

Supporting Information for

Cu-substituted NiF₂ as Cathode Material for Li-Ion Batteries

Cesar Villa^{†,◇}, Sungkyu Kim^{‡,¶,◇}, Yixue Lu[†], Vinayak P. Dravid^{,†}, Jinsong Wu^{*,†,‡}*

[†] Department of Materials Science and Engineering, NUANCE Center, Northwestern University, Evanston, Illinois 60208, United States

[‡] Nanostructure Research Centre, State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070, China

[¶] Mechanical Engineering and Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, United States

*E-mail: wujs@whut.edu.cn; v-dravid@northwestern.edu

◇These authors contributed equally.

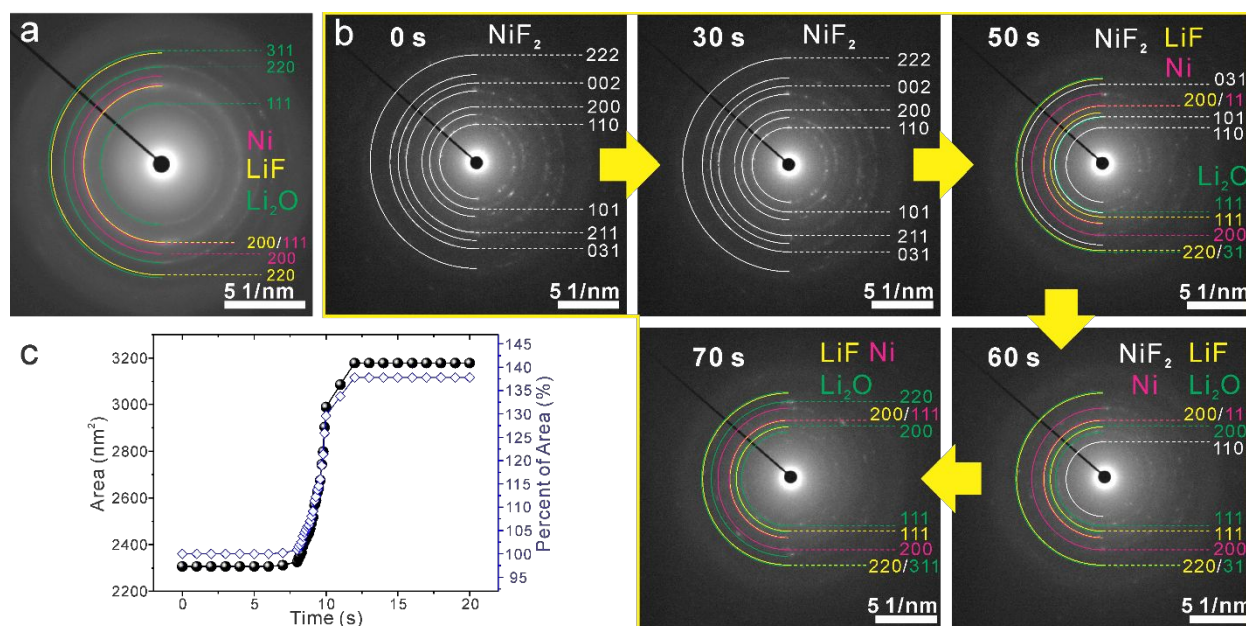


Figure S1. (a) SAED patterns of NiF₂ nanoparticles after lithiation process. This SAED patterns obtained from NiF₂ nanoparticles in Figure 4a. NiF₂ nanoparticles are changed to LiF and Ni phases by conversion reaction. (b) Time-lapse SAED patterns of NiF₂ nanoparticles during lithiation process. NiF₂ phase is gradually reduced, and Ni and LiF phases are formed by the inserted Li ions. Intensity profiles in Figure 4d is plotted based on (b). (c) Area changes of NiF₂ particles obtained from Figure 4a.

