Electronic Supplementary Material (ESI) for Green Chemistry.

Electric potential-determined redox intermediates for effective recycling of spent lithium-ion batteries

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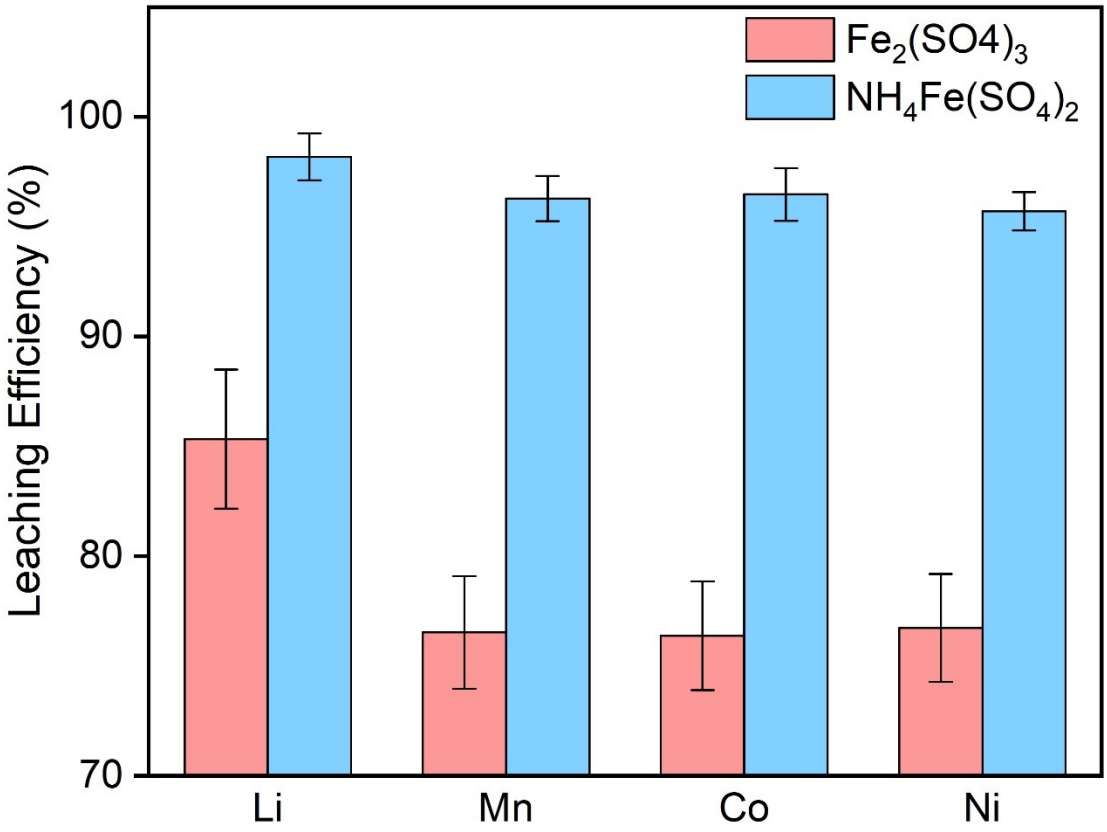

Fig. S1 Behaviors of synergistic metal leaching from NCM622 and LFP cathode by $Fe_2(SO_4)_3$ and $NH_4Fe(SO_4)_2$ (LFP/NCM622=1.8; $NH_4Fe(SO_4)_2 \cdot 12H_2O$/mixed cathode powder = 3:1, or $Fe_2(SO_4)_3$/mixed powder=1.1:1 (all in g/g) at 50°C for 30 min)

