

Bioinspired and Zwitterionic Materials

864. WE-Heraeus-Seminar

23 – 26 August 2026

at the Physikzentrum Bad Honnef, Germany

**WILHELM UND ELSE
HERAEUS-STIFTUNG**



Introduction

The Wilhelm und Else Heraeus-Stiftung is a private foundation that supports research and education in science with an emphasis on physics. It is recognized as Germany's most important private institution funding physics. Some of the activities of the foundation are carried out in close cooperation with the German Physical Society (Deutsche Physikalische Gesellschaft). For detailed information see <https://www.we-heraeus-stiftung.de>

Aims and scope of the 864. WE-Heraeus-Seminar:

Zwitterionic materials like phosphorylcholines and betaines, but also ampholytes, ionic liquids, and layer-by-layer materials have emerged as promising materials in biomedical and engineering applications. The conference will bring together experts from the fields of bioinspired and naturally occurring materials with zwitterionic key building blocks used to tailor the properties of synthetic bioinspired materials and provide an overview of the research state of the art and latest developments on this research topic.

The topics include bioinspired and zwitterionic materials, surfaces and nanoparticles, materials synthesis and characterization, applications in medicine, biosensors and cell culture, applications in nanomedicine and nanodiagnostics, technical applications like protective coatings for ocean and freshwater applications, membrane technology, construction, energy research, and pharmaceutical applications like controlled drug delivery.

The conference will cover fundamentals on the physics and physicochemical properties of the materials up to applied and translational research. The participation of young researchers is encouraged, and a young scholar poster prize will be awarded.

Scientific Organizers:

Prof. Dr. Axel Rosenhahn

Ruhr-Universität Bochum

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Prof. Dr. Karen Lienkamp

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Prof. Dr. André Laschewsky

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Introduction

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Registration:

Mojca Peklaj (WE Heraeus Foundation)
at the Physikzentrum, Reception Office
Sunday (16:00 h - 21:00 h) and Monday morning

Program

Program

Sunday, 23 August 2026

16:00 – 21:00 Registration

18:30 *BUFFET SUPPER and informal get together*

20:00 André Laschewsky **Polyzwitterions: Achievements, Myths
& Fancy**

Get together

Monday, 24 August 2026

08:00 *BREAKFAST*

Session chair: Axel Rosenhahn

09:00 – 09:20 Organizers **Opening**

09:20 – 10:00 Kazuhiko Ishihara **MPC Polymer Science and Engineering:
Shaping the Future of Biointerfaces
and Medical Devices**

10:00 – 10:20 Rong Yang **Vapor-Phase Engineering of Zwitter-
ionic Polymer Interfaces for Biofouling
Control and Electrochemical Stability**

10:20 – 10:40 Jessica Schiffman **Bioinspired and Fermented Materials
for Environmental Applications**

10:40 – 11:20 *COFFEE BREAK*

Session chair: Joe Schlenoff

11:20 – 11:40 Masaru Tanaka **Understanding Water-Mediated
Interactions at BioInterfaces:
Intermediate Water Concept**

Program

Monday, 24 August 2026

11:40 – 12:00	Esben Thormann	Specific Ion Effects in Zwitterionic Polymer Coatings: From Antipoly-electrolyte Swelling to Ice Adhesion
12:00 – 12:20	Elena Vassileva	Polyzwitterionic Hydrogels as Multi-functional Biomaterials: From Chronic Wound Management to Ocular Drug Delivery
12:20 – 12:40	Conference Photo (in front of the main entrance)	
12:40 – 14:10	<i>LUNCH</i>	

Session chair: Kazuhiko Ishihara

14:10 – 14:50	Yung Chang	Molecular Engineering of Zwitterionic Membranes for Preferential Blood Cell Separation
14:50 – 15:10	Karen Lienkamp	Polyzwitterionic Hydrogels: Beyond Biofilm Formation and Biocompatibility
15:10 – 15:50	Flash Presentations I	
15:50 – 16:30	<i>COFFEE BREAK</i>	
16:30	Flash Presentations II	
	Poster Session	
18:30	<i>DINNER</i>	
19:30	Poster Session cont.	
	Get together	

Program

Tuesday, 25 August 2026

08:00 *BREAKFAST*

Session chair: André Laschewsky

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|---------------|---------------------|--|
| 09:00 – 09:40 | Joe Schlenoff | How Are “Electrostatic” Interactions Driven by Charge Pairing and Dehydration? |
| 09:40 – 10:00 | Tao Wei | Molecular Mechanisms for Zwitterionic Antifouling |
| 10:00 – 10:20 | Lucas Kreuzer | Zwitterionic Diblock Copolymer Thin Films: Responsive Behavior Under Different External Stimuli |
| 10:20 – 10:40 | Wolfgang Maison | Photochemical Grafting of Cationic–Zwitterionic Polymer Brushes from Titanium: Deciphering the Role of Charge for Biofilm Formation |
| 10:40 – 11:20 | <i>COFFEE BREAK</i> | |

Session chair: Karen Lienkamp

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|---------------|----------------------------|--|
| 11:20 – 11:40 | Jian Ji | Molecularly Interlaced Amphiphilic Block Copolymers for Robust Biofouling Resistance |
| 11:40 – 12:00 | Hana Vaisocherová-Lísalová | Programmable Zwitterionic Biointerfaces: From Molecular Mechanisms to Biomedical Applications |
| 12:00 – 12:20 | Ta-Chung Liu | Zwitterionics in Tuning Water State for Energy Storage |
| 12:20 – 14:10 | <i>LUNCH</i> | |

Session chair: Kevin Neumann

- | | | |
|---------------|---------------|---|
| 14:10 – 14:50 | Ayse Asatekin | Zwitterionic Amphiphilic Copolymers for Membranes for Ion and Organic Molecule Separations |
|---------------|---------------|---|

Program

Tuesday, 25 August 2026

14:50 – 15:10	Andriy Kuzmyn	SI-PET-RAFT Polymer Brushes for Biointerfaces and Scent Distinction
15:10 – 15:30	Roy Bernstein	Polyzwitterionic Brushes for Enhancing Membrane Antifouling Properties
15:30 – 15:50	Zhibin Ye	Fouling-resistant Zwitterionic Thin-film Nanocomposite Membranes for Dye-contaminated Wastewater Treatment
15:50 – 16:30	<i>COFFEE BREAK</i>	

Session chair: Ayse Asatekin

16:30 – 16:50	Kevin Neumann	Poly(ylides): A new Class of Overall Charge-neutral Hydrophilic Polymers
16:50 – 17:10	Julia Rho	Dynamic Stealth Polymers for Adaptive Cell Surface Engineering
17:10 – 17:30	Matthew Bernards	Synthesis of Peptide-based Zwitterionic Cross-linkers and their Evaluation in Polyampholyte Hydrogels
17:30 – 19:00	Poster Session cont.	
19:00	<i>HERAEUS DINNER</i> (social event with cold & warm buffet with complimentary drinks) Get together	

Program

Wednesday, 26 August 2026

08:00 *BREAKFAST*

Session chair: Shaoyi Jiang

09:00 – 09:40 Shaoyi Jiang **Low Immunogenic, High-efficacy, and Targeted Zwitterionic-based Lipid Nanoparticles for mRNA Delivery**

09:40 – 10:00 Zhan Chen **Probing Surface Hydration, Salt Resistance, and Protein Interaction of Zwitterionic Polymers in Situ to Elucidate Antifouling Mechanisms**

10:00 – 10:20 Amy Bassett **Tailoring Dielectric Properties of Polyimides through Zwitterion Functionalization**

10:20 – 10:40 Chen-Tsyr Lo **Zwitterionic Polymer Electrolytes for Lithium Metal Batteries: From Coordination Chemistry to Directional Ion Transport**

10:40 – 11:20 *COFFEE BREAK*

Session chair: Zhan Chen

11:20 – 11:40 Tomohiro Hayashi **Interfacial Interactions Induced by Anti-biofouling Self-assembled Monolayers**

11:40 – 12:00 Peng Zhang **Anion- π Interlocking Induced Supra-macromolecular Zwitterionic Hydrogel for Bioelectronics**

12:00 – 12:20 Ji-Hun Seo **Zwitterionic Polymer-inorganic Hybrid Hydrogel Coated Surfaces for Anti-icing/Frosting Application**

12:20 *LUNCH*

End of the seminar and departure

Posters

Poster Session

Katrin Ademmer	Exploring Swelling Behaviour and Antifouling Properties of Highly Crosslinked Zwitterionic
Aabha Bajaj	Zwitterionic copolymer nanodiscs for homogenous solubilization of liposomes - Efforts towards Membrane Protein Stabilization
Romina Berger	Tuning the UCST-Type Liquid-Liquid Phase Separation Behavior of Sulfobetaine Copolymers via Charge-Asymmetry
Negin Bouzari	Zwitterionic Stimuli-Responsive Hydrogels for Soft Robotic Applications
Michelle Buchholz	Interpolyelectrolyte Complexes of Chitosan and Poly(methacrylic acid): Tuning Nanostructure through Hydrophobic and Quaternary Ammonium Modifications
Chia Chih Chang	Light-Driven Polymerization to Afford Functional Hydrophilic Polymers with Anti-fouling Property, Programmable Degradation and Thermoresponsiveness
Tao Chen	Mimicking Color-Changing Organisms to Enable the Multicolors and Multifunctions of Smart Fluorescent Polymeric Hydrogels
Ying Nien Chou	Surficial Bonded Zwitterionic Modification for Anti-fouling and Biosensor Applications
Tatsuro Goda	Conformation-Dependent Cytosolic Translocation of Oligonucleotides Mediated by a Phospholipid-Mimicking Zwitterionic Copolymer
Changhao Hu	Thermo-responsive interpolyelectrolyte complexes (IPECs): molecular design, mesoscopic structure, drug solubilisation

Poster Session

Xiaohao Hu	Precision Functionalization and Electrophoretic Sorting of Amine-Functionalized Polymer-Coated Gold Nanoparticles
Chun Jen Huang	Functional Silatranes as a New Type of Building Blocks for Surface Engineering and Polymer Synthesis
Andrey Iljin	What if to cope ferroelectric nematics with zwitterionics?
Seokhyun Jang	Photoinduced Immobilization of Zwitterionic Polymer Coatings for Enhanced Antifouling Performance of Composite Resins
Markus Klapper	Twitter ionic polymers as surface active molecules on inorganic particle
Michelle Kobus	Plasma/Click-Functionalization of Plastics: Systematic Evaluation of Structure-Performance Relationships in Low-Fouling Zwitterionic Polymer Brushes
Yuto Kusunoki	Synthesis of Zwitterionic Surfactants for Nanoparticle Extraction and Structural Analysis of Self-Assembled Structures by Small-Angle Neutron Scattering
Eunji Lee	Comparative Mechanistic Analysis of Zwitterionic and Pseudo-Zwitterionic Hydrogel Coatings for Anti-Icing Applications
Kejing Li	Ionomer–Polyelectrolyte Complexes as Tunable Nanofiltration Membrane Materials
Mingzhong Li	Pharmaceutical Cocrystals: Route of Molecules to Patient-Centric Medicine

Poster Session

Peixun Li	Structure-property relationships in ethoxylated quaternary cationic and tertiary amine surfactants: impacts of molecular architecture and pH on interfacial behavior and self-assembly
Kunpeng Liu	Polyglycerol-based zwitterionic-like copolymer coating for Antifouling and Bactericidal Functions on Blood-Contacting PVC Devices
Ana Elena Moran Espinoza	Phosgene-Free Route to High-Purity Sarcosine N-Carboxyanhydride for the Controlled Synthesis of High-Molecular-Weight Polysarcosine
Alexander Münch	Toward Reusable Particle-Based Materials: Selective Recovery of Distinct Nanoparticulate Species from Model and Device Surfaces using Poly(phosphorylcholines) coatings
Nahieli Preciado Rivera	Injectable dynamic crosslinked zwitterionic hydrogels as a viscosupplement for osteoarthritis treatment
Martin Reifarth	Hydrophilic polymer brush surfaces for cell interface patterning
Noel Rene Schneider	Hydrophilic Poly(Iminopyridinium Ylide)s: Defining a New Chemical Space for Poly(ylide)s
Vladislav Semak	Polyzwitterionic Coating of Porous Adsorbents for Therapeutic Apheresis
Rene Soenderbaek Joergensen	A role for zwitterionic surfaces in advanced hull coatings -helping to decarbonize the shipping industry
Johannes Steffen	Enhancing the mechanical properties of a polyzwitterionic hydrogel

Poster Session

George Stiubianu	A role for zwitterionic surfaces in advanced hull coatings -helping to decarbonize the shipping industry
Kristopher Waynant	Synthesis, optimization, and evaluation of peptide-based zwitterionic cross-linkers and their use in polyampholyte materials
I Hsuan Yang	Zwitterionic Functional Polysaccharide Hydrogels via Charge-Balanced Design for Postoperative Adhesion Prevention
Kai Zhang	Bioinspired native nanostructured materials for energy generation

Abstracts of Lectures

(in alphabetical order)

Zwitterionic amphiphilic copolymers for membranes for ion and organic molecule separations

Ayse Asatekin¹

¹Tufts University, Department of Chemical and Biological Engineering, Medford, MA, USA

Most commercial membranes today are composed of a handful of polymers, developed decades ago. These formulations do not take advantage of the wide parameter space available to polymer scientists and engineers, including the self-organization capabilities of polymeric materials. Our group aims to rationally design polymeric materials that self-assemble into functional, biomimetic nanostructures to achieve enhanced membrane performance, with a particular focus on selectivity and fouling resistance. For example, zwitterionic amphiphilic copolymers (ZACs), comprising hydrophobic and zwitterionic monomers, form self-assembled zwitterionic domains that act as a network of nanochannels for water permeation. The effective pore size of ZAC-based thin film composite membranes can be tuned between 0.9-1.5 nm by controlling polymer chemistry and cross-linking. ZAC-based membranes with smallest pore sizes (~0.9 nm) can exhibit unprecedented selectivity between equally charged ions (e.g. Cl⁻/F⁻), arising from zwitterion-ion interactions amplified by nanoconfinement. Varying zwitterionic monomer chemistry in ZACs can lead to a wide range of ion selectivity patterns, linked with a complex range of effects apparently mediated by factors that include the chemical structures of ions forming the zwitterion, their order from the polymer backbone, the presence of pendant groups, and functional groups linking the zwitterion to the backbone. In some cases, this results in highly unusual selectivity patterns that are not predicted by common membrane transport models emerge, including ability to separate like-charged ions. In others, selectivity between uncharged small molecule solutes of similar size (e.g. sugars and anti-oxidants) can be achieved. As such, well-designed ZAC-based membranes have the potential to impact a wide range of industrial and environmental separations, from metal recovery from mine drainage to the separation of rare earth elements to chemical manufacturing.

Tailoring Dielectric Properties of Polyimides through Zwitterion Functionalization

Amy E. Honnig Bassett^{1,2}, Emily C. Davidson², Rodney D. Priestley²

¹*Andlinger Center for Energy and the Environment, Princeton University, Princeton, USA*

²*Department of Chemical and Biological Engineering, Princeton University, Princeton, USA*

Conventional dielectric polymers used in capacitors suffer from two significant drawbacks: (1) limited operating temperatures ($< 105\text{ }^{\circ}\text{C}$) and (2) low dielectric permittivity ($\epsilon' \leq 3.5$). These challenges hinder their application in advancing the electrification of transportation, where temperatures can approach $300\text{ }^{\circ}\text{C}$. Additionally, low ϵ' results in reduced energy density, inefficient energy utilization, and the need for bulkier devices. To address these challenges, a series of carboxybetaine zwitterionic polyimides with increasing zwitterion content was synthesized. Zwitterions were selected to functionalize polyimides because their large dipole moment is expected to increase ϵ' . Furthermore, pendant zwitterions on flexible polymers are known to increase the glass transition temperature (T_g) through dipole-dipole associations. We sought to combine the benefits of zwitterions (increased ϵ' and T_g) with those of polyimides (high T_g and thermal stability) to simultaneously address the two main concerns of conventional dielectric polymers.

