

SUPPLEMENTARY INFORMATION

Size-dependent magnetophoresis of native single super-paramagnetic nanoparticles in a microchip

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The AVI files show dynamic of native single super-paramagnetic nanoparticles under magnetophoresis (160 nm, Movie S1; 500 nm, Movie S2; model polydispers mixture, Movie S3) in microchip.

Movie S1 Typical real-time dynamics of native single 160 nm SPMNPs in a microchip under magnetophoresis. The distance from the separation region to the permanent magnet was set at 2.0 mm. The 160 nm SPMNPs were suspended in 1.0% PVP solution with 0.015 mg/mL.

Movie S2 Typical real-time dynamics of native single 500 nm SPMNPs in a microchip under magnetophoresis. The distance from the separation region to the permanent magnet was set at 2.0 mm. The 500 nm SPMNPs were suspended in 1.0% PVP solution with 0.015 mg/mL.

Movie S3 Typical real-time dynamics of the polydisperse model (160 nm and 500 nm SPMNPs) in a microchip under magnetophoresis. The distance from the separation region to the permanent magnet was set at 2.5 mm. The polydisperse mixture of 160 nm and 500 nm SPMNPs was prepared by mixing the suspensions of 160 nm and 500 nm SPMNPs at a 1:1 ratio.

Experimental

Chemical and reagents

The Fe₃O₄@SiO₂ core/shell SPMNPs with carboxyl and hydroxyl functional groups (160 nm and 500 nm in diameter) were obtained from Nanobrick (Suwon, Korea) at a 30 mg/mL original concentration. Poly(vinyl pyrrolidone) (PVP, $M_r = 1\,000\,000$) (Polyscience, England) was dissolved in ultra-pure water at 1.0% w/v. Individual 160 nm and 500 nm SPMNP suspension samples were prepared by suspending 160 nm and 500 nm SPMNPs individually in 1.0% PVP solution at 0.015 mg/mL and treated ~30 min by sonication. The rare earth neodymium-iron-boron (NdFeB) permanent magnet coated with nickel and copper (N35, 6 mm length × 4 mm width, × 3 mm thickness) was purchased from G-market in Korea (http://item2.gmarket.co.kr/English/detailview/Item.aspx?goodscode=229974755&pos_shop_cd=RC&pos_class_cd=111111111&pos_class_kind=T).

Characterization of SPMNPs

The powdered SPMNPs were characterized by X-ray diffraction (XRD) using an X-ray diffractometer (D/Max-2400, Rigaku, Japan) equipped with a Cu K α monochromatic radiation source ($\lambda = 0.154187$ nm). The morphologies were observed using a scanning electron microscope (SEM) (S4800, Hitachi, Japan) operating at an accelerating voltage of 15 kV. The size distributions were measured using a dynamic light scattering (DLS) meter

(802DLS, Viscotek, USA). Magnetic properties were measured using a vibrating sample magnetometer (model 7034, Lakeshore, USA).

Lab-made microchip magnetophoresis system

The magnetophoresis system consisted of three main parts: (1) detection based on DIC, (2) a microchip for SPMNPs separation, and (3) a permanent magnet as a magnetic field source (Fig. 1). The DIC system consisted of an inverted microscope (IX-71, Olympus, Japan) equipped with a DIC slider (U-DICT, Olympus, Japan), a 60× objective lens (LCPlanFI 60×/0.70 N.A., W.D. 1.70, Olympus) and a 100× objective lens (UPLANFL 100×/1.3 N.A., W.D. 0.2, Olympus). For real-time single particle detection, a charge-coupled device (CCD) camera (Cascade 512B, Photometrics, USA) was installed on the top of the microscope. The camera exposure time was 10-100 ms. MetaMorph (Version 7.0, Universal Imaging, USA) and Matlab software were used for image acquisition and data processing.

The lab-made glass microchip was used to determine the real-time dynamics of native SPMNPs at the single-molecule level. The 69 mm × 18 mm microchip had a 60 mm long separation channel (5 μm in depth and 100 μm in width) and 4.0 mm long injection channel (ESI†, Fig. S1). The injection design was a double-T channel with 300 μm offset. The reservoirs were 2.0 mm in diameter and 1.0 mm deep. The channel length was 60.0 mm from reservoir 1 to reservoir 3. Before sample injection, the channel was washed with ultra-pure water for ~30 min and then filled with 1.0% PVP solution to suppress adsorption of the SPMNPs.

