

# Supporting Information

## **Synthesis and hydrogen storage properties of Mg-based complex hydrides with multiple transition metal elements**

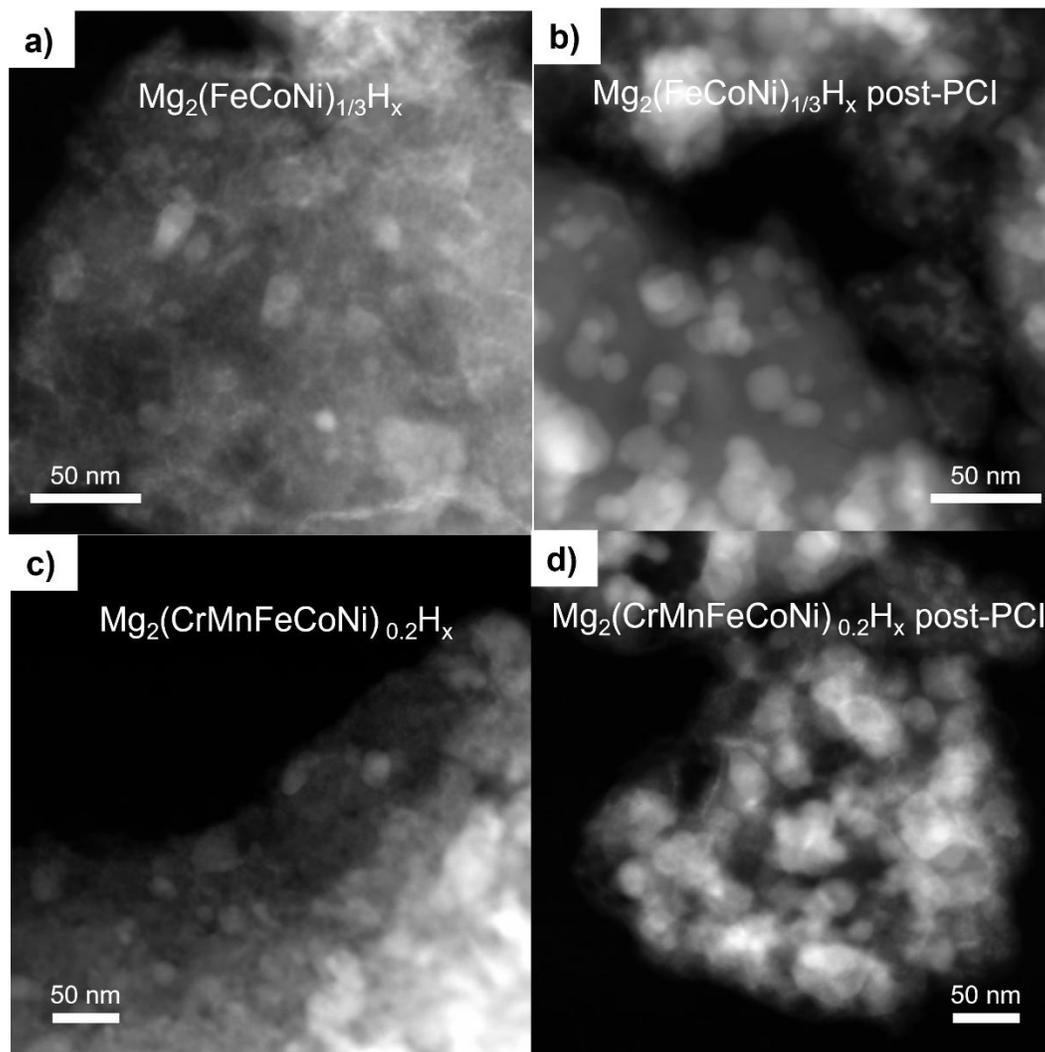
*Evans Pericoli,<sup>1</sup> Alessia Barzotti,<sup>1</sup> Raffaello Mazzaro<sup>1,2</sup>, Romain Moury,<sup>3</sup> Fermin Cuevas<sup>3</sup> and Luca Pasquini<sup>\*1,2</sup>*

<sup>1</sup> Department of Physics and Astronomy “Augusto Righi”, University of Bologna, Bologna, 40127, Italy

