

## Supplementary Materials for

### **Anisotropic structural color particles from colloidal phase separation**

Huan Wang, Yuxiao Liu, Zhuoyue Chen, Lingyu Sun, Yuanjin Zhao\*

\*Corresponding author. Email: yjzhao@seu.edu.cn

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Fig. S14. The effect of various isoproterenol concentrations.

Table S1. Zeta potential characterization of silica nanoparticle solution, GO solution, and silica nanoparticles/GO solution, respectively.

Legends for movies S1 to S4

**Other Supplementary Material for this manuscript includes the following:**

(available at [advances.sciencemag.org/cgi/content/full/6/2/eaay1438/DC1](http://advances.sciencemag.org/cgi/content/full/6/2/eaay1438/DC1))

Movie S1 (.avi format). The spherical SCPs in fluid.

Movie S2 (.avi format). The Janus SCPs in fluid.

Movie S3 (.avi format). The photothermal response process of a pNIPAM hydrogel Janus SCP.

Movie S4 (.avi format). The cardiomyocytes beating derived color change of a GelMA hydrogel SCP.

## The calculation of the size of the nanoparticles in the mix solution

The sedimentation of the nanoparticles resulted from the force  $F_g$  caused by the difference between the weight and buoyancy, which could be shown as following

$$F_g = (\rho_{silica} - \rho_{water})g \frac{4}{3}\pi r^3 \quad (1)$$

where  $\rho_{silica}$  and  $\rho_{water}$  were the density of silica and water respectively,  $r$  was the radius of the nanoparticles. According to the Stokes' law, the resistance  $F_d$  of nanoparticles in the solution was

$$F_d = 6\pi\eta vr \quad (2)$$

where the  $\eta$  was the dynamic viscosity of the solution,  $v$  was the sedimentation rate. Thus, the terminal sedimentation rate of the nanoparticles in the droplets could be reached when

$$F_g = F_d \quad (3)$$

at this moment the terminal rate  $v_t$  was

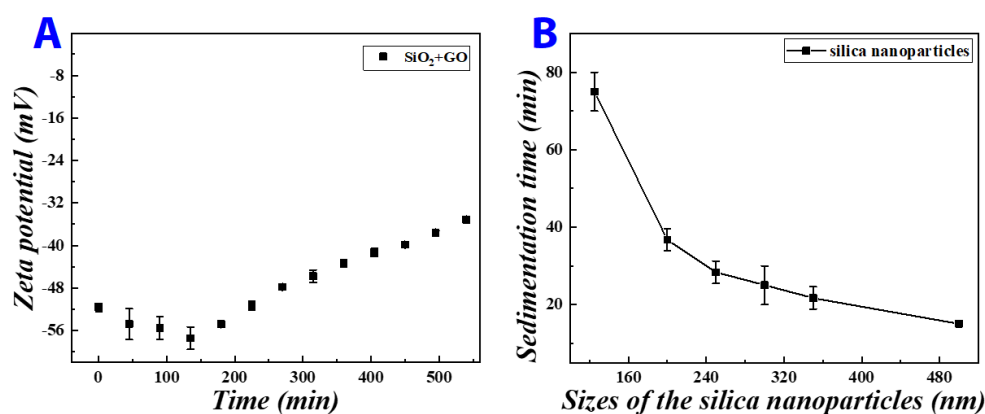
$$v_t = \frac{2r^2(\rho_{silica} - \rho_{water})g}{9\eta} \quad (4)$$

