

# Supporting Information

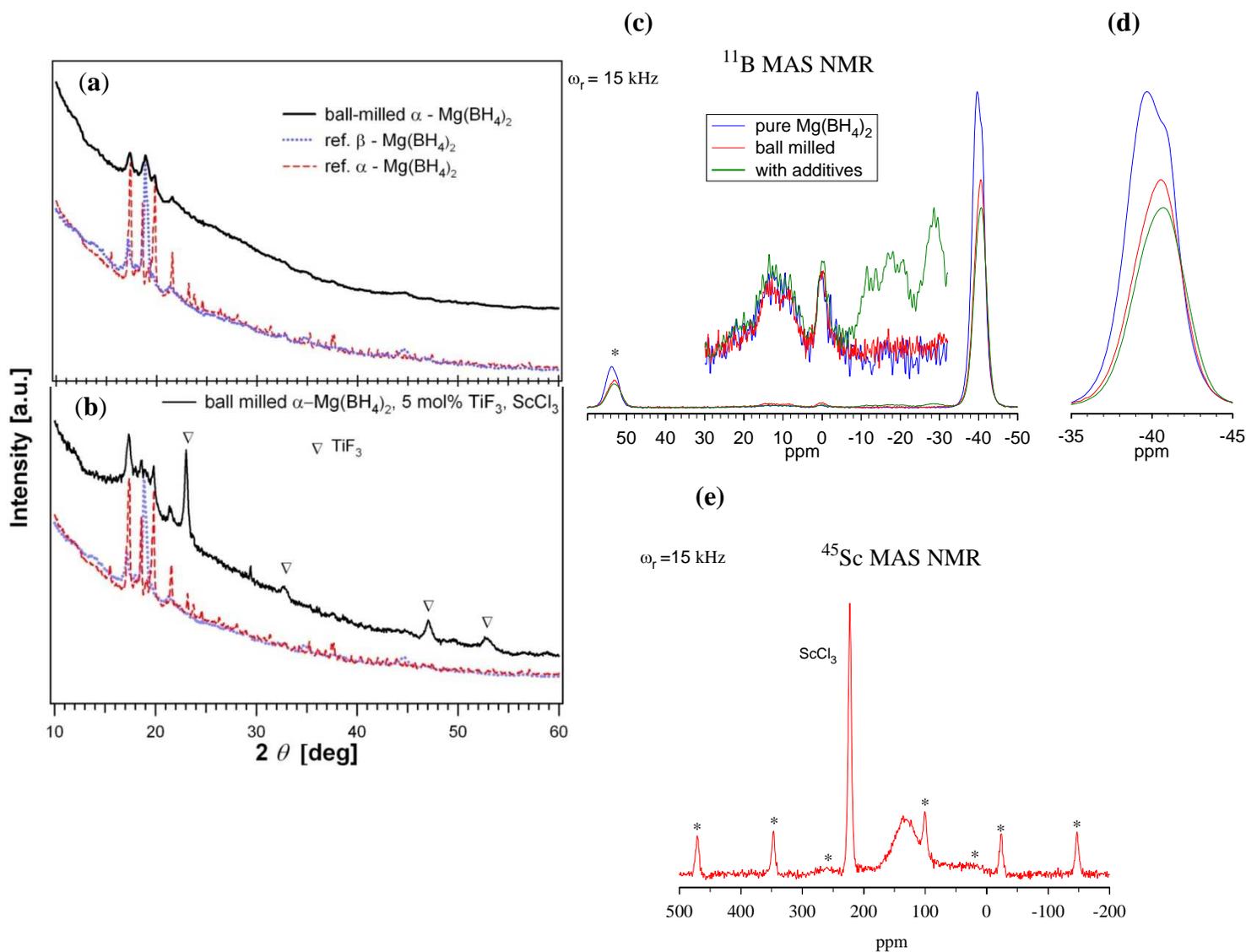
## **Reversibility and Improved Hydrogen Release of Magnesium Borohydride**