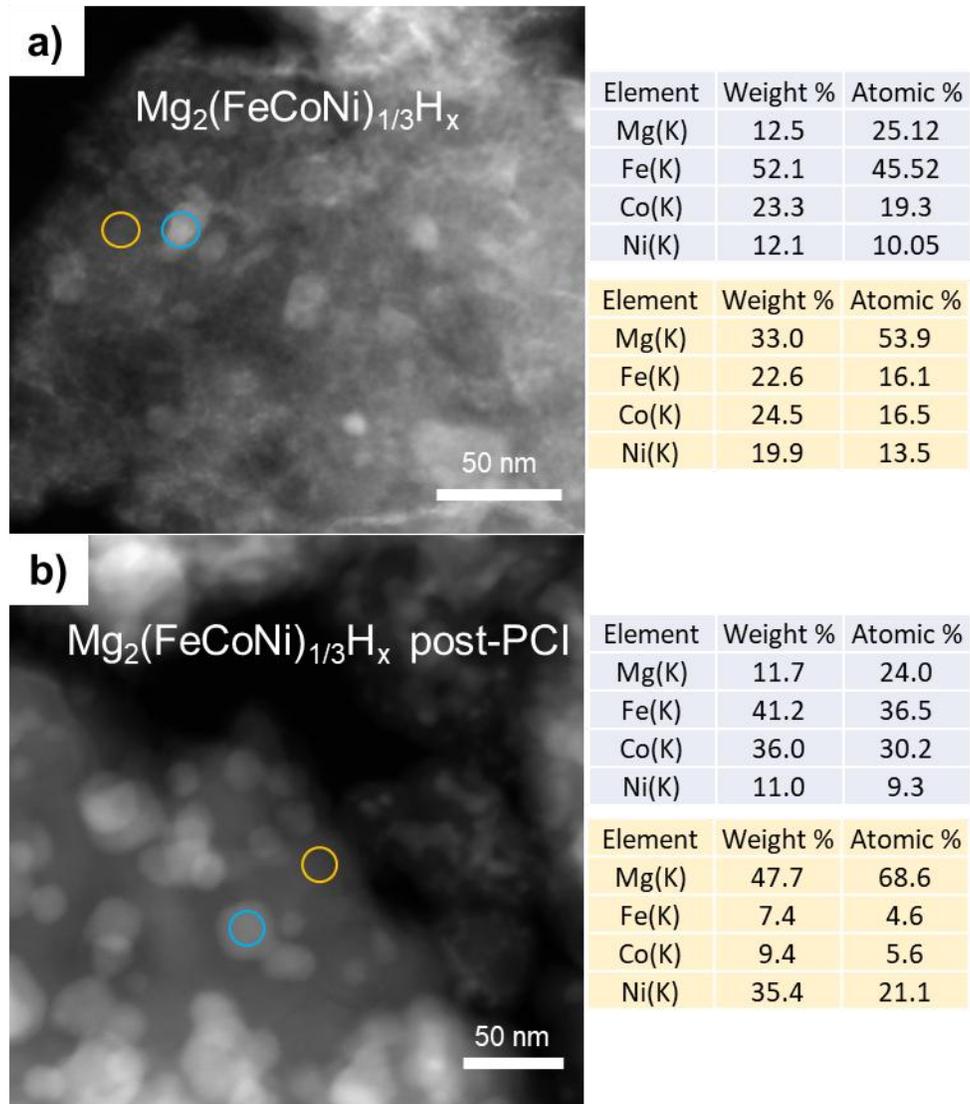
<sup>2</sup> Istituto per la Microelettronica e Microsistemi (IMM), Consiglio Nazionale delle Ricerche (CNR), Bologna, 40129, Italy

<sup>3</sup> University Paris-Est Creteil, Centre National de la Recherche Scientifique (CNRS), Institut de Chimie et des Matériaux Paris-Est (ICMPE), Thiais, 94320, France