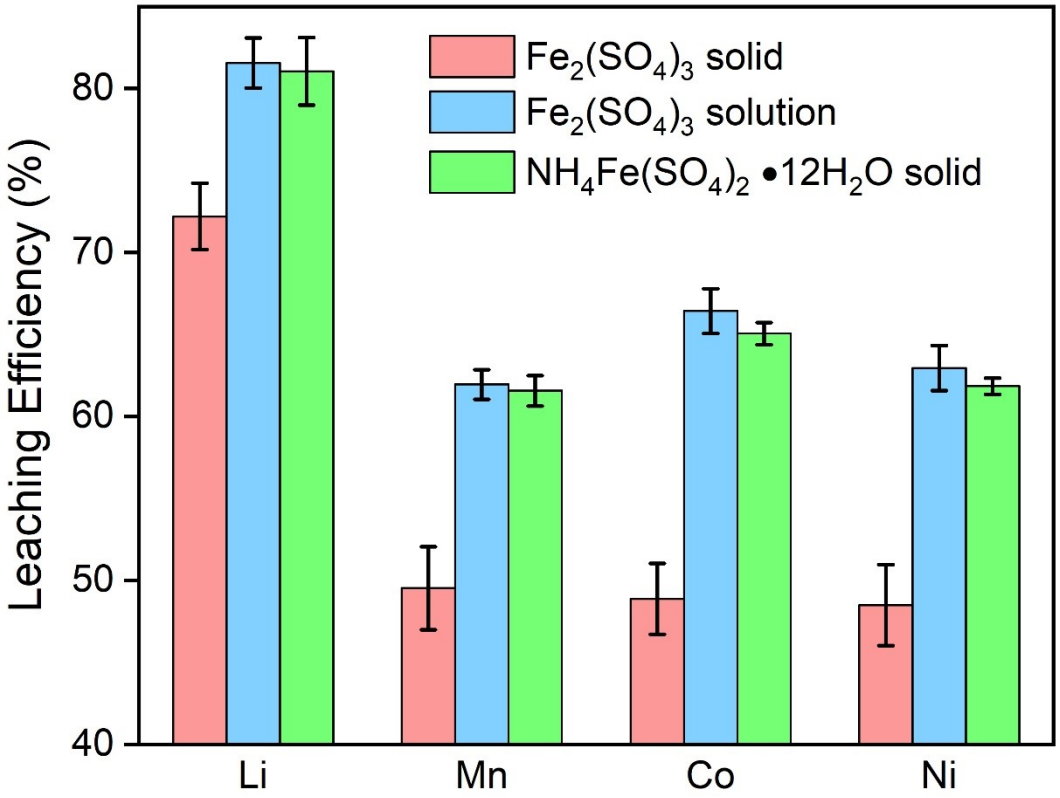

Fig. S2 Comparison of leaching efficiency by adding water with $Fe_2(SO_4)_3$ as solid powder and by preparing $Fe_2(SO_4)_3$ aqueous solution in advance, $NH_4Fe(SO_4)_2 \cdot 12H_2O$ also added for comparison (LFP/NCM622=1.8, $Fe_2(SO_4)_3$/mixed powder=1.2:1, $NH_4Fe(SO_4)_2 \cdot 12H_2O$/mixed cathode powder = 3:1 (all in g/g) at 50°C for 10 min)

