Soft Matter and Polymers

1 Sustainable Materials

Building a circular, eco-friendly and self-sustaining economy in the present that ensures the security, well-being and prosperity of our future is a grand challenge for all scientific disciplines. Soft matter and polymers mimic natural materials across the smallest to largest scales making them a critical part of the solution. However, future material innovation in soft matter and polymers will not only benefit from a deeper understanding of existing and natural materials but also require improvement in bioinspired material discovery, processing and performance for a targeted application. Therefore, a start-to-end lifecycle analysis of new and existing soft materials and polymers is essential, and the STS will play a central role in achieving this goal.

1.1 Scientific and Technological Impact

Soft materials and polymers are being employed in multiple areas of research and technology and hold the key to achieving our sustainability goals. We have identified the following areas of research and innovation that align with this grand challenge:

a. **Clean Energy:** Comjugated and charged polymers are being used in clean-energy research to device fuel cells for a hydrogen economy. Polymer-based membranes also play a crucial role in solid-state batteries which reduce reliance on conventional petroleum energy sources. A key role that polymers can play in this field is in improving the efficiency and service life of clean-energy fuel cells and batteries. [1.1]

b. **Food and water security:** Soft matter research is directly contributing to security and availability of water and food for our future generations. Improvements in water harvesting and reverse-osmosis technology by designing next-generation polymer membranes that are cheap and yield efficient potable water generation is key to solving the looming water crisis. Bioinspired polymers are also going to play a critical role in enhancing shelf lives of food and during storage, thus contributing to food security. Designer biopolymer based artificial topsoil that retains water and macronutrients as well as provides a friendly environment for nitrogen, carbon and phosphorous fixing micro-organisms can also contribute to solving the agricultural problems faced by the US farmer today. [1.2]

c. **Additive manufacturing (AM):** The future of manufacturing and housing hinges on innovations in AM technology. In the future, soft materials that flow while printing and harden to form custom-made parts will be required. Thus, to ensure a safe and healthy environment in the future, discovery of sustainable, low-carbon footprint soft materials is essential, especially in the field of bio-cement and polymer nanocomposite engineering. These classes of soft materials are hierarchical in structure and the final performance of the material is dependent on their flow properties and processing history. [1.3]

1.2 Relevance to STS

Since neutrons are non-abrasive, non-ionizing and non-destructive, soft and polymer materials especially benefit from innovations in neutron scattering techniques. Furthermore, these materials are abundant in hydrogen that strongly scatter neutrons. Neutron scattering measurements at STS will leverage its high-flux and multi-faceted capabilities in neutron imaging, tomography, spectroscopy and scattering to gain further insight into molecular-design driven structure-property-performance relations in soft matter and polymers. These efforts will unlock the discovery of next generation sustainable materials because (i) the materials of interest are multi-component that can benefit from detailed- study of interfaces as well as contrast-variation experiments with good signal-to-noise ratio (ii) their inherent hierarchical and non-uniform structure exhibit a wide-range of dynamics that can only be probed with high-flux cold neutrons (iii) processing of polymers is intimately linked to their performance that can be probed by investigating structure evolution using event-mode data acquisition with high-flux neutrons.

1.3 Example of Game-Changing Outcomes

Block copolymer membranes have been used to harvest water from the atmosphere. [1.4] It has been postulated that the precipitation of water on the membrane is an interfacial phenomenon and can be hastened by wrinkling or patterning of the membrane surface or by introducing additional hydrophilic-hydrophobic interfaces on the surface. A key experiment is measuring the impact of the smoothness at the interface on the gas-to-liquid phase transition of water as well as measuring the transport at the interface which can inform future designs of membranes with better water harvesting efficiency. Neutron imaging of PS-PAA membranes in a controlled humidity environment coupled with *in operando* small-angle neutron scattering measurements would be uniquely suited to not only visualize the process but also answer these interface design and transport questions. However, currently, investigating the *in operando* interfacial phenomena between hydrophobic and hydrophilic polymers is inaccessible due to the low flux and poor length resolution of the neutron imaging instruments. STS will therefore be a leader in interfacial transport phenomena research with its next generation neutron-imaging station.

