Electronic Supplementary Information (ESI)

Liquid-liquid interface-mediated room-temperature synthesis of amorphous NiCo pompoms from ultrathin nanosheets with high catalytic activity for hydrazine oxidation

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Experimental

Synthesis of NiCo alloys

All reagents were of analytic grade and ultrapure water was used throughout the experiments. In a typical synthesis of NiCo alloys with a molar ratio of Ni:Co=1:3 (denoted as Ni_1Co_3), 50 mg of $NiCl_2 \cdot 6H_2O$ and 150 mg of $CoCl_2 \cdot 6H_2O$ were dissolved in 30 mL $CHCl_3$ in a 100 mL flask. 30 mL 0.1 mol L^{-1} $NaBH_4$ solution were slowly added into the above solution. The mixed solution was sealed for 8 h. The black resultant precipitate was collected by centrifugation, washed with ethanol, then washed with deionized water several times and stored in ethanol solution. To obtain

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NiCo alloys of different composition, NiCo alloys with various precursor ratios were listed in Table S1. For comparison, Ni_1Co_3 (Ni:Co=1:3) was also synthesized by the similar process used for preparing Ni_1Co_3 pompoms, in which $CHCl_3$ was replaced by 30 mL of ultra-pure water.

Table S1. Synthesis conditions and the mass of the precursors.

Sample	$NiCl_2 \cdot 6H_2O$ (mg)	$CoCl_2 \cdot 6H_2O$ (mg)	Initial ratio Ni:Co	Actual ratio Ni:Co
Ni_3Co_1	150	50	3:1	2.95:1
Ni_1Co_1	100	100	1:1	1:1
Ni_1Co_3	50	150	1:3	1:3.02
Ni_1Co_5	33.3	166.7	1:5	1:5
Ni_1Co_3 sheets	50	150	1:3	1:3.01

Characterization

XRD patterns were recorded on a Shimadzu XD–3A (Japan) using filtered Cu-Kα radiation ($\lambda = 0.15418$ nm) generated at 40 kV and 30 mA. Scans for 2θ values were recorded at 5° min^{-1}. Scanning electron microscopy (SEM) images were obtained using a JSM-6701F (Japan). Transmission electron micrographs (TEM), high angle annular dark field scanning transmission electron microscopy (HAADF-STEM) images of the catalysts were taken on a JEOL (JEM-2000 FX) microscope operating at 200 kV. Energy dispersive spectroscopy (EDS) in the STEM mode was employed for elemental composition using either individual particles or several particles. The average chemical compositions of the catalysts were obtained using an IRIS

advantage inductively coupled plasma atomic emission spectroscopy (ICP-AES) system (Thermo, America). The specific surface area was determined by Brunauer-Emmett-Teller (BET) method on Quantachrome Autosorb-1 volumetric analyzer.