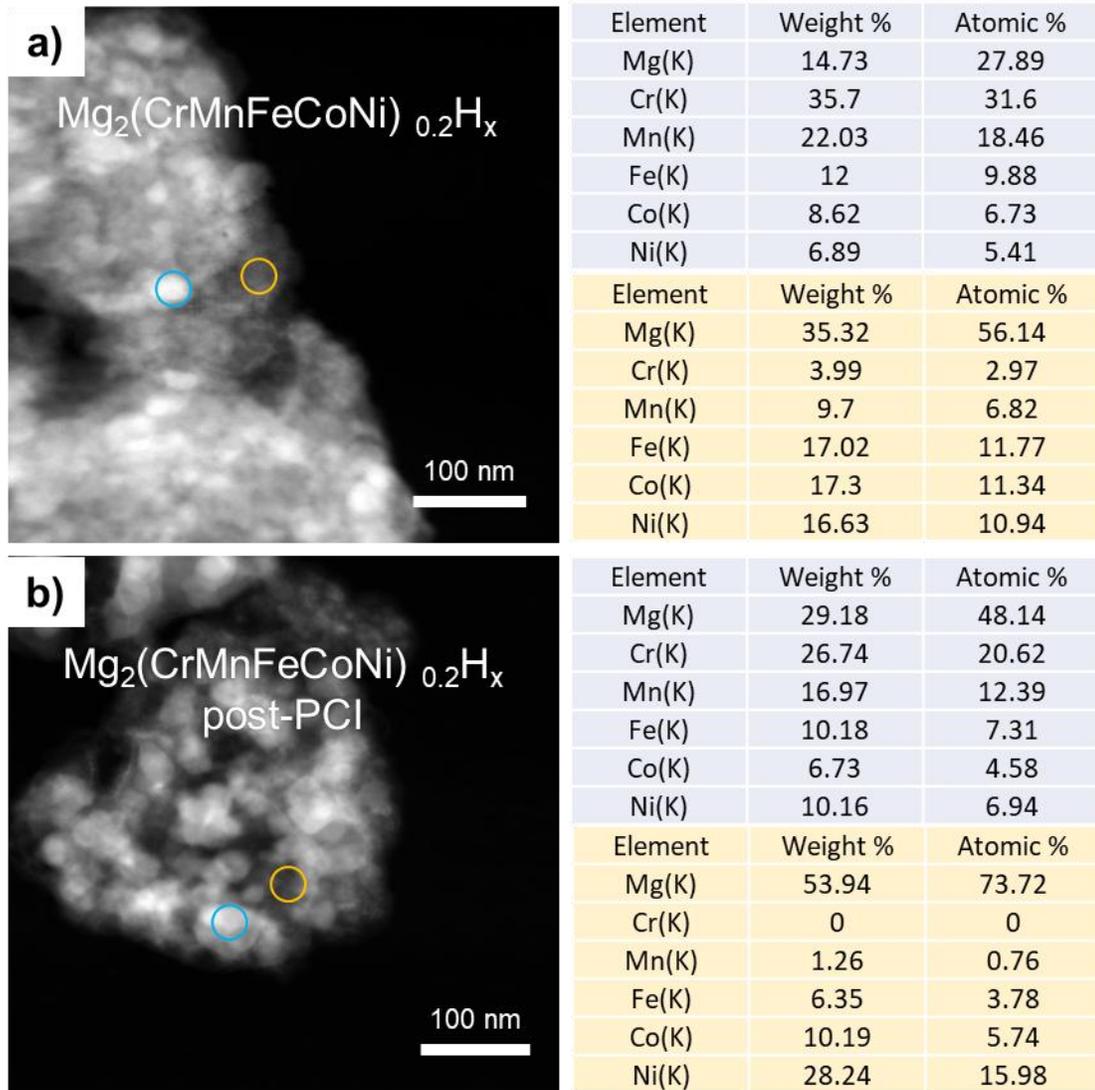
E-mail: [luca.pasquini@unibo.it](mailto:luca.pasquini@unibo.it)



