Papilla-like Magnetic Particles with Hierarchical Structure
for Oil Removal from Water

By Li-Ping Xu, Xiwen Wu, Jingxin Meng, Jitao Peng, Yongqiang Wen,
Xueji Zhang, and Shutao Wang

Research Center for Bioengineering & Sensing Technology, University of Science
and Technology Beijing, Beijing 100083, China.

Beijing National Laboratory for Molecular Sciences (BNLMS), Institute of
Chemistry, Chinese Academy of Sciences, Beijing 100190, China. E-mail:
stwang@iccas.ac.cn.
Experimental Section

Materials: Iron microparticles and oil red were purchased from Alfa-Aesar. Corn oil (JinLongYu) was purchased from supermarket. All other reagents and solvents were of analytical reagent grade and obtained from Sinopharm Chemical Reagent co. Ltd, China. They were used without further purification. All solutions were prepared with ultrapure water (Milli-Q, 18.2MΩcm).

Fabrication of LA-papilla-like particles: Lotus seeds inspired papilla-like particles were fabricated via the oxidation of iron microparticles by being heated at 400 °C for 4 hr. After thermal treatment, the iron microparticles were oxidized into papilla-like particles with nanoflakes. Then the papilla-like particles were rinsed with alcohol for three times. After rinsing, the papilla-like particles were immersed into 1 mM lauric acid in ethanol for 8 hr, followed with rinsing with ethanol for three times to remove free lauric acid and drying.

A field-emission scanning electron microscope (JSM-6700F, Japan) was used for characterizing the morphologies of the as-prepared film. X-ray photoelectron spectroscopy (XPS) data were obtained with an AXIA ULTRA DLD electron spectrometer (Kratos, UK) equipped with Al Kα monochromatized radiation. The X-ray diffraction was performed on a Rigaku D/max-2500 diffractometer using Cu Kα radiation.

Contact angles (CA) were measured on an OCA20 system (Data-Physics, Germany) at ambient temperature. The particles were deposited on a filter paper by filtration and dry. Enough particles were used to form a dense pack to eliminate the influence of the
substrate. The water droplets with 2 µL in volume were dropped onto the surface. For oil CA measurement, the corn oil droplets were syringed out and dropped carefully onto the surfaces, which were immersed in water. The average CA values were obtained by measuring at three different points on the five different samples. Corn oil was chose to assess the oil absorbing capacity of LA-papilla-like particles and Fe microparticles. A permanent magnet was used to track and remove those oil absorbents.
Figure S1 a) XRD patterns of papilla-like particles and Fe microparticles and XPS profiles of b) C 1s region, c) Fe 2p region and d) O 1s region for LA-papilla-like particles.

The obtained papilla-like particles were characterized by XRD and XPS. As shown in Fig. S1a, the XRD spectrum of the Fe microparticles is identified as that of body centered cubic (bcc) structure in α-Fe (Figure S1a, JCPDS # 65-4899). The spectrum of papilla-like particles is more complicated. Besides the peaks corresponding to α-Fe, more peaks were observed, which can be ascribe to Fe$_2$O$_3$ (JCPSD # 33-0664) and Fe$_3$O$_4$ (JCPSD # 65-3107). Above discussion indicates that the material is
composed of Fe$_3$O$_4$, α-Fe$_2$O$_3$, and α-Fe. Fe is the core of the sphere and the presence of Fe$_3$O$_4$ was probably due to the incomplete oxidation of Fe powder. XPS was used to characterize the surface states of LA-papilla-like particles. The C 1s spectrum (Fig. S1b) showed two peaks located at 284.8 eV and 288.5 eV. The 288.5 eV signal is assigned to the carbon of coordinated COO group, which indicates lauric acid was absorbed on papilla-like particles successfully via C-O bond. The Fe 2p spectrum (Fig. S1c) showed three peaks located at 710.68 eV, 718.89 eV and 724.24 eV, which can be attributed to Fe 2P3/2 of the α-Fe$_2$O$_3$, satellite α-Fe$_2$O$_3$ and Fe 2P1/2 of the α-Fe$_2$O$_3$ respectively. A high-resolution O 1s XPS spectrum of LA-papilla-like particles (Figure S1d) can be resolved into peaks located at 529.68 and 531.39 eV. The former can be attributed to oxygen in the lattice, i.e., oxygen atoms that were bound to only iron atoms (Fe–O), and the latter can be ascribed to oxygen atoms on the surface, i.e., oxygen atoms in surface hydroxyl groups (H–O). The peak intensity of Fe–O species was significantly higher than that of H–O, confirming that there were many most carboxyl group have been attached to α-Fe$_2$O$_3$. 

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Figure S2 Wetting properties of LA-Fe microparticles. a) Image of a water droplet sitting on LA-Fe microparticles film; b) a corn oil droplet on LA-Fe microparticles film.