Electrochemical measurements

Electrochemical measurements were carried out using an electrochemical work station CHI 650D. A common three-electrode electrochemical cell was used at ambient temperature for the measurements. The counter and reference electrodes were a platinum wire and a Hg/HgO reference electrode (KOH 1.0 mol L^{-1}). The working electrode was prepared on a 5-mm diameter glassy carbon disk, 2 mg of catalyst was dispersed ultrasonically in 0.4 mL Nafion/ethanol (0.25% Nafion) solution for 15 min, and 8 μL was pipetted and air dried on the glassy carbon surface. The total metal loading on the electrode was 20.4 mg cm^{-2}. Before each experiment, all solutions were deoxygenated by purging with pure argon for at least 15min and then, potential sweep voltammetry was carried out using a solution without hydrazine until the waves were sufficiently stable for electrode conditioning. All potentials are reported with respect to a reversible RHE.
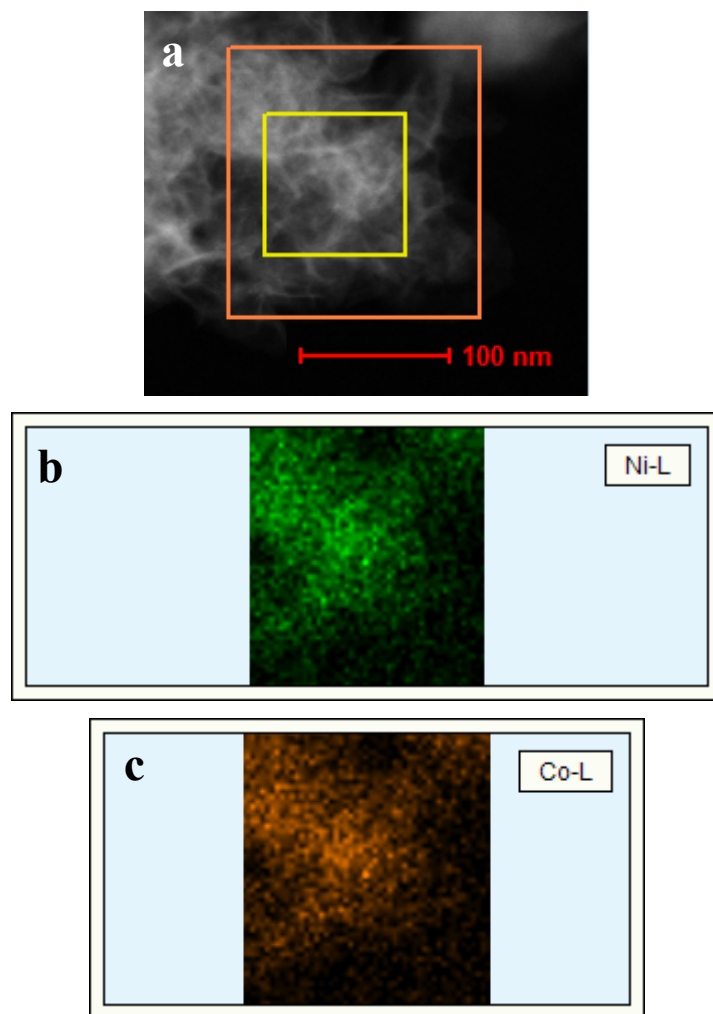
Figure S1. (a) STEM image and the elemental mapping of Ni (b) and Co (c) of the region in yellow box in figure a.
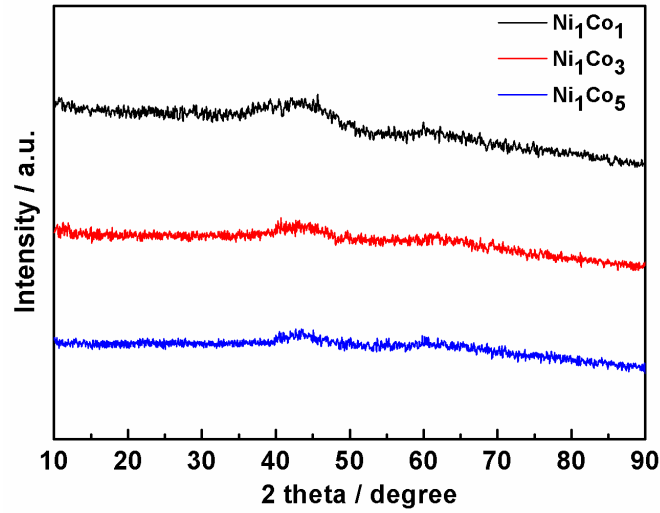
Figure S2. XRD patterns of Ni_1Co_1, Ni_1Co_3, Ni_1Co_5.

