CECAM Flagship School

Simulating energy materials with ESPResSo and waLBerla

Book of Abstracts

Stuttgart, Germany
October 9–13, 2023

2023 ESPResSo Summer School

CECAM Flagship School "Simulating energy materials with ESPResSo and waLBerla"

Stuttgart, Germany
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Organizing committee

Jean-Noël Grad, Christian Holm,
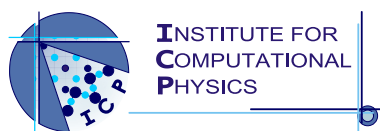Alexander Schlaich, Rudolf Weeber

Edited by

Jean-Noël Grad

Publisher
Institute for Computational Physics
University of Stuttgart
Allmandring 3
70569 Stuttgart
Germany

Conference Program

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Course description

Scientific content

This school will teach the physics and simulation methods used to study energy materials. We will explore topics such as electrostatics in confinement, chemical reactions and catalysis, electrophoretic mobility, diffusion, and electrokinetics.

We will first introduce particle-based approaches and Monte Carlo schemes to model reactions in chemical systems. Then, we will cover the lattice-Boltzmann method for hydrodynamic interactions and a diffusion-advection-reaction solver for modelling electrokinetics and catalysis.

Lectures will provide an introduction to the physics and model building of these systems as well as an overview of the necessary simulation algorithms. During the afternoon, students will practice running their own simulations in hands-on sessions.

Many of the lectures and hands-on sessions will be taught by developers of the software. Hence, the school will also provide a platform for discussion between developers and users about the future of the software. Moreover, users can get advice on their specific simulation projects. Time will also be dedicated to research talks, which illustrate how the simulation software is applied, and which provide further background in the physics of energy materials.

Participants have the opportunity to bring a poster to introduce their work to their peers. We welcome submissions on both planned and ongoing research projects, done with or without ESPResSo, as long as they fit to the general themes of energy materials, fluid dynamics or soft matter physics. The poster session will open with 1-minute lightning talks from all presenters. The poster boards will remain up for the entire duration of the school.

Teaching material

The teaching material used during the school is available online. The ESPResSo software and Jupyter notebooks are free and open-source, available on GitHub (github.com/espressomd/espresso). The lecture slides can be found on the CECAM page for the event (www.cecam.org/workshop-details/1229). The recorded lectures have been published on the YouTube channel ESPResSo Simulation Package.

Hands-on sessions

We use interactive Jupyter notebooks to teach concrete applications of the simulation methods introduced in the lectures. These notebooks outline physical systems relevant to soft matter physics and sketch simulation scripts written for the ESPResSo package using the Python language. A few parts of these scripts are hidden and need to be completed by participants, with the help of the ESPResSo user guide and tutors.

We offer tutoring to all on-site participants and to a small number of online participants via Zoom. These exercises can also be carried out in self-study using the web browser via Binder or Gitpod, and all exercises have hidden solutions that can be revealed at any time.

Software

In this school, students learn to conduct coarse-grained and lattice-based simulations suitable for modeling energy materials, but which can easily be transferred to other fields of statistical physics and soft matter physics, using the software ESPResSo (espressomd.org) and waLBerla (walberla.net). ESPResSo is an open-source particle-based simulation package with a focus on coarse-grained molecular dynamics models. In addition, it offers a wide range of schemes for solving electrostatics, magnetostatics, hydrodynamics and electrokinetics, as well as algorithms for active matter and chemical reactions[60, 58].

ESPResSo consists of an MPI-parallelized simulation core written in C++ and a scripting interface in Python which integrates well with science and visualization Python packages, such as numpy and PyOpenGL. ESPResSo relies on waLBerla, a high performance lattice-Boltzmann library, for hydrodynamics and other lattice-based schemes for electrokinetics and related fields[6, 5].