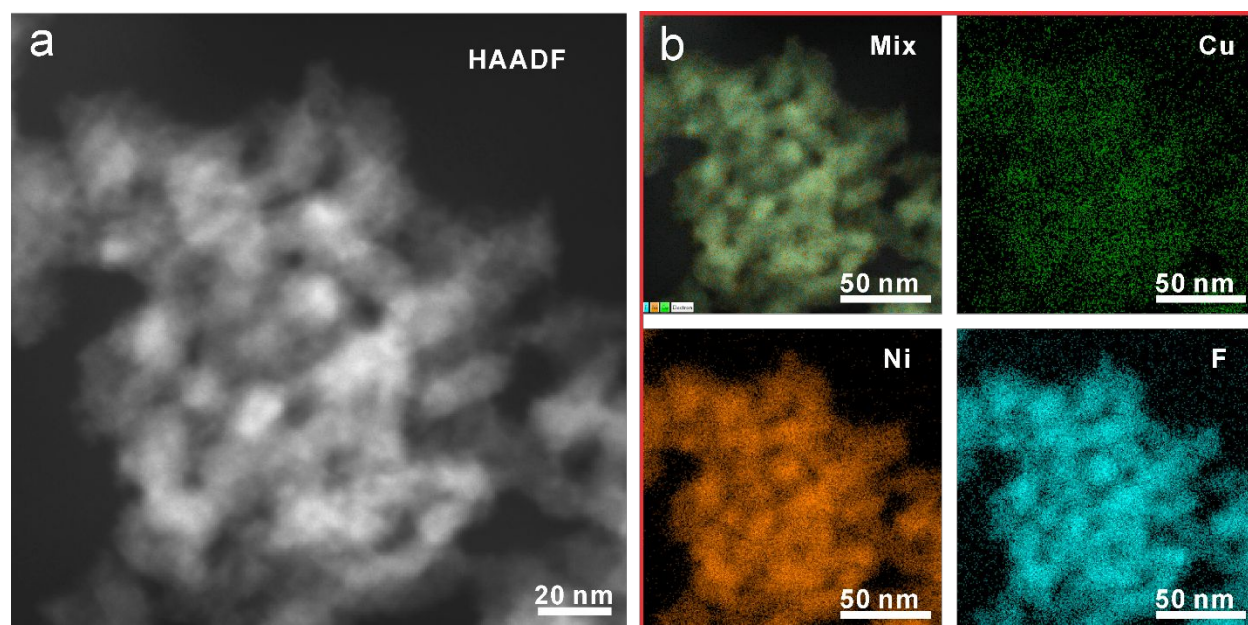


Figure S2. (a) HAADF-STEM image of $\text{Cu}_{0.1}\text{Ni}_{0.9}\text{F}_2$ nanoparticles at the pristine state. (b) EDS map images obtained from (a). All elements are uniformly distributed.