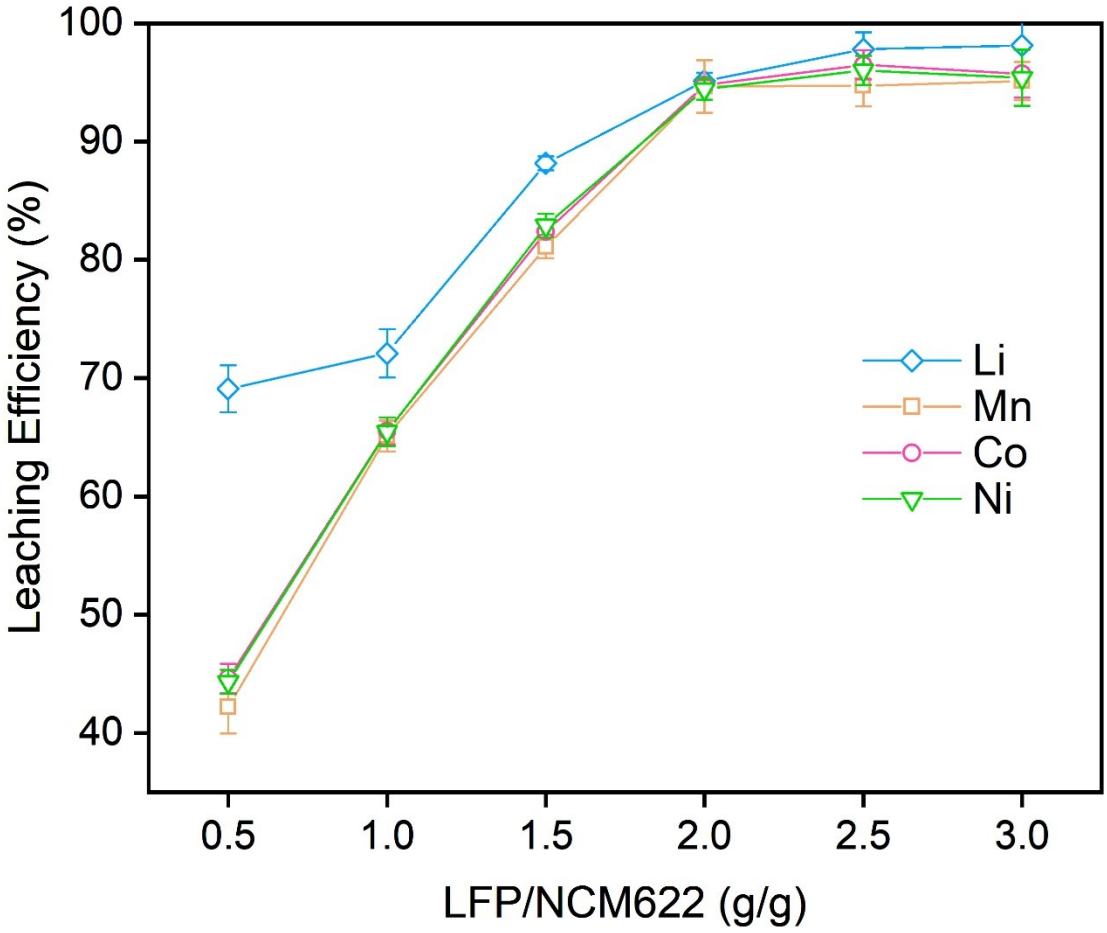

Fig. S3 Leaching efficiencies of metals under different LFP/NCM622 mass ratios

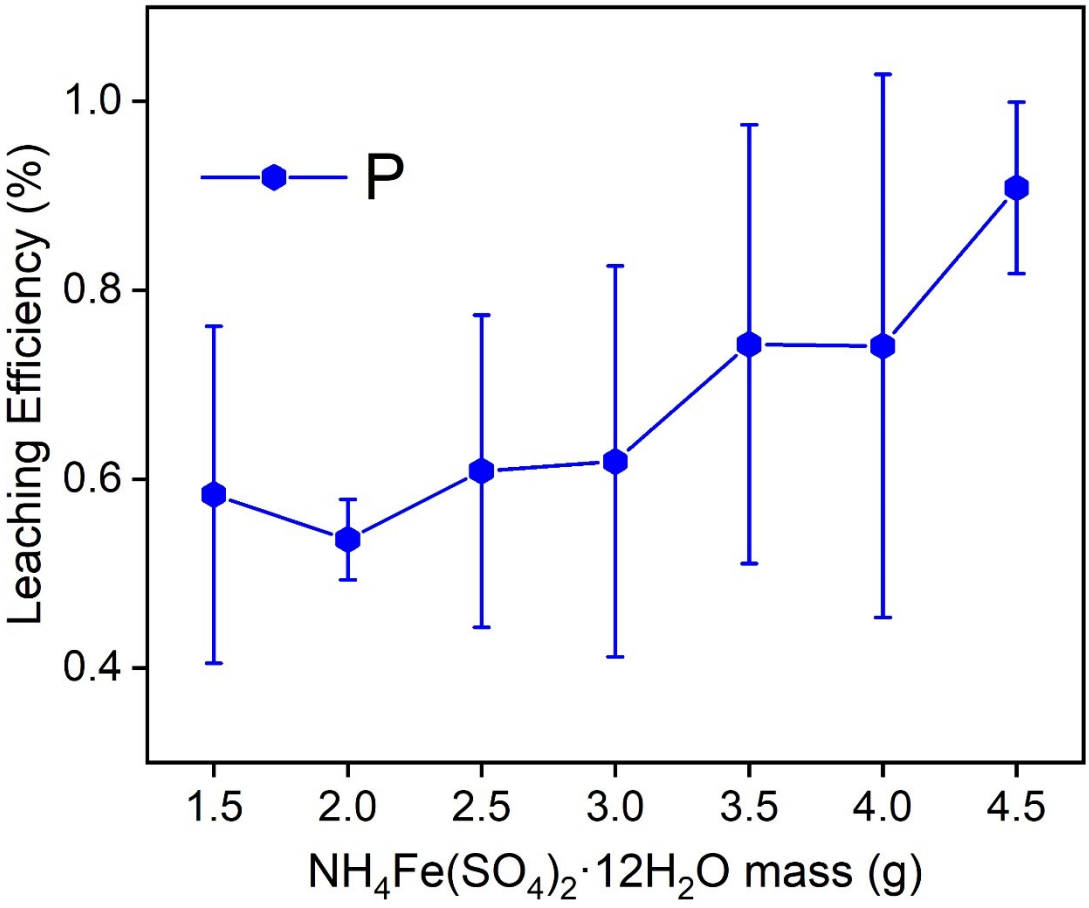

Fig. S4 Leaching efficiencies of phosphate with different added amount of $NH_4Fe(SO_4)_2 \cdot 12H_2O$. The phosphate was hardly extracted at all time.