Magnetic field strength measurement in the microchip

The magnetic field strength of the separation region in the microchip, which was generated by the NdFeB permanent magnet in the magnetic axis direction, was measured by a hand-held Gaussmeter (model 410, LakeShore, USA) (Figs. 2A and 2B). The permanent magnet was immobilized by the holder which was connected to the guide rail. The sensor of the Gaussmeter and the microchip were fixed, while the magnet holder could glide along the guide rail. With the gliding of the magnet holder along the guide rail, variations of the magnetic field strength in the separation region of the microchip were detected by the Gaussmeter.

Velocity measurement of native single-SPMNPs in the microchip

The 1.0 μL SPMNPs suspension (160 nm and 500 nm individually) was loaded in reservoir 4 and driven by applied magnetic force to fill the double-T section of the microchip. Then the magnetic force was applied in the separation channel direction to drive the SPMNPs across the separation channel. The magnetic force in the separation channel was controlled by adjusting the knob of the X-stage, which was linked to the permanent magnet holder. The magnetophoretic velocities of single-SPMNPs were measured by analyzing the continuous images of the detection, which were recorded by the DIC detection system.

Separation of the polydisperse mixture of SPMNPs by microchip magnetophoresis

The polydisperse mixture sample of SPMNPs was prepared by mixing the 160 nm and 500 nm SPMNPs individual suspensions (0.015 mg/mL) at a 1:1 ratio. The polydisperse mixture sample (1.0 μ L) was loaded in reservoir 4 and injected in the double-T region as described above. Then the magnetic force was applied by setting the permanent magnet at a distance of 2.5 mm in the separation channel direction. Separation images were recorded by a DIC detection system.

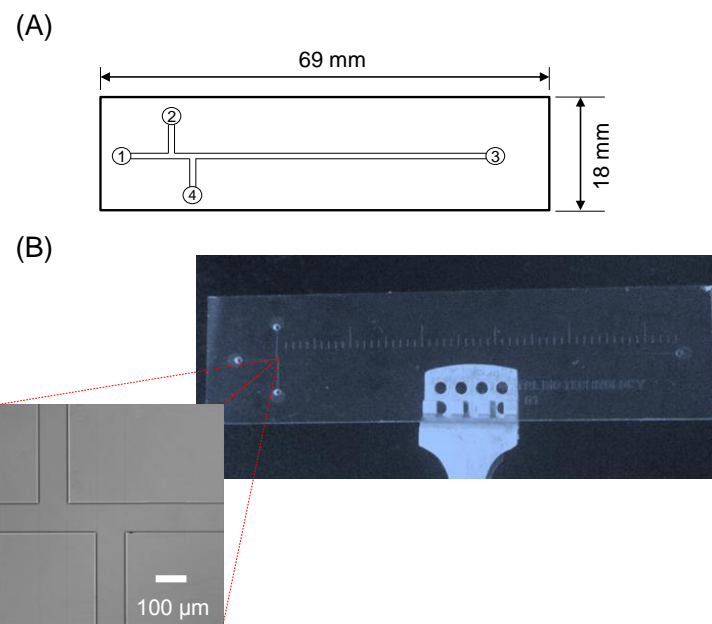


Fig. S1 (A) Schematic diagram of the glass microchip used for microchip magnetophoresis of native SPMNPs. (B) Photograph of the entire glass microchip, and DIC images of the double-T injection region of the microchip.