and the radius of the nanoparticles could be demonstrated by

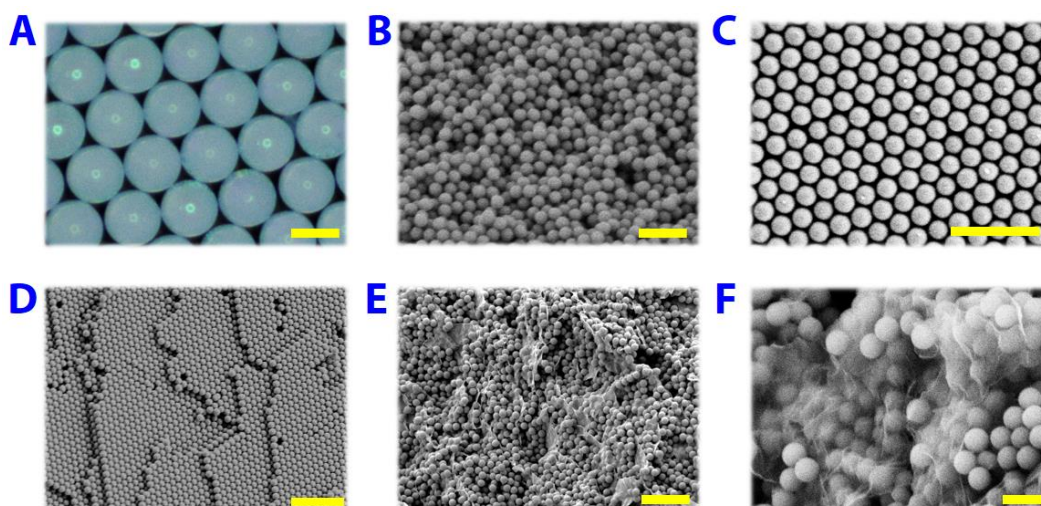
$$r = \sqrt{\frac{9\eta v_t}{2(\rho_{silica} - \rho_{water})g}} \quad (5)$$

In the droplets of silica nanoparticles with size of 250 nm ( $r_0$ ), the obvious stratification formed after 30 min and the thickness of the GO layer was about 100  $\mu\text{m}$ .

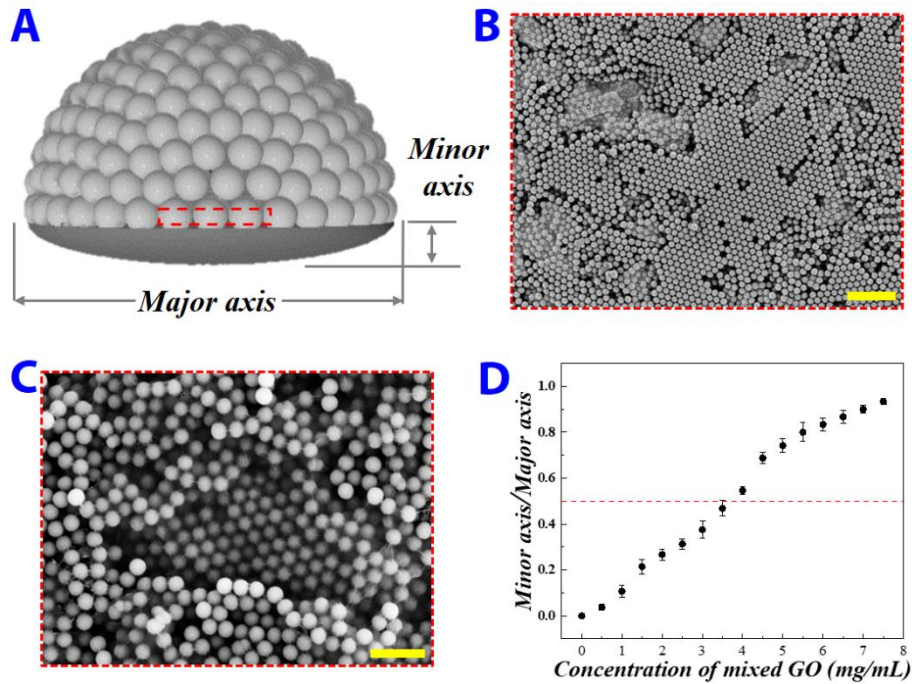
According to this result, assuming that there were no silica nanoparticles existed in the GO layer and the nanoparticles reached sedimentation rate in a short time, the  $v_t$  could be estimated about 200  $\mu\text{m/h}$ . The dynamic viscosity  $\eta$  of the solution was measured by a rheometer and the value was 2.1  $\text{mPa}\cdot\text{s}$ ; the densities of silica and water was 2.65  $\text{g/cm}^3$  and 1  $\text{g/cm}^3$  respectively; the gravitational acceleration was 9.8  $\text{m/s}^2$ . Plug all these data to the equation (5) we could find the value of  $r$  was 360 nm. The value of  $(r/r_0)^3=1.728$ , meaning that the volume of the silica nanoparticles in the mix solution was only a little larger than the initial silica nanoparticles, hence the silica nanoparticles hadn't aggregated in large groups. In addition, it was worth mention that there were many silica nanoparticles exist in the GO layer, indicating that the actual  $v_t$  was lower and the calculated  $r$  was larger than actual situation, therefore actual  $(r/r_0)^3$  was smaller than 1.728, which meant that aggregation was occurred less than calculated situation in the mix solution. The diameters measured by the granulometer ( $287.3 \pm 2.7$  nm for silica solution and  $298.4 \pm 4.3$  nm for mix solution) also confirmed the results.



