Electronic Supplementary Information

Fe-Ni Foams Self-Heal during Redox Cycling via Reversible Formation/Homogenization of a Ductile Ni Scaffold

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Table S1. Nickel content in Fe-Ni foams prepared from corresponding freeze casting suspensions.

Target Ni Content (at. % of Fe+Ni)	Freeze Casting Suspension Composition (vol. % of suspension)				
	Fe_2O_3	NiO	Zephrym	PEG	Water
0	10	0	0.5	2	87.5
6.6	9.51	0.49	0.5	2	87.5
19	8.52	1.48	0.5	2	87.5
25	8.03	1.97	0.5	2	87.5

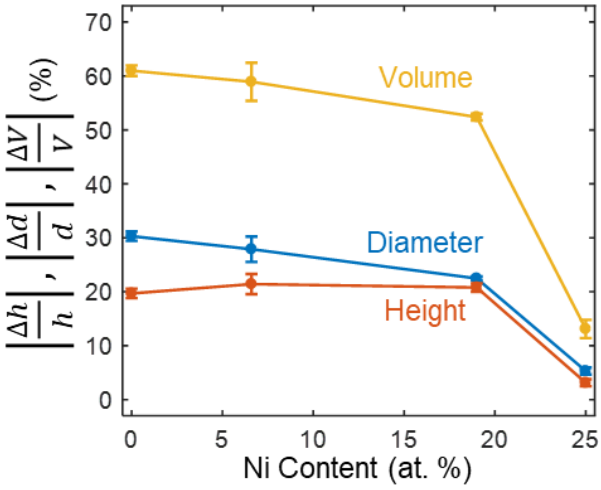

Figure S1. Effect of Ni content on shrinkage of foam height (orange), diameter (blue), and volume (yellow) after 5 redox cycles, relative to as-prepared foams. Error bars: ± std. error (N = 3).

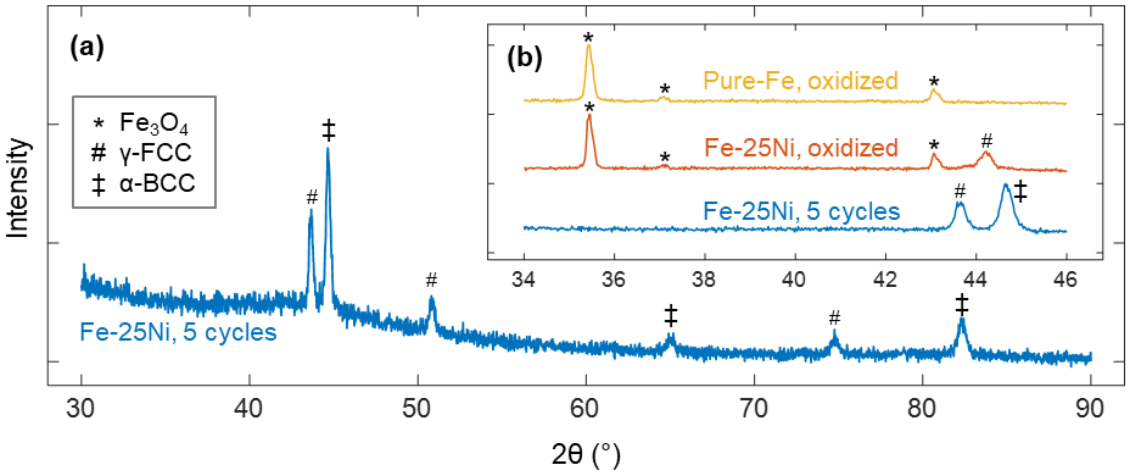

Figure S2. (a) X-ray diffraction pattern of Fe-25Ni foam cooled to room temperature after 5 redox cycles. Inset (b) shows detail of first peaks, compared with pure-Fe (yellow) and Fe-25Ni (orange) after single oxidation half-cycles. The γ-FCC (#) peak shift in Fe-25Ni from the oxidized state to after 5 cycles is due to an increase in Fe content in the γ-phase.