For the Ni_1Co_3 sample, a broad weak peak in a wide 2θ range (34 - 48°) appears and no other clear peak can be observed, implying that the structure of Ni_1Co_3 pompoms is amorphous, which is in accordance with the SAED result. From Ni_1Co_5 to Ni_1Co_1, the relative intensity of the broad peak increases, suggesting the degree of crystallinity increases.
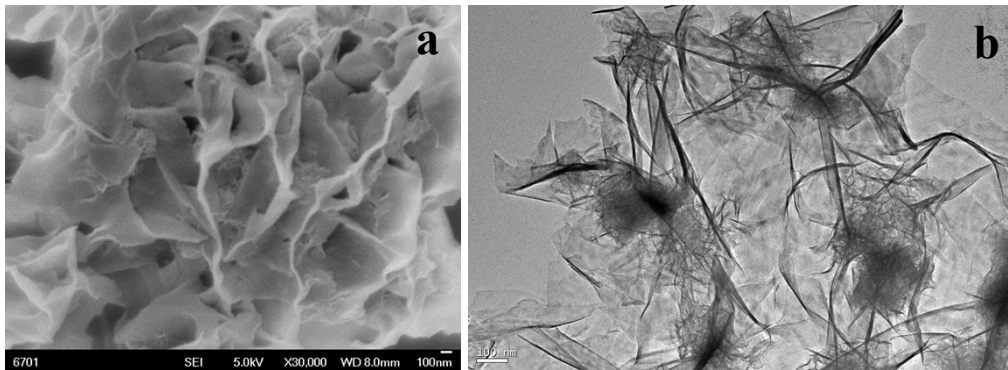

Figure S3. SEM and TEM images of NiCo sheets obtained in one-phase system (ultrapure water).
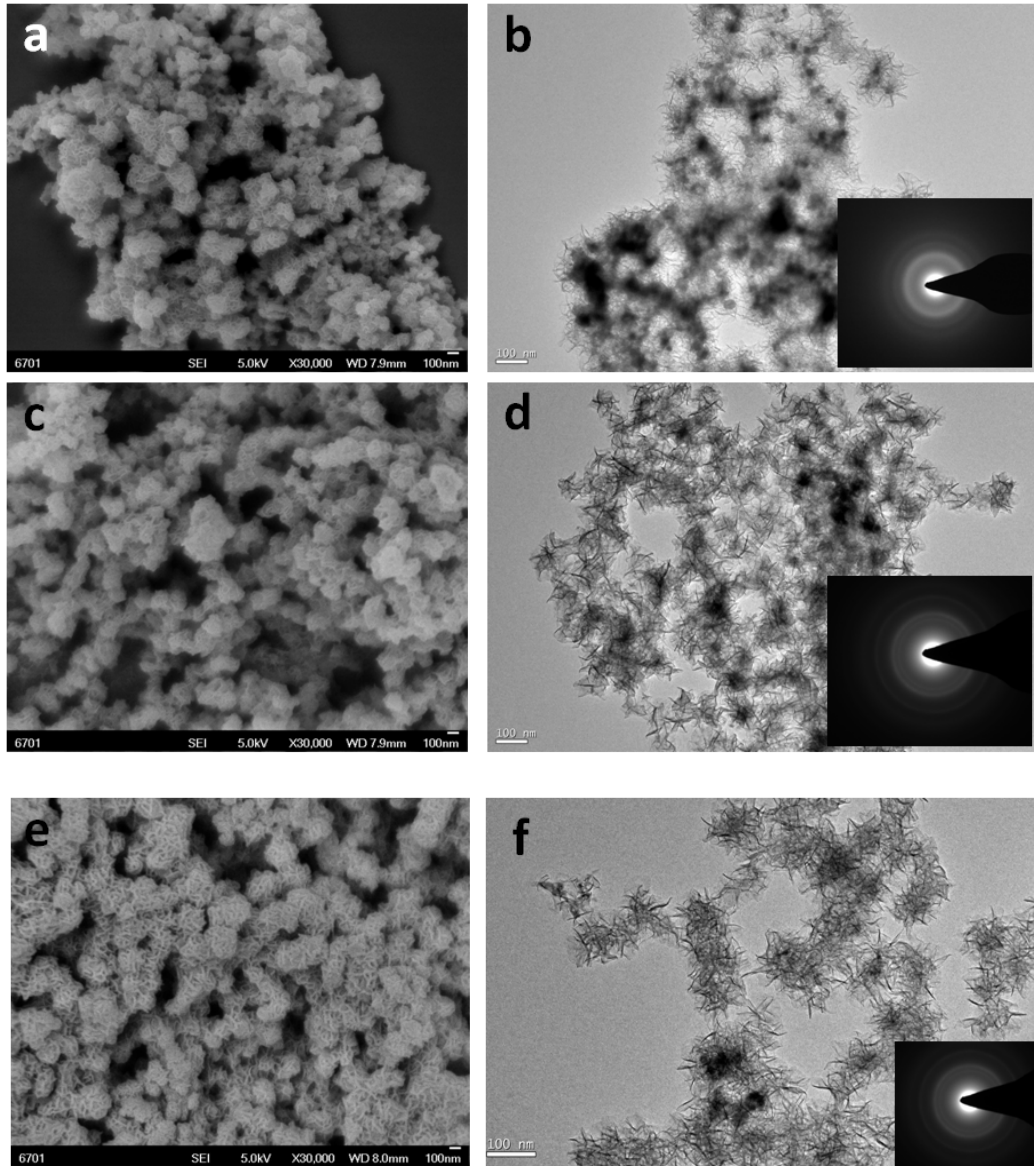
Figure S4. SEM and TEM images of the Ni_3Co_1 (a,b), Ni_1Co_1 (c,d) and Ni_1Co_5 (e,f);

