

Investigating the effects of interface dynamics on the positioning of catalyst in SILP catalysis

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This work investigates the effects of interface dynamics on the task-specific catalyst enrichment within SILP (Supported ionic liquid phase) catalysts at the molecular level. SILP catalysts are composed of a molecular catalyst which is dissolved in a thin film of ionic liquid (IL) covering a porous support material. Thus, SILP combines the flexibility of homogenous catalysis with a solid, long-term stable, macroscopically dry, and free flowing material behaviour. It allows task-specific tuning via variation of the IL, the support and, especially, chemical modification of the molecular catalyst towards enhanced enrichment at the SILP interfaces. The latter converts the classical SILP into an interface enhanced SILP catalyst with superior catalytic properties.

In order to pinpoint the effects of interface dynamics on the catalyst enrichment under realistic reaction conditions within interface enhanced SILP, we conduct Molecular Dynamics simulations and free energy calculations were performed on mixtures of IL and ligand-modified catalysts with pressurized molecular hydrogen over a broad range of temperature and pressure. Effects of the H₂ atmosphere on the properties of IL were studied, along the placement of the catalyst on the IL-vacuum interface. Finally, the cooperative effects of both the H₂ atmosphere and the catalyst functionalisation was explored.

Our results indicate that a mild surface enrichment of H₂ at the interface with the IL affects the surface tension of IL in a pressure-dependent manner.^{1,2} Enrichment effects at the IL-vacuum interface, now of the catalysts are also observed depending on the functionalisation of the catalysts' ligands. Specifically, the fluorinated catalyst exhibited a tenfold increase in the surface affinity over the unmodified Wilkinson catalyst, also reflecting in the surface tension of the IL film. Finally the synergistic effect of both H₂ and other small gasses enrichment in conjunction with the catalyst functionalisation are explored. These findings suggest that by altering the surface properties of the interface and modifying the functionalities of the catalyst we can control the positioning of the catalyst at the interface, which can, in turn enhance the efficiency of SILP catalysis.

[1] Zhai, Z., et al., International Journal of Hydrogen Energy, 72,2024, 1091-1104, DOI:10.1016/j.ijhydene.2024, 05, 249.

[2] Zhai, Z.,et al., Journal of Molecular Liquids, 2020, 41, 122388, DOI:10.1007/s10765-020-02720-w.