The organizing committee
Jean-Noël Grad, Christian Holm, Alexander Schlaich, Rudolf Weeber
(Institute for Computational Physics, University of Stuttgart, Germany)

Talks

Challenges in the modeling of energy storage devices

Mathieu Salanne[1,2,3]

There is a strong need for energy storage devices with enhanced performances. Batteries and supercapacitors are in the forefront of this research field. They are characterized by different charging mechanisms, and display complementary energy/power performances. Although many experimental approaches allow to study them, they lack a microscopic view of the different phenomenon at play. They are therefore well complemented by atomic-scale simulations, such as classical molecular dynamics and density functional theory.

In the first part of this lecture, I will shortly describe the main features of batteries and supercapacitors. I will then describe the state-of-the-art approaches used to model them[31]. I will particularly focus on the study of electrified interfaces and of the adsorption of charge carriers inside nanoporous electrodes and intercalation materials. In the last part I will discuss the main challenges that remain to be addressed in the field.

Electrostatics in confinement: From charged soft matter to energy materials

Alexander Schlaich[4]

In this talk I will introduce some basic concepts of soft matter and polymer physics before introducing charged surfaces and their description within the Poisson-Boltzmann mean-field theory. I will then show how this can be related to the electric double layer (EDL) capacitance and put related concepts in their physical context[24]. I will introduce the concepts of anisotropic and inhomogeneous electrostatic interactions at interfaces[42], which significantly improve the EDL capacitance when compared to experiments through local dielectric permittivity profiles[50]. In the context of charges strongly confined between metallic surfaces this leads to strong deviations from the usual 1/r Coulomb scaling. Last, I will show how electrostatic interactions close to a charge polymer can be approximated through a one-dimensional cell model.

Catalysis and reactive force fields

Timo Jacob[5,6,7]

This lecture gives an overview of the ReaxFF[57] forcefield, and presents ongoing research of Ostwald ripening on gold surfaces, catalysis on platinum surfaces presenting defects, and reactions on lithium, sodium, and magnesium nanoparticles, using DFT calculations.

Simulating chemical reaction equilibria in ESPResSo

Peter Košovan[8]

In the lecture we present a toolbox for simulating chemical reaction equilibria in ESPResSo. We start by introducing the thermodynamic background of chemical reaction equilibria. Next, we focus on two key simulation algorithms implemented in ESPResSo: the constant-pH ensemble and reaction ensemble. In the sequel, we discuss the grand-reaction method which is a generalization of the reaction ensemble that enables simulations of reaction equilibria in two-phase systems. Finally, we briefly introduce another generalization of the reaction ensemble, which can be applied to simulate the formation of reversible crosslinks. The discussion of individual methods is supplemented by examples of their applications, based on our recent research results.

[1]Sorbonne Université, CNRS, Physico-chimie des Électrolytes et Nanosystèmes Interfaciaux, PHENIX, Paris, France

[2]Institut Universitaire de France, Paris, France

[3]Réseau sur le Stockage Electrochimique de l'Énergie, FR CNRS 3459, Amiens, France

[4]Institute for Computational Physics, University of Stuttgart, Germany

[5]Biofluid Simulation and Modeling, University of Bayreuth, Germany

[6]Helmholtz-Institute Ulm, Electrochemical Energy Storage, Germany

[7]Karlsruhe Institute of Technology, Germany

[8]Department of Physical and Macromolecular Chemistry, Charles University, Czech Republic

Electrostatic solvers

Christian Holm[1]

This talk introduces the Ewald sum method[20] and the P^{3}M method[16, 17] for fully periodic systems, followed by the Yeh–Berkowitz[62] and ELC[2, 15] methods for partially periodic systems. Then, advanced methods for planar and curved dielectric interfaces will be presented (ICC* algorithm, ELC-IC method)[54, 55, 56, 3], as well as methods available via the ScaFaCoS library[4].

The lattice-Boltzmann method

Stephan Gekle[2]

The lecture will start with an introduction to the physical foundations of hydrodynamics and their mathematical description. We will then move on to a short sketch of kinetic gas theory from which the governing equations of the lattice-Boltzmann method (LBM) will be derived. Some technical aspects such as boundary conditions or the coupling with immersed cells will be briefly introduced. Finally, I will show some examples from our work how LBM can be used for biophysical problems.