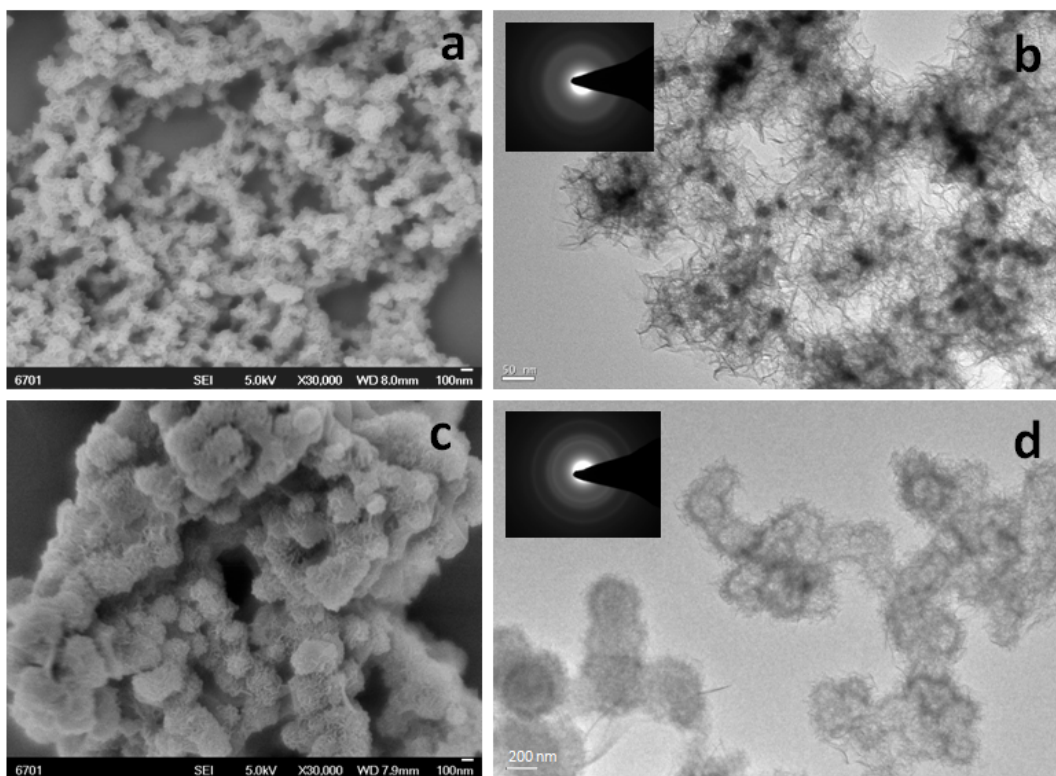Inset: The corresponding SAED pattern of Ni and Co.

