Electronic Supplementary Material

Selective Pb^{2+} removal and electrochemical regeneration of fresh and recycled FeOOH

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Supporting information to https://doi.org/10.1007/s12274-023-5569-2

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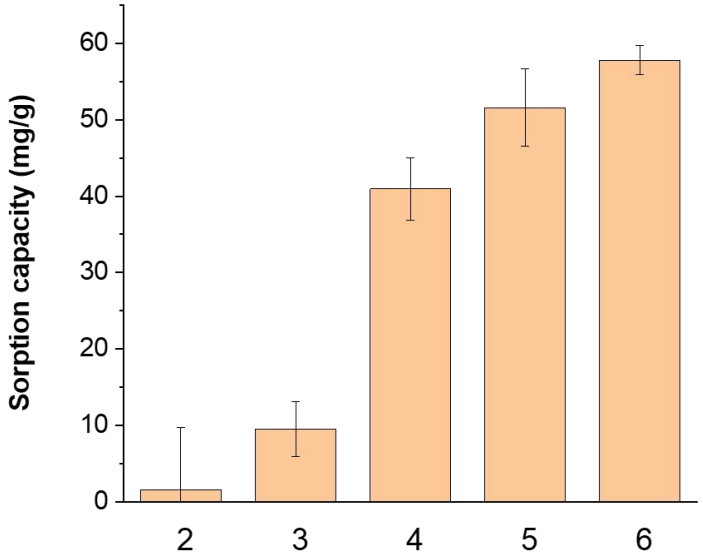

Figure S1 The sorption capacity of FeOOH powder in 10 mM $Pb(NO_3)_2$ at different pH values.

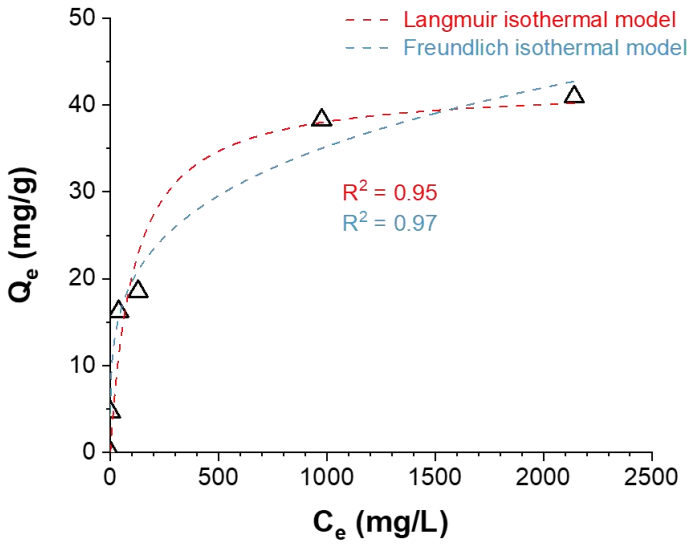

Figure S2 Adsorption isotherm of Pb^{2+} by FeOOH and the fitting results according to the Langmuir (**Equation 6**) and Freundlich models (**Equation 7**).

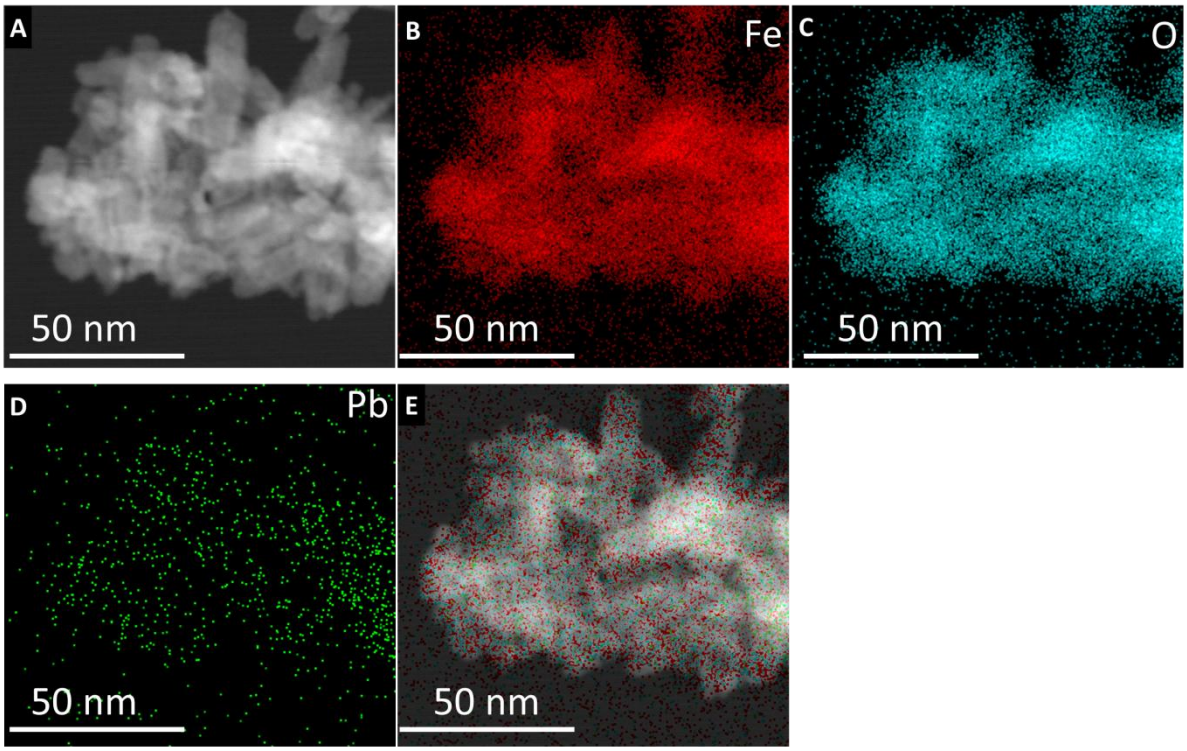

Figure S3 A HAADF-Stem image (A) of FeOOH powder after immersing in $Pb(NO_3)_2$ solution corresponding to the EDX mapping of Fe (B), O (C), Pb (D) and overlay image of Fe, O, and Pb (E); possible Pb^{2+} existing sites in β-FeOOH.