After synthesizing the zwitterionic polyimides, the dielectric and thermal properties were characterized. To evaluate ϵ' , broadband dielectric spectroscopy (BDS) was utilized, while the thermal stability was assessed using thermogravimetric analysis (TGA), and the T_g was determined via differential scanning calorimetry (DSC). Introducing zwitterions onto the polyimide backbone increased ϵ' at $25\text{ }^{\circ}\text{C}$ and 1 kHz relative to the non-zwitterionic polyimide, and ϵ' remained constant with increasing frequency. However, zwitterionic polyimides exhibited a reduction in T_g and onset of decomposition relative to the non-zwitterionic polyimide, suggesting a lack of dipole-dipole associations. Nevertheless, the zwitterionic polyimides exhibited T_g values greater than $185\text{ }^{\circ}\text{C}$, maintaining the favorable high T_g characteristic of polyimides. The lack of dipole-dipole associations likely resulted from the system being in a glassy state due to the high T_g and the highly aromatic polyimide backbone, which impeded the mobility of the zwitterions. This work illustrates the potential of zwitterionic polymers to enhance the dielectric properties and marks the first report evaluating the dielectric properties of a zwitterionic polyimide.

Synthesis of peptide-based zwitterionic cross-linkers and their evaluation in polyampholyte hydrogels.

Adrienne Shea¹, Skyler Oneida², Robert Hubbard², Kristopher Waynant², and Matthew Bernards¹

¹Chemical and Biological Engineering and ²Chemistry, University of Idaho, Moscow, ID

Polyampholyte chemistries are widely demonstrated to have nonfouling properties when their charged monomer subunits remain balanced and well distributed. However, long-term applications of these materials are hampered by the lack of zwitterionic cross-linker species available both commercially and in literature. Our recent efforts have focused on synthesizing peptide-based zwitterionic cross-linkers groups and evaluating the influence of these cross-linkers on the nonfouling performance of polyampholyte hydrogels. Specifically, we have synthesized cross-linkers comprised of serine (Ser-Ser) and diaminopropanoic acid (Dap-Dap) and incorporated these cross-linkers into polyampholyte hydrogels composed of equimolar mixtures of [2-(acryloyloxy)ethyl] trimethylammonium chloride (TMA) and 2-carboxyethyl acrylate (CAA) (TMA/CAA). Physical characteristics (swelling, percent hydration, elastic modulus, etc.) have been quantified and compared to control hydrogels synthesized with diethylene glycol dimethacrylate (DEGDMA) as a historic nonfouling control and carboxybetaine dimethacrylate (CBMAX) as the literature standard. Nonfouling performance has been evaluated for these cross-linkers following repeat exposure to *Ralstonia (R.) pickettii* over 28 days. In these studies, all zwitterionic cross-linkers (<0.04% surface coverage) significantly outperform the DEGDMA cross-linked systems (3.7% surface coverage). Further, after 28 days of repeat exposure, the Ser-Ser cross-linked hydrogels statistically distinguish themselves from the other zwitterionic cross-linkers (0.03-0.04% versus <0.005% surface coverage). Similar performances were seen in evaluations of bacterial surface coverage for thin film hydrogels formed on stainless steel substrates in gravity impacted, simulated microgravity, and microgravity-based experiments. Collectively, the results demonstrate the importance of cross-linker design, especially for long duration and complex media.

Polyzwitterionic Brush Architectures for Advanced Water Treatment Membranes

E Ziemann, H Huang, M Yang, M Halakarn and R. Bernstein

Zuckerberg Institute for Water Research, Jacob Blaustein Institutes for Desert Research, Ben-Gurion University of the Negev, Sde Boker Campus, 84990, Israel

Polyzwitterions have been extensively investigated as antifouling surface coatings due to their strong hydration capacity and charge neutrality. When grafted as polymer brushes—polymer chains tethered to a surface at high grafting density—polyzwitterions form a dense hydration layer and a steric barrier that reduce foulant–surface interactions and facilitates foulant removal, making them attractive for developing antifouling membranes.

Despite their potential, research on grafting polyzwitterionic brushes onto membranes has been relatively limited. Conventional membrane materials are often chemically inert and have limited chemical stability, making surface-initiated reversible-deactivation radical polymerization techniques difficult to implement. In addition, direct characterization of brush layers on porous membranes is challenging. Moreover, the relationships between brush chemistry, grafting density, architecture, and membrane performance under realistic permeation conditions have not been systematically studied.

In recent years, we have been investigating polyzwitterionic brush-modified ultrafiltration and reverse osmosis membranes for various water treatment applications. We have been studying how linear, branched, three-dimensional, and bottle-brush architectures influence fouling and scaling resistance, as well as membrane permeability and selectivity, under realistic filtration conditions. We also employed surface-sensitive characterization techniques to better understand the antifouling behavior. These studies highlighted the excellent antifouling properties achievable through grafting polyzwitterionic brushes. They also showed that membrane performance strongly depends on brush architecture, thickness, and grafting density, which can help guide the design of advanced antifouling membranes for water treatment applications. We continue investigating the role of zwitterionic chemistry, other brush architectures, the use of brushes to enhance selectivity, and simpler methods for grafting polymer brushes onto membranes.

References

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- [2] H. Huang, Y. Tian, D. AbuKhadra, N. Maman, R. Bernstein, *Journal of Membrane Science*, 124537 (2025).
- [3] E. Ziemann, T. Coves, Y. S. Oren, N. Maman, R. Sharon-Gojman, et al., *Science Advances* 10, eadm7668 (2024).
- [4] E. Ziemann, J. Qin, T. Coves, R. Bernstein, *Journal of Membrane Science* 672, 121422 (2023).

Molecular Engineering of Zwitterionic Membranes for Preferential Blood Cell Separation

Yung Chang

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Blood cell separation is a fundamental enabling technology for transfusion medicine, immunotherapy, regenerative medicine, and precision healthcare. Current separation platforms largely depend on differences in cell size, density, or affinity-based recognition; however, these approaches frequently involve complex processing, limited selectivity, and undesirable activation of blood cells. Inspired by the highly hydrated and dynamically regulated interfaces found in biological systems, we have established a molecular engineering strategy for zwitterionic membranes that enables preferential blood cell separation through precise manipulation of interfacial hydration and electrostatic interactions. This presentation will highlight our recent advances in the molecular design of bio-inspired zwitterionic membrane interfaces based on well-defined polymer brushes and charge-regulated pseudozwitterionic polymer networks. By precisely tailoring molecular architecture, grafting density, and charge distribution, the hydration layer and interfacial physicochemical properties can be systematically engineered to regulate protein adsorption and cell–surface interactions. Rather than functioning solely as antifouling materials, these zwitterionic interfaces provide a programmable platform for selectively modulating the adhesion and transport of specific blood cell populations. We further demonstrate that pseudozwitterionic membrane systems with tunable positive and negative charge compositions generate controllable charge-biased hydration environments that promote preferential interactions with targeted blood cells. The synergistic regulation of electrostatic interactions and interfacial hydration enables highly selective cell capture, enrichment, and separation while maintaining excellent hemocompatibility and cellular integrity. These molecularly engineered membranes have been successfully translated into a variety of biomedical applications, including leukocyte depletion, platelet enrichment, immune cell isolation, extracellular vesicle purification, and membrane-based blood processing technologies. In summary, our work demonstrates that molecular engineering of zwitterionic membranes establishes a new design paradigm for controlling cell–material interactions beyond conventional size-exclusion and affinity-based separation mechanisms. By integrating membrane science, polymer chemistry, and biomimetic interfacial engineering, this research provides a universal molecular design framework for next-generation bioseparation membranes and opens new opportunities for precision blood processing, regenerative medicine, immunoengineering, and advanced cell manufacturing.

References: G. V. Dizon, K.-L. Liang, A. Venault, A. Doche, T.-Y. Yin, Y.-Y. Tan, C.-J. Chou, C. Causserand, P. Bacchin, Y. Chang*, *Chemical Engineering Journal*, **2026**, 537, 176328; C.-C. Yeh, A. Venault, G. V. Dizon, P.-R. Lin, K.-L. Liang, S.-H. Liou, C.-J. Chou, P. Bacchin, Y. Chang*, *Separation and Purification Technology*, **2025**, 354, 129013; A. Venault, B. C. Wu, I. V. Maggay, Y. Chang*, *Journal of Membrane Science*, **2024**, 708, 123076; A. Venault, Y.-C. Chen, S.-H. Tang, T. C. Wei, C. F. Weng, P. Bacchin, Y. Chang*, *Journal of Membrane Science*, **2023**, 683, 121803.; Y. Fu, H.-J. Hong, A. Venault, Y. Chang*, *Journal of Membrane Science*, **2023**, 662, 121014.; A. Venault, R.-J. Zhou, T. A. Galeta, Y. Chang*, *Journal of Membrane Science*, **2022**, 658, 120760.

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Probing Surface Hydration, Salt Resistance, and Protein Interaction of Zwitterionic Polymers in Situ to Elucidate Antifouling Mechanisms

Zhan Chen

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Zwitterionic materials exhibit outstanding antifouling properties and have been extensively investigated for applications in biomedical devices, drug delivery systems, and marine coatings. Their exceptional antifouling performance is generally attributed to the strong hydration layer formed at their surfaces. However, direct in situ characterization of this hydration layer is challenging because it exists at buried solid/liquid interfaces. Our research has successfully employed sum frequency generation (SFG) vibrational spectroscopy to investigate the surface hydration of several zwitterionic polymers, including poly(carboxybetaine), poly(sulfobetaine), and poly(trimethylamine N-oxide) (pTMAO). Strong surface hydration was observed for all of these zwitterionic polymers. SFG spectroscopy was also applied to zwitterionic polymers prepared by initiated chemical vapor deposition (iCVD), revealing substantially stronger surface hydration than that observed for corresponding non-zwitterionic control polymers.

SFG studies further demonstrated that protein adsorption does not disrupt the strongly hydrated interfacial water layer on zwitterionic polymer surfaces, whereas the hydration layer on polyethylene oxide (PEO) surfaces can be significantly perturbed by proteins. These findings highlight an important advantage of zwitterionic materials over conventional antifouling polymers. Our investigations also revealed that mussels are reluctant to settle on zwitterionic polymer surfaces. However, when mussels are forcibly maintained on such surfaces (e.g., using a rubber band), they can develop strong adhesion. SFG measurements showed that mussels utilize both interfacial water molecules and adhesive proteins to establish strong attachment to zwitterionic polymer surfaces. This behavior differs markedly from that observed on conventional fouling surfaces, where mussels typically remove interfacial water and dehydrate the surface before adhesion. SFG spectroscopy was further applied to study barnacle attachment on zwitterionic polymers. In contrast to mussels, barnacles were unable to exploit interfacial water to enhance adhesion on zwitterionic polymer surfaces.

In addition, our SFG studies demonstrated that dissolved salts can weaken the surface hydration of zwitterionic polymers through electrostatic charge-screening effects, potentially compromising their antifouling performance. Nevertheless, the results showed that zwitterionic materials with short charge-separation distances can maintain strong surface hydration even under high-salt conditions. Poly(trimethylamine N-oxide), which possesses an exceptionally short charge distance, exhibits remarkable resistance to salt-induced screening effects and retains robust surface hydration in saline environments.

Interfacial interactions induced by anti-biofouling self-assembled monolayers

Tomohiro Hayashi

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Bioinert coatings are critical in fields such as medical biosensors, artificial blood vessels, and marine antifouling coatings. Among the various anti-biofouling chemistries, zwitterionic surfaces carrying balanced positive and negative charges are regarded as among the most effective. Although many anti-biofouling strategies have been reported, their underlying mechanisms differ. For polymer brushes, the rejection of biomolecules and microorganisms is attributed to steric repulsion from hydrated chains—the “volume exclusion effect.” This picture, however, is inadequate for anti-biofouling self-assembled monolayers (SAMs), including zwitterionic mixed-charge (MC) SAMs of equimolar positively and negatively charged thiols, where the molecules are too densely packed for such an effect to operate. [1, 2]

In this presentation, we explore the fundamental interactions responsible for the bioinertness of zwitterionic MC-SAMs. We directly probed the interface by atomic force microscopy (AFM) to measure surface forces and used surface-sensitive infrared absorption spectroscopy to characterize the interfacial water structure. Our results demonstrate that the robust anti-biofouling performance of these zwitterionic SAMs originates from a stable interfacial water layer, several nanometers thick, that effectively prevents bio-adhesion. [3-5]

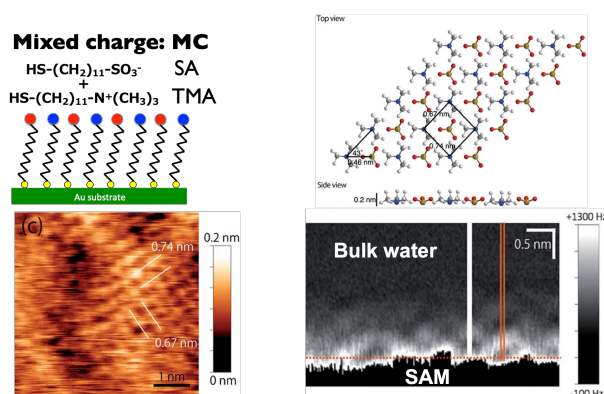


Fig. 1. (upper left) Chemical structure of the molecules constituting the anti-biofouling zwitterionic mixed-charge (MC) SAM. (upper right) A possible packing configuration of the molecules based on the AFM images. (bottom left) Frequency-modulation (FM) AFM image of the MC-SAM. (bottom right) Cross-sectional FM-AFM image of the water-SAM interface.

References

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MPC Polymer Science and Engineering: Shaping the Future of Biointerfaces and Medical Devices

Kazuhiko Ishihara

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Medical devices are essential in modern medicine, playing a central role in life-saving treatments and improving patients' quality of life (QOL). Achieving high clinical performance and long-term reliability requires precise control of biological reactions at the device-tissue interface. From a top-down perspective, the functional requirements of medical devices define the necessary interface properties, which guide the molecular design of the materials. This presentation will provide a comprehensive overview of the science and engineering of 2-methacryloyloxyethyl phosphorylcholine (MPC) polymers as a platform for advances in interface design based on biomimetic principles [1,2]. Since the first report of smart processes for synthesis and purification in 1990 and the commencement of industrial production in 1999, MPC polymers have driven significant progress in the field of interface science. MPC polymers exhibit excellent biocompatibility by effectively inhibiting protein adsorption and cell adhesion, minimizing unfavorable biological reactions. These superior properties on the MPC polymer surface are based on the unique hydration state of the phosphorylcholine groups in the MPC polymer. As a result, MPC polymers are now widely used worldwide as a surface modification technology for a wide range of medical devices, making a significant contribution to improving clinical outcomes and enhancing patients' quality of life [3,4]. Furthermore, MPC polymers provide a versatile platform for producing functional nanoparticles and hydrogels with excellent cytocompatibility, opening up new directions in cell engineering for controlling cellular function [5,6]. How MPC polymer science and engineering are driving innovation and shaping the future of biointerfaces and medical devices by linking clinical needs with interface control and molecular design will be highlighted.