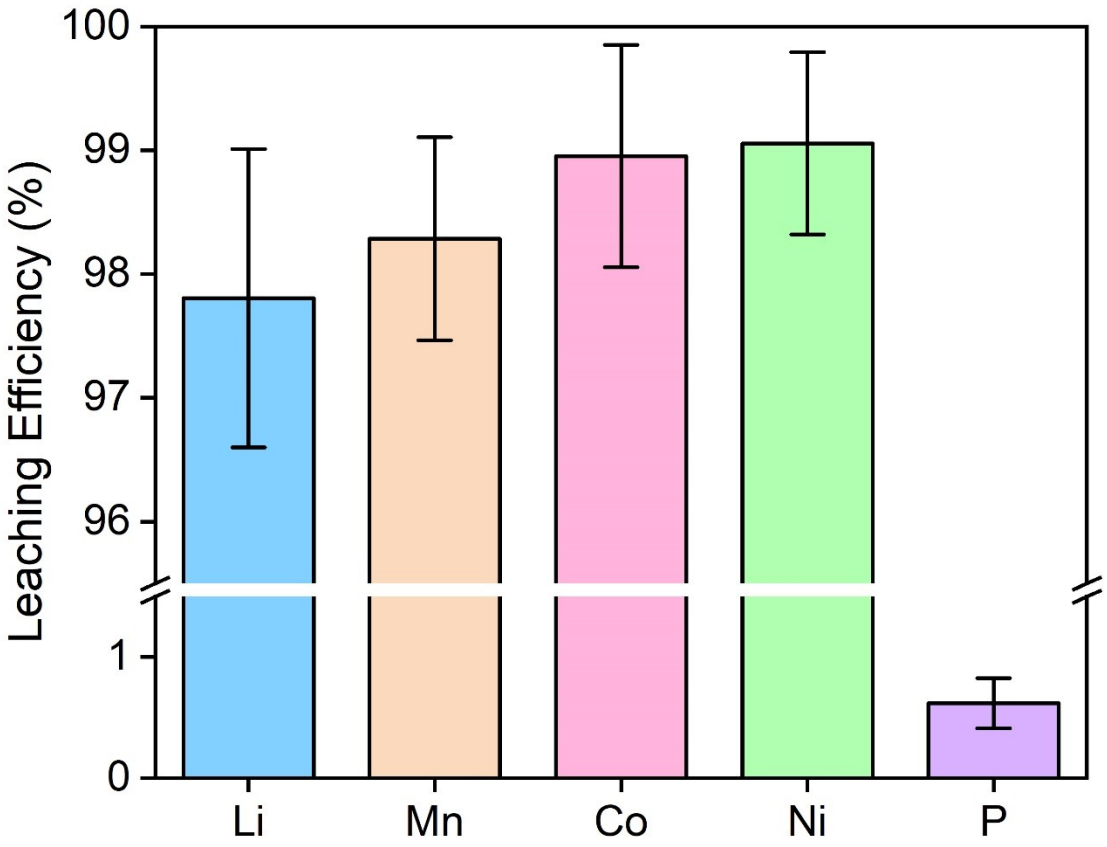

Fig. S5, Leaching efficiencies of Li, Mn, Co, Ni and P under optimized condition (LFP/NCM622=1.8 (g/g), $NH_4Fe(SO_4)_2 \cdot 12H_2O$/mixed cathode powder =3:1 (g/g), 50g/L, 50˚C, 30min).

