

# Enhanced Magnetization in Highly-Crystalline and Atomically-Mixed bcc Fe-Co Nanoalloys Prepared by Hydrogen Reduction of Oxide Composites

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### 1. Experimental details

**Preparation:** For preparation of a  $\text{Fe}_{50}\text{Co}_{50}$  nanoalloy, 1 mmol of  $\text{Fe}(\text{OAc})_2$  and 1 mmol of  $\text{Co}(\text{OAc})_2$  were taken in 200 mL of triethylene glycol in presence of 40 mmol polymer (monomeric unit). To dissolve the reactants properly the reaction mixture were put in ultrasonic bath for about 30 min at 25°C. Then argon gas was bubbled into the reaction mixture for 30 min while stirred mechanically. 4 ml of aqueous solution of  $\text{NaBH}_4$  (5M) was added into the reaction mixture at temperature 140°C and a black dispersion was obtained. The solution was kept at 180°C for 5 min. After cooling the dispersion to room temperature, the black precipitate was obtained by adding 600 ml mixed solvent of acetone and diethyl ether (1:1) and collected by centrifugation. The precipitate was washed with two kinds of mixed solvents: water and acetone with 1:3 ratio and ethanol and diethyl ether with 1:5 ratio. Due to natural oxidation during washing polymer stabilized mixed oxide ( $\text{Fe}_2\text{O}_3\cdot\text{CoO}$ ) was obtained. Hydrogen reduction of the prepared oxides was conducted with thermogravimetry

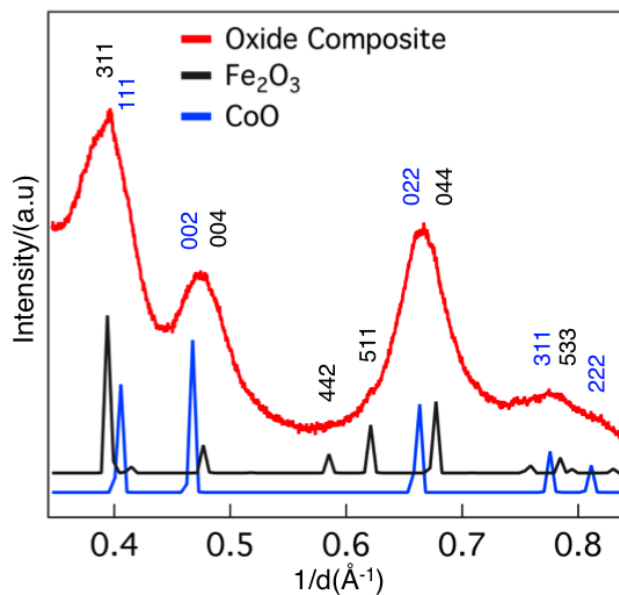
(TG) instrument (Bruker, MTC1000SA) by hydrogen flow at 200 mL/min at different heating conditions.

**Table S1.** Preparation conditions and averaged sizes determined by analyzing XRD patterns of Fe<sub>50</sub>Co<sub>50</sub> nanoalloys.

Polymer type	Temperature/°C	Time/min	Size/nm
PVP K30	450	30	18
PEG 1540	450	30	30
PEG 1540	475	30	55
PVP 1540	500	30	70
PEG 1540	500	30	80