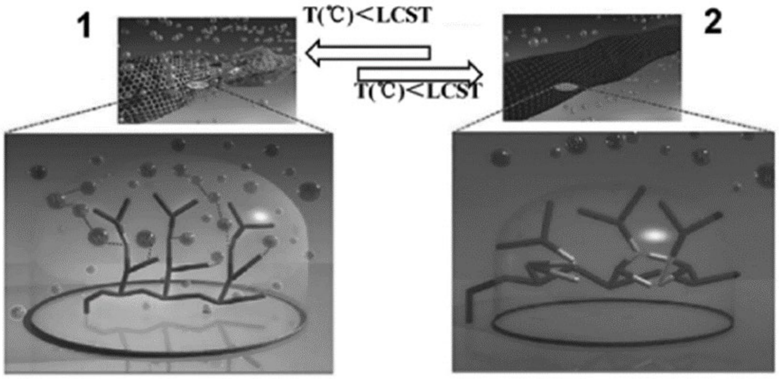

Figure 1: Water collection and release at the interface for PNIPAAm polymer. PNIPAAm is a temperature-responsive polymer that is widely used in polymer membranes. (1) Water collection in “superhydrophilic state” by hydrogen bonding between PNIPAAm and water, and (2) Water release in “superhydrophobic state” by the formation of PNIPAAm intermolecular bonds. *In operando* investigation of the structure and dynamics of the polymer-water interface will result in design of next-generation water harvesting technologies.

2. Dynamics and Evolution

Disorder and the presence of light elements such as hydrogen, carbon, and nitrogen are common features of all soft materials. A grand challenge is to design new soft materials for applications in the emerging clean energy landscape. New materials are defined by atoms and ions and the connections between them. While AI offers new avenues to design combining the atoms and ions into new motifs for enhancing properties such as ion transport, the proposed scattering and imaging instruments at the STS are necessary for validating these designs.

2.1 Scientific and Technological Impact

Computer simulations, based on established force fields and interaction energies, provide enormous insight into system properties on the nanometer and nanosecond scales. Equilibrated simulations on large enough scales provide insight into macroscopic phenomena. While the predicted structure (locations of atoms and ions) can be tested by methods such as cryo-EM and microdiffraction, inelastic neutron scattering at the STS will provide the only test of the simulated timescales. Practical devices contain features such as pores and catalyst particles on the micron length scale. We will track the transport of mass and charge using the mask-based STS tomography instrument which will have the requisite resolution. High brightness will enable rapid transient measurements on the second timescale, commensurate with practical applications. Our objective is to compare device-level observations with theoretical predictions based entirely on intermolecular interactions. It is common to use different force-fields such as CHARMM general force field (CGenFF) or the optimized potentials for liquid simulations with all atom model (OPLS-AA) in different disciplines. By additional comparisons between simulated *dynamics* with experiments at STS, we aim to converge on a single set of interaction energies that are used in all simulations relevant to both biology and technology. This may require abandoning the present approach wherein complexities such polarizability and other quantum effects are approximated by constructs such as partial charges.

2.2 Relevance to STS

While time scales relevant for biological and technological applications is typically larger than 10^{-3} s, most of the factors that underlie observations occur on time scales between 10^{-12} (small molecule relaxation) to 10^{-5} s (relaxation of polymer modes). The timescales accessible by spectroscopic techniques (*e.g.*, NMR, IR and Raman) lie below 10^{-12} s, and thus experimental determination of relevant time scales remain controversial. For example, estimates of solvent relaxation times in lithium-ion battery electrolytes, first estimated to be 10^{-12} s [3.1], has been revised up to 10^{-10} s with no experimental validation [3.2]. The lack of such fundamental knowledge impedes rational design; the major components of lithium battery electrolytes used today are the same as those used in the original patents [3.3]. The suite of inelastic and quasi-elastic instruments at STS will cover timescales from 10^{-12} to 10^{-5} s, thereby bridging the crucial dynamical gap between spectroscopy and conventional relaxation experiments such as dynamic light scattering. While AI-based materials designs are accelerating, their impact on practical applications remains limited. While it is easy to introduce a new material and measure device performance, determining the reason for the performance change is very challenging.

2.3 Example of Game-Changing Outcomes

The emerging hydrogen economy will rely on electrolyzers to produce the hydrogen and fuel cells to provide clean energy. We show a fuel cell in Figure 3 wherein H_2 is used to produce electrons and water. Let us assume we are interested in testing a new membrane that is supposed to limit the crossover of hydrogen through the device. A thorough understanding of device performance requires quantifying the flux of H^+ that will be primarily driven by migration, and the fluxes of H^+, H_2 and H_2O that are primarily controlled by diffusion. The crossover of both H_2 and H_2O impact energy conversion efficiency but little is known about these important effects. How does one distinguish between these 3 different hydrogen-containing species, especially inside the electrodes that contain heavy elements like iron and platinum? The STS tomography instrument with suitable isotope substitution is the only approach for mapping H^+, H_2 and H_2O in a device, as shown in Figure 3. Time-resolved images will give the flux of the non-deuterated species as a function of space and time. Such instrumentation will provide the missing fundamental knowledge necessary to create the hydrogen economy.

