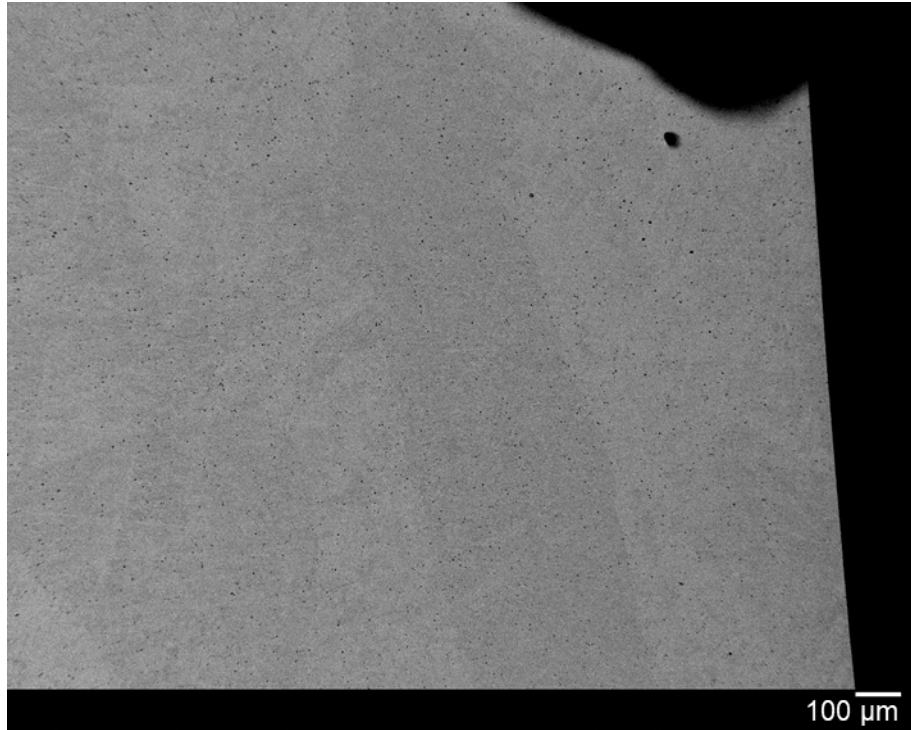


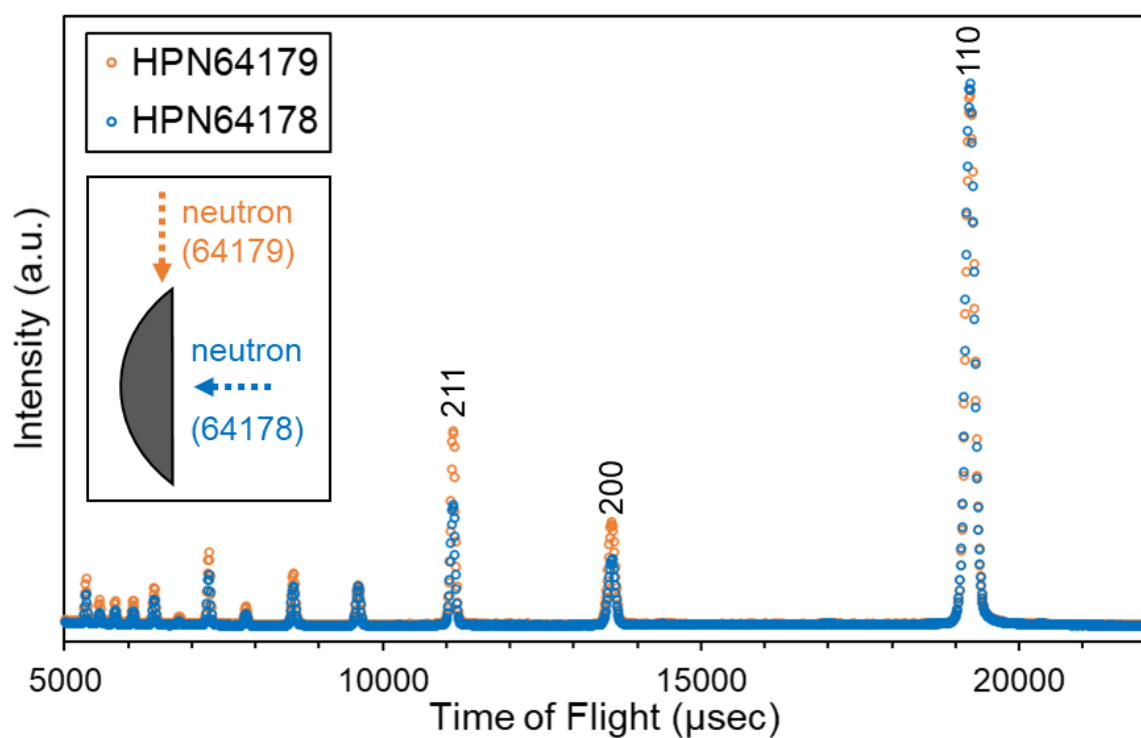
## Supplementary information

**Table S1.** Unit cell volumes of *fcc* Fe<sub>0.9</sub>Ni<sub>0.1</sub> obtained using XRD at various *P-T* conditions.

