

Students' Annual Seminar

Tailoring the Interfacial Water Structure by Electrolyte Engineering for Selective Electrocatalytic Reduction of Carbon Dioxide- A Spectroelectrochemical Perspective

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Engineering aqueous electrolytes with low amounts of additives to achieve a tunable CO₂ reduction product is an underexplored territory in electrocatalysis. Here, we show the enhancement of the Faradaic efficiency (FE) of CO₂ reduction to CO on unmodified polycrystalline gold from ~67% to ~94% by the addition of up to 15 mol % of N,N-dimethylformamide (DMF) to an aqueous electrolyte. The role of electrolyte structure modification near the electrode-electrolyte interface was studied using in situ surface-enhanced infrared absorption spectroscopy in the attenuated total reflection mode (ATR-SEIRAS). In addition to the expected detection of the adsorbed CO (CO_{ad}) intermediate present on the Au surface, in both the linearly- and bridged-bonded forms, we observed changes in the structure of interfacial water induced by the addition of DMF. The changes in the water bending band and the DMF carbonyl band indicate an increase in the strongly hydrogen bonded DMF-water pairs with increasingly negative potential near the interface in the presence of DMF. We hold this interfacial water structure modification by DMF responsible for increasing the CO₂RR FE and decreasing the competing hydrogen evolution reaction (HER). Furthermore, the suppression of the HER is observed in other electrolytes and also when platinum was used as electrode, and hence can be a potential method for increasing the product selectivity of complex electro-catalytic reactions.

Friday, Mar 24th 2023

10:30 AM (Tea / Coffee 10.15 AM)

Seminar Hall, TIFR-H