Electronic Supplementary Information

Experimental Section

1. Synthesis

All reagents were of analytic grade, and double-distilled water was used throughout the experiments. To prepare nanostructured Ni film, 2 mmol NiCl₂·6H₂O was dissolved into 20 mL deionized water under intense stirring, followed by addition of 0.5 mL ethylene diamine (ca. 8 mmol) to the NiCl₂ solution. Then, 10 mL NaOH (10M), 10 mL hydrazine hydrate solution (50%) and 40 mL ethanol were dropped in sequence to the above solution. Afterwards, the solution was transferred into a Teflon-lined steel autoclave, in which a piece of 5 cm×12 cm copper foil with a thickness of ca. 10 µm and a weight of 0.54±0.02g (obtained from Guangzhou Great Power Energy &Technology CO.,LTD) closely attached to the inner wall of vessel was used as support. Then the autoclave was put in the center of a circular column magnet with the magnetic field intensity of ca. 0.2 Tesla around the inner wall of the vessel (The reactor equipment will be illustrated in Fig. S2). Finally, the autoclave, together with the magnet, was kept in an air oven at 100 °C for 8 h. After reaction, the whole Cu foil changes from light yellow to gray black in color, suggesting that the Cu support was coated with Ni. The as-prepared nanostructured film was rinsed with double-distilled water and ethanol, and finally dried in vacuum oven at 50 °C for 6 h. The synthetic process of nanostructured Co and CoNi alloy films is similar to the above-mentioned procedure except that some parameters were altered based on our design. To clearly describe our experimental design, some reaction conditions in the synthesis process were listed in Table S1.

Samples	Metal source (mmol)		Complexant/dosage (mmol)	Mass of magnetic layer (g) a	Atomic ratio ^b	Crystallite sizes (nm) ^c
	NiCl ₂	CoCl ₂				
Ni	2	0	Ethylene diamine/8	0.0581	Cu:Ni=1:0.12	29
Co	0	2	Triethylenetetramine/4	0.0750	Cu:Co=1:0.15	25
Co ₅₀ Ni ₅₀	1	1	Ethylene diamine/8	0.0325	Cu:Co:Ni=1:0.032:0.033	22
Co ₇₅ Ni ₂₅	0.5	1.5	EDTA/8	0.0409	Cu:Co:Ni=1:0.060:0.021	16

Table S1. Reaction receipt for the synthesis of nanostructured films and the mass of magnetic layer/atomic ratios of them.

^{*a*} The mass of Co, Ni or CoNi alloy deposited on the Cu substrate was determined by calculating the mass difference of the Cu foil before and after reaction. ^{*b*} Evaluated by ICP-AES analysis. ^{*c*} Estimated by calculation of the width of (002) or (111) diffraction peak in the XRD patterns according to Scherer equation.

2. Characterization

X-Ray diffraction (XRD) patterns were recorded using a Shimadzu XD-3A X-ray diffractometer with Cu K α radiation (λ =1.5406Å). The micromorphology of the film samples were studied using a Hitachi S-4800 field

emission scanning electron microscope (FE-SEM). Transmission electron microscopy (TEM) images were obtained on a JEOL JEM-2100 transmission electron microscope operated at 200 kV. M/H hysteresis loops were recorded with a Quantum Design MPMS XL-7 superconducting quantum interference device (SQUID) magnetometer. Contact angle was measured by a contact angle meter (OCA20, DataPhysics Instruments GmbH, Germany) at ambient temperature. The average of at least five measurements taken on different spots on each film sample was adopted as static contact angle. The average chemical compositions of films were determined using an IRIS advantage inductively coupled plasma atomic emission spectroscopy (ICP-AES) system (Thermo, America).

3. Catalytic performance testing

The catalytic reduction of p-nitrophenol (PNP) to p-aminophenol with sodium borohydride was carried out at 25° C in a glass reactor. In a typical reaction, 8 mL of PNP solution (2×10⁻⁴ M) and 8 mL of NaBH₄ solution (2×10⁻² M) was mixed in the reactor, followed by addition of a piece of 5 cm × 3 cm film tailored from the as-prepared film. The absorbance of the reaction solution was measured at λ =400 nm periodically on a Shimadzu UV-3600 spectrophotometer, from which the catalytic activity was calculated. The reusability of the film catalysts was assessed by repeating the catalytic reaction for 20 times (The reaction time for each catalytic reaction was 20 min). After each cycle, the catalyst was isolated from reaction solution and then washed thoroughly with double-distilled water and dried.

Additional figures



Fig.S1 SEM images of Cu foil substrate (a), Co film (b), Ni film (c), Co₇₅Ni₂₅ film (d) and Co₅₀Ni₅₀ film (e).



Fig.S2 (a) side view and (b) vertical view of the reaction equipment, and (c) the direction of magnetic force exerted on the magnetic bodies in the reaction vessel.

(Blue dotted cylinder denotes the reactor; the red dotted cylinder denotes the copper foil support, which closely clings to the inner wall of the Teflon cup; and the orange cylinder denotes the tube column magnet.)



Fig.S3 SEM images of the powdery samples of Co (a), Ni (b), $Co_{50}Ni_{50}$ (c) and $Co_{25}Ni_{75}$ (d) prepared in the absence of magnetic field.



Fig. S4 Plots of ln(At/A0) versus time for Co (a), Ni (b), Co₅₀Ni₅₀ (c) and Co₂₅Ni₇₅ (d) films.



Fig. S5 Room-temperature magnetic hysteresis loops of Co (a), Ni (b), Co₅₀Ni₅₀ (c) and Co₂₅Ni₇₅ (d) films.



Fig. S6 The saturation magnetization (Ms) of the films at different Co fraction.



Fig.S7 Photographs of water droplet on the surface of Co (a), Ni (b), Co₇₅Ni₂₅ (c), and Co₅₀Ni₅₀ (d) films.