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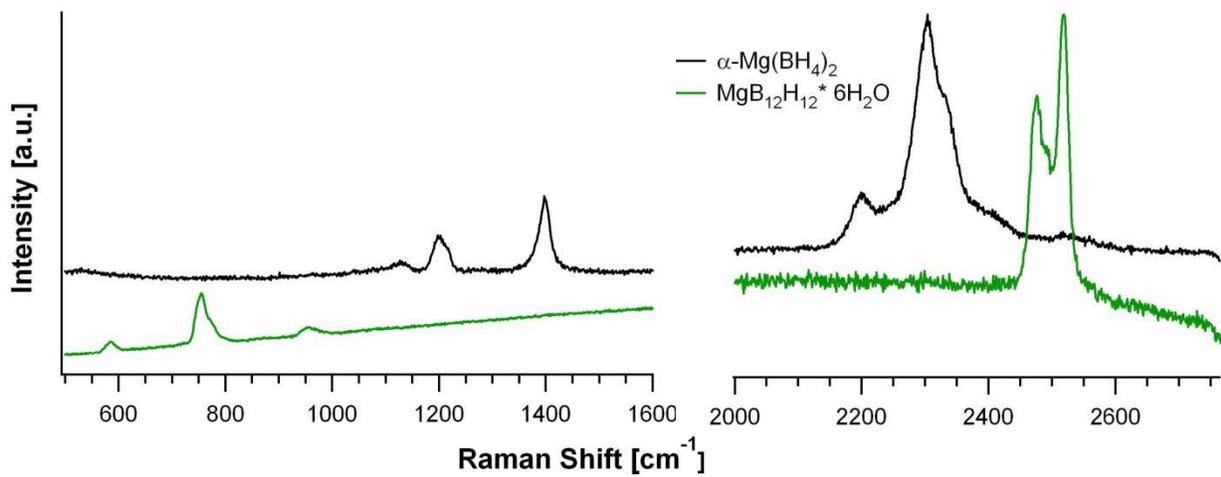
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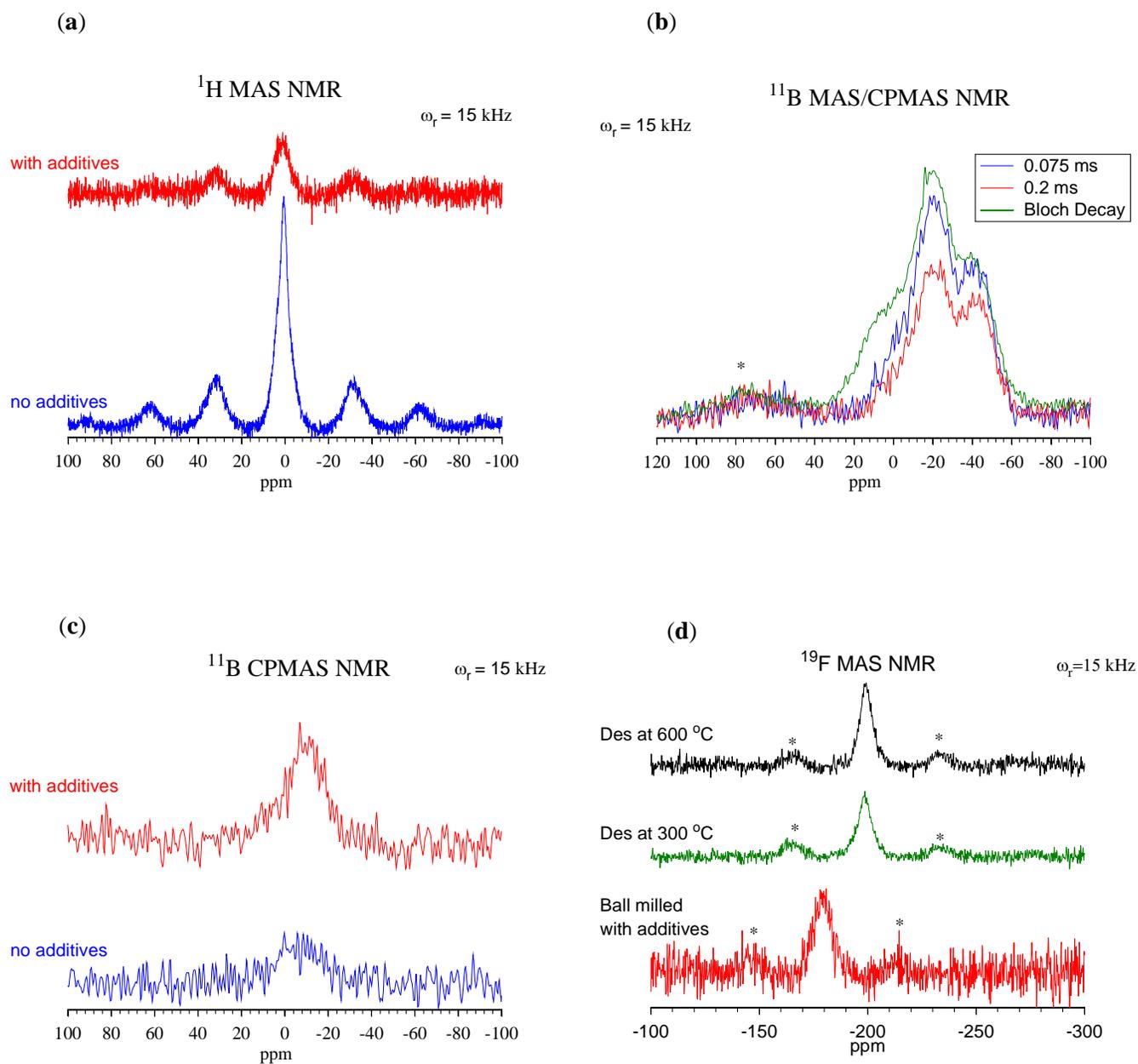