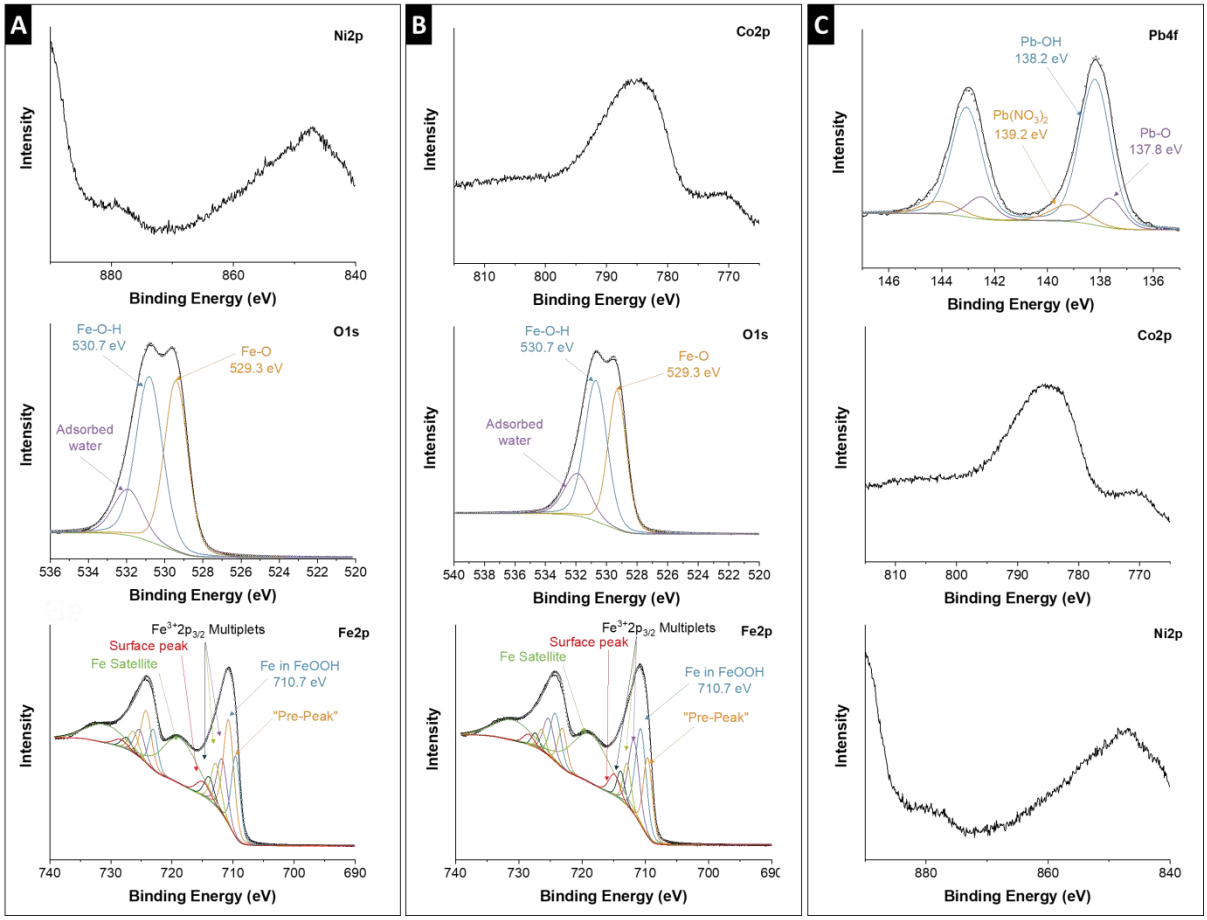

Figure S4 XPS spectra of FeOOH after immersing in Ni^{2+}(A), Co^{2+} (B), and mixed solution (C).

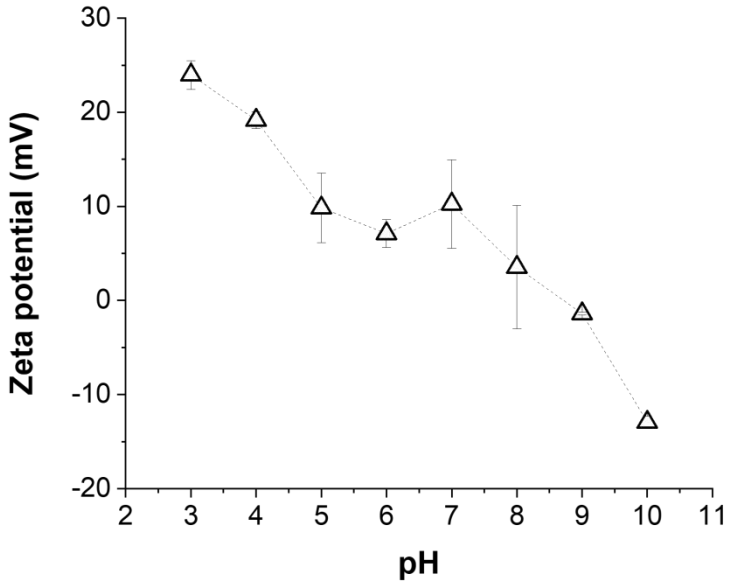

Figure S5 Zeta potential of FeOOH in 100 mM $NaNO_3$ at different pH.

