswitchable nanoparticles are used, however, the oscillating signal can be selectively amplified in an unlimited manner and effectively measured after sufficient photoswitching cycles are completed. Here we show that photoswitchable nanoparticles containing photoactive spiropyran-merocyanine could be imaged not only in time domain (like most fluorescent probes), but also in frequency domain (unlike most fluorescent probes). Applying frequency-domain imaging (FDI) to biological applications, we have clearly detected, monitored, and tracked photoswitchable probes in living cells over the matrix of overwhelming non-switching fluorescence interferences. Therefore, FDI reveals molecular behavior that is not visible or measurable in real-time fluorescence imaging, thus a powerful tool in biomolecular imaging.

IL-10

MODELING THE MECHANICAL RESPONSE OF DOUBLE-NETWORK GELS UNDER CYCLIC DEFORMATION

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Mechanical behavior of double-network (DN) gels with covalent and non-covalent bonds depends strongly on time, strain rate and deformation program. A constitutive model is derived¹ for the viscoelastic and viscoplastic response of a polymer network with permanent and transient junctions under multi-cycle deformation. The viscoelastic response reflects breakage and reformation of temporary junctions in the transient network (transition of chains connected by physical bonds from their active to dangling state and vice versa). The viscoplastic behavior reflects slippage of junctions in the permanent network with respect to their reference positions. Motion of a junction starts when it becomes unbalanced due to transformation of one of the chains connected by this junction from its active state into the dangling state. The junction proceeds to slide with respect to the network (plastic flow) until it reaches a new equilibrium state. Approximation of experimental data in tensile tests with various strain rates, relaxation tests, loading-unloading tests, and multi-cycle tests with various deformation programs on a series of DN gels shows that the stress-strain diagrams are described correctly by the governing equations, material parameters evolve consistently with experimental conditions, and predictions of the model are in quantitative (where sufficient data are provided) and qualitative agreement with experimental data. In particular, numerical simulation demonstrates the ability of the model to describe the Mullins effect in DN gels.

Financial support by the Danish Innovation Fund (project 5152-00002B) is gratefully acknowledged.

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IL-11

DNA SUPRAMOLECULAR HYDROGELS

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The reversible responsiveness of DNA secondary structures to environmental stimuli has enabled to facilitate responsive devices and materials based on pure DNA or hybrid systems. Based on sequence and structure design, we have prepared kinds of pure or hybrid DNA supramolecular hydrogels, which could be formed under physiological condition within a minute at room temperature and without using any organic solvents. By tailoring the length of “sticky ends” of DNA linker, mechanical property of the hydrogel could be varied from hundreds to thousands Pa (G' , storage modulus); we also found that the viability of cell in a 4 mm diameter hydrogel is nearly 100% after 24 hours incubation from top in plastic tubes. These hydrogels possess extraordinary healing and fast-responding thixotropic properties, which make them injectable and writable. Because the formations of such hydrogels are based on DNA assembly, by DNA sequence design, they could be easily conferred excellent responsiveness including pH, DNA restriction enzymes, temperature etc., and enable easy removal after cell culture. In addition, we will show their application in 3D cell printing.

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IL-12

SELF-HEALING ALGINATE GELS THAT DO NOT FAIL ON STRETCHING TO 16000%

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Recently, there has been a concerted effort to develop hydrogels that are capable of sustaining large strain and self-heal upon rupture. Here, we report a super-stretchable hydrogel based on boronic acid-modified alginate that resists failure even at strains of 16000%. These gels also display outstanding self-healing behavior after fragmentation, recovering their original state within a minute of being the pieces held together. Small details in the formulation of these hydrogels such as the polymer/base ratio make big differences in their behavior. We suggest a model to rationalize how the dynamics of association/dissociation of boronate ester bonds in this optimal formulation allow restructuring of the gel during stretching so that failure is prevented. Remarkably, there are no qualitative differences between the equilibrium structure of super-stretchable gels and other crosslinked alginate hydrogels. In both cases, the gels are characterized by boronate ester crosslinks. Super-stretchability arises from a balance between the time scales of association/dissociation of boronate ester bonds and that of alginate chain orientation on stretching.

IL-13

MULTIFUNCTIONAL, RESPONSIVE HYDROGELS REACTION COMPARTMENTS AND IN MICROFLUIDIC APPLICATION

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Responsive polymeric materials are becoming a highly studied field not only in basic synthetic approaches and sophisticated characterization, but especially with regard to their use in technical applications, e.g. as sensors and actuators in microsystems and microfluidic applications. In addition, compartmentalization becomes an issue for being able to localize specific actions and reactions e.g. by preparing nano- and microcapsules equipped with stimuli-responsive shells and membranes.¹

Multiresponsive hydrogels are developed making use of an interpenetrating network approach² as well as the concept of graft copolymer networks³ for retaining responsivity for up to four different stimuli in one hydrogel. The hydrogels have been adapted for integration into microfluidic devices as volume-changing gels, adjusting degree of swelling as well as mechanical stability and allowing their use as effective sensors and actuators as needed for a chemical valve function.³ First examples of their use as chemical transistors and the development of logical modules for achieving basic logic gate functions through a microfluidic setup will be also presented.⁴ In addition, cascade enzyme reaction could be carried out in microreactors by integrating the enzymes in suitable hydrogel dots.⁵

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IL-14

POLYMERIC CAPSULES PRODUCED VIA MINIEMULSION OR AEROSOL CATIONIC PHOTOPOLYMERIZATION

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The synthesis of polymeric particles is of high interest for their applications going from drug delivery, paints, self-healing systems and reinforced materials. Emulsion and mini-emulsion polymerization is the most used polymerization technique for the polymeric particles production but cationic process is strongly inhibited by the presence of water, so in principle it would not be possible to carry a miniemulsion polymerization cationic reaction in aqueous media. In this study cationic miniemulsion photopolymerization of triethylene glycol divinyl ether (DVE3) in aqueous phase was demonstrated to be possible. by including the oil phase n-hexadecane which provides the interface with water. As an alternative route, a photo-induced cationic polymerization in aerosol was applied. A monomer solution was sprayed and irradiated with UV-light. During the reactor passage, both polymerization and phase separation occurred inside the single droplet. By adjusting the solvents amounts and ratios, different structures could be realized. Nano-structured particles with tunable pore shapes and dimensions were obtained. Capsule-like structures were obtained with the addition of a co-solvent able to get involved in the polymerization.

IL-15

RELATING MECHANICS TO CHAIN-LEVEL ARCHITECTURE IN GLASSY CROSSLINKED POLYMERS

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The relationship of chain-level network architecture and mechanical response are explored via graph-theoretic analysis of bead-spring molecular simulations. Model polymer networks were created from a modified Watts-Strogatz graph-generation scheme and the corresponding physical properties examined with molecular dynamics simulations. In particular, the degree of architectural disorder is systematically varied and linked with the linear and nonlinear uniaxial stress response, the nature of bond scission, and non-affine displacements of fully-cured glassy networks. We show that the chain architecture strongly affects the flow stress, the onset of bond breaking, and ultimate stress, but leaves the modulus and yield point unchanged. This behavior is attributed to internal restrictions imposed by architectural disorder, which alter crosslink dynamics in the flow regime and the degree of coordinated chain extension and failure near the ultimate stress. The mechanical properties are shown to be sensitive to even incremental changes in topology. Thus, the overall network architecture is predicted to be a meaningful parameter in glassy networks. Further, we find that network architectures assembled via a dynamic curing approach are an intermediate between lattice and random graphs as a result of the restrictions imposed by the finite contour length of the chains.¹



Figure 1. Snapshot of a disordered polymer network taken at a strain value near the ultimate stress of the structure.

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IL-16

FIXED VOLUME HYDROGELS FROM EMULSION TEMPLATED POLYACRYLATE NETWORKS

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Porous polymer monoliths (PPM) prepared using high internal phase emulsion precursors, can reach very high porosities, usually between 70% and 95%^{1,2} and have a unique interconnected cellular morphology where micron sized pores are formed due to the removal of the droplet phase of emulsion after the polymerisation of continuous phase while a second level of interconnecting pores are formed during and after the polymerisation. While water in oil emulsions are typically used for the synthesis of PPMs, termed polyHIPEs (High Internal Phase Emulsions, oil in water emulsions can also be used for the preparation of polyHIPEs from water soluble monomers, such as poly(acrylic acid) and poly(2-hydroxyethylmethacrylate).^{3,4} Such PMMs can be used as an interesting class of hydrogels where a large volume of aqueous media can be accommodated within the polymer network while the expansion of the network does not necessarily correspond to a large overall volume change. This is due to the large initial porosity of the PMM offering space for internal network expansion. The principle has been demonstrated in the case of polyHIPEs by Silverstein et al, using acrylamide and acrylic acid salts.^{5,6} On the other hand, we have used methacrylates, namely dimethylaminoethyl methacrylate and di(ethylene glycol) methyl ether methacrylate, to prepare PMMs of polyHIPE type and tested the water uptake at various pH values and temperatures. Up to 5 times polymer weight was absorbed without any visible volume change, at still high crosslinking degrees of PMM, namely 20% mol.

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ORAL COMMUNICATIONS

O-01

FRACTURE OF INTERPENETRATED NETWORKS: FROM BOND SCISSION TO MACROSCOPIC FRACTURE

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Soft but tough materials emerge in many new applications such as large amplitude actuators, flexible electronics or prostheses and the conditions of macroscopic failure are important. We investigate the initiation and propagation of a crack, and more precisely dissipative mechanisms at the crack tip with a multi-scale approach linking molecular bond scissions to macroscopic fracture of the sample. We prepared interpenetrated multiple network elastomers as model soft and tough materials to introduce a controlled fraction of sacrificial bonds that can break before the material fails macroscopically. We introduce in this sacrificial network as a crosslinker a mechanophore, developed by Göstl *et al.*,¹ based on a 9π -extended anthracene Diels Alder adduct, that releases anthracene as the molecule breaks due to an applied force. The fluorescence; which is intense and resistant to photobleaching, can then be detected by confocal microscopy as the crack opens and propagates, as shown in the example 3D-image above. These microscopic observations provide a convenient tool to map where and when chemical bonds break during a mechanical test.

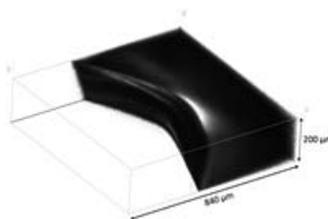


Figure 1. 3D reconstruction from confocal observation of a crack in a multiple network. Light color represents bond scission

Acknowledgement: We are grateful to R. Göstl and R. Sijbesma for their kind help with the mechanophore synthesis.

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INSPIRED BY NATURE: REINFORCING COILED COIL HYDROGEL BUILDING BLOCKS WITH HISTIDINE-METAL COORDINATION

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In nature, α -helical coiled coil (CC) protein-folding motifs often occur in proteins with mechanical function, such as myosin or α -keratin. In the field of bioinspired materials, naturally occurring and synthetic CCs are emerging as highly versatile material building blocks for biomimetic hydrogels. Despite their prevalence in natural and bioinspired materials, the relationship between the molecular mechanical properties of CCs and the bulk viscoelastic behavior of CC-crosslinked hydrogels remains poorly understood. Yet, this information is critical for controlling and tuning the dynamic mechanical performance of CC-based materials in the linear and non-linear range.

Toward the goal of generating tunable CC-crosslinked hydrogels, we equipped a heterodimeric CC with histidine-metal coordination bonds, which are strong, non-covalent interactions with self-healing properties. Specifically, two histidine residues were introduced into terminal helical turns in a geometry that favors metal coordination. Using circular dichroism spectroscopy and atomic force microscopy-based single molecule force spectroscopy, we demonstrate that histidine-metal coordination increases the overall stability of the CC thermodynamically and mechanically. Serving as crosslink between branched poly(ethylene glycol) molecules, this CC is now being used for the development of tunable, self-healing biomimetic hydrogels. Oscillatory shear rheology reveals that the dynamic mechanical properties of these CC building blocks can be transferred to the hydrogel, which can reversibly be tuned using transition metal ions and chelators. These results provide a novel starting point for engineering hydrogels with precisely tunable linear and non-linear viscoelastic behavior.

IONIC LIQUID-BASED GELS WITH SPECIFIC INORGANIC/ORGANIC DOUBLE NETWORK

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Ion gels are promising soft materials that contain a large amount of ionic liquids (ILs) in a polymer network. Ion gels have recently attracted considerable interest for their applications in electrochemical devices, actuators, and gas separation membranes due to the unique ILs-based properties, such as their non-volatility; non-flammability; and thermal, chemical, and electrochemical stabilities. In practical applications, the high mechanical strength of ion gels is an indispensable property of this soft material. Development of tough ion gels with a large amount of ILs is a critical challenge for overcoming the limitations in practical applications. In this research, we developed robust ion gels having interpenetrating inorganic/organic network, named double network (DN) ion gel.¹ The DN ion gels were prepared by first silica particle network formation via sol-gel reaction and following organic polydimethylacrylamide (PDMAAm) network formation via free radical polymerization. Because the brittle inorganic silica particle network was formed by physically inter-particle interaction, the inorganic network was easily ruptured and dissipated the loaded energy as sacrificial bonds, resulting high toughness. In addition, owing to the physical bonding between silica nanoparticles, the ruptured silica nanoparticle network, could be significantly recovered by annealing.

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pH RESPONSIVENESS OF HYDROGELS FORMED BY TELECHELIC POLYAMPHOLYTES

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We present the pH dependent properties of hydrogels formed by hydrophobic association of the sticky ends of the triblock terpolymer poly(methyl methacrylate)-*b*-poly(2-(diethylamino)ethyl methacrylate-*co*-methacrylic acid)-*b*-poly(methyl methacrylate) (PMMA-*b*-P(DEA-*co*-MAA)-*b*-PMMA).¹ The middle block is a weak polyampholyte having a pH dependent charge density and sign, which enables tuning of the rheological and structural properties by pH variation. Small-angle neutron scattering (SANS) studies of hydrogels reveal a network with charged fuzzy PMMA spheres interacting via a screened Coulomb potential at low pH values. With increasing pH, the gel disintegrates. The hydrophobic aggregation of micelles is observed near the isoelectric point. Rotational dynamic light scattering reveals a frozen gel at low pH values, whereas at high pH, the diffusion of single micelles is observed in addition.

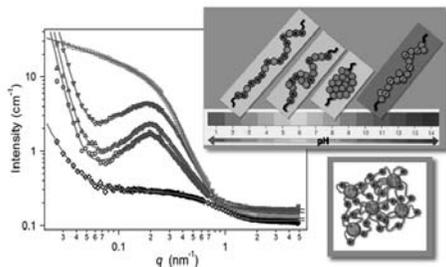


Figure 1. SANS data from a 3 wt% solution in D₂O at different pH values. Inset: Schematics of the pH induced charge variation of DEA (red) and MAA (green) along the middle block. Grey: hydrophobic moieties.

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O-05

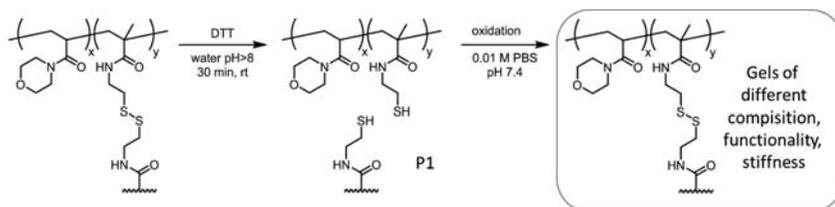
A MODULAR HYDROGEL PLATFORM WITH TUNABLE MECHANICAL AND BIOLOGICAL PROPERTIES

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Here, we present a novel approach to fabricate well-defined hydrogels as potential substrates for cell culture application. The approach is based on three steps: Firstly, disulfide containing hydrogels of different composition were prepared by *in-situ* copolymerization. Afterwards the disulfide crosslinker of the gels were cleaved with dithiothreitol (DTT) to form soluble copolymers that were then analyzed by conventional analytical solution methods.



Scheme. In situ hydrogel formation, cleavage with DTT and reformation with oxidation agents.

Finally, gel fabrication of different stiffness and functionality was carried out by mixing one or more soluble copolymers at pre-defined concentration in the presence of an oxidation agent for 2D substrates. A small library of 25 gels with distinct copolymer compositions were fabricated and cell compatibility was tested with HEK293T cells.

1. P. Bolduan, F. Pätzold, C. Carrillo Garcia, D. Schade, R. Weberskirch, *submitted for publication*.

HYDROGEL POSSESSING THERMO REVERSIBLE ROBUSTIZING

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Phase separation, which is driven by various stimuli, such as temperature, pressure, pH, solvent affinity, salt, and so on, is one of unique phenomena for hydrogels to drastically change their physical features. As poly (isopropylacrylamide) (PNIPAm) gel, the volume phase separation of gel has been paid attention so much, while formation of hydrophobic interaction must contribute thermal robustizing of gel matrix.

Here, we develop the novel thermoresponsive hydrogel, which is easily prepared from conventional poly (acrylic acid) (PAAc) and calcium acetate (CaAc), shows isochoric phase separation, while it is capable thermal super jumping of stiffness and toughness without any volume change. Material preparation is quite simple that the polymerized PAAc gel was just soaked in CaAc solution by equilibrium. Figure 1 shows appearance of PAAc-CaAc gel before and after phase separation. The transparent gel got opaque white and stiff so much at low critical solution temperature (LCST). This phenomenon is completely reversible. The LCST can be controlled from around 20 to 90, by fraction of polymer and salt. The mechanical characteristics were evaluated from uniaxial tensile test. The Young's modulus and work of extension at 70 °C were around 430 times and 60 times higher than those of 20 °C. This super robustizing would be achieved from not only hydrophobic interaction but also reinforced electrostatic interaction. At high temperature, PAAc side chain would form PAAc-Ca-OOCCH₃ complex that is totally hydrophobic moiety. Therefore, the sidechain is encapsulated in hydrophobic dense region, and electrostatic interaction became large so much due to low dielectric constant environment.

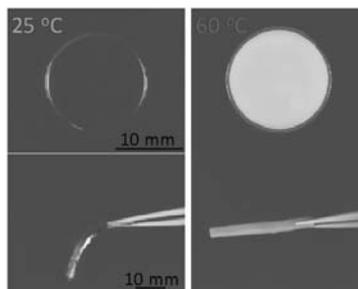


Figure 1. Appearance of PAAc-CaAc hydrogel before and after LCST.

O-07

TOUGH AND MULTI-RESPONSIVE POLYMER HYDROGELS CROSSLINKED BY BLOCK COPOLYMER MICELLES

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The lecture presents on novel tough polymer hydrogels crosslinked by triblock copolymer micelles. Amphiphilic block copolymer PEO65-PPO99-PEO65 (Pluronic F127) functionalized with double bonds on both ends form reactive micelles in aqueous solution. The multi-functional micelles are copolymerized with hydrophilic soluble monomers to form three-dimension networks that contain a large amount water, or polymer hydrogels. With covalent bonding with the micelles, the hydrogel show outstanding strength, ultra high stretchability, and fatigue resistance.¹ Small angle X-ray scattering studies reveal the oriented deformation of micelles in the network matrix along the stretching direction, exhibiting ellipsoidal scattering patterns.² This behavior suggests an energy dissipation or toughening mechanism through the deformation and chain pull-out of the micelles. As ionic or ionizable monomers are used for the synthesis, hydrogels that are responsive to pH, temperature, ionic strength,³ and electric field⁴ are obtained. Interesting results will be showed to demonstrate great potentials of such micelle-crosslinked hydrogels for smart stimulus-responsive devices.

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PROGRAMMABLE VOLUME PHASE TRANSITION OF THE HYDROGELS FOR SMART SOFT MATTER DEVICES

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Poly(*N*-isopropylacrylamide) hydrogel undergoes a reversible volume phase transition from a swollen hydrated state to a shrunken dehydrated state at the critical temperature. To optimize the temperature responsiveness on demand, we have tuned the thermal behavior of the PNIPAM based hydrogels by modifying the chemical structure of them. Shifting of transition temperature,¹⁻² stepwise transition over wide temperature range³, and large thermal hysteresis⁴ for the crosslinked hydrogels could be achieved by copolymerization with hydrophobic¹ or hydrophilic² comonomers, preparation of multi-components composition,³ and grafting of PNIPAM onto the methylcellulose,⁴ respectively.

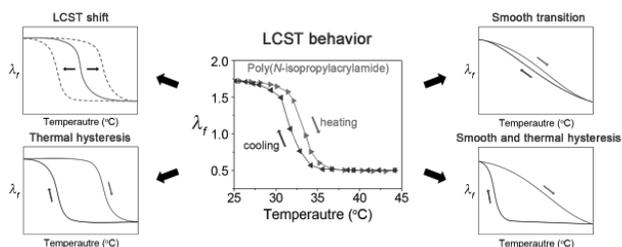


Figure 1. Various volume phase transition behavior of the hydrogels

These achievements enable to develop the light responsive actuators of various motion,^{1,4} efficient solar control windows³, and transdermal hydrogel patches.²

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ANTIMICROBIAL COMPOSITE WOUND DRESSING

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Nowadays, a wound dressing is no longer a passive material, it must interact with the wound and improve the healing process. In fact, the actual requirements for a wound dressing are quite challenging. To achieve these demanding goal, wound dressing' research have been focus on the development of composites that combine the best of two or more polymeric materials. Thus, our group developed a composite material of cotton functionalized with hydroxypropyl methylcellulose / cyclodextrins hydrogel to be used as antimicrobial wound dressing. Hydrogel polymer will improve the wearability and the drug delivery capacity of cotton textiles. And cotton will enhance the mechanical properties of hydrogel, facilitating the handle process.

The composite synthesis was performed by one-step chemical crosslinking. The reaction parameters, such as crosslinker concentration and polymeric solution concentration, were optimized. The obtained composites were characterized base on their physicochemical and biological properties.

To the best of our knowledge, loading and release of gallic acid (as antibacterial agent) into composites wound dressings, and its release for control wound infections, have not been evaluated until now. The developed composites have the combined properties of cotton and hydrogel. The gallic acid was successfully loaded into the polymeric network, and it release was sustained for 48 h. The loaded composites can destroy bacterial cells preserving the gallic acid antibacterial activity. Thus, the developed composites showed suitable properties for the incorporation of gallic acid and utilization as antibacterial wound dressing.

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PREDICTIONS AND LIMITATIONS OF A PURELY TOPOLOGICAL METHOD FOR CALCULATING POLYMER NETWORK CONNECTIVITY

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Understanding the impact of network topology on the properties of polymer hydrogels and elastomers is one of the longest standing challenges in the science of polymer networks. Based on the early work of Stepto, we have implemented kinetic graph theories and kinetic Monte Carlo theories to model the formation of loops within polymer networks. These methods are able to match measurements performed by the Johnson group with no adjustable parameters for both primary loops and secondary loops for PEG gels produced using polycondensation chemistry. In addition, they can match much of the classical data on gel point suppression for polycondensation polymer gels and provide a basis for new theories to account for network topology in calculating the linear elastic modulus of polymer networks.

Due to the effectiveness of these approaches in matching key experimental results, it is also useful to examine their predictions of critical behavior in the vicinity of the gelation transition. Using very large kinetic Monte Carlo (KMC) simulations, estimates of critical exponents can be produced. For low loop densities, the KMC simulations predict exponents in good agreement with the Flory-Stockmayer predictions. However, as loop density increases above a threshold, the exponents continuously change value, suggesting a fundamental change in the nature of the underlying network at high loop densities. The use of these theories also provides an opportunity for critical examination of their approximations, most notably the family of approximations stemming from neglect of spatial positions of the reactive molecules. Despite effectiveness in matching several experimental data sets, other predictions of the theories clearly show limitations of this approach that must be addressed with further advances.

O-11

HOW TO PREPARE GEL FIBERS

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Gel fiber based membranes have a unique structure mimicking the structure of native soft tissues. As such, they can be used as materials for cell cultivation and in tissue regeneration. However, there is a constant need for new methods for the preparations of such materials, since there are no general strategies for their synthesis. Electrospinning technique enables the preparation of a wide variety of matrices built of fibers in the nano or micrometer diameter range. However, the use of water soluble polymers in electrospinning is inappropriate because of their fast dissolution in biological fluids. By utilizing reactive electrospinning, chemically cross-linked gel fibers can be created, which, without enzymatic or chemical degradation, will not dissolve in physiological conditions. In the present work two techniques are presented for the preparation of gel fibers based on electrospinning and the anhydrous form of poly(aspartic acid) (Figure 1.). One is based on thiol disulfide chemistry,¹ whereas the other on coaxial electrospinning.²

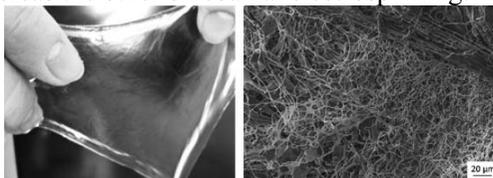


Figure 1. Macroscopic and microscopic picture taken of an electrospun poly(aspartic acid) based gel fiber matrix

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O-12

NON-LINEAR RHEOLOGICAL PROPERTIES OF RIGID POLYMER NETWORK MADE OF A GIANT POLYSACCHARIDE

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Rigid filaments of proteins play important roles in cell mechanics, and networks of such rigid filaments exhibit commonly strain-hardening. Here we used a polysaccharide “sacran” as a model rigid polymer and studied the non-linear rheological properties of its chemical gels. Extracted from a freshwater unicellular cyanobacterium, *Aphanothece sacrum*, this giant polysaccharide ($M_w > 10^7$ g/mol), show liquid crystalline properties even in dilute aqueous solutions.¹ The microrheologically measured intrinsic persistence length of the sacran chain (~ 60 nm) are comparable to or shorter than the mesh size of sacran networks. Chemical gels of sacran were prepared by cross-linking sacran chains with divinylsulfone in basic conditions. A significant strain hardening is observed for the chemical gels at different cross-linking ratios. The strain hardening behavior is well explained with an elasticity model which describes the stress – strain relationship of polymeric networks as a function of the chain rigidity and extensibility (Figure).²

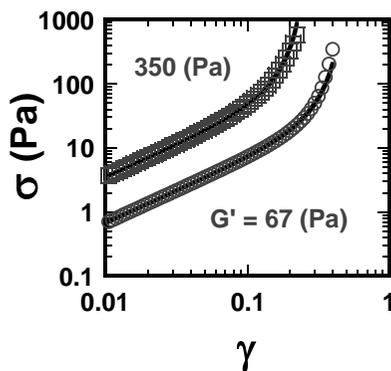


Figure 1. Shear stress as a function of strain fitted with the model.