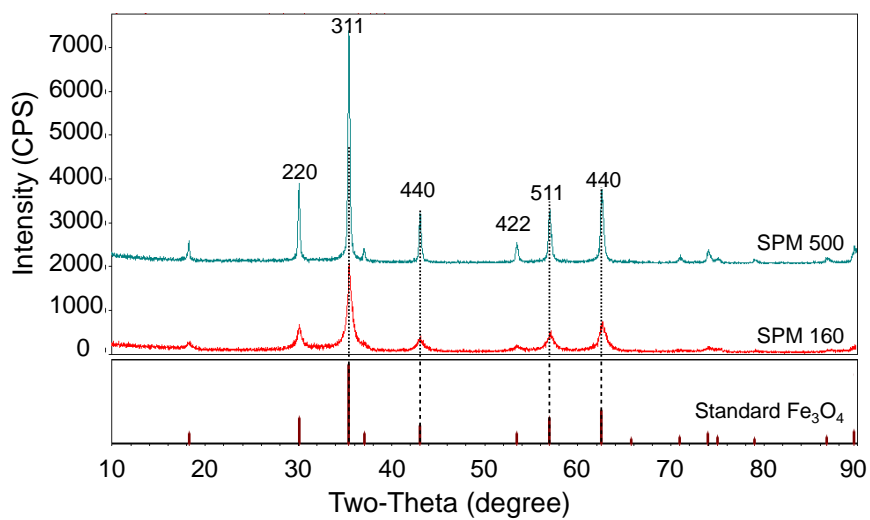


Fig. S2 XRD patterns of 160 nm and 500 nm SPMNPs. All the peaks are in conformity with the standard Fe₃O₄ crystal. No diffraction peaks corresponding to SiO₂ are observed because the SiO₂ is amorphous.

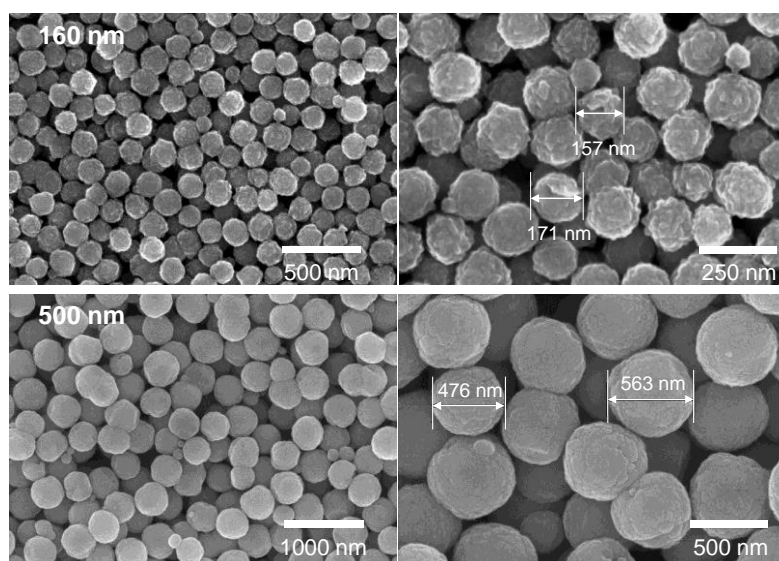


Fig. S3 Scanning electron microscope (SEM) images of 160 nm and 500 nm SPMNPs. These images confirm the spherical geometry of powdered SPMNPs and average size distribution of 160 nm and 500 nm.

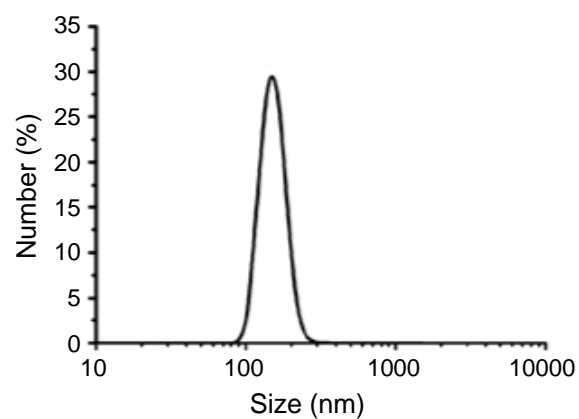


Fig. S4 The dynamic light scattering (DLS) pattern of 160 nm SPMNPs with the carboxyl functional group. The particle diameter is 260.3 nm on average. Due to the hydration effect, the particle size value obtained from DLS was larger than that of SEM. The 45.13 nm peak width indicates the size distribution.