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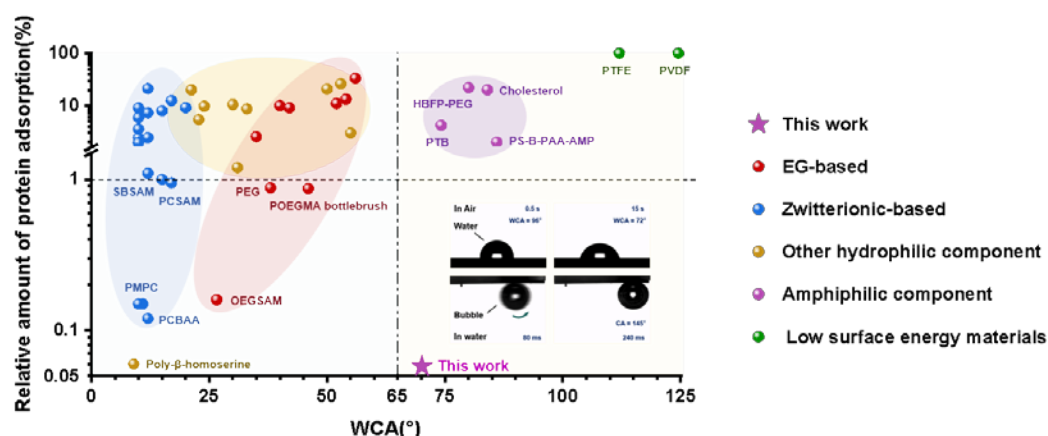
Molecularly Interlaced Amphiphilic Block Copolymers for Robust Biofouling Resistance

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Although minimizing nonspecific bioadhesion is of crucial importance in biomedical, marine, and industrial applications, current strategies rarely employ hydrophobic materials due to their limited water-binding capacity¹⁻². In this work, we developed molecularly interlaced triblock copolymers (PLA-PEG-PLA, PLEL) to fabricate hydrophobic, biofouling-resistant materials. Despite having an 80% hydrophobic content, this polymer demonstrates remarkable protein resistance, comparable to that of zwitterionic materials, with protein adsorption as low as 0.5% of that of PLA. Wetting and adsorption experiments substantiate that the repellent properties of the interface stem from its specific molecular architecture and block composition. Different from conventional anti-fouling interfaces that depend on pre-designed templates, water molecules actively engage in the dynamic adjustment of the PLEL interface, triggering chain rearrangement to establish an optimal anti-fouling state. Moreover, PLEL analogs with substituted hydrophobic monomers also display exceptional anti-fouling performance while preserving interfacial chain mobility. These findings may necessitate a reevaluation of the current consensus on nonfouling structures, providing new perspectives for the molecular design of next-generation anti-fouling materials.



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Low Immunogenic, High-efficacy, and Targeted Zwitterionic-based Lipid Nanoparticles for mRNA Delivery

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Lipid nanoparticle (LNP)-based mRNA vaccines have shown great success against COVID-19. However, there are three major barriers to using mRNA-LNPs for applications beyond COVID-19 vaccines such as cancer vaccines and autoimmune diseases. The first challenge is LNP immunogenicity, resulting from the inflammatory response triggered by LNP components, particularly ionizable and poly(ethylene glycol) (PEG) lipids. In addition, injection-site inflammation is a concern. The second challenge comes from inefficient endosomal escape of the mRNA. Once internalized by cells, much of the mRNA remains trapped in endosomes and fails to reach the cytoplasm. The current LNP formulations have very low endosomal escape efficacy. The third challenge is off targeting. For an intravenous injection, LNPs accumulate in the liver. Even though LNPs are taken up by the spleen, they are often cleared by macrophages before they can reach the immune cells within the spleen.

In this talk, I will discuss our efforts to tackle these barriers using the concept of zwitterionic materials. A poly (carboxybetaine) (PCB) lipid was used to replace the PEG lipid, enabling multiple injections of mRNA-LNPs [1]. A membrane-destabilizing zwitterionic (MeDZ) lipid was designed, synthesized, and applied to improve endosomal escape [2,3]. Various zwitterionic LNPs were developed to achieve secondary lymphoid organs (SLOs)-targeting delivery [4,5] and extrahepatic, non-phagocytic and splenic immune cell-resolved mRNA delivery. Furthermore, zwitterionic peptides or polymers are shown to extend protein circulation from mRNA-LNPs [6] and reduce inflammation around intramuscular (IM) injection sites. These mRNA-LNPs are developed for applications associated with vaccines and immunotherapies for infectious diseases, cancer, and autoimmune diseases.

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Zwitterionic Diblock Copolymer Thin Films: Responsive Behavior Under Different External Stimuli

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Zwitterionic polymer thin films exhibit responsive behavior in confined geometry that differs markedly from solution. This talk presents a systematic study of orthogonally dual thermo-responsive diblock copolymer thin films composed of a zwitterionic poly(sulfobetaine) (PSB) block and a nonionic poly(N-isopropylmethacrylamide) (PNIPMAM) block. The two blocks respond inversely to temperature: PSB undergoes a coil-to-globule transition upon cooling (UCST-type), while PNIPMAM collapses upon heating (LCST-type).

Using time-of-flight neutron reflectometry, grazing-incidence small-angle neutron scattering, and FTIR spectroscopy, the thin film behavior is tracked in situ, linking mesoscopic film characteristics such as swelling degree and solvent content to molecular-level polymer–solvent interactions and hydration states.

Key findings include a pronounced isotope sensitivity of the PSB block, leading to preferential H₂O uptake over D₂O. In mixed water/methanol vapors, both blocks independently exhibit co-nonsolvency-type behavior reflecting their distinct chemical natures. Phase transitions in thin film geometry are significantly shifted and slower than in solution, consistent with increased polymer concentration and restricted chain mobility. Salt addition (NaBr) selectively modulates the PSB transition temperature via a salting-in effect, leaving PNIPMAM unaffected. These results establish a highly tunable set of thin film regimes with potential applications in nanosensors, nanoswitches, and functional surfaces.

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SI-PET-RAFT Polymer Brushes for Biointerfaces and Scent Distinction

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Surface-initiated photoinduced electron-transfer reversible addition–fragmentation chain-transfer polymerization (SI-PET-RAFT) has emerged as a versatile and scalable method for producing well-defined polymer brush coatings under mild, oxygen-tolerant, and metal-free conditions. The approach enables precise control over brush chemistry, thickness, and architecture, allowing the fabrication of functional surfaces for sensing, antifouling, and biointerface applications.

Polymer brushes prepared via SI-PET-RAFT can be designed with diverse chemical functionalities, including zwitterionic, hydrophilic, and hydrophobic structures. Arrays of such coatings were employed to generate distinct mass-absorption “fingerprints” for volatile organic compounds (VOCs) and complex scents, enabling proof-of-concept olfactory sensing using quartz crystal microbalance with dissipation monitoring (QCM-D). This concept demonstrates how chemically tuned polymer brush interfaces can act as reusable sensing elements for detecting scent mixtures and environmental VOCs.

Beyond chemical sensing, SI-PET-RAFT brushes also enable dynamic biointerfaces. Thermoresponsive poly(di(ethylene glycol) methyl ether methacrylate) brushes synthesized via this method exhibit reversible conformational switching with temperature, enabling controlled cell adhesion and release without harsh regeneration procedures. Such switchable coatings allow repeated use of biosensing platforms and facilitate studies of cell–surface interactions.

Furthermore, polymer brushes produced by SI-PET-RAFT can undergo thermally induced depolymerization. Complete removal is observed for ultrathin layers ($\approx 3\text{--}4$ nm), whereas thicker brushes show partial depolymerization with decreasing conversion as thickness increases. These properties highlight the potential of SI-PET-RAFT coatings as reusable sensing interfaces and reprocessable surface materials for detecting protein traces, scents, and environmental analytes.

Polyzwitterions: Achievements, Myths & Fancy

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Polyzwitterions are neutral compounds bearing pairs of electrical charges of opposite sign. While first described in the 1950ies, the interest in polyzwitterions (originally considered rather as "exotic academic fruits") set in only slowly during the late 1970ies, but has been growing markedly ever since. This is reflected by the increasing numbers of review and feature articles dealing with these polymers –reaching more than 100 papers in peer reviewed chemical journals during the past five years–, the annual numbers growing steadily (NB: their scope and quality varying vastly). Much of the interest is purely application-driven, stretching nowadays from the originally aspired mimics of gelatine and lipid cell membranes via absorbents and robust hydrogels to coatings for solar cells, solid state electrolytes, or highly polarizable materials for energy storage. Still, the focus of polyzwitterions' has been on the use for low-fouling, "stealth", bio- and hemocompatible surfaces. Impressive practical results have been achieved with polyzwitterions over the years in various fields, while our theoretical understanding and knowledge of structure-property relationships remain limited.

Yet, the –often uncritical– enthusiasm about this polymer class risks to obscure our understanding, what are genuine or even truly unique features of polyzwitterions, when their role has rather a "me, too" character, or where the term is merely employed as buzz word. Moreover, the label "polyzwitterion" is increasingly misattributed, due to misinterpreted or incomplete chemical analytical data, improper formalistic generalizations, or confusions about the meaning of chemical formulas (and the limits of an intuitively interpreted valence bond theory). Before this background, the contribution is intended as a thought-provoking impulse, to value but demystify polyzwitterions and their uses, and to revisit our putatively 'established' knowledge of polyzwitterions to identify weak spots where we still stand on shaky grounds.

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Polyzwitterionic Hydrogels: Beyond Biofilm Formation and Biocompatibility

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Polyzwitterionic molecules have been intensely studied with respect to their chemical, physical and biological properties. In this context, important properties such as their unusual solubility and anti-polyelectrolyte effect, their prevention of protein adhesion and biofilm formation, and lack of toxicity for various cell types have been described. [1, 2, 3] Yet despite their excellent bioactivity profile, the uses of polyzwitterions in practice are limited both in number and type. Currently, applications focus on thin, surface-attached polymer films, and on the well-known poly(2-methacryloyloxyethyl phosphorylcholine), which was commercialized in numerous products such as contact lenses and stents.[4]

The aim of this contribution is to highlight the challenges involved in turning polyzwitterionic molecules into bulk materials, e.g., polyzwitterionic hydrogels, for applications such as wound dressings, soft robotics, highly ion conductive materials or low friction surfaces. The dominating challenge in this context is improving the poor mechanical properties, with elastic moduli typically in the mid-kPa range.[5] So far, this is remedied by double or interpenetrating network strategies using other polymers, which however may hamper the typical polyzwitterion properties. We present how molecular engineering contributes to enhanced mechanical properties of the resulting polyzwitterionic hydrogels. Special emphasis is placed on achieving maximum molar masses and long-chain entanglements, and on studying the shear-thinning properties of these polymers, which are important for processing of polyzwitterions from solutions into the desired shapes.

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Zwitterionics in Tuning Water State for Energy Storage

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Our previous work first demonstrated a quasi-solid-state hydrogel electrolyte based on the concept of interfacial wetting water, where ion transport is governed by two-dimensional migration along hydrated interfaces rather than conventional diffusion through bulk-like water. This interfacial water-dominated transport mechanism enabled a 2.5 V electrochemical window together with a low water content of 24 wt%. The hydrogel also exhibited excellent mechanical robustness, with an ultimate tensile strength of 420 kPa and an elongation at break of 6000%, allowing flexible supercapacitors to deliver high energy density, high power capability, and suppressed self-discharge.

Building upon this concept, we further developed an ultra-water-deficient hydrogel electrolyte (UWDHE) for flexible zinc batteries by integrating a zwitterionic-bias polymeric network with the chaotropic salt $\text{Zn}(\text{ClO}_4)_2$. This strategy extends the interfacial water concept by simultaneously regulating the hydrogen-bond network, Zn^{2+} solvation structure, and interfacial ion distribution, thereby decoupling ionic transport from water content. The optimized electrolyte contains only ~11 wt% water, yet achieves a high ionic conductivity of 60 mS cm^{-1} and a Zn^{2+} transference number of 0.84. In an open-cell Zn//Zn configuration, it enables stable Zn plating/stripping for over 2,000 h.

Together, these studies establish a coherent design strategy for lean-water hydrogel electrolytes, in which engineering the interfacial water environment and hydrogen-bond network enables high ionic conductivity, superior mechanical flexibility, and highly reversible electrochemical reactions under extremely low water content. This work provides a promising foundation for the development of safe, flexible, and high-performance aqueous energy-storage systems.

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Zwitterionic Polymer Electrolytes for Lithium Metal Batteries: From Coordination Chemistry to Directional Ion Transport

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Lithium metal batteries are promising energy-storage systems with high energy density; however, their development remains limited by the insufficient ionic conductivity, low lithium-ion transference number, and unstable electrode interfaces of polymer electrolytes. Zwitterionic polymers contain covalently linked cationic and anionic groups and can promote lithium-salt dissociation, restrict anion migration, and regulate interfacial ion distribution through strong dipolar and coordination interactions.

This presentation first introduces the molecular structures and synthetic strategies of zwitterionic polymers and discusses their structure–ion transport relationships. Zwitterion-based solid, gel, and composite polymer electrolytes for lithium metal batteries are then systematically reviewed. A zwitterionic–single-ion conducting nanofiber composite electrolyte is subsequently presented, in which the continuous nanofibrous framework and immobilized anionic groups improve selective Li⁺ transport and membrane stability. Finally, coordination-driven zwitterionic core–shell polymers are discussed as functional nanostructured transport units. Controlled polymerization, self-assembly, and selective crosslinking are employed to construct continuous and low-tortuosity ion-conduction pathways. By integrating molecular coordination, nanostructural engineering, and interfacial regulation, this presentation highlights the development of zwitterionic polymer electrolytes from lithium-salt dissociation toward directional Li⁺ transport.

Keywords: zwitterionic polymers, lithium metal batteries, solid polymer electrolytes, single-ion conductors, directional ion transport

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Photochemical Grafting of Cationic–Zwitterionic Polymer Brushes from Titanium: Deciphering the Role of Charge for Biofilm Formation

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Titanium is an important implant material, and long-term clinical performance critically depends on how its surface is functionalized. Here we report a light-driven strategy for covalently attaching cationic¹ and zwitterionic² polymer brushes to titanium using surface-initiated photoiniferter-mediated synthesis. Starting from photochemically activatable phosphonate anchors,³ we synthesized zwitterionic and cationic polymer brushes, as well as hierarchical copolymer brushes that incorporate quaternary ammonium groups and zwitterionic moieties (sulfobetaines and *N*-oxides). We have systematically varied both the fraction of cationic vs. zwitterionic groups and their spatial position within the brush architecture. This tunability gave a spectrum of materials with contact-active to non-adhesive properties. These materials were tested against clinically relevant microorganisms. The amount and position of positively charged quaternary ammonium sites was gradually modified to dissect how charge density and hydration of the polymer brushes affect (i) bacterial biofilm formation, (ii) biofilm growth kinetics, (iii) microbial viability, and (iv) microbial adhesion. Our data reveal subtle yet significant effects of charge distribution on microbial colonization. Quantitative confocal microscopy combined with SEM-EDX demonstrates that adhesion is strongly influenced not only by the number of cationic groups but also by their positioning within the polymer brush.

