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Electrochemical Regeneration of LiAlH₄

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Hydrogen, a promising clean energy vector, surpasses traditional fuels in energy content (140 MJ/kg) but faces storage challenges. Aluminum-based complex metal hydrides like LiAlH₄ offer solutions with high volumetric (>80g H₂/L) and gravimetric (>8 wt %) energy densities, along with low decomposition temperatures (<423 K). However, their synthesis involves costly organometallic reactions and high hydrogenation pressures (>1 kbar). Consequently, a significant interest is in cost-effective approaches for regenerating LiAlH₄ from its spent (hydrogen-depleted) material and H₂.

Graetz^[1] proposed its first chemical regeneration, examining dehydrogenated LiH and Al with a Ti catalyst in THF under high H₂ pressure (50 bar). Humphries^[2] explored sub-ambient regeneration (<0°C) using Me₂O and 95 bar H₂. However, their practicality is hindered by reliance on high H₂ pressure. Our research group investigates the viability of an electrochemical approach for its regeneration and potential elimination on the use of high H₂ pressure, a novel concept not previously reported in the literature..

Electrochemical properties were studied in THF-based electrolyte and Ar atmosphere at RT. Figure 1 shows a cyclic voltammogram of 1M LiAlH₄ in THF to which we identified essential reactions, including Li and Al deposition, and irreversible anodic polarization of [AlH₄⁻] associated with H₂ release^[3]. Applying a constant reduction potential (-3.10 V vs. standard hydrogen electrode) resulted in dehydrogenated LiAlH₄ (Li-Al alloy formation) and H₂ release, demonstrating room-temperature electrochemical dehydrogenation with a Faradaic efficiency of 69%. We explore the effect of H₂ pressure on these electrochemical reactions using our custom electrochemical pressure cell. The next step involves regenerating the deposited Li-Al alloy under moderate H₂ pressures (<10 bar) and applying oxidative potentials to induce [AlH₄⁻] reformation using Pd, Pt, Ti, or Ni electrocatalysts. Reformed LiAlH₄ is quantified using ²⁷Al NMR coupled with ICP-MS. Fundamental parameters will be investigated, including its kinetics, diffusion coefficients, and regeneration mechanisms, using various electroanalytical techniques like chronoamperometry, voltammetry, rotating disk electrode, and electrochemical-impedance spectroscopy.

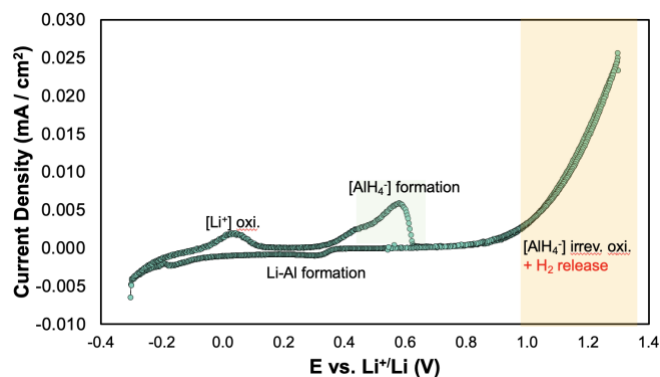


Figure 1. CV of a 1M LiAlH₄ – THF solution under 7 bar of Ar with a scan rate of 20 mV/s.

References

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