Electronic Supplementary Material (ESI) for Soft Matter.

Supporting information for

Self-assembly of Anisotropic Red Blood Cell (RBC)-like colloidal Particles

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Experimental Section

Materials

The RBC-like particles were synthesized according to our previous work[1, 2]. Sodium dodecyl sulfate (SDS, Guangdong Guanghua Science and Technology Ltd.) was used as surfactant. Anhydrous ethanol (Guangdong Guanghua Science and Technology Ltd.) was used as solvent. Glass slide (2 cm*8 cm) and Petri dish (D = 6 cm) were soaked in freshly prepared piranha solution (18 M H_2SO_4 and 30 wt% H_2O_2 solutions in a 2 : 1 v/v mixture) for 10 min, rinsed several times with water, and dried under nitrogen. Deionized water (18.2 MΩ) was employed in all experiments.

Preparation of 2D and 3D colloidal crystals under convection and capillary force

First of all, 0.5wt% of SDS aqueous solution, particles suspension of different concentration was prepared and sonicated for 5 minutes. Subsequently, 5 ml deionized water was dropped into a clean Petri dish, and then the suspension was dropped on the water surface carefully. Since the effective density of RBC-like polystyrene particles was lower than that of water, the RBC-like particles were capable of floating on water surface. Followed by several drops of SDS surfactant solution, a dense white film was formed at the air-water surface. Finally, with the evaporation of water, self-assembled colloidal crystals films were obtained on the bottom of Petri dish. 3D colloidal crystals were fabricated by repeat above process several times.

Preparation of binary colloidal crystals by vertical deposition technique

Polystyrene spheres with 280 nm were synthesized by soap-free emulsion polymerization[3]. Then the colloidal particles suspension was diluted to desired volume fractions without any purification (0.1 wt%, 0.2 wt%, 0.3 wt%, 0.4 wt%). 100 ml of particles suspension were added into a clean beaker and sonicated

for 5 minutes. A large area highly ordered colloidal crystals film based on above optimized conditions was immersed into the above beaker and fixed with tape. Subsequently, the beaker and the RBC-like colloidal crystals film was putted into the oven at 60 ℃. When water evaporated completely, the self-assembly of binary colloidal crystals were fabricated ultimately.

Characterization of RBC-like particles and multilayer colloidal crystals film

Morphology of RBC-like particles and multilayer colloidal crystals films were sputtered with platinum and characterized in details by field-emission scanning microscope (FE-SEM, ZEISS ULTRA 55). Transmission electron microscope (TEM, HITACHI H-7650) was running at 80 kV to further confirm its structure. Atomic force microscope (AFM) was running by the Multimode 8, Bruke, Germany. The fast Fourier transform (FFT) was executed by Matlab software to illustrate the symmetry and periodicity of colloidal crystals. Optical photographs of the as-prepared monolayer colloidal crystals films were taken by a digital camera (Canon, 1300D).

Based on optimal conditions (the concentration of particles equals to 1.5 wt%, the mass of 0.5 wt% surfactant suspensions equal to 0.05 g and the proportion of anhydrous ethanol is 60 wt%), the highly ordered anisotropic colloidal crystals based on our RBC-like particles were fabricated. Figure S2 shows the microstructure of highly ordered colloidal crystals film obtained on glass slides.

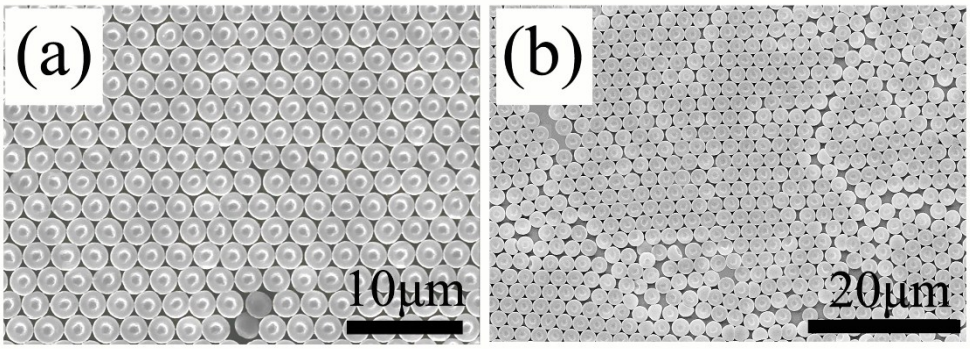

Figure S1. SEM images of well-ordered colloidal crystals based on above optimal conditions.

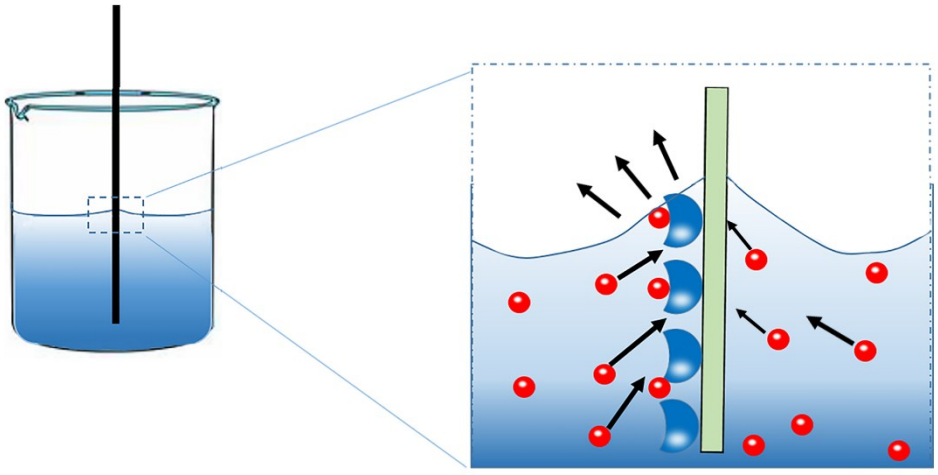

Figure S2. Schematic of the self-assembly of binary colloidal crystals via vertical deposition technique.

References:

1 D. Xie, X. Ren, Y. Xie, X. Zhang, S. Liao, *ACS Macro. Lett.* **2016**, *5*, 174.

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