

## **Chemically synthesizing anisotropic SmCo<sub>5</sub> nanomagnets with a large energy product**

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## Experimental Procedures

**Synthesis of  $\text{Co}_3\text{O}_4@\text{Sm}_2\text{O}_3\text{-CaO}$ :** The precursor was synthesized *via* decomposition of cobalt acetate tetrahydrate ( $\text{Co}(\text{Ac})_2 \cdot 4\text{H}_2\text{O}$ ), samarium nitrate hexahydrate ( $\text{Sm}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ), calcium nitrate tetrahydrate ( $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ ) with Sm : Co molar ratio of 1 : 3.0. In a typical synthesis, 0.5135 g of  $\text{Co}(\text{Ac})_2 \cdot 4\text{H}_2\text{O}$ , 0.3054 g of  $\text{Sm}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$  and 0.5 g of  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$  were dissolved in mixture solvent which includes 50 ml of deionized water, 50 ml of ethanol, and 15 ml dimethyl formamide (DMF). Then, 0.5 g of polyvinylpyrrolidone (PVP) and 1.0 g of citric acid were added into the system to stabilize the solvent. This system was heated to 70 °C to evaporate all solution to obtain the purple powder. The powder was further annealed at 500 °C for 2 h under air at a heating rate of 2 °C·min<sup>-1</sup>. When it was cooled to room temperature, the black  $\text{Co}_3\text{O}_4@\text{Sm}_2\text{O}_3\text{-CaO}$  was collected. For the synthesis of  $\text{Co}_3\text{O}_4\text{-Sm}_2\text{O}_3$  precursor, it is same as with above process except for the addition of  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ .

**Synthesis of  $\text{SmCo}_5$  particles:** For the synthesis of  $\text{SmCo}_5$  particles, previously prepared precursor powders were mixed with 0.6 g of calcium (Ca) powders. Subsequently, the mixture was transferred to a steel crucible which was then moved into a steel tube and degassed three times to remove air and moisture. The tube was flushed with Ar and heated to 900 °C at a rate of 8 °C·min<sup>-1</sup>. The reaction was maintained for 90 min before cooling down to room temperature within 1 h. Afterwards, the sample was washed with  $\text{NH}_4\text{Cl}$  methanol solution in a glovebox under argon atmosphere and collected by a magnet.

**Preparation of aligned  $\text{SmCo}_5$  nanomagnets:** 200 mg  $\text{SmCo}_5$  powders were suspended in 10 ml ethanol, and then were mixed with epoxy quickly. The fluid was placed in a cylindrical mold

under a static magnetic field of 22 kOe. After 6 h solidification, the black bulk was obtained. And  $\Phi 3 \times 3$  samples were cut from the bulk for further measurement.

**Characterization:** The crystallographic structure was identified by X-ray diffraction (XRD, D/MAX 2200 PC) with Cu-K $_{\alpha}$  radiation ( $\lambda=0.15418$  nm). The microstructure and morphology of the above samples were investigated using scanning electron microscopy (SEM, ZEISS - SUPRA55) and transmission electron microscopy (TEM, Tecnai G2 F20). The magnetic properties were measured at room temperature using a Physical Property Measurement System (PPMS) under a maximum applied field of 70 kOe.