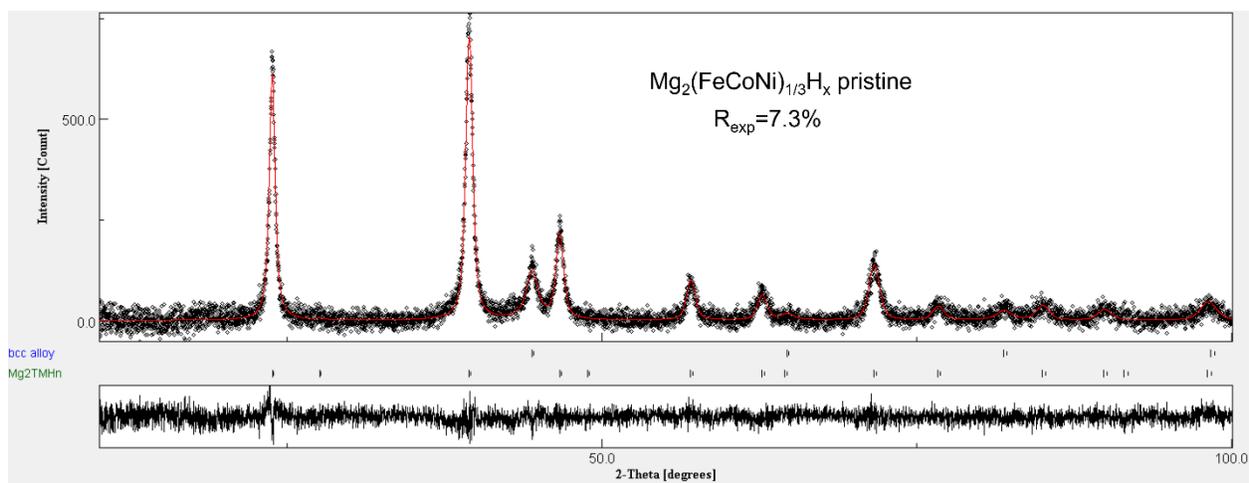
**Figure S1:** Dark field HAADF-STEM images of  $\text{Mg}_2(\text{FeCoNi})_{1/3}\text{H}_x$  and  $\text{Mg}_2(\text{CrMnFeCoNi})_{0.2}\text{H}_x$  in the as-milled state (**a,c**) and after the PCIs (**b,d**).



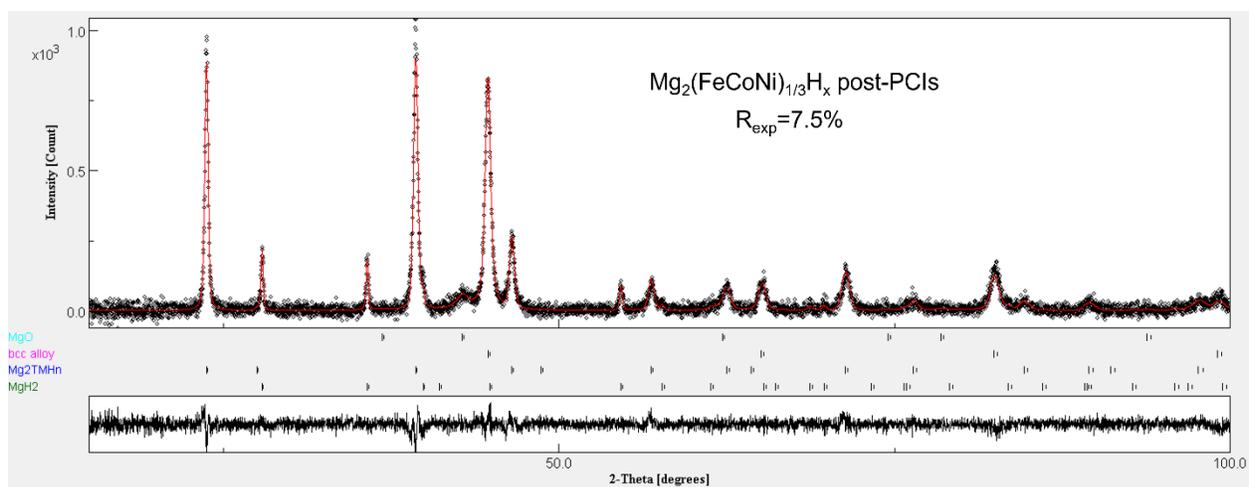
**Figure S2:** STEM-EDX quantification of  $\text{Mg}_2(\text{FeCoNi})_{1/3}\text{H}_x$  in the as-milled state (**a**) and after the PCIs (**b**).