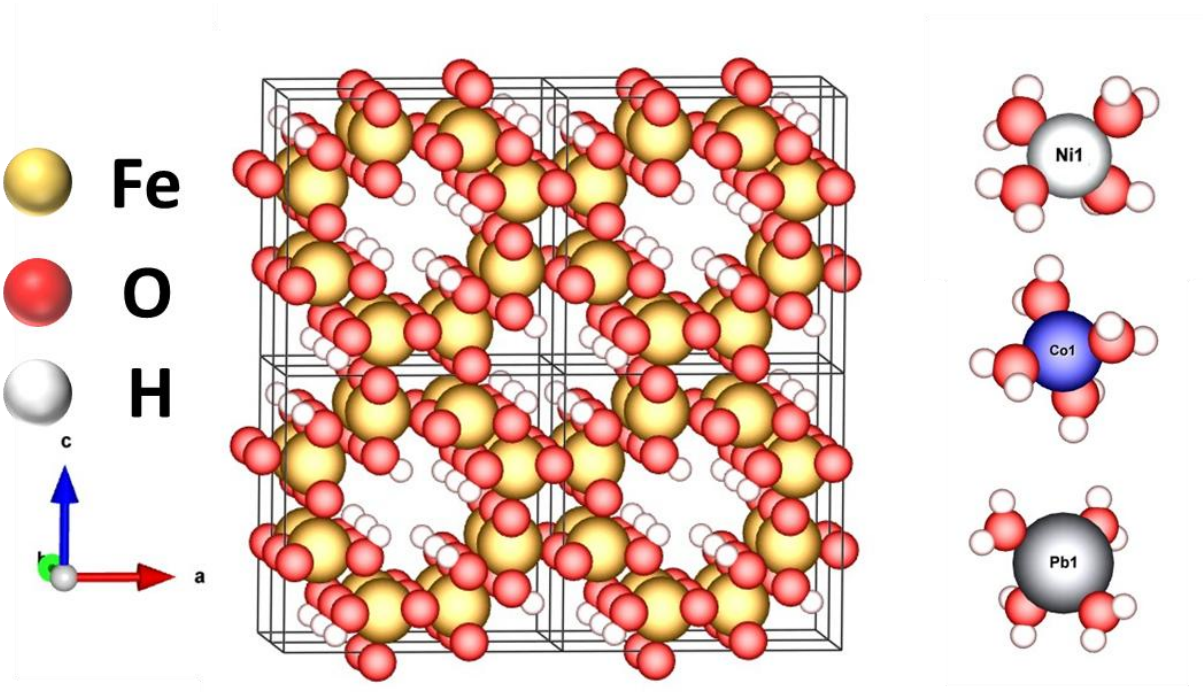

Figure S6 The crystal structure of FeOOH and Ni^{2+}, Co^{2+}, and Pb^{2+} clusters in the DFT calculations.

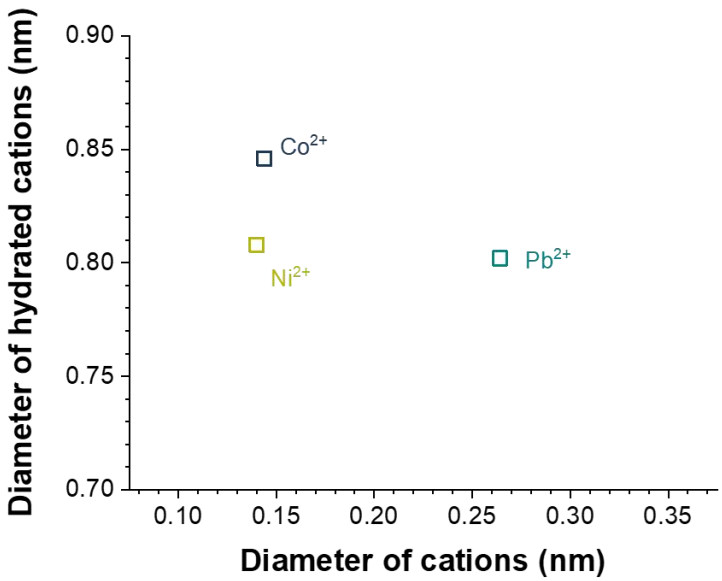

Figure S7 The theoretical diameter of Ni^{2+}, $Co^{2+,}$ and Pb^{2+}. Data from: Transformation in a Homoionic Form on the Removal of Some Heavy Metal Ions from Wastewater, Journal of Environmental Science and Health, Part A, 38 (2003) 545-554.

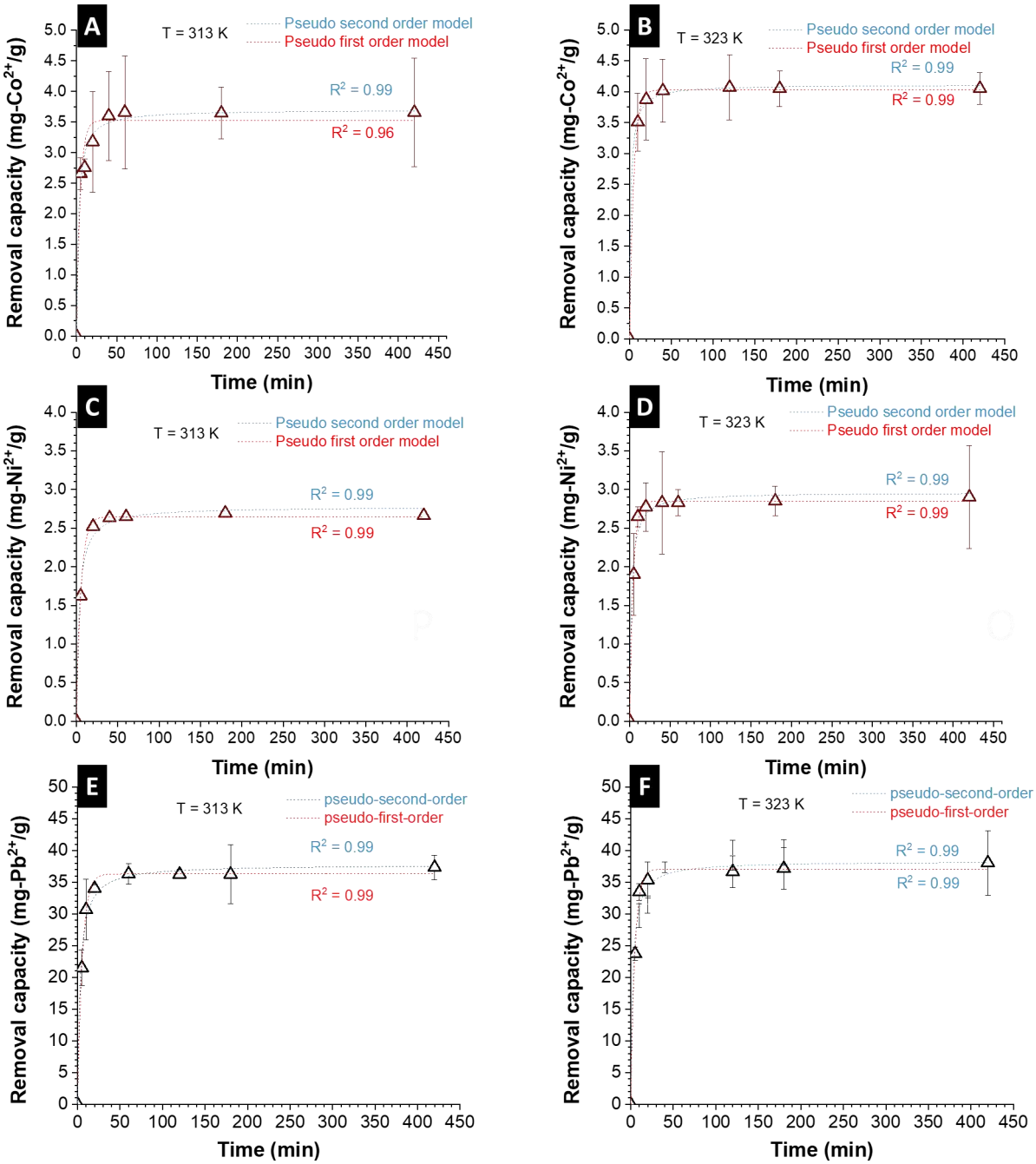

Figure S8 Sorption kinetics of Co^{2+} by FeOOH at 313 K (A) and 323 K (B); Sorption kinetics of Ni^{2+} by FeOOH at 313 K (C) and 323 K (D); Sorption kinetics of Pb^{2+} by FeOOH at 313 K (E), and 323 K (F).

