Research Project #15

Combining large scale atomistic simulations with neutron scattering experiments for understanding of interfacial dynamics of water, ligand and iron oxides for energy applications (IET-3 & JCNS-3)

At IET-3 we pursue research in theory and computation that complements experimental materials research on novel electrocatalysts. Crucial scientific challenges in this context are addressed with numerical atomistic modelling, either *ab initio* or molecular mechanics-based. These challenges encompass processes that alter structural, mechanical and thermodynamic properties of solid materials and their interfaces with the aqueous phase, including microscopic charge transfer, degradation via catalyst dissolution or support corrosion, and interfacial diffusion. To assure reliability of the computational workflows we develop accurate and efficient computational methodologies and evaluate their feasibility by comparison with experimental data provided by our partners. Among them, JCNS-3 applies neutron scattering and X-ray methods as a unique probe to characterize the structureand dynamics of materials over many length and time scales, studies energy systems in-situ and operando, as well as develops and operates neutron instruments at neutron sources.

Despite being omnipresent in various applications spanning fuel cells, heterogeneous catalysis and biomedicine, the dynamics of water molecules diffusing on nanostructured iron oxide surfaces are not yet well understood. This is in part because of the presence of organic ligand molecules that stabilize the nanostructured surfaces, producing complex interfacial behavior. Because a neutron favorably interacts with hydrogen, quasielastic neutron scattering (QENS), mastered at JCNS-3, is an ideal technique to investigate interfacial diffusion dynamics of hydrogen-containing species. It allows us to gain insight into the relaxation times, activation energies and geometry of motion of water molecules on nanoparticle surfaces. This information can be interpreted with the aid of atomistic simulations mastered at IET-3 and enabled by superior supercomputing facilities of the research center, particularly the exascale JUPITER supercomputer. This allows us to disentangle the diffusion dynamics of ligand molecules from that of light and heavy water molecules.

The aim of this project is to perform advanced, large-scale molecular dynamics simulations based on machine learning (ML) to interpret the neutron spectroscopy experiments on iron oxide nanoparticle powders. These nanoparticles will be synthesized according to a strategy well-established at JCNS-3, and stabilized with either citrate, lactic acid or diethylene glycol molecules, with samples equilibrated at different relative humidities of D_2O, which reflects a different number of water layers on the nanoparticle surface. The foreseen computational results will be integrated into the data analysis of the neutron spectroscopy data, in order to achieve a global understanding of the interfacial dynamics of D_2O and H_2O molecules. Part of the neutron data is already available from a running project; further neutron experiments shall be complemented by this project. Combining data from computation and experiment will allow to achieve new insights into interfacial dynamics of water-ligand-iron oxide surfaces.

This highly interdisciplinary and collaborative project will be executed by combining the long-standing experience of your hosts (IET-3 and JCNS-3) and collaboration partners. You will learn state-of-the-art high-performance simulation and data analysis techniques. Studies will be performed on world-class facilities at Forschungszentrum Jülich with the JUPITER exascale machine and world-wide available neutron scattering facilities as main resources.

Specific tasks are:

- to employ high-performance exascale supercomputing resources for nanoscale simulations of iron-oxide interfaces,
- to develop AI/ML force fields for use in computer-based simulations of iron-oxide comprising tens of thousands to millions of atoms and validate it against *ab initio* simulations on smaller scale models (up to a few hundreds of atoms),
- to perform neutron spectroscopy experiments at neutron sources worldwide including sample preparation and complementary physicochemical characterization (JCNS-3),
- to apply the devised force fields to large-scale molecular dynamics simulations of the particle-level processes such as interfacial diffusion, to provide support in interpreting the neutron scattering data,

- based on simulated and measured data to characterize diffusion mechanisms and identify differences of H_2O and D_2O diffusive properties at iron-oxide interfaces,
- to effectively collaborate on the topic with the internal and external partners,
- to publish results in peer-reviewed scientific journals and presentations at conferences and workshops.

Location of the HITEC Fellow	Forschungszentrum Jülich, Institute of Energy Technologies - Theory and Computation of Energy Materials (IET-3), Director: Prof. Dr. Michael Eikerling, Computational Materials Modelling Division (Head: Dr. Piotr Kowalski) https://www.fz-juelich.de/en/iet/iet-3
Partners of the HITEC Project	Forschungszentrum Jülich, Jülich Center for Neutron Science - Neutron Analytics for Energy Research (JCNS-3), Director: Prof. Dr. Mirijam Zobel https://www.fz-juelich.de/en/jcns/jcns-3
Specific **requirements**	M.Sc. in Chemistry, Physics, Computational Materials Science, Geoscience or related disciplines; Experience in high performance computing as well as in X-ray or neutron scattering and data analysis will be an advantage; Good command of written and spoken English
For **project specific** questions please **contact**	Dr. Piotr Kowalski, IET-3, p.kowalski@fz-juelich.de (simulations/modelling) Prof. Dr. Mirijam Zobel, m.zobel@fz-juelich.de (neutron scattering, experiments)