Introduction to multi-phase models for the lattice-Boltzmann method

Alexander Reinauer[1]

This lecture gives a brief introduction into various modeling techniques for multi-phase and multi-component simulations within the lattice-Boltzmann framework[35], namely the Shan–Chen method[51], the color gradient method[26, 47, 40] and the free-energy method[52].

The lattice electrokinetics method

Christoph Lohrmann[1], Ingo Tischler[1]

In this lecture, we will introduce an algorithm to simulate the diffusion, advection and reaction of dilute charged species in a solvent. The corresponding electrokinetic equations will be presented and their physical interpretation highlighted. We then outline the lattice electrokinetics algorithm as implemented in ESPResSo[11, 48]. With this algorithm, the equations are efficiently and accurately solved on a regular grid, which also allows coupling to particle simulations. We finish with an overview of various systems which have successfully been simulated using lattice electrokinetics and ESPResSo[36, 53].

ESPResSo applications

Christoph Lohrmann[1]

In this lecture, I will show an overview of the different types of systems that can be simulated using the ESPResSo software package. Instead of diving deep into the details, I will rather focus on the broad variety of applications and the algorithms involved. The examples presented will range from the scale of single atoms up to robots on the centimeter scale and will combine particle-based and continuum-based methods[9, 39, 29, 41, 49].

Introduction to particle-based simulations

Rudolf Weeber[1]

This lecture gives a general overview of various simulation methods used to model the dynamics of physical systems a different time-scales and length-scales, followed by a deeper dive into coarse-grained simulations: Newtonian mechanics, Lennard-Jones potential, thermodynamic ensembles, molecular dynamics integration schemes and trajectory analysis methods.

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[1] Institute for Computational Physics, University of Stuttgart, Germany
[2] Biofluid Simulation and Modeling, University of Bayreuth, Germany

Managing simulation data

Rudolf Weeber[1]

This lecture gives an overview of everyday tools to manage simulation scripts and simulation data: 1) tracking changes to simulation scripts with git, 2) adding command line arguments with argparse, 3) storing simulation results with pickle and pandas, and 4) aggregating results with pandas. Motivation: 1) easier access to simulation results, 2) reproducibility, and 3) resilience against user errors

Error estimation in time-correlated data

Jean-Noël Grad[1]

This lecture gives an overview of statistical tools to estimate the standard error of the mean for physical quantities derived from simulation data. The first chapter introduces stochastic processes, random variables, and estimators. Sources of bias for independent measurements are discussed, along with the concept of bias-corrected estimators. The second chapter is dedicated to time-correlated measurements, which require constructing new estimators to avoid underestimating the standard error of the mean. Common techniques such as auto-covariance analysis, binning analysis, and jackknife analysis are presented[59, 30]. Practical implications for data collection and data analysis are explored using a typical Lennard-Jones fluid simulation as example.

ESPResSo user and developer meeting

Jean-Noël Grad[1], Rudolf Weeber[1], Alexander Reinauer[1], Christian Holm[1]

The ESPResSo core developers will meet with the community of users to report plans for the next software release: waLBerla-based lattice-Boltzmann and diffusion-advection-reaction methods, better support for Monte Carlo schemes, improved code scaleability for HPC systems, and more control over equations of motion and propagation schemes. Results from the ESPResSo benchmark on HPC Vega will be reported[25].

Metallic interfaces: from surface tension to catalysis

Laura Scalfi[2]

Electrode/electrolyte interfaces play an essential role in systems such as capacitors or electrochemical cells. Molecular dynamics simulations and statistical mechanics provide a tool of choice in this field and offer a microscopic picture of the mechanisms involved. In a first part, I will discuss how microscopic charge fluctuations on the electrode surface can modify macroscopic properties such as the surface tension. In a second part, I will present recent work on reactivity at the interface with an electrode where metal ions are embedded on a graphene support.