Overall, the presentation highlights a photochemical method for engineering metal surfaces with charged and zwitterionic brush polymers. It provides also a mechanistic insight into the influence of charged polymer surfaces on the colonization of microorganisms and the tailored design of contact-active *and* non-adhesive implant surfaces.

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Poly(ylides): A new class of overall charge-neutral hydrophilic polymers

K. Neumann¹

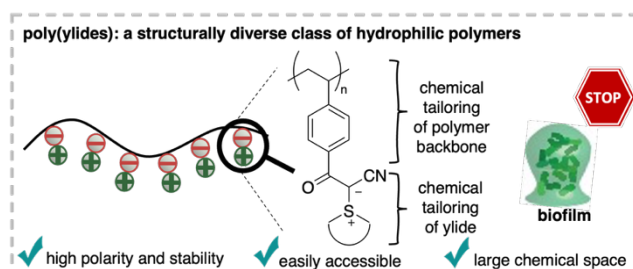
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My group rationally designed polymeric ylides as a new form of overall-charge neutral polymers with zwitterionic character and demonstrated that polymeric sulfur and phosphorus ylides successfully prevent biofilm formation by pathogenic bacteria.¹⁻³

Structurally, polymeric ylides differ significantly from polyampholytes and polybetaines by having the negative directly adjacent to the positive charge, minimizing the dipole and providing sterically non-demanding residues. While polymeric sulfur ylides display toxicity towards bacteria, they do not affect eukaryotic cells, making them attractive materials for medical coatings. In addition, we gained an in-depth understanding of structure-property relationships by accessing diverse polymer structures with varying backbone- and ylide scaffolds.⁴ Most recently, we fabricated ylide-terminated PEG segments, termed *y*PEGs, and examined their potential to prevent immune recognition.⁵

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Dynamic Stealth Polymers for Adaptive Cell Surface Engineering

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Living cell therapeutics hold immense promise for treating cancer, regenerating tissues, and correcting genetic disorders. However, a fundamental challenge to their clinical translation is that the immune system rapidly recognises and eliminates engineered cells before they can exert therapeutic effects. Polymer-based stealth coatings have emerged as a promising strategy to extend cell circulation times by shielding surface antigens from immune surveillance.^{1,2} Whilst current approaches using poly(ethylene glycol) and zwitterionic polymers successfully prolong cell survival, these static coatings present a critical trade-off: they indiscriminately mask cells, often blocking essential receptor-mediated functions such as tumour targeting, antigen recognition, and tissue homing.

Natural cell membranes are inherently dynamic, constantly remodelling their surface composition in response to environmental cues. Our research aims to replicate and enhance this adaptive behaviour through rational polymer design. We are developing stimuli-responsive polymer coatings that intelligently modulate their stealth properties in response to biological triggers, including pH gradients in tumour microenvironments, enzymatic activity at inflammatory sites, and receptor engagement during cellular activation. Our initial findings demonstrate that block copolymer architecture critically influences polymer-cell interactions and, in turn, the cell-surface landscape. By bridging polymer chemistry, super-resolution imaging, and cell biology, we believe this research will open new pathways for precision medicine.

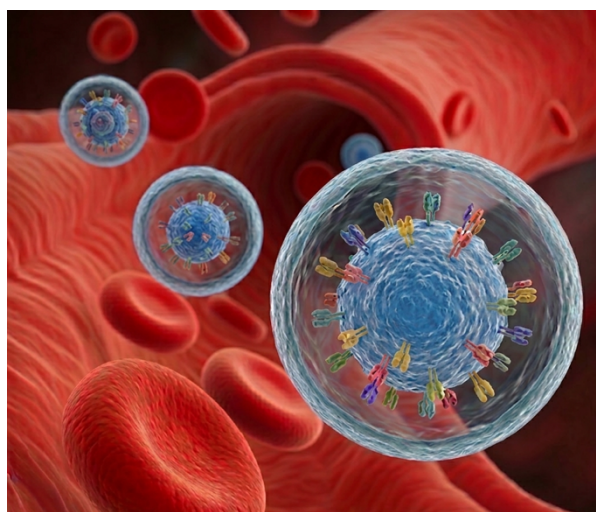


Figure 1. Schematic depiction of dynamic polymer stealth coating for cell surface modification.

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Bioinspired and Fermented Materials for Environmental Applications

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The reliability and ease of operation of membrane-based water purification systems has led to their increased use in water and wastewater treatment. However, water and energy are mutually dependent critical resources; to produce clean water requires energy and the production of energy requires large volumes of water. The overall goal of this talk is to illustrate how green chemistry and microbially-derived approaches can be used to transform the manufacturing industry. First, I will provide an overview of how we use only water and salt to manufacture materials from pairings of oppositely charged synthetic and natural polymers. We have demonstrated that by exploiting salt-driven plasticization, we can enable the formation of membranes, as well as additional materials, such as robust textiles and antifouling coatings. If time allows, I will also introduce our newest non-woven membranes that are a byproduct of the kombucha tea fermentation process. These cellulose membranes hold great promise for use as ultrafiltration membranes. The overall goal of this talk is to highlight some of our recent findings and how these results can guide the green engineering of multifunctional materials that improve human health and the environment.

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How Are “Electrostatic” Interactions Driven by Charge Pairing and Dehydration?

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Charge neutrality and hydrophilicity are recognized as the main contributors to the effectiveness of zwitterionic coatings at reducing nonspecific adsorption. Attractive or repulsive interactions between opposite- or like-charges are intuitively termed “electrostatic.” However, in aqueous environments, charges on macromolecules or surfaces are generally compensated by counterions, which neutralize electric fields. Thus, long-range Coulombic interactions, which lead to strong interactions between charged species in vacuum, are difficult to translate to salty solvents without a few significant approximations. Coulombic attractions between opposite charges should be exothermic in nature, whereas many “electrostatic” associative processes such as complex coacervation and polyelectrolyte adsorption are endothermic, implying significant entropic contributions.

The importance of counterion rearrangement or release in driving interactions between opposite/like charges was recognized decades ago. For example, after measuring no heat changes in polyelectrolyte complexation, Michaels attributed complexation/coacervation to the “escaping tendency of microions.” We prefer the term “charge pairing” to “electrostatics” because it more clearly describes the short-range association of one positive with one negative charge, leaving open the possibility that the process is *not* driven by long-range Coulombic attraction but instead by concentration gradients and more specific interactions such as hydration, π -cation, π - π and hydrophobic forces.

The main contributions to entropy changes during association/condensation are counterion loss and water loss (dehydration). Of these, counterion loss is easier to quantify: one simply compares ion concentrations in the charge paired material (CPM) and the solution. Accounting for entropic contributions from water loss is more difficult. Water moves from a lower concentration (in the CPM) to higher (bulk water), so rotational and vibrational entropy gains must be invoked. There is an indirect mechanism: loss of water increases the charge density of the CPM therefore enhancing the entropy gain from ion loss.

This talk will focus on quantitative aspects of entropic and enthalpic driving forces for charge pairing, including the role of hydration.

Zwitterionic polymer-inorganic hybrid hydrogel coated surfaces for anti-icing/frosting application

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The formation of ice and frost on chilled surfaces operating in humid environments is a persistent challenge, particularly in heat exchangers where frost layers severely impede heat transfer and reduce overall energy efficiency. Although polymer gels have recently attracted considerable attention as anti-icing materials, conventional hydrogels often exhibit mechanical robustness issues and swell in humid environments. Achieving reliable anti-icing performance, therefore, requires materials capable of regulating interfacial water structure while maintaining stable behavior during continuous operation. This study proposes a silica–zwitterionic polymer hybrid gel that addresses both requirements through hydration-state confinement enabled by an inorganic–organic co-network. The hybrid gels are fabricated with various swelling ratios by tuning the composition ratios of silica and the zwitterionic polymer, 2-methacryloyloxyethyl phosphorylcholine (MPC), followed by a detailed characterization of water contents, quantification of icing and frost growth, and evaluation of thermal efficiency during repeated frosting–defrosting cycles. Incorporating silica nanoparticles into the MPC matrix yields a compact gel architecture with a substantially reduced free-water fraction, decreasing the swelling ratio to ~ 70%. This confined network enhances stiffness and increases the energy activation for ice formation by reducing the amount of free water. Consequently, the hybrid gel effectively suppresses ice nucleation, exhibiting ice-free performance for 60 min at –10 °C and maintaining over 95% thermal efficiency over repeated cooling cycles. These findings demonstrate that the engineered inorganic–organic gel provides a thermally stable, high-performance anti-icing and anti-frosting platform, offering a promising strategy for improving thermal efficiency in practical thermal management.

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Understanding Water-Mediated Interactions at Biointerfaces: Intermediate Water (IW) Concept

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When materials encounter blood, the body's innate defense mechanisms recognize them as foreign. Water plays a crucial role in biointerfacial interactions, including protein adsorption and cell adhesion on material surfaces¹. However, water is often neglected as a medium at the interface between materials and biological systems². Here, we discuss the fundamental concepts underlying protein and cell interactions with hydrated materials, together with selected examples from our recent studies on poly(2-methoxyethyl acrylate) (PMEA) and its derivatives. We found that intermediate water (IW), which is loosely bound to a material, serves as a key indicator of the biocompatibility of soft material surfaces. Our findings highlight the critical role of IW in modulating interfacial interactions. Furthermore, the concept of IW is extended to zwitterionic polymers, poly(ethylene glycol), poly(*N*-vinyl-2-pyrrolidone), poly(methyl vinyl ether) and poly(2-oxazoline)s, including bio(macro)molecules such as proteins, polysaccharides, and nucleic acids. The degree of denaturation of adsorbed proteins was found to be influenced by the IW content. This concept of IW, which is common to hydrated bio(macro)molecules and synthetic biocompatible materials, provides a valuable framework as a key screening factor and for the rational design of biomaterials and medical devices. Ongoing research further explores molecular designs that balance multiple functional requirements, including suppression of nonspecific adsorption and fouling, alongside properties such as ionic conductivity, mechanical strength, responsiveness, biodegradability, selective cell interactions, protein stabilization, and drug delivery capability. These advances extend the applicability of the IW concept beyond biomedical systems to broader fields involving aqueous environments³.

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Specific Ion Effects in Zwitterionic Polymer Coatings: From Antipolyelectrolyte Swelling to Ice Adhesion

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Zwitterionic polymers are attractive for bioinspired surface modifications because they combine strong hydration with overall charge neutrality, central features of many low-fouling, lubricious, and ice-repellent interfaces. In sulfobetaine-containing materials, this hydration arises from closely coupled quaternary ammonium and sulfonate groups, whose internal ion pairing also makes salt responses difficult to rationalize by electrostatic screening alone. In this presentation, I will discuss sulfobetaine polymer layers using a molecularly matched platform in which the cationic group, the anionic group, and the complete zwitterionic motif are studied separately and compared within the same polymer chemistry (see Figure) [1,2].

The first part will focus on the swelling behavior of cross-linked terpolymer films with comparable charge fractions, cross-link densities, and hydrophobic comonomer content. Cationic and anionic polyelectrolyte films collapsed with increasing salt concentration, whereas the polyzwitterionic films swelled with increasing ionic strength, consistent with the antipolyelectrolyte effect.

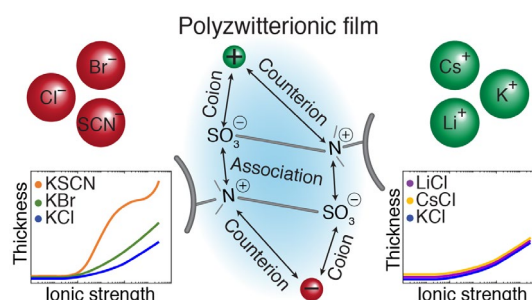


Figure adopted from ref [1]

The magnitude of the zwitterionic swelling was strongly anion-dependent, following the order $\text{SCN}^- > \text{Br}^- > \text{Cl}^-$, but was essentially insensitive to the identity of the tested cations (Li^+ , K^+ , and Cs^+). By comparing the zwitterionic system with the corresponding single-charge analogues, we assign this behavior mainly to ion-pair formation between polarizable anions and quaternary ammonium groups, whereas the sulfonate groups show much weaker ion-pairing with the tested cations.

The second part connects this ion-specific hydration picture to ice adhesion. Cationic, anionic, and zwitterionic thiol-ene curable coatings with tunable cross-link densities were used to show that temperature-dependent ice adhesion is governed by the physical state of hydration water. Step-like increases in adhesion coincided with freezing transitions estimated from ellipsometry and differential scanning calorimetry. At lower temperatures, the remaining adhesion correlated with the fraction of non-freezable water, with zwitterionic coatings displaying lower plateau adhesion than the corresponding single-charge analogues.

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Programmable Zwitterionic Biointerfaces: From Molecular Mechanisms to Biomedical Applications

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How can we engineer biointerfaces that remain fully functional in complex biological fluids while enabling nanometer-scale control of molecular recognition and cellular interactions? Biofouling remains a major barrier preventing advanced biointerfaces from operating in real biological environments. We address this challenge through zwitterionic polymer brush scaffolds that combine exceptional antifouling performance, cytocompatibility, and programmable biological functionality. Using an integrated experimental–computational approach combining SPR, mass spectrometry, and molecular dynamics simulations, we uncovered molecular mechanisms of interactions between human plasma proteins and zwitterionic or non-ionic polymer brushes. High-density lipoprotein (HDL) apolipoproteins were identified as dominant constituents of the hard corona across a broad range of brush chemistries, providing new design principles for antifouling biointerfaces.

To translate these concepts beyond model systems, we developed scalable microfluidics-based surface-initiated polymerization strategies enabling highly dense, homogeneous coatings on planar substrates, nanoparticles, and D-shaped optical fibers. More recently, we integrated DNA origami nanostructures within zwitterionic brush scaffolds, combining the bioinspired hydration of zwitterionic materials with the programmability of DNA nanotechnology. This approach enables precise nanoscale control of ligand presentation while maintaining operation in complex biological media. These architectures serve as versatile platforms for label-free bioanalytics, including optical-fiber sensors for direct biomarker detection in blood plasma and reusable sensing systems for complex environmental samples. Our results demonstrate how zwitterionic polymer brushes can evolve from passive antifouling coatings into multifunctional bioinspired scaffolds bridging molecular design, programmable nanoscale organization, and real-world biomedical applications.

**Polyzwitterionic Hydrogels as Multifunctional Biomaterials:
From Chronic Wound Management to Ocular Drug Delivery**

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Polyzwitterionic (PZI) hydrogels have emerged as a versatile class of biomaterials that synergistically combine high hydration, intrinsic antibiofouling properties, and tuneable stimuli responsiveness, making them particularly attractive for biomedical applications. Through rational design of PZI networks, these materials can be tailored for a broad range of uses - from advanced wound dressing materials for chronic wounds to stimuli-responsive drug delivery systems.

We have demonstrated that PZI-based wound dressings efficiently combine high fluid uptake with controlled removal of enzymes (protease and collagenase) from chronic wounds. Poly(sulfobetaine methacrylate) (PSB) and poly(carboxybetaine methacrylate) (PCB) hydrogels, synthesized via crosslinking with poly(ethylene glycol) dimethacrylate, exhibit reduced bacterial adhesion and strong resistance to biofouling and biofilm formation. Moreover, an interpenetrating network combining PSB and PCB demonstrated how a dual PZI architecture can be engineered to respond to biologically relevant changes in temperature, pH, and ionic strength. These smart networks display swelling behaviour governed by pH, salt concentration, and temperature variations, while also exhibiting non-cytotoxicity and antibiofouling activity - revealing their potential as adaptive interfaces in contact with complex biological fluids.