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Figure S5. SEM and TEM images of the Ni (a,b) and Co (c,d); Inset: SAED pattern

of Ni and Co.

Table S2. A benchmark of the catalytic activity of as-prepared Ni_1Co_3 pompoms for hydrazine oxidation with the value obtained from some other independent literaures in terms of the current of oxidation peak at the positive scan or/and the onset potential of hydrazine oxidation.

Catalyst	The literature	The current of oxidation peak (mA)	The onset potential of hydrazine oxidation (V vs. RHE)	The concentration of the electrolyte
Ni_1Co_3 pompoms	--	42.7 mA cm^{-2}	-0.19	1.0 mol L^{-1} KOH + 0.1 mol L^{-1} N$_2$H$_4$
Ni/MWNTs-textile	Ref. 6	43 mA cm^{-2}	-	1.0 mol L^{-1} NaOH + 60 mmol L^{-1} N$_2$H$_4$
$Ni_{0.5}Co_{0.5}$	Ref. 8	-	0.12	1.0 mol L^{-1} KOH + 0.1 mol L^{-1} N$_2$H$_4$
Co-CFC	Ref. 33	22 mA cm^{-2}	-0.08[a]	1.0 mol L^{-1} KOH + 20 mmol L^{-1} N$_2$H$_4$

[a] The value was obtained by calculating the given value of -1.1 V vs. Ag/AgCl.
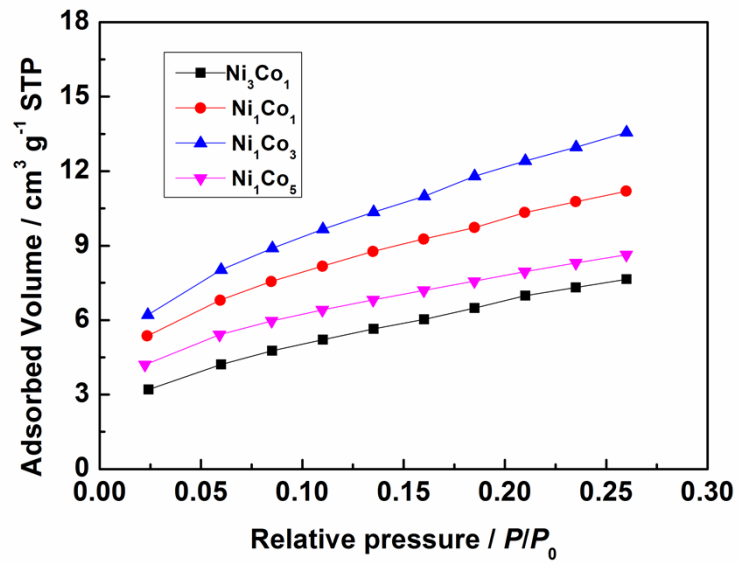
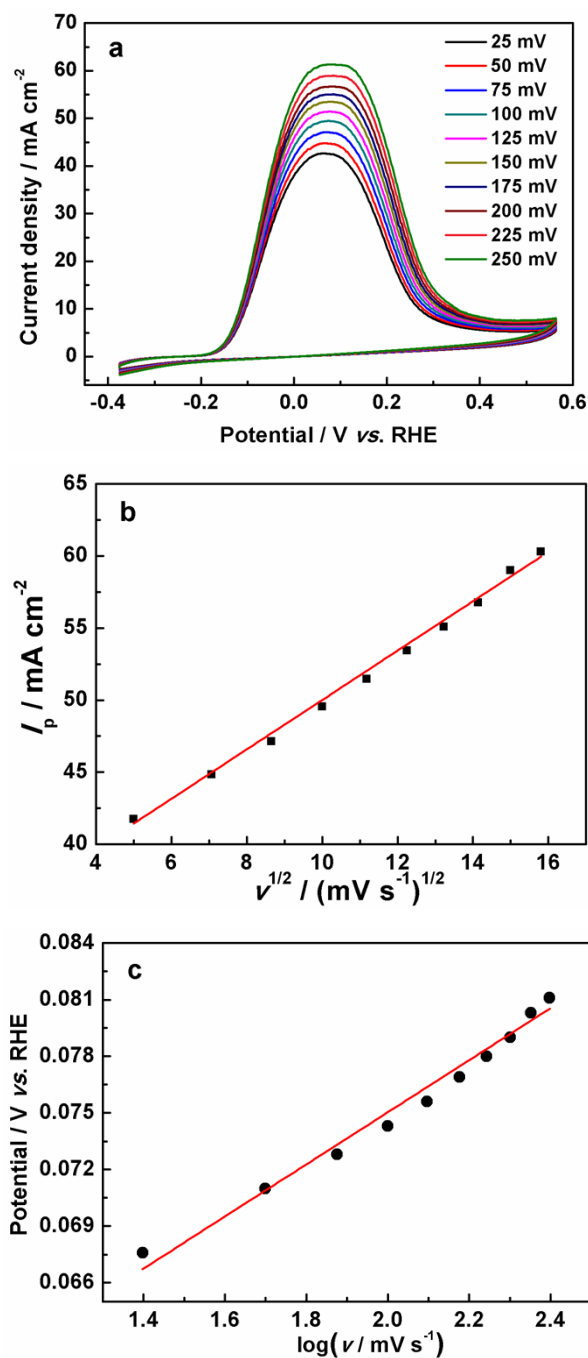**Figure S6.** Nitrogen adsorption isotherm of Ni_3Co_1, Ni_1Co_1, Ni_1Co_3 and Ni_1Co_5.