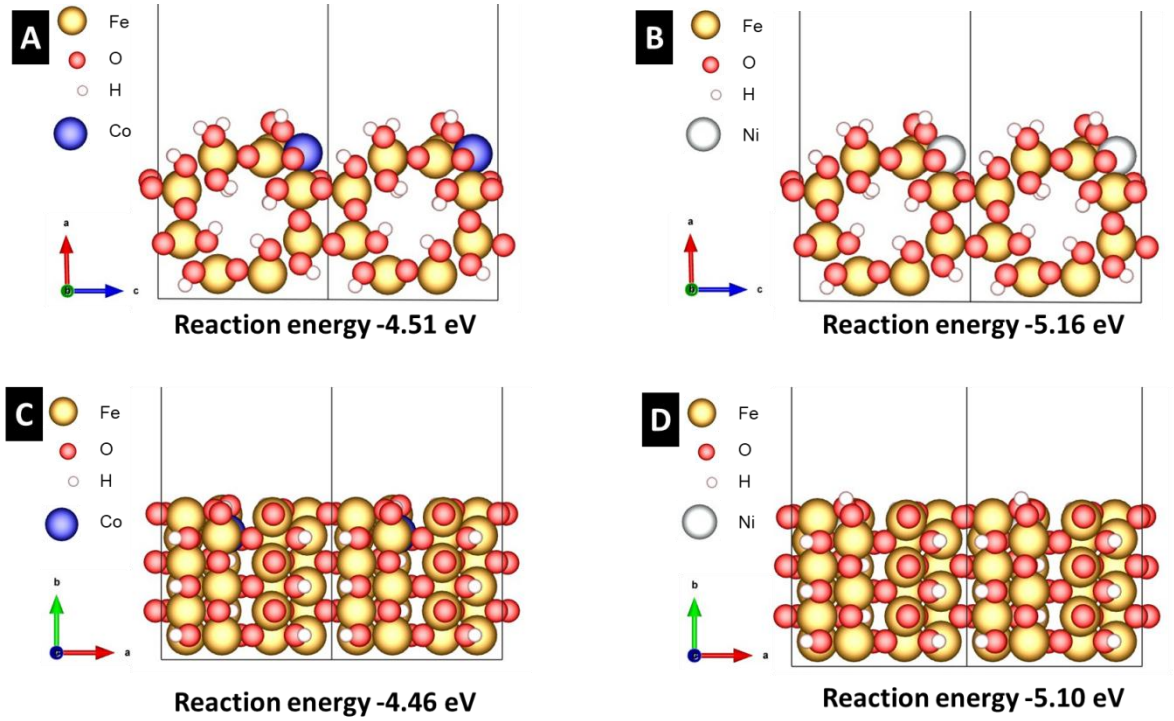

Figure S9 Calculated optimized structure and corresponding reaction energy of FeOOH for Co^{2+} (A), Ni^{2+} (B) on the (100) surface and Co^{2+} (C), Ni^{2+} (D) on the (010) surface.