Using the approach for PZI copolymer development, we generated poly(sulfobetaine methacrylate-co-vinyl pyrrolidone) hydrogels suitable as soft contact lens drug carriers for controlled ocular delivery of timolol maleate. These materials leverage their high-water content and optical transparency ensuring also an extended drug release.

Our studies reveal that zwitterionic functionality enables multifunctionality - antifouling behavior, high hydration, diverse stimuli responsiveness, and drug delivery capability - across a broad spectrum of biomedical applications.

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Molecular Mechanisms for Zwitterionic Antifouling

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Biofouling is initiated by the adsorption of biomolecules onto material surfaces. Zwitterionic polymers have emerged as a leading class of ultralow-fouling materials for biomedical and marine applications. To uncover the molecular mechanisms underlying their antifouling properties, we combined ab initio molecular dynamics (AIMD) and atomistic molecular dynamics (MD) simulations to investigate protein-stabilizing trimethylamine N-oxide (TMAO)-derived polymers (pTMAO) and carboxybetaine-based polymers with varying charge-separation distances at both quantum and atomistic scales.

Our AIMD simulations revealed that fouling resistance is governed by the interplay of hydration characteristics, including hydrogen bonding, net charge, and dipole moment. Reducing the charge-separation distance strengthens hydrogen bonding with water through enhanced electrostatic and induction interactions, increased charge transfer, and improved hydration-layer stability, thereby inhibiting biomolecule attachment. At the same time, shorter charge separation lowers the dipole moment of near-neutral zwitterions, reducing electrostatic interactions with biofoulers. Among the materials studied, TMAO exhibited the shortest charge separation, strongest hydration, smallest net charge, and lowest dipole moment, making it an exceptionally effective nonfouling material.

Consistent with sum-frequency generation (SFG) vibrational spectroscopy measurements, atomistic MD simulations showed that pTMAO surfaces support a condensed and highly ordered hydration layer that creates a substantial free-energy barrier against protein adsorption. The addition of salt had only a minor effect, highlighting the robustness of the interfacial hydration structure. Furthermore, comparisons between sulfobetaine methacrylate (SBMA) and carboxybetaine polymers demonstrated that polymer packing structure also plays a critical role in determining antifouling performance.

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Vapor-Phase Engineering of Zwitterionic Polymer Interfaces for Biofouling Control and Electrochemical Stability

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Zwitterionic and mixed-charge polymers represent powerful interfacial design tools for controlling hydration, ion transport, and biological adhesion. However, their translation into functional coatings is often limited by challenges in processing, solubility, nanoscale compositional control, and long-term stability. In this talk, I will discuss our recent efforts to address these challenges using initiated Chemical Vapor Deposition (iCVD) as solvent-free routes to conformal zwitterionic nanofilms. First, I will present fluorine-free amphiphilic copolymers that combine hydrophobic cyclosiloxane units with pyridinium-based sulfobetaine zwitterions. These coatings replace persistent perfluoroalkyl chemistries while retaining broad-spectrum marine biofouling deterrence. The vapor-phase process yield chemically stable, elastic, and ultra-smooth films, which substantially reduce bacterial attachment and biofilm formation while enabling efficient fouling release of marine diatoms under gentle shear, demonstrating a synthetic route to PFAS-free amphiphilic antifouling coatings. Second, I will describe how noncovalent molecular interactions can be deliberately engineered during vapor-phase polymerization to tune the properties of polyampholytes. By designing Lewis acid-base comonomer pairs with different interaction strengths, we show that vapor-phase complexation can alter polymerization kinetics, sequence distribution, wetting behavior, and biofilm outcomes. Strong interactions suppress biofilm formation by common bacteria, whereas weaker interactions can enhance biofilm growth, establishing noncovalent interaction strength as a design parameter for both antifouling surfaces and engineered living materials. Finally, I will discuss gradient zwitterionic polymer nanofilms as artificial solid-electrolyte interphases for rechargeable metal batteries. Combining conformal iCVD coatings with diffusion-limited vapor derivatization creates a zwitterion-rich top layer for rapid ion transport and an inner electrochemically stable layer that blocks solvent access to the metal electrode. These nanofilms regulate interfacial transport and reaction kinetics, enabling dense metal deposition and long-term cycling of lithium-metal anodes under demanding conditions, with broader applicability to sodium and zinc electrodeposition.

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Designing fouling-resistant zwitterionic thin-film nanocomposite membranes for treatment of dye-contaminated wastewaters

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Organic dyes from textile industries are becoming the second largest pollutants in the world. Membrane technologies are promising for the treatment of dye-contaminated wastewaters. However, current polyamide thin film composite nanofiltration membranes have some major restrictions, such as membrane fouling, low flux, and inefficient separation of dyes and salts. In our works, inspired by seawater fish, we have designed a range of zwitterionic thin-film nanocomposite membranes for the treatment of dye-contaminated wastewaters. In our strategy, zwitterionic monomers have been synthesized and used to fabricate the thin-film active layer of the composite membranes through interfacial polymerization. Featured with the zwitterionic functionalities in the thin film layers, the resulting membranes show, through systematic characterizations and testings, enhanced hydrophilicity and water permeability, along with significantly reduced fouling, outstanding dye/salt separation. Our works demonstrate the potential of this strategy for producing zwitterionic membranes of improved performance for the treatment of dye-contaminated wastewaters.

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Anion- π Interlocking induced supramacromolecular zwitterionic hydrogel for bioelectronics

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Advances in implantable bioelectronics have improved the interaction between intelligent systems and biological tissues. Yet, the mechanical-immunological mismatch between rigid electrodes and soft tissues continues to limit long-term device stability. Here, we present SSPH, an immunocompatible, injectable, conductive hydrogel bridge that enables minimally invasive delivery and stable tissue integration. By forming a compliant interfacial bridge, SSPH reduces mechanical-biological mismatch and immune stress on electrodes. It is formed by the spontaneous co-assembly of PEDOT:PSS and the zwitterionic polymer poly(sulfobetaine methacrylate) (PSBMA), resulting in a 3D network stabilized by anion- π interactions, electrostatic interactions, and PEDOT-rich nanostructures. This self-healable architecture allows SSPH to maintain its intrinsic conductive pathways after deformation. Experiments confirmed that SSPH exhibits stable electrochemical properties and favorable immunocompatibility. In an acute muscle injury model, SSPH restored signal transmission across the damaged region, demonstrating its potential to serve as a bridge across disrupted tissue. Furthermore, in both electromyography recording and spinal cord stimulation models, SSPH preserved electrode performance for up to four weeks, supporting reliable bidirectional signal conduction. In addition to the hydrogel bridge connecting the electrodes and tissue, we further developed a high-resolution patterned hydrogel electrode based on this formulation for brain-machine interfacing.

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Abstracts of Posters

(in alphabetical order)

Exploring the Swelling Behaviour of Highly Crosslinked Zwitterionic Thin Films

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Zwitterionic polymers have gained significant attention for antifouling applications due to their exceptional hydration and tunability of properties.^[1] However, their inherent softness in the swollen state can lead to mechanical instability and facilitate the incorporation of particles, which can act as anchor points for fouling, thereby limiting their long-term performance. Increasing crosslinking density while preserving high hydrophilicity offers a strategy to enhance robustness without compromising antifouling efficiency.^[2]

In this study, zwitterionic [*N*-2-(methacryloyloxy)ethyl-*N,N*-dimethyl]ammonio-propane-1-sulfonate) (SPE) was copolymerized with varying amounts (1 - 50%) of a crosslinker comprising both 2-(4-benzoylphenoxy)ethyl methacrylate (BPMA) and an SPE moiety. The swelling behaviour of the resulting copolymer coatings as well as their antifouling performance was investigated in different electrolyte conditions. The swelling response was primarily governed by the antipolyelectrolyte effect, where increasing salinity modulates electrostatic interactions within the copolymer network. Even at crosslinker concentrations as high as 50%, which significantly restricted swelling, the hydrogels maintained excellent antifouling properties, effectively preventing organism adhesion. These findings highlight the potential of highly crosslinked zwitterionic hydrogels as durable, fouling-resistant materials suitable for applications in challenging environments.

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Zwitterionic copolymer nanodiscs for homogenous solubilization of liposomes– Efforts towards Membrane Protein Stabilization

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Membrane proteins represent more than 60% of therapeutic drug targets. However, their structural and functional characterization remains challenging because they rapidly lose stability once removed from their native phospholipid environment. Polymer-based nanodiscs have emerged as a promising platform for preserving membrane proteins in a near-native state while overcoming several limitations of protein-based membrane scaffold systems ⁽¹⁾. However, currently available copolymer nanodiscs still suffer from limited tolerance to pH variations, divalent ions, and structural heterogeneity. Additionally, efficient surface immobilization of nanodisc-isolated membrane proteins for biophysical analysis continues to present a major challenge in the field.

Here, we present the development of zwitterionic-containing amphiphilic copolymers designed to improve nanodisc tolerance to pH and divalent ions by virtue of better solvation and charge-density optimization. Bioinspired zwitterionic functionalities, including carboxybetaine and sulfobetaine groups, were introduced into RAFT-synthesized diisobutylene-maleic anhydride copolymers to provide controlled molecular architecture and narrow size distribution. The resulting copolymers were evaluated for the solubilization of 100 -150 nm liposomes into nanodiscs and their preliminary physicochemical characterization. This work aims to elucidate how zwitterionic modifications influence nanodisc formation, membrane compatibility, and protein stabilization. The developed platform may facilitate more robust membrane protein studies and expand applications in bioanalytics, including kinetic measurements and protein - ligand interaction analysis.

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Tuning the UCST-Type Liquid-Liquid Phase Separation Behavior of Sulfobetaine Copolymers via Charge-Asymmetry

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Polyzwitterions (PZs), a class of polyampholytes (PAs), exhibit upper critical solution temperature (UCST)-type phase behavior in aqueous solution, arising from the electrostatic interactions of cationic and anionic groups within a single repeating unit. However, electrostatic control over their phase transitions remains largely unexplored. While charge-asymmetry is well established in theoretical frameworks for charge-separated PAs, its impact on PZs has not been systematically investigated. Current strategies for tuning PZ phase transitions rely primarily on empirical adjustments of hydrophilicity, hydrophobicity and spacer groups. Here, we introduce a charge-asymmetry-driven design concept by incorporating 5–20% cationic comonomers into sulfobetaine-based copolymers via reversible addition-fragmentation chain transfer (RAFT) polymerization. The resulting copolymers exhibit both anti-polyelectrolyte and polyelectrolyte behavior, governed by charge-asymmetry and salt concentration. In water, increasing charge-asymmetry lowers the cloud-point temperature, which disappears at 20% cationic content and re-emerges in saline solution. In contrast, copolymers with 5–10% excess charge display salting-in behavior. This demonstrates that UCST modulation in PZs can be guided by theoretical concepts for PAs. Strikingly, turbidimetry revealed a pronounced hysteresis (~20 °C) in saline media, arising from the formation of large, transparent appearing coacervate droplets, which induce a previously unrecognized optical transition that is not captured by existing theory. The identified coacervate swelling mechanism introduces a new understanding of phase behavior and optical response in PZ systems. Together, these findings establish a framework for electrostatically controlling UCST-type phase separation, enabling the rational design of responsive PZ materials with tunable phase behavior and optical properties for bioinspired applications.

Zwitterionic Stimuli-Responsive Hydrogels for Soft Robotic Applications

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Stimuli-responsive hydrogels are a prominent class of soft materials with tissue-mimetic physicochemical properties. They are promising material candidates for soft robotic constructs and capsules to perform minimally invasive medical interventions, such as localized cell delivery and biopsies. To bypass the historical trade-offs between dynamic actuation and biocompatibility, this work introduces a novel bioinspired material platform utilizing zwitterionic/acrylate chemistry. This formulation imparts inherent self-healing capabilities, stimuli-responsiveness, low cytotoxicity, and highly tunable mechanical properties. In this work, we used biocompatible, high-aspect-ratio cellulose nanocrystals to impart microstructural anisotropy by applying a unidirectional shear force [1], alongside a multilayering approach at the macroscale to achieve heterogeneous physicochemical properties [2]. Through this approach, we achieved controlled differential swelling as a robust strategy to program 2D-to-3D shape changes upon exposure to environmental cues [1], [2], [3]. Additionally, by utilizing these responsive zwitterionic hydrogels, we successfully fabricated miniature, small-scale robotic constructs using digital light processing (DLP) additive manufacturing. High-resolution 3D printing enables the fabrication of custom-designed architectures optimized for low Reynolds number regimes, bypassing navigation challenges in hard-to-reach, fluidic physiological environments. As a proof-of-concept, we demonstrated soft robotic functionalities in confined, flooded media, including shape transformation, magnetic locomotion, and targeted cargo transport. This versatile zwitterionic hydrogel platform significantly expands the portfolio of functional biomaterials capable of driving next-generation miniaturized soft actuators for targeted biomedical operations.

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Interpolyelectrolyte Complexes of Chitosan and Poly(methacrylic acid): Tuning Nanostructure through Hydrophobic and Quaternary Ammonium Modifications

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Interpolyelectrolyte complexes (IPECs) formed from oppositely charged polymers represent a versatile platform for designing bioinspired soft materials with tunable physicochemical properties. In this work, we investigate complexes based on hydrophobically modified poly(methacrylic acid) (PMAA) and quaternized chitosan as a model system to understand how hydrophobic interactions and quaternary ammonium functionalization govern complex formation and nanostructure. The combination of pH-dependent carboxylate groups on PMAA, pH-responsive amine groups on chitosan, and permanent quaternary ammonium charges creates a multicomponent, ampholytic system, positioning these IPECs as a bioinspired bridge between classical polyelectrolyte complexation and zwitterionic materials design, with implications for biocompatible coatings, nanocarriers, and responsive soft materials.

Quaternized chitosan derivatives with quaternization degrees up to 68%, confirmed by NMR and potentiometric titration, are combined with hydrophobically modified PMAA to introduce associative driving forces beyond pure electrostatics. Complex formation is studied as a function of charge stoichiometry, pH, and ionic strength. Static and dynamic light scattering (SLS/DLS) reveal the formation of IPECs with radii of 40–100 nm depending on charge ratio, while small-angle neutron scattering (SANS) probes the internal structure across multiple length scales. Stopped-flow kinetics coupled with time-resolved transmittance measurements further resolves the rapid association dynamics as a function of molecular design and solution conditions. Together, these complementary techniques allow us to disentangle the electrostatic and hydrophobic contributions driving IPEC assembly, providing insight into the underlying molecular mechanisms and offering a framework that may be extended to more complex polyelectrolyte systems such as nucleic acids and proteins.