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FAST pH-RESPONSIVE SUPRAMOLECULAR HYDROGEL FROM PEPTIDE-POLYMER HYBRID

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Block copolymers are potentially fascinating soft materials in both industrial and biomedical fields. Especially, a hybridization of structurally and functionally controlled peptides and conventional synthetic polymers is a very attractive strategy for constructing new classes of smart polymeric materials with tailored characters and structures. We have reported the synthesis of various peptide-polymer hybrids with diblock, triblock and multiblock architectures, and their structural and functional properties.¹⁻⁴

Here, we report a novel stimuli-responsive supramolecular hydrogel fabricated from a self-assembling peptide-poly(ethylene glycol) (PEG) star-shaped hybrid block copolymer. Four β -sheet-forming oligopeptides with alternating hydrophobic Leu/ionizable Glu repeat were conjugated to flexible 4-armed PEG by thiol-maleimide reaction. A detailed analysis of conformation and self-assembly behavior of the hybrid in water was conducted by circular dichroism and rheological measurements. As a result, the star-shaped peptide-PEG hybrid was found to self-assemble into β -sheet networks and form hydrogels with various rheological properties under certain pH conditions. Interestingly, this hydrogel showed rapid and reversible gel-to-sol transition based on pH-induced conformational change from β -sheet to random coil structure. In addition, the peptide-PEG hydrogel was found to possess self-healing and injectable capabilities due to reversible self-assembly of the peptide blocks, demonstrating the potential of this hybrid as a smart biomaterial.

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HIGH-STRENGTH GELS COMPOSED OF AN IONIC LIQUID AND INORGANIC/ORGANIC HYBRID NETWORKS FORMED VIA ONE-POT/ONE-STEP PROCESS

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Two types of high-strength ion gels with a large amount of ionic liquid and inorganic/organic hybrid network were prepared by one-pot/one-step preparation process.¹ The inorganic component of the inorganic/organic hybrid network is silica nanoparticles formed by condensation polymerization of tetraethyl orthosilicate (TEOS). The silica nanoparticles formed in an ionic liquid were connected with each other by physical interactions to form network structure. On the other hand, the organic network is a polydimethylacrylamide network formed by thermal initiated free radical polymerization of dimethylacrylamide (DMAAm). The specific inorganic/organic hybrid networks were formed by just heating the mixture of an ionic liquid, TEOS, solvolytic agent for polycondensation reaction of TEOS, DMAAm, cross-linker and thermal radical initiator. Both of the prepared ion gels have very high mechanical strength, but their mechanical properties are completely different. One is a double-network gel showing high strength based on energy dissipation mechanism via fracture of inorganic silica particle network. The other is an inorganic/organic composite ion gel showing elastic deformation. These high strength ion gels can be selectively prepared by controlling the preparation temperature to control the formation rate of the inorganic and the organic networks.

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**MICROGEL-POLYELECTROLYTE COMPLEXES: TOWARD
STIMULI-SENSITIVE CONTAINERS FOR CAPACIOUS UPTAKE
AND TRIGGERED RELEASE OF MULTI-FUNCTIONAL
PAYLOADS**

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We highlight the use of microgels as containers of high capacity for uptake and triggered release of multi-functional guests. As a model guest, heteroarm star-shaped copolymers (miktoarm stars) are chosen, as their certain arms could carry different active moieties, while other arms could act as “stickers” to the microgel host. Electrostatic attraction is used as a binding principle, wherein positively charged bis-hydrophilic miktoarm stars interact with negatively charged microgels. Functioning of microgels as containers with high capacity offers the possibility to construct promising stimuli-responsive uptake and release systems. Hence, aqueous mixtures of these oppositely charged polymeric components, the microgels and the miktoarm stars, were investigated in dependence on their initial charge ratio ($icr = [\text{cationic charges}]/[\text{anionic charges}]$) and on the concentration of a low molecular weight salt. The miktoarm stars are able to penetrate into the microgels to compensate their negatively charged groups. The amount of the miktoarm stars bound to the microgels can be controlled *via* ionic strength of the system, with limiting cases from quantitative binding to no binding. Then, such a microgel- polyelectrolyte complex consists of one microgel and more than 1000 miktoarm stars. Furthermore, a jump-wise increase of ionic strength in solutions of the complexes triggers the complete release of the miktoarm stars from the microgel, and the system stays always colloidally stable. Thus, microgel-polyelectrolyte complexes provide opportunities for many important applications, especially in targeted/controlled delivery.

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HYDRO-GEL-METALLURGY

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Many industrial wastewaters such as electroplating factory effluents contain heavy metals ions. Controlled chemical precipitation is widely used in hydrometallurgy to recover the useful metal ions from the wastewater selectively by the reaction with the sodium hydroxide or sodium carbonate to produce insoluble metal salts. However, the separation of the produced salts from the solution requires multiple huge sedimentation tanks. We reported a simple recovery method of metal ions using a hydrogel.¹ The hydrogel with tertiary amino group produced hydroxide ions by protonation of their amino group and retained them by ionic interaction. When the hydrogel was immersed in metal ion aqueous solution, the metal ion diffused into the hydrogel and produce metal hydroxide in the hydrogel. The hydrogel within the metal hydroxide was separated from the solution easily. In this study, a hydrogel retaining the anions other than hydroxide ion was prepared and the recovery of cation by the hydrogel were investigated.

The hydrogel was prepared from (3-Acrylamidopropyl)trimethylammonium chloride by the radical polymerization with cross-linker. The gel was immersed in sodium sulfonate or sodium carbonate solution to exchange the chloride ion. After the exchange, the hydrogel was immersed in calcium chloride solutions. Figure 1 shows the effect of the anions on the amount of calcium ion in the hydrogel. The amount Ca^{2+} reacted with carbonate ion was seven times higher than that of the sulfate ion because the solubility product of CaCO_3 was smaller than that of CaSO_4 . The recovery amount was changed by the kind and/or concentration of anion retained in the hydrogel at preparation. It suggests that the hydrogel can selectively recover cations from the solution as well as the controlled chemical precipitation.

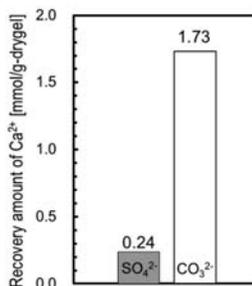


Figure 1. Effect of anion in hydrogel on removal amount

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O-17

„TEXTILE MINING“ - RECOVERY OF NOBLE METALS FROM INDUSTRIAL PROCESS WATERS BY THE USE OF TEXTILE-FIXED POLYELECTROLYTES

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The recycling and recovery of high-priced noble metals such as platinum, gold, palladium and silver or rare and strategic metals like indium, gallium, and rare earth metals from scrap metals and wastewaters will be from steadily increasing importance within the next years. Therefore, the focus has to be set on the detection of potentially usable secondary resources and the development of inexpensive and energy-saving processes to separate and recover the metals selectively (urban mining). Beside electronic scrap industrial process and wastewaters represent a considerable source for noble metals. Recently, we have successfully developed an innovative metal-adsorbing textile filter material based on various polyelectrolytes. The surface modification of the fibrous material is easy to realize with common methods in textile finishing yielding a durable, high-performing and even cheap fiber-fixed polymer network. We present results on the general textile finishing procedure and the pH-depending adsorption of noble metal ions. The feasibility of the overall process is demonstrated on palladium containing process waters obtained from a German producer of circuit boards. Moreover, the same innovative adsorber material is useful for the decontamination of chromate-polluted ground waters and soils. Our latest investigations focus their use in the selective recovery of rare earth metals from large-scale FCC catalyst production for the petroleum refining industry.

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O-18

SYNTHESIS OF MICRO-HYDROGEL PARTICLES CONSISTING OF HYPERBRANCHED POLYAMIDOAMINE FOR THE CAPTURING OF HEAVY METAL IONS AND CO₂

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Polyamidoamine (PAMAM) dendrimers and hyperbranched polymers have attracted considerable interest in recent years. The highly branched structures together with a large number of functional groups in the main chains and end groups make these materials ideal for applications in host-guest encapsulation, nanoreactors, and delivery devices. Amine and amide functional groups of PAMAM dendrimers can serve as nanoscale high-capacity containers for heavy metal ions such as Cu²⁺, Cd²⁺, and others. However, drawbacks of PAMAM dendrimers and hyperbranched polyamidoamines (HPAMAM) include their small size, the hydrodynamic diameter of the dendrimers, and the radius of gyration of the hyperbranched polymers, which is typically less than 10 nanometers. Thus, additional processes such as ultrafiltration are necessary for separation from an aqueous solution. In this study,¹ we used the A₂+B₄ polycondensation method for hyperbranched polymers to make micro-sized HPAMAM hydrogel particles through the dispersal of an aqueous HPAMAM solution obtained by A₂+B₄ polycondensation into an immiscible liquid and then by carrying out the polycondensation until reaching the point of critical gelation. This simple method allowed us to make micro-sized hydrogel particles which wholly consisted of hyperbranched polyamidoamine without any additional crosslinking reagents. The crosslinked HPAMAM hydrogel particles were found to be highly effective when used to remove heavy metal ions from an aqueous solution and could easily be separated from water by a simple filtration process.

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VARIOUS POLYELECTROLYTE HYDROGEL ARCHITECTURES – SYNTHESIS, CHARACTERIZATION AND THEIR APPLICATION FOR SALT WATER DESALINATION

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We are investigating a novel approach for water desalination using charged polymer hydrogels based on poly(sodium acrylate) under externally applied pressure.¹ The desalination mechanism is based on the unequal distribution of an added salt between gel and surrounding solution phase as schematically displayed in Figure 1B.^{2,3}

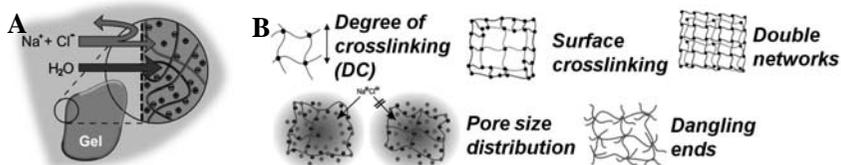


Figure 1. A) Salt rejection of mobile ions by the fixed charges from the polyelectrolyte hydrogels. B) Various network structures that are synthesized and characterized in this work.

The aim of this particular work is to study the influence of the network topology on the desalination performance (see Figure 1B). We synthesized different model systems for the polyelectrolytes to investigate the effect of the network structure on several hydrogel properties such as mechanical strength, water uptake, polymer mobility, salt repulsion and desalination efficiency.

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POLYDIMETHYLSILOXANE HYBRID POLYMERS AS SORBENT FOR THE REMOVAL OF OIL/ORGANIC SOLVENT FROM THE ENVIRONMENT

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Oil is one of the main energy resources. There is a risk of oil spillage during its production, transportation and storage.^{1,2} Oil and oil derived organic liquids need to be cleaned effectively in case of a sudden spillage. In recent years, the usage of absorbents for the removal of oil from water appears as an attractive method. A good sorbent material should have some properties such as fast oil sorption rate, high oil sorption capacity, hydrophobicity, low water pickup, good reusability, high buoyancy, and excellent physical and chemical resistances.³

In this work, poly(dimethylsiloxane) (PDMS) hybrid polymers were synthesized by the condensation of different molecular weights of PDMS macromonomers and a crosslinker as a sorbent for the cleaning of oil. The characterization of PDMS sorbents was determined by Fourier transform infrared spectroscopy, solid-state ¹³C and ²⁹Si cross-polarization magic angle spinning nuclear magnetic resonance and thermal gravimetric analysis. The swelling features of the synthesized sorbents in several organic solvents (dichloromethane, tetrahydrofuran, benzene, toluene) and oil derived fuels were evaluated by the swelling test. The reusability and the water-oil separation efficiency of PDMS sorbents were also investigated.

***Acknowledgment:** We thank the Scientific and Technological Research Council of Turkey - TUBITAK for the support of this work through Grant KBAG/ 116Z365.*

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STIMULI-RESPONSIVE THIN FILMS PREPARED FROM PNIPAM/PAAC BASED MICROGELS

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Synthetic hydrogels are promising biomaterials for tissue engineering as they are highly versatile. Some of their characteristics can be suitably tuned, for instance elasticity, topography and biochemical cues. They can thus represent a bio-realistic environment necessary for cellular growth *in vitro*.¹ Furthermore, smart microgels have been recently employed as switchable platforms to mimic tissue stiffness during ageing.² Among them, poly(isopropylacrylamide) (PNIPAM) is largely used due to the fact that its elastic properties are similar to the extracellular matrix of the cells. However PNIPAM may result too soft and the addition of a polyelectrolyte such as poly(acrylic acid) (PAAc) can help to increase the stiffness. On the other hand, PAAc also adds fixed and mobile charges to the polymeric network thus rendering the material electroactive and pH-responsive.³ In this work we have developed a simple and versatile technique to form a well-stabilized stimuli-responsive thin film by using click-chemistry. In particular, ene-functionalized PNIPAM/PAAc microgels with different chemical compositions have been synthesized⁴ and characterized in size and chemical composition. These particles in water dispersion have been then used to prepare spin-coated films on functionalized glass substrates. The film biocompatibility has been finally investigated after stabilization via thiol-ene promoted crosslinking.

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A HYDROGEL DEVICE FOR OIL/WATER SEPARATION

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Massive oil spill incidents cause catastrophic environmental damage and serious energy and economic losses. Only through efficient and fast retrieval of the spilled oil can there be a hope to decrease the environmental damage, while energy and economic losses can be minimized if cost savings during the retrieval process and reuse of the retrieved oil are possible. We devised a hydrogel bowl with macroscopic openings in its sidewall and a surface with hydrophobic, oleophilic, and high oil wettability properties based on the mechanical principles describing the interaction between water and the openings in the bowl. The macroscopic openings of the bowl prevent water from flowing into the bowl, help any water in the bowl flow out easily, and can collect a large amount of spilled oil of various viscosities at a high rate. Since the long-term usability, storage, and scalability of the hydrogel bowl have all been verified, it can be a highly effective solution for the environmental and economic problems caused by oil spill accidents.

THE SWELLING OF PNIPAM MICROGELS: EFFECT OF CHARGE AND SOLVENT QUALITY

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We report dynamic light scattering data for the hydrodynamic radius of neutral and a weakly charged PNIPAM based microgels as a function of temperature and added salt concentration.

Incorporation of 2% ionic co-monomers leads to a significant increase in microgel swelling, which can be partially suppressed by addition of salt. Scaling theories predict a power-law variation of the microgel size with added salt $R_H \approx c_S^{-0.2}$, which is stronger than the experimentally observed exponents of $R_H \approx c_S^{-(0.05-0.1)}$. The difference between theory and experiment may be related to the neglect of intrinsic excluded volume in the former.

The charge induced swelling data are interpreted within the framework of the Flory-Rehner model, modified to include a Donnan term to account for the additional osmotic pressure arising from entropy of dissociated counterions. This model strongly under-predicts microgel swelling, particularly at low salt concentrations and low temperatures (good solvent). Repulsion between charged polymer segments appears to play a leading role in the swelling of ionic microgels. Mean field theories for polyelectrolyte gels do not quantitatively capture this feature.

**PHASE TRANSITION IN HYDROGELS
OF THERMORESPONSIVE INTERPENETRATING POLYMER
NETWORKS**

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Collapse transition in hydrogels of acrylamide-based semi- and fully interpenetrating polymer networks (IPNs) was studied by combination of swelling and mechanical experiments, NMR spectroscopy, small-angle neutron scattering (SANS) and DSC. The effect of various network composition and reverse sequence in the preparation procedure of the network on the volume phase transition was examined. Transition temperature linearly increased with the content of hydrophilic polyacrylamide component in IPNs and the fraction of collapsed polymer units detected by NMR as well as the enthalpy change in DSC measurements were reduced. SANS results showed that compact 3-dimensional multi-chain particles with gyration radius ~ 20 nm are formed in collapsed IPN hydrogels.

A certain portion of spatially restricted bound water (HDO) was established for studied IPNs at temperature above the volume phase transition from measurements of ^1H NMR spectra, spin-spin relaxation times T_2 and diffusion coefficients D of water molecules. Slow exchange regime between bound and free water was revealed. The content of bound water correlates with swelling experiments and lower values of the shear mechanical modulus and shows the decisive role of bound water in this respect. The fraction of bound water slowly decreases with time.

RESPONSIVE TOUGHENING IN PHASE-SEPARATED GELS

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By using concepts of polymer physics, we developed some strategies to design tough “hybrid” gels that combine covalent (permanent) cross-links and physical (reversible) sacrificial interactions. Thus, polymer adsorption onto silica nanoparticles¹⁻³ can be a remarkably simple and efficient means for gel’s mechanical toughening either in bulk or for gel adhesion. More recently, a novel mode of fracture toughening by crack bifurcation has been highlighted in phase-separated hydrogels, by exploring the coil-to-globule transition.⁴⁻⁵ We designed original gel topologies that phase-separate at constant macroscopic volume and quite high level of hydration, independently of the phase-separation process. The polymer network combines a conventional network with thermo-responsive domains which act as reinforcing fibers operating at a targeted temperature. Network topology is crucial to enhance efficiently the fracture resistance of the gels.

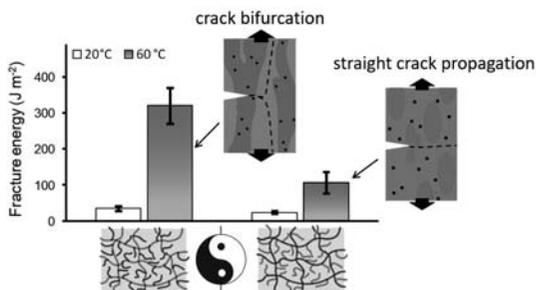


Figure 1. Impact of gel topology on fracture properties, from ref. 4.

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REDOX RESPONSIVE HYDROGELS FOR DRUG DELIVERY APPLICATIONS

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Over the years, the interest of Stimuli Responsive Polymers (SRPs) grown exponentially.¹ In the biomedical fields, gels and especially hydrogels have attracted attention due to the simplicity and the reduced cost of the gels synthesis compared to polymeric micelles. The drug release from Stimuli Responsive Hydrogels (SRHs) was largely realised with pH, temperature and light stimuli, however only a small number of redox responsive hydrogels were developed.²

In this project, we desire to develop a SRH that can release its guest molecules upon the trigger of an electrical stimulus for biomedical applications. To obtain this redox responsive hydrogel, we have introduced into the hydrogel the stable nitroxide radical 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO) that can be reversibly oxidized into an oxoammonium cation. A precursor of the hydrogel was firstly synthesized by the free radical polymerization of 2,2,6,6-tetramethylpiperidin-4-yl methacrylate (TMPM). In the second step, the TMPM units were oxidised into TEMPO-methacrylate (TMA) to obtain the desired redox responsive.

The redox activity of the accordingly obtained hydrogels was evidenced by cyclic voltammetry, confirm the presence of TMA in these hydrogels. Also rheology and swelling tests show that those gels exhibit promising characteristics. To conclude, this data show a promising hydrogel that can be tested to be used in drug delivery applications in our future work.

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PREDICTING THE ONSET OF BUCKLING DURING TRANSIENT SWELLING OF HYDROGELS USING FEM

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The phenomenon of swelling induced buckling in hydrogels is an intriguing scientific problem that has generated considerable attention over the last decades. However, most of the analytical and numerical studies in the literature investigate buckling caused by homogeneous swelling. In this work, we aim at quantifying the effect that the transient nature of swelling has on the point of onset of buckling for confined hydrogel plates with gradient stiffness.

A user defined material model (UMAT) for the commercial finite element software Abaqus describing transient hydrogel swelling was implemented based on the work by Hong et al.,¹ Kang and Huang,² and Toh et al.³ It is demonstrated that the implemented model predicts the time dependency in swelling hydrogel spheres at various overall sizes as reported from experimental studies.⁴ A novel procedure for quantifying the point of onset of buckling in finite element models of hydrogel swelling is outlined.

Using the proposed framework, the effect of the diffusion parameter for the onset of buckling was studied. A pronounced influence on the extent of swelling prior to the onset of buckling was found, with an increased extent of swelling at the point of buckling for higher values of the diffusion parameter. Accounting for this effect can be of great importance when aiming for finite element modelling procedures that can predict the behavior of buckling hydrogels in a quantitative manner.

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DOUBLE NETWORK CRYOGELST. Sedlacik^a, T. Nonoyama^a, J. P. Gong^{a,b}

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Porous hydrogels are highly desired materials in many applications of biotechnology and medicine. However, preparing hydrogels with an open-cell structure and a suitable mechanical strength can be challenging. One of the promising methods is based on gelation in a frozen state, resulting in sponge-like hydrogels with an interesting mechanical behavior, despite of high pore volume and water content.¹ These materials, cryogels, can be compressed to high level without any structural damage, while water present in pores drains out. After deformation, cryogels quickly retain their original shape as the water is drawn back into the porous structure due to capillary forces. Despite these unique properties among hydrogels, cryogels are relatively soft. In recent years, cryogels of interpenetrating polymer networks (IPN) have been prepared to tune their biological and/or physical properties.² Double network (DN) gels, as a subcategory of IPN, combine a brittle and a ductile network and have been shown to be remarkable stronger and tougher when compared with corresponding single network gels.³ The 1st network serves sacrificial bonds dissipating a mechanical energy, while the 2nd network effectively distributes the mechanical energy and avoids a crack propagation by allowing the brittle network to break only into small clusters. In this work, we have focused on toughening cryogels based on sodium 2-acrylamido-2-methylpropane sulfonate (NaAMPS) by introduction of the 2nd network based on acrylamide. We have been introducing a preparation method based on UV irradiation of frozen NaAMPS cryogels, first swollen in monomer solution of 2nd network, using a photoinitiator. We have described relations between mechanical properties and composition of DN cryogels.

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**MECHANICS AND FRACTURE OF TOUGH HYDROGELS
BELOW WATER-FREEZING TEMPERATURES**

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Hydrogels consists of hydrophilic polymer networks dispersed in water. Many applications of hydrogels rely on their unique combination of solid-like mechanical behavior and water-like transport properties. However, if the temperature is decreased below 0°C, the water in the hydrogel freezes and the hydrogel becomes rigid, brittle and non-conductive, thereby losing its desirable properties. Here we demonstrate a class of hydrogels that do not freeze at temperatures far below 0°C, while retaining high stretchability and fracture toughness. These hydrogels are synthesized by soaking in a suitable amount of ionic compound into the hydrogel. The present study focuses on a polyacrylamide-alginate double network hydrogels equilibrated with aqueous solutions of calcium chloride. The resulting hydrogels can be cooled to temperatures as low as -57°C without freezing. In this temperature range, the hydrogels can still be stretched more than four times their initial length and have a fracture toughness of 5000 Jm⁻². In particular, we show that the mechanical properties of these salts-containing hydrogels reach an optimum by going through a “slurry” state, coexistence of a mixed phase of ice crystals and salt solution inside the gel network, before fully freezing. We give a comprehensive explanation of the reasons of this optimum through a micro-mechanical understanding of the observed modified failure scenario. We anticipate that this new class of hydrogels will prove useful in the development of applications operating under a broad range of atmospheric conditions, and we further demonstrate this through a proof-of-concept of an ionic touch sensor operating well below the freezing point of water.

SYNTHESIS OF FUNCTIONAL SMART HYBRID MATERIALS

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Although technological and scientific importance of functional polymers have been well established over the last few decades, the most recent focus that attracted much attention are stimuli-responsive polymers. This group of materials is of a particular interest due to their ability to respond to internal and/or external chemico-physical stimuli that is often manifested by large macroscopic responses. Aside for scientific challenges of designing stimuli-responsive polymers, the main technological interest are numerous applications ranging from reactive surfaces to drug delivery and separation systems, or from chemo-mechanical actuators to other applications that have been extensively explored.¹

Here, different approaches to prepare devices based on thin layers of smart hydrogels will be presented. For instance, temperature responsive films with LCST or UCST behavior were combined with pH responsive hydrogels.² Their application in sensors and actuators were demonstrated. To increase the scope of such hydrogels a novel dual crosslinking system combining photo cross-linkable covalent bonding with special molecular recognitions sites was introduced. When the noncovalent bond was broken or formed, the swelling ratio of the polymer gel will be changed significantly. Further, an alternative way to synthesize mesoporous Al₂O₃ by using hydrogels as porogenic material is presented. Hydrogels can easily be patterned by light and used to imprint their structure onto alumina opening a new approach to fabricate patterned Al₂O₃. Followed by a nanocasting process mesoporous alumina samples were synthesized.^{3,4}

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DYNAMICS OF ADSORBED LAYER IN HYBRID HYDROGELS

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Our group developed hybrid model networks (Fig 1.) The principle relied on polymer adsorption onto nanoparticles (NPs) to design reversible cross-links while chemical cross-links permanently fix the network topology. The reversible interactions between poly(*N,N*-dimethylacrylamide) chains and NPs have been demonstrated to drastically improve the dissipative processes, providing nanocomposite hybrid gels with highly enhanced mechanical properties together with self-recovery processes in bulk¹ or at the gel interfaces by spreading NPs solution to provide robust adhesion between covalently crosslinked gels.²

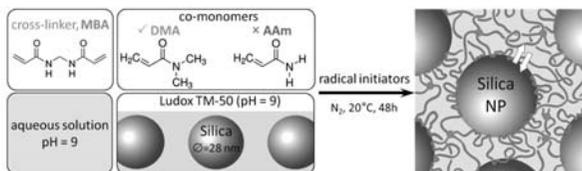


Figure 1. Hybrid networks combine sacrificial exchangeable bonds (arrows) and permanent cross-links (dots). Tuning of interactions is done by replacing DMA by non-adsorbing AAm.

The aim of this work is to better understand the role of the dynamics of adsorbed gel layer at the NPs interface, since by analogy with filled rubbers³ this layer is believed to be a key aspect for mechanical reinforcement⁴. Here, we first synthesized linear polymer chains using controlled polymerization to quantify polymer adsorption efficiency onto silica NPs. Gels were then designed with a fine tuning of the polymer/NPs interactions by replacing adsorbing DMA units by non-adsorbing AAm units while controlling the NPs dispersion state independently. The gel layer dynamics was studied by NMR, Large Amplitude Oscillatory Shear and fracture properties.

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KINETICALLY CONTROLLED CROSSLINKING OF METALLO-SUPRAMOLECULAR NETWORKS DRIVEN BY THE BELOUSOV-ZHABOTINSKY REACTION

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The supra- and macromolecular dynamics in supramolecular polymer systems are complex, and we aim at gaining rational understanding of their underlying interplay. To better separate these two different dynamics from one another, we control the periodicity and synchronicity of metallo-supramolecular crosslinking of poly(ethylene glycol) (PEG) with terpyridine-functionalized side-chains using the autocatalytic Belousov–Zhabotinsky (BZ) reaction.^{1,2} The precisely tuned BZ reaction allows us to obtain control over the otherwise random complexation process of the metal–ligand bonding by generating periodical variations of the transient network density, translating into variations of its viscoelastic properties, and therefore to delimit random from coordinated supramolecular crosslinker blinking. In particular, we work toward obtaining insight into the relevance of bond-lifetime renormalization due to sticker cooperativity in transiently crosslinked systems. This approach will bring us to a better understanding of polymer chain dynamics through comprehension of the role and impact of macro- and supramolecular contributions.