## 2. XRD pattern of oxide composite ( $\text{Fe}_2\text{O}_3\cdot\text{CoO}$ )

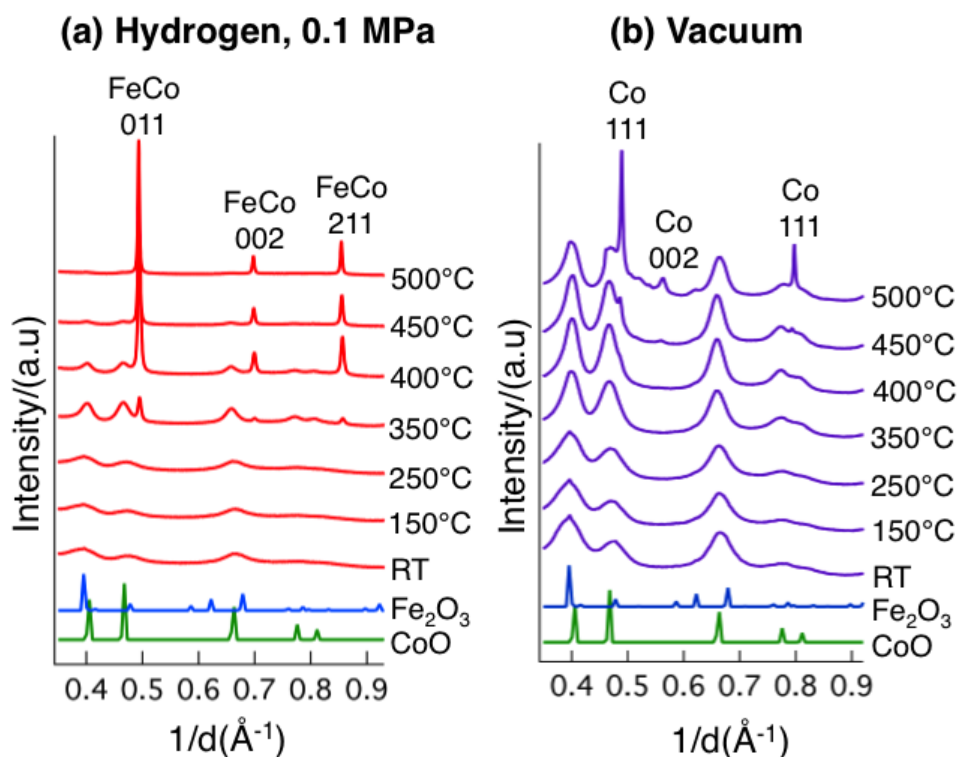
XRD patterns were measured by using synchrotron radiation at  $\lambda = 0.579088(3) \text{ \AA}$ .



**Fig. S1.** XRD pattern of an oxide composite ( $\text{Fe}_2\text{O}_3\cdot\text{CoO}$ ) at Fe:Co=50:50 composition.

### 3. *In-situ* XRD patterns of Fe<sub>50</sub>Co<sub>50</sub>O<sub>y</sub>/PEG

XRD patterns were measured by using synchrotron radiation at  $\lambda = 0.579088(3)$  Å.

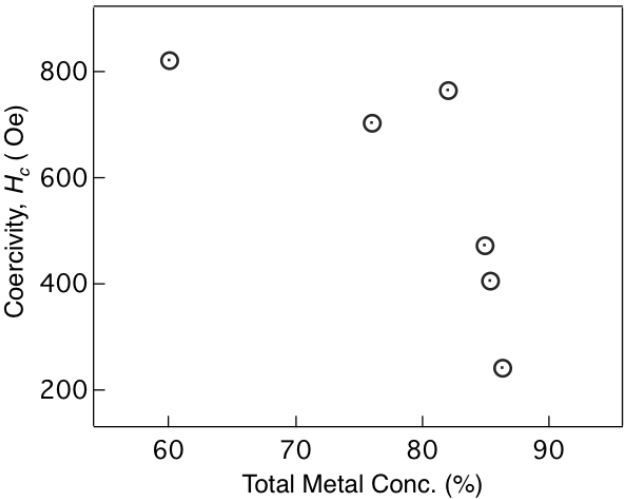


**Fig. S2.** *In-situ* XRD patterns of Fe<sub>50</sub>Co<sub>50</sub>O<sub>y</sub>/PEG during heating under (a) 0.1 MPa hydrogen and (b) vacuum. XRD patterns of Fe<sub>2</sub>O<sub>3</sub> and CoO are indicated at the bottoms.

**Table S2.** Percentages of component solid phases determined by Rietveld analysis of *in-situ* XRD patterns under 0.1 MPa hydrogen at different temperatures.