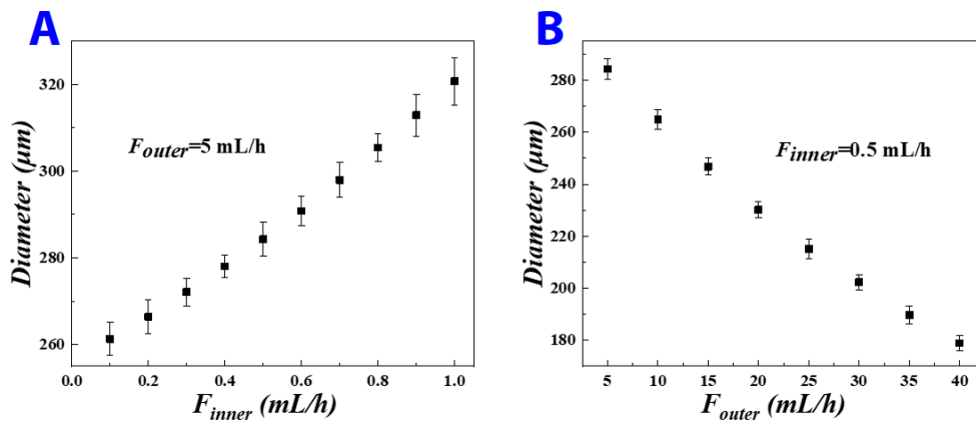
**Fig. S1. The change of charge (zeta potential) of the mixed solution and sedimentation time of the silica nanoparticles.** (A) The change of charge (zeta potential) during the solvent evaporation of the mixed solution. (B) The relationship between the sedimentation time and the sizes of the silica nanoparticles.



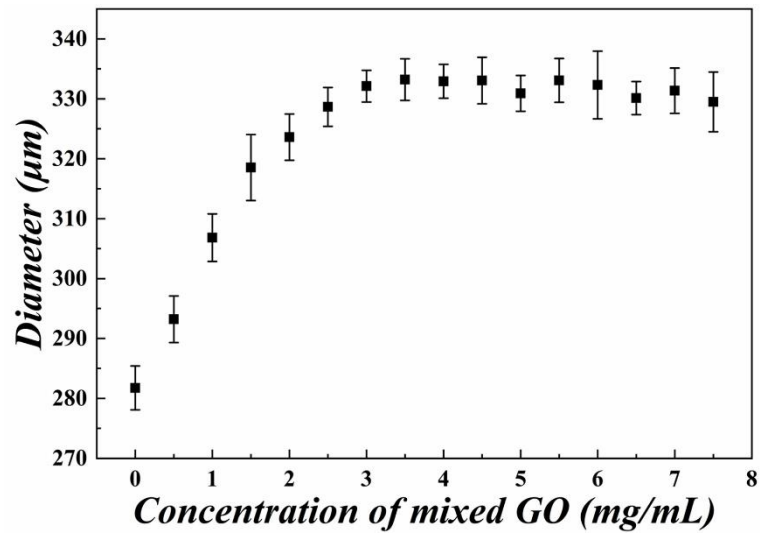
**Fig. S2. Optical image and SEM image characterization of spherical particles and Janus SCPs.** (A) The optical image of the particles assembled by silica nanoparticles in acid solution. (B) The microstructure of a particle in fig. S2A. The bars are 200  $\mu\text{m}$  in A, 1  $\mu\text{m}$  in B, (C, D) The surface and inner microstructures of a particle composed of silica nanoparticles with sulfonic groups on the surface. (E, F) The inner microstructure of the “dark section”. The bars are 200  $\mu\text{m}$  in A, 1  $\mu\text{m}$  in B and D, 500 nm in C and F, and 2  $\mu\text{m}$  in E.