Ion dynamics in conductive nanosized pores

Svyatoslav Kondrat[3,1]

Conductive nanosized pores are ubiquitous in scientific and technological domains, finding diverse applications, particularly in the field of capacitive energy storage. In my presentation, I will concisely overview the primary challenges of modelling conductive nanopores[33]. The presentation will encompass studies of ion mobility inside such pores and its dependence on the potential difference applied to the pores relative to the bulk electrolyte. We will see that a consensus has yet to be achieved between various simulation results and experimental observations[32, 46, 22]. Furthermore, I will discuss the dynamics of nanopore charging, revealing the existence of four distinct charging regimes[9]. We will see that the charging process is inherently sluggish, primarily due to pore clogging. Consequently, I will elucidate several strategies that have been proposed to expedite this process[9, 10, 43]. Finally, we will examine the discharging dynamics and explore how to accelerate it[10].

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[1] Institute for Computational Physics, University of Stuttgart, Germany
[2] Freie Universität Berlin, Germany
[3] Institute of Physical Chemistry, Polish Academy of Sciences, Warsaw, Poland

Multi-scale models for a better understanding and performance prediction of energy storage materials

Céline Merlet[1,2]

Progress in the development of novel energy storage systems is hampered by our lack of understanding of the microscopic mechanisms that determine their performance. The key issue is that phenomena on the atomistic scale have consequences on macroscopic length and timescales. In particular, the effects of ionic confinement and diffusion are crucial for device performance, yet experiments that probe properties related to local structure and diffusion are challenging and difficult to interpret without a parallel modelling approach. I will focus on carbon-carbon supercapacitors which store energy through ion adsorption at the electrode surface. To understand such systems, it is essential to characterize finely the materials used and the properties of the fluid adsorbed. However, in order to screen materials, computationally efficient methods are needed. I will present insights from two approaches: i) molecular simulations which provide a microscopic understanding of the charging mechanisms and ii) a mesoscopic model we develop, 10,000 times faster than molecular simulations, for the prediction of electrochemical performance. The mesoscopic model allows in particular to bridge the gap between the time and length scales of atomistic simulations, accurate but computationally expensive, and experimental results such as electrochemical measurements and nuclear magnetic resonance spectroscopy.

Machine learning with ESPResSo

Samuel Tovey[3]

Machine learning has begun to permeate almost all fields of science. Underneath these machine learning applications is usually an engine connecting a trained model to some environment for training or deployment. In this talk, I will demonstrate three research areas in which we are using ESPResSo to accelerate our machine-learning research, spanning fields of machine-learned inter-atomic potentials, multi-agent reinforcement learning for micro-robotics, and swarm-driven reservoir computing.

Lightning overview of the MultiXscale project

Alan O'Cais[4,5], Kenneth Hoste[6]

The MultiXscale project of the European High Performance Computing Joint Undertaking aims at increasing the performance, productivity and portability of scientific software used in multiscale simulations. The binaries are built for multiple computer architectures by EESSI[18] using EasyBuild[28] and hosted on the CernVM-FS[8, 13] repository, from where they can be downloaded to the target system with full integration to the Lmod module manager[23]. To showcase the scientific and industrial potential of multiscale simulations, MultiXscale is also carrying out *in silico* pilot experiments in helicopter blade design, batteries design, and ultrasound diagnostics.

Predicting cell stress and deformation during bioprinting

Stephan Gekle[7]

In this talk, I will present simulations on cell deformation during extrusion bioprinting[45, 44].

Ferrofluids and ferrogels: how they respond to magnetic fields and how we capture this in simulations

Rudolf Weeber[3]

This is an introduction to soft magnetic materials such as ferrofluids and ferrogels. I will discuss the applications that arise from the ability to control the material properties by means of external magnetic fields, and provide an overview on the simulation techniques used to model them and present some of our current research.

