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## **Dual-Responsive Colloidosome-like Microgels as the Building Blocks for Phase Inversion of Pickering Emulsions**

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## 1. Materials

Ludox TMA colloidal silica (34 wt% suspensions in H<sub>2</sub>O) was purchased from Sigma-Aldrich. Methacryloxyethyl thiocarbamoyl rhodamine B (PolyFluor 570) was obtained from Polysciences. N, N'-methylene bisacrylamide (BIS) (99%), Isobutyl(trimethoxy)silane(97%), and Perylene were bought from Aladdin. N-Isopropyl acrylamide (NIPAM) was purchased from TCI. Absolute ethanol and Fluorescein sodium was provided by Titan Scientific Co., Ltd. (Shanghai, China). Potassium persulfate (KPS), Methylacrylic acid (MAA), Toluene, and Sodium dodecyl sulfate (SDS) were bought from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Ammonium hydroxide solution (25-28%) was supplied by Shanghai Macklin Biochemical Co., Ltd. Deionized water was used for all experiments.

## 2. Characterizations

The morphology of the modified hydrophobic silica, the microstructure of the colloidosome-like microgels at different pH values and temperature were observed by scanning electron microscopy (SEM; Hitachi S-4800). The wettability was measured by a contact angle analyzer (Dataphysics OCA15EC) in air. The prepared emulsions were observed by optical microscopy (Nikon Ni-U) and confocal laser scanning microscopy (Nikon AX equipped with Eclipse Ti2 body), the excited wavelengths for Perylene, Fluorescein disodium salt, and PolyFluor 570 are 405 nm, 488 nm, and 561 nm, respectively.

## **Results and Discussion**

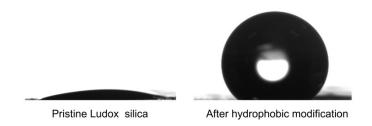


Fig. S1 Contact angle measurementes of the pristine Ludox silica and the modified silica.

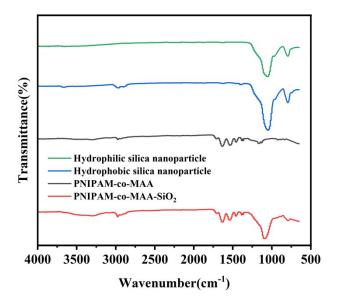


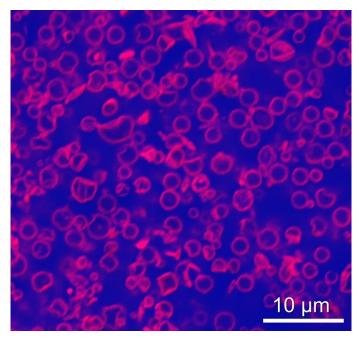
Fig. S2 FTIR analysis of the silica naoparticles, PNIPAM-co-MAA polymer, and the silica coated colloidosome-like microgel.



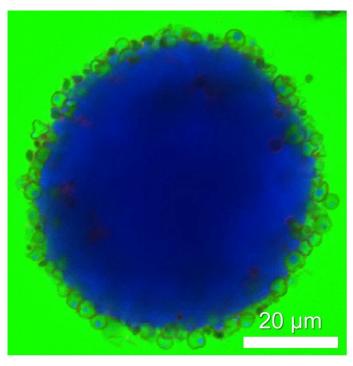
**Fig. S3** Contact angles of water drop at the films of colloidosome-like microgels dried at 25  $^{\circ}$ C (left) and 50  $^{\circ}$ C (right) respectively.



**Fig. S4** Apperance of the dispersion of the colloidosome-like microgels with toluene as the solvent with different concentrations.



**Fig. S5** CLSM image of the dispersion of colloidosome-like microgels in toluene, the polymer matrix of the microgel was labelled with Rhodamine (red) while the toluene was dyed with perylene (blue).



**Fig. S6** CLSM images of an oil-in-water emulsion droplet with the colloidosome-like microgels as particulate stabilizers in merged channels in cross-section focus.