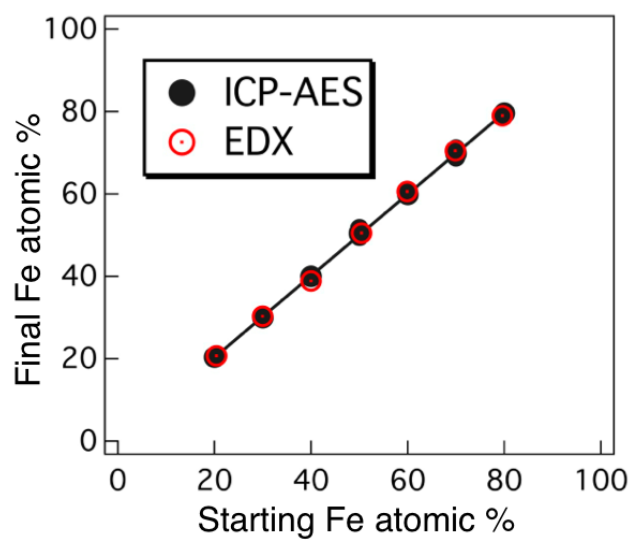
Temp. (°C)	FeCo (wt%)	Fe <sub>2</sub> O <sub>3</sub> (wt%)	FeO (wt%)	CoO (wt%)
350	22	5	35	38
400	78	1	11	10
450	90	0	5	5

**4. Metal concentration dependence on coercivity of Fe<sub>50</sub>Co<sub>50</sub> NAs**



**Fig. S3.** Metal concentration dependence on coercivity of Fe<sub>50</sub>Co<sub>50</sub> NAs.

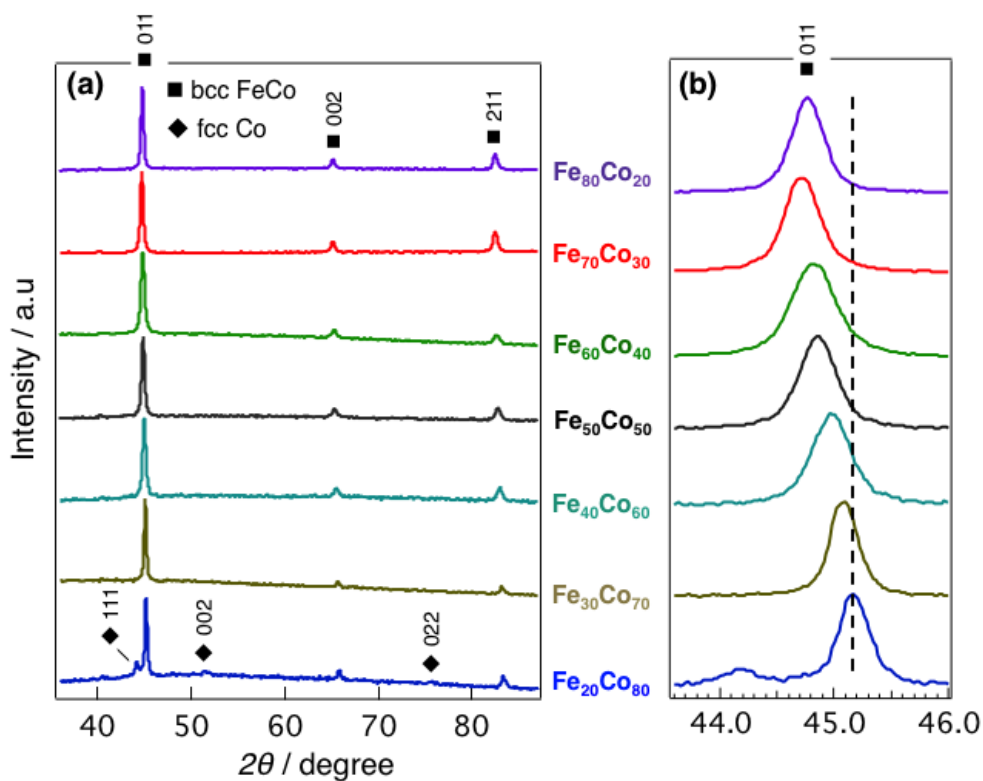
## 5. Relationship between starting and final compositions of $\text{Fe}_x\text{Co}_{100-x}$ NAs