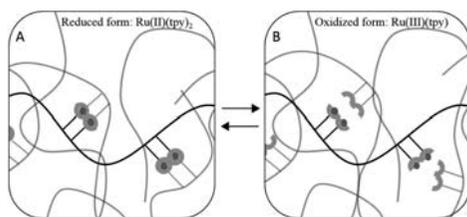


Figure 1. Ruthenium-mediated metal–ligand complexation of supramolecular PEG with terpyridine-functionalized side-chains. Reversible complex formation under BZ conditions of (A) Ru(II)-bis-terpyridine-PEG metallo-supramolecular network and (B) Ru(III)-terpyridine-PEG macromonomers.

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EFFECTS OF CROSSLINKING ON THE EDGE MORPHOLOGY OF PATTERNED POLYMER BRUSHES

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Polymer brushes, a collection of polymer chains whose one end is covalently fixed on a substrate, are important in tuning various surface properties.¹ When polymer brushes are grown from a patterned surface, chains near the pattern edge tend to be extruded out onto the bare substrate, making the pattern edge dull.² It is expected that introduction of crosslinks restricts the lateral conformational freedom of the brush chains and suppresses the chain extrusion. In this study, we aimed to reveal the effects of crosslinking on the microscopic morphology near the edge of patterned brushes.

The model crosslinked and non-crosslinked patterned brushes were prepared by a combination of photolithography technique and surface-initiated atom- transfer radical polymerization (SI-ATRP) of *N*-Isopropylacrylamide (NIPAAm, monomer) and *N,N'*-methylenebisacrylamide (BisAAM, crosslinker) (Figure, top). Atomic force microscopy (AFM) was employed to obtain the height image and cross-sectional profile near the brush edge (Figure, bottom). It is evident from the cross-sectional profile that the edge of the crosslinked patterned brush is steeper than that of the non-crosslinked counterpart with the similar brush height. The result suggests that crosslinking suppresses the chain extrusion and makes the pattern edge steeper. Quantitative analyses of cross-sectional profiles and the effects of the brush height will also be discussed in the presentation.

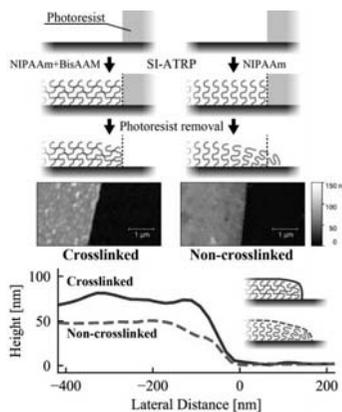


Figure 1. Preparation method, AFM height images, and cross-sectional profiles of the model patterned brushes.

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MICRORHEOLOGY ON WEAKLY ASSOCIATED PEG CHAINS IN SEMI-DILUTE SOLUTION

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Aqueous solutions of flexible poly(ethylene glycol) (PEG) are widely used in numerous biomedical and technical applications. However, the structural properties of the PEG chains in these and also in organic solvents are not

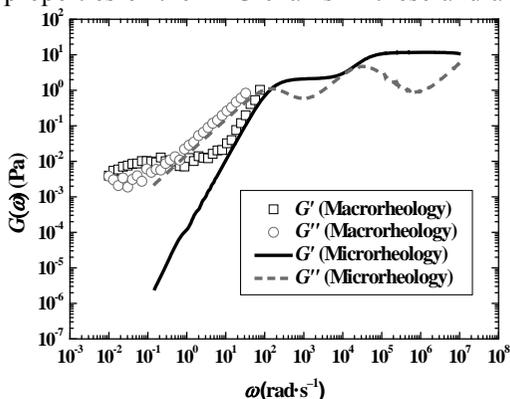


Figure 1. Storage and loss moduli of a 20 kg·mol⁻¹ PEG sample in water obtained from macrorheological measurements using a rheometer (open symbols) and microrheological measurements using DLS (lines).

fully understood, as there is experimental evidence and various theories about the formation of helical- or cluster-based chain associations.^{1,2} In rheological measurements of aqueous PEG solutions (PEG with a molar mass range of 2,000 – 20,000 g·mol⁻¹) we observe a frequency-independent low-frequency

second plateau of the storage modulus G' , indicating a weak transient network structure. To investigate this and to gain a better understanding on the network structure of chemically uncrosslinked PEG chains in semi-dilute solution, we take advantage of microrheological methods based on dynamic light scattering with spherical PEG-coated gold nanoparticles as small probe particles.

understanding on the network structure of chemically uncrosslinked PEG chains in semi-dilute solution, we take advantage of microrheological methods based on dynamic light scattering with spherical PEG-coated gold nanoparticles as small probe particles.

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CONTROLLED SYNTHESIS OF ZIF-8, ZIF-67 AND THEIR MIXED METAL DERIVATIVES BY A REACTION DIFFUSION PROCESS IN AGAR GEL

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Zeolitic imidazolate frameworks (ZIFs), a subclass of metal-organic frameworks (MOFs), have been recently employed in various fields such as gas separation, catalysis, water purification and drug delivery.¹ ZIFs have been synthesized solvothermally or at room temperature using organic solvents or pure water.² The control of size and morphology of crystals has been achieved using microwave, ultrasound-assisted syntheses and coordination modulation methods.¹⁻³ Herein, we investigate a new synthesis method where ZIF crystals are produced using the reaction-diffusion framework (RDF) in Agar gel medium at room temperature. The method is based on the diffusion of an outer solution of the organic linker or mixed linkers into Agar gel matrix containing the inner metal ions Zn(II) where a precipitation reaction takes place leading to the formation of the ZIF crystals. A propagating supersaturation wave, initiated at the interface between the outer solution and the gel matrix leads to a precipitation front endowed with a gradient of crystal sizes ranging between 100 nm and 55 μ m along the same reaction tube. The effects of temperature, the concentration of the reagents, and the thickness of the gel matrix on the growth of the ZIF crystals are investigated. We also show that by using RDF, we can envisage the formation mechanism of the ZIF crystal. Moreover, the formation of a solid-solution ZIF via the incorporation of Co(II) and Zn(II) cations within the same framework is achieved in a controlled manner. Finally, we demonstrate that doping ZIF-8 by Co(II) enhances the photodegradation of methylene blue under visible light irradiation without using hydrogen peroxide.

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TUNABLE PROPERTIES OF BETAINE-BASED MATERIALS

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Betain-based materials increase attention in biomedical and technological applications as a smart material.¹ Betaine polymers possess zwitterionic character with overall neutral charge, however, negative and positive charge in the same monomer units. They utilization is mainly in prevention and biofouling, bioconjugation and drug delivery systems.

This study is focused on synthesis hydrogel polymeric networks based on combination of zwitterionic monomers as well as different character of polymeric backbone. Combination of methacrylic zwitterionic monomer can lead to modulation of upper critical solution temperature (UCST) of prepared copolymers and hydrogels. Mechanical (compression strenght, stress at break...) and optical properties (transparency, refraction) can be adjusted and tailored by combination and concentration of these monomers and crosslinker in hydrogels. Some derivatives as carboxybetaine ester with positive charge can undergo pH-induced and -controlled hydrolysis of esteric moieties and formation of a neutrally charged zwitterionic structure. This feature allows polyionic complexation with negatively charges species and release after pH controlled hydrolysis, which will be showed by interactions with DNA molecules. Furthermore, investigation on polymerization for formation of poly(disulfides) network on selected betaines will be discussed. Polysufobetains material allows pH as well as thermally and chemically controlled depolymerization which will be demonstrated by means on UV, circular dichroism and dynamic light scattering measurement.

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**EXPERIMENTAL OBSERVATION OF TWO FEATURES
UNEXPECTED FROM THE CLASSICAL THEORIES OF RUBBER
ELASTICITY**

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Although the elastic modulus of a Gaussian chain network is thought to be successfully described by classical theories of rubber elasticity, such as the affine and phantom models,¹ verification experiments are largely lacking owing to difficulties in precisely controlling of the network structure. We prepared well-defined model polymer networks experimentally, and measured the elastic modulus G for a broad range of polymer concentrations and connectivity probabilities, p . In our experiment, we observed two features that were distinct from those predicted by classical theories. First, we observed the critical behavior $G \sim |p-p_c|^{1.95}$ near the sol-gel transition. This scaling law is different from the prediction of classical theories, but can be explained by analogy between the electric conductivity of resistor networks and the elasticity of polymer networks. Here, p_c is the sol-gel transition point. Furthermore, we found that the experimental $G - p$ relations in the region above C did not follow the affine or phantom theories. Instead, all the $G/G_0 - p$ curves fell onto a single master curve when G was normalized by the elastic modulus at $p = 1$, G_0 . We show that the effective medium approximation for Gaussian chain networks explains this master curve.

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SHEAR DEFORMATION OF ENTANGLED AND UNENTANGLED POLYMER NETWORKS: A MONTE-CARLO-STUDY

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Using a highly efficient GPU version of the bond-fluctuations model, we study the elastic properties of end-linked 4-functional model networks where we can switch off either entanglements or both entanglements and excluded volume. This allows to separate the contribution of the connectivity and the topological constraints to the shear modulus and to test the presumed additivity of both terms for a simple shear deformation. For the connectivity contribution, we take into account network defects, finite cyclic structures and the effect of excluded volume. We estimate the entanglement degree of polymerization and compare its scaling with the polymer volume fraction. Similar to a preceding study,¹ we observe a weaker power law as predicted in text-books.² This discrepancy may be caused by the cross-linking process, which induces an extra stress in the networks that grows with decreasing overlap number of the network strands.

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O-39

ON THE ELASTICITY OF PHANTOM NETWORKS WITH CYCLIC AND LINEAR DEFECTS

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The impact of finite cycles on the phantom modulus of otherwise perfect networks is computed exactly correcting a recently published approach.¹ These results are compared with theoretical estimates on the impact of linear and branched defects for some particular model networks structures (end-linked model networks, networks of star polymers, etc ...). It is further discussed that the formation of cyclic structures and other processes may lead to the chain conformations in a network that are deformed with respect to the reference state of a free chain at same polymer concentrations. A precise estimate of phantom modulus, therefore, requires information on conversion and on the overlap number of strands, the latter controlling simultaneously the formation of finite loops and excess strain in the network.

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O-40

NEW DEVELOPMENTS IN RANDOM GRAPHS

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The application of graph theory to chemistry has a long-standing tradition. On many occasions a reduction of a molecular structure to simply a diagram of bonds and nodes, or a graph, does still contain the information on the physical properties of the molecule. Notably, differences in boiling temperatures of isomers, formation heats of conjugated hydrocarbons, and vibrational potential energy of proteins has been successfully explained by the graph theory. Of course, to make use of the graph theory one must exactly know what connectivity structure is formed by the covalent bonds. In the field of polymer networks, which feature no pre-defined regular structures, such a prerequisite might be quite restrictive. The recently introduced idea of random graphs can help us to recast the known theories to face the inherent irregularity of polymer networks.

In this talk, I will review the latest developments in the field of random graphs,¹⁻³ and show how these can be of an aid to a modeller trying to connect experimental observables with the polymerization conditions. As a bonus, I will also discuss interesting peculiarities that the theory predicts but until now has not been experimentally reported.

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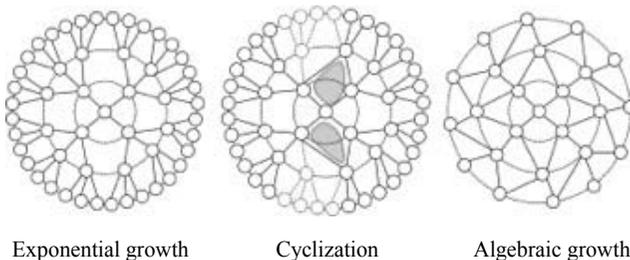
THE EFFECT OF EUCLIDEAN SPACE ON GRAPH-LIKE MODELS OF HYPERBRANCHED POLYMER NETWORKS

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Since the development of the first models for hyperbranched polymer networks by Flory, there is a heated discussion about the effects of three-dimensional space on polymer networks. Flory's model¹ and many models developed afterwards, e.g. random graphs, assume a tree-like structure in the sol and infinite cycles in the gel. For instance, for functionality $f=3$ the percolation threshold is predicted to be $p=0.5$. These assumptions, however, predict (1) no small cycles and (2) exponential growth with the number of generations when explored from an arbitrary node. Another alternative is lattices, which predict a higher percolation threshold than Flory and are space-embeddable, but the percolation threshold strongly depends on the chosen lattice. Additionally, Eichinger proposed a model that predicts no percolation for $f=3$.²

We propose a model that features algebraic growth of the networks and yields a constant density in Euclidean space while still sustaining random structures (in contrast to lattices). By not constraining the number of parents of a node we allow cyclization in the network. The model was developed as an extension of our previous work.³



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MODELING OF ETHYL LINOLEATE POLYMER NETWORKS VIA AUTOMATED REACTION MECHANISM

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Complex polymerization problems arise in the field of restoration and conservation of art. Many paints and coatings consist of triacyl glycerides (TAG) that polymerize and form hard films at the room temperature and atmospheric pressure. These films are known to be unstable within decades and centuries. Our starting point to explore this process is to model polymerization of ethyl linoleate (EL), which follows reaction pathways similar to the TAG molecule. This polymerization is not trivial and cannot be treated by means of existing polymer reaction engineering approaches. EL undergoes series of transformations before it even starts polymerizing and forms 3 types of crosslinks with other monomers.¹ Due to the complexity of the monomer structure we face a combinatorial explosion of the number of states of one EL unit outside and within the polymer network. For this purpose we developed a methodology based on so-called ‘monomer approach’. In this approach we do not consider the polymer formation explicitly, but rather keep statistics over the basic units that form polymer network. The number of molecular species scales down from infinity to the number of unique fragments that contribute to the network. We developed an automated reaction mechanism² that reconstructs all possible configurations of these fragments and captures all possible reactions between them in a form of a reaction network. This reaction network is then transformed into the population balance equations. Solving them, we obtain the distribution of crosslinks over the drying time of EL. Applying random graph theory, this information can be used further to uncover properties of the polymer network, such as predicting gel point and obtaining molecular weight distribution. This methodology is generic and can be applied to other complex polymerization problems.

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THERMODYNAMIC MODEL FOR A REVERSIBLE DESALINATION CYCLE USING WEAK POLYELECTROLYTE HYDROGELS

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We propose a novel desalination scheme which employs the hydrogels as a tool to draw salt ions from a brine with low salinity to a brine with high salinity. As a driving force we use the fact that the volume of polyelectrolyte gel does depend on the surrounding salinity. While the salinity defines the gel volume in open system, i.e. in equilibrium with big bath, in closed system salinity is defined by the gel volume. The compression and swelling in open and closed system processes are combined into four stages thermodynamic cycle working between two bathes of different salinities. The cycle implies reversibility at any stage, so, in principle, the method can achieve the maximum thermodynamic efficiency. We have shown that for weak polyelectrolyte gels the dependence between the salinity and gel volume appears to be non-monotonic. Depending on the model parameters the surrounding salinity may increase or decrease during compression. In both cases we consider the possible use of this relation in desalination cycle.

SELF-CONSISTENT FIELD MODEL OF WEAK POLYELECTROLYTE GELS

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We present a novel approach¹ to modeling polyelectrolyte gels, exploiting the analogy between star-branched polymers and polymer networks as a computationally inexpensive yet reliable alternative to full-scale simulations. In the numerical mean-field model of a star-like polymer we modify the boundary conditions to represent an infinite network. We validate the predictions against a coarse-grained simulation model. The mean-field model explicitly considers local density gradients and agrees with the simulation results in a broad range of parameters, beyond that of the analytical model. Finally, we use the mean-field model for predictions of the swelling behaviour of weak polyelectrolyte gels under different pH conditions. We demonstrate that the local density gradients are important and that the ionization of the weak polyelectrolyte gel is significantly suppressed with its effective pK_A being about one unit higher than that of the free monomer.

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QUANTIFICATION OF LIGATION POINTS IN PHOTOCHEMICALLY LINKED POLYMER NETWORKS

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We introduce a facile fluorescence-based methodology for the counting of linking points in polymer networks (Figure 1).

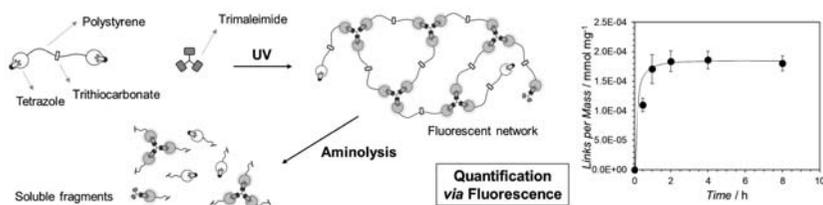


Figure 1. Fluorescent networks prepared under UV irradiation and subsequently degraded *via* aminolysis. Fluorescence correlates directly with the amount of ligation points of the network.

Tetrazole end-capped polystyrene chains were prepared *via* RAFT polymerization.¹ These chains were crosslinked under UV irradiation by a trimaleimide *via* nitrile imine-mediated tetrazole-ene cycloaddition (NITEC).^{1,2} A strongly fluorescent adduct is generated for each one of the links formed. Quantification of the ligation points is done by simple fluorescence readouts of the soluble network fragments, obtained after network degradation *via* aminolysis of the trithiocarbonate RAFT groups.³ This methodology offers the possibility of a quantitative approach to the structural characterization of a range of polymer-based networks.

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A MODEL PHYSICAL GEL CROSSLINKED BY DOUBLE STRANDED DNA

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Physical polymer gels are familiar materials for humankind, including jelly, boiled eggs, and even the living organisms. They are composed of a small number of polymer chains (~10wt%) and a large number of water (~90%). Because the polymer chains are crosslinked with each other to form a giant 3D-networks, the physical gel does not have fluidity even though a large amount of water is contained as liquid. The cross-linker in physical gels are formed by reversible non-covalent bonding: hydrogen bond, Coulomb interaction, or hydrophobic interaction. The physical gels show reversible sol/gel transition by adding the external stimuli (temperature, pH, solvent quality, etc.). These bonding are generally formed randomly and non-specifically, easily resulting the aggregation and heterogeneous network structure.

In this study, we synthesized a new physical polymer gel with a DNA-terminated tetra-functional polyethylene glycols. Because the branching point are located at the center of the module polymer and the physical bonding of DNA are extremely specific, the resultant physical gel is expected to be an ideal physical polymer gel. The physical properties and static structure are investigated by rheological measurement and small angle neutron scattering technique. The polymer gel was stable and showed rubber elasticity at room temperature and exhibited reversible sol/gel transition at around 70°C. The sol/gel transition temperature is reasonable considering the melting temperature (a temperature for double-helix to come apart to 50%) of DNA (~63°C). The scattering profile at room temperature was resemble to that of chemically crosslinked polymer gel, suggesting the universal nature of semi-dilute polymers as a continuous body. However, a peak was found at high-temperature (~80°C), suggesting micelle-like structure.

MECHANICAL PROPERTIES OF TISSUE-LIKE HYDROGELS PRODUCED BY INJECTION OF SPRAY-DRIED COLLAGEN

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Simple injection of concentrated collagen to obtain biomimetic hydrogels is desirable for regenerative medicine such as dermal fillers or wound healing. It is well-known that highly concentrated collagen solutions can form tissue-like organizations¹ with required mechanical behaviours after fibrillogenesis. Yet injection of such solutions is still challenging due to their high viscosity. Conversely, chemically crosslinked collagen-based gels can be readily injected.² Although their mechanical properties are better than that of un-crosslinked injectable collagen gels,³ their composition may lead to inflammatory responses. Recently, highly concentrated fibrillar collagen gels (18wt%) were injected – however their microstructure lacks the structural features characteristic of native tissues.⁴ How to combine biomimetism and injectability of 100% collagen-based hydrogels?

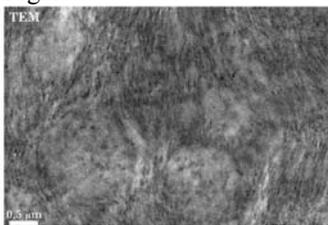


Fig. 1: TEM micrograph of 8wt% collagen

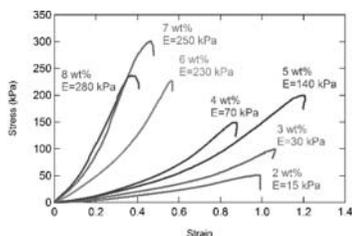


Fig. 2: Tensile properties of collagen gels

To address this issue we inject⁵ highly concentrated collagen beads into a mould mimicking a tissue defect. Electron microscopy observations and mechanical tests results show self-assembly capability and promising mechanical behaviour compared to collagen-rich native tissues.

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IMMUNOMODULATORY EFFECTS OF SCHIZOPHYLLAN ON RAW264.7 CELL IN THREE-DIMENSIONAL CULTURE MATRIX

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Schizophyllan (SPG), one of water-soluble β -glucans obtained from *Schizophyllum commune*, has immunomodulatory activity as binding on several membrane receptors of immune cells.¹ In most cases, in vitro immune cell and SPG influence studies has relied on tissue culture plate (TCP).²⁻⁴ However, such a 2D culture platform often has limitation to mimic 3D microenvironment of native tissue, in which immune cells reside or perform immune responses.⁵ In this study, PEG hydrogel-based cell culture matrix was therefore utilized in macrophage culture and the immunomodulatory effects of SPG on encapsulated macrophage were evaluated. In 3D hydrogel matrix, SPG induced inflammatory responses of RAW264.7 cells with upregulation of M1 macrophage phenotype markers (i.e., iNOS, COX2, TNF- α). Compared to 2D cultured cells, 3D-cultured cells were a lower sensitivity to SPG. It was probably due to less cellular uptake of SPG in 3D. We found SPG recognition receptors of encapsulated RAW264.7 cells were downregulated. On the other hand, SPG slightly increased M2 macrophage phenotype markers (i.e., CD206, Arg-1, and TGF- β) in 3D, indicating that macrophages tend to have immune-regulation function than in 2D environment. These results indicate that PEG-based hydrogel 3D culture system would be a promising cell culture platform for investigation of the immunomodulatory effects of SPG on macrophage.

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DESIGN OF THIOL-MODIFIED CHITOSAN HYDROGELS WITH DIFFERENT MALEIMIDE-BASED CROSS-LINKERS

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Biomaterials that are rapidly formed are highly desirable in many biomedical applications.¹ For this purpose, *in situ* forming hydrogels have attracted wide attention as excellent 3D scaffolds. Thiol-Michael addition reaction, which is an efficient click chemistry reaction due to its mild reaction conditions and high yields,² was selected in this research work for the formation of chitosan-based hydrogels.

Thiol moieties were covalently incorporated into chitosan by reaction with thiolactic acid in aqueous medium. The modification was demonstrated by infrared and nuclear magnetic resonance spectroscopy, where new signals appeared in both cases. Besides, thiol group content was determined by Ellman's test. Subsequently, various maleimide-functionalized cross-linkers were employed for the *in situ* hydrogels formation via click chemistry in PBS buffer at 37°C; namely, previously synthesized bis- and tris-maleimide and commercial tetra-maleimide. The obtained biomaterials are expected to be good candidates for tissue engineering applications.

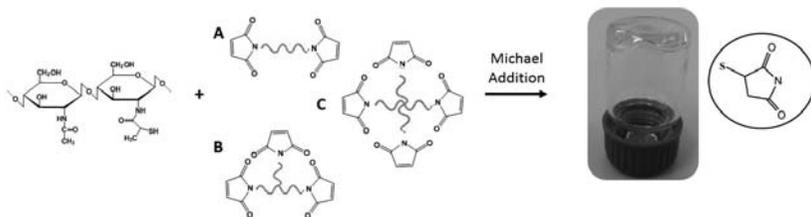


Figure 1. Schematic representation of the synthetic process based on thiol-maleimide Michael addition for hydrogels formation.

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WATERBORNE POLY(URETHANE-UREA) SYNTHESIZED FROM BIOBASED POLYOL AND TRIBLOCK COPOLYMERS CONTAINING HYDROPHILIC BLOCK

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Bio-based polyols are good option to prepare poly(urethane-urea)s from renewable sources. On the other hand, given the variety of morphologies triblock copolymers can offer, leading to well-defined structures at the nanometric level,¹ they are suitable to be used as polyols in the synthesis of nanostructured poly(urethane-urea)s. Is for these reasons that both, a bio-based polyol and different triblock copolymers, have been used as polyols to synthesize waterborne poly(urethane-urea)s following a solvent-free synthesis procedure.² Bio-based polyol was used in order to prepare poly(urethane-urea) from renewable sources; whereas different triblock copolymers, which contained an hydrophilic block, were used to synthesized poly(urethane-urea)s with nanostructured morphology, which can make them useful for incorporation of nanofillers. A poly(ethylene oxide) homopolymer was also used. This hydrophilic polyol was needed in order to obtain waterborne poly(urethane-urea) dispersions with the followed synthesis procedure. Characterization of the obtained dispersions and films was carried out.

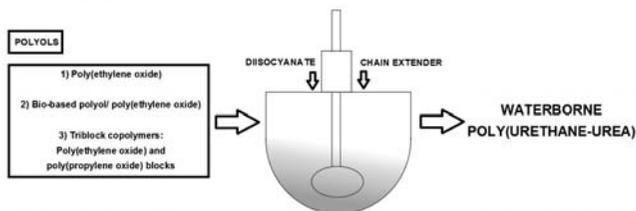


Figure 1. Synthesis of waterborne poly(urethane-urea) dispersions.

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TOOTH DERIVED STEM CELL CULTIVATION ON POLY(ASPARTAMIDE) BASED HYDROGELS

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Biodegradable and biocompatible hydrogels are one of the most promising scaffold materials in tissue engineering. They mimic the extracellular matrix (ECM) due to their 3 dimensional, fibrous polymer network structure, permeability for small nutrients and appropriate mechanical properties.

The natural ECM mainly contains polymers composed of amino acids, such as collagen, elastin, fibronectin, and proteins.² Therefore, synthetic poly(amino acid) (PASP) based materials, such as poly(aspartic acid), are good candidates for this application.³

In this work the effect of the mechanical and chemical properties of the PASP based hydrogels on cell viability, migration of differentiation have been investigated. The physical and chemical properties of the hydrogels were determined mechanical testing, SEM and different analytical methods. For in vitro experiments, MG-63 human osteoblast-like and two different (periodontal ligament and Dental pulp) stem cell lines were chosen. The viability, migration and differentiation of the cells were followed either by quantitative (WST-1, ALP) and qualitative methods (Phase contrast and two photon microscopy).

Poly(aspartic acid) based hydrogels can support proliferation as well as migration of human cells, which implies that our system could be a good candidate for 3D cell culturing of osteoblast type cells.