Let us assume, for example, that a company designs a membrane to improve the efficiency of an electrolyzer by reducing hydrogen crossover. At present, one would introduce this membrane and see if the efficiency of the electrolyzer improves. However, the STS approach allows for independent measurement of H_2 crossover and H^+ flux simultaneously, thereby providing fundamental understanding that cannot be obtained otherwise. One can construct similar experiments to probe other parts of the hydrogen economy such as hydrogen storage and release.

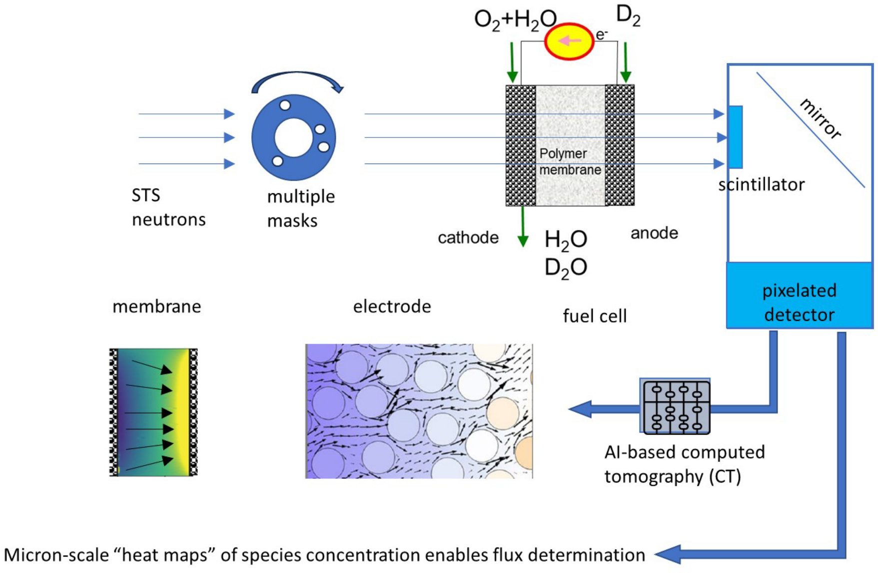

Fig. 3.3. *Operando* time-resolved STS tomography experiment for imaging hydrogen in a fuel cell with using the ghost imaging Hanbury-Brown-Twiss approach. The fuel cell is rotated in the beam with multiple static masks to give a series of 2D images that will be analyzed by AI to give heat maps of hydrogen concentration on the micron length scale.

References

[1.1] Luo S, Zeng Z, Wang H, Xiong W, Song B, Zhou C, Duan A, Tan X, He Q, Zeng G, Liu Z. Recent progress in conjugated microporous polymers for clean energy: Synthesis, modification, computer simulations, and applications. Progress in Polymer Science. 2021 Apr 1;115:101374.

[1.2] Brown LR. The global loss of topsoil. Journal of Soil and Water Conservation. 1984 May 1;39(3):162-5.

[1.3] Al Rashid A, Khan SA, Al-Ghamdi SG, Koç M. Additive manufacturing of polymer nanocomposites: Needs and challenges in materials, processes, and applications. Journal of Materials Research and Technology. 2021 Sep 1;14:910-41.

[1.4] Zhang S, Huang J, Chen Z, Lai Y. Bioinspired special wettability surfaces: from fundamental research to water harvesting applications. Small. 2017 Jan;13(3):1602992.

[3.1] Lee, K.-K.; Park, K.; Lee, H.; Noh, Y.; Kossowska, D.; Kwak, K.; Cho, M., Ultrafast fluxional exchange dynamics in electrolyte solvation sheath of lithium ion battery. *Nat. Commun.* **2017,** *8* (1), 14658.

[3.2] Dereka, B.; Lewis, N. H.; Zhang, Y.; Hahn, N. T.; Keim, J. H.; Snyder, S. A.; Maginn, E. J.; Tokmakoff, A., Exchange-mediated transport in battery electrolytes: Ultrafast or ultraslow? *J. Am. Chem. Soc.* **2022,** *144* (19), 8591-8604.

[3.3] Yoshino, A.; Sanechika, K.; Nakajima, T.; “Secondary Battery”, US Patent 4,668,595, **1987**.

[3.4] Padgett, M.J.; Boyd, R.W.; “An introduction to ghost imaging: quantum and classical”, Phil. Trans. Royal Soc. A, **2017**, 375, 20160233.