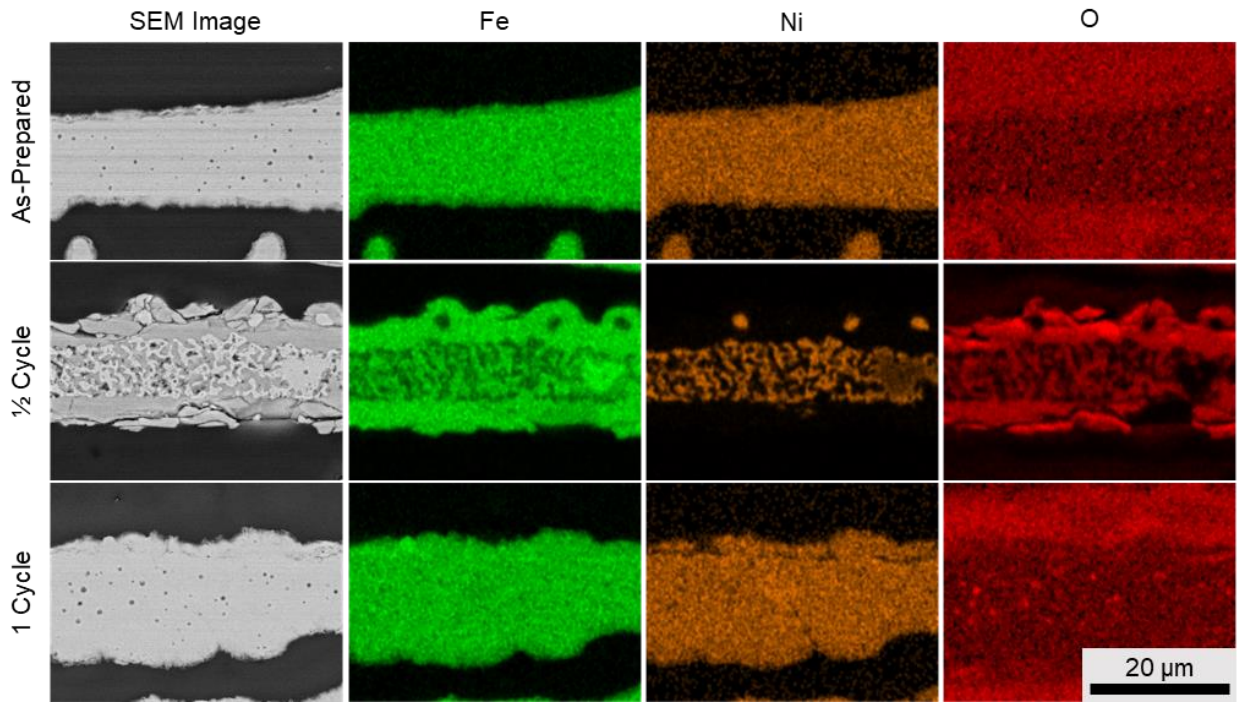

Figure S3. Elemental spatial maps of individual lamellae in Fe-25Ni foams as-prepared, oxidized (½ cycle), and after 1 redox cycle, measured by EDS of foam cross-sections. Maps are not group-normalized, so mounting epoxy creates strong oxygen signal for foams as-prepared and after 1 cycle.

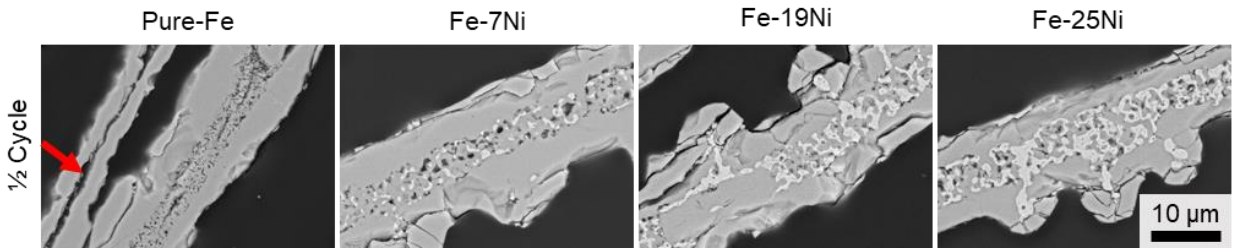

Figure S4. SEM of cross-sections of lamellae in fully oxidized foams (after ½ cycle). Metal and oxide phases are light- and dark-gray, respectively; pores and cracks are black. Pure-Fe shows two adjacent lamellae, a thin one with a central crack (red arrow) and a thicker one with Kirkendall micropores in its core.