Figure S7. (c) CVs on the Ni_1Co_3 electrode in 0.1 mol L^{-1} N_2H_4 + 1.0 mol L^{-1} KOH solution at scan rates of 25, 50, 75, 100, 125, 150, 175, 200, 225 and 250 mV s^{-1} (from bottom to top); (b) Ip $vs.$ $v^{1/2}$ plot; (c) Ep $vs.$ log(v) plot.
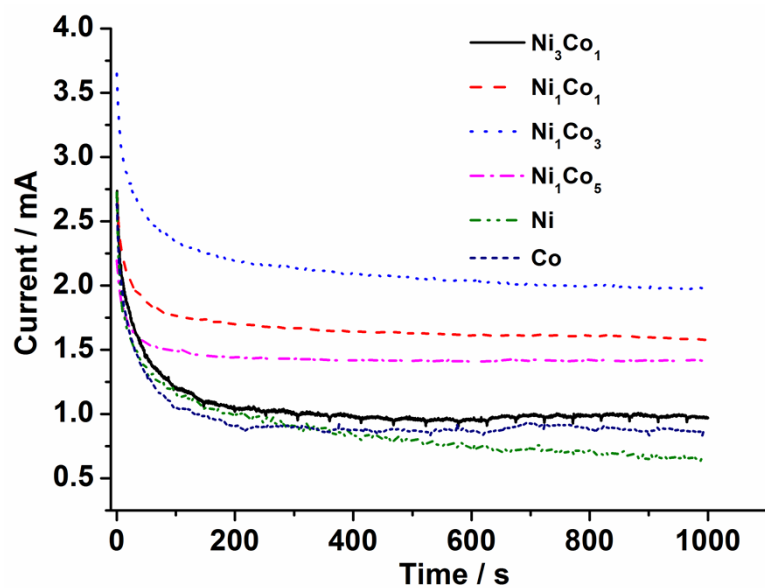
Figure S8. Chronoamperometric curves of Ni, Ni_3Co_1, Ni_1Co_1, Ni_1Co_3, Ni_1Co_5, and Co