Hydrophilic Zwitterionic Polymers with Programmable Degradation and Thermoresponsiveness

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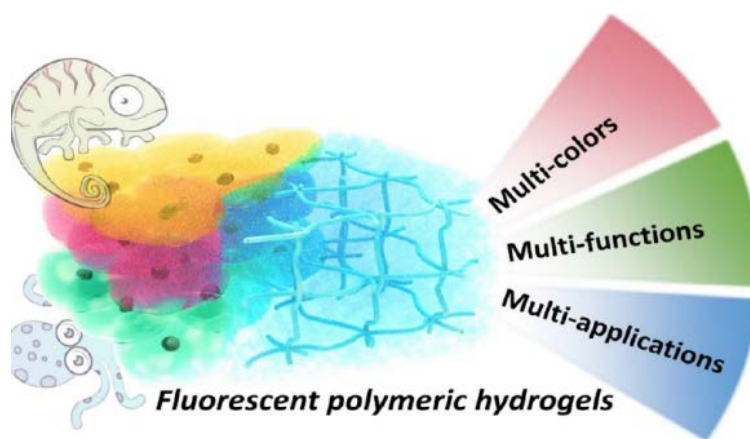
Development of functional hydrophilic polymers with tailorable properties is essential for biomedical research. α -Lipoic acid (LA) has emerged as a versatile and cost-effective monomer for constructing degradable polymers because of cleavable disulfide bonds are present along the polymer backbone. LA and its derivatives undergo radical ring-opening polymerization via both conventional and controlled radical processes. The disulfide bonds can readily undergo bond scission under mild reductive conditions. While reversible addition–fragmentation chain transfer (RAFT) copolymerization of LA with vinyl monomers provides degradable polymers with controlled molecular weights, conventional thermally initiated RAFT systems often suffer from oxygen sensitivity and limited LA incorporation. Herein, we report the synthesis of degradable vinyl copolymers incorporating α -lipoic acid derivatives via photoiniferter-RAFT polymerization. This light-mediated approach eliminates the need for external initiators and exhibits enhanced oxygen tolerance, enabling a simplified and spatiotemporally controlled polymerization process under ambient conditions. The method demonstrates broad comonomer compatibility and allows efficient block copolymer synthesis with predictable molecular weight. Introduction of Isopropyl group to the terminal of LA impart thermoresponsiveness to hydrophilic poly(*N,N'*-dimethylacrylamide) and zwitterionic polymers, such that lower critical solution temperature can be adjusted by the polymer composition and side chains.

Mimicking Color-Changing Organisms to Enable the Multicolors and Multifunctions of Smart Fluorescent Polymeric Hydrogels

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Fluorescent polymer hydrogels (FPHs) combine the advantages of fluorescent polymers and hydrogels, making them attractive for displays, camouflage, and soft robotics. The key challenge is no longer synthesis but achieving robust multicolor tunability and multifunctional synergy in a single system. Nature offers a powerful blueprint: chameleons and cephalopods exhibit dynamic color change via hierarchical chromatophore layers and integrate diverse soft tissues for complex behaviors. Mimicking these strategies can greatly enhance FPH performance. In this talk, we first replicate reptile skin multilayer structures to expand FPH fluorescence across the visible spectrum. We then introduce supramolecular interactions or integrate FPHs with other soft materials to create multifunctional systems. These enable bioinspired actuators with synergistic color/shape switching, on-demand fluorescent patterning for displays and encryption. Our work aims to deepen understanding of natural color-change systems and accelerate the development of bioinspired smart materials.



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Surficial Bonded Zwitterionic Modification for Anti-fouling and Biosensor Applications

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We report the development of functionalized zwitterionic copolymers featuring epoxy reactive groups for robust and stable covalent immobilization on diverse material surfaces. Poly(GMA-co-SBMA) (PGSM) and poly(GMA-co-SBAA) (PGSA) were synthesized via free radical polymerization, enabling durable modification of chemically inert substrates including nitrile butadiene rubber, polyamide (PA), polyurethane (PU), and various biosensing platforms. To further enhance process efficiency and scalability, a one-step acid-catalyzed surface activation strategy was introduced, allowing direct covalent bonding onto PA and PU substrates without conventional multistep pretreatment. This streamlined approach preserves strong interfacial adhesion while significantly reducing processing complexity. The resulting zwitterionic coatings exhibit outstanding antifouling performance driven by the formation of a dense hydration layer, achieving up to 95% reduction in nonspecific protein adsorption, substantial improvement in hemocompatibility, and over 98% suppression of *E. coli* adhesion. Beyond passive antifouling, this work highlights the critical role of zwitterionic interfaces in enhancing biosensor performance under complex biological environments. Specifically, surface modification of cellulose-based substrates with PGSA significantly improves capillary-driven transport and minimizes nonspecific adsorption in lateral flow immunoassay (LFIA) systems, resulting in enhanced signal clarity, improved detection sensitivity, and reduced background interference. Furthermore, integration of zwitterionic coatings onto surface-enhanced Raman scattering (SERS) chips effectively suppresses fouling from complex media such as serum or whole blood, thereby prolonging sensing stability and enabling more reliable detection of target analytes in real-world conditions. The covalent “graft-to” strategy ensures long-term stability of the antifouling layer, overcoming limitations associated with physically adsorbed coatings in dynamic sensing environments. By bridging antifouling surface engineering with advanced sensing platforms, this study demonstrates a versatile and industrially viable approach for developing next-generation biosensors with improved robustness, sensitivity, and operational reliability. The proposed methodology holds significant potential for applications in medical diagnostics, wearable sensing devices, and point-of-care testing systems.

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Conformation-Dependent Cytosolic Translocation of Oligonucleotides Mediated by a Phospholipid-Mimicking Zwitterionic Copolymer

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Bioinspired zwitterionic polymers that emulate the amphiphilic architecture of phospholipids offer unique opportunities to engineer non-disruptive biointerfaces [1,2]. Here, we investigate a phospholipid-mimicking copolymer, poly(2-methacryloyloxyethyl phosphorylcholine-co-*n*-butyl methacrylate) [poly(MPC_{30-r}-BMA₇₀), PMB30W], composed of the zwitterionic phosphocholine monomer 2-methacryloyloxyethyl phosphorylcholine (MPC) and the hydrophobic monomer *n*-butyl methacrylate (BMA), that enables direct cytosolic translocation of oligonucleotides without reliance on endocytosis. Although passive membrane-crossing phenomena have been reported for certain amphiphilic polymers, the influence of cargo molecular structure, particularly conformational rigidity and flexibility, on cytoplasmic accessibility remains poorly understood. To elucidate this structure–function relationship, we synthesized well-defined PMB30W via RAFT polymerization, enabling precise control of molecular weight and end-group functionality. Single-stranded DNA (ssDNA) and double-stranded DNA (dsDNA) were covalently conjugated using copper-free click chemistry to construct structurally defined carrier–cargo systems. Systematic cellular studies revealed that DNA–PMB30W conjugates achieved cytoplasmic delivery under conditions that exclude classical endocytotic pathways. Notably, cargo conformation played a decisive role: rigid dsDNA maintained high cytosolic accessibility even at extended chain lengths, whereas flexible ssDNA exhibited pronounced length-dependent attenuation of delivery efficiency. Importantly, membrane integrity and cell viability assays confirmed that this translocation occurred without detectable membrane disruption or cytotoxicity. These findings demonstrate that the interplay between bioinspired amphiphilic polymer architecture and nucleic acid conformational mechanics critically governs cytoplasmic accessibility. This work provides mechanistic insight into energy-independent membrane translocation and establishes molecular design principles for zwitterionic polymer systems aimed at controlled intracellular delivery.

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Thermo-responsive interpolyelectrolyte complexes (IPECs): molecular design, mesoscopic structure, drug solubilisation

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When two oppositely charged polyelectrolytes are mixed in an aqueous medium, cooperative electrostatic interactions lead to spontaneous formation of interpolyelectrolyte complexes (IPECs). IPECs can adopt various colloidal structures, which will depend on how the polymers interact with each other and the solvent (water). This means by adjusting the copolymer's Mw and segment ratio, the properties of the IPECs can be effectively tuned, thereby establishing a connection between polymer design and IPECs properties. [1-3] To develop IPECs for “targeted drug delivery” that have tunable solubilization properties and temperature-responsiveness, diblock copolymers (poly(2-acrylamido-2-methyl-1-propanesulfonic acid)-b-poly(N-isopropylacrylamide) (PAMPS-PNIPAM) and poly(2-methacryloyloxyethyltrimethylammonium chloride)-b-poly(ethylene oxide) (PMTAC-PEO)), with varying molecular weights and length ratios have been synthesized by us to form thermo-responsive IPECs with diverse morphologies. Light scattering and small angle neutron scattering experiments show that the co-assembled IPECs behavior varies not only with the mixing ratio, but also with molecular design and, most importantly, temperature. UV-Vis experiments show that the solubilization property of IPECs can be controlled by temperature. These experiments enable a deeper understanding of how the IPECs structure can be precisely controlled, paving the way for their further development in targeted drug delivery.

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PMA-DDA-DMAPA-Coated Gold Nanoparticles with Controlled PEG Valency: Functionalization and Electrophoretic Isolation

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¹

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Nanoparticle labels with a discrete and controlled number of functional groups are highly desirable for applications in nanomedicine, biosensing, and the assembly of complex nanostructures. However, the direct synthesis of such well-defined conjugates remains challenging because of the statistical nature of ligand attachment. This contribution presents a robust strategy to overcome this “statistical barrier” by separating the functionalization and purification processes through gel electrophoresis.

In our study, 4 nm gold nanoparticles (AuNPs) were encapsulated in a versatile amphiphilic polymer shell based on poly(isobutylene-alt-maleic anhydride) grafted with dodecylamine (PMA-DDA) and further modified with 3-(dimethylamino)-1-propylamine (DMAPA). The incorporation of DMAPA introduces pH-responsive amine groups and gives the polymer shell ampholyte-like surface properties. The remaining carboxyl groups on the polymer shell provide reactive sites for subsequent functionalization under mild conditions.

By optimizing the concentration of 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide (EDC), amino-modified poly(ethylene glycol) (PEG) chains of sufficient length (10000 Da) were covalently attached to the polymer surface. Using 2% agarose gel electrophoresis, we successfully resolved discrete bands corresponding to AuNPs bearing exactly zero, one, or two PEG molecules per particle. These conjugates were extracted and purified from the gel. Their discrete valency and stability were subsequently verified by re-electrophoresis, during which the isolated conjugates migrated to positions matching the corresponding bands in the original samples.

This approach demonstrates the potential of DMAPA-modified PMA-DDA polymers as building blocks for tailoring the surface charge and functionality of bioinspired nanomaterials. By providing a route to isolate nanoparticles with a well-defined number of ligands, this work may facilitate the development of diagnostic probes and targeted drug-delivery systems with reduced nonspecific interactions and controlled binding behavior.

Functional Silatranes as a New Type of Building Blocks for Zwitterionization and Polymer Synthesis

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The precise architecture of functional interfaces is a fundamental challenge in surface chemistry, particularly when seeking to balance robust stability with high-performance functionality. While traditional trialkoxysilanes have been widely utilized for modifying oxides and creating superhydrophilic surfaces, they are often hindered by rapid, uncontrolled hydrolysis and self-condensation. These factors lead to thick, non-uniform layers that compromise the molecular regularity of functional groups. This presentation showcases a decade of progress in replacing traditional precursors with functional silatranes—a "caged" class of silane molecules that redefine the standards of controlled silanization. The unique pentacoordinate structure of silatranes, characterized by a transannular N→Si coordinate bond, provides inherent hydrolytic stability. This allows for the formation of thin, homogeneous nanolayers with exceptional molecular regularity.¹ These ultra-smooth, antifouling coatings have enabled critical breakthroughs in medical devices, such as anti-clogging hemofiltration membranes for the mass collection of circulating tumor cells.² Our recent work has expanded the silatrane library to include methacrylate and amino-functionalized variants, enabling the synthesis of dense polymer brushes via click chemistry and radical polymerization.³ The latest frontier in this research involves sophisticated bioinspired designs. We introduce mixed zwitterionic silatranes as a superior platform for creating biointerfaces that resist complex biological media.⁴ Collectively, these advancements establish functional silatranes as a highly predictable and versatile toolkit for modern surface engineering and polymer science.

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Advancing quantum optics with LC

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Promises and prospects of optical information technologies have been getting an essential boost due to recent developments in quantum photonics, in which non-classical light sources are used for the implementation of optical quantum protocols and algorithms. Large-scale parallel information processing suggests implementation of compact, multifunctional and, of course, tuneable elements that provide wide-broadband and extremely fast operation – constituents of the ‘flat optics’ paradigm.

Liquid crystals, with their ability to self-assemble, strong response to external fields and integrability into complex systems, were for long considered as key materials in light-beam manipulation. The recently discovered ferroelectric nematic liquid crystals (FNLC) have extremely large spontaneous polarization and considerable second-order optical nonlinearity that make these a great candidate for nonlinear optics and photonics. The use of these as sources of quantum light could considerably extend the boundaries of photonic quantum technologies [1].

Strong intrinsic polarization of FNLC introduces additional constraints and peculiar requirements on boundary conditions. On the other hand, engineering special surface patterns of electrostatic coupling should allow for creation of comprehensive LC structures with specific nonlinear optical properties that will pave the way for greater efficiency and new phenomena impossible with nonpolar LC materials. Zwitterionic materials due to a very high content of ionic groups should play a crucial role in control of self-organization processes of novel ferroelectric nematic liquid crystals and alignment of these in peculiar specific structural geometries of interest. There are high hopes, at least.

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Photoinduced Immobilization of Zwitterionic Polymer Coatings for Enhanced Antifouling Performance of Composite Resins

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Biofilm accumulation on composite resin restorations is a major challenge because it contributes to secondary caries and restorations. Although 2-methacryloyloxyethyl phosphorylcholine (MPC) has shown excellent antifouling properties, previous approaches have mainly relied on bulk blending, which may impair the mechanical properties of composite resins. Here, we developed a photo-crosslinkable zwitterionic polymer coating that enables stable surface immobilization on composite resins while enhancing biofilm resistance. A photoreactive MPC-based copolymer (PMBAz), consisting of MPC, n-butyl methacrylate (BMA), and azide-functionalized methacrylate (MPAz), was synthesized and introduced onto composite resin surfaces through UV-induced photografting. Copolymers with different MPC contents (30%, 60%, and 90%) were compared based on surface hydrophilicity, protein adsorption, and bacterial adhesion. The 60% MPC composition demonstrated the optimal balance between hydration-layer formation and coating stability, exhibiting significantly reduced protein adsorption and bacterial adhesion compared with other formulations. The optimized PMBAz coating maintained cytocompatibility and showed strong antifouling performance in both single-species and microcosm biofilm models, reducing biofilm biomass and thickness by approximately 75% and 71%, respectively. In addition, an in vivo rat intraoral mucosal attachment model confirmed reduced biofilm accumulation on PMBAz-coated composite resin surfaces after one week of oral exposure compared with uncoated controls. These results demonstrate that zwitterionic polymer surface photografting is an effective strategy for improving the biofilm resistance of composite resins while preserving their intrinsic material properties, suggesting its potential application for long-term dental restorative materials.

Twitter ionic polymers as surface active molecules on inorganic particle

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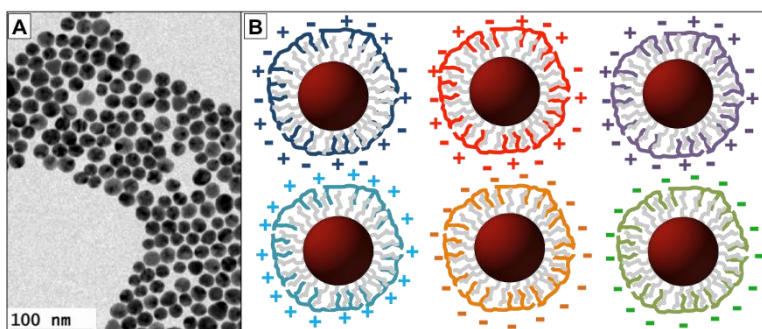
Various zwitterionic polymers are discussed for two different applications.

In the first example, zwitterionic polymers are used to make inorganic particles hydrophobic for use in composites. The polymers are employed both as random copolymers and as block copolymers to form micelles in hydrophobic media. Within the micelles, various inorganic particles such as sulfides, sulfates, or silicates are mineralized.