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MODIFICATION OF HYDROGELS BY A FEMTOSECOND LASER: FROM MECHANISM TO OPHTHALMIC APPLICATIONS

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Femtosecond lasers can locally alter the refractive index of many transparent materials by the process of multiphoton absorption and allow to write waveguides or optical lenses. Writing such structures into hydrogels used in ophthalmology was already reported, but the mechanism of refractive index change remains unclear.

We investigated the modification of the refractive index induced by femtosecond laser pulses in pHEMA based hydrogels with various types and concentration of UV absorbers and other additives. We characterized the modified structures using Raman spectroscopy and studied the induced optical phase change using interferometry.¹ To demonstrate the application potential of the femtosecond laser modification in ophthalmology, we report on writing optical corrections into hydrogels suitable as an intraocular lens material.

Interferometry shows predictable and reproducible negative phase shift in areas modified by the femtosecond laser. Raman spectroscopy indicates that increase in water content, rather than alteration of the polymer structure, causes the decrease of the refractive index. The magnitude of induced changes strongly depends on the material composition. We attribute the refractive index change to depolymerization of the hydrogel material.

¹ Gandara-Montano G.A., Stoy V., Dudic M., Haskovcova K., Knox W.H., *Opt. Mater. Express* 7, 2017, p. 3162-3180.

POLYMER NETWORKS FOR ENZYME-INDUCED MINERALIZATION

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Hydrogels are representing a widely used class of polymer materials that generally lacks mechanical stability. Here, we present a way to obtain fully transparent, ultrastiff hydrogels by synthesizing polymer matrices with finely dispersed enzyme aggregates that allow the formation of a “near perfect” percolated inorganic/organic double network from an aqueous inorganic precursor solution at room temperature. This is realized by prepared polymer networks with entrapped enzymes, such as urease and alkaline phosphatase. These enzymes are then converting water-soluble precursors into water-insoluble salts, such as CaCO_3 and Ca_3PO_4 .

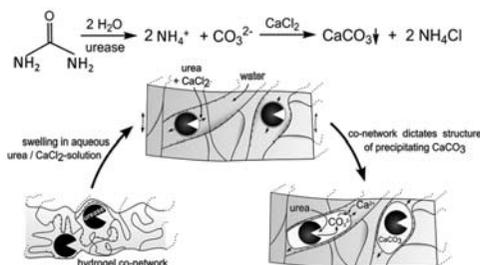


Figure 1. Concept of bio-induced mineralization in polymer networks

The stiffness of the water-swollen hydrogels goes up to 400 MPa. Also the toughness of the hydrogels is increased in the best cases to more than 1000 J/m^2 , which is comparable to human cartilage. Tuning of the properties, mechanistic studies, and application potential of these new materials are discussed as well.

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DESIGN OF NOVEL EPOXIDE MONOMERS FOR pH-SENSITIVE POLY(ETHYLENE GLYCOL) HYDROGELS VIA ACID-LABILE AND CROSSLINKABLE ALLYL SIDE GROUPS

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Poly(ethylene glycol) (PEG) is the "gold standard" for biocompatible polymers, due to its remarkable characteristics such as low toxicity, immunogenicity and unique aqueous solubility.¹ The range of PEG-applications is greatly expanded by the incorporation of crosslinkable side groups, which can be used for the synthesis of PEG-hydrogels.² These insoluble, biocompatible materials can absorb considerable amounts of water due to their hydrophilicity, rendering them promising for instance for drug delivery or as scaffolds for tissue engineering.³ One possibility to form covalent, polymeric networks is thiol-ene click chemistry.⁴

In this work, we introduce the novel, acid-cleavable ethoxy ally glycidyl ether (EAGE) monomer. Using anionic ring-opening polymerization P(EAGE-*b*-EO-*b*-EAGE) triblock copolymers were prepared on the basis of PEG-macroinitiators with different molecular weights. Hydrogels were formed by thiol-ene photo-click chemistry. The materials were optimized regarding their water absorption capacity by varying molecular weights of the blocks. To gain a deeper understanding of the materials properties, the triblocks were cross-linked and analyzed directly on the rheometer. In addition, random P(EO-*co*-EAGE) copolymers were synthesized and functionalized with *N*-(*tert*-butoxycarbonyl)-L-cysteine methyl ester by thiol-ene click reaction for post-polymerization modification. All copolymers were characterized by common analytical methods (SEC, NMR, DSC).

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**RATIONAL DESIGN OF ADVANCED ELECTROCHEMICAL
ENZYME BIOSENSORS VIA SURFACE FUNCTIONALIZATION
BY STIMULI-SENSITIVE MICROGELS**

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The thin (nanosized) microgel and microgel/enzyme films were fabricated on conductive substrates (graphite, gold) via two-steps, sequential adsorption of dual (pH and temperature) stimuli-responsive functional microgels, viz., poly(*N*-isopropylacrylamide-*co-N*-[3-(dimethylamino)propyl]methacrylamide), P(NIPAM-*co*-DMAPMA), or poly(*N*-isopropylacrylamide-*co-N*-(3-aminopropyl)methacrylamide), P(NIPAM-*co*-APMA), followed by adsorption of enzymes (choline oxidase, tyrosinase, butyrylcholine esterase) under different pH and temperature conditions. Both stimuli (pH and temperature) can synergistically control the surface coverage by microgel particles and therefore determine efficiency of the following enzyme uptake. Further, we highlight that the loading of the microgel films by the enzymes strongly depends on the microgel architecture, on the state of the adsorbed microgel particles (swollen vs. collapsed) and on the molecular weight (globule size) of the enzyme. With help of the P(NIPAM-*co*-DMAPMA) microgel, the bienzymatic biosensor system with spatially separated enzymes was prepared in one-step, while the stabilized biosensor system with high operational stability was designed using the P(NIPAM-*co*-APMA) microgel *via* covalent cross-linking of enzyme globules to its primary aminogroups.

This work was supported by the Deutsche Forschungsgemeinschaft within the SFB 985 "Functional Microgels and Microgel Systems" and the Russian Foundation for Basic Research within the RFBR project 17-08-01151.

POLYOXYMETHYLENE/FUNCTIONALIZED HYDROXYAPATITE NANOCOMPOSITES WITH IMPROVED THERMAL STABILITY

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Polyoxymethylene (POM) is well known engineering thermoplastic polymer widely used in many technical and biomedical applications like in joint replacement components and other long-term bone implants.

POM nanocomposites with both unmodified hydroxyapatite (HAp, $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$) and HAp functionalized using diisocyanates as coupling agents were investigated as potential biomaterials for orthopedic applications. For unmodified HAp, a dramatic decrease of POM thermal stability after incorporation of HAp nanoparticles during melt processing has been observed. In the next approach polymer chains such as poly(ethylene glycol) (PEG) or poly(ϵ -caprolactone) (PCL) have been grafted on HAp nanoparticles surface using an 1,6-hexamethylene diisocyanate as a coupling agent to obtain hybrid inorganic-organic system HAp-graft-PEG (or HAp-graft-PCL). Functionalized HAp was incorporated to POM copolymer in the same way as before for unmodified HAp using melt processing methods. The obtained results revealed that both HAp-graft-PEG and HAp-graft-PCL, in contrast to unmodified HAp, significantly improved the thermal stability of POM - from 296°C for POM to 326°C for POM modified with 10% of HAp-graft-PEG. This effect can be attributed to the reduction of the number of active centers in HAp which were active sites involved in catalytic reactions of POM depolymerization in POM/HAp nanocomposites.

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DYNAMICS AND STRUCTURE OF PVDF – SOLVENT GELS FILLED WITH SILICA

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VDF-based copolymers are widely used materials for example as Lithium battery separators. Silica nanoparticles can be added to improve their properties (thermal, mechanical...),¹ but the silica dispersion state must be carefully controlled to ensure the performances of final materials.

Homogeneous silica dispersion in PVDF by a solvent route was developed. For this purpose, silica must first be transferred in an appropriate solvent of the polymer.² Then, PVDF and VDF-based copolymers (filled or not with silica) form thermoreversible gels in solvents, the morphology of which strongly depends on the nature of the solvent. The gelation process must be mastered to obtain the desired material performances at the end.

PVDF gelation was studied in various solvents.³ For PVDF gels in methylethylketone (MEK), these features have hardly been addressed in the literature, and only in dried gels,⁴ assuming that drying does not affect the gel morphology.

Combination of ¹⁹F NMR, SAXS, WAXS and rheology on undried gels will be used to probe both the gelation mechanism and the structure of these systems. For binary PVDF/MEK mixtures, NMR evidences the presence of both elastically active chains and rigid zones in the gel state. Rigid regions are found to be crystalline through X-ray diffraction. As silica nanoparticles are added, the fractions of rigid zones and elastically active chains increase. These data will be compared to rheological measurements.

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WHAT CAN BE LEARNED FROM POLYMERIZATION IN A TWO-COMPONENT PYRIDINE-BASED SYSTEM

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A solution of poly(4-vinylpyridine) in pyridine (1:1 molar ratio) is known to undergo spontaneous gelation when left for several days either under illumination or in the dark. The resulting gel possesses completely different properties from those of the initial solution.¹ In particular, the gel exhibits unusual electro-optical properties, such as a large difference in the mobility of the photoinduced charges: proton mobility $\sim 1.8 \times 10^{-3} \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ and that of electrons $\sim 25 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$.² This difference suggests the existence of two distinct conductivity mechanisms and charge carriers. The observation of Stark splitting with the comparatively low energy (3.4eV) of the 1S electron level of the pyridine nitrogen² and the presence of the ¹H NMR signal at 10.5 ppm, indicates the existence of a species containing a protonated nitrogen. Thus, the gel involves a network of N⁺-H⁺-N hydrogen bonds as a result of H⁺ transfer and the formation of betaine-like structures. The DFT calculated energy gain upon formation of such a bond in a model dimer complex is $\sim 30 \text{ kJ/mol}$ and the respective N⁺-H distance is 1.91 Å. A second chemical reaction is the light-induced ring opening of the pyridine cycle in the presence of trace amounts of water, giving rise to aminopentadienal and its subsequent oligocondensation. The azaoligoacetylene conjugated molecules can serve as molecular wires to provide electron conductivity. The consistently reproducible gelation of poly(4-vinylpyridine)/pyridine solutions produces materials with a variety of dielectric/semiconductor/ionic/electron conductive properties which are suitable for different technologies for energy conversion in charge transfer based devices.

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METALLIC NANOPARTICLES IMMOBILIZED AT THE PORE SURFACE OF POLYMERIC MATERIALS: TOWARDS VERSATILE AND EFFICIENT CATALYTIC SYSTEMS

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This presentation will be focused on hybrid systems consisting in metallic nanoparticles immobilized at the pore surface of three types of porous polymers: (i) nanoporous polystyrenes stemming from the selective degradation of diblock copolymers possessing a cleavable chemical junction between both blocks,¹ (ii) bulk macroporous monoliths prepared from a dimethacrylate-based crosslinker possessing a disulfide bridge² and (iii) in-capillary macroporous monoliths prepared from a functional monomer, *i.e.* *N*-acryloxysuccinimide.³ After immobilization of metallic nanoparticles, such hybrid functional materials have been successfully applied as catalytic supports in diverse model organic reactions, including the reduction of dyes or nitroaromatic compounds, notably under flow through conditions, but also for more complex C-C homocoupling reactions or even smart cascade reactions consisting of two successive supported nanogold-catalyzed reactions (Figure 1).

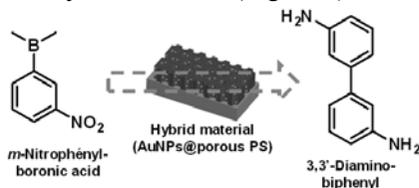


Figure 1. Cascade reaction carried out at the surface of porous polystyrene immobilized nanogold.

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MESOPOROUS POLYMERS CREATED BY MICROSYNTERESIS AND THEIR APPLICATION

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Recently there was found¹⁻³ that during polymerization of monomers with high content of crosslinking component diluted with 2 – 10-times volume surplus of porogenic solvent able to swell there is created stable morphology with up to 90 % porosity. So high porosity can be created during polymerization by phase separation of microsineretic rather than macrosineretic type. The expanded state of these polymers does not depend on nature of surrounding solvent but the porous structure is prone to partial collapse during drying. Probably that is the reason why this phenomenon was not described sooner, as approach to the investigation of relations between preparation conditions and resulting polymer morphology has been traditionally based on dry-state morphology assessment. However, substantial increase of resistance to the collapse during drying is possible to achieve by additional, post-polymerization crosslinking (hypercrosslinking) by e.g. by Friedel-Craft's reaction of the residual double bonds. After this morphology "reinforcement" the exceptional properties of these polymers are evident even in results of conventional nitrogen adsorption porosimetry.

Polymers created by microsineresis offer exceptional morphological properties - high volumes of wide pores in 10 - 80 nm range, high surface area 200 – 900 m²/g in absence of micropores and rigid morphology stable in wide variety of solvents. We investigated possibilities of application of these materials and there was already proved that as solid state peptide synthesis supports they can offer quite competitive advantages against existing commercial types.⁴

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« COLORED » MULTI-STIMULI RESPONSIVE HYDROGELS

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Controlled deformation processes are ubiquitous both in biological systems and materials science. For example, muscles may undergo expansion or contraction as a result of an external physiological stimuli. Similarly, synthetic smart polymer gels, capable of swelling or deswelling in response to various physical and/or chemical stimuli, have been developed during the last decades. Complexes fabricated from the electron deficient cyclobis(paraquat-*p*-phenylene) (CBPQT⁴⁺) and electron-rich guests have become one of the most important building blocks for the synthesis of colored self-assembled architectures.¹ Here, we report on the successful engineering of new multi-stimuli responsive macromolecular hydrogels featuring CBPQT⁴⁺ based complexes. More particularly, we have exploited these colored CBPQT⁴⁺ based interactions to i) control the swelling/shrinking processes of materials by applying different stimuli (T, V, competitive macromolecules)² i) to impart both thermal and temporary memory function to hydrogels³ and iii) to develop polymeric hydrogel systems capable of swelling *via* a supramolecular transmission. An important practical aspect of these new functional materials is that all relevant phenomena (swelling/shrinking processes, memory function) have an associated visible readout.

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APCNS BASED ON CRYSTALLIZABLE POLY(2-OXAZOLINES)

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Poly(2-methyl-2-oxazoline) (PMOx) and poly(2-heptyl-2-oxazoline) (PHepOx) ABA triblock copolymers were used to form APCNs with distinguished nanostructures. Therefore two different crosslinking strategies were used, that both focus on the end groups of the polymers. It was found that the PHepOx blocks partially crystallize over 19 vol.% of PHepOx in the APCNs.

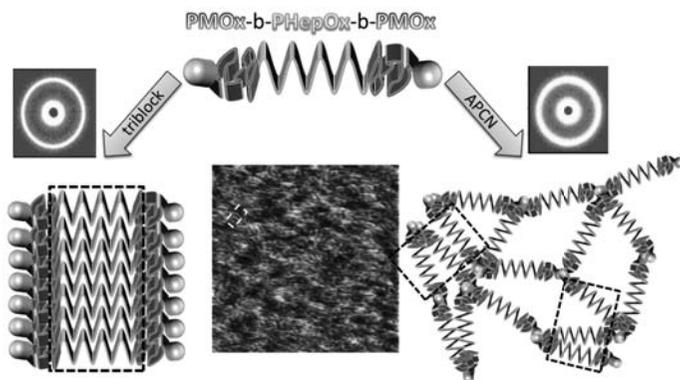


Figure 1. Schematic overview of the expected arrangement of the crystalline PHepOx blocks, in PMOx-b-PHepOx-b-PMOx triblock copolymers and in the APCNs.

The crystals in the APCNs were found to be smaller than those formed by the ABA triblock copolymers (Figure 1), but the scale of the crystals in the APCNs is independent from the composition of the polymer networks. Moreover, the crosslinking reaction using the end groups leaves the inner PHepOx block enough freedom to form these crystal domains in the APCNs.

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INTEGRATION OF INDUSTRIAL BY-PRODUCTS IN BIO-BASED RESINS AND COMPOSITES

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In the context of resource depletion, bio-sourced materials are a very promising alternative to conventional solutions. We propose here the use of bio-based resins formulated from humins, epoxidized linseed oil (ELO) and an industrial hardener Capcure 3-800. Humins,¹ a by-product of bio-refineries, are complex mixtures of furan derivatives presenting varying degrees of modification and polymerization. ELO² is a type of epoxidized vegetable oil with an 8-9% epoxide content, viewed as an eco-friendly alternative to synthetic epoxides, and has previously been used in resin formulations. The objective of the RECYSITE project (LIFE15 ENV/BE/000204) is the development of composite materials based on vegetable fibers using a resin based on humins and ELO. This type of composites aims to have an up to 85% bio-sourced composition, thus representing a viable solution in guaranteeing sustainability in the plastics sector and contributing to circular economy. A wide range of mechanical properties has been obtained for the materials, from elastomers to rigid thermosets. Figure 1 displays the results of the project.

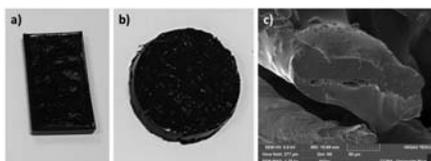


Figure 1. a) Humins/Capcure/ELO resin; b) resin/hemp composite; c) SEM micrograph of a resin/hemp composite

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INFLUENCE OF PROCESSING VARIABLES ON THE RHEOLOGICAL PROPERTIES OF ONE-STEP PROCESSED CASTOR OIL/LIGNIN-BASED GEL-LIKE POLYURETHANES

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The increasing interest in harmless and renewable materials has propelled the research on bio-based polyurethanes. The aim of the present work is to understand and compare lignin/castor oil-based polyurethane performing through a comprehensive study of a facile one-step preparation protocol, free of any harmful catalyst or solvent, by analysing the effect of related processing variables in the structural and thermo-rheological properties. Different temperatures (25, 45, 70°C), stirring speeds (23, 70, 140 rpm) and two kinds of diisocyanates (hexamethylene diisocyanate (HDI) and tolylene-2,4-diisocyanate (TDI)) were selected to process the different polyurethane samples. Processing variables have demonstrated to greatly influence the viscoelastic response, temperature dependence and curing process. Fourier transform infrared spectroscopy (FTIR) and differential scanning calorimetry (DSC) techniques were used to follow the curing process. Comparing the oleogels thus obtained with previously developed two-step polyurethanes¹, outstanding properties were generally obtained using lower lignin concentration and higher lignin/diisocyanate ratio, also by applying shorter reaction times. Lignin proved to have a powerful effect on the viscoelastic moduli of final polyurethanes.

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RHEOLOGICAL BEHAVIOUR OF EPOXIDE-FUNCTIONALIZED CELLULOSE PULP GEL-LIKE DISPERSIONS IN CASTOR OIL

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Nowadays, as a result of the environmental awareness, the lubricant industry is greatly involved in the replacement of petroleum-derived products with vegetable oils and renewable thickening agents from natural resources. In this work, several di-functional epoxides have been employed to chemically modify an industrial grade cellulose pulp from *Eucalyptus globulus* to be used as thickener in a castor oil medium. The resulting gel-like dispersions can be proposed as promising bio-lubricating grease formulations. The influence of thickener epoxy index on the rheological properties of gel-like dispersions was analyzed by using different amounts of neopentyl glycol dyglycidyl ether (NPGDGE) in the functionalization reaction. Furthermore, di-epoxide chemical structure was also evaluated. The resulting epoxidized cellulose pulp samples were characterized by means of epoxy index determination, thermogravimetric analysis (TGA) and Fourier transform infrared (FTIR) spectroscopy. Moreover, gel-like dispersions were fully characterized from a rheological point view. According to the results obtained, the values of the linear viscoelastic functions initially decreased by increasing the epoxy index but increased above a certain critical epoxy index. Finally, higher values of viscoelastic moduli were achieved when using aromatic di-epoxides to modify cellulose pulp.

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HYBRID SILICA HYDROGEL MATRICES FOR AN IMMOBILIZATION OF GLUCOSE SENSITIVE PROTEINS

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The preparation, structure and physical properties of hybrid silica hydrogel based on the organofunctional silanes is presented in this work. Tetraethoxysilane, 3-aminopropyl triethoxysilane and 3-epoxypropyl trimethoxysilane were employed as building blocks, whereas glycerol and various polymeric additives were used as processing agents.

The silica hydrogels prepared at physiological conditions were characterized with respect to the glucose diffusion properties and the porosity by employing various approaches. A diffusion coefficient of glucose in silica hydrogel was determined by two complementary techniques. The confocal laser scanning microscopy in a time-lapse imaging mode was employed to measure the ingress of fluorescently labeled glucose analogue inside the hydrogel. In addition, a method for direct glucose release from the hydrogel was established. The BET analysis and permeation of fluorescently labeled dextrans of various molecular weights were used to characterize the porosity of silica hydrogel. The radius of pores accessible for diffusion of dextran molecules in prepared silica hydrogel ranges between 1 and 6 nm.^{1,2} The mechanical testing was performed in the compression mode determining the mechanical characteristics as a function of aging time in TRIS buffer up to 14 days. A new phenomenological model was proposed to describe the stress – strain behaviour.

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PEROVSKITE NANOPARTICLE COMPOSITE FILMS BY SIZE-EXCLUSIVE MASS FLOW LITHOGRAPHY

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On account of their exceptional electronic and optical properties, organometal halide perovskite materials have been extensively explored in the various optoelectronic applications for last few years. For example, alkylammonium lead halide perovskite nanoparticles exhibit superior optical properties such as high color purity with narrow band width, broad color tunability covering entire visible region either by varying halide composition or particle size, and almost unity photoluminescence quantum yield (PLQY).¹ However their practical applications are often hindered by the poor stability of perovskite materials against water and polar solvents.² Additionally, patterning technology of perovskite materials is highly demanded for creating the high resolution pixels.

As a way of addressing above mentioned problems, perovskite nanoparticle composite films have been fabricated by using size-exclusive mass-flow lithography. Colloidal perovskites nanocrystals based on formamidinium lead halide (FAPbX₃) have been synthesized by ligand assisted reprecipitation method using PbX₂-DMSO complexes as precursors at room temperature. The synthesized nanoparticles were composited with various polymers. With application of size-exclusive mass-flow induced by photo-polymerization, perovskite particles were assembled within the polymer matrix. By choosing the properties of monomers and kinetics of photo-polymerization process, the well-defined patterns of perovskite particles were prepared.

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**A NOVEL STRATEGY FOR PRECISION NETWORK
FORMATION BASED ON PARA-FLUORO-THIOL LIGATION**

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In this work, we introduce the *para*-Fluoro-Thiol reaction (PFTR) chemistry as a new strategy for network synthesis.¹ Due to their light weight and their attitude to be recovered and re-used, polymer networks find application in several field such as energy, wastewater treatment, and biomedical drug carriers. Aside the composition, structural imperfections and the degree of cross-linking strongly influence the performance of the material itself.² As a consequence, the achievement of 'ideal network' has recently received significant attention from material scientists.³ Here, we show how, by designing a novel fluorinated linker, one could tune and introduce selectivity to the reaction. To assist the elucidation of the PFTR mechanism, a model reaction with a monothiol supported by a kinetic study is presented. The versatility of the concept is further demonstrated by employing several bifunctional thiols. A novel modelling tool is being developed for a characterization of the evolution of the polymer microstructure of individual macromolecules/segments during network formation. In addition, X-ray photoelectron spectroscopy (XPS) and time of flight secondary ion mass spectrometry (ToF-SIMS), are suggested as analytical tools for the confirmation of network formation.

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THERMALLY HEALABLE AND REPROCESSABLE POLYMER NETWORKS BASED ON DYNAMIC COVALENT CHEMISTRY OF BIS(HINDERED AMINO)DISULFIDES

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A facile approach to polymethacrylate networks that contain thermally exchangeable bis(2,2,6,6-tetramethylpiperidin-1-yl)disulfide¹ (BiTEMPS) cross-linkers is reported, and the easily inducible healability and reprocessability of the obtained networks are discussed (Figure 1). The free radical polymerization of BiTEMPS cross-linkers and methacrylate monomers afforded insoluble and colorless networks of the corresponding poly(methacrylate) films.

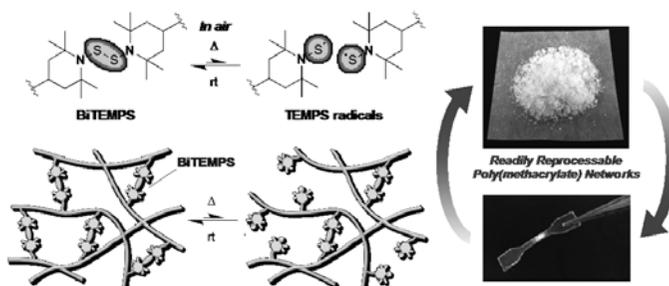


Figure 1. Polymer networks containing thermally exchangeable BiTEMPS.

Swelling experiments and stress-relaxation measurements at elevated temperatures revealed the contribution of BiTEMPS as a polymer chain exchanger both in the gels and in the bulk. The BiTEMPS-cross-linked PHMA networks showed damage healability and repeatable reprocessability in the bulk by simple hot pressing at 120 °C under mild pressure. These results should grant facile access to various vinyl polymer networks with on-demand malleability.²

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NOTES

POSTERS

P-01

EOR OF THE COMBINATION FLOODING SYSTEMS CONSISTING OF POLYMER MICROSPHERES AND NONIONIC SURFACTANT FOR BOHAI OILFIELD

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A new microspheres-surfactant flooding system consisting of sub-microscale polymer microspheres and nonionic surfactant is detailed to enhance oil recovery of Bohai oilfield by increasing the sweep efficiency of water and the oil displacement efficiency of surfactant synchronously. The surface topography and particle size distribution of sub-microscale polymer microspheres were investigated through scanning electron microscopy and dynamic lighting scattering. In addition, plugging performance, oil displacement efficiency and the deep profile control process of combination system were studied by utilizing assembled parallel double cores models and core displacement experiments. The experimental results showed that the initial appearance of the sub-microscale polymer microspheres was regularly spherical; the particle size distribution was from 200 nm to 6 μm . The particle size of the sub-microscale microspheres after swelling was increased by nearly five times compared with the original particle size. The combination system exhibited an effective injectivity and plugging effect for cores with different permeability from 0.5 μm^2 to 3.0 μm^2 . This system can significantly reduce fractional flow through high permeability channel, and also present good profile control effect and oil displacement efficiency in parallel double cores. By the injection of the 2.5 PV microsphere solution, the microspheres flooding recovery could improve about 34% more than water flooding. The slug composition, with 400 mg/L polymeric microspheres and 0.3% nonionic surfactant, could obtain a higher oil recovery by 4 wt% original oil than the microspheres system. The combination system can be recommended as a potential combination for profile control in Bohai heterogeneous reservoirs.