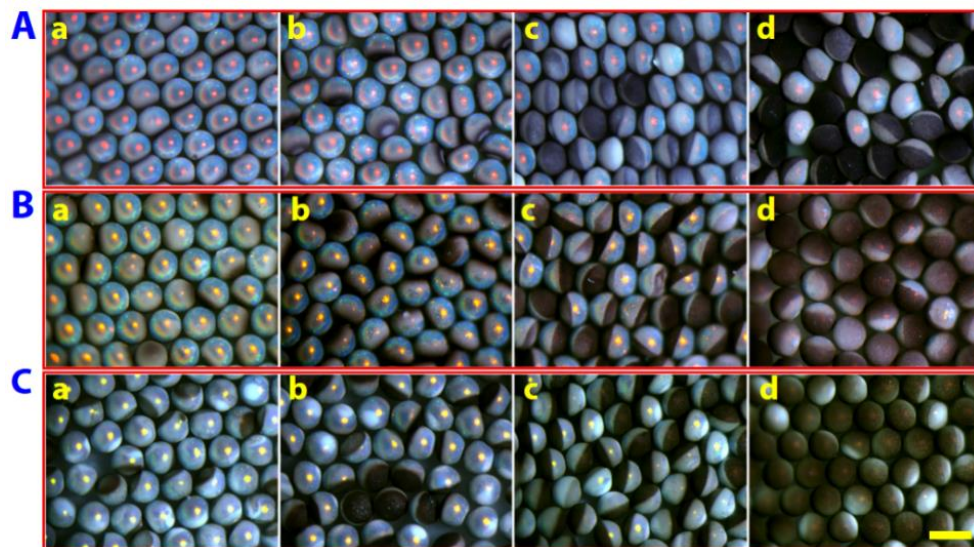
**Fig. S3.** The microstructure of the junction of the two domains and the effect of GO concentration on the ratio of the major axis and minor axis. (A) The schematic of the definitions of major axis and minor axis. (B, C) The microstructure of the part of “photonic section” boxed by red dashed line in fig. S3A. (D) The relationship between the concentration of mixed GO and the ratio of the major axis and minor axis of the Janus particles. The bars are 2  $\mu\text{m}$  in B and 1  $\mu\text{m}$  in C.



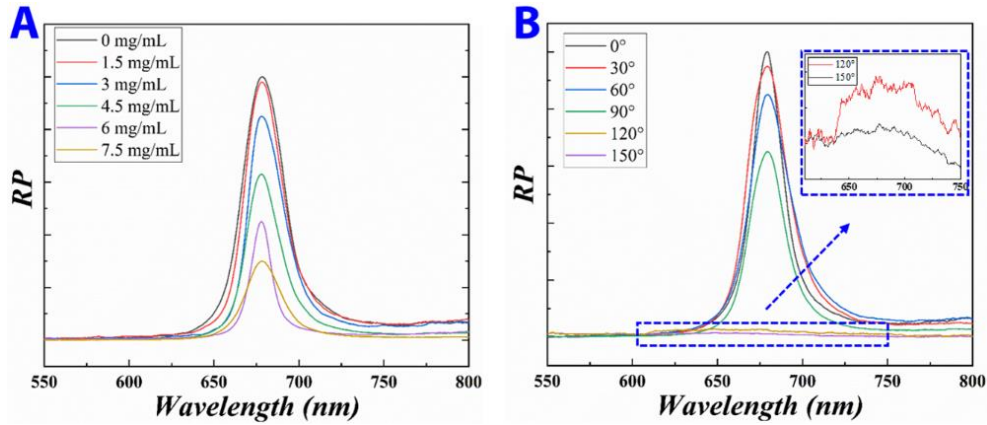
**Fig. S4.** Relationships between the Janus SCP radius and the flow rate. (A) The influence of the inner flow rate, (B) The influence of the outer flow rate.