The results obtained are compared with experiments in which amphiphilic positively or negatively charged random or block copolymers are used in the mineralization process. The aggregation behavior and micelle formation of these different polymers are measured using DLS. In addition, the particles are characterized using TEM.

In the second example, particles surrounded by a hydrophobic shell — mostly gold — are hydrophilized using zwitterionic copolymers, and their stability in biological media (serum) is studied. Effects on protein adsorption are investigated. Here, too, the results are compared with those of other ionic and nonionic surfactants.

In both cases, zwitterionic polymers demonstrate significant advantages in the functionalization and stabilization of particles, both in nonpolar media and in water. The reasons for this include altered dynamics in solution as well as the internal saturation of positive and negative charges.



AU Nanoparticles surrounded by twitterionic polymers (TEM) + Schematic illustration of charged particles

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Plasma/Click-Functionalization of Plastics: Systematic Evaluation of Structure-Performance Relationships in Low-Fouling Zwitterionic Polymer Brushes

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A central objective in zwitterionic materials research is to establish molecular design principles that link the architecture of zwitterionic brush layers to antifouling performance.^[1] The latter depends on many factors such as the zwitterionic functionality involving the chemical functional groups and their charge spacing, the polymer backbone, brush layer thickness and homogeneity.^[2-5] Current understanding of the antifouling behavior of different zwitterions is primarily derived from molecular dynamics simulation and comparative protein adsorption assays on idealized substrates such as silicon wafers, gold, and glass. While these approaches have provided valuable insight into hydration-layer formation and resistance to nonspecific protein adsorption, they do not capture the complexity of biological fouling involving living microorganisms. Consequently, structure-performance relationships on clinically relevant polymer surfaces remain insufficiently explored, particularly with respect to microbial adhesion and biofilm development.

We present a versatile three-step surface modification strategy that enables the formation of dense polymer brush layers with various zwitterionic functionalities. The method involves a plasma-mediated functionalization of base materials such as polyethylene and polyamide-6 with “clickable” polymer brushes and a highly efficient late-stage zwitterionization. The resulting set of materials allows a detailed comparison of different zwitterionic groups and their charge spacing with respect to the adhesion of microorganisms to zwitterionic brush layers. A thorough microbiological analysis with different bacterial strains revealed clear structure-dependent differences in biofilm retention, demonstrating how subtle variations in zwitterionic architecture determine antifouling performance on polymeric substrates.

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Synthesis of Zwitterionic Surfactants for Nanoparticle Extraction and Structural Analysis of Self-Assembled Structures by Small-Angle Neutron Scattering

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3-Alkyl dimethyl ammonio propyl sulfates (C_n -APSO₄) are zwitterionic surfactants that exhibit a characteristic phase separation behavior. Above the cloud point (CP), C_n -APSO₄ forms a homogeneous aqueous solution, whereas below the cloud point it separates into an aqueous phase and a surfactant-rich phase containing highly concentrated C_n -APSO₄. This temperature-responsive phase separation can be utilized in extractions, termed cloud point extractions (CPE). CPE using C_n -APSO₄ is particularly attractive for nanoparticle concentration because nanoparticles can be extracted while still maintaining their dispersibility. This is an important advantage, as nanoparticles often tend to aggregate during conventional concentration procedures. Therefore, C_n -APSO₄ based extraction has significant potential as a mild, green, and effective method for handling dispersed nanomaterials. However, the relationship between the molecular structure of C_n -APSO₄, the micellar structures formed before phase separation, and the resulting cloud point behavior has not yet been sufficiently investigated. In this study, C_n -APSO₄ molecules with different alkyl chain lengths and related molecular structures were synthesized to determine how molecular design influences micelle formation and phase separation. The self-assembled structures of the synthesized surfactants were determined by small-angle neutron scattering (SANS), which provides specific structural information on the micelles present in solution. The results provide new insights into how the molecular structure of C_n -APSO₄ affects micellar structure and CP behavior. These findings should allow for the rationale design of zwitterionic surfactants more suitable for nanoparticle extraction and contribute to a deeper understanding of the relationship between molecular structure, self-assembly, and functional phase separation.

Comparative Mechanistic Analysis of Zwitterionic and Pseudo-Zwitterionic Hydrogel Coatings for Anti-Icing Applications

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Thermal management, including heat transport and heat exchange, is essential for maintaining performance and improving energy efficiency in systems ranging from household appliances, such as refrigerators and air conditioners, to industrial applications, including machinery and data centers. Unwanted ice and frost accumulation on surfaces significantly increases thermal resistance, leading to a drastic reduction in heat-transfer efficiency and, consequently, higher energy consumption. To date, zwitterionic hydrogels have attracted considerable attention as promising anti-icing coatings because their strongly hydrated ionic groups can form a stable water-rich interfacial layer that suppresses ice nucleation and reduces ice adhesion. However, the synthesis of conventional zwitterionic materials can be limited by monomer availability, structural tunability, and processing complexity.

In this study, we comparatively investigate zwitterionic and pseudo-zwitterionic hydrogel coatings to evaluate whether a pseudo-zwitterionic structure, designed to mimic the charge-balanced nature of zwitterions, can provide comparable enhanced anti-icing performance. The pseudo-zwitterionic hydrogels were constructed by incorporating spatially associated cationic and anionic groups within the hydrogel.

Through systematic comparison of hydration states, freezing delay, frost formation, and heat-transfer efficiency, the pseudo-zwitterionic hydrogel coating exhibited anti-icing properties comparable to those of the zwitterionic hydrogel. To elucidate the origin of the behavior, the hydration states of water within the hydrogel networks were analyzed using DSC and FTIR. The amount of free water increased in the order of pseudo-CBMA < CBMA < pseudo-SBMA < SBMA, demonstrating that subtle differences in ionic architecture significantly influence water organization within the hydrogel network. Interestingly, the anti-icing performance exhibited the reverse trend, suggesting that minimizing free water is more critical for suppressing ice nucleation and propagation. These findings highlight the important role of hydration-state engineering in the design of advanced anti-icing coatings.

These results demonstrate that anti-icing performance is governed not only by zwitterionic functionality but also by the hydration state arising from the underlying ionic architecture, highlighting pseudo-zwitterionic hydrogels as a versatile alternative platform for anti-icing surface design. This work provides mechanistic insight into the relationship between ionic hydration structure and ice formation behavior and establishes hydration-state engineering as a key strategy for the development of advanced anti-icing coatings.

Ionomer–Polyelectrolyte Complexes as Tunable Nanofiltration Membrane Materials

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Polyelectrolyte complexes (PECs) have recently emerged as promising materials for nanofiltration membranes because their ionic structure, hydration, and net charge can be adjusted to control molecular transport. Most PEC membranes are currently prepared by layer-by-layer (LbL) assembly, in which oppositely charged polyelectrolytes are deposited sequentially. Each adsorption step must be followed by a rinsing step to remove weakly bound polymer and prevent uncontrolled complex formation in the bulk solution.

Here, we present an alternative approach for preparing PEC nanofiltration membranes in only two main steps. First, a continuous layer of a water-insoluble cationic ionomer is formed on a porous support. The ionomer layer is subsequently converted into a polyelectrolyte complex through contact with polyelectrolyte solution. Because the preformed ionomer is already insoluble, the selective-layer thickness can be established before complexation, avoiding the repeated deposition cycles required in conventional LbL assembly.

The resulting ionomer–polyelectrolyte complex acts as a dense but hydrated nanofiltration selective layer. Its composition, charge, swelling, permeability, and separation properties can be tuned through the complexation conditions and the properties of the polyelectrolyte. This approach therefore provides a simplified platform for investigating and designing charged polymer complexes as selective membrane materials.

Multi-component crystal forms of nutraceuticals for bioavailability improvement

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Nutraceuticals, also referred to as functional foods, are widely recognised for their roles in the prevention and treatment of various diseases. A broad range of food-related products, including vitamins, soy-derived compounds, glucosamine, chondroitin, polyphenols, and flavonoids, fall within this category. Despite their growing popularity and therapeutic potential, many nutraceuticals suffer from inherent physicochemical limitations, such as poor solubility and low dissolution rates, which result in reduced oral bioavailability and consequently limit their clinical applications. One promising strategy to address these challenges is the development of cocrystal formulations. A cocrystal is a crystalline material composed of two or more components, typically an active pharmaceutical ingredient (API) or nutraceutical and a coformer, present in a definite stoichiometric ratio. By carefully selecting appropriate coformers, cocrystals can be rationally designed to enhance key properties of the parent compound, including solubility, dissolution rate, physicochemical stability, and mechanical behaviour. As a result, cocrystallisation offers a viable pathway for improving the performance and applicability of nutraceutical products.

Two case studies employing cocrystallisation approaches to generate multicomponent crystal forms of nutraceuticals, specifically daidzein (DAI) [1] and apigenin (API) [2], are presented. For daidzein, a cocrystal screening with pyridine-derived molecules, i.e., nicotinamide, isonicotinamide, caffeine, d-proline, l-proline and 4,4'-bipyridine was conducted. A new cocrystal of daidzein and 4,4'-bipyridine (DAI-BIP) was successfully generated via grinding and solvent methods. For apigenin, a large scale cocrystal screening with 80 coformers was conducted. In order to reduce the number of the experimental screening tests, three computational prescreening tools, i.e., molecular complementarity (MC), hydrogen bond propensity (HBP), and hydrogen bond energy (HBE), were used to provide an initial selection of 47 coformer candidates, leading to the discovery of seven APG cocrystals. Among them, six APG cocrystal structures have been determined by successful growth of single crystals, i.e., apigenin-carbamazepine hydrate 1:1:1 cocrystal, apigenin-1,2-di(pyridin-4-yl)ethane hydrate 1:1:1 cocrystal, apigenin-valerolactam 1:2 cocrystal, apigenin-(DL) proline 1:2 cocrystal, apigenin-(D) proline/(L) proline 1:1 cocrystal. All of the DAI and APG cocrystals showed improved dissolution performances with the potential to be formulated into drug products.

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Structure-property relationships in ethoxylated quaternary cationic and tertiary amine surfactants: impacts of molecular architecture and pH on interfacial behavior and self-assembly

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Cationic surfactants are essential in industrial and biomedical sectors due to their surface affinity and antimicrobial properties. While performance is typically tuned via alkyl chain modification or mixing, headgroup ethoxylation—common in anionic systems—offers a sophisticated, underutilized route for controlling quaternary ammonium salts and their tertiary amine precursors. This study integrates results from two series of surfactants: ethoxylated quaternary ammonium salts and pH-responsive ethoxylated tertiary amines. Using Small-Angle Neutron Scattering (SANS) and Neutron Reflectivity (NR), we characterized the evolution of micellar microstructures and adsorption layers. The influence of alkyl chain length (C12, C16) and the precise degree of ethoxylation (EO_n) was systematically evaluated alongside pH variation. Our findings demonstrate that ethoxylation enables the fine-tuning of surfactant packing. In quaternary systems, increasing the EO sequence provides a predictable shift in the hydrophilic-hydrophobic balance, affecting micellar shape and surface density. In tertiary amine systems, a dramatic pH-dependent transition occurs: surfactants are nonionic at high pH but become cationic upon protonation at low pH. This triggers a structural shift from large, high-aggregation number aggregates to smaller, globular charged micelles. Notably, this evolution correlates with biological performance; antimicrobial activity is "switched on" at low pH, while remaining inactive at neutral pH. The integration of precise synthesis and neutron scattering demonstrates how ethoxylation, chain length, and charge density can be tuned to design 'smart' surfactants for diverse biological applications.

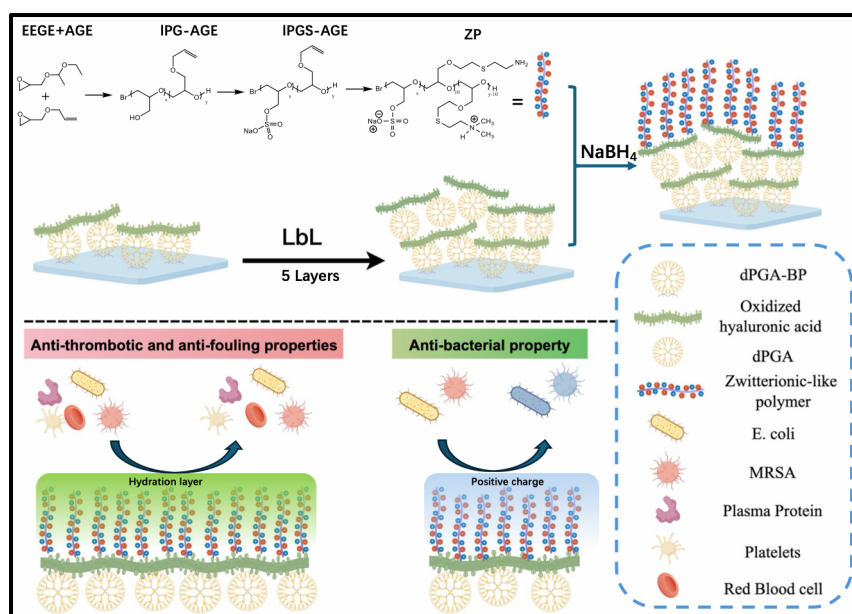
Bioinspired Zwitterionic-like Polyglycerol Coatings for Antifouling and Bactericidal Biomedical Surfaces

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Bioinspired zwitterionic materials create hydrated, biocompatible interfaces that resist nonspecific adhesion. Here, we report a zwitterionic-like polyglycerol/hyaluronic acid multilayer coating (shown in Figure) for PVC, a blood-contacting material prone to protein adsorption, platelet activation and bacterial colonization. Mixed-charge polyglycerol copolymers bearing sulfate and amine groups were synthesized by anionic ring-opening copolymerization, sulfation and thiol-ene functionalization, then covalently grafted onto dendritic polyglycerol/oxidized hyaluronic acid (OHA) multilayers via Schiff-base chemistry and reductive stabilization. The resulting uniform, hydrophilic coatings showed excellent cytocompatibility, low hemolysis and improved hemocompatibility. Charge-balanced ZP2 gave the strongest antifouling performance, reducing platelet coverage from $96.26 \pm 0.80\%$ to $0.38 \pm 0.34\%$ and suppressing MRSA and *E. coli* adhesion to $0.04 \pm 0.01\%$ and $0.07 \pm 0.01\%$. Positively biased ZP3 showed contact-killing activity, reducing viable MRSA and *E. coli* to 23 ± 12 and 36.67 ± 35.11 CFU. This platform enables tunable antifouling and antibacterial functions for polymer-based biomedical devices and blood-contacting applications broadly.



Schematic illustrations of polymer synthesis and coating. Dendritic polyglycerol amine with benzophenone (dPGA-BP) was immobilized onto a polymer substrate followed by layer-by-layer assembly of dPG and OHA via Schiff base bonds. The respective ZP was grafted on the top subsequently. Then the imine bonds were reduced to stable amines.

Phosgene-Free Route to High-Purity Sarcosine N-Carboxyanhydride for the Controlled Synthesis of High-Molecular-Weight Polysarcosine

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Poly(ethylene glycol) (PEG) has long been considered the standard hydrophilic polymer for biomedical applications. However, the growing prevalence of anti-PEG antibodies and the resulting immunogenic responses have prompted the search for alternative stealth polymers that offer enhanced biological performance [1]. Polysarcosine (pSar), a biocompatible and biodegradable polypeptide, has emerged as a promising candidate thanks to its high-water solubility, low immunogenicity, resistance to non-specific protein adsorption and capacity to extend circulation times. However, the synthesis of high-molecular-weight pSar with a narrow molecular weight distribution remains challenging and is highly dependent on the purity of the sarcosine N-carboxyanhydride (Sar-NCA) monomer used in ring-opening polymerization (ROP) [2, 3].