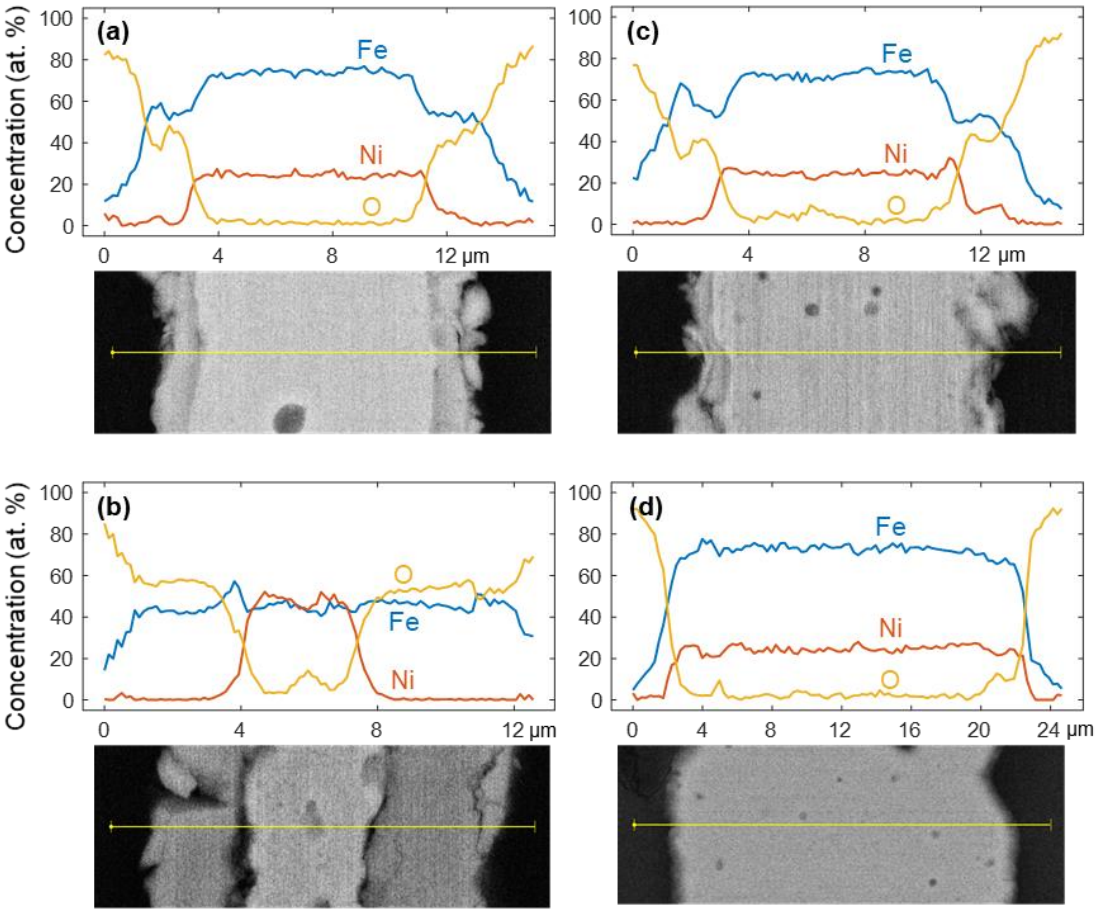

Figure S5. Elemental concentrations from EDS line scans of lamellae in Fe-25Ni foams removed at various points during redox cycling. (a) First, initial oxidation, (b) mostly oxidized, (c) partially reduced, and (d) fully reduced after 20 cycles (using 90 min reduction half-cycles). Metal and oxide phases are light- and dark-gray, respectively; pores are black. High oxygen concentration in pores is due to mounting epoxy.