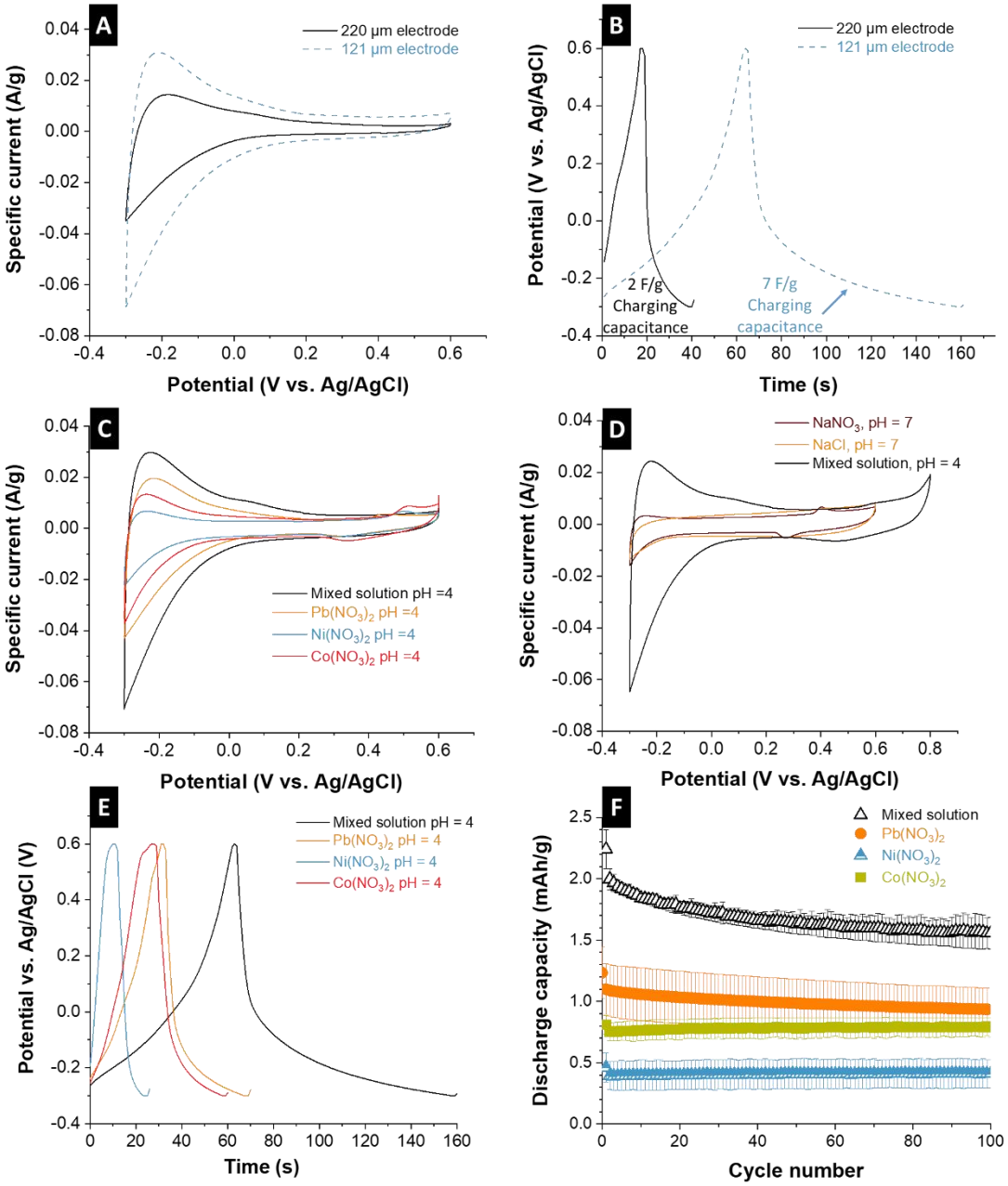

Figure S10 Cyclic voltammogram (A) and galvanostatic charge/discharge profiles (B) of FeOOH with different thicknesses of the electrode in the mixed solution with 10 mM $Pb(NO_3)_2$, 10 mM $Co(NO_3)_2$, and 10 mM $Ni(NO_3)_2$; cyclic voltammogram curves (C, D) and galvanostatic charge/discharge of 121 µm FeOOH electrode (E) measured in various electrolytes; discharge capacity of 121 µm FeOOH during 100 cycles in different electrolytes.

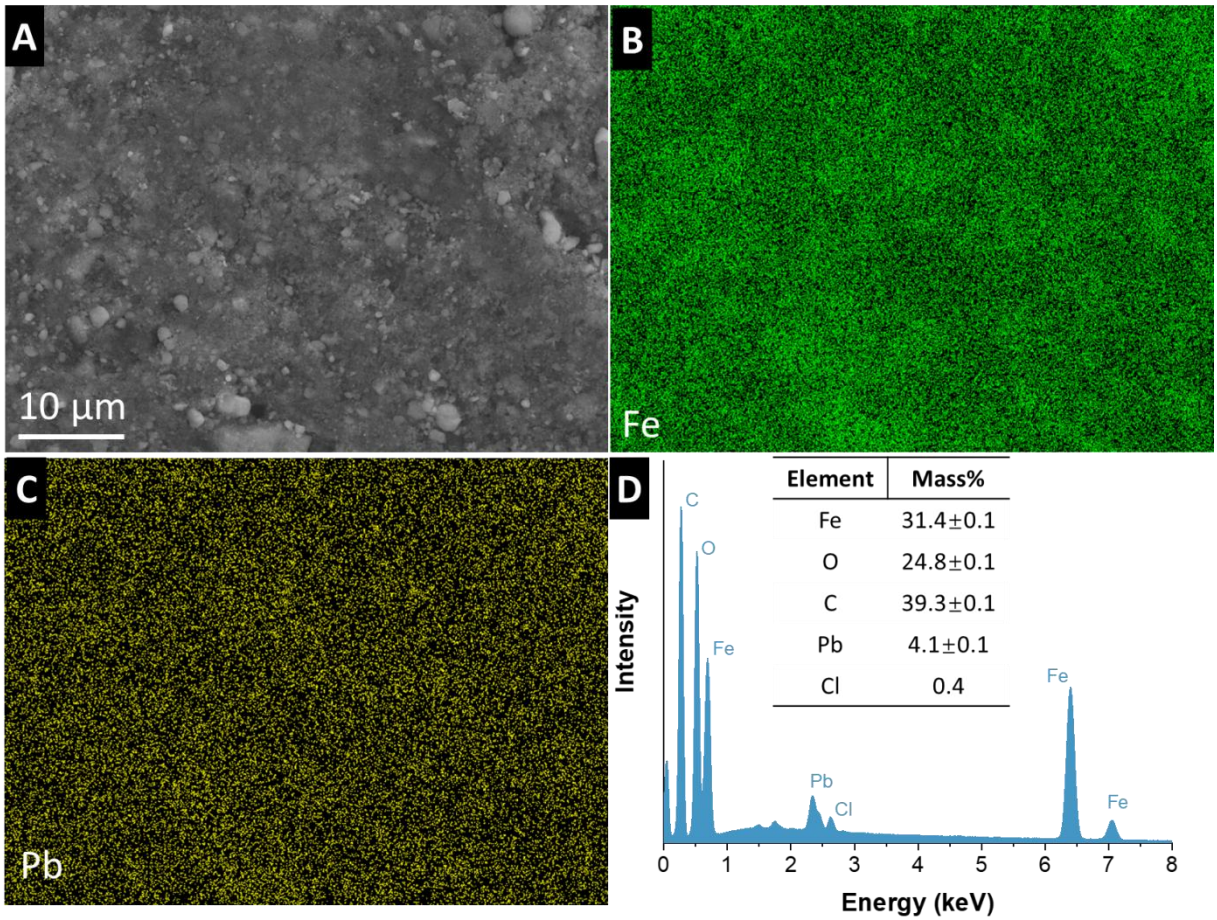

Figure S11 A scanning electron micrograph of the FeOOH electrode at discharging state (A) and the EDX mapping of elements: (B) Fe, (C) Pb; (D) EDS spectrum with an insert table showing the mass ratio of each element.