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[1] CIRIMAT, Université Toulouse 3 Paul Sabatier, Toulouse INP, CNRS, France

[2] Réseau sur le Stockage Electrochimique de l'Énergie, FR CNRS 3459, Amiens, France

[3] Institute for Computational Physics, University of Stuttgart, Germany

[4] Department of Condensed Matter Physics, University of Barcelona, Spain

[5] Centre Européen de Calcul Atomique et Moléculaire (CECAM), EPFL, Lausanne, Switzerland

[6] Department of ICT, Ghent University, Belgium

[7] Biofluid Simulation and Modeling, University of Bayreuth, Germany

Poster session

Investigating a graphene biosensor at the nanoscale using molecular dynamics simulations

Louis Tremblay[1], Colin Rousseau[2], Madline Sauvage[3], Sébastien Côte[1,2], Delphine Bouilly[1,3]

Graphene field-effect transistor biosensors (GFET) are used to detect various types of biological molecules with great sensitivity[7]. To do so, GFETs leverage the unique electronic properties of graphene – a nanomaterial made of a single layer of carbon atoms – making it very sensitive to the presence of nearby electric charges such as found in biomolecules. While many promising GFET platforms were demonstrated over the last few years, it remains difficult to precisely pinpoint the nanoscale physical mechanisms at the origin of the electrical signal generated by a given biomolecule to detect[14]. Here, we use molecular dynamics simulations to interpret the electrical signal generated by a GFET designed to detect cancer biomarkers consisting of 20-nt DNA sequences. We address two questions: (1) how the DNA-graphene interactions change as the probe DNA attached to the graphene binds to the complementary target DNA, and (2) how the potential at the which the graphene is held with respect to the solution in the GFET affects these DNA-graphene interactions? We observe that the DNA in its single-stranded state interacts strongly with graphene, while it lies relatively perpendicular to the graphene in its double-stranded state. Furthermore, we observe that the potential at which the graphene is held doesn't significantly affect these DNA-graphene interactions, over the range of potential values used experimentally. Now, we are improving our modelling of the potential at which the graphene is held. Instead of setting fixed charges on the graphene, we are looking at setting a fixed potential difference between the graphene and the solution using the algorithm ICC* implemented in ESPResSo. We expect that our simulations will provide a better understanding of the nanoscale interactions responsible for the electrical signal generated by this GFET designed to detect cancer biomarkers.

Anomalous bacterial transport in confined geometries

Peixin Zhang[4], Eric Clement[4], Anke Lindner[4]

Motile bacteria are known to interact with flows exhibiting in the bulk active Betherton–Jeffery trajectories or rheotactic drift due to the helical flagella shapes. In the vicinity of bounding surfaces, persistent upstream swimming is enhanced by the presence of edges. Statistically, the combination of hydrodynamic interactions and flow-induced orientation, leads to a strong density increase in the surface vicinity. In disordered environments, geometrically complex surfaces make large-scale dispersion properties of active bacteria a challenging issue. Based on previous study, here we developed experimental model systems suited to observe individual trajectories and to assess the emerging dispersion processes in a funnel-shaped microfluidic device using motile *E. coli* bacteria. An extremely strong anomalous accumulation is observed in the presence of smooth edges and by controlling the shear rate in the system. This work aims to identify the optimal conditions for regulating and amplifying the anomalous transport phenomenon in the presence of geometrically complex surfaces.

Influence of motility and hydrodynamics on phage bacteria encounters

Christoph Lohrmann[5], Sujit Datta[6], Christian Holm[5]

Bacteriophages - or "phages" for short - are viruses that can infect and kill bacteria. They are small particles that rely on thermal diffusion to find target cells, but are also advected in the flow-field generated by motile bacteria. We use coupled lattice-Boltzmann and coarse-grained molecular dynamics simulations to investigate the encounter between phages and bacteria. We find that while motility increases the encounter rate at the cell body, the increase is much smaller than what would be predicted if hydrodynamic interactions were neglected. On the flagella, the opposite effect is observed. This has important implications for our understanding of the evolutionary cost that bacteria have to pay for their motility.