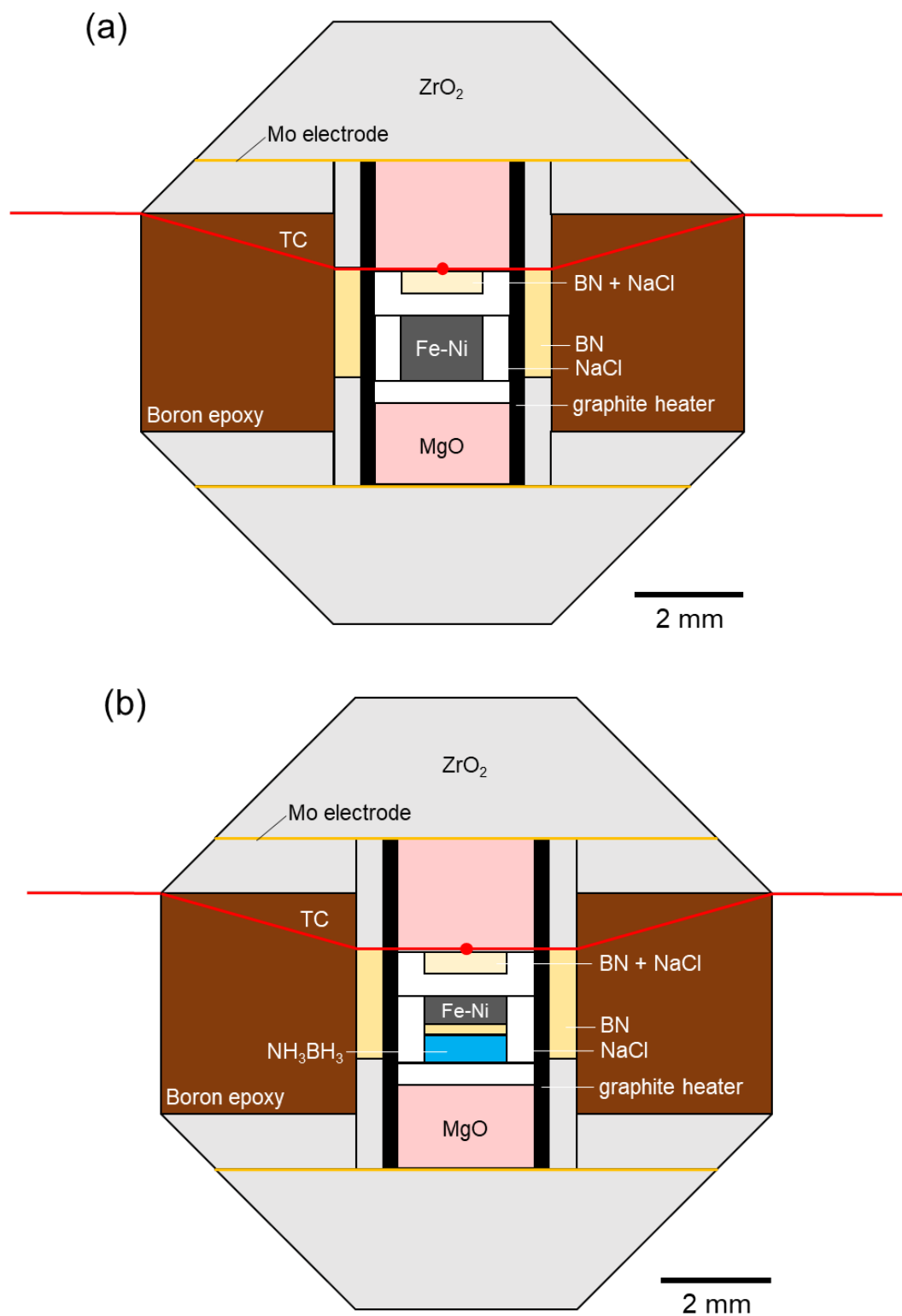
<i>P</i> (GPa)	<i>T</i> (K)	<i>V</i> (Å <sup>3</sup> )
3.13	900	46.29(2)
4.83	850	45.71(7)
3.23	800	46.20(5)
6.32	800	45.16(5)
7.91	800	44.67(6)
3.35	700	45.70(11)
6.12	700	44.95(4)
7.63	700	44.44(6)
3.16	600	45.54(9)
5.93	600	44.73(4)
7.47	600	44.21(7)
5.74	500	44.52(5)
7.36	500	44.01(8)
5.64	450	44.42(6)
5.56	400	44.32(5)
7.12	400	43.85(9)
7.04	300	43.65(6)