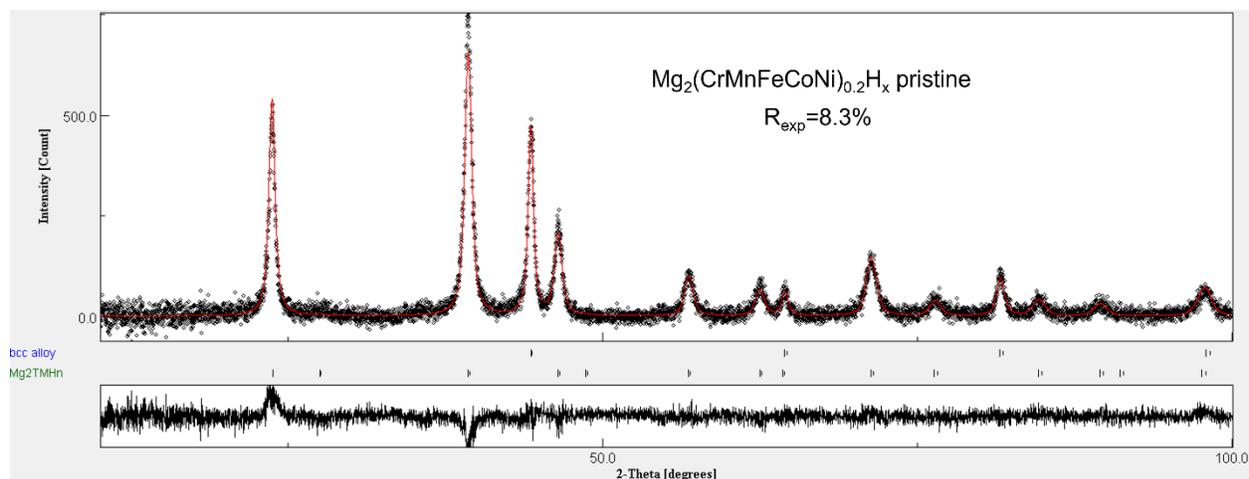
**Figure S3:** STEM-EDX quantification of  $\text{Mg}_2(\text{CrMnFeCoNi})_{0.2}\text{H}_x$  in the as-milled state (**a**) and after the PCIs (**b**).



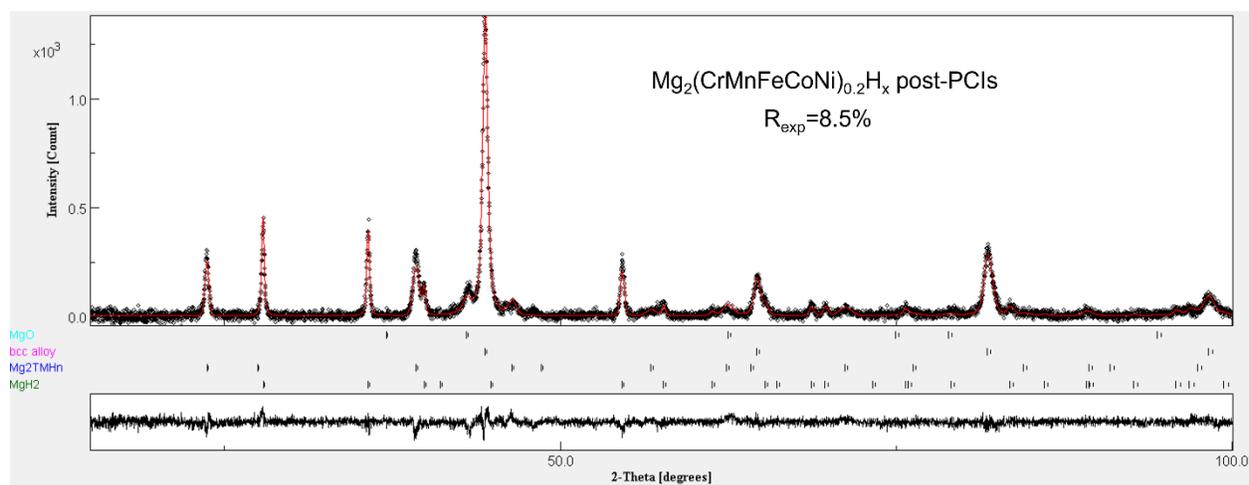
**Figure S4:** XRD profile and Rietveld refinement of the as-milled  $\text{Mg}_2(\text{FeCoNi})_{1/3}\text{H}_x$ .