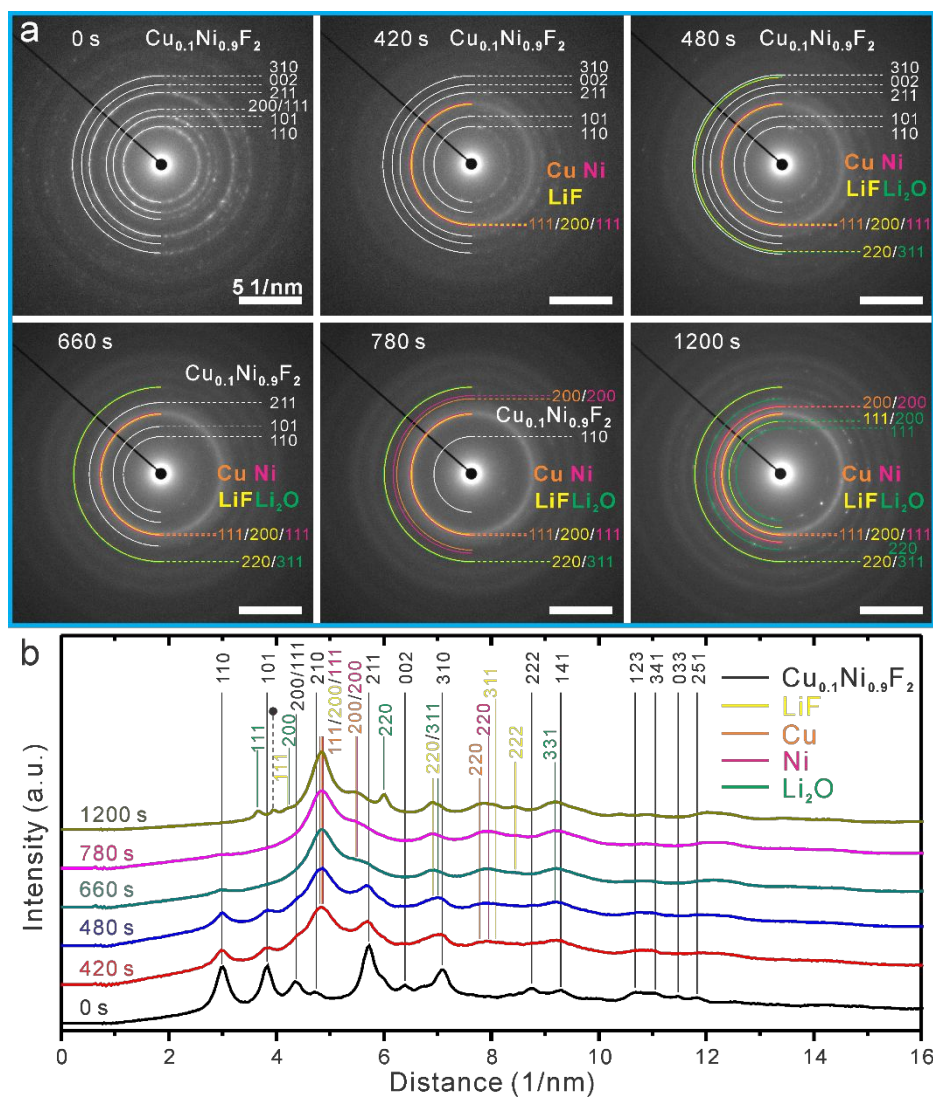


Figure S3. (a) Time-lapse SAED patterns of $\text{Cu}_{0.1}\text{Ni}_{0.9}\text{F}_2$ nanoparticles. (b) Intensity profiles converted from each SAED patterns in (a) as a function of distance. $\text{Cu}_{0.1}\text{Ni}_{0.9}\text{F}_2$ nanoparticles are gradually changed to Cu, Ni, and LiF phases during lithiation process.

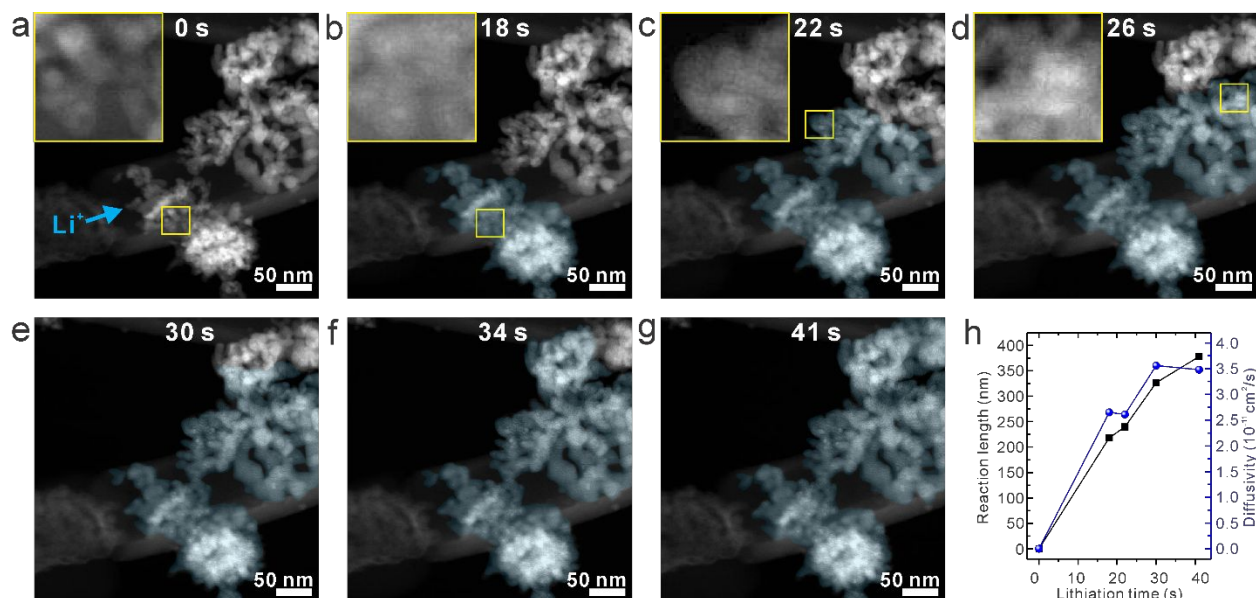


Figure S4. (a-g) Time-lapse HAADF-STEM images of $\text{Cu}_{0.1}\text{Ni}_{0.9}\text{F}_2$ nanoparticles during lithiation process. Each inset shows magnified images obtained from the yellow square region. (h) The diffusion length of Li ions along nanoparticles and the calculated diffusivity as a function of lithiation time. The diffusivity is estimated based on the equation $D = d^2/t$, where D is diffusivity, d is diffusion distance, and t is the lithiation time.