P-02

POLYMERIZABLE MICROSPHERE-INCLUDED HIGH MECHANICAL STRENGTH OF COMPOSITE HYDROGEL COMPOSED OF ACRYLAMIDE

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Polymerizable microspheres were introduced to prepare the high mechanical strength composite hydrogels with a novel honeycomb structure. Rheological properties, compressive stress-strain, tensile property and compression strength of both microspheres composite hydrogels and normal hydrogels without microspheres were investigated by HAKKE rheometer, TA.XT PLUS texture analyzer and compression strength test device. The test results illustrated that viscous moduli (G'') and elastic moduli (G') of composite hydrogel containing polymerizable microspheres (HPM) reached the maximum values. When the hydrogels were squeezed, the normal hydrogel was easily fractured under high strain (99%), whereas the HPM was not broken, and it quickly recovered its initial morphology after the release of load. The HPM showed excellent tensile properties, with an elongation at break up to 90% and a tensile strength greater than 220 g. The compression strength of the normal hydrogel was $100.44 \text{ kPa}\cdot\text{m}^{-1}$, while the resulting strength of HPM was enhanced to be $248.00 \text{ kPa}\cdot\text{m}^{-1}$. Therefore, the various performances of the normal hydrogel were improved by adding polymer microspheres. In addition, scanning electronic microscopic (SEM) was used to observe the surface topography of hydrogel samples and the SEM examination revealed that normal hydrogel has a general three-dimensional network structure, the conventional network structure did not exist in the HPM, it is a gel network structure similar to the honeycomb in the spherical projection, and the channels are very dense, which lead to the compaction of the space between the honeycomb network layers and reduced the flowing of free water wrapped in the honeycomb, therefore the mechanical strength of hydrogel was enhanced.

AN EMERGING THERMO-ELECTRIC GENERATOR DRIVEN BY TRAMPOLINING ELASTIC GELS

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In this research, we demonstrate an emerging thermo-electric generating technology based on Leidenfrost effect induced trampolining of elastic gel on hot surface,¹ which was occurred under a coupling effect from the elasticity of gel and Leidenfrost effect.² The thermal energy was transferred into electric energy through a Leidenfrost activated mechanical motion, supported by a self-constructed piezoelectric-hydrogel system. After carefully optimizing the system by selecting gel spheres with different solvent concentrations, sizes (4 mm – 25 mm), elasticity (0.5 kPa – 500 kPa), and studying other factors such as mass loss, impact dynamics, etc, we achieved a conversion unit to produce an optimum 3V and expected to get higher output (> 30V) by integrating a number of same units. This finding opens a window to create an energy-harvesting device that can generate electrical power from thermal sources and offer a potential solution for those regions with rich geo-thermal resources to acquire electric energy, such as Iceland or volcanic islands.

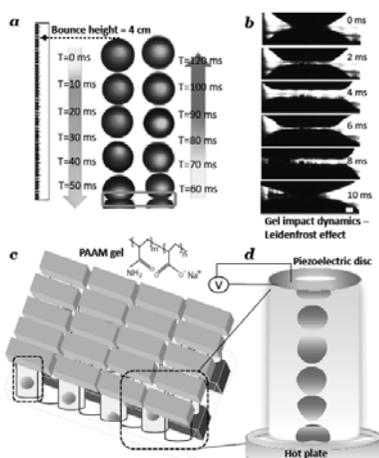


Figure 1. a) Elastic trampolining of gel with Leidenfrost activation occurred at b) the scale bar is 1mm; c) A conceptual integrated thermal energy harvester design with single electric generator unit in d), the inset shows molecular structure of PAAM

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SYNTHESIS OF MAGNETIC NANOCOMPOSITES WITH TUNABLE EPOXY MATRIX

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Smart polymer nanocomposites (PNCs) based on elastomeric epoxy matrix with exchangeable covalent bonds (vitrimers) and iron oxide magnetic nanoparticles (MNPs) were prepared. The multi-functionality of these PNCs is due to the combined effect of a tunable epoxy matrix and magnetic properties of embedded MNPs. The MNPs can induce change of the epoxy material shape and generate a local anisotropy of mechanical properties in a particular area of elastomeric nanocomposites in order to increase the strength of this area in the predetermined direction. One of the main parameters affecting the properties of PNCs is the strength of interactions between the particles and the matrix, which can be tuned by introduction of different functional groups on the surface of the particles. In this study, we evaluated PNCs with MNPs containing hydroxyl, phenyl and anhydride functional groups. Hydroxyl and phenyl groups provide non-covalent interactions (hydrogen bonding, Van-der-Waals) interactions with matrix, whereas anhydride groups form covalent bonds with polymer. PNCs were prepared by curing reaction of the diglycidyl ether of bisphenol A and a mixture of fatty dicarboxylic and tricarboxylic acids in the presence of modified MNPs and zinc acetate as transesterification catalyst. It was shown that most pronounced effect of MNPs is observed in the case of covalently bonded MNPs.

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P-05

MANUFACTURE AND CHARACTERIZATION OF PIEZOELECTRIC COMPOSITES USING PAR/PVDF SHEATH-CORE FIBER

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Piezoelectric materials have a wide range of applications as materials that can obtain electrical energy from mechanical energy such as pressure or vibration.¹ PVDF is a representative piezoelectric polymer material exhibiting the largest permittivity. However, the piezoelectric material using only PVDF has a relatively low strength.² In this study, thermoplastic composites were prepared using poly(vinylidene fluoride) (PVDF) having piezoelectric property and highly oriented liquid crystal polymer Polyarylate (PAR). The PAR/PVDF sheath-core fibers were prepared by melt conjugate spinning for manufacture of the PAR/PVDF thermoplastic composites. The PAR/PVDF piezoelectric composite was prepared by compression molding of the PAR/PVDF sheath-core fiber. The composite was molded by hot-press under various molding condition and materials were analyzed to determine whether the properties and piezoelectricity of the composites were related to the molding conditions. The piezoelectric properties of the composites were confirmed by various polarization conditions.

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P-06

HYDROGENATION OF *P*-NITROBENZOIC ACID BY GOLD AND PALLADIUM NANOPARTICLES IMMOBILIZED WITHIN MACROPOROUS AMPHOTERIC CRYOGELS IN AQUEOUS SOLUTION

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Aqueous solutions of sodium salt of *p*-nitrobenzoic acid (*p*-NBA) were hydrogenated in mild conditions in the presence of palladium (PdNPs) and gold nanoparticles (AuNPs) immobilized within macroporous amphoteric cryogel based on N,N-dimethylaminoethylmethacrylate (DMAEM) and methacrylic acid (MAA). Both cryogel matrix DMAEM-*co*-MAA and macroporous catalyst DMAEM-*co*-MAA/AuNPs were characterized by SEM. The pore sizes of sponge-like porous structure of cryogels are in the range of 40-80 μm . The sizes of AuNPs immobilized within macroporous amphoteric cryogel are varied from 3 to 10 μm . The reduction of *p*-NBA to *p*-aminobenzoic acid (*p*-ABA) and further to by-products sodium 4,4'-(diazene-1,2-diyl)dibenzoate and sodium 4-(4-aminobenzamido)benzoate was confirmed by Raman and ¹H NMR spectroscopy. The kinetic parameters and activation energies of hydrogenation of *p*-NBA have been calculated. The application of amphoteric macroporous cryogels with immobilized metal nanoparticles as effective flow-through catalytic reactor for one-step synthesis of aminoaromatic compounds has been realized for the first time.

Acknowledgement: *Financial support from the Ministry of Education and Science of the Republic of Kazakhstan (IRN AP05131003, 2018-2020) is greatly acknowledged.*

TRANSPORT PROPERTIES OF THE HYDROGEL THIN FILM ELECTRODEPOSITED ON CONDUCTING SURFACE

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Intelligent hydrogel layers, due to their unique properties, can be used in the construction of switchable ON-OFF electrodes or logic gate systems. The understanding of the mechanism of mass transport through the hydrogel layer under the different condition is a crucial issue in all applications of modified electrodes. We obtained temperature and pH sensitive electrodes modified with poly(N-isopropylacrylamide) (pNIPA) or sodium polyacrylate (pAS) layers, respectively. The pAS and pNIPA gels were deposited on micro- and regular-electrode surface via electrochemical induced free radical polymerization.

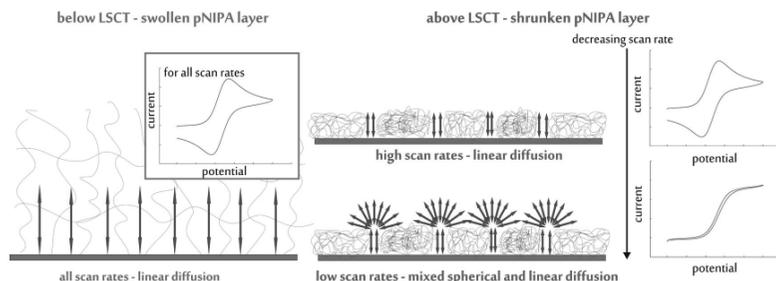


Figure 1. Scheme of mass transport through the electroinactive hydrogel layer in swollen and shrunken state to electrode surface and corresponding voltammograms.

Transport of charged probes to the surface of an electrode modified with a pH-sensitive polymer network was investigated. It was found that charging of the hydrogel film could act as an electrostatic barrier for negatively charged probes and could be penetrable for positively charged redox probe. For electrodes modified with pNIPA layers it is possible to obtain different voltammetric responses, depending on the structure of the layer and the scan rate. Transport of external redox probe was investigate for swollen and shrunken state of pNIPA hydrogel layer.

MODIFICATION OF GOLD ELECTRODE WITH MONOLAYER OF ENVIRONMENTALLY SENSITIVE MICROGELS

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Modification of the surface of an electrode with a polymer layer was frequently done to achieve desired electrochemical and physicochemical properties. A new trend in this activity is formation of the so called intelligent interface; it can be obtained by deposition of a layer of an environmentally-sensitive hydrogel on the electrode surface. The attachment of smart gels to the surface of an electrode widens their usefulness in, for instance, the construction of switchable sensors/biosensors, switchable electrochemical systems and signal-responsive interfaces.¹⁻³

We modified a surface of gold electrodes with monolayer of environmental sensitivity microgels. For this purpose the microgels contained disulfide bonds were synthesized using the surfactant-free emulsion polymerization. Thanks to the strong interactions between gold and sulfide atoms (presented in microgels) the strongly adhered monolayer of the microgels on the gold electrode was obtained. Additionally, introduction to microgels conducting polymer, metal nanoparticles or electroactive groups lead to obtaining new electrochemical systems. Influence of changes in environmental conditions on electrochemical properties modified electrodes will be presented.

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SKIN COLLAGEN FIBER/POLYURETHANE BIOCOMPOSITE WITH WATER-INDUCED SHAPE MEMORY ABILITY

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Most of the existing reported shape memory polymers (SMPs) have focused on sole thermo-responsiveness.¹ To realize the application of SMPs in mild conditions for special applications in biomedical and soft robotics fields, non-thermal stimulus such as water, pH values, acids and salts are required.^{2,3} In this study, skin collagen fiber/polyurethane (SCF/PU) biocomposite was prepared by a green approach resembled paper-making. The obtained SCF/PU possesses interpenetrating network where collagen fiber provides stiffness while PU matrix offers elasticity. Compares to pure PU, SCF/PU biocomposite showed significantly increased hydrophilicity due to the existence of collagen. Notably, the combination of reversible formation and disruption of hydrogen bonds within SCF in the elastomeric PU matrix leads to a shape memory effect that can be easily induced by water. Meanwhile, SCF/PU possess desirable flexibility and strength which can be modulated by controlling the SCF content. It is believed that this type of biocomposite can be applied as sensors or biomedical materials.

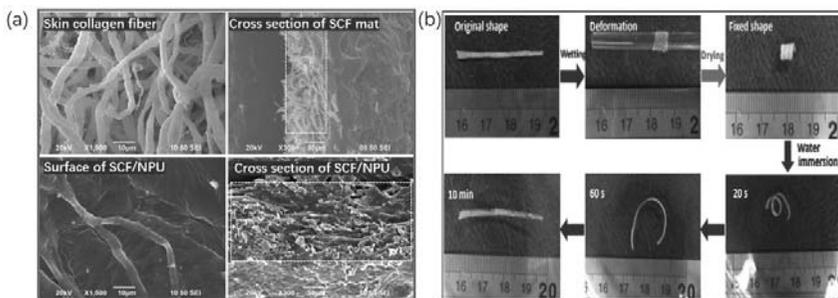


Figure 1. (a) SEM images of SCF and SCF/PU; (b) water induced shape memory effect of SCF/PU biocomposite

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THE ROLL OF ORGANOMETALIC SOL-GEL FILMS
FOR STRETCHABLE TRANSPARENT ELECTRODES

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As demands on wearable and biomedical devices increase, stretchable transparent electrodes have been regarded as the fundamental part for stretchable optoelectronic devices. Although lots of researches have been reported transparent electrodes with silver nanowire (AgNW) on stretchable polymer substrates like polyurethane and polydimethylsiloxane, the adhesion property between AgNW and polymer substrates has been still challenging.¹ In this work, we adopted titanium oxide (TiO₂) sol-gel films as a buffer layer through simple coating and heating in 60°C. The fabricated stretchable electrodes show sheet resistance of 14 Ω/□, transmittance at 550 nm of 83 %, and the surface roughness of around 1nm, which are similar with those of conventional ITO substrate. Also, the fabricated electrodes with the buffer layer showed enhanced stretchability compared with a reference without the buffer layer. The roll of titanium oxide buffer layer for AgNW-based stretchable electrodes will be focused.

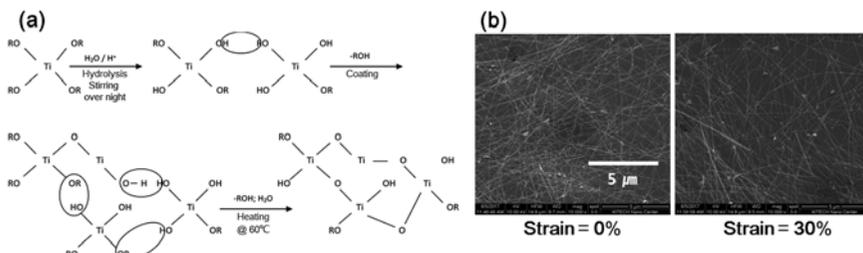


Figure 1. (a) The utilized titanium oxide sol-gel reaction starting from titanium isopropoxide, (b) the fabricated stretchable transparent electrodes with silver nanowires after stretching tests

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P-11

DYNAMICS OF SEMI-FLEXIBLE FILAMENT IN VISCOELASTIC MEDIA: MICRORHEOLOGY USING SEMIFLEXIBLE POLYMER

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The mechanics and dynamics of soft matter and living system often cannot be described by the classical physical framework of flexible polymers or rigid rods. The basic components are semi-flexible polymers, whose bending stiffness leads to nontrivial dynamics and mechanical response. Here, we investigate the bending dynamics of single-walled carbon nanotubes (SWNTs), which are model semi-flexible polymers with non-photobleaching fluorescence. We embedded SWNTs in viscoelastic media and analyze the thermally driven fluctuations in their shape. We discuss how the bending dynamics of SWNTs embedded in soft media can be used to probe the viscoelastic properties of such media.

P-12

A VINYLIC ROTAXANE CROSS-LINKER TOUGHENING NETWORK POLYMERS VIA RADICAL POLYMERIZATION OF VINYL MONOMERS

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Rotaxane-cross-linked polymers (RCPs) having the rotaxane structures at the cross-link points have been attracting considerable attention due to their unique properties such as high swellability, extensibility and well stress-relaxing ability, caused by the movable cross-link points.^{1,2} In this paper, we describe the synthesis of [2]rotaxane cross-linker (RC) having a methacryloyloxy group as the polymerizable group on each component and its application to the vinyl polymer-based RCPs.

Butyl acrylate-based RCP was obtained by the radical polymerization in the presence of a small amount of RC. Covalently cross-linked polymer (CCP) was also obtained by using 1,6-hexanediol dimethacrylate as covalent cross-linker for comparison. Resulting RCP and CCP were characterized by DSC, swellability, and tensile strength. The stress-strain curves of RCP and CCP showed much larger fracture stress and strain of RCP than those of CCP, even though Young's moduli of RCP and CCP were almost same. The results suggested that movable cross-link points enhance mechanical property of cross-linked polymer.³

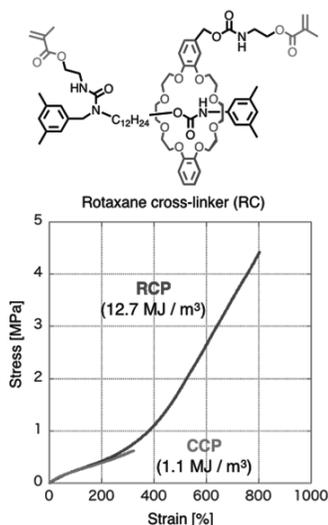


Figure. Stress-strain curves of RCP and CCP

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P-13

“QUENCHED” POLYAMPHOLYTE HYDROGELS BASED ON (3-ACRYLAMIDOPROPYL)TRIMETHYL AMMONIUM CHLORIDE AND SODIUM SALT OF 2-ACRYLAMIDO-2-METHYL-1-PROPANESULFONIC ACID

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A series of polyampholyte hydrogels (PAHs) based on fully charged cationic monomer (3-acrylamidopropyl) trimethylammonium chloride (APTAC) and anionic monomer sodium salt of 2-acrylamido-2-methyl-1-propanesulfonic acid (AMPSNa) in the presence of N,N-methylenebisacrylamide (MBAA) were synthesized by free radical (co)polymerization. The physico-chemical, thermal and mechanical properties of PAHs have been studied by FTIR, TG, DTA, DSC and mechanical testing. The isoelectric points of PAHs were determined by swelling experiments. The sorption and desorption ability of PAHs was evaluated with respect to dyes and surfactants. The swelling degree of PAHs at 20 mol.% of MBAA changes in the order: [APTAC]:[AMPSNa]=25:75 > [APTAC]:[AMPSNa]=75:25 > [APTAC]:[AMPSNa] = 50:50 mol.%. The mechanical properties of PAHs depend on the composition of copolymers. The Young modulus changes in the following order: [APTAC]:[AMPSNa]=75:25 > [APTAC]:[AMPSNa]=50:50 > [APTAC]:[AMPSNa]=25:75 mol.%.

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TEMPERATURE-SENSITIVE DOUBLE NETWORK HYDROGELS

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We used concept of double-network (DN)¹ to design thermoresponsive hydrogels composed of two highly asymmetrically crosslinked components, the tightly crosslinked first network of thermoresponsive poly(*N*-isopropylacrylamide) (PNIPAAm) or poly(*N,N*-diethylacrylamide) (PDEAAm) and the loosely crosslinked second network of hydrophilic polyacrylamide or poly(2-acrylamido-2-methyl-1-propanesulfonic acid). A series of DN hydrogels were synthesized with different crosslinking densities of the first network. Swelling, DSC experiments and NMR spectroscopy were combined to examine parameters of the phase transition, such as volume phase transition temperature (VPTT), enthalpy change, fraction of monomeric units directly involved in collapsed globular structures and portions of bound and released water. The behavior of the prepared DN hydrogels was compared with the behavior of one-component single networks (SNs).

It was found that presence of second hydrophilic network in DN hydrogels has a strong impact on their behavior. Both, deswelling with increasing temperature and specific enthalpy of demixing in DN hydrogels are strongly reduced relative to corresponding SN hydrogels. The hydrogels with the higher crosslinking density tend to swell less and their VPTT is shifted to lower values. Detailed analysis of NMR spectra and their variable temperature dependencies showed that at lower temperatures most PNIPAAm or PDEAAm units are involved in tightly crosslinked network regions and thus they are significantly reduced in their mobility.

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P-15

HYBRID HYDROGEL BASED ON κ -CARRAGEENAN AND METHACRYLATES FOR BIOMEDICAL APPLICATIONS

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The blending of natural and synthetic polymers allows developing new biomaterials that exhibit combinations of properties such as high gel strength, biocompatibility and biodegradability that could not be obtained from individual polymers.¹ Hybrid hydrogels were obtained by the addition of κ -carrageenan (κ C) to methacrylate monomers to obtain biocompatible hydrogels and to improve mechanical and swelling properties. The swelling ratio of hydrogels was investigated and the results indicated that the incorporation of κ -carrageenan produces an appreciable increase in the swelling behavior, which increased with the amount of κ C, compared to synthetic hydrogel. The morphologies of the surfaces of hydrogels showed that the addition of κ -carrageenan favours the formation of larger and more defined pores. The mechanical properties indicated that the addition of κ C improves the mechanical properties of synthetic hydrogel reaching up 400 N compression load. Therefore, the addition the κ -carrageenan to synthetic hydrogels improves both mechanical and swelling properties. The biocompatibility of the prepared hydrogels was evaluated and the results indicated that could be considered as candidate for biomedical use, such as tissue engineering and drug delivery.

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P-16

STUDY ON THE CURE BEHAVIORS OF THE MODIFIED EPOXY RESINS AND MECHANICAL PROPERTIES OF ITS CARBON FIBER COMPOSITES

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Among the thermosetting resins used as a matrix of carbon fiber reinforced composite materials, epoxy resins have excellent properties such as excellent chemical resistance, specific strength and specific modulus. As a representative epoxy resin, the DGEBA (diglycidylether of bisphenol A) type epoxy has been used for many applications. However, its fiber composite has relatively low properties of flexural strength and interfacial shear strength, and the long curing time of the epoxy resin restricts widening of its applications. Therefore, in this study, tetra-functional epoxy resin was blended to the DGEBA epoxy to improve the interfacial strength and also, a rapid curing agent was added to reduce the curing time of the resin. Curing behaviors and the thermal properties of the modified epoxy resins were analyzed according to each mixing ratio. Changes in viscosities of the modified resins were investigated with temperature. The carbon fiber reinforced epoxy composites were prepared by a compression molding process. The mechanical properties of the Carbon/Modified Epoxy composites were analyzed according to their mixing ratios. Final results were analyzed and discussed with mixing ratios and curing conditions of the modified epoxy resins.

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P-17

SURFACE STRUCTURE ANALYSIS OF SILICONE HYDROGEL BY NEUTRON REFLECTIVITY MEASUREMENT

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Contact lenses are the most frequent example of industrial use of hydrogels. Current mainstream of its materials is the so-called “silicone hydrogel” which is copolymer of silicone-based monomer used for a hard lens and hydrophilic components. Low biocompatibility of the hydrophobic silicone component is a deficiency although silicone possess high permeability of gases. Therefore, control of the interfacial properties between the lenses and tear film becomes particular an important issue.

The precise analysis (in nanometer order) of gels under water swelling near the surface (interface) is a rather difficult generally because of rough surface. Various silicone hydrogels swollen with heavy water set on the flat surface of silicon wafer, which made the interface between the wafer and the gel flat. We conducted the neutron reflectivity measurement (NR, BL16 SOFIA, J-PARC) in order to reveal water content near the gel surface (actually interface between the wafer and the gels). Neutron beams was incident from silicon substrate side, traveled in the wafer, and reflected the interface. The reflectivity depended on the amount of heavy water in the gels.

It was confirmed that water rich layer near the surface of treated silicon. It was pointed out the possibility of detecting the difference in the depth of moisture which shows the increase in moisture content and the change near the surface.

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E-BEAM CROSS-LINKING OF COLLAGEN-CO-POLY(VINYLPYRROLIDONE)-POLY(ETHYLENE OXIDE) TRIBLOCK SUPERABSORBANT HYDROGELS

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In the present study, superabsorbant hydrogels based on collagen–polyvinylpyrrolidone (PVP)–polyethylene oxide (PEO) were cross-linked by electron beam irradiation in aqueous solution and in the absence of oxygen. The effect of irradiation dose and the PEO concentration on hydrogels properties was investigated. The sol-gel analysis was used to determine gelation dose, correlated with radiation and chemical cross-linking yields. The hydrogels network structure and their composition was investigated using equilibrium swelling degree, rheological analysis and FT-IR spectroscopy. Rheological analysis was performed to determine the elastic (G') and viscous (G'') moduli, the average molecular weight between cross-links (M_c), cross-link density (ϱ_c) and the mesh size (ξ).

The hydrogels showed superabsorbant capacity up to 7000 % and high gel fraction, above 94%. The radiation-chemical yields increased as function of absorbed dose and the cross-linking processes predominated, $G(X) > G(S)$. The magnitude of G' and G'' moduli was strongly depended on the applied radiation dose and PEO concentration, were $G' = 9800$ Pa at 0.25 % PEO and 25 kGy.

The network parameters (M_c) and (ξ) have decreased with absorbed dose, while the ϱ_c increased as function of absorbed dose, confirming that the triblock polymeric system have been cross-linked by e-beam irradiation.

DIRECT OBSERVATION OF SINGLE POLYMER STRAND BASED ON DOUBLE NETWORK STRATEGY

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Hydrogel is a polymer material containing large amount of water. Until now, many researchers tried to clarify structure of hydrogel to explain its unique properties. However, nano-scale structure is still not well understood due to difficulty on direct observation of thin polymer.

In this study, inhomogeneous nucleation, nucleation starts from the functional group, was used to stain polymer network of hydrogels. If we carried out inhomogeneous nucleation in the hydrogel, mineral crystals easily forms complex with hydrogel polymer network (Fig. 1) and this mineral distribution should represent the original polymer structure. Therefore, the purpose of this study is to stain hydrogel polymer network with mineral and to observe its nano-scale structure by using transmission electron microscope (TEM).

Double network (DN) hydrogel, which contains sulfonic functional group, was synthesized by 2-step UV radical polymerization.¹ Then, Bernalite (Fe(OH)₃), whose nucleation accelerates by sulfonic group, was mineralized into DN gel. Nano scale network structure was successfully observed (Fig. 2) and this structure well agreed with previous prediction from neutron scattering. The detailed results will be presented in conference.

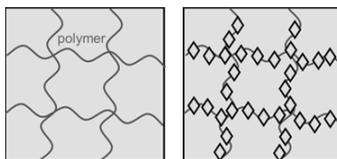


Figure 1. Schematic illustration

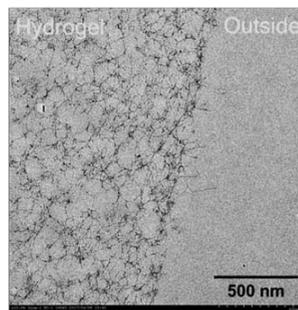


Figure 2. TEM image

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**INFLUENCE OF OLIGODIOL CONTENT AND CHAIN LENGTH
ON PROPERTIES OF BETA-CYCLODEXTRIN/OLIGODIOL
CROSS-LINKED POLYURETHANES**

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Nowadays cyclodextrin (CD) based polymers are broadly utilized in multiple areas (e.g. as sorbents) including macromolecular design. Previously, a possibility of synthesizing polyurethane (PU) elastomers with a predetermined degree of cross-linking based on β -CD as a cross-linking agent and various oligodiols was demonstrated.¹ It is of a certain importance to investigate the dependence of the properties of CD-based cross-linked PUs from the length of linear segments and from the CD/oligodiol ratio. For these purposes PUs comprising β -CD and polyethylene glycol (PEG) and polypropylene glycol (PPG) of molecular masses from 250 to 20000 were obtained and their properties were studied. Molecular masses of soluble CD-containing samples are 2-3 times greater than of pure PEG (PPG) PUs, which confirms that CD is introduced into polymer chains. Glass-transition temperatures (T_g) decrease as length of oligodiol segments increases. T_g reaches -50°C for PUs containing PPG-2025 and PEG of weights more than 4000. Stress-strain properties of a series of samples correspond to typical elastomers: Young's moduli of 10-18 MPa, elongations of 200-400%. However, there appears to be a tendency of turning from elastic to brittle as oligodiol chain length increases. The work was supported by Russian Foundation for Basic Research (grant 16-29-01041 ofr-m).