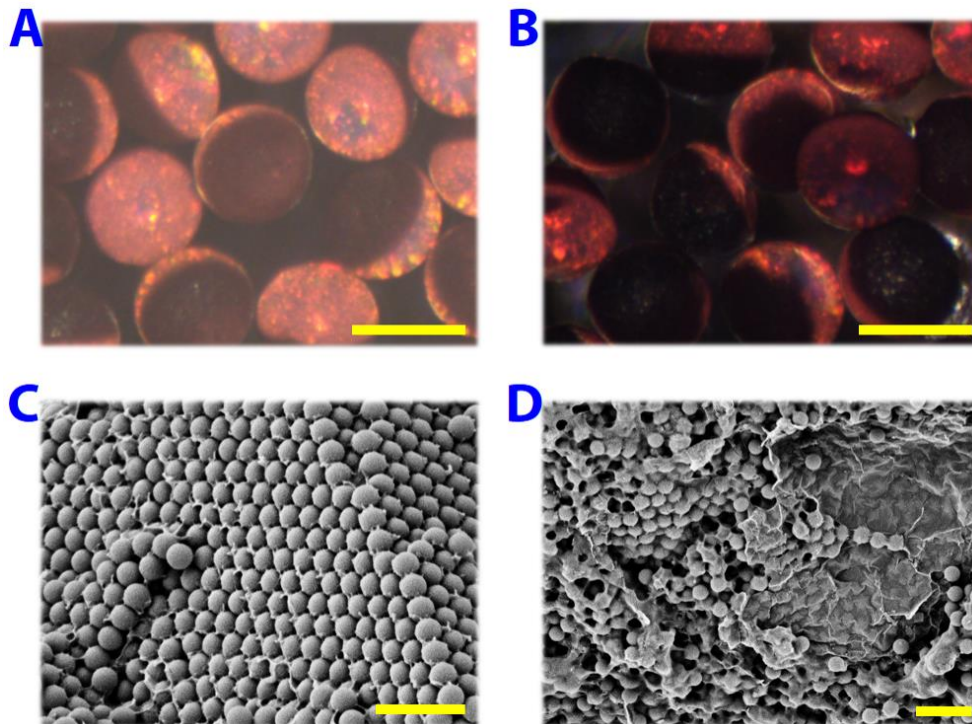
**Fig. S5.** Relationships of the Janus SCP radius with the concentration of mixed GO.



**Fig. S6.** Reflection microscope images of Janus SCPs. Four kinds of anisotropic (A) red, (B) orange and (C) yellow Janus SCPs with doping concentration of GO at 0.5, 1, 2, 4 mg/mL, respectively. The scale bar is 300 μm.