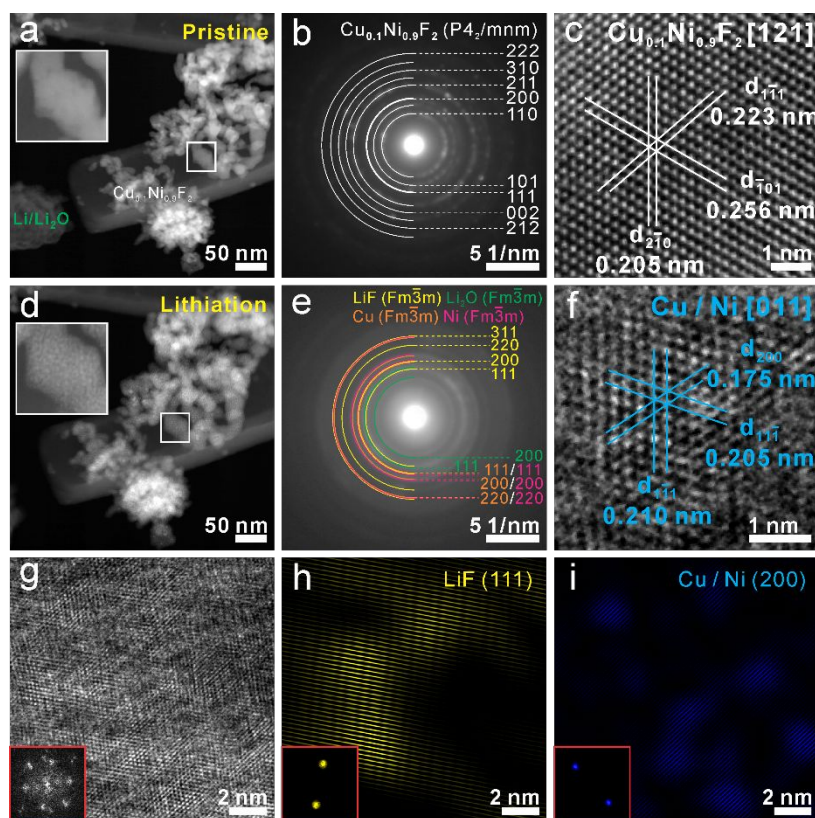


Figure S5. HAADF-STEM images, SAED patterns, and HRTEM images of $\text{Cu}_{0.1}\text{Ni}_{0.9}\text{F}_2$ (a-c) at the pristine state and (d-f) after lithiation process, respectively. (g) HRTEM image obtained after conversion step. (h,i) The inverse FFT images based on the FFT spots (inset) which are selected from FFT patterns in (g). The formed Cu and Ni nanoparticles are embedded in LiF matrix after lithiation.