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VOLUME TRANSITION OF POLY(OLIGO ETHYLENE GLYCOL METHACRYLATE)-BASED HYDROGELS

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Polymer hydrogels are soft and water-swollen materials. Physicochemical properties can be changed greatly by the stimuli-responsiveness of hydrogels, which make hydrogels promising materials for the advanced applications. Recently, we discovered that new types of hydrogels allow the selective separation and release of halogen-containing compounds.¹ The hydrogels are based on water-immiscible poly(2-methoxyethyl acrylate) as the halogen bonding acceptor, which is incorporated in swollen hydrogels of poly(oligo ethylene glycol methacrylate), pOEG (Figure 1). Due to the properties of pOEG gel matrix, it is thus possible to control the amount of adsorbed halogen-containing compounds that is released.

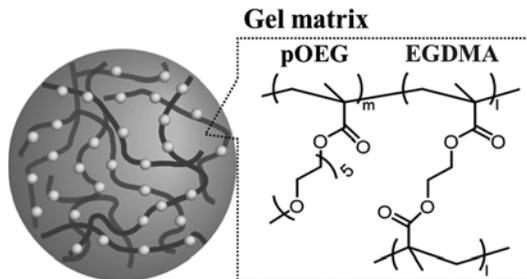


Figure 1. The scheme and chemical structure of pOEG hydrogels

However, the release of halogen compounds was not observed when the other thermo-responsive gel species were used as the gel matrix. The release mechanism for the adsorbed halogen compounds as well as the thermo-responsive behavior of pOEG gels is still unclear. In this study, in order to quantitatively evaluate the volume phase transition behavior of pOEG bulk gels, we perform DLS measurements on a series of the hydrogels at different temperatures.

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**ROLE OF DISTINCT MICROSTRUCTURES
ON THE RHEOLOGICAL BEHAVIOR OF PECTIN-Ca GELS**

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Pectin, a polysaccharide widely used in food industries, is an important constituent of plant cell walls. It can form an ionically crosslinked gel in the presence of divalent cations like calcium. It is known that, based on the concentration of divalent calcium ions, the gel structure that is formed, differs. At low Ca^{2+} concentrations, the gel microstructure predominantly consists of egg-boxes, which are crosslinks formed by parallel alignment of two chains, as well as aggregates of such structures.

However at high Ca^{2+} concentrations, monocomplexes of single pectin chains form the predominant microstructure. We observe that the rheological behavior shifts from one type to another, as the gel microstructure goes from the former to the latter by increasing the calcium ion content. In the presence of higher number of egg-boxes, application of an oscillatory shear deformation leads to rearrangement of microstructures at intermediate strain amplitudes, characterized by an overshoot in the loss modulus. Strain stiffening as well as negative normal stress appear as a consequence of the aggregated structure formed. These characteristics of the gel progressively disappear as the egg box content decreases. Modifications done to the networks by salt addition confirm the role played by egg-boxes in the deformation behavior observed.

SPEEDING UP THE THE STUDY OF DIPOLAR SYSTEMS GELATION

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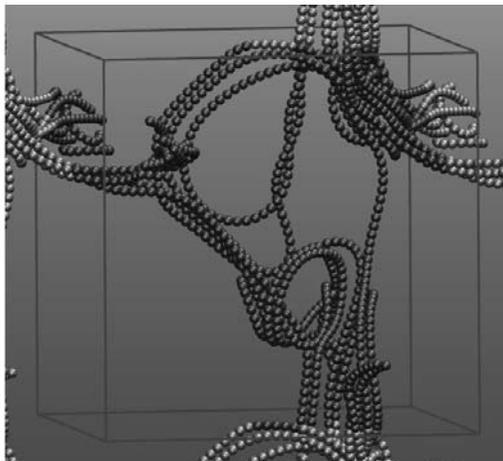
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We study the structure changes of a diluted dipolar system during gelation and cristalization through Monte Carlo simulations in the NVT ensemble.

In order to speed up the dynamics, several approximations and accelerated algorithms are proposed and tested. Originally, it turns out that “Chain Move” algorithm is roughly one decade faster than the well known and typical Monte Carlo. We find regimes similar to an anti-parallel crystal, a gel, a short-chain-liquid, a ring-liquid and a gas. Such regimes are studied and characterized through positional, orientational and thermodynamical observables.



**DESIGN OF A REACTIVE POLYVINYLALCOHOL GEL
AND RIGID MACROPOROUS SORBENTS FOR BIOMEDICAL
APPLICATION**

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A novel method for poly(vinylalcohol)-based macroporous network (MP-PVA) preparation in aqueous media without usage of an outer porogens has been elaborated.¹ The method includes selective oxidative cleavage at 1,2-diol fragments of PVA by periodic acid and self-polycondensation of generated telechelic oligomers followed by crosslinking with an excess of glutaraldehyde (GA). MP-PVA represents a highly crosslinked matrix that is non-swelling in water or organic solvents. It has a high pore volume and high pore surface, and contains reactive aldehyde groups. The rigid polymer network is characterized by heterogeneous globule-like structure with open macropores and large through pores. Light scattering study has elucidated the phase separation during the first self-crosslinking step as the main factor responsible for macroporous structure generation. The two-phase structure of gel is fixed on the second step by crosslinking with GA. The prepared macroporous network has been successfully applied as the parent platform for preparation of a boronate affinity sorbent useful for differentiation and quantification of glycosylated and non-glycosylated haemoglobin in human blood of diabetics patients. In addition, the modification of affinity sorbent by magnetite nanoparticles was performed. The magnetically-controlled microparticles were used for the extraction of oligosaccharides from aqueous media due to their reversible adsorption onto the affinity sorbent.

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DEGRADABLE PEG-HYDROGELS

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Despite its outstanding properties, the use of poly(ethylene glycol) (PEG) for biomedical applications is limited to molecular weights up to 30,000 g mol⁻¹ to permit renal excretion.¹ This major drawback can be overcome using cleavable PEG, which must be degradable under physiological conditions. Recently, our group established the copolymerization of ethylene oxide (EO) and 3,4-epoxy-1-butene (EPB) by anionic ring-opening copolymerization (AROP), resulting in multi allyl ether-functional PEG.² In this work we present the synthesis and characterization of P(*iso*EPB-*co*-EG) copolymers via AROP. As these polymers are obtained via controlled polymerization, well-defined hydroxyl end groups are obtained. Various novel polymer architectures based on degradable PEG can be achieved using different multifunctional initiators. The polymers are subsequently functionalized and crosslinked to generate hydrogels that are stable under neutral conditions and show rapid hydrolysis at physiologically relevant pH. The strategy is applicable to macroscopic hydrogels as well as nanoscopic structures.

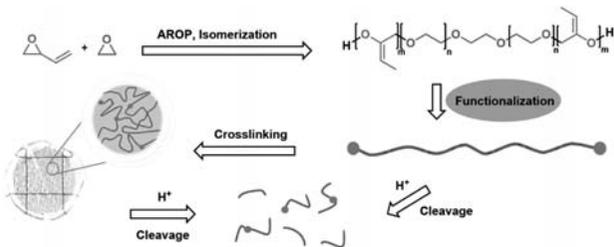


Figure 1. Synthesis and functionalization of P(*iso*EPB-*co*-EG) resulting in acid labile hydrogels.

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METHACRYLATE-BASED HYDROGELS FOR TRANS-SCLERAL ADMINISTRATION OF TOPOTECAN AND VINCRIStINE IN RETINOBLASTOMA THERAPY

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Retinoblastoma is the most common primary intraocular malignancy in childhood.¹ It is a tumor which arises from the stem cells of the developing retina. There are many treatment options for retinoblastoma but the best scenario would be the local application of an active chemotherapeutic agent.² Nowadays, scientists have studied different approaches for the delivery of chemotherapeutic agents into the vitreous humor of eyes with tumors.³ Materials, such as liposomes, lipid nanoparticles, and hydrogels, have been studied for sustained drug release.^{2,4}

The aim of this study was to develop new hydrogel implants for trans-scleral drug administration. The 2-hydroxyethyl methacrylate (HEMA) based hydrogels were synthesized and tested for their capacity of sorption and release properties for two anti-carcinogenic drugs, topotecan (TPT) and vincristine (VCR). HEMA based hydrogels were prepared with various cross-linker degree and with addition of various functional groups to the polymer backbone. It was observed that the addition of different comonomers to HEMA doesn't improve the sorption capacity of the hydrogels. The release profiles of both drugs showed two steps, first a burst release of the drugs in the first hours followed by a slow sustained release in the following days. In both cases the sorption capacity, as well as the release kinetics are in accordance with therapeutic requirements.

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**BIO-INSPIRED CORONA FORMATION MIMICKING
THE OCULAR ENVIRONMENT FOR CONTROLLED *IN VIVO*
THERAPEUTIC APPLICATION OF POLYMER PARTICLES**

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Polymer particles adsorb biomolecules to form corona when they enter the biological environment.¹ In the vitreous of the eye, the composition of the corona is determined by the electrostatic and hydrophobic properties of the associated proteins, regardless of the core material or size of the particles. This phenomenon is different from that in serum or plasma, demonstrating tissue-specific corona formation. To assess the biological impacts of tissue-specific coronas, we incubated polymer particles with the 5 most common proteins in vitreous-based coronas. Priming polymer particles with tissue-specific corona proteins allowed them to exert their therapeutic effects *in vivo*. Our results suggest that controlled corona formation that mimics *in vivo* processes may be useful in the therapeutic use of polymer materials via local administration by inhibiting nonspecific, uncontrolled protein adhesion.²

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**POLYMER-DNA CONJUGATES WITH AMPLIFIED VEGF
APTAMERS INHIBIT RETINAL VASCULAR
HYPERPERMEABILITY**

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In this study, nanoconstructs consisting of a DNA-amplified aptamer with a biocompatible polymer backbone for capturing target biomolecules are presented. First, the polymer-DNA nanoconstructs were prepared by hybridization of two complementary single-stranded DNAs that were each conjugated to a dextran polymer backbone. The designed polymer-DNA amplified aptamer nanoconstructs (PA-aNCs) were then prepared by utilizing polymer-DNA nanoconstructs conjugated with an aptamer (PA-NCs) using a rolling circle amplification reaction to amplify the aptamer. These PA-aNCs were successfully applied to alleviate VEGF-induced retinal vascular hyperpermeability *in vivo* through the highly effective capture of human VEGF as a target molecule. These PA-aNCs could be used as therapeutic agent for anti-VEGF therapy by efficiently capturing human VEGF.

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NOVEL HYDROGELS FOR POST-OPERATIVELY ADJUSTABLE INTRAOCULAR LENSES

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Cataract surgery restores vision in millions of patients every year, but many are not satisfied because optical characteristics of the implanted intraocular lens (IOL) does not match their needs. Emerging technology of the femtosecond laser modification (FLM) aims to non-invasively correct optical characteristics of IOLs by precisely localized adjustment of the refractive index. However, many current IOL materials may not be suitable for FLM. In this work, we aimed to optimize synthetic hydrogels for high FLM sensitivity.

Several pHEMA-based hydrogels with various types and concentration of UV absorbers and other additives were modified by a femtosecond laser. Interferometry, Raman spectroscopy, fluorescence microscopy and chromatography were used to characterize the modified hydrogels.

Interferometry shows reproducible negative refractive index changes due to nonlinear absorption of femtosecond laser light. The induced refractive index change strongly depends on hydrogel composition. Raman spectroscopy indicates higher water content in modified areas.¹

We propose that some material components are essential for the nonlinear energy absorption (“dopants”), while other components (“quencher”) are beneficial in redirecting the absorbed energy to cause chemical reactions that change the polymer network and result in the change of the refractive index of the material.

The presented hydrogels promise IOLs with better postoperative adjustment.

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CARBOXYMETHYL CELLULOSE GEL SYSTEMS WITH SAPROPEL EXTRACT

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Carboxymethylcellulose salts (CMS) are well known as a thickener and binder in food industry, as lubricant for drilling in oil industry and as a stabilizer and binder in cosmetics. Because of its non-toxicity, biodegradability and biocompatibility, CMS solutions have been used as a carrier for biologically active substances.¹ In this study carboxymethylcellulose sodium salt was used to produce a carrier for sapropel extract. Sapropel has a unique structure with wide biological and biochemical variety depending on its origin. Its characteristics are determined by organic, mineral and biological compounds with a multitude of effects on skin.² Sapropel has a high heating capacity and it is a mixture of chemical elements, hormones, various organic acids and vitamins (C, B1, B2, B5, B6, B9, B12, E, D and P).³ Previously sapropel has generally been used in raw forms. To extract the biologically active substance from raw sapropel, it was subjected to alkaline solution. Alkaline extraction was followed by filtration, and the water present in the aqueous mixture was evaporated.

The sapropel extract was mixed in the CMS gel matrix, and the stability and obsolescence of the gel system was measured. The results demonstrated that after 24h there are no changes, but after 7 days the gel without preservatives shows signs of molding.

Additionally, a further evaluation of CMS-Sapropel gel system can help develop new a pharmaceutical gel to treat skin diseases and joint pain.

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THREE-DIMENSIONAL CARBON-BASED POLYMER SCAFFOLD FOR BONE TISSUE ENGINEERING

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Bone tissue engineering is devoted to the development of biocompatible materials suitable for the replacement of defect bone tissue.¹ Bones represent a very dynamic structure and its understanding is the basis for designing materials mimicking the extracellular matrix.² A porous 3D carbon based material was produced by thermal modification of polyacrylonitrile at 360°C with air access. Morphological structure is created by interconnected pores that were characterized by scanning electron microscope. Communicating pores of sizes suitable for the growth and proliferation of human osteoblasts were formed in the structure by incorporation and subsequent washing of the porogen particles - fractionate crystals of sodium chloride. The mechanical properties of the material were improved by introduction of a secondary interpenetrating chitosan network. Proposed material and its hydroxyapatite composite have been studied in biological experiments for their osteoconductivity. The materials proved to be non-cytotoxic and their chemical composition and morphological structure promote the adhesion, proliferation and growth of human osteosarcoma cell line MG 63 strain that formed a confluent layer on the scaffolds.

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NOVEL BIOSYNTHETIC CAF1-BASED HYDROGELS WITH POTENTIAL AS CELL SCAFFOLDS

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Caf1 (Figure 1a) is a polymeric protein obtained from the bacteria *Yersinia pestis* which shares its 3D structure with the majority of the mammalian extracellular proteins. The Caf1 protein polymer is formed by 15 kDa protein monomer subunits which donate a single β -strand to the preceding monomer in the chain, linking together the subunits through very strong non-covalent interactions into a continuous polymer chain. The “non-stick” phenotype characteristic of the protein can be modified by inserting a cell adhesion motif as RGDS, highlighting the capability to engineer the Caf1 polymer for cell culture purpose. These properties make Caf1 an interesting material from which to develop biomaterials for use as cell scaffolds.

Our approach to the preparation of hybrid hydrogels combines the benefits of using a biologically-active Caf1 protein with the features of synthetic polymers to obtain well-defined and highly tunable hydrogels which have potential as biomaterials for mimicking the extracellular matrix. We have cross-linked of Caf1 protein polymer with a selection of functionalised PEG polymer crosslinkers by mixing of the components under mild conditions to yield hydrogel. RGDS-variants of the Caf1 polymer showed biocompatibility with 3T3 fibroblast cells which bound to the surface of the gels. These preliminary results suggest Caf1-based materials are promising candidates in cell culture applications.

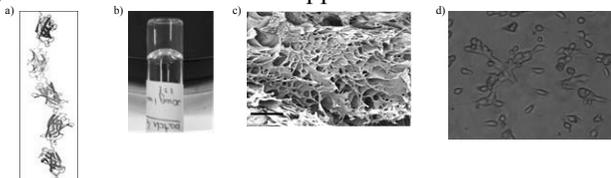


Figure 1. a) Modelled structure of polymeric Caf1 polymer. b) Picture of real hydrogel. c) Representative SEM images of hydrogel with 4-arm PEG crosslinker. Scales bars represent 50 μm . d) In vitro 2D cell culture studies using 3T3 fibroblast cells in the top of the hydrogel.

EXPERIMENTAL EVALUATION OF THEORETICAL MODEL OF IPN HYDROGEL ELASTICITY

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The model homogeneous hydrophilic methacrylate-based IPN hydrogels were prepared sequentially to evaluate the theoretical model of elastic behavior based on the two-networks hypothesis proposed by Andrews, Tobolsky, and Hanson.¹ The theory accounts for different coiling states of chains due to swelling of the first network in the second network monomer. The contributions are considered additive with no extra increase of internetworks entanglements. The first PHEMA network was prepared at 40 vol.-% of dilution and various crosslinker content (0.2 – 6 mol-%), while the second PHEMA network was embedded by swelling of monomer and crosslinker (0.3 to 2 mol-%) into the first network. The elastic modulus of equilibrium swollen IPN hydrogels was determined from tensile responses and the experimental values of the Young's modulus (E_{exp}) were compared with the calculated ones (E_{calc}). It was found that the experimental data fit the theory reasonably well, however with some limits: for the less crosslinked PHEMA first networks (<1 mol-% crosslinker), the E_{exp} was above the E_{calc} . Entanglements between interpenetrating networks are assumed to be the reason. For PHEMA first networks with >3 mol-% of crosslinker, the E_{exp} was below that of E_{calc} : the reason for this was the onset of the reaction-induced phase-separation manifested by appearance of turbidity.

Acknowledgement: *The authors acknowledge the financial support provided by the Czech Science Foundation – project No. 17-08531S.*

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MODELING SOLUTE DIFFUSION THROUGH A HYDROGEL

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We present a finite-difference model of solute diffusion between two chambers separated by a hydrogel membrane. We aim to model a specific experimental setup where the solute is initially in one chamber, and diffuses across a thin HEMA-based hydrogel membrane into the other chamber which initially contains pure solvent. Two different solutes investigated in the experiments (Topotecan and Vincristine) exhibit different dependence of concentration on time: an initial plateau is followed by a much faster decay at later times. Our goal is to understand and predict how the gel-solute interactions affect the concentration profiles. We study the influence of solute accumulation/depletion in the gel, and the ratio of solute diffusion coefficients inside the gel and in the bulk.

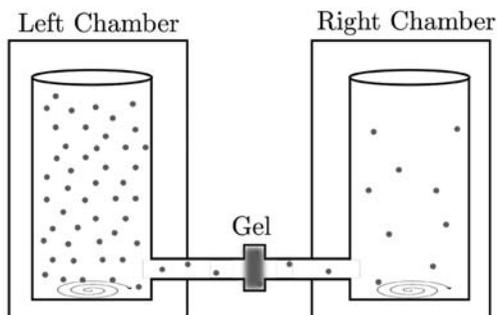


Figure 1. Schematic representation of the experimental setup.

**MECHANICAL PROPERTIES OF RANDOMLY ORIENTED
ELECTROSPUN FIBRE TEXTURE**

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Electrospun nano- and micro-fiber network as novel artificial extracellular matrix have attracted an intensive research area over the past decades. Although the electrospinning technology is well developed, little is known on the deformation mechanism of spun fiber networks. The strength and load bearing capacity of these materials are important factors for many biomedical applications. We have studied the fundamental mechanical properties of fibrous scaffold, prepared from poly(succinimide) and other polymer molecules. A home-made electrospinning instrument was used to prepare fibers from the polymer solutions. 2D randomly oriented fibre mats were prepared from the polymers. In order to elucidate mechanical behaviour of non-woven, stiff randomly oriented fibrous systems, unidirectional stress-strain measurements have been performed. The deformation mechanism including the occurrence of failure and rupture of fibres as well as the dependence of adhesion between fibres have been investigated. Whenever a fiber in the mat experiences a force equal to or greater than its strength threshold, it breaks and results in redistribution of the local stresses. Due to the complex interplay of non-bonded adhesive interactions and the load sharing behaviour of fibres, after deformation the electrospun texture can not fully recover its original dimensions. We have observed that the individual fibers obey Hookean elasticity right up to the breaking of the non-bonded interactions between the fibers and the load distributes itself among the surviving neighbouring fibers. As a result, the load-displacement curve of fiber mats show a maximum type dependence with many abrupt changes in the stress.

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FERINGA TYPE ENGINES IN POLYMER MODEL SYSTEMS: FOLDING, COILING, MOLECULAR STIRLING ENGINES, AND ACTIVE GELS

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We use the bond fluctuation model to study the contraction process of different polymeric model systems with attached Feringa engines. For these engines, the top part of the molecule rotates unidirectional with respect to the bottom part upon irradiation with light. For a figure of 8 shaped model system, we study a topology where each of the two attached strands is connected to bottom and top part of the engine. For this setup, a contraction process is observed that consists of two steps: folding followed by coiling. Double stranded polymers are studied in a slit geometry where either both strands connected to bottom and top of engine are connected to the bottom and top wall respectively (X geometry) or one strand at each end of the engine connects to the bottom wall while the second strand on this end connects to the top wall (H-geometry). We show that the engine in these two geometries exhibit distinct work cycles that allows to consider a Stirling machine that is either efficient at a small wall separation (H-geometry) or at a large wall separation (X-geometry). This difference highlights the relevance of the particular connectivity for the function of the engines. A random connectivity is studied by inserting the engines into the centers of 4-functional stars in star polymer nano-gels. These gels shrink upon irradiation with light as a function of the quantity of adsorbed light. The results of the first two model systems are used to elucidate the effectiveness of the engines inside the gel.

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THE ELASTICITY OF REAL POLYMER NETWORKS WITHOUT ENTANGLEMENTS

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We simulate 4-functional model networks at a different polymer volume fraction and with a different degree of polymerization between the cross-links. Entanglements are switched off by allowing the strands to cross at no change in excluded volume. Simple shear deformation is simulated to measure the bulk modulus of the networks. The local deformation of individual chains, the fluctuations of cross-links, and the segment orientations are analyzed to learn more about the relation between network elasticity and structure, in particular concerning the impact of finite cycles. Additional contributions by coupling of segment orientations via excluded volume are monitored by detecting the orientation of the inactive chains in a deformed network, which indicates that “inactive” chains still contribute partially to elasticity. The formation of finite loops decouples the cross-link fluctuations from elastic contribution of the attached strands such that considering fluctuations underestimates modulus. This comparison also reveals that there is some pre-load of the network chains that is generic for end-linked model networks.

**FLORY-REHNER AND SCALING DESCRIPTIONS
OF THERMORESPONSIVE PNIPAM GELS**

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Thermoresponsive polymeric gels are a class of smart materials which can absorb or release large amounts of solvent upon small changes in their environment. Two popular approaches to model the swelling of gels and microgels are the Flory-Rehner model, and scaling theories (de Gennes ν theorem and Obukhov's model). We examine data on linear and cross linked poly(N-isopropylacrylamide) (PNIPAM) in aqueous solutions of different molar masses for different degrees of cross-linking and solvent quality. While the power-law exponent for the swelling equilibrium as a function of degree of cross-linking density is close to the scaling and Flory-Rhener predictions, the swelling observed in cross linked networks is much larger than expected from the aforementioned models. We consider some possible reasons for the discrepancies observed, and conclude that network inhomogeneities may play a large role in the enhance swelling of polymeric networks.

**PRECISE SYNTHESIS AND THERMAL PROPERTIES
OF HOMOGENEOUS COPOLYMER GELS WITH DIFFERENT
MONOMER SEQUENCE**

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In general, polymer networks in gels have tangled, because the synthesizing process is extremely complicated in the reaction solution. The network structures including homogeneity, sequence of monomers, and cross-linking points were hardly controllable so far, despite those are deeply related to the properties of gels. It is one of the important problem to be solved clarifying the connection between macroscopic properties and microscopic networks.

Here we sequentially employed two types of preparative methods to obtain star-shape polymers using living radical polymerization for making controlled polymer networks. Firstly, star-shape polymers are synthesized by 'Core-first method'(Fig.1-a) using multi-fuctional initiator.

Secondly, with 'Linking methods'(Fig.1-b), the star-shape polymers are converted to the polymer networks by adding linking agents in one-pot. The combination of these methods facilitates to control the homogeneity and the sequence of monomers on the polymer networks.

In this report, we synthesized network controlled copolymer gels using the polymers exhibit temperature-responsiveness or not, composed of *N*-isopropylacrylamide (NIPA) and/or *N,N*-dimethylacrylamide (DMA). Furthermore, conventional inhomogeneous copolymer networks are simply synthesized by a free radical polymerization, because of using vinyl monomers. Static and dynamic thermal properties of these different gels were investigated.

In our presentation, we will discuss more details about the synthesis and the results of the static and dynamic thermal properties of these gels.

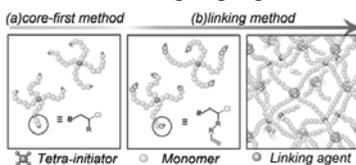


Figure 1. Combination of preparative methods synthesizing star polymers Core-first method (a) and Linking method (b) for constructing networks.

**CRYSTALLIZATION BEHAVIORS OF GLASS
FIBER/POLYAMIDE 6 COMPOSITES**

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Fiber reinforced composites have been applied in various industrial fields because of their excellent physical and mechanical properties. Especially, in the field of automotive, glass fiber (GF) reinforced polyamide 6 (PA6) composites have been studied for weight reduction and fuel efficiency improvement. Since the mechanical properties of the GF/PA6 composites are influenced by the crystallization of PA6, extensive knowledge of the crystallization behavior of PA6 is required. The nucleation ability of glass fibers plays an important role in the crystallization behaviors of the GF/PA6 composites among various parameters related to crystallization.

In this study, the isothermal crystallization behaviors of PA6 matrix in the GF/PA6 composites were conducted with a differential scanning calorimetry(DSC) and analyzed by Avrami's kinetic theory. Due to the heterogeneous nucleation, the incorporation of glass fibers improved the crystallization rate and the nucleation effect of the GF/PA6 composite to some extent. Microstructure and mechanical properties of the GF/PA6 composites were varied depending on the fiber content. Details of the crystallization behaviors of the GF/PA6 composites were analyzed and discussed according to the mechanical properties.