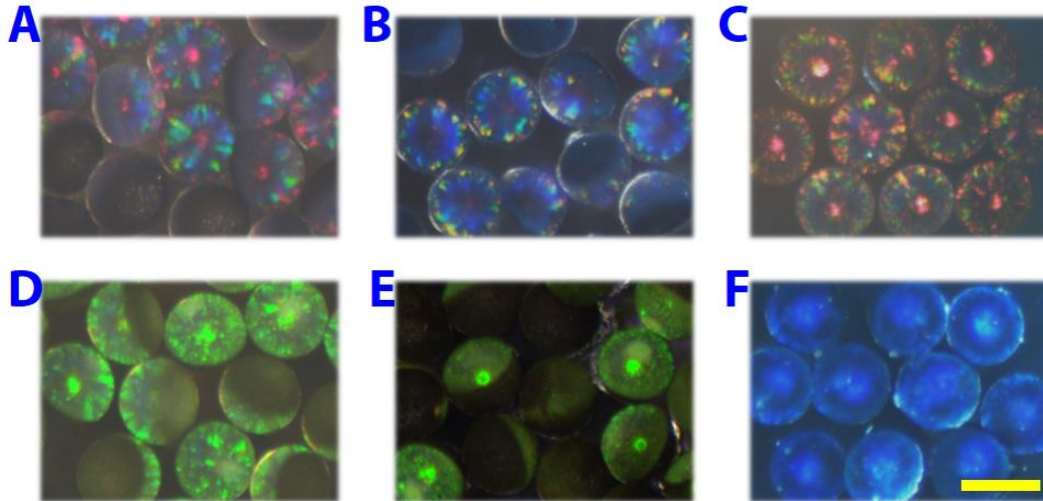


**Fig. S7.** The relationships between the spectra and GO concentration or observed angles. (A) the concentration of GO; (B) the different observed angles.

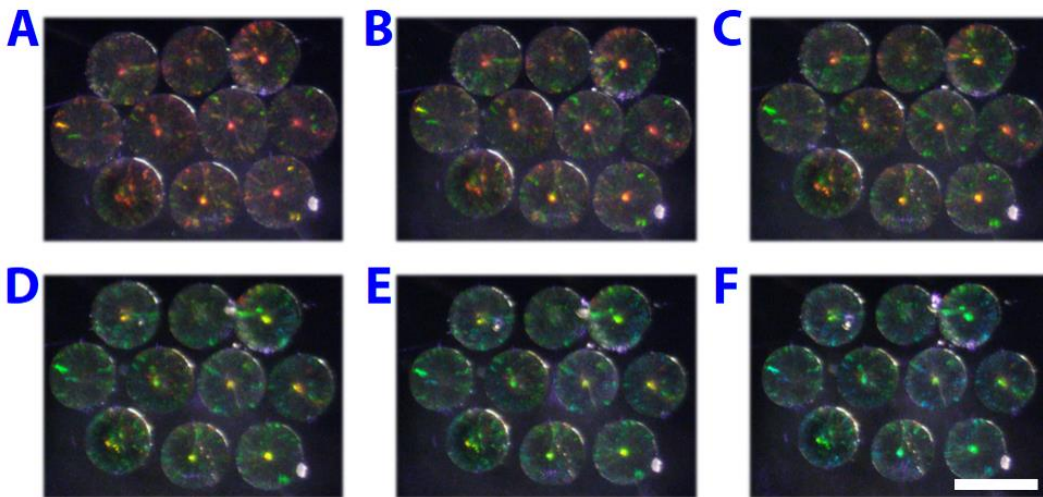


**Fig. S8.** The reflection microscope images and microstructure characterization of the orange Janus particles and the hybrid structure with pNIPAM. (A, B) The corresponding reflection microscope images of Fig. 5b: (A) The corresponding Janus SCPs, (B) the corresponding hybrid Janus SCPs. (C, D) The SEM images of the inner microstructure of the “photonic section” (C) and “dark section” (D) of a hybrid Janus SCP. The scale bars are 300  $\mu\text{m}$  in A, B and 1  $\mu\text{m}$  in C, D.