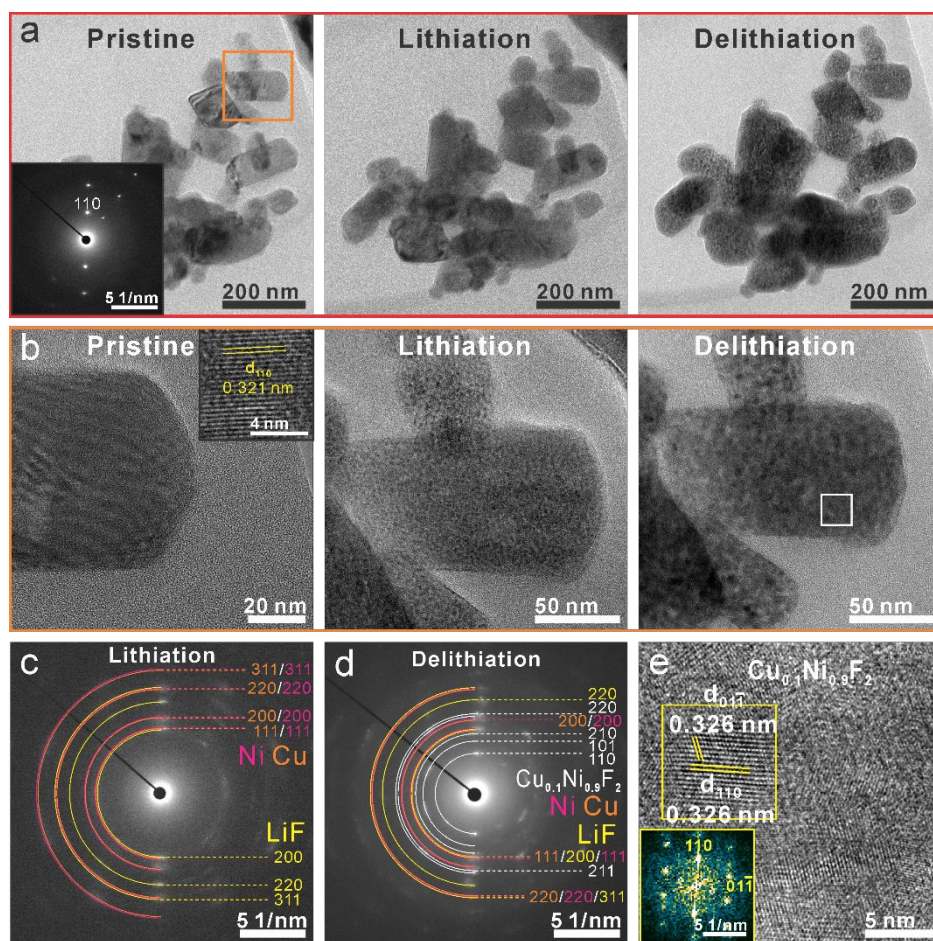


Figure S6. (a) Sequential TEM images of single-crystalline $\text{Cu}_{0.25}\text{Ni}_{0.75}\text{F}_2$ nanoplates at the pristine state, after lithiation, and after delithiation process. The inset shows SAED patterns of $\text{Cu}_{0.25}\text{Ni}_{0.75}\text{F}_2$ nanoplate at the pristine state. (b) The morphology changes of single nanoplate obtained from the orange square region in (a) during the first discharge/charge cycles. The inset shows HRTEM image of $\text{Cu}_{0.25}\text{Ni}_{0.75}\text{F}_2$ nanoplate at the pristine state. SAED patterns of $\text{Cu}_{0.25}\text{Ni}_{0.75}\text{F}_2$ nanoplate (c) after lithiation and (d) after delithiation obtained from (b). After extracting Li ions from matrix, the converted phases are recombined to the original structure. (e) HRTEM image shows the reversible phase transformation of $\text{Cu}_{0.25}\text{Ni}_{0.75}\text{F}_2$ phase after charge step.