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[1] Department of Physics, Faculty of Arts and Sciences, Université de Montréal, Montréal, Canada
[2] Department of Physics, Cégep de Saint-Jérôme, Saint-Jérôme, Canada
[3] Institute for Research in Immunology and Cancer, Université de Montréal, Montréal, Canada
[4] Laboratoire de Physique et Mécanique des Milieux Hétérogènes, UMR 7636, CNRS, ESPCI Paris-PSL, Sorbonne Université, Université Paris Cité, France
[5] Institute for Computational Physics, University of Stuttgart, Germany
[6] Department of Chemical and Biological Engineering, Princeton University, New Jersey, United States

Modelling electrode interfaces via multi-scale simulations

Henrik Jäger[1], Philipp Stärk[1], Alexander Schlaich[1]

The structure of electrode/electrolyte interfaces is routinely addressed using first principles simulations based on density functional theory. Yet, (first principle) molecular dynamics simulations at controlled electrode potential with fluctuating number of electron remains challenging. Combining first principle-calculations with force field molecular simulations opens length- and timescales to allow the description of dynamics of confined electrode systems with high accuracy. Kohn–Sham DFT is the first principles method with which the lower bound of the force field based molecular dynamics timescales can be reached. By obtaining the material dependent Thomas–Fermi screening lengths from first principles simulations, one can bridge the gap between the length-scales of ab-inito and force field-based methods.

Modelling hierarchical porous electrode materials via molecular dynamics

Philipp Stärk[2]

Electrode systems are fundamental components of energy storage and catalysis devices, and their properties at different length scales are essential for optimizing their performance. Nano-porous materials are particularly promising due to their advantageous physical properties and very high specific surface area. However, the physics of confined fluids in nanometer-scale pores differs significantly from that of bulk fluids, as the atomistic nature of fluid molecules and pore-walls becomes important. In such cases, continuum assumptions break down, and simulations and experimental evidence must be used to understand what is truly happening at that length scale. This necessitates consideration of quantum mechanical details, which become important at this length scale.

To incorporate quantum mechanical details into our models while still achieving the necessary time-scales, we use force-fields that are optimized using quantum mechanical data. Additionally, for materials with metallic properties such as graphene, we utilize a constant potential scheme with parameters that are tuned to quantum density functional theory.

However, since most of the technically interesting observables are of macroscopic nature, such as for instance the capacitance, reactivity, and transport, it is essential to bridge the scales between atomistic simulations and macroscopic observables. To achieve this, we use coarse-graining procedures that allow us to retrieve continuum models with modified parameters which capture the atomistic details effectively. These procedures involve integrating out the degrees of freedom of individual atoms or molecules to obtain a simplified model that captures the essential features of the system at larger length and time scales.

In order to accurately model the behavior of electrode pores, it is crucial to consider them as open systems, where only a small subsystem is explicitly simulated, allowing for particle exchange with the outside. This approach enables fluctuations in particle numbers and changes in composition due to complex interactions between the wall, fluid, and any applied potential on the electrodes in the system. Thermodynamically, open systems are best described by a grand canonical ensemble, where both the number of particles and energy can fluctuate with the environment.

When modeling the grand canonical ensemble in simulations, Grand Canonical Monte Carlo methods are commonly used. However, these methods can be slow and inefficient, especially for larger systems and when multiple species of fluid molecules are involved. For this reason, we employ the Wang–Landau meta-sampling method. The Wang–Landau sampling method allows us to efficiently determine the density of states of the system, by iteratively approximating the free energy surface. By using this method, we hope to accurately capture the behavior of electrode systems across different length scales and investigate the behavior of confined fluids in binary and even tertiary mixtures. This is essential because the behavior of confined fluids in these mixtures can differ significantly from bulk fluids, and the behavior of the electrode can also be affected.