In this study, we present a scalable phosgene free synthesis of highly pure Sar-NCA using an oxalyl chloride/silyl-mediated route and compare it with a conventional PCl_3 -based method. Although both the comparable outcomes of the two methods, the oxalyl chloride approach achieve Sar-NCA with a sarcosine residue below the detection threshold of ^1H NMR following double sublimation. Contrary, the PCl_3 derived monomer exhibited traceable impurities indicative of decomposition and hydrolysis. These differences had a pronounced impact on polymerization behavior. Utilizing benzylamine initiation and benzoic acid catalysis, oxalyl chloride-derived Sar-NCA facilitated the controlled ring-opening polymerization (ROP) over a wide range of targeted degrees of polymerization ($\text{DP} = 50\text{--}2250$), yielding polymers with monomodal molecular-weight distributions and low dispersity. However, PCl_3 -derived monomers yielded broadened distributions, reduced molecular weights, and chromatographic shoulder formation at elevated DPs. A large-scale synthesis of high-purity Sar-NCA (12.5 g, 73% yield) was conducted, enabling the preparation of pSar with different average molecular weights up to 186 kDa and low dispersity ($\text{Đ} = 1.08$). The SEC-MALS analysis confirmed the presence of defined polymer structures across a wide molecular weight range. The findings of this study demonstrate that monomer purity is imperative for achieving controlled ROP and accessing hitherto unattainable high molecular weight pSar. This provides a practical platform for the large-scale production of polysarcosine for drug delivery, nanomedicine, and bioconjugation applications.

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Toward Reusable Particle-Based Materials: Selective Recovery of Distinct Nanoparticulate Species from Model and Device Surfaces using Poly(phosphorylcholines) coatings

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Functional polymer films are a class of smart surface coatings used to design smart interfaces. Due to their specific physicochemical properties this form of surface modification is promising for various applications. In this context, zwitterionic polyphosphorylcholines (PMPCs) and their use as multifunctional coatings have received increasing attention in recent years. Discussed applications include anti-fog, anti-fouling, easy-to-clean, and anti-freeze coatings are based on the interaction between PMPC and water.

While PMPCs exhibit high solubility and undergo swelling in pure water and ethanol, the chains collapse at an ethanol content of 80% water which is known as co-nonsolvency effect. This work will demonstrate a novel application that uses this effect to selectively detach functionalized gold nanoparticles (AuNPs) on demand from model surfaces and real electronic devices, such as Si-FETs. Due to their unique optical properties, which manifest in a pronounced localized surface plasmon effect, AuNPs are being studied and used in various applications. However, the preparation of such functionalized AuNPs is time-consuming, synthetically complex, and highly resource-intensive. Therefore, new recovery strategies are considered. We demonstrate, through a series of desorption experiments, that by meticulously controlling the solvent composition, AuNPs can be regained from the surface functionalized by adaptive PMPC coatings selectively and spatially resolved.

Obstacles to realize these applications include transferring PMPC coatings from small model surfaces to technically relevant substrates, ensuring scalability, and address stability concerns. The lecture will also address these issues and present application-oriented coating processes for films for well-known as well as new applications. Additionally, strategies for enhancing stability against mechanical stress and degrafting by water will be discussed.

Acknowledgments: The authors acknowledge the funding from the Federal Ministry for Economic Affairs and Energy (BMWE) of Germany (22614 BG) and by the German Research Foundation (Deutsche Forschungsgemeinschaft, DFG) via the "Responsible Electronics in the Climate Change Era – REC²" Cluster of Excellence (EXC 3035, Project-ID 533607596).

Injectable dynamically crosslinked zwitterionic hydrogels as a viscosupplement for osteoarthritis treatment

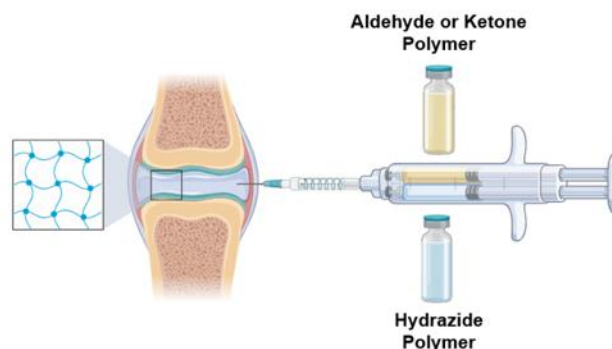
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While hyaluronic acid-based viscosupplements have been widely used clinically for osteoarthritis treatment, their rapid *in vivo* degradation and variable biological interactions have led to inconsistent patient outcomes. Herein, we report the use of a dynamically-crosslinked hydrazone-based zwitterionic hydrogel based on mixing of hydrazide (ZH) and aldehyde (ZA) or ketone (ZK)-functionalized precursor polymers as an alternative to hyaluronic acid-based viscosupplements. Upon co-injection via a double barrel syringe, the water-like precursor polymer solutions gel within ~1-3 min. Relative to a leading hyaluronic acid-based product (Synvisc®), the dynamic zwitterionic hydrogels showed 10-50× higher compressive moduli beneficial to mechanically support the joint while at the same time exhibiting a 5-9× higher reduction in the coefficient of friction at walking frequencies against an alumina surface and a >25% lower coefficient of friction against *ex vivo* cartilage samples, all while also being easier to inject given the low initial viscosity of the precursor polymers. The hydrogel and its degradation products were non-cytotoxic and showed minimal inflammatory responses when injected *in vivo*, with the ZA formulations persisting for >90 days post-injection compared to days for conventional HA-based viscosupplements. Overall, zwitterionic dynamic hydrogels provide a combination of mechanical support, lubricity, and *in vivo* inertness/stability that suggest promise for clinical translation for osteoarthritis treatment.



Hydrophilic Poly(Iminopyridinium Ylides):

Defining An Unexplored Chemical Space

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With the increase in hypersensitive reactions to PEG as well as the occurrence of the accelerated blood clearance phenomenon, the search for alternative, overall charge-neutral and hydrophilic polymers becomes increasingly crucial.^{1,2} Poly(ylides), a class of hydrophilic polymers with strong zwitterionic character, show potential as bio- and nanomedical building blocks because of their high water-solubility and antifouling properties, among others.^{3,4} In this context, we here define so far unexplored chemical space for poly(ylides) by establishing poly(iminopyridinium ylide) as a versatile tool for bio- and nanomedical applications. Next to showing excellent solubility in both water and saline as revealed by measurements of the critical aggregation concentration, poly(iminopyridinium ylide) shows full stability across a wide pH-range. DLS, DOSY and zeta-potential analysis reveal that its surface charge and aggregation patters resemble those of established zwitterionic polymers, underlining its potential applicability as a PEG replacement. Furthermore, poly(iminopyridinium ylide) shows only minimal to no binding to biomolecules, and also proves capable to protect insulin-degradation via heat-shock. In combination with detailed studies on its synthesis and characterization, we believe that this work advances the knowledge on poly(ylides) and establishes poly(iminopyridinium ylide) as a functional hydrophilic polymer.

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Polyzwitterionic Coating of Porous Adsorbents for Therapeutic Apheresis

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Objectives: Blood compatibility is crucial in haemapheresis, where adsorbent polymers are in direct contact with whole blood. Here, we present blood-compatible zwitterionic polymers that can be applied as surface coatings on porous adsorbents to enhance blood compatibility, i.e., to minimise the activation of blood cells on the biomaterial surface.

Methods: We developed blood-compatible matrices by surface modification with polyzwitterionic polysulfobetainic and polycarboxybetainic coatings. Photoreactive zwitterionic terpolymers were synthesised by free-radical polymerisation of zwitterionic, photoreactive and fluorescent monomers. Upon UV irradiation, the terpolymers were photo-deposited and mutually crosslinked on the surface of hydrophobic polystyrene-co-divinylbenzene (CG161c, Amberchrom) and hydrophilic polyacrylamide-co-polyacrylate (DALI, Fresenius Medical Care) beads. Blood compatibility was evaluated by assessing polymer-induced haemolysis, coagulation parameters, and cell adhesion to the material surface under static and flow conditions. The remaining adsorption capacity of coated adsorbents was studied in human whole blood.

Results: Fluorescence microscopy revealed coatings with an average thickness of 5 µm, which were limited to the bead surface. The plain synthesised polyzwitterions (1.0 mg/mL), as well as coated adsorbents (10% v/v), did not induce haemolysis (haemoglobin <5 mg/dL). The polyzwitterionic coating of the hydrophobic CG161c adsorbents improved coagulation parameters by significantly shortening prothrombin time (PT), activated partial thromboplastin time (aPTT), and diminishing the adsorption of antithrombin (AT)-III and protein C. In the case of DALI, the coating effect was less pronounced. Coated CG161c maintained an adsorption capacity of 39-67% for IL-6 and 25-35% for TNF-α, while the coated DALI beads preserved 80% of the adsorption capacity for low-density lipoprotein.

Conclusions: Coating enhanced the blood compatibility of hydrophobic adsorbents but not of hydrophilic ones. The most pronounced effect was observed on coagulation parameters (PT, aPTT, AT-III, protein C) and neutrophil count. Polycarboxybetaine with a charge spacer of 5 carbons was the most promising polyzwitterion for the coating of adsorbents for whole blood apheresis.

Keywords: polyzwitterions, coat, adsorbents, extracorporeal therapies, blood compatibility.

Reference: Semak, V.; Eichhorn, T.; Weiss, R.; Weber, V. Polyzwitterionic Coating of Porous Adsorbents for Therapeutic Apheresis. *J. Funct. Biomater.* **2022**, *13*, 216. <https://doi.org/10.3390/jfb13040216>.

A role for zwitterionic surfaces in advanced hull coatings - helping to decarbonize the shipping industry

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Biofouling increases ship drag, fuel consumption, and greenhouse gas emissions. With shipping contributing approximately 2-3% of global anthropogenic CO₂ emissions, the IMO has strengthened decarbonization requirements through measures such as EEXI and CII [1,2]. Effective fouling management and improved hull smoothness can deliver fuel savings of up to 20%, making hull coatings an important contributor to fleet-wide energy efficiency [3,4]. Conventional fouling-control strategies have largely relied on biocidal self-polishing coatings. Although effective, these systems depend on the controlled release of biocides, VOCs, and binder degradation products into the marine environment, creating increasing environmental and regulatory concerns [5].

Recent advances in polymer chemistry and surface science have enabled silicone-based fouling-release coatings that contain little or no biocide. Their performance is based on low surface energy silicone matrices combined with amphiphilic polymers that form a hydrated, hydrogel-like interface in seawater. This dynamic surface reduces biological adhesion and helps maintain low drag throughout service life [6]. However, amphiphilic additives may gradually lose effectiveness through degradation, depletion, or leaching. Zwitterionic surface chemistries offer a promising next step because of their exceptional ability to strongly bind water and resist adsorption of proteins, microorganisms, and other fouling precursors. By creating a highly stable hydration layer on an already smooth hydrophobic surface, zwitterionic technologies could provide longer-lasting fouling protection and further reduce vessel energy consumption. Though significant challenges remain, including scalable manufacturing, durability, application practicality, and cost competitiveness. Nevertheless, zwitterionic coatings represent a promising route toward the next generation of environmentally responsible fouling-control technologies supporting shipping decarbonization.

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Improving the Mechanical Properties of Polyzwitterionic Hydrogels

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Polyzwitterionic hydrogels have attracted significant attention in recent years owing to their excellent antifouling behaviour and biocompatibility.¹ This property combination makes them promising candidates for a wide range of applications, especially in the field of biology and medicine, e.g., for improved wound treatment, skin-attached strain sensors and implants.²⁻⁴ However, the practical implementation of polyzwitterionic hydrogels is held back by their poor mechanical properties.⁵ Such hydrogels are generally brittle and have a significantly lower toughness and resistance to tensile stress than comparable non-zwitterionic hydrogels. Enhancing the mechanical properties of polyzwitterionic hydrogels comes with the additional challenge that their anti-adhesive properties should not be compromised, and many current strategies to strengthen these materials rely on adding non-zwitterionic components, either by making copolymers or polymer double networks, or additives that are not optimized for antifouling applications.⁶ This work aims to resolve that trade-off by strengthening polyzwitterionic hydrogels with the implementation of stronger interchain interactions. The aim of this study is to investigate how an increase in entanglement in the material⁷ and an increase in chain length can maximize the material's mechanical toughness.

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TITLE: Synthesis of peptide-based zwitterionic cross-linkers and evaluation in polyampholyte materials.

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ABSTRACT:

There has been an increased demand for biocompatible materials that exhibit decreased non-specific biological adsorption (i.e., non-biofouling). Polyampholytes have emerged as excellent candidate polymer platforms for these applications as the ionic components attract a strong water layer that inhibits biofouling. The addition of zwitterionic cross-linkers to these materials provides an uninterrupted charged network throughout the material and increasing non-fouling over traditional PEG-based cross-linkers. Peptides are naturally zwitterionic at biological pH and peptide coupling chemistry has been extensively studied and optimized providing ample literature precedence for synthesis. A library of zwitterionic cross-linkers of various lengths, chemistry, and charge density will be presented as to highlight both the synthetic route and how small adaptations can fine tune the resulting polyampholytes for desired physical properties. For example: the dipeptide cross-linker composed of two serine amino acids (Ser-Ser dimethacrylate) has been synthesized at the multigram scale from common amino acid starting materials and has been studied extensively in both bulk hydrogels and in thin-films of 2-carboxyethyl acrylate (CAA) and [2-(acryloyloxy)ethyl] trimethylammonium chloride (TMA) polyampholytes. The results show an increased non-biofouling effect over PEG-based (DEGDMA) systems. Recent expansion into bis(methacrylamides) and polypeptide cross-linkers with additional spacing units has allowed for the examination of the extent of non-fouling while tuning macroscopic properties (i.e., swelling, % hydration, shore 00 hardness, contact angle). Bis(methacrylamide) peptide cross-linker synthesis can be adapted for solid-phase techniques, simplifying the process and allowing for larger scale reactions. Overall, the straightforward synthetic route and high utility of these cross-linkers allows for multiple potential applications.

Bioinspired multivalent nanostructured native materials for energy generation

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In nature, the flux of ions through various channels and ion separation are important processes for generating potential, which could be further converted into usable energy. This inspires the development of materials containing nanochannels with multiple and/or zwitterionic charges at inner surface of nanochannels for energy generation. To fabricate such materials, native nanostructures, in particular cellulose-based, can be used either in their initial states or reconstructed manner. In their initial states, e.g. in wood or other plant tissues, cellulose nanochannels can be obtained after removing major or all amounts of lignin and hemicelluloses. Thanks to the versatile chemical functionalization potential on cellulose surface, multiple charged functional groups and/or zwitterionic charges can be introduced onto such channel inner surface as homogeneous or gradient distribution, using different oxidation methods and further coupling reactions.[1,2,3,4] Such charged nanochannels can be further used for mechanical energy generation and long term discharging. Reconstructed nanochannels can be embedded in membranes using modified nanocellulose, e.g. bacterial cellulose, while charges can be introduced either onto their surface via chemical modification or via doping with charged nanoparticles.[5,6] Using such nanofluidic membranes for local ion separation allows their application for osmotic energy harvesting.

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