**Figure S1.** pXRD patterns (a and b),  $^{11}\text{B}$  MAS NMR spectra (c and d) of  $\alpha\text{-Mg}(\text{BH}_4)_2$  ball-milled for 30 minutes and  $\alpha\text{-Mg}(\text{BH}_4)_2$  ball-milled for 30 minutes with 5 mol %  $\text{TiF}_3$  and  $\text{ScCl}_3$  added, and  $^{45}\text{Sc}$  MAS NMR spectrum (e) for the sample with additives. For pXRD, as-made  $\alpha\text{-Mg}(\text{BH}_4)_2$  (---), as-made  $\beta\text{-Mg}(\text{BH}_4)_2$  (....) and  $\text{TiF}_3$  peaks ( ▽ ) are shown for comparison. For NMR, the spectrum of as-synthesized pure  $\text{Mg}(\text{BH}_4)_2$  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  $\text{Mg}(\text{BH}_4)_2$  peak from the  $\alpha$ -phase to the  $\beta$ -phase is presented in the extended view (d). Spinning sidebands are marked with an asterisk (\*).

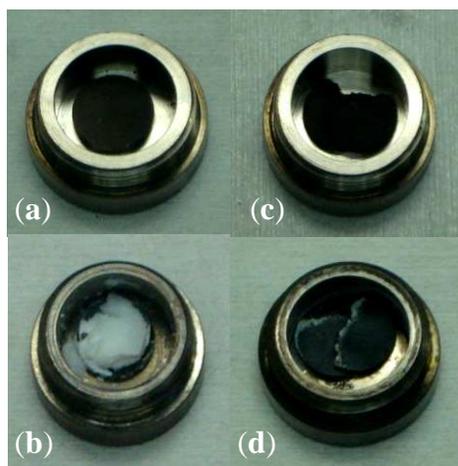
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**Figure S2.** Raman spectra for hydrated  $\text{MgB}_{12}\text{H}_{12} \cdot 6\text{H}_2\text{O}$  (—) and desolvated  $\alpha\text{-Mg(BH}_4)_2$  (—).



**Figure S3.** NMR spectra measured for samples with and without additives after desorption reactions. a)  $^1\text{H}$  MAS NMR spectra after desorption at low temperature ( $300\text{ }^\circ\text{C}$ ). b)  $^{11}\text{B}$  CPMAS NMR and MAS (Bloch decay) NMR spectra of sample with additives after desorption at low temperature ( $300\text{ }^\circ\text{C}$ ). Broad peak at 40-80 ppm range is a spinning sideband from the major peak in the upfield. c)  $^{11}\text{B}$  CPMAS NMR spectra of both samples after desorption reaction at  $600\text{ }^\circ\text{C}$ . d)  $^{19}\text{F}$  MAS NMR spectra before and after desorption reactions showing the formation of  $\text{MgF}_2$ .



**Figure S4.**  $\text{MgB}_2$  with and without additives before the high pressure experiment (**a**) and (**c**), respectively, and after exposure to 900 bar  $\text{H}_2$  at 390 °C for 72 hours ((**b**) and (**d**), respectively).