Our research has important implications for a range of applications in energy storage and catalysis, including the conversion of CO_2 to fuels. By improving our understanding of the behavior of open electrode pores, we can optimize the design of electrode systems and improve their performance for these applications. Our poster highlights the importance of understanding the behavior of open pores in electrode systems and the usefulness of the Wang–Landau method in this context.

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[1]Institute for Computational Physics, University of Stuttgart, Germany
[2]GS SimTech, Stuttgart, Germany

Mixed ionic-electronic transport – GRK 2948

Richard Schömig[1], Alexander Schlaich[1]

We will present some aspects of mixed electronic/ionic transport in conjugated polymer systems and our strategy to include them in coarse-grained simulations. We will also present the recently established graduate school between Heidelberg and Stuttgart universities.

Path integral molecular dynamics simulations using ESPResSo

Devashish Tiwari[2], David Beyer[1], Christian Holm[1]

There are many physical systems (e.g. quantum gases) that require a simultaneous treatment of thermal and quantum fluctuations. The path integral formalism, originally developed by Feynman to study the time evolution of quantum systems[21], is a useful tool in the study of such systems. Via a so-called "Wick rotation" to imaginary times, path integrals can be used to study quantum statistical mechanics. Numerically, time-discretized paths have to be considered, which are isomorphic to classical ring polymers[12]. This isomorphism allows for a straightforward numerical evaluation of quantum properties using classical simulation techniques such as molecular dynamics. Here, we use path integral molecular dynamics in ESPResSo to study the statistical mechanics of a quantum harmonic oscillator[61]. Our simulation results are in excellent agreement with analytical predictions for the density matrix and energy levels.

Simulation of polyelectrolyte hydrogels with various charge densities and network defects

Somesh Kurahatti[1], Mariano Brito[1], David Beyer[1], Christian Holm[1]

The properties of homogeneous gel network structures have been extensively studied using MC and MD simulation, alongside their synthesis in the laboratory. Polyelectrolyte hydrogels possess a distinctive capacity to function as desalination agents when placed in saline solution and they possess remarkable water absorption capacity. In recent experimental investigations[27], hydrogels have been employed to desalinate saline water through a forward osmosis process without explicit membrane. The charges within the polymer result in an uneven distribution of salt between the gel and the solution phase due to Donnan partitioning, consequently leading to higher salt concentration in the supernatant. Homogeneous network structures can be easily prepared in the lab[1] and they are studied extensively through theoretical models and computer simulation[34]. We want to investigate the effect of network heterogeneity on the macroscopic quantities such as swelling capacity, mechanical properties and the salt partition. In many applications such as superabsorbers, it is desirable to have increased swelling capacity without compromising the bulk modulus. The current project is concerned with decoupling of swelling capacity and bulk modulus of the hydrogel by the introduction of various charge densities in the form of defects. To better understand the effect of added charge densities, salt-free simulation of hydrogels with attached bottle-brushes and floating linear chains are performed. The preliminary results show floating chains of suitable length seem to be a promising way to decouple swelling capacity from bulk modulus of the hydrogel. We also plan to design various network structures and study hydrogels with dangling ends, dendrimers, charged inclusion of polyelectrolyte stars. These structures allow to realize high charge density without significantly increasing the bulk modulus. In the previous studies, polydispersity in the chain length was added in the mean field gel models[38, 37] and swelling results showed pronounced differences when compared to a simulation which takes explicit polydispersity[19] and further work is needed to understand this mismatch better. In the simulation, the gel would be imposed with different volumes to obtain a pressure-extension curve which allows us to calculate the isothermal bulk modulus. The system in contact with ionic solution will be modelled using Grand Canonical Monte Carlo ensemble. All simulations will be carried out using home-grown software ESPResSo[60]. In the future, we will also investigate the effect of multi-valent ions on the mechanical properties and salt partitioning of the hydrogel.

[1] Institute for Computational Physics, University of Stuttgart, Germany
[2] Indian Institute of Science Education and Research, Bhopal, India

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