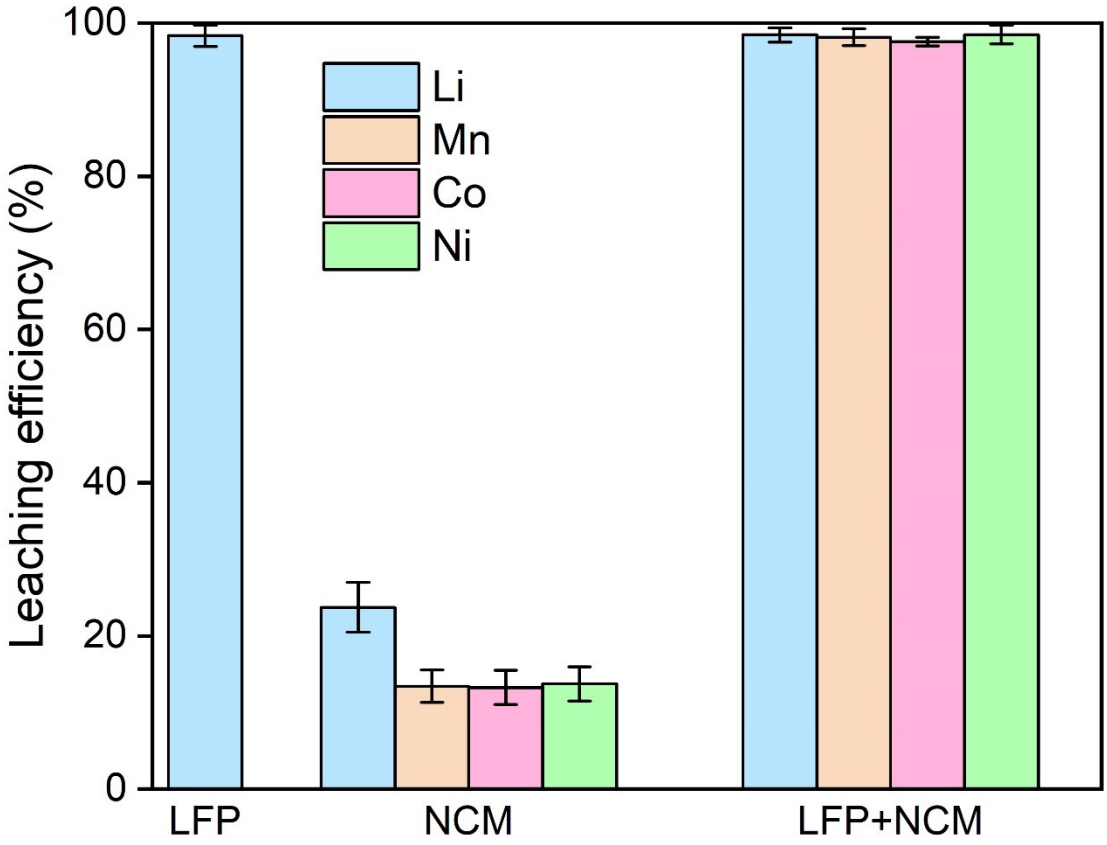

Fig. S6 Comparison of leaching effects using $NH_4Fe(SO_4)_2 \cdot 12H_2O$ + LFP alone, $NH_4Fe(SO_4)_2 \cdot 12H_2O$ + NCM alone, and $NH_4Fe(SO_4)_2 \cdot 12H_2O$ + mixed powder.

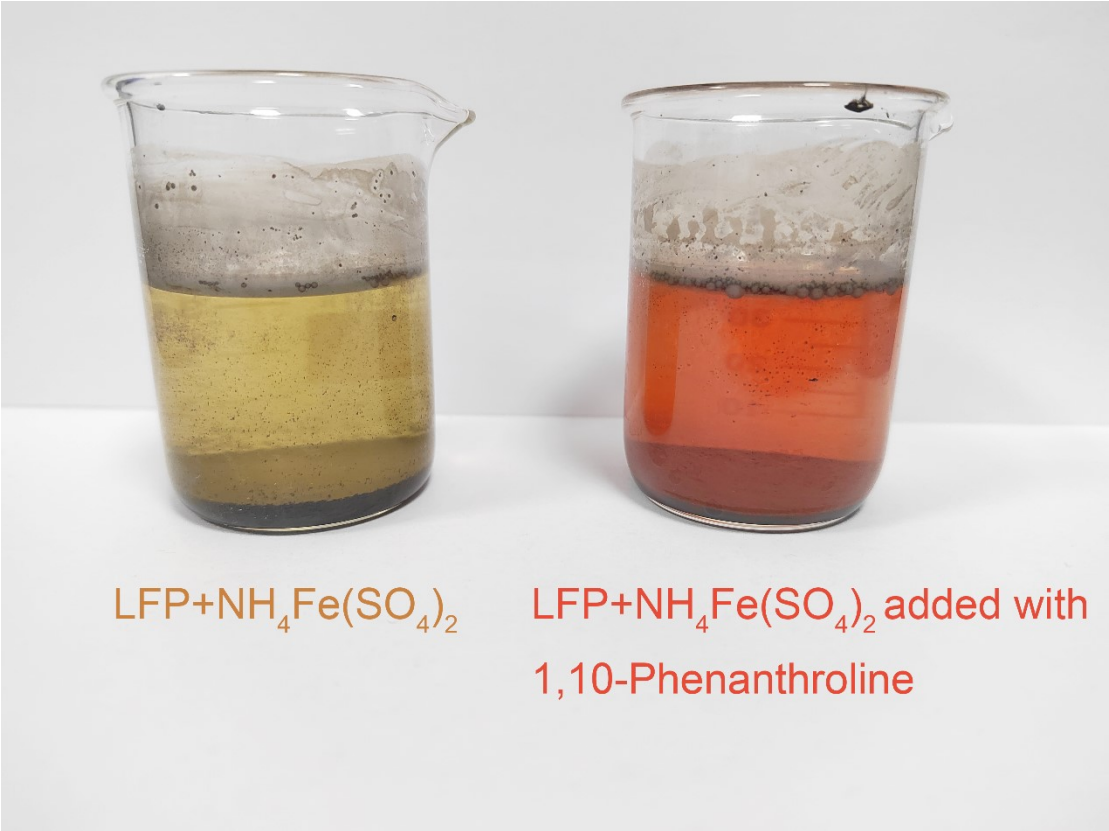

Fig. S7 Solution after reaction between LFP and $NH_4Fe(SO_4)_2 \cdot 12H_2O$. 1,10-Phenanthroline is added and the red color verifies the generation of Fe^{2+}.