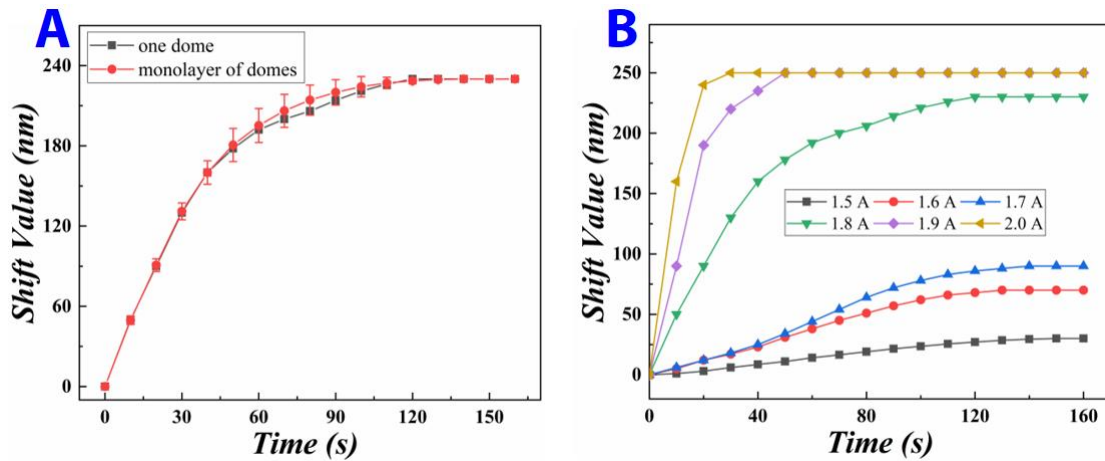




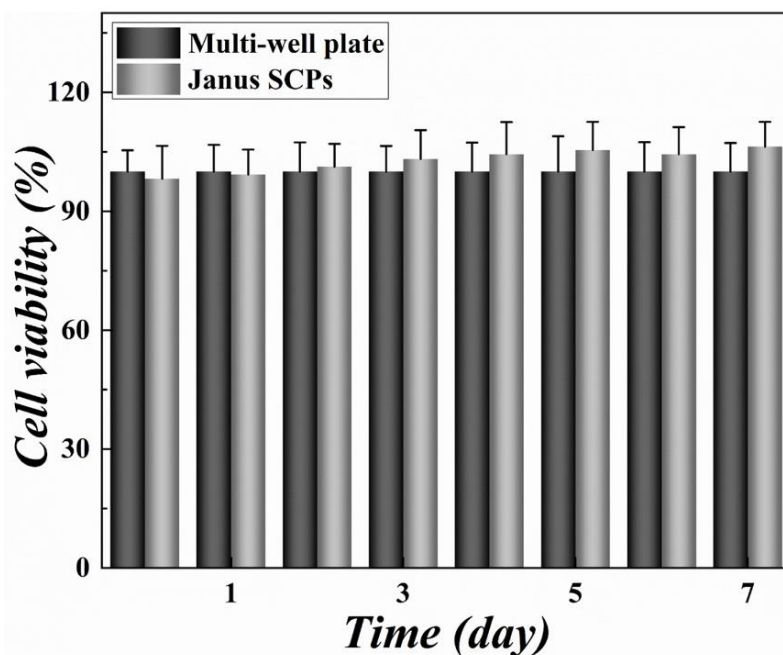
**Fig. S9.** Reflection microscope image characterization of the red and green Janus SPCs, the hybrid structure with pNIPAM, and the corresponding inverse opal pNIPAM Janus SPCs. (A, D) the red and green structural color Janus SPCs, (B, E) the corresponding hybrid Janus SPCs, and (C, F) the corresponding inverse opal Janus SPCs. The scale bar is 300  $\mu\text{m}$ .



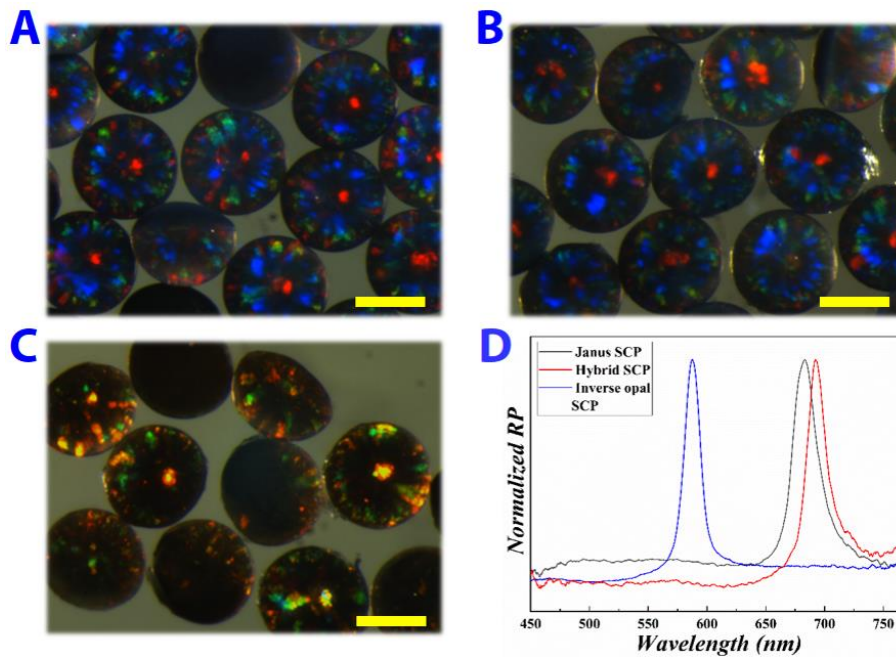
**Fig. S10.** The optical microscope images of the structural color variation process of the hydrogel Janus SPCs during half photothermal response cycles. The bar is 300  $\mu\text{m}$ .



