Supporting Information

Reversibility and Improved Hydrogen Release of Magnesium Borohydride

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Figure S1. pXRD patterns (a and b), $^{11}$B MAS NMR spectra (c and d) of $\alpha$-Mg(BH$_4$)$_2$ ball-milled for 30 minutes and $\alpha$-Mg(BH$_4$)$_2$ ball-milled for 30 minutes with 5 mol % TiF$_3$ and ScCl$_3$ added, and $^{45}$Sc MAS NMR spectrum (e) for the sample with additives. For pXRD, as-made $\alpha$-Mg(BH$_4$)$_2$ ( ), as-made $\beta$-Mg(BH$_4$)$_2$ ( ) and TiF$_3$ peaks ( ) are shown for comparison. For NMR, the spectrum of as-synthesized pure Mg(BH$_4$)$_2$ sample is included. The spectral range between -30 and 30 ppm was scaled up by 15 times to show the presence of boron oxide contaminants (peaks between -5 to 20 ppm) and boron species formed after the ball milling in the presence of additives (c). The position shift of the major Mg(BH$_4$)$_2$ peak from the $\alpha$-phase to the $\beta$-phase is presented in the extended view (d). Spinning sidebands are marked with an asterisk (*).

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Figure S2. Raman spectra for hydrated MgB₁₂H₁₂·6H₂O (—) and desolvated α-Mg(BH₄)₂ (—).
Figure S3. NMR spectra measured for samples with and without additives after desorption reactions. a) $^1$H MAS NMR spectra after desorption at low temperature (300 °C). b) $^{11}$B CPMAS NMR and MAS (Bloch decay) NMR spectra of sample with additives after desorption at low temperature (300 °C). Broad peak at 40-80 ppm range is a spinning sideband from the major peak in the upfield. c) $^{11}$B CPMAS NMR spectra of both samples after desorption reaction at 600 °C. d) $^{19}$F MAS NMR spectra before and after desorption reactions showing the formation of MgF$_2$. 
Figure S4. MgB$_2$ with and without additives before the high pressure experiment (a) and (c), respectively, and after exposure to 900 bar H$_2$ at 390 °C for 72 hours ((b) and (d), respectively).