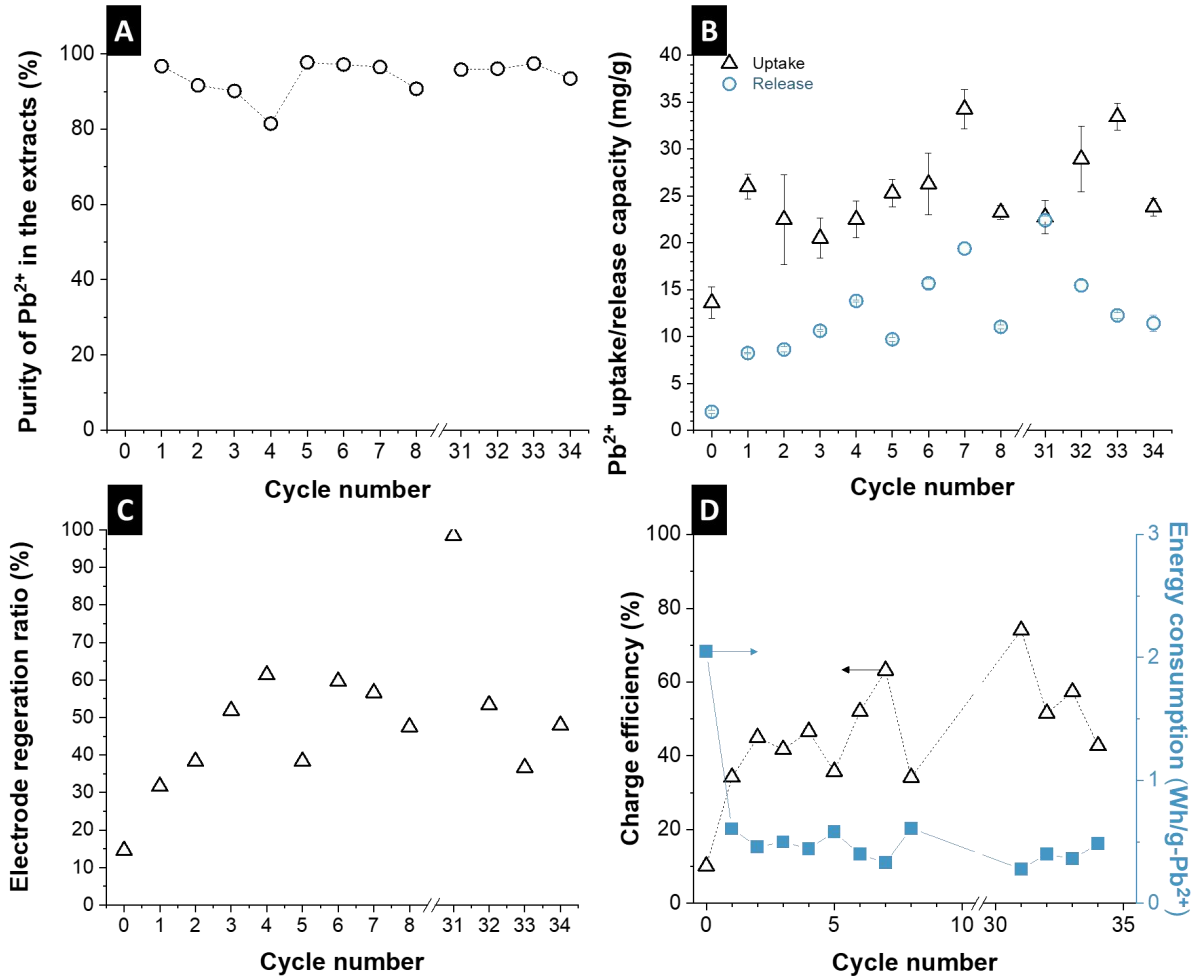

Figure S12 The electrochemical experiment with an electrode with a thickness of 220 μm in a mixed solution of 10 mM Co^{2+}, 10 mM Ni^{2+}, and 10 mM Pb^{2+}: (A) the purity of Pb^{2+} in the extracts; (B) the uptake/release capacity of Pb^{2+} different cycles; (C) the capacity regenerate percentage at different cycles; (D) the charge efficiency and energy consumption at different cycles.

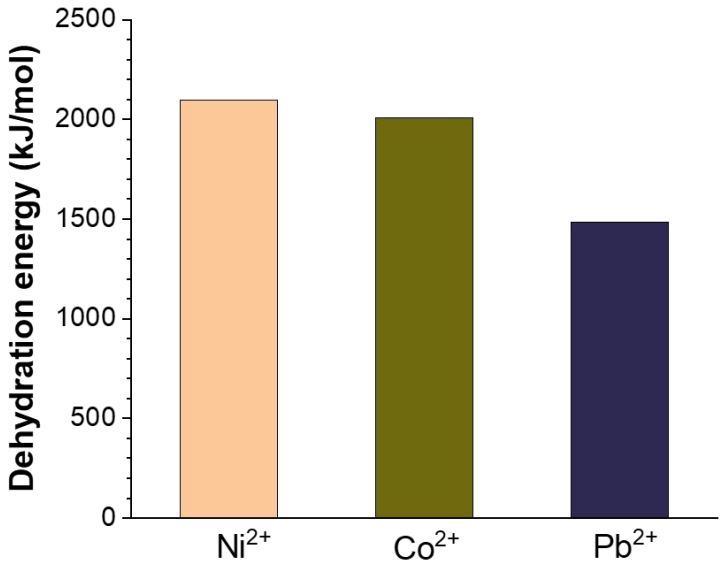

Figure S13 The theoretical dehydration energy of Ni^{2+}, $Co^{2+,}$ and Pb^{2+}. Data from: Transformation in a Homoionic Form on the Removal of Some Heavy Metal Ions from Wastewater, Journal of Environmental Science and Health, Part A, 38 (2003) 545-554.

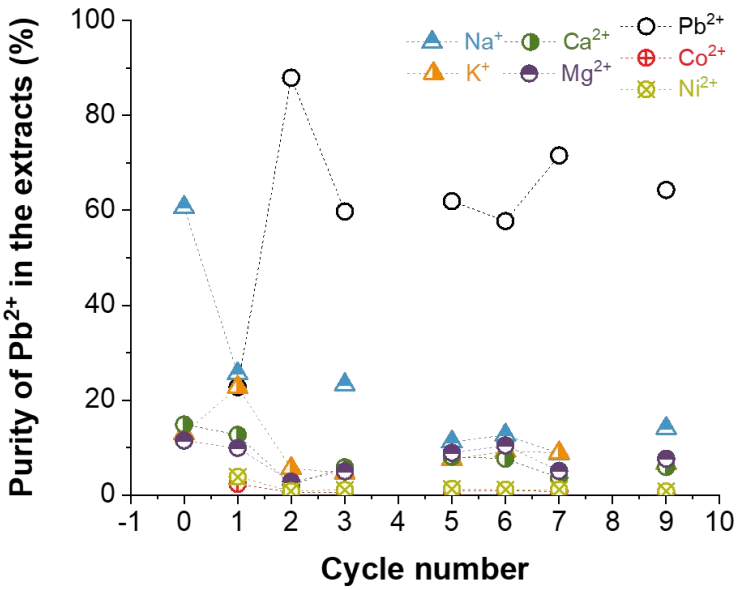

Figure S14 The ratio of each element in the extracts in the experiment conducted in the feed water containing 10 mM Pb^{2+}, 10 mM Co^{2+}, 10 mM Ni^{2+}, 100 mM Na^{+}, 100 mM K^{+}, 100 mM Mg^{2+}, and 100 mM Ca^{2+}.