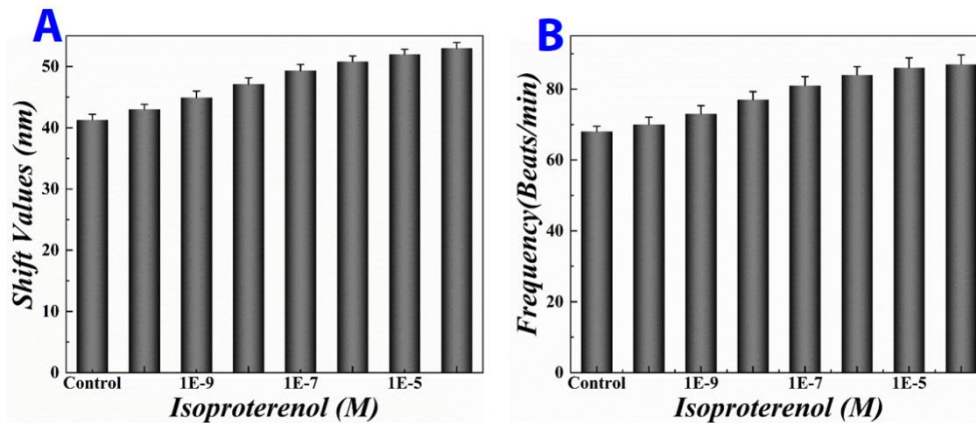
**Fig. S11. The photothermal responsive capability of the pNIPAM hydrogel Janus SCPs under NIR light.** (A) The compare of the photothermal responsive capability of one hydrogel particle and a monolayer of domes. (B) The photothermal responsiveness of one hydrogel particle under different NIR intensity. As the reflective spectra are messy at the wavelength range under 400 nm, the blue shift of reflective peaks more than 250 nm could not be detected.



**Fig. S12. The results of the cardiomyocyte MTT assays.** The cardiomyocytes were cultured on multi-well plate and GelMA inverse opal structural-color hydrogels for 1 day, 3 days, 5 days, and 7 days, respectively. The MTT values of the cardiomyocytes on multi-well plate in different time were set as control and the cells viability were set as 100%.



**Fig. S13. Reflection microscope images and spectra characterization of the Janus SCPs, the hybrid structure with GelMA, and the inverse opal GelMA Janus SCPs.** (A-C) Reflection images of (A) Janus SCPs, (B) GelMA hydrogel hybrid Janus SCPs, and (C) the GelMA inverse opal Janus SCPs. (D) Reflection peaks of the corresponding particles in (A-C). The bar is 200  $\mu\text{m}$ .



**Fig. S14. The effect of various isoproterenol concentrations.** Relationships of the average peak shift values (A) and the beating frequency (B) of the beating cycle of the hydrogel Janus SCPs with various isoproterenol concentrations.

**Table S1. Zeta potential characterization of silica nanoparticle solution, GO solution, and silica nanoparticles/GO solution, respectively.**

<b>Solution</b>	<b>Zeta potential (mV)</b>
SiO <sub>2</sub>	-51.27±0.45
GO	-43.27±1.27
SiO <sub>2</sub> +GO	-21.6±1.6

**Movie S1. The spherical SCPs in fluid.**

**Movie S2. The Janus SCPs in fluid.**

**Movie S3. The photothermal response process of a pNIPAM hydrogel Janus SCP.**

**Movie S4. The cardiomyocytes beating derived color change of a GelMA hydrogel SCP.**