## Supplemental Data

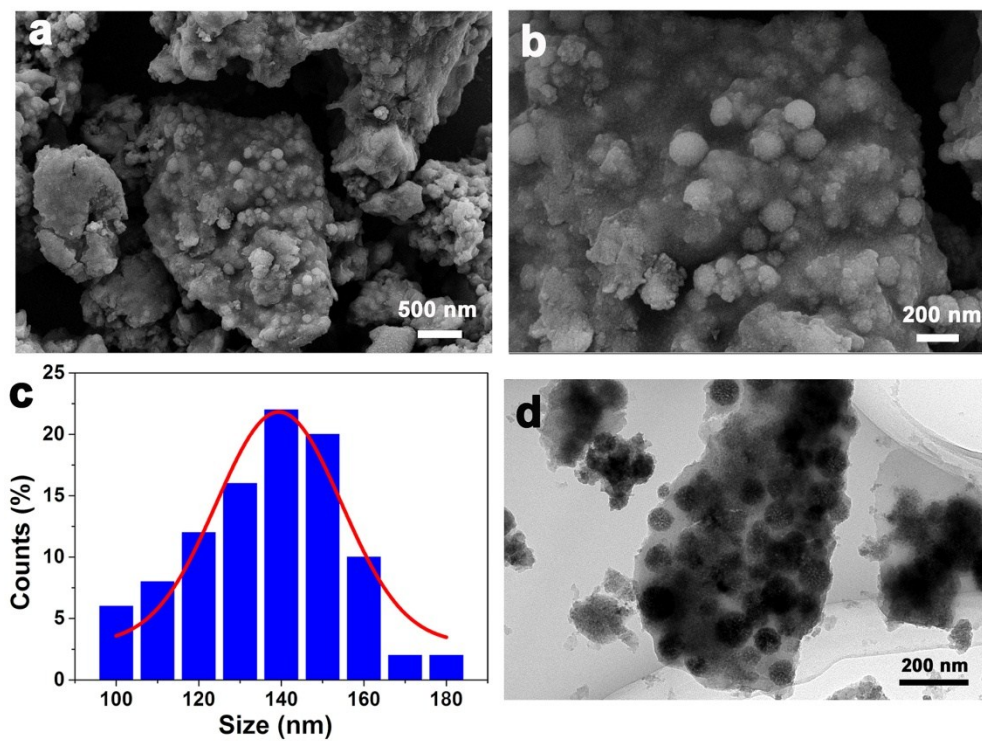


Fig. S1 (a) SEM image of  $\text{Co}_3\text{O}_4@ \text{Sm}_2\text{O}_3\text{-CaO}$ ; (b) an amplified SEM image of (a); (c)  $\text{Co}_3\text{O}_4$  particle size distribution histogram from (a); (d) TEM image of  $\text{Co}_3\text{O}_4@ \text{Sm}_2\text{O}_3\text{-CaO}$ .

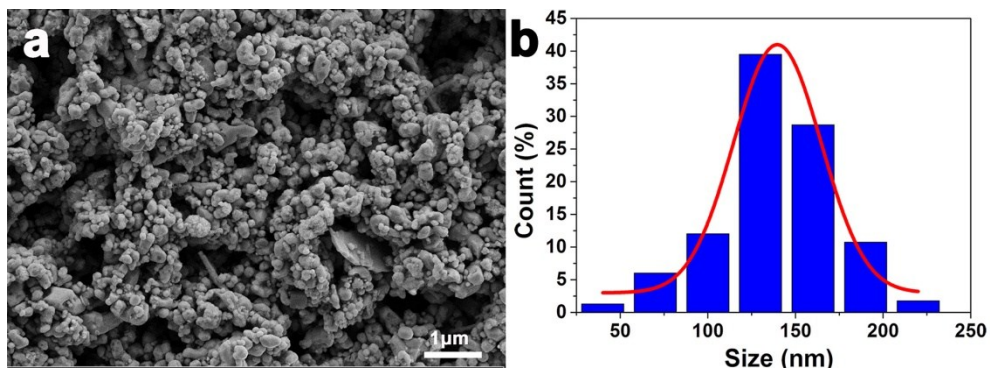


Fig. S2 (a) SEM image of  $\text{SmCo}_5$  particles by reductive annealing of  $\text{Co}_3\text{O}_4@\text{Sm}_2\text{O}_3\text{-CaO}$ ; (b)  $\text{SmCo}_5$  particle size distribution histogram