HIGH FREQUENCY DYNAMICS OF A LIQUID CRYSTALLINE, CYANOBACTERIAL, SULFATED POLYSACCHARIDE STUDIED BY DLS/DWS MICRORHEOLOGY

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A novel polysaccharide “sacran” was extracted from a freshwater unicellular cyanobacterium, *Aphanothece sacrum*. This polysaccharide, containing carboxylate and sulfonate groups, has an extremely high molecular weight (more than 10^7 g/mol). Aqueous solutions of sacran form huge domains of liquid crystals at very low concentrations (> 0.5 %), suggesting high rigidity of the chains. In order to characterize the sacran chain rigidity at a single chain scale, we studied microrheological properties of aqueous sacran solutions by dynamic light scattering (DLS) and diffusing-wave spectroscopy (DWS). The mean square displacement of probe particles was calculated from DLS and DWS autocorrelation functions. The complex modulus was calculated by using the generalized Stokes-Einstein relation. The dynamic shear moduli measured by microrheology show good agreement with those measured by classical mechanical rheometry. At high frequencies we found Zimm-Rouse mode showing $G^* \sim C^1 \omega^{5/9}$ as well as bending mode of the rigid polymer chain showing $G^* \sim C^1 \omega^{3/4}$. The persistence length, l_p , was determined from the transition frequency of the two modes. Odijk-Skolnick-Fixman theory suggests the intrinsic persistence length of 50 nm. The interchain distance was measured from high frequency dielectric relaxation of counterion. We found that at higher concentration the entanglement length was shorter than l_p , scaling as $G^* \sim C^{1.4}$.

COMPLEX VISCOSITY MODELING OF IONOGELS REINFORCED WITH CHITOSAN

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Chitosan is often used to improve properties of materials, such as biodegradability, biocompatibility, biocidal, etc¹. This carbohydrate can be used employed as mechanical reinforcement of materials², such as ionogels. The aim of this work is to model the complex viscosity of the ionogels with a matrix of 1-butyl-3-methylimidazolium chloride (BMIMCl) and cellulose, reinforced with chitosan with different deacetylation degree (DD).

The matrix of the ionogels was synthesized by dissolving microcrystalline cellulose in BMIMCl (6 wt. %) under magnetic stirring and N₂ atmosphere at 100 °C. A 3 % (w/w of matrix) chitosan reinforcement at different DD (54, 60, 69, 78 and 84 %) was added to the matrix. The gelling process took place in a climatic chamber for 5 days at 25 °C. The ionogels were characterized in an ARES rheometer through dynamic frequency sweep (frequency from 0.01 to 50 Hz, applying a 0.5 % strain at 25 °C). The Power-law, the Ellis, and Cross models were applied to the complex viscosity data.

The results showed a clear influence of the DD on the complex viscosity on the ionogels, since the complex viscosity increased as the DD was increased. This fact is due to a major interaction between the amine groups of the chitosan and the matrix of the ionogels.

The obtained materials presented a pseudoplastic behavior. The Ellis model was discarded because of physical meaningless values of its parameters. The Power-law and Cross models presented a suitable fit of the complex viscosity ($R^2 > 0.998$). However, the Power-law model exhibited the lowest standard deviation values ($\sigma < 0.438$).

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OSMOTIC ENGINE – ENERGY RECOVERY FROM SALT GRADIENTS VIA POLYELECTROLYTE HYDROGELS

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Polyelectrolyte hydrogels, like crosslinked poly(sodium acrylate) can swell up to 1,000 time their own weight in desalinated water. In seawater-like saline solutions (4.3 wt% NaCl), the degree of swelling is usually a factor of 2-12 lower. Hence, cycling swelling and shrinking can be used to move a piston in an osmotic engine.¹

This presentation addresses the energy recovery from solutions with different salinities. The poly(sodium acrylate) hydrogels were specially synthesized for this research to investigate systematically the influence of chemical composition and process parameters at three different self-constructed setups for the potential osmotic engines. The experiments were evaluated to find the best parameters for the highest power production.²

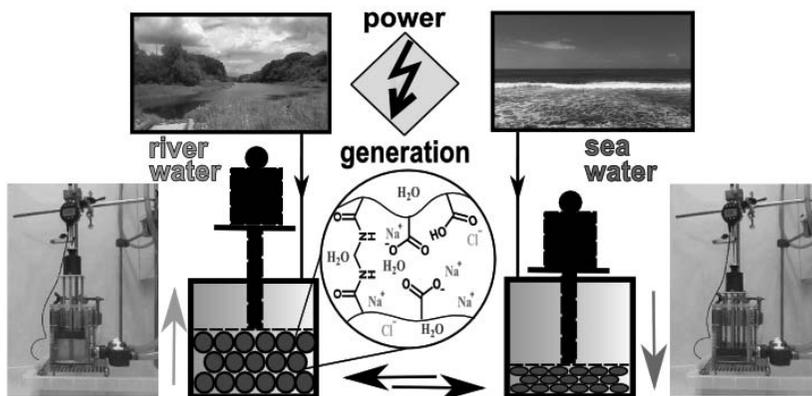


Figure 1. Energy recovery from salt gradients via swelling and shrinking of poly(sodium acrylate) hydrogels in an osmotic engine.

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SPATIOTEMPORAL FEATURES OF RESPONSIVE HYDROGEL MATERIALS CHARACTERIZED BY QUANTITATIVE PHASE CONTRAST MICROSCOPY

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Hydrogels, consisting of a cross-linked network immersed in liquid, adopt their swelling equilibria depending on molecular parameters of the network, topology, properties of the immersing bath and external fields. In the present contribution, we describe the application of quantitative phase contrast optical imaging (QPI) to determine properties of surface attached hydrogels such as thickness and creation of patterns when the hydrogel is subjected to conditions altering its swelling volume. The capacities of the QPI approach are illustrated employing acrylamide-based hydrogels with various molar concentrations of BIS fabricated in two layers by spin coating method. The bilayered structure consists of a stiff swelling layer with the cationic monomer N-(3-dimethylaminopropyl)acrylamide on top of a soft non swelling layer. These structures were investigated in solutions of different pH using a time-lapse feature. QPI renders high contrast images of samples that otherwise exhibit low contrast in common bright field imaging. The quantitative nature of this imaging modality allows measurements of refractive index and thickness with great precision. A precise phase measurement with sensitivity down to 1 mrad, can yield thickness change measurements of approximately 1 nm along the optical axis and sampled with lateral resolution corresponding to the numerical aperture of the microscope. In this way, QPI is a very precise tool for studying dynamics of swelling of hydrogel materials over broad temporal and spatial scales.

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SWELLING AND DEFORMATION RESPONSES OF POROUS HYDROGEL SIMULATED WITH FINITE ELEMENT METHOD

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Swelling and deformation responses of macromolecular hydrogels are important parameters in biological applications. Macroscopic deformation of simple geometry involves deformations of various geometries of pore walls dependent on object morphology. Local stresses and gradients of swelling exist even in undeformed state and are changed as a result of external deformation. The changes on microscale are important for applications, for instance, for cell cultivation in bioengineering. In this work, the behavior of hydrogel was modelled by finite element method (FEM). The model is developed for the calculation of equilibrium swelling and the deformation characteristics of swollen gels. The model is based on changes of Gibbs energy of a swollen network as described by a statistical-mechanical model of cross-linked polymeric hydrogel. Contributions to Gibbs energy due to polymer segments – solvent molecules mixing (Flory-Huggins type) and due to stretching of network chains of finite extensibility (elastic contribution) were used in the model. The elastic contribution is similar to the Gent model of hyperelasticity but the fully stretched chain limits are described more rigorously. The complete FEM model is programmed in the MATLAB. The open code allows monitoring of the progress of the calculation as well as implementation of any non-standard descriptions of changes of the Gibbs energy. So far, the model was applied to simple porous gel structures.

The research was supported by the grant agency of the Czech Republic by the project No. 17-08531S “Computational design of hydrogel cell scaffolds”.

**MODELLING OF POROUS HYDROGEL TOPOLOGY IN 3D:
WHEN AND HOW MUCH THE PORES COMMUNICATE?**

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Macroporous hydrogels with high volume of interconnected pores are widely used in biomedical fields. We simulated gel topology resulting from a specific pore formation method, and compared with a 3D image reconstruction. There has been a successful simple method of fabrication of highly porous hydrogels based on addition of salt particles insoluble

during gelation and removed from the final gel by dissolution.¹ To describe “templated gel” topology, we formulated a mathematical simulation model based on random placement of the particles into the gel volume. The model allows evaluating a number of topological parameters such as the volume fraction of pores contributing to paths percolation through the gel, number of not connected pores and

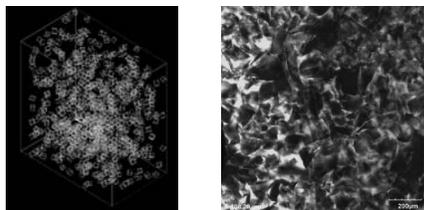


Figure 1. Structure of porous hydrogel fabricated by particle templating method. Simulated structure vs. laser scanning confocal microscopy image of gel in swollen state.

internal volume of pores formed by overlaps. The simulation model can be used first for determining the morphology, and subsequently for analysis of the gel deformation responses in ANSYS environment. The research is supported by the Grant Agency of the Czech Republic by project No GA17-08531S – “Computational design of hydrogel cell scaffolds“.

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SWELLING OF WEAK POLYELECTROLYTE GELS IN THE PRESENCE OF HYDROPHOBIC COUNTERIONS

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In this work we study swelling of weak polyelectrolyte gels in the presence of monovalent hydrophobic counterions. Previous works studied swelling of strong polyelectrolyte gels with monovalent salt.¹ Current combined experimental and simulation study² reported that Cobalt boron cluster (negatively charged amphiphile) induces conformational changes and ionization of weak polyelectrolytes. Therefore we wanted to estimate similar effect on weak polyelectrolyte gels.

We use coarse-grained model, with soft spheres representing individual monomer units and explicit ions in the system. We employ our in-house implementation of Hybrid Monte Carlo in the reaction ensemble.³ We use non-virial pressure estimator⁴.

We realised that hydrophobic counterions abound more in the gel comparing to 'normal' hydrophilic salt ions. They concentrate near the network nodes, where they induce ionization of the polymer segments. At longer chains (lower crosslinking) they induce to form the pearl structures.

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DEGRADABLE, THERMO-, pH- AND REDOX- SENSITIVE HYDROGEL MICROCAPSULES FOR BURST OR SUSTAINED RELEASE OF THE DRUGS

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Polymer microcapsules offer great potential for formulating high performance biomaterials. The mass of encapsulated particles is very high compared with the mass of the microcapsules wall, because of that hollow microparticles have a higher cargo loading capacity than smaller solid particles. Moreover, microcapsules containing specific cargo in the interior are promising model structures to gain a better understanding of drug transport through such systems. When they are sensitive to stimuli that exist in diseased and cancer cells they are promising materials of application in controlled intracellular drug delivery. Most of cancer cells exhibit mildly acidic pH and reducible environment (high concentration of glutathione). Moreover, reactive oxygen species (ROS) are overproduced in cells affected by inflammation and also in most of cancer cells. We fabricated hydrogel microcapsules by precipitation polymerization of *N*-isopropylacrylamide with *N,N'*-bis(acryloyl)cystine via the semi-batch process. Degradation of microcapsules by glutathione and hydrogen peroxide and controlled release of doxorubicin, from microcapsules, was investigated.

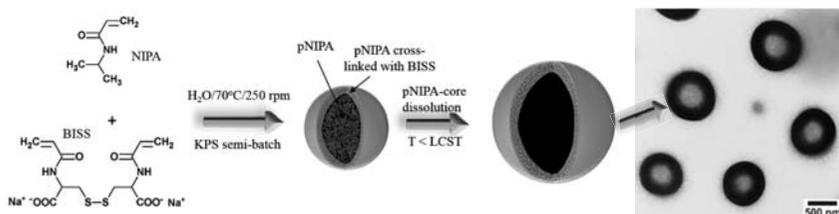


Figure 1. Scheme of synthesis and TEM images of microcapsules based on poly(*N*-isopropylacrylamide) crosslinked with derivative of cystine.

**THERMO-RESPONSIVE HYDROGEL FROM AMINO
ACID-DERIVED TRIBLOCK VINYL POLYMERS
VIA FLOWER-LIKE MICELLE FORMATION**

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Thermo-responsive hydrogels are attractive materials for applying to drug delivery system because a mechanical property of the hydrogels can be easily changed by thermal stimuli from outside. Here, we report novel thermo-responsive ABA-type triblock copolymers composed of alanine and glycine-derived vinyl polymers (poly(NAAMe_n-b-NAGMe_{2m}-b-NAAMe_n) which show a gelation property (Fig. 1). The triblock copolymers were successfully synthesized *via* atom transfer radical polymerization (ATRP) in a living manner, which were carried out in DMSO/2-propanol mixture solution at RT, by use of bifunctional alkyl bromide as an initiator. Thermal behaviors of the resultant triblock polymers were analyzed in dilute aqueous solutions by turbidimetry. The turbidity curves provided two-step lower critical solution temperature (LCST) transitions at *ca.* 18 and 60°C, and a flower-like micelle formation was confirmed at the temperature region between the first and second LCST transitions by dynamic light scattering, AFM and TEM analyses. At higher copolymer concentrations (>30 wt%), the triblock copolymer, whose degrees of polymerization are *n*=53, *m*=120, formed hydrogel at temperatures above the first LCST due to network formation of the flower-like micelles. The hydrogel showed self-healable and injectable properties based on rearrangement of polymer chains.

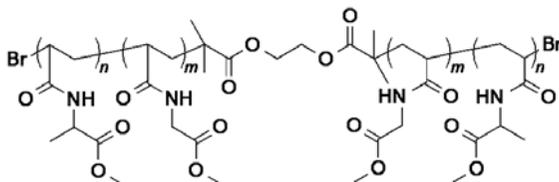
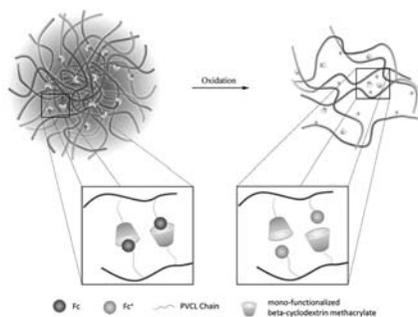


Figure 1. Chemical structure of poly(NAAMe_n-b-NAGMe_{2m}-b-NAAMe_n) synthesized by ATRP.

DEGRADABLE SUPRAMOLECULAR COLLOIDAL GELS

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Among the broad variety of nanomaterials, microgels have raised strong interest in many scientific areas. These colloidal polymer networks show a controlled responsive behavior, high stability, and bio-compatibility depending on their molecular structure.¹ Their tunable chemical structure and interesting properties in solution and at interfaces offer a large diversity of biomedical applications.² Also, introducing supramolecular functionalities enables the degradation of microgels *via* stimuli such as light, redox or pH, which can be used for the controlled release of drugs. Herein, we demonstrate a facile method to design redox-responsive aqueous microgels containing supramolecular compounds. The degradable crosslinker was formed using mono-modified β -cyclodextrin (host) and vinylferrocene (guest) based on host-guest interactions. A series of microgels using poly(N-vinylcaprolactam) (PVCL) with variable content of the degradable crosslinker was synthesized *via* precipitation polymerization. The selective degradation of microgels upon chemical and electrochemical stimuli could be demonstrated by TEM, AFM and DLS.³

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THERMORESPONSIVE POLY(N-ISOPROPYLACRYLAMIDE) AND ITS 3-(TRIMETHOXYSILYL)PROPYL METHACRYLATE BASED COPOLYMERS, HYBRID NETWORKS AND GELS

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One of the most intensively investigated class of smart materials belongs to poly(N-isopropylacrylamide) (PNIPAAm)¹ containing materials, mainly because of their broad application possibilities. The gel free synthesis of copolymers of NIPAAm and 3-(trimethoxysilyl)propyl methacrylate (TMSPMA) up to relatively high TMSPMA contents, capable to undergo sol-gel reactions, was accomplished.² Due to the hydrophobic nature of TMSPMA, the cloud point and clearing point decrease with increasing TMSPMA content. Unprecedented hysteresis behavior of P(NIPAAm-co-TMSPMA) copolymers with higher TMSPMA contents was found, indicating catalyst free and composition dependent gelation above the cloud point, i.e. under mild conditions. Inorganic-organic hybrid gels of P(NIPAAm-co-TMSPMA) copolymers with high gel fractions by sol-gel processes were successfully prepared in both the absence and presence of tetraethyl orthosilicate (TEOS). The resulting materials were characterized by their gel collapse temperature (GCT), SEM and SAXS indicating the absence of large scale phase separation in these thermoresponsive hybrid networks. Drug release experiments surprisingly show that these hybrid gels are suitable drug carrier matrices for sustained release under and above the GCT as well.³

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ELECTROSPUN POLYVINYLPIRROLIDONE/ESSENTIAL OIL COMPOSITES FIBERS

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Over the recent years there has been a growing interest in the development of composite fibers based on essential oils (EO) due to a wide range of their potential applications.¹ Among the different methods to obtain composite fibers in a controlled manner from the micro to the nanometer scale (1 μm to 100 nm diameter), electrospinning has become a very useful technique. There are several synthetic polymers used for electrospinning processing, such as polyvinyl alcohol, polyethylene oxide or polyvinylpyrrolidone (PVP). PVP is a biodegradable polymer that presents a good environmental stability and easy processability. Hence, it has been considered as a promising polymer for numerous applications such as shielding devices, adhesives, medical devices and biological engineering materials.

Therefore, the present work aims to report the effect of EO load on the electrospinning of PVP solutions under different concentrations and types of essential oils (clove, cinnamon, white thyme and oregano). To this end, characteristics of the electrospun fiber mats including morphology, chemical and thermal properties were studied. Additionally, the rheological behavior of the polymer solutions was analyzed to understand the electrospinnability of the PVP/EO solutions. This allowed gaining insight into potentially of these blends to develop composite fibers with enhanced functional properties.

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CARBOXYLIC ACID FUNCTIONAL POLY(ETHYLENE GLYCOL) FOR NON-COVALENT CROSSLINKED HYDROGELS

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Poly(ethylene glycol) (PEG) is the gold standard of biocompatible polymers in medical, pharmaceutical and cosmetic applications due to its outstanding properties. PEG exhibits a unique high aqueous solubility, a low immunogenicity, antigenicity and toxicity. Ethylene oxide (EO) can be copolymerized with various functional epoxides to achieve multifunctionality of the polyether chain, leading to multifunctional PEG.¹ Hydrogels are used in a variety of biomedical applications. In case of physical gels, the gelation is due to hydrogen bonding or ionic interaction. Therefore, the gelation is reversible and responsive to changing *pH*.² We present the design of ester functional epoxides for the synthesis of ester functional PEG via ring opening polymerization. Cleavage of the ester groups results in biocompatible polyelectrolytes, that can be used to form *pH*-responsive gels trough ionic interactions. Since certain tissues of the body have a slightly more acidic *pH* than normal tissue,^{3,4} the resulting hydrogels can provide an efficient way for *pH*-controlled drug release.

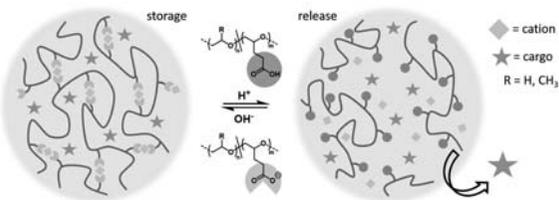


Figure 1. *pH*-responsive storage-release system.

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THERMO- AND pH-SENSITIVE INTERPENETRATING NETWORK MICROGELS FOR COATINGS

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Polymer microgels responsive to the influence of temperature and acidity of the medium can be used to obtain different functional materials such as superabsorbents, carriers of drugs and dyes. Using microgels in front of macrogels is more preferable because of shorter response times to external stimuli. The aim of our study is to investigate the properties of microgels based on poly(*N*-isopropylacrylamide) (PNIPAM) and poly(acrylic acid) (PAA) interpenetrating networks (IPN) and to check the possibility to use such microgels for the development of polymer films and coatings. To obtain the microgels with IPN structure, in the first stage, the PNIPAM microgels were synthesized by thermo-induced precipitation polymerization, then the second subnetwork was synthesized by in situ copolymerization of acrylic acid and *N,N'*-methylenebisacrylamide in the volume of the initial PNIPAM microgels. The influence of different polymerization conditions was studied. The thermo- and pH-responsive behavior of obtained IPN microgels was investigated by dynamic and static light scattering. The formation of second subnetwork didn't affect the transition temperature (34°C). The pH-induced transition was observed around pH 4.5. It was shown that at the room temperature the IPN particles have a core-shell structure. The sizes and shapes of IPN particles were observed by scanning and transmission electronic microscopy. In the next steps, polymer films will be produced by assembling of IPN microgels on different substrates during solvent evaporation and further temperature treatment or crosslinking by multivalent ions. Such films can be used as containers for immobilization of organic dyes and drugs.

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DOUBLE-NETWORK HYDROGELS IMPROVE pH-SWITCHABLE ADHESION

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A double-network hydrogel is repeatedly used in environmentally-switchable adhesion systems. The main goal of using double-network hydrogels rather than single-network hydrogels is that the integrity of the interface must be retained after each adhesion cycle. This is because of that the single-network hydrogels can be reused only once since the cohesive failure leads to an accumulation of the hydrogel on the brush surface and the hydrogel can only be reused at different parts of that surface. Here, a double-network hydrogel of poly(methacrylic acid) and poly[oligo(ethylene glycol)methyl ether methacrylate] is shown to undergo adhesive failure during pH-switchable adhesion with a grafted (brush) layer of polycationic poly[2-(diethyl amino)ethyl methacrylate], and can be reused at least seven times. At pH 6, the adhesion is strong at the gel/brush interface. While the interface is detaches as the adhesion becomes weak at pH 1. The double-network hydrogel can even attach and detach at least three times at the same point on the brush surface at pH 6 without an environmental stimulus, whereas the single-network hydrogel cannot be reused under such circumstances.

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RECOVERY OF CO₂ USING TEMPERATURE-RESPONSIVE AMINE GEL SLURRY

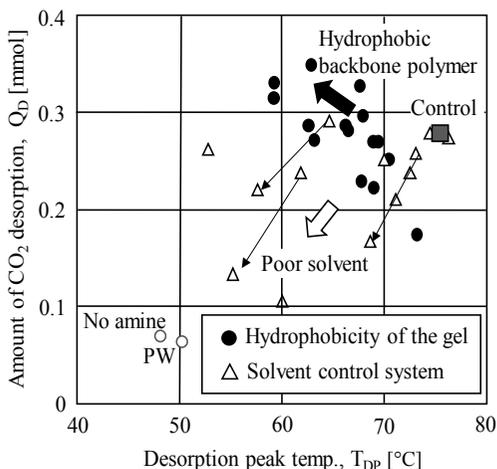
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Amine-based chemisorption process is reliable method for the recovery of CO₂ from industrial plants. However, desorption of CO₂ and regeneration of adsorbent are energy consuming processes with high-temperature treatment. Thus, reduction of the desorption temperature is the key subject to develop eco-process in the recovery system. In the present study, we focused on slurry of cross-linked LCST polymer micro hydrogels to realize the reduction of desorption temperature in the amine-based CO₂ recovery system. Temperature responsive volume change in the LCST polymer gels enables control of pKa of the amine groups immobilized in the gels,¹ which would also enable desorption of CO₂ at lower temperature than ordinary amine system. The gel slurries were prepared using N-isopropylacrylamide (NIPA), N,N-diethylaminopropylacrylamide (DMAPAA) and some other hydrophobic monomers. adsorption and desorption behaviours for CO₂ were investigated as a function of hydrophobicity of backbone polymer, amine content and composition of solvent in the slurry system. The slurry conditions with effective CO₂ recovery was identified from the T_{DP} vs. Q_D trends shown in Fig.1.



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Figure 1. Relationship between the desorption temperature of CO₂ and the amount of CO₂ desorption.

POLYMER NETWORKS IN CONTINUOUS FLOW REACTORS FOR ORGANOCATALYTIC REACTIONS

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Flow reactors are useful tools to enable hazardous reactions and are, hence in agreement with *Green Chemistry*.^{1,2} Important benefits of flow reactions are on one hand the used catalyst is part of continuous flow system and will permanently generate product, on the other hand the resulting product is free of catalyst and could be easily isolated by solvent evaporation.³

In order to prevent the bleeding of catalyst, covalently bonded catalysts are used for organic reactions like Aldol- or Mannich-reaction. Thus catalyst immobilized on polystyrene particles are often used, but this is restricting the catalytic active sites to the particle surface.⁴⁻⁶ To enlarge the catalytic activity inside of the flow reactor, such particles will be replaced by polymer networks. Cross-linked polymers based on acrylates and acrylamides, enable a reaction inside of swollen polymer networks, which increased the amount of immobilized catalyst from only surface area to bulk volume.

To optimize the reactor performance special emphasis is given to the polymer network size and shape as well as composition (to be active in a selected reaction). The characterization of synthesized polymer networks is important to find correlations between structure and activity.

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PREPARATION AND PROPERTIES OF ENR-40/SILICA NANOCOMPOSITES VIA SAFE SULFUR CURING SYSTEM

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In this study, ENR-40 was synthesized via 'in situ' performic epoxidation of NR using a molar ratio of HCOOH/H₂O₂ to isoprene unit at 1/0.75 at 50°C for 8 h. To prepare ENR-40/nano-SiO₂ nanocomposites, the obtained ENR-40 was blended with three loadings of nano-SiO₂ (1–3 phr) in the latex stage, followed by compounding with various curing ingredients in an internal mixer and then compression curing. Tetrabenzylthiuram disulphide (TBzTD) was used as a non-carcinogenic accelerator for the efficient sulfur curing system. It was found that the incorporation of an appropriate amount of nano-SiO₂ into the ENR-40 had improved the mechanical properties of the nanocomposites. Among them, the one that filled with 2 phr nano-SiO₂ exhibited the highest tensile properties and tear strength both before and after thermal aging at 70°C for 72 h, indicating the better dispersion of nano-SiO₂ in the ENR-40 matrix.

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WATER UPTAKE BY ULTRATHIN CROSSLINKED FILMS STUDIED BY QCM

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Water diffusion into cured networks of high performance polymeric coatings always occurs under application conditions. Diffusing water influences the mechanical resistance, durability, and adhesion of coatings even at very low concentration. The mechanism is related to formation of H-bonds acting in different ways: they may bridge the network structure or hydrate the polar moieties causing plastification, and diffuse further causing network relaxation – depending on network chemistry. To test the water uptake kinetics and equilibrium in very thin coatings we have set up a method based on quartz crystal balance monitor (QCM) capable of detection of very low mass changes, as low as 10^{-9} grams and working under various humidity, pressure and temperature. Here, we present the QCM apparatus designed for water uptake studies, and the results obtained so far with spin coated films.

