

Electronic Supplementary Material (ESI) for Materials Horizons.

Magneto-Sensitive Photonic Crystal Ink for Quick Printing of Smart Device with Structural Colors

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Table S1 The DLS, TEM and reflection spectra measurement result of the MNCs with different size.

Sample	TEM-ave. [nm] ^{a)}	Z-ave. [nm] ^{b)}	PDI ^{b)}	Zeta Potential [mV] ^{b)}	Reflection Peak Position [nm] ^{c)}
MNCs-200	164	287	0.052	-38.4	819
MNCs-300	143	222	0.026	-56.5	643
MNCs-400	136	208	0.068	-57.8	594
MNCs-500	112	184	0.034	-43.3	552
MNCs-600	71	168	0.077	-52.9	478

^{a)} Particle diameters measured from TEM images; ^{b)} Results from DLS measurement reports;

^{c)} Collected from the reflection spectra in Figure 1f.

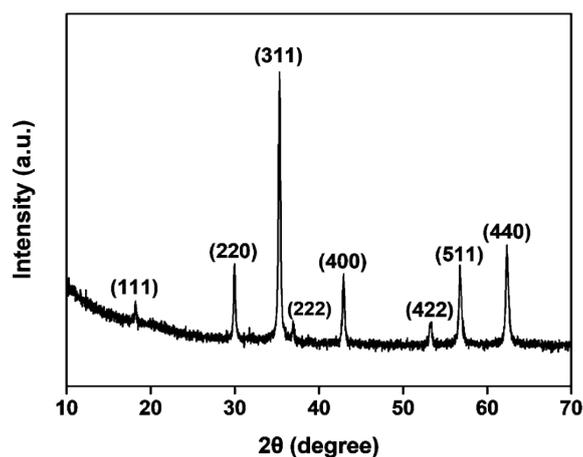


Fig. S1 The XRD pattern of the MNCs-400.

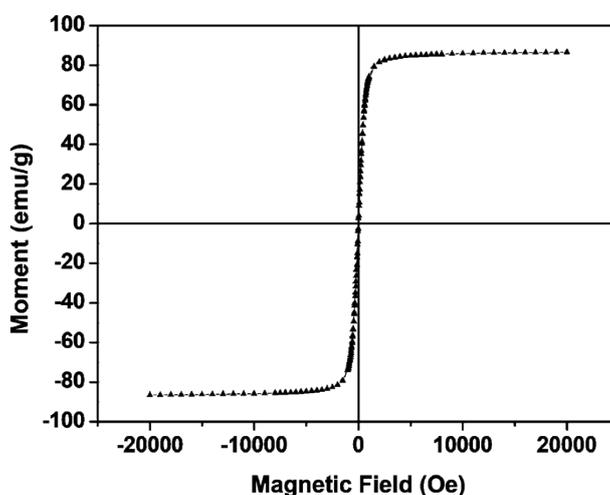


Fig. S2 The hysteresis curves of the MNCs-400.