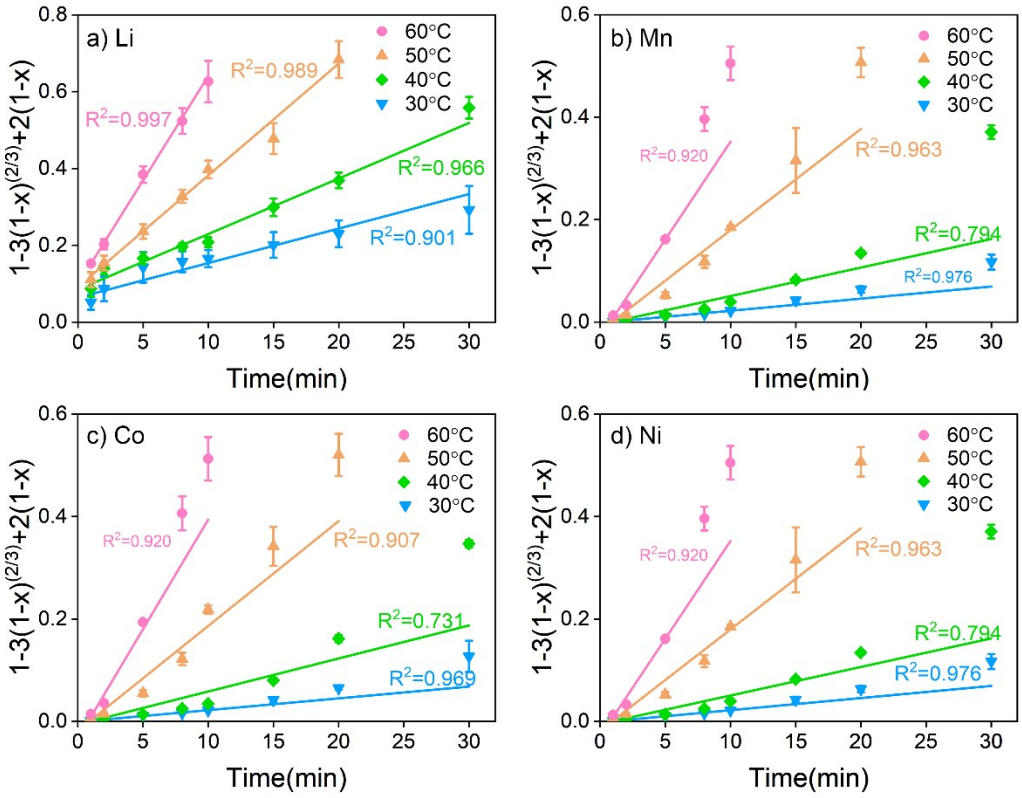

Fig. S8 Fitting results of a) Li, b) Mn, c) Co and d) Ni according to surface diffusion model.

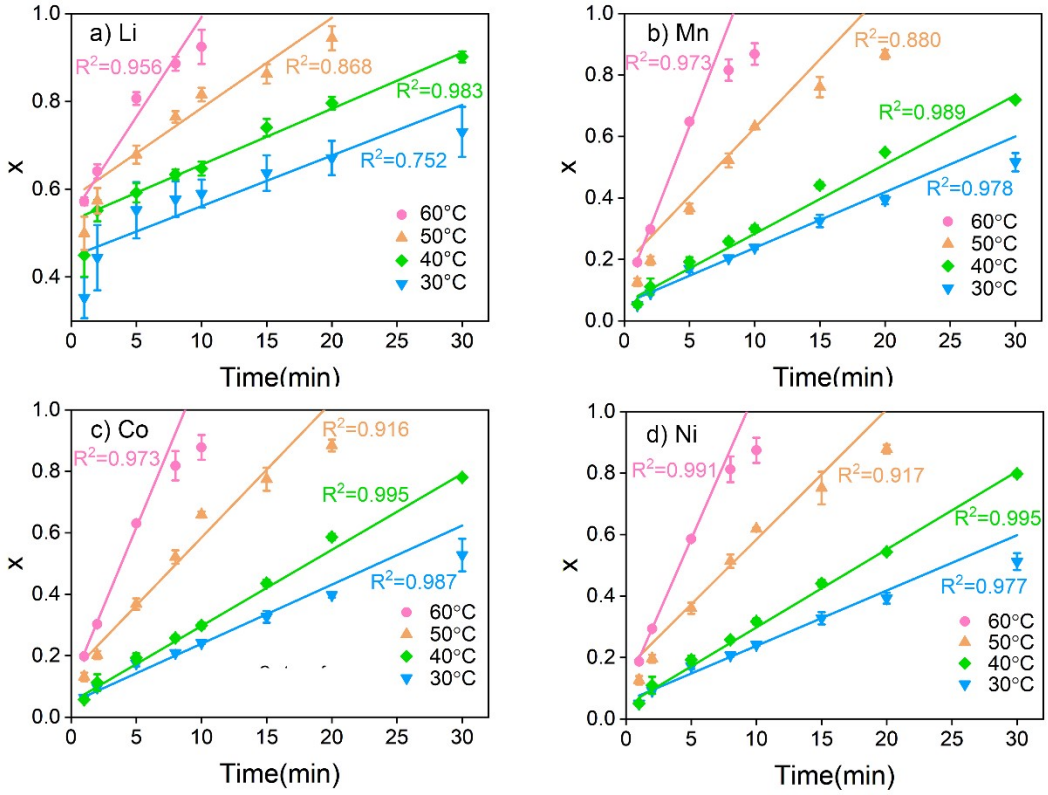

Fig. S9 Fitting results of a) Li, b) Mn, c) Co and d) Ni according to mass transfer model.