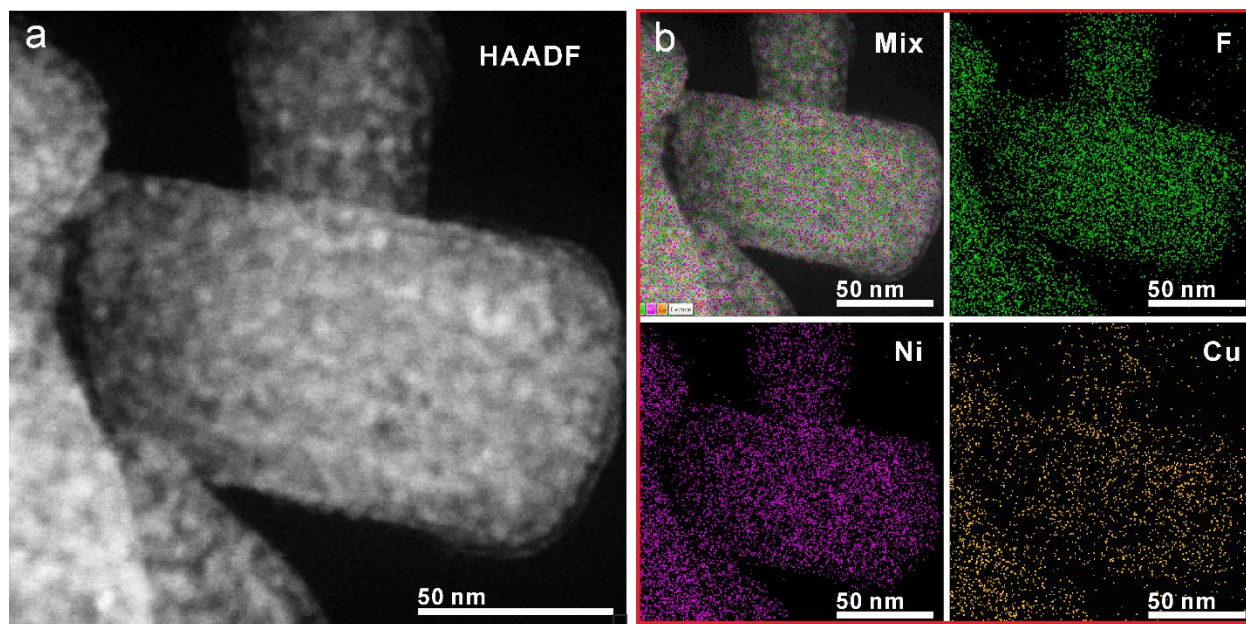


Figure S7. (a) HAADF-STEM image of $\text{Cu}_{0.25}\text{Ni}_{0.75}\text{F}_2$ nanoplate obtained from the square region in Figure S6a after delithiation process. (b) EDS map images corresponding to the region in (a). All elements are well re-distributed after delthiation process.

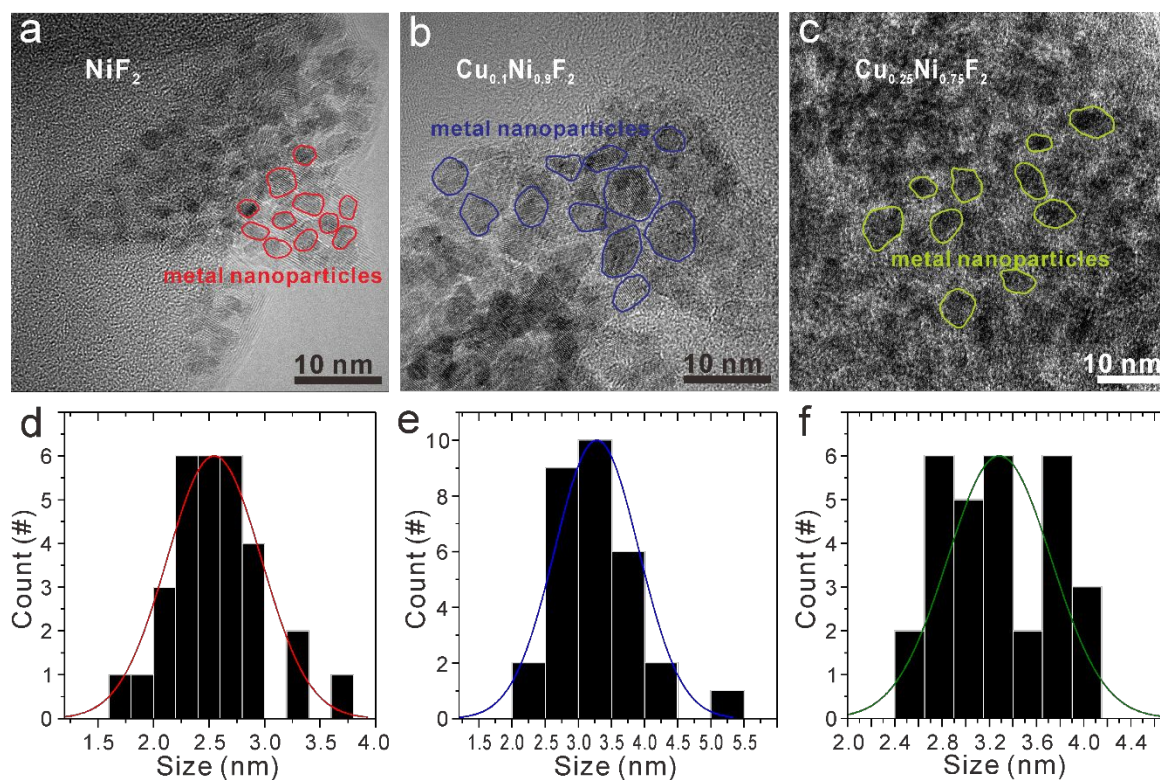


Figure S8. HRTEM images of (a) NiF_2 , (b) $\text{Cu}_{0.1}\text{Ni}_{0.9}\text{F}_2$, and (c) $\text{Cu}_{0.25}\text{Ni}_{0.75}\text{F}_2$ materials after the first lithiation process. (d-f) Metal nanoparticle size distribution and normal fitting corresponding to (a-c).