**Fig. S4.** Linearity between starting and final compositions of  $\text{Fe}_x\text{Co}_{100-x}$  NAs.

## 6. Powder XRD patterns of $\text{Fe}_x\text{Co}_{100-x}$ NAs

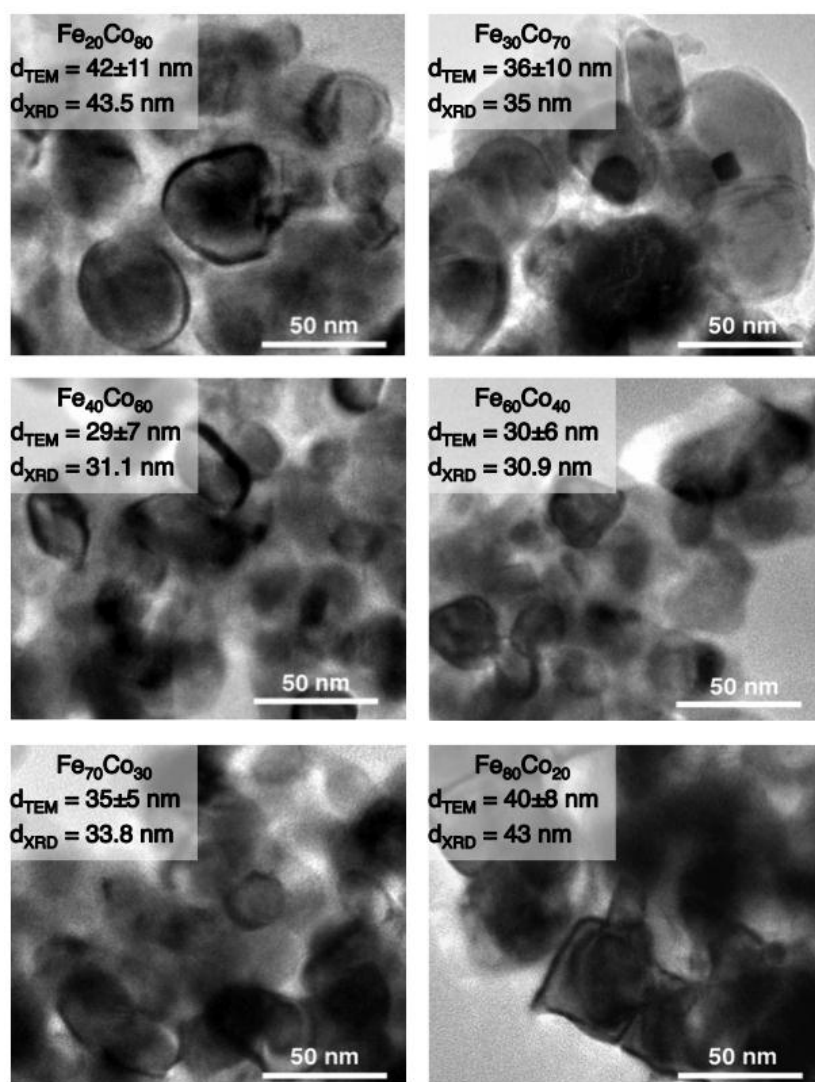
XRD patterns were measured by using Cu  $K\alpha$  radiation.



**Fig. S5.** (a) XRD patterns of  $\text{Fe}_x\text{Co}_{100-x}$  NAs and (b) its expanded view at 011 position.



## 7. TEM images of Fe<sub>50</sub>Co<sub>50</sub> NAs



**Fig. S6.** TEM images of Fe<sub>x</sub>Co<sub>100-x</sub> NAs.

**Table S3.** Diameters of Fe<sub>x</sub>Co<sub>100-x</sub> NPs determined by TEM and XRD analyses.

Sample	Diameter	
	<i>d</i> <sub>TEM</sub> (nm)	<i>d</i> <sub>XRD</sub> (nm)
Fe <sub>20</sub> Co <sub>80</sub>	42 ± 11	44
Fe <sub>30</sub> Co <sub>70</sub>	36 ± 10	35
Fe <sub>40</sub> Co <sub>60</sub>	29 ± 7	31
Fe <sub>50</sub> Co <sub>50</sub>	30 ± 5	32
Fe <sub>60</sub> Co <sub>40</sub>	30 ± 6	31
Fe <sub>70</sub> Co <sub>30</sub>	35 ± 5	34
Fe <sub>80</sub> Co <sub>20</sub>	40 ± 8	43