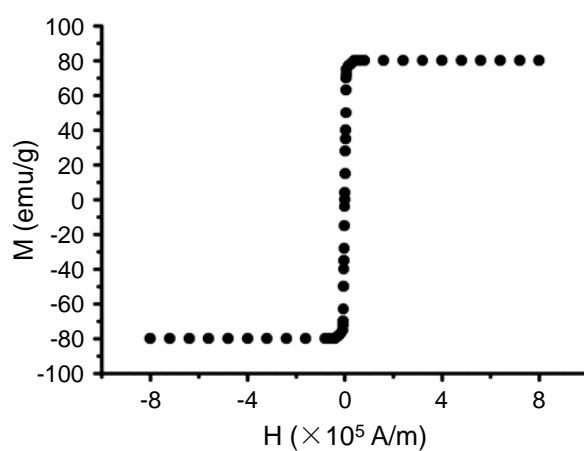


Fig. S5 The magnetization property of 160 nm SPMNPs at 298 K was measured by a vibrating sample magnetometer with the applied magnetic field between -8.0×10^5 and $+8.0 \times 10^5$ A/m. Absence of an open loop in the M-H curve indicates the excellent super-paramagnetic behavior of the SPMNPs.

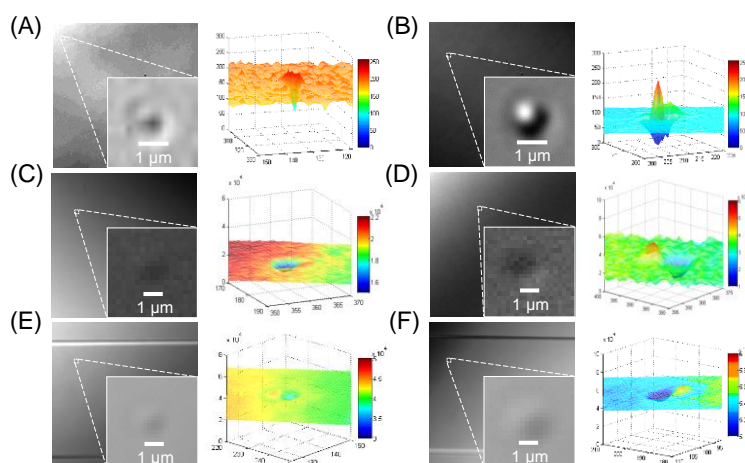


Fig. S6 The typical DIC images of 160 nm and 500 nm SPMNPs. Conditions: (A) 160 nm and (B) 500 nm SPMNPs on glass plate with 100× objective lens; (C) 160 nm and (D) 500 nm SPMNPs on glass plate with 60× objective lens; (E) 160 nm and (F) 500 nm SPMNPs in microchip with 60× objective lens. The 3-D diagram shows the DIC intensity. Objective lens, Olympus UPLANFL 100×/1.3 N.A., W.D. 0.2 and Olympus LCPlanFI 60×/0.70 N.A., W.D. 1.70; camera, Cascade 512B CCD.

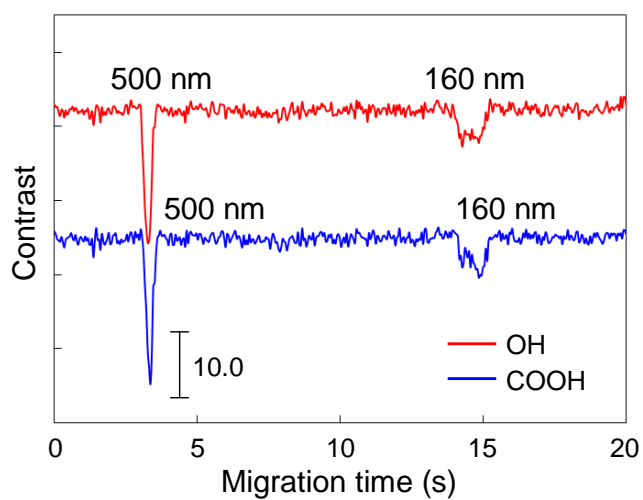


Fig. S7 Representative magnetopherograms of the polydisperse SPMNPs mixture (160 nm and 500 nm) with different functional groups (-OH and -COOH) in microchip with the DIC detection system.