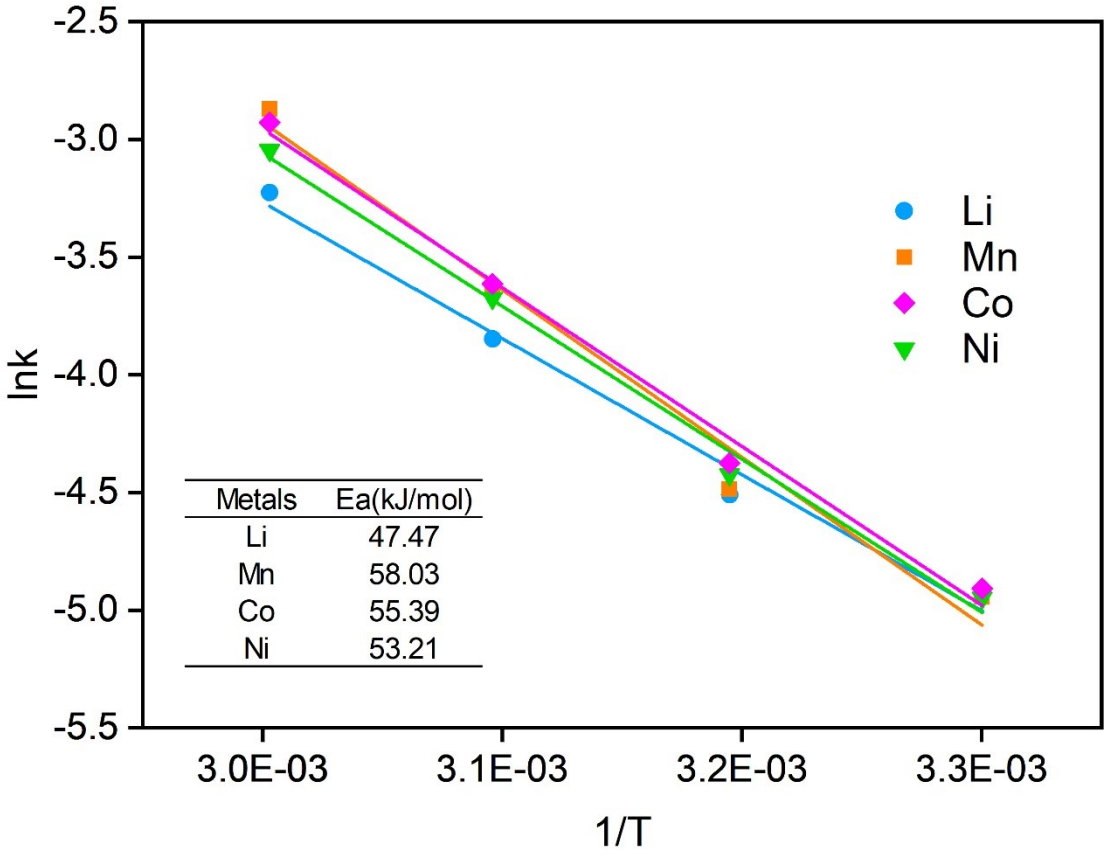

Fig. S10 Activation energy calculation of Li, Mn, Co and Ni according to Arrhenius equation in the range of 30-60°C

