First-principles calculations of mass transport in magnesium borohydride CHAO YU, VIDVUDS OZOLINS, Department of Materials Science and Engineering, UCLA — Mg(BH$_4$)$_2$ is a hydrogen storage material which can decompose to release hydrogen in the following reaction: Mg(BH$_4$)$_2$(solid) → $\frac{1}{6}$MgB$_{12}$H$_{12}$(solid) + $\frac{5}{6}$MgH$_2$(solid) + $\frac{13}{6}$H$_2$(gas) → MgH$_2$(solid) + 2B(solid) + 4H$_2$(gas). However, experiments show that hydrogen release only occurs at temperatures above 300 °C, which severely limits applications in mobile storage. Using density-functional theory calculations, we systematically study bulk diffusion of defects in the reactant Mg(BH$_4$)$_2$ and products MgB$_{12}$H$_{12}$ and MgH$_2$ during the first step of the solid-state dehydrogenation reaction. The defect concentrations and concentration gradients are calculated for a variety of defects, including charged vacancies and interstitials. We find that neutral [BH$_3$] vacancies have the highest bulk concentration and concentration gradient in Mg(BH$_4$)$_2$. The diffusion mechanism of [BH$_3$] vacancy in Mg(BH$_4$)$_2$ is studied using the nudged elastic band method. Our results shows that the calculated diffusion barrier for [BH$_3$] vacancies is $\approx$ 0.2 eV, suggesting that slow mass transport limits the kinetics of hydrogen desorption.