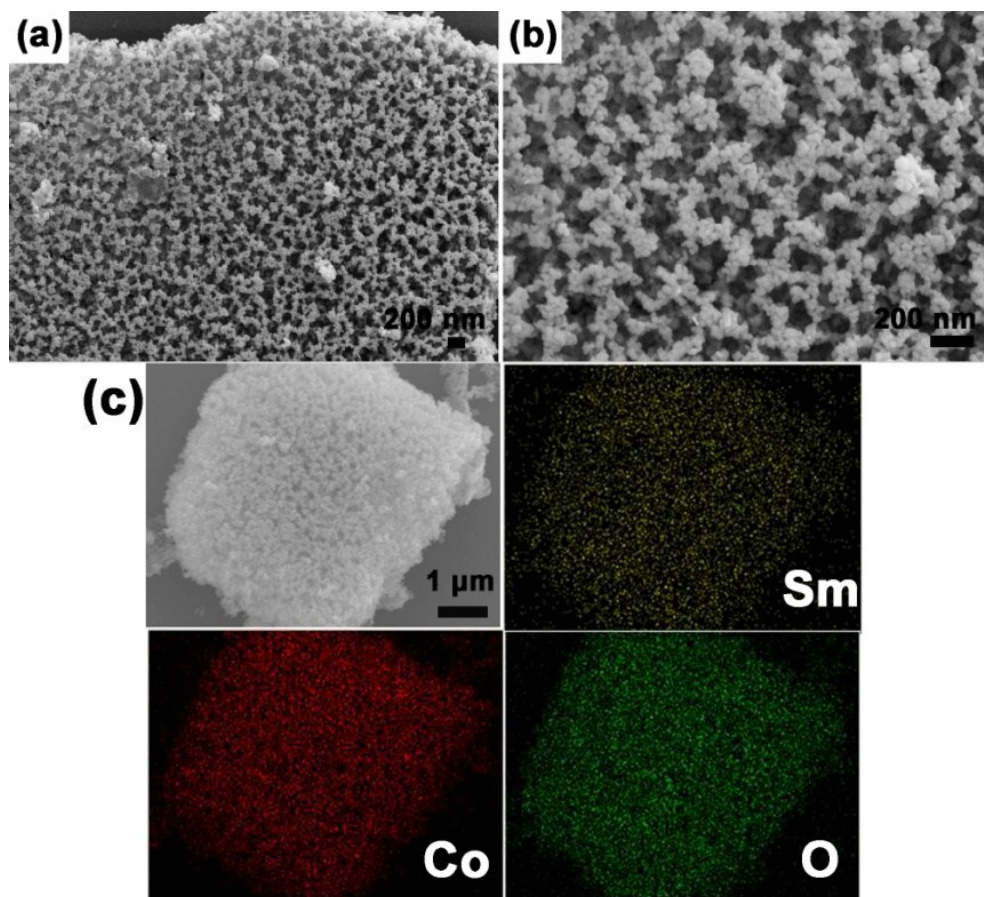


Fig. S3 (a) SEM image of  $\text{Co}_3\text{O}_4\text{-Sm}_2\text{O}_3$  precursor without addition of  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ ; (b) an amplified SEM image of (a); (c) Elemental mapping: Sm (yellow), Co (red) and O (green).

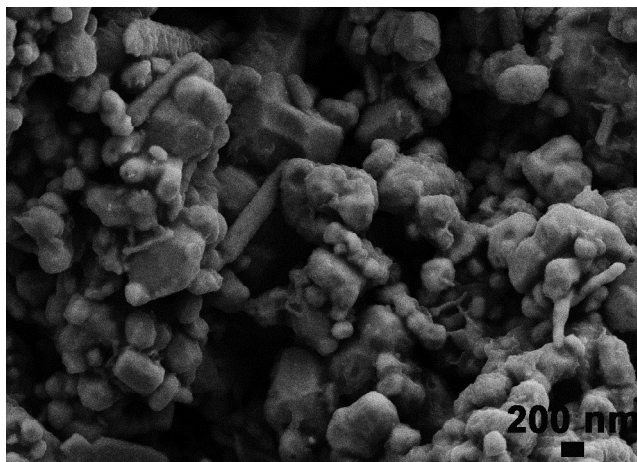


Fig. S4 SEM image of hexagonal SmCo<sub>5</sub> particles prepared by reductive annealing of Co<sub>3</sub>O<sub>4</sub>-Sm<sub>2</sub>O<sub>3</sub> precursor, which exhibit obvious agglomeration and growth without stabilization of CaO.

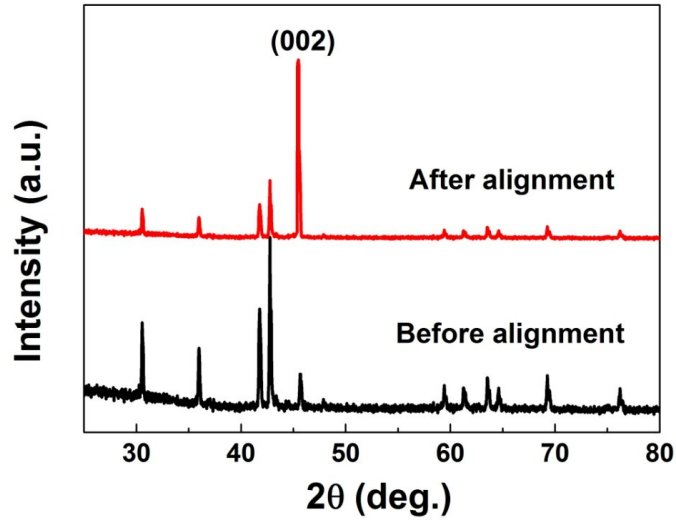


Fig. S5 XRD patterns of SmCo<sub>5</sub> prepared by reductive annealing of Co<sub>3</sub>O<sub>4</sub>-Sm<sub>2</sub>O<sub>3</sub> before alignment (a) and after alignment (b). The aligned particles show an obvious enhancement at (002) diffraction peak, but there is a certain intensity for other peaks, which implies SmCo<sub>5</sub> particles without the protection of CaO represent bonding and are difficult to achieve full orientation.