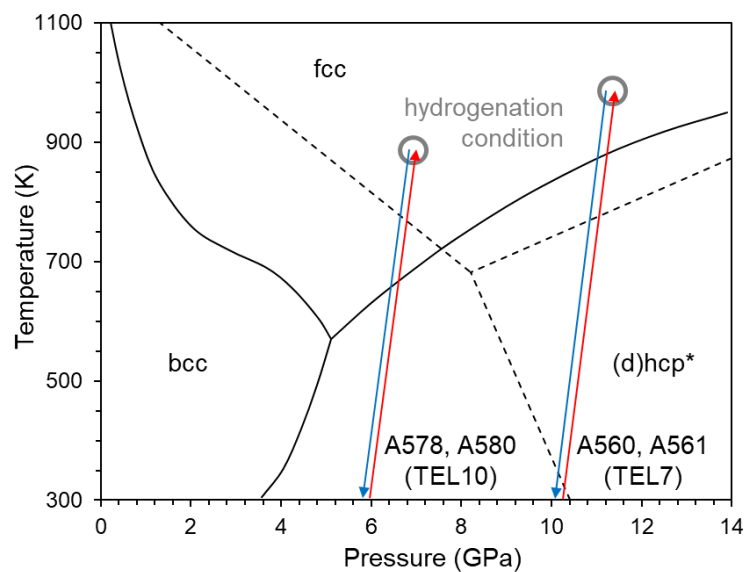
**Figure S1.** BSE image of synthesized Fe<sub>0.9</sub>Ni<sub>0.1</sub> alloy. The tiny black spots are metal oxides.



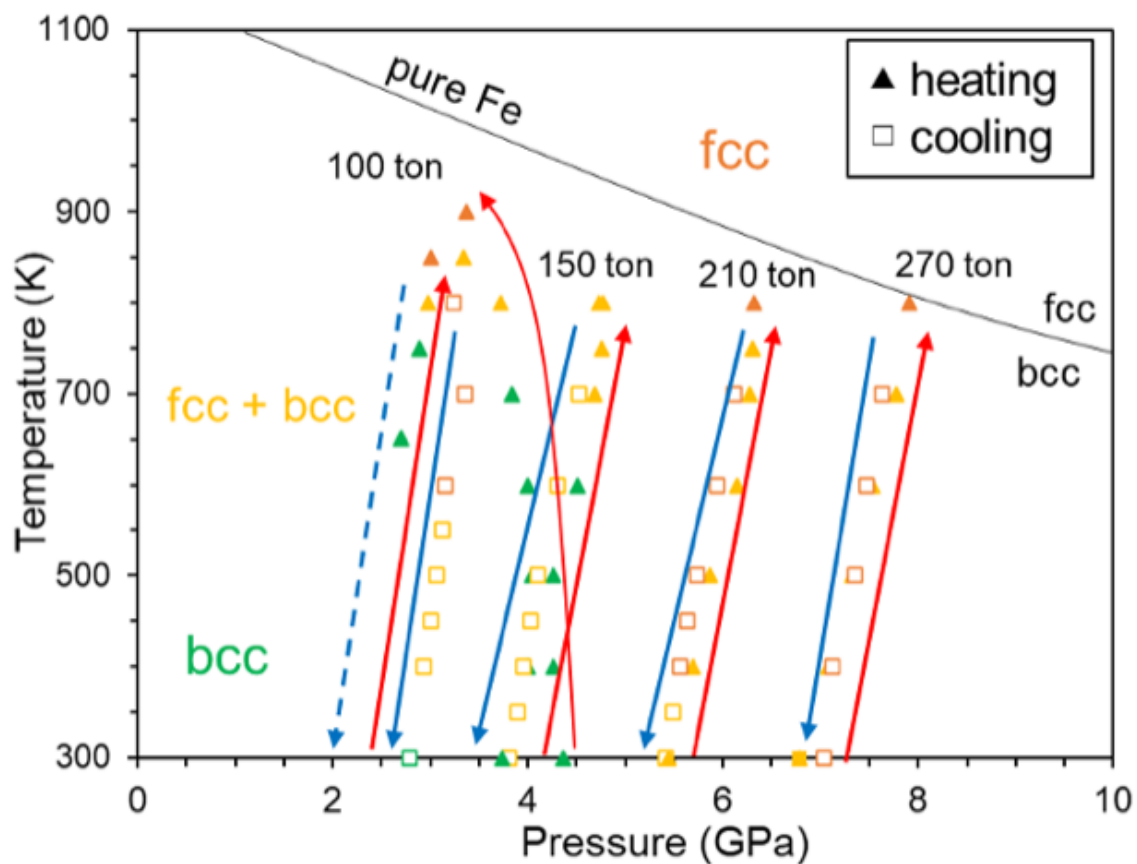
**Figure S2.** Neutron diffraction profiles of Fe<sub>0.9</sub>Ni<sub>0.1</sub> alloy taken with different geometries. Both profiles are normalized by the height of 110 peak. The inset shows directions of incident neutron beam to the sample in the two measurements (HPN64178, and HPN64179). The irradiation area of the incident neutron beam was larger than the sample size (3 mm × 1.5 mm × 5 mm).



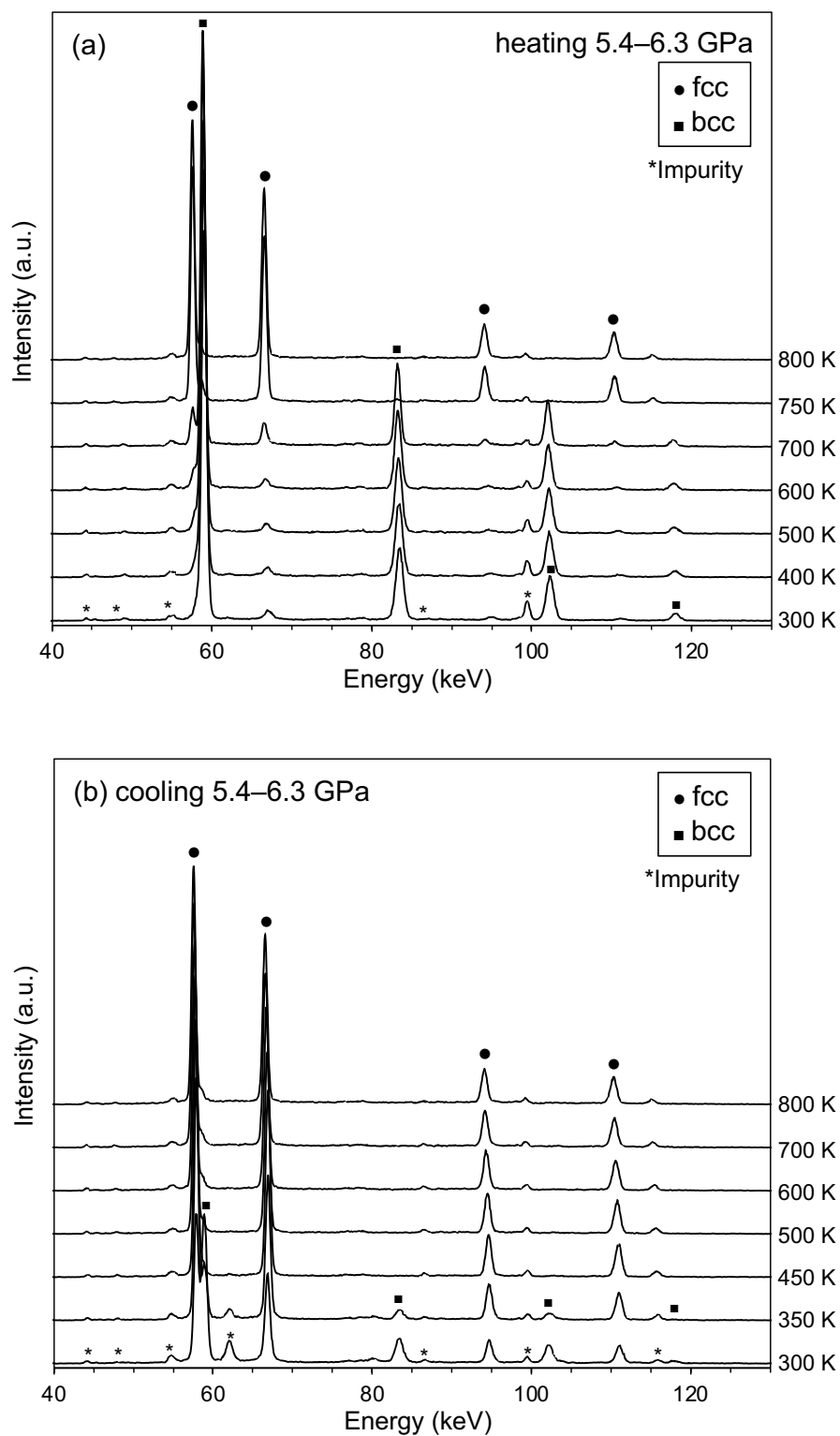
**Figure S3.** Cell assemblies for XRD experiments of (a)  $\text{Fe}_{0.9}\text{Ni}_{0.1}$  and (b)  $\text{Fe}_{0.9}\text{Ni}_{0.1}\text{H}_x$ .



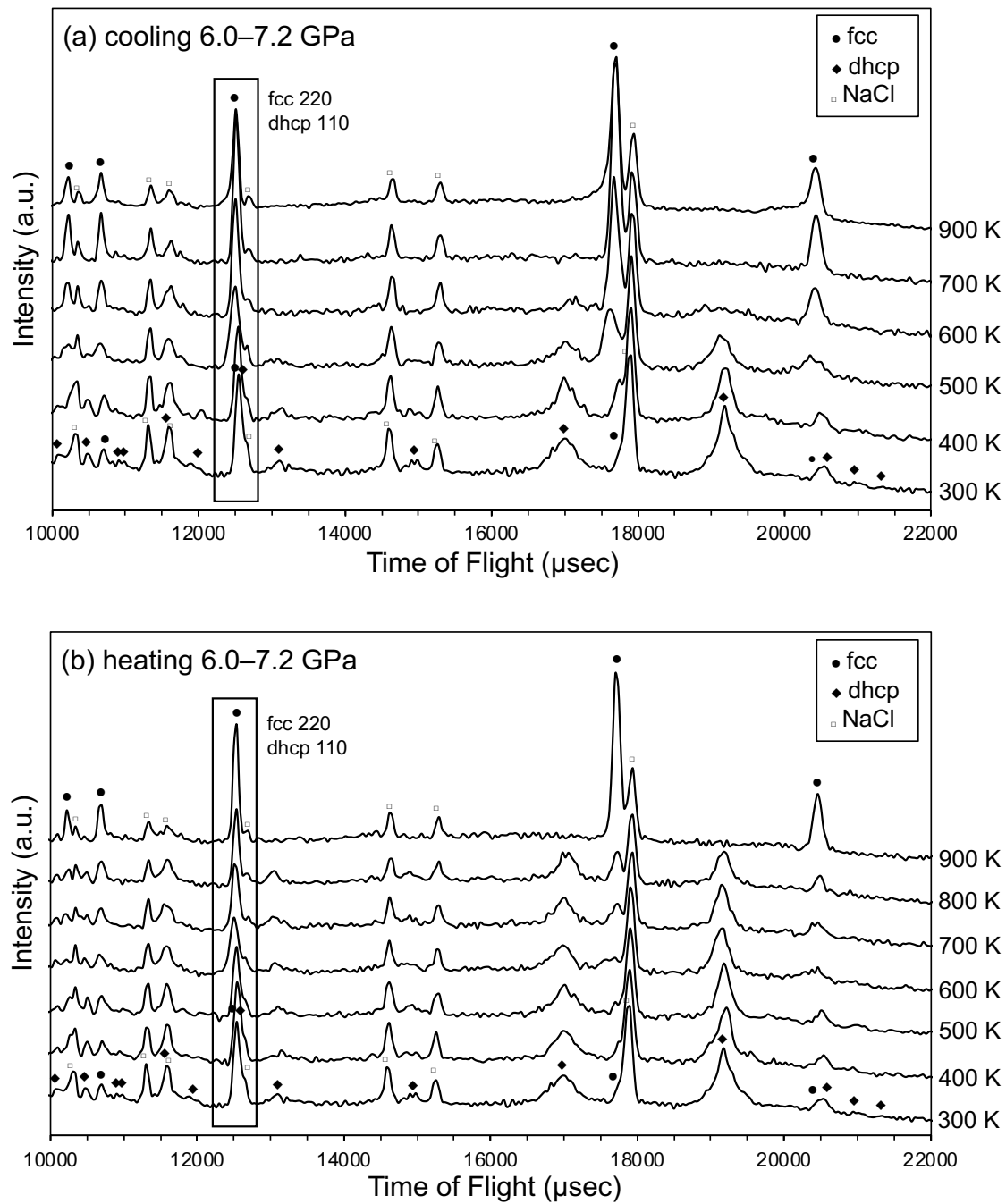
**Figure S4.** *P-T* conditions of neutron diffraction measurements of  $\text{Fe}_{0.9}\text{Ni}_{0.1}\text{D}_x$ .  $\text{Fe}_{0.9}\text{Ni}_{0.1}$  alloys were kept at the hydrogenation conditions (gray circles) for about one hour to make the amount of hydrogen uniform in the entire sample. Solid lines and dashed lines indicate phase boundaries of  $\text{FeH}_x$  (Fukai et al. 2003) and Fe (Bundy 1965), respectively. The hexagonal phase shown by asterisk is double hexagonal close-packed (*dhcp*) and *hcp* structures for Fe–H system and pure Fe, respectively.



**Figure S5.** *P-T* conditions of XRD measurements and observed phases of  $\text{Fe}_{0.9}\text{Ni}_{0.1}$ . The red filled and opened plots indicate the *P-T* conditions measured in heating and cooling processes, respectively. Orange, yellow, and green symbols indicate the *PT* conditions where *fcc*, *fcc + bcc* and *bcc* phases were observed, respectively. Phase boundary between *fcc* and *bcc* of pure Fe (Bundy, 1965) is shown by black line.