electrodes in 0.1 mol L^{-1} N_2H_4 + 1.0 mol L^{-1} KOH solution at a rotation rate of 100 rpm, at a constant potential of 0.1 V, and at ambient temperature.
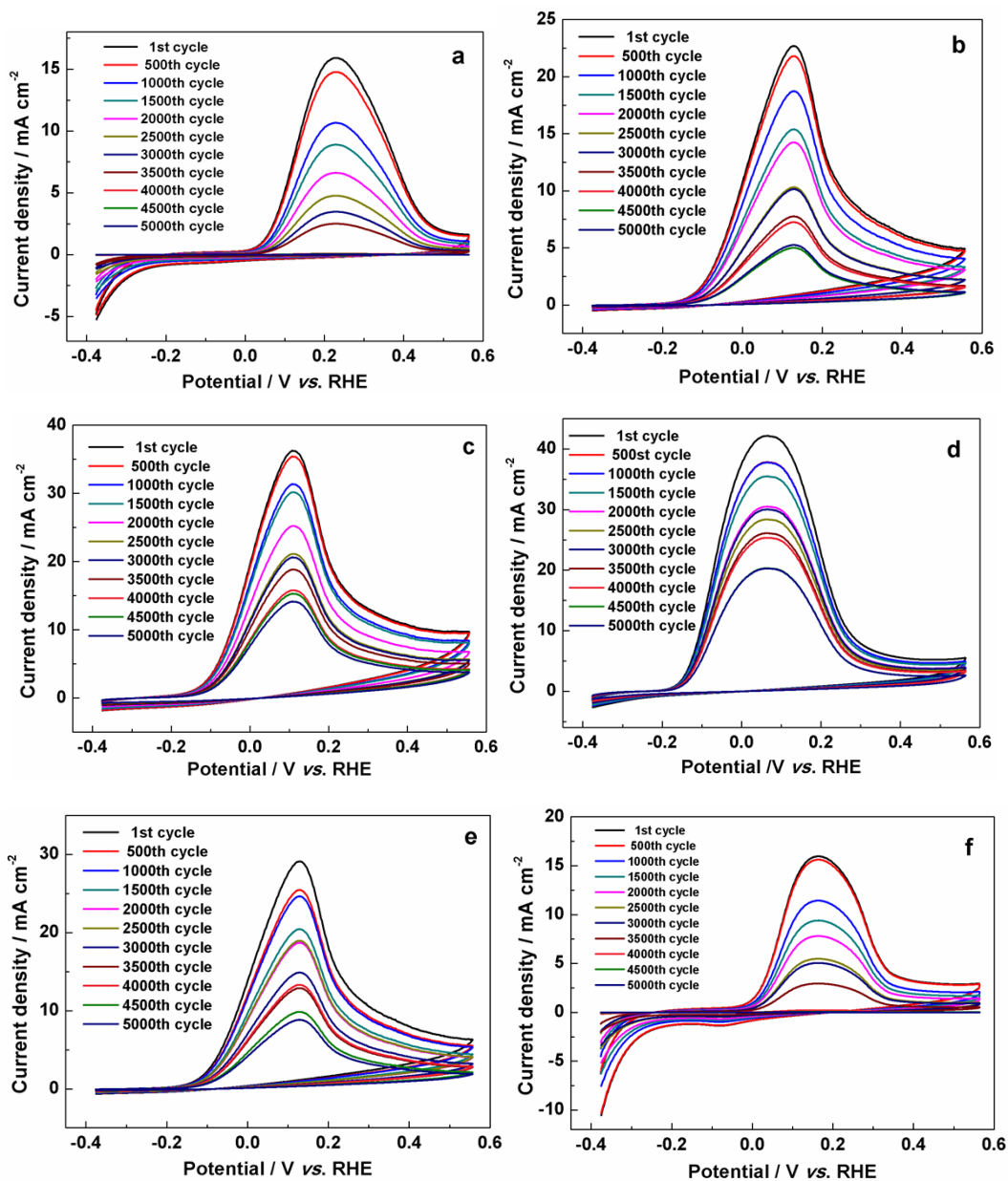
Figure S9. (c) CVs on Ni (a), Ni_3Co_1 (b), Ni_1Co_1 (c), Ni_1Co_3 (d), Ni_1Co_5 (e), and Co (f) electrodes in 0.1 mol L^{-1} N_2H_4 + 1.0 mol L^{-1} KOH solution at a scan rate of 20 mV s^{-1}.