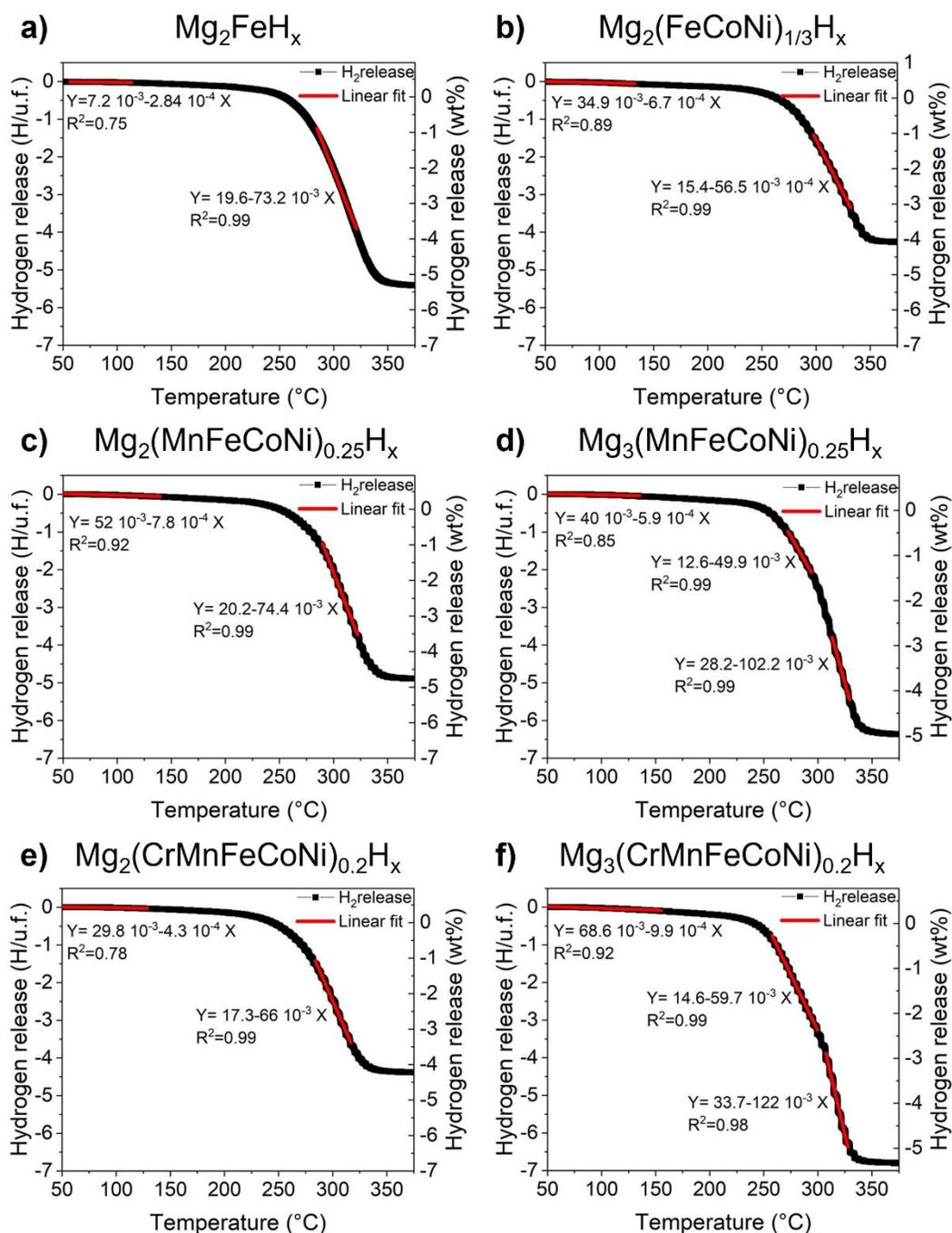
**Figure S5:** XRD profile and Rietveld refinement of  $\text{Mg}_2(\text{FeCoNi})_{1/3}\text{H}_x$  after PCI.



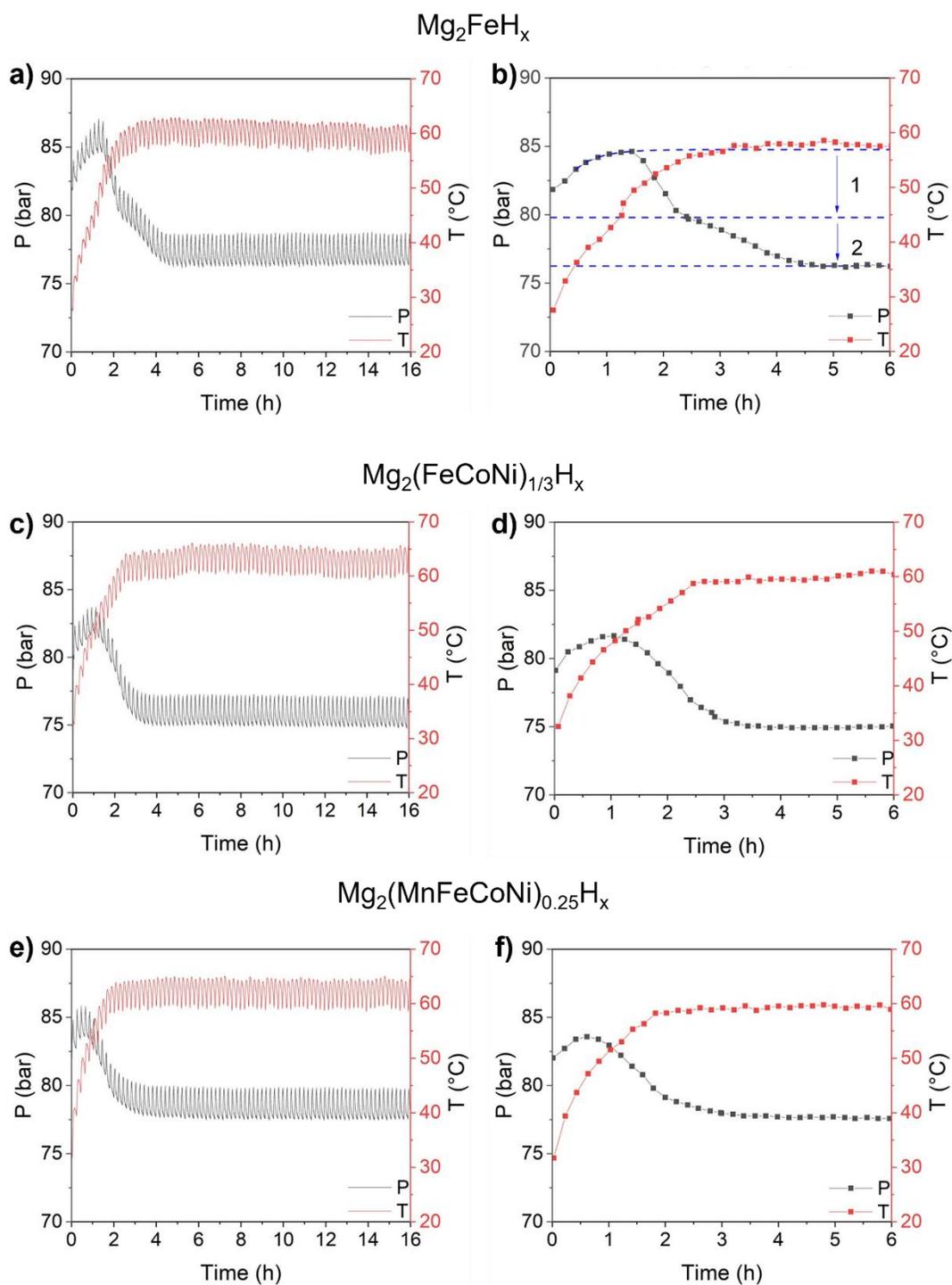
**Figure S6:** XRD profile and Rietveld refinement of the as-milled  $\text{Mg}_2(\text{CrMnFeCoNi})_{0.2}\text{H}_x$ .



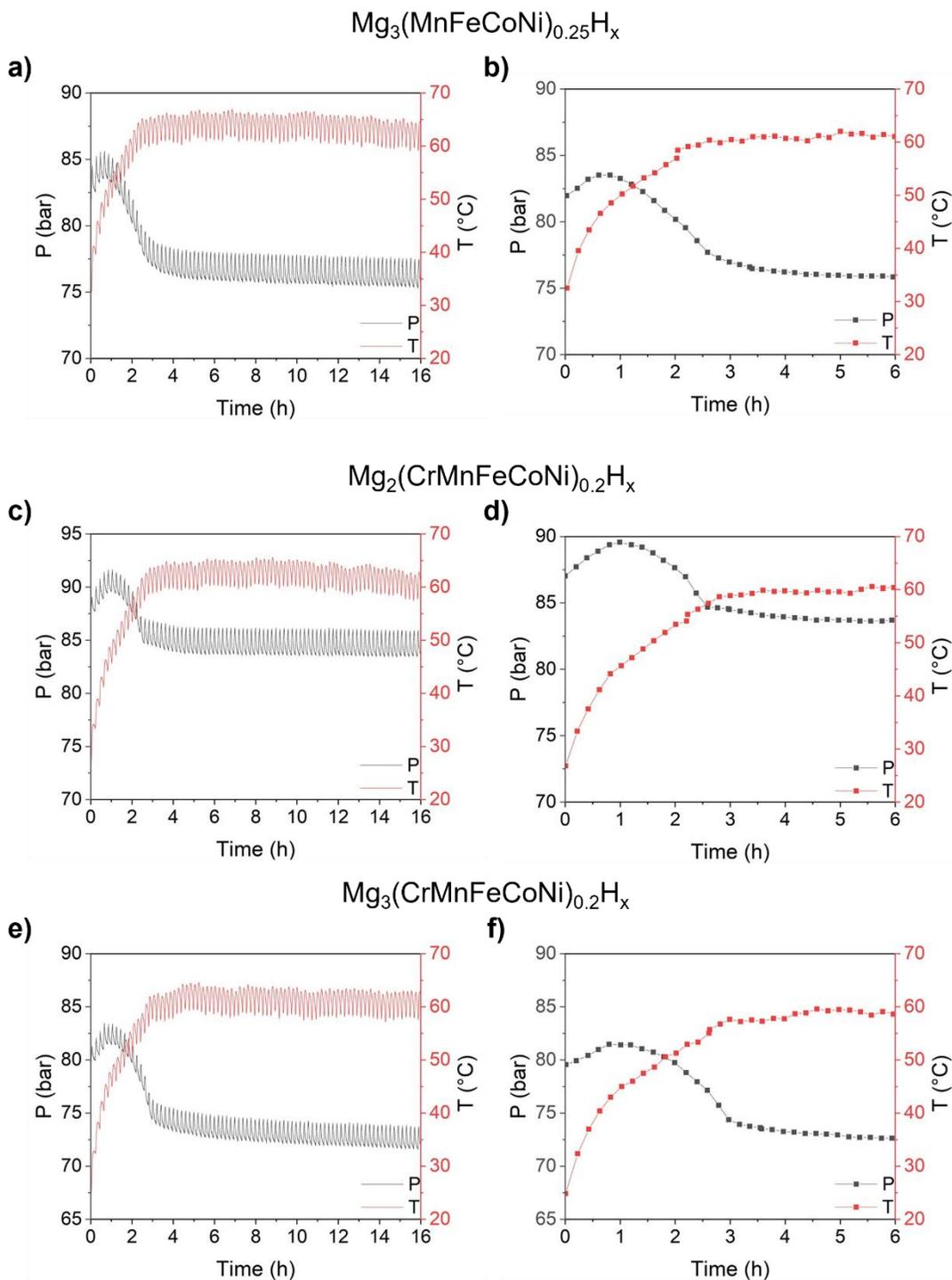
**Figure S7:** XRD profile and Rietveld refinement of  $\text{Mg}_2(\text{CrMnFeCoNi})_{0.2}\text{H}_x$  after PCIs.



**Figure S8:** Determination of the onset temperature in TPD experiments by the intersection of linear fits to the ramp start and to the linear decreasing trends that represent  $H_2$  release for  $Mg_2FeH_x$  (a),  $Mg_2(FeCoNi)_{1/3}H_x$  (b),  $Mg_2(MnFeCoNi)_{0.25}H_x$  (c),  $Mg_3(MnFeCoNi)_{0.25}H_x$  (d),  $Mg_2(CrMnFeCoNi)_{0.2}H_x$  (e),  $Mg_3(CrMnFeCoNi)_{0.2}H_x$  (f).



**Figure S9:** Pressure and temperature time-profile of the RBM synthesis process. Raw data of the full process (left) and p-T points collected at the end of each three-minutes cycle for  $\text{Mg}_2\text{FeH}_x$  (**a,b**),  $\text{Mg}_2(\text{FeCoNi})_{1/3}\text{H}_x$  (**c,d**),  $\text{Mg}_2(\text{MnFeCoNi})_{0.25}\text{H}_x$  (**e,f**). The blue arrows and dotted lines highlight the two-step process ( $\text{Mg}/\text{MgH}_2$  and  $\text{MgH}_2/\text{Mg}_2\text{TMH}_2$ ) the metallic mixture undergoes during the synthesis.



**Figure S10:** Pressure and temperature time-profile of the RBM synthesis process. Raw data of the full process (left) and p-T points collected at the end of each three-minutes cycle for  $\text{Mg}_3(\text{MnFeCoNi})_{0.25}\text{H}_x$  (**a,b**),  $\text{Mg}_2(\text{CrMnFeCoNi})_{0.2}\text{H}_x$  (**c,d**),  $\text{Mg}_3(\text{CrMnFeCoNi})_{0.2}\text{H}_x$  (**e,f**).