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Neutron reflectometry for in operando characterisation of the solid electrolyte interphase (SEI) in electrochemical lithium-mediated nitrogen reduction reactions (Li-NRR)

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As the global population grows, the demand for essential resources such as food and energy will place a significant strain on the climate owing to the high levels of anthropogenic emissions generated during production. Despite advancements in renewable energy sources, such as solar power, the limitations of existing infrastructure hinder our ability to efficiently distribute harvested energy. Converting energy into chemical fuels allows for reliable long-term storage, enabling locally harvested renewable energy solutions to be utilised worldwide. Carbon-free options, such as ammonia, can be engineered to ensure negligible emissions at the point of use1,2. To produce economically viable, truly carbon-free ammonia fuel, it is essential to develop an energy-efficient synthesis process.

An ammonia synthesis method growing in popularity is the electrochemical reduction of nitrogen from the atmosphere. Whilst protons would ideally be obtained from a sustainable resource such as the electrolysis of water, the delivery of protons must be precisely controlled due to direct competition from the hydrogen evolution reaction (HER)3,4. To date, strictly aprotic, lithium mediated systems are the only systems that exhibit performance relevant to the industrial scale5,6. Further improvements are still required (alternative mediators7,8, electrolyte decomposition9,10 proton donors10) to enhance the sustainability of this technology. Accurate assessment of these changes will require comprehensive investigation of the solid electrolyte interface (SEI).

This research, conducted at ANSTO's Open Pool Australian Light-water Reactor (OPAL), focuses on the development of high-performance electrochemical cells optimised for in operando analysis using neutron reflectometry. This approach allows for characterisation of the SEI layer composition across various mediators, electrolytes, and proton delivery systems. The advanced procedures and analysis techniques developed by this work will be discussed. Importantly, this methodology constitutes a versatile and comprehensive ongoing research project, which will continuously be adapted to accommodate future advancements in electrochemical Li-NRR system development.

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Topics

Neutron Instruments and Techniques

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