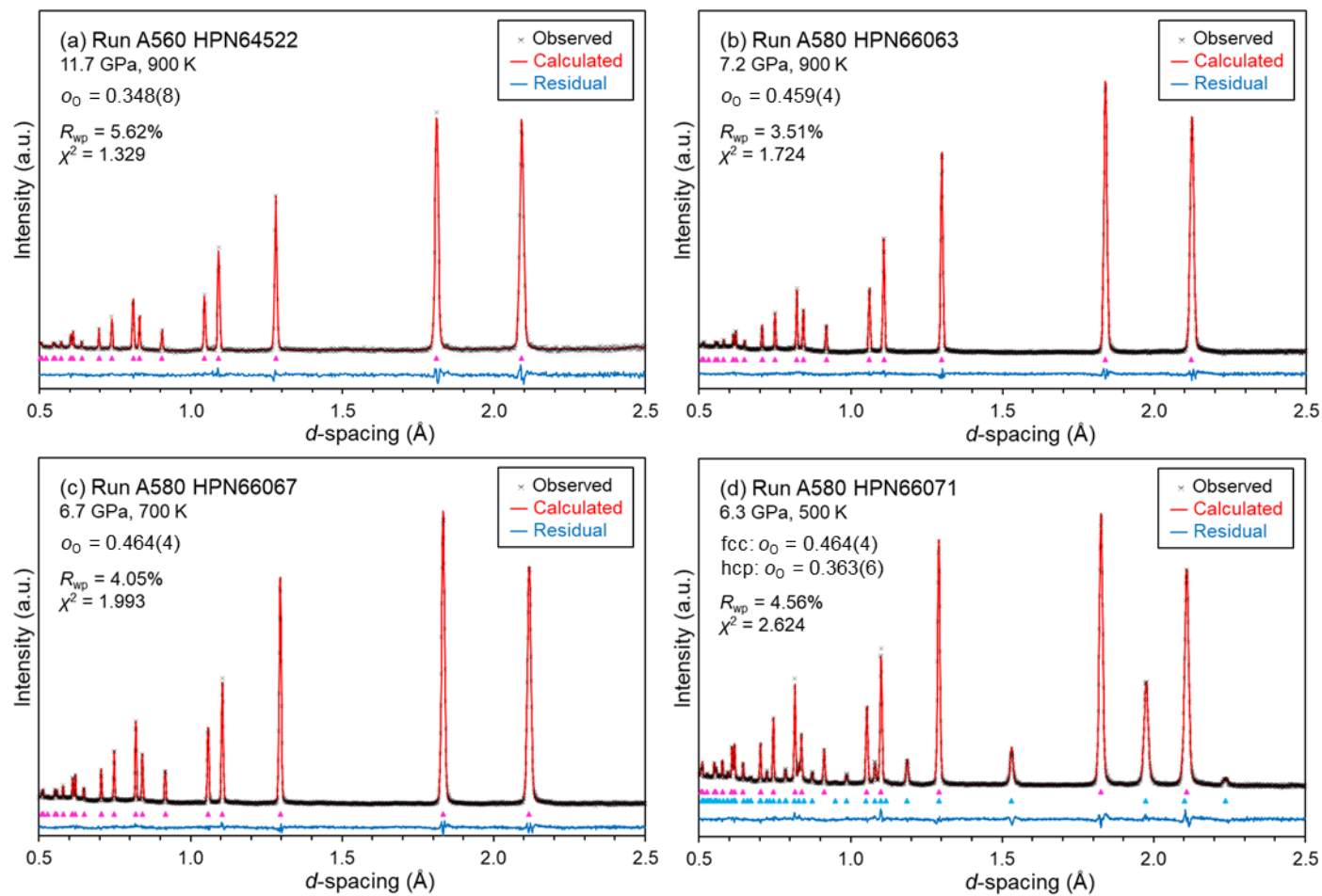


**Figure S6.** XRD profiles of  $\text{Fe}_{0.9}\text{Ni}_{0.1}$  obtained during (a) heating and (b) cooling at 5.4–6.3 GPa. The impurity peaks have not been identified.



**Figure S7.** Neutron diffraction profiles (raw data) obtained during (a) cooling and (b) heating processes in Run A561. Each profile was obtained for about 15 minutes. The peak framed by a rectangle (*dhcp* 110) is sharper than the other peaks from *dhcp* phase.





**Figure S8.** (a)–(d) Neutron diffraction profiles after intensity correction and the results of the Rietveld fitting. Pink and blue triangles indicate the expected positions of the peaks of *fcc* and *hcp*  $\text{Fe}_{0.9}\text{Ni}_{0.1}\text{D}_x$ , respectively.