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Particle-size dependence of the activation energy for decomposition of lithium amide¹ CHRIS VAN DE WALLE, Materials Department, University of California, Santa Barbara, KHANG HOANG, Naval Research Laboratory, ANDERSON JANOTTI, Materials Department, University of California, Santa Barbara — Lithium amide (LiNH_2) is a promising material for reversible hydrogen storage, yet atomistic mechanisms behind the dehydrogenation process are unknown. The activation energy for LiNH_2 decomposition has been observed to strongly vary with ball milling, suggesting a dependence of the thermodynamics and kinetics of the decomposition on the particle size. We have examined these mechanisms based on first-principles calculations for native point defects and defect complexes in LiNH_2 . We propose that the decomposition of LiNH_2 into lithium imide (Li_2NH) and ammonia (NH_3) occurs through two competing mechanisms, one involving the formation of native defects in the interior of the material and the other at the surface. As a result, the prevailing mechanism and hence the activation energy depend on the surface-to-volume ratio, or the specific surface area, which changes with the particle size. We explain the observed variations of activation energy, and address the role played by LiH in the dehydrogenation of (LiNH_2+LiH) mixtures. The relationship between the structure of hydrogen-related defects and the end products in the decomposition reaction can be extended to other complex hydrides.

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