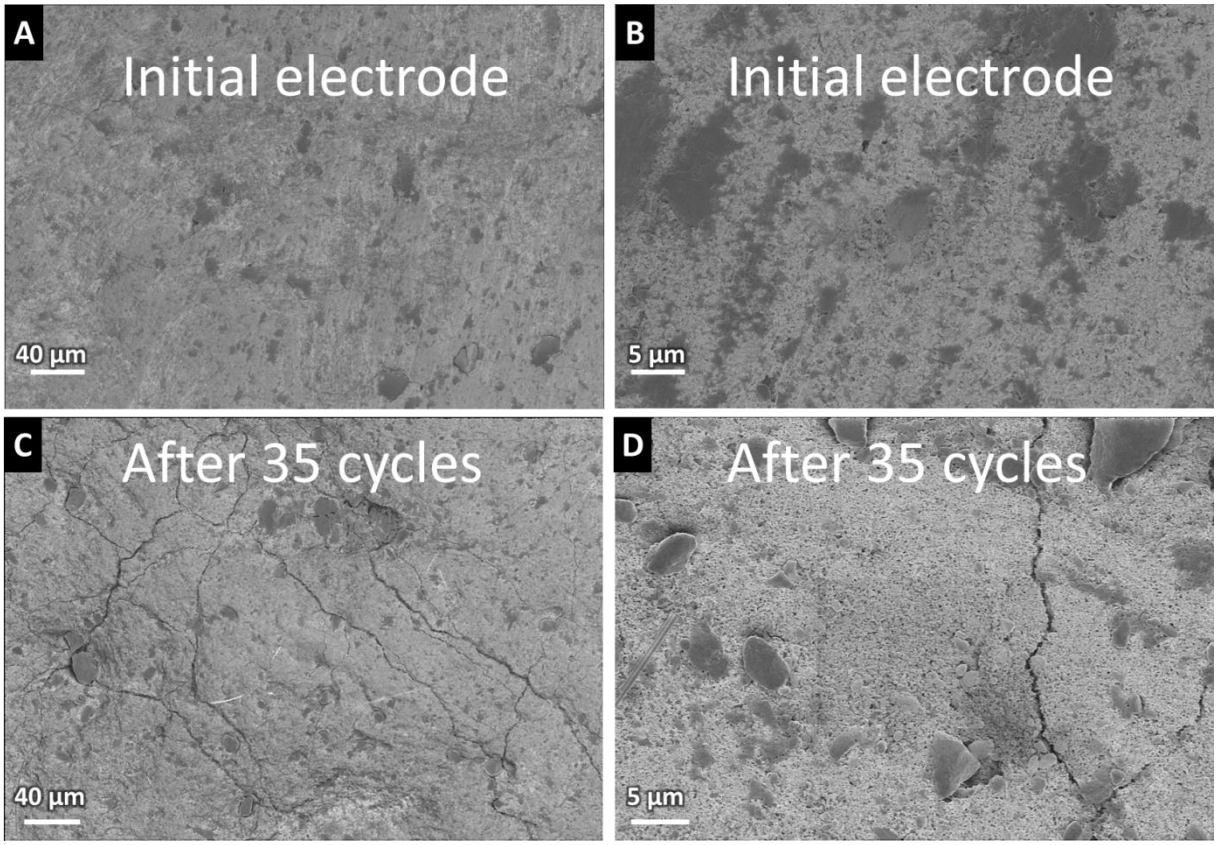

Figure S15 SEM images of initial FeOOH electrode (A, B) and electrodes after 35 cycles (C, D).

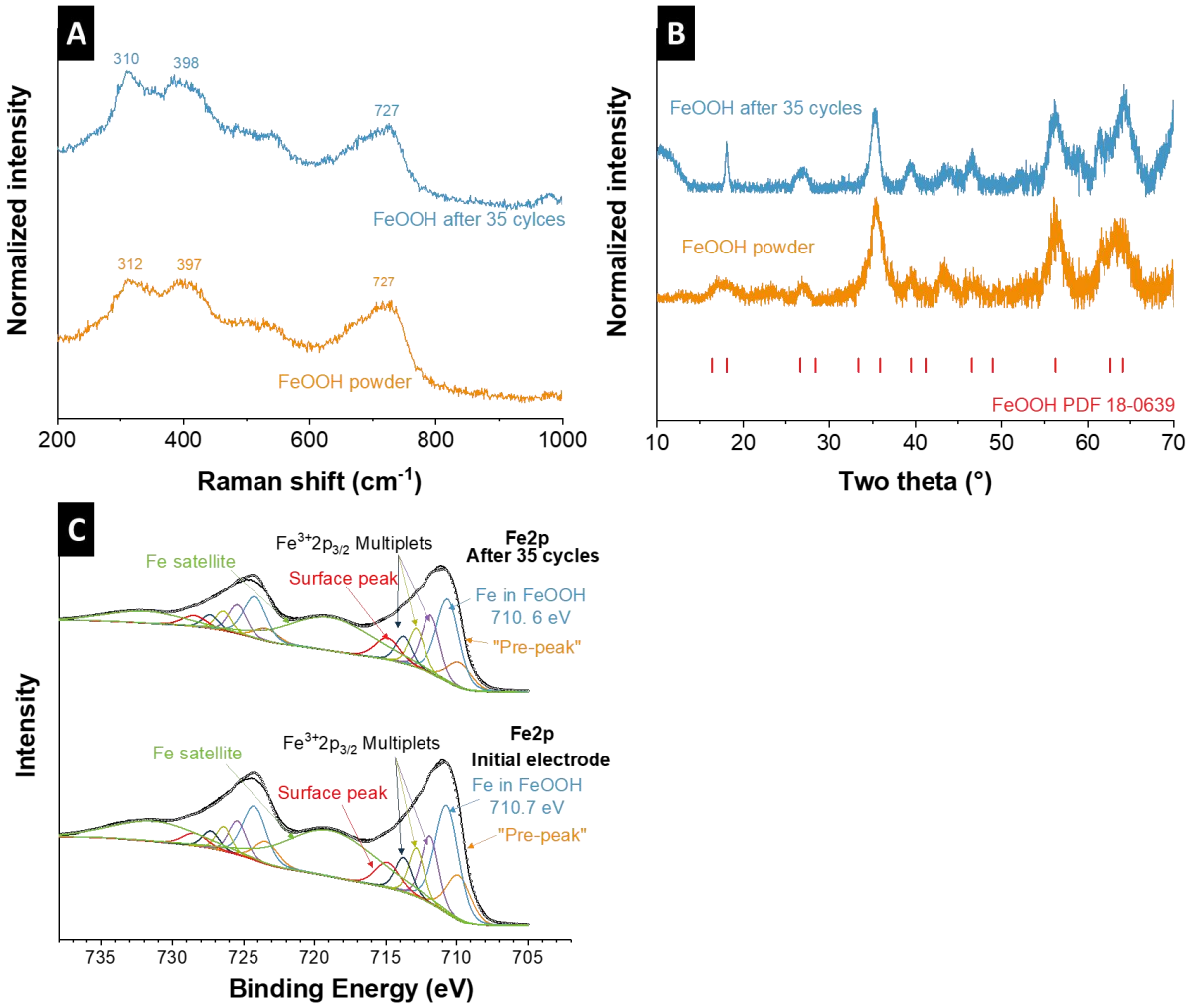

Figure S16 Post-mortem Raman spectra (A), X-ray diffraction patterns (B), and XPS spectra of Fe 2p (C) of the FeOOH powder and of the electrode after cycling.