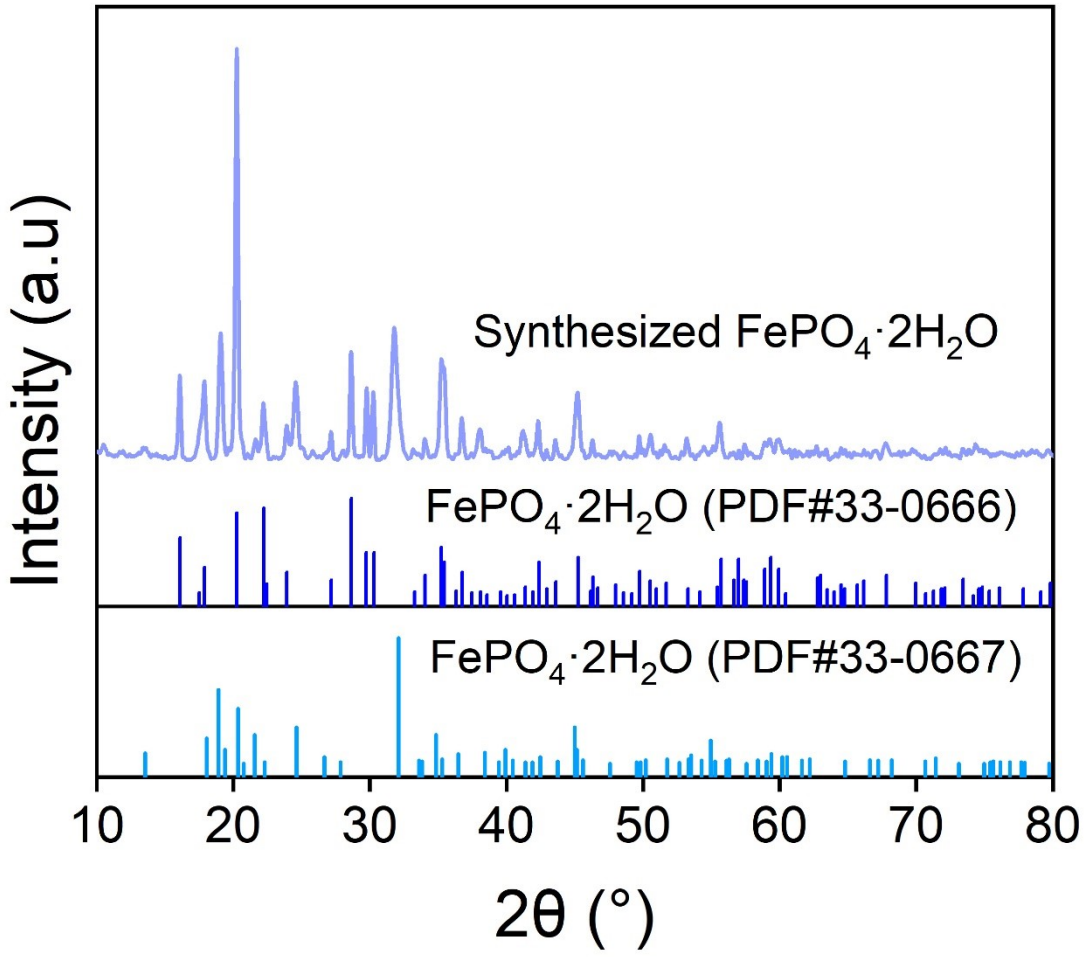

Fig. S11 XRD pattern of recovered $FePO_4 \cdot 2H_2O$ after washing the residue with H_3PO_4+$NH_4H_2PO_4$ at PH=2-2.5

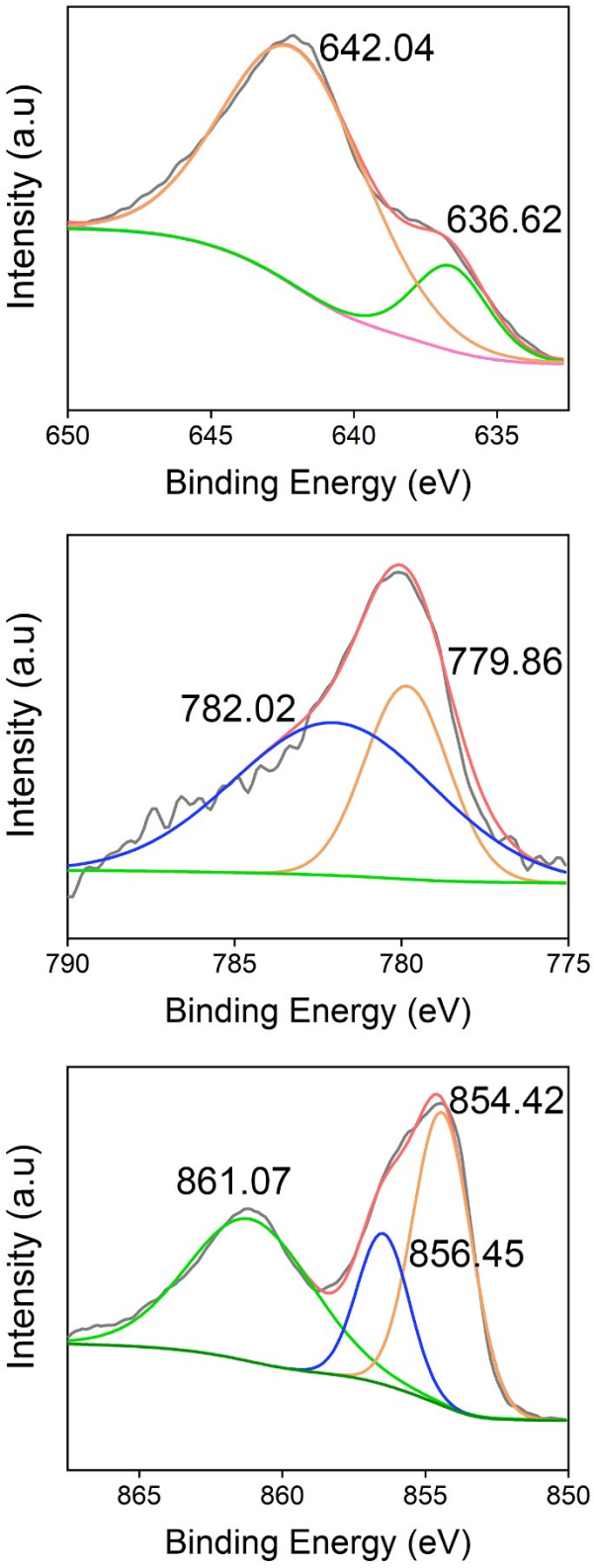

Fig. S12 XPS analysis of recovered MnCoNi oxide after 900°C calcination