We have studied water uptake by films based on polyurethane/polyurea and evaluated parameters such as network composition, crosslink density, film thickness, external humidity, and pressure. The process revealed very fast initial uptake step and hysteresis behavior. The diffusion coefficients estimated using Crank model spanned the range of $1-5 \times 10^{-9}$ cm^2/s and the equilibrium amount of absorbed water was surprisingly high, around units of percent. The hysteresis effects were observed in parallel in sorption-diffusion measurement done with a sorption balance and were attributed to film morphology.

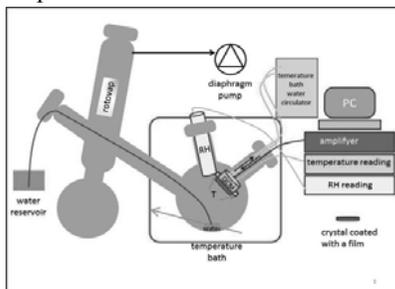


Fig. 1: QCM set-up for monitoring water uptake.

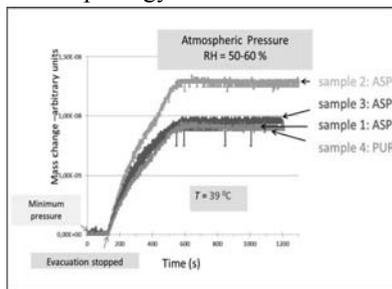


Fig. 2: Water uptake by thin films monitored by QCM.

ADHESION AND RHEOLOGICAL ASSESSMENT OF BIO-SOURCED POLYURETHANE NETWORKS FROM CELLULOSE ACETATE/VEGETABLE OIL MIXTURES

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The replacement of the petroleum-based adhesives with more eco-friendly materials has become in recent decades one of the main industrial interests. Additionally, polyurethanes have drawn the attention in the adhesive production due to its outstanding adhesion performance, along with exceptional flexibility, high resistance, etc.¹ On the other hand, cellulose derivatives and vegetable oils have been proposed to synthesize new eco-friendly alternatives to traditional polyurethanes.^{2,3} In this research, cellulose acetate was functionalized with 4,4'-diphenylmethane diisocyanate at different NCO:OH ratios, ranging from 2 up to 4.53, and the resulting biopolymers were mixed with castor oil at 1:1 weight ratio. The completely cured bio-sourced polyurethanes were rheologically characterized by applying dynamic oscillatory torsional tests at different temperatures (from -30 up to 200°C). Moreover, their adhesion performance on stainless steel and wood substrates was analysed, by conducting standardized mechanical tests studying single-lap and 180° peel strengths at room temperature and 100°C. Fourier-transform infrared spectroscopy-attenuated total reflectance, along with differential scanning calorimetry and thermogravimetric analysis were also performed. In most cases, a predominant simple thermo-rheological behaviour could be found within the whole temperature range studied, being able to apply the t-T superposition principle. However, an increase in the temperature led to a depletion in their mechanical performance, thus reducing their temperature range of application. Thermal and spectroscopic analysis corroborated the complete disappearance of free isocyanate during the first few days of curing.

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NCO-FUNCTIONALIZED WHEAT AND BARLEY STRAWS-BASED OLEOGELS: INFLUENCE OF SOLID-STATE FERMENTATION

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Wheat and barley straws are one of the main agricultural residues with great potential because of its wide availability and low cost. These straws were subjected to solid-state fermentation (SSF) with *Streptomyces* sp. MDG301. Subsequently, control and fermented straws were functionalized with hexamethylene diisocyanate (HDI) to be used as thickener agents in the production of bio-based lubricating greases. Lubricant performance of oleogels was assessed by means of thermo-rheological characterization, unworked and worked penetration and tribological properties. At room temperature, all of them showed rheological characteristics similar to those found in lithium-based commercial greases.¹ Nonetheless, several differences among these oleogels could be found, as *Streptomyces* had contrary effects for both straws; whereas wheat straw viscoelastic moduli were negatively affected, barley straw moduli potentially increase when processing. On the other hand, temperature and rolling-shear exert a higher influence on straw-based oleogels than on lithium-based greases. On the contrary, regarding tribological properties, a significant reduction in the friction coefficient was achieved when using straw-based oleogels as lubricants.

Acknowledgment: This work is part of two research projects (CTQ2014-56038-C3-1R and CTQ2014-56038-C3-2R) sponsored by MINECO-FEDER.

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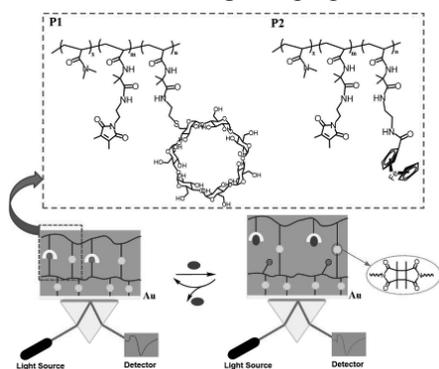
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DUALLY CROSSLINKING SUPRAMOLECULAR POLYMER TOWARDS SMART GEL ACT AS MOLECULAR SENSORS

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In the last decade, polymer networks containing dually crosslinked systems were paid much attention due to their special properties and promising applications in both chemical and biological fields.¹ Despite significant developments in this field, novel visualizing strategies of dually crosslinking polymer gels that offer various applications are still imperative.² Here, a novel dually crosslinking supramolecular polymer combining the photo cross-linkable covalent bonding with host-guest noncovalent binding was prepared.



Based on this concept, a stable azlactone based copolymer P(DMAAm-VDMA) was obtained via RAFT copolymerization and further modified with DMIAm as photo crosslinker and β -CD and ferrocene as host-guest interaction pair. After mixing the solution of two copolymers, supramolecular network was constructed by spin coating and photo crosslinking forming a thin gel layer anchored

on gold surface. Optical waveguide (OW) as well as surface plasmon resonance (SPR) spectroscopy were combined to observe the layer thickness and refractive index independently at the same time.³ Interestingly, the supramolecular network swelling was triggered by target molecules, which could break the host-guest interaction pair through competitive interaction with β -CD. Thus, the thin gel layer was used as a molecular sensor monitored by the SPR spectroscopy.

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INVESTIGATION OF ACID ORANGE 8 ADSORPTION ONTO VINYL IMIDAZOLE-BASED COPOLYMERIC HYDROGELS

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Textile dyes, which are toxic, carcinogenic and mutagenic for humans and living organisms, have a significant proportion of pollutants in industrial wastewater. Adsorption is one of the most effective and preferable method among the other chemical and biological methods for the wastewater treatment.

In this study, vinyl imidazole-based copolymeric hydrogels were synthesized by using free radical polymerization. [2-(Methacryloyloxy)ethyl]dimethylnonylammonium bromide (DMA_C8) which was obtained by quaternization of 2-dimethylaminoethyl methacrylate with 1-bromononane was used as comonomer for the hydrogel synthesis. The functional groups of the hydrogels were analyzed by using FTIR; the surface morphologies were investigated by SEM. The equilibrium swelling ratio of hydrogels was determined in pure water at room temperature by gravimetric method. Hydrogels were used as an adsorbent for the removal of Acid Orange 8 (AO8) from aqueous solutions. Adsorption experiments were carried out in a batch system and the experimental parameters were evaluated with respect to pH, agitation time and initial dye concentration. Langmuir and Freundlich adsorption models were used to describe the adsorption isotherms. The kinetic data were analyzed using the pseudo-first-order, pseudo-second-order and intraparticle diffusion equations. The adsorption was fitted to pseudo-second-order kinetics, and intraparticle diffusion until 90 min. Copolymeric hydrogels exhibited remarkable adsorption performance at the pH 2.0. The adsorption capacity of the copolymeric hydrogel was found to be significantly higher than the most of the other common adsorbents using in the adsorption of anionic dye pollutants have.

MODIFICATION OF POLY(N-ISOPROPYLACRYLAMIDE): SYNTHESIS AND CHARACTERIZATION OF NOVEL THERMOSENSITIVE COPOLYMERIC HYDROGELS

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Hydrogels are three dimensional, cross-linked, polymeric structures. They do not dissolve in water and they swell by absorbing high levels of water in aqueous solutions. Hydrogels are sensitive to heat, pH, ionic strength, and electric and magnetic fields, and may exhibit dramatic and reversible volume changes with a variance under these conditions. Poly(N-isopropylacrylamide) (PNIPAAm) is the one of the most investigated thermosensitive hydrogels. It has a low critical solution temperature (LCST) above which they undergo transition from a solution to gel state.^{1,2} In this study, N-isopropylacrylamide based thermosensitive homopolymeric and copolymeric hydrogels were synthesized by free radical solution polymerization. [2-(methacryloyloxy)ethyl]dimethyltetradecylammonium bromide (DMAC14), N,N'-methylenebisacrylamide (BAAm), potassium persulfate (KPS), N,N,N',N'-tetramethylethylenediamine (TEMED) were used as comonomer, crosslinking agent, initiator and catalyst, respectively. Scanning electron microscope (SEM) and fourier transform infrared spectrometer (FTIR) were used for the characterization of the hydrogels. The dynamic swelling experiments were carried out in distilled water at different temperatures. It was observed that the presence of DMAC14 in the polymeric matrix reduced the thermal sensitivity and swelling degree of the copolymeric gels.

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A MECHANICAL STUDY OF TOUGH HYDROGELS WITH CHEMICAL AND PHYSICAL CROSSLINKS

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Hydrogels are promising candidates for biomedical applications, such as targeted drug-delivery or tissue engineering. However, the heterogeneity of the polymer network and the lack of dissipative mechanisms in the material makes them very fragile. In order to improve the mechanical properties of hydrogels, reversible physical crosslinks have been introduced into a chemically crosslinked network. Inspired by the mussel byssus, an organ of the mussel which allows it to attach to coastal surfaces thanks to metal ligand coordination complexes,^{1,2} we developed a poly(acrylamide-co-vinylimidazole) hydrogel physically crosslinked by Ni²⁺ – Imidazole coordination bonds, chemically crosslinked by methylene bisacrylamide (MBA). The rheological modulus, tensile modulus and extensibility of this dual crosslink gel were found to be much higher than those of the bare chemical gel. These properties also increased when increasing the concentration of metal ions. By increasing [Ni²⁺], we can increase the energy dissipated in loading/unloading cycles, which might explain the increase in extensibility observed. The next step would be to introduce mecanophores in the chemical network, to better understand the mechanics of fracture of this dual-crosslink hydrogel.



Figure 1. Schematic presentation of the formation of the dual crosslink gel.

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SYNTHESIS AND CHARACTERISTICS OF ELECTROSWITCHABLE MICROGEL

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Microgels are polymer hydrogels or macromolecules that form, in a micro scale, three-dimensional networks filled with a solvent. We synthesized a microgel which size could be controlled electrochemically. The starting material was a thermoresponsive microgel built from *N*-isopropylacrylamide, sodium acrylate and *N,N'*-bisacryloylcystine as the crosslinker. The gel was modified with aminoferrocene through amid bond formation. The properties of the obtained materials were examined with dynamic light scattering technique, UV-Vis spectroscopy, SEM and TEM microscopies and cyclic voltammetry. The influence of oxidation state of the ferrocene groups in the microgel on volume phase transition temperature was determined. The -S-S- groups from the *N,N'*-bisacryloylcystine linker were employed in chemisorption of the microgels on the Au electrode surface and formation of gel monolayers. It appeared that the microgel layers could be either in the shrunken or the swollen state depending on the oxidation number of iron atoms in the ferrocene groups. The oxidation number could be changed electrochemically and the shrunken-swollen transformation could be repeated reversibly many times.

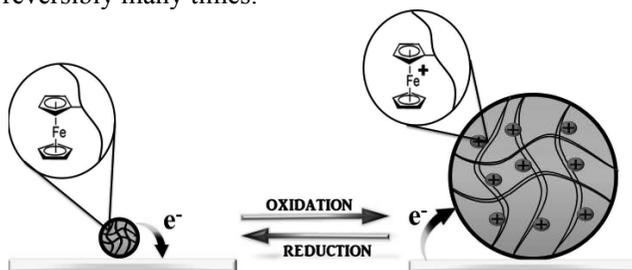


Figure 1. Scheme of the microgel volume phase transition induced electrochemically.

LIGHT AND X-RAY SCATTERING STUDY OF A TEMPERATURE AND pH RESPONSIVE PHYSICAL HYDROGEL

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Stimuli-responsive physical hydrogels strongly change their properties upon a small change of the environment and may be used as fast sensors, for drug delivery or for tissue engineering.¹

In the present work, the structure of a multiresponsive CBABC type pentablock terpolymer in aqueous solution is investigated in dependence of temperature and pH using light and X-ray scattering methods. The end blocks are statistical copolymers of the hydrophobic *n*-BuMA and the thermoresponsive TEGMA, the intermediate middle blocks are the weak cationic polyelectrolyte PDMAEMA, and the middle block is the hydrophilic PEG. In aqueous solution, either flower-like micelles or a physical network, with the hydrophobic end blocks acting as crosslinks, are expected. The dynamics of the network may be tuned by varying the temperature around the transition temperature of the endblocks.² pH allows stretching of the intermediate blocks and thus tunes the gelation behavior.

Large hydrodynamic radii (R_h), which decrease with increasing temperature, are found for high pH values, where PDMAEMA is uncharged, whereas at low pH, R_h is smaller and temperature-independent. The micellar radius, R , obtained from small-angle X-ray (SAXS) measurements, which presumably probe single micelles, is also found to be higher for high pH values, indicating a higher aggregation number. Furthermore, for high pH values, R decreases at higher temperatures, while at the lowest pH, R continuously increases with increasing temperature.

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PHOTOPOLYMERIZED DOUBLE NETWORK HYDROGEL OF HYALURONIC ACID AND SILK FIBROIN

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Hyaluronic acid (HA) and its derivatives have been used in hydrogel fabrication for biomedical applications. In general, HA-based hydrogel rapidly degrades in physiological condition by hyaluronidase or hydrolysis.¹ Therefore, various techniques have been tried for slow degradation of HA hydrogel. In this study, silk fibroin (SF) was incorporated in HA hydrogel network via both chemical and physical cross-linking. HA and SF were modified with methacrylic group for photocrosslinking. Both methacrylated HA and SF were dissolved in PBS and the solutions were blended at various volume ratios. For chemical cross-linking, the blended solution was photopolymerized with a photoinitiator under ultra-violet light irradiation. Then, the hydrogel was treated with ethanol for physical cross-linking of SF chains. By addition of SF in hydrogel, not only was the hydrolytic degradation retarded, but the characteristic physical property change was observed. Before ethanol treatment, shear elastic modulus slightly increased with an increase of SF content in hydrogel. On the other hand, the hydrogel modulus was significantly elevated after ethanol treatment. Such an elevation of shear modulus became higher with an increase of SF content. The result of FTIR analysis indicated that the property change via ethanol treatment was due to beta-transition of incorporated SF chains.² The beta-sheet structure of SF induced the formation of physical cross-linking, resulted in double-network in HA hydrogel.

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THERMORESPONSIVE BEHAVIOR OF POLY(N-ISOPROPYLACRYLAMIDE)S WITH DODECYL AND CARBOXYL TERMINAL GROUPS IN AQUEOUS SOLUTION: pH-DEPENDENT CLOUD POINT TEMPERATURE

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It was recently reported that poly(N-isopropyl acrylamide) (PNIPAm) polymers synthesized by RAFT polymerization using S-1-dodecyl-S'-(α,α' -dimethyl- α'' -acetic acid)trithiocarbonate as a chain transfer agent form micelles in aqueous solutions with the core of hydrophobic terminal dodecyl groups and the corona of PNIPAm chains with carboxylic groups at the periphery, the ionization of which prevents the micelles from phase separation above the lower critical solution temperature of PNIPAm in water¹. In this project², we study the pH- and ionic strength-dependence of the aggregation behavior of two HOOC-PNIPAm-C12 polymers, differing in the degree of polymerization, in aqueous solutions. We show that the cloud point temperature (CPT) of HOOC-PNIPAm-C12 can be shifted up to several tens of K by changing pH of the solution. The aggregation of the PNIPAm above the CPT can be efficiently accelerated by screening electrostatic repulsion between PNIPAm micelles by changing ionic strength of the solution.

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FREEZING-INDUCED MECHANOLUMINESCENCE OF CROSS-LINKED POLYMER GELS THAT CONTAINS A LIGHT-EMITTING MECHANOCROMOPHORE

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Polymer mechanochemistry is a fundamental research field to understand how macroscopic mechanical forces are applied to polymeric materials. As a branch of mechanochemistry, mechanochromism, changing color in response to mechanical stress, has been achieved by various stimuli such as tension, compression, shearing, and sonication. Surprisingly freezing also induces mechanical stress. Previous study of our research group showed freezing-induced mechanochromism (FIM) of polymer gels, where

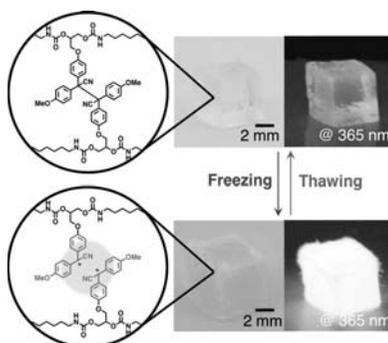


Figure 1. Freezing-induced mechanoluminescence of **TASN gel**.⁴

diarylbibenzofuranone (DABBF) derivatives at the cross-linking points acting as the color-changing mechanophores was homolytically cleaved to form blue-colored radical in response to shear force induced by freezing.¹ In this study, freezing-induced mechanoluminescence was achieved by the introduction of tetraarylsuccinonitrile (TASN)² skeleton as a yellow light-emitting mechanophore into the cross-linking points of polymer gels (**TASN gel**, **Fig. 1**). FIM of **TASN gel** enhanced capability to see mechanical stress with not only pink coloration but also yellow light emission under UV irradiation. The mechanism and characteristics of FIM were studied with the quantitative evaluation by electron paramagnetic resonance (EPR) measurements, revealing that the network structure³ and solvent affinity with polymer chain had good correlations with the dissociation behavior of TASN in FIM.⁴

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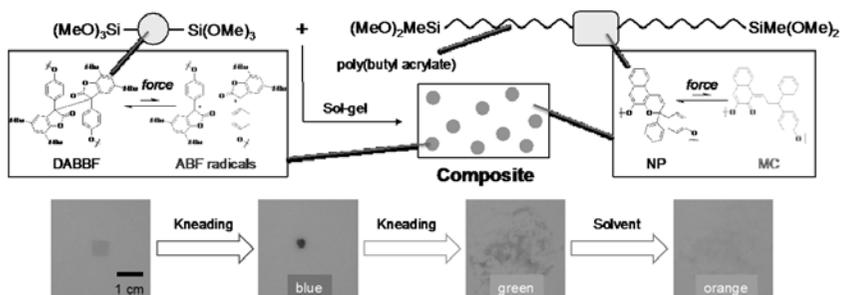
**POLYMER-INORGANIC COMPOSITE NETWORKS SHOWING
MULTICOLOR MECHANOCROMISM WITH DUAL
MECHANOCROMOPHORES**

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Diarylbibenzofuranone (DABBF) can generate blue-colored stable arylbenzofuranone (ABF) radicals¹ and naphthopyran (NP) can be converted to orange-colored merocyanine (MC)² in response to mechanical stimuli. In a high mobility region, ABF radicals quickly revert back to colorless DABBF¹, on the other hand MC can remain its colored state.² Here we developed the multi-color mechanochromic polymer-inorganic composite containing DABBF units in silica-rich domains and NP units in polymer domain, based on difference between the characters of DABBF and NP. It showed different colors according to strength of mechanical stimuli, lapse of time, and solvent addition.



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THE STRUCTURE AND PHYSICAL PROPERTIES OF MIXED STEROL ORGANOGELS

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In many foods, crystallized saturated fats provide texture and mechanical strength, but are linked to an increased risk of heart disease. The replacement of saturated fats with alternative materials which offer the same mechanical properties but without the associated health risks is therefore a topic of much research. An alternative method for structuring unsaturated triglyceride oils is the formation of so-called oleogels. Several potential oleo-gelators have been identified including mixtures of phytosterols and sterol esters. The use of phytosterols as oleogelators is particularly appealing as these have also been linked to cholesterol lowering properties, and have been incorporated into some spreads for these reasons. However, understanding the structure of how these gels and the kinetics of their formation are key to successfully incorporating organogels into food.

To this end, we have investigated phytosterol organogels over a range of length scales. A combination of spectroscopy, atomic force microscopy (AFM) and molecular dynamics simulations give insight into the structure of individual self-assembled fibrils and the driving force behind their formation.¹⁻³ Using the same range of experimental techniques we also investigate the interactions between fibrils to form larger, fibrous bundles.

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**SHAPE MEMORY, SPONTANEOUS ACTUATION,
AND PROGRAMMABLE ACTIONS OF HYDROGELS**

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Generally, the temporary shape of shape memory polymers is formed by an external force and fixed by some reversible interactions, which are triggered by gentler conditions. For the hydrogels containing plenty of water, the motion of polymer chains becomes fast and the fixation for the temporary shape is weakened. At the same time, all the actuations are one-off, and the original shape cannot convert to the temporary shape spontaneously and reversibly. For this purpose, hydrogels are usually fabricated with anisotropic structures to transfer the isotropic volume expansion into anisotropic movements. However, these hydrogels usually require a complicated preparation or/and subsequent treatment, limiting their applications. Therefore, the spontaneous shape change from the original shape to the temporary shape is essentially required for the hydrogel to perform a continuous actuation.

The hydrogels exhibited pH responsive shape memory behavior and spontaneous shape change after the first shape memory cycle were prepared. When the recovered hydrogel with a little residual deformation was immersed in HCl again, it twisted spontaneously and rapidly to the previous temporary shape. The spontaneous twisting and recovering can be repeated for 10 times simply by alternately immersing in NaOH and HCl solutions without any external force. This unique synergistic effect of fast swelling, residual helical deformation, and increased strength played a significant role to the spontaneous shape alternation.

The Janus bilayer and multilayer hydrogel actuators were fabricated from tough and adhesive PAA-clay hydrogel. This PAA gel and Fe³⁺ reinforced PAA gel showed different moduli and ionic strengths. Elaborate hydrogel actuators such as bidirectional bending flytrap, palm with grasp-open, gesturing hand, and word-writing actuator were realized.

POLY(DIMETHYLSILOXANE)-BASED CROSSLINKED FILM WITH HIGH DIELECTRIC CONSTANT AND LOW MODULUS

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We synthesized high molecular weight poly(dimethylsiloxane-*co*-methylvinylsiloxane) (**1**) and poly(dimethylsiloxane-*co*-methylsiloxane) (**2**) for obtaining a crosslinked film with high dielectric constant and low modulus. Once the liquid copolymers **1** and **2** are mixed together, platinum (Pt)-catalyzed hydrosilylation reaction¹ occurs between vinyl and hydrosilane groups in the polymer chains. The resulting crosslinked film showed comparatively high dielectric constant (4~5 @ 1 kHz) and low initial modulus (~ 100 kPa), which depended on its composition. Our film would be superior to that fabricated from Sylgard[®] or Elastosil[®] as an actuator material.

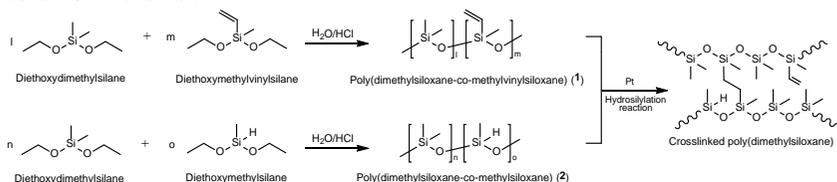


Figure 1. Synthesis of copolymers **1** and **2** and hydrosilylation reaction between them.

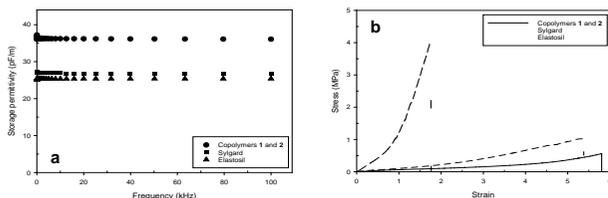


Figure 2. Dielectric constant (a) and mechanical properties (b) of the crosslinked films fabricated from Sylgard[®], Elastosil[®], and copolymers **1** and **2**.

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ENHANCING THE MECHANICAL DURABILITY OF ICEPHOBIC SURFACES BY INTRODUCING AUTONOMOUS SELF-HEALING FUNCTION

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Ice accretion presents a severe risk for human safety. Although great efforts have been made for developing icephobic surfaces (the surface with an ice adhesion strength below 100 kPa), expanding the lifetime of state-of-the-art icephobic surfaces still remains a critical unsolved issue. Herein, a novel icephobic material is designed by integrating an interpenetrating polymer network (IPN) into an autonomous self-healing elastomer, which is applied in anti-icing for enhancing the mechanical durability. The molecular structure, surface morphology, mechanical properties, and durable icephobicity of the material were studied. The creep behaviors of the new icephobic material, which were absent in most relevant studies on self-healing materials, were also investigated in this work. Significantly, the material showed great potentials for anti-icing applications with an ultralow ice adhesion strength of 6.0 ± 0.9 kPa, outperforming many other icephobic surfaces. The material also exhibited an extraordinary durability, showing a very low long-term ice adhesion strength of ~ 12.2 kPa after 50 icing/deicing cycles. Most importantly, the material was able to exhibit a self-healing property from mechanical damages in a sufficiently short time, which shed light on the longevity of icephobic surfaces in practical applications.

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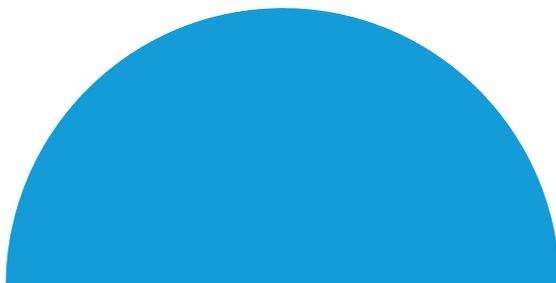
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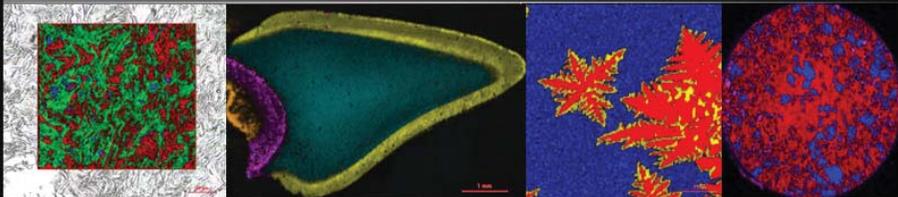
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