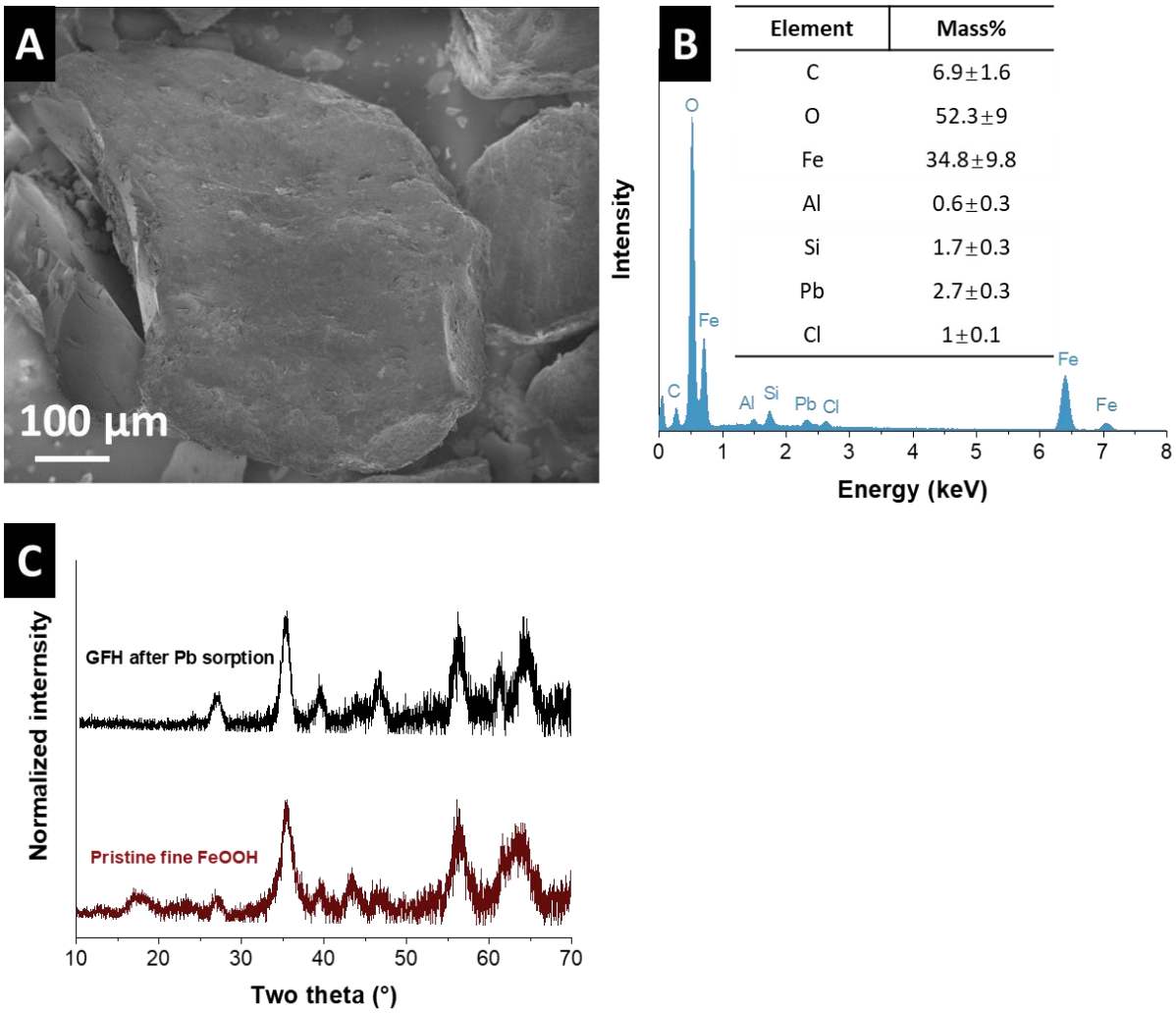

Figure S17 SEM (A) EDX (B) and XRD (C) of exhausted GFH.

Table S1 Element component of GFH before and after treating drinking water

Element	Before treatment (mg/kg)	After Treatment (mg/kg)
Fe	597000	580000
Mn	1405	730
Cu	11	265
Cr	37	34
Ni	53	27
V	150	138
Pb	< 1	7000
As	< 1	<1
Cd	< 0,05	0,05

Table S2 Experimental details for FeOOH and E-FeOOH electrode

Electrode	Feed water	Recovery solution	Cycle number	Operations		Note
				Feed water	Recovery solution	
121 µm-FeOOH	Pb^{2+}, 10 mM Co^{2+}, 10 mM Ni^{2+}, 10 mM	10 mM NaCl	35	Keeping 17 mL for each cycle. Taking 2 mL before and after treatment for ICP	Initial volume 80 mL. Taking 2 mL before and after treatment for ICP.*	11^{th}-30^{th} cycle charged/discharged in the feed water
124 µm-FeOOH	Pb^{2+}, 5 mM Co^{2+}, 5 mM Ni^{2+}, 5 mM Na^{+}, 100 mM K^{+}, 100 mM Ca^{2+}, 100 mM Mg^{2+}, 100 mM	10 mM LiCl	10		Initial volume 60 mL. Taking 2 mL before and after treatment for ICP	-
116 µm-E-FeOOH	Pb^{2+}, 10 mM Co^{2+}, 10 mM Ni^{2+}, 10 mM	10 mM NaCl	10			
220 µm-FeOOH	Pb^{2+}, 10 mM Co^{2+}, 10 mM Ni^{2+}, 10 mM	10 mM NaCl	34	Keeping 19 mL for each cycle. Taking 2 mL before and after treatment for ICP	Taking 3 mL before and after treatment for ICP. The initial volume of the 0^{th}, 3^{rd} and 8^{th} are 50 mL.	9^{th}-30^{th} cycle charged/discharged in the feed water

* At the 10^{th} cycle, extra 16 mL is taken out to enhance the concentration change in the following cycles by reducing the volume. And at the 34^{th} and 35^{th} cycles, only 1 mL is taken out for ICP measurement.