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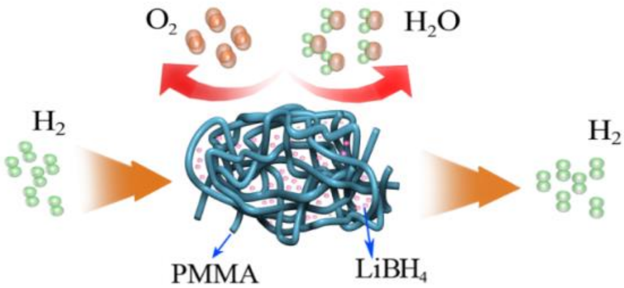
Dehydrogenation mechanism of $LiBH_4$ by Poly(methyl methacrylate)

Hydrogen is considered as a promising alternative energy carrier owing to its high-energy density, abundance, light weight and pollution-free burning [1]. Developing a safe and efficient hydrogen storage material is one of the key challenges for the mobile application of hydrogen [2]. Due to the high gravimetric (18.5 wt.%) and volumetric (121 kg H_2/m^3) hydrogen density, lithium borohydride ($LiBH_4$) has been acknowledged as a potential candidate for hydrogen storage materials [3]. However, due to the unfavorable high thermal stability (e.g. decomposition peak temperature of ~ 470 °C), the practical utilization of $LiBH_4$ as hydrogen storage medium is hampered [4]. Hence, several approaches including reactant destabilization, catalyst/additive introduction, nanostructuring, and anion/cation substitution have been applied to decrease the dehydrogenation temperature and accelerate the kinetics [5].

Nanoengineering had been demonstrated to be a useful method to reduce the dehydriding/rehydriding temperature of $LiBH_4$ by decreasing diffusion path lengths and increasing surface areas [6]. However, nanoscale $LiBH_4$ is too reactive and very sensitive to the water and oxygen in the air, which impede its practical utilization. PMMA ((Poly (methyl methacrylate))), with a high permeability ratio of H_2/O_2, was reported to have good gas selectivity [7]. Therefore, in this project, PMMA was applied to protect $LiBH_4$ from oxygen and water but let the hydrogen get in or out freely (Scheme 1). Furthermore, the nanoconfinement of $LiBH_4$ in the fine network pore of PMMA and the interaction between the B atom in $LiBH_4$ and the O atom in C=O of PMMA resulted in a much lower hydrogen release temperature of $LiBH_4$. $LiBH_4$ PMMA composite started to dehydrogenate at 53ºC and released 5.2 wt.% of hydrogen at 162ºC within 1 h. This project provides a general strategy to utilize a gas-selective polymer to protect air-sensitive hydrogen storage compounds and improve their hydrogen storage properties.



Scheme 1. Schematic illustration of $LiBH_4$ protected from oxygen and water by PMMA.

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