**Supporting information**

Belonging to the manuscript

**Thermodynamics and stability of the Mg-H-F system for thermochemical energy storage applications**


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**Fig. S1.** Ex-situ XRD data for samples ball milled for 40 hours (L) collected at room temperature. \(\lambda = 1.5418 \text{ Å}\). Red and black dot line’s refers to main peaks of MgF\(_2\) and MgH\(_2\) respectively.
Fig. S2. *In situ* SR-XRD at room temperature of Mg(HₓF₁₋ₓ)₂-L samples ball milled for 40 hours and annealed. λ = 1.000389(1) Å.

Fig. S3. Comparison between SR-XRD of samples ball milled for 10 and 40 hours and then annealed (A). ●=MgₓFeH₆, ✧=Si. λ = 1.5418 Å, at room temperature.
Fig. S4. *In situ* XRD for Mg(H$_{0.70}$F$_{0.30}$)-L performed under vacuum using a $\Delta T/\Delta t = 10^\circ$C/min before 200 $^\circ$C and 5 $^\circ$C/min after 200 $^\circ$C. $\lambda = 1.000389(1)$ Å.

Fig. S5. *In situ* XRD for Mg(H$_{0.50}$F$_{0.50}$)-L performed under vacuum using a $\Delta T/\Delta t = 10^\circ$C/min before 200 $^\circ$C and 5 $^\circ$C/min after 200 $^\circ$C. $\lambda = 1.000389(1)$ Å.
**Fig. S6.** *In situ* XRD for Mg(H$_{0.95}$F$_{0.05}$)-L performed under vacuum using a $\Delta T/\Delta t = 10^\circ$C/min before 200 °C and 5 °C/min after 200 °C. $\lambda = 1.000389(1)$ Å.

**Table S1.** Summary of parameters and data collected from PCT desorption measurements of Mg(H$_{0.85}$F$_{0.15}$)-S Pressure and H$_2$ wt% uncertainties

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Observed Desorption H$_2$ (wt%)</th>
<th>Final Pressure (bar)</th>
<th>Theoretical H$_2$ wt%</th>
<th>Difference between observed vs theoretical H$_2$ wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>438</td>
<td>4.57 ± 0.14</td>
<td>14.5</td>
<td>5.4</td>
<td>0.83</td>
</tr>
<tr>
<td>444</td>
<td>4.37 ± 0.29</td>
<td>24.1</td>
<td>5.4</td>
<td>1.03</td>
</tr>
<tr>
<td>450</td>
<td>4.85 ± 0.19</td>
<td>4</td>
<td>5.4</td>
<td>0.55</td>
</tr>
<tr>
<td>461</td>
<td>4.61 ± 0.15</td>
<td>19.8</td>
<td>5.4</td>
<td>0.79</td>
</tr>
</tbody>
</table>
Fig. S7. Simultaneous thermal analysis of Mg(H\textsubscript{x}F\textsubscript{1-x})\textsubscript{2}-L samples by (a) DSC, (b) TGA and (c) MS. $\Delta T/\Delta t = 10 ^\circ $C/min. DSC and MS data are normalised to the mass of the sample.
Fig. S8. Simultaneous thermal analysis of Mg(H_{0.50}F_{0.50})-L by (a) MS and (b) TGA. $\Delta T/\Delta t = 10 ^\circ C/min$.

Fig. S9. In situ XRD for Mg(H_{0.50}F_{0.50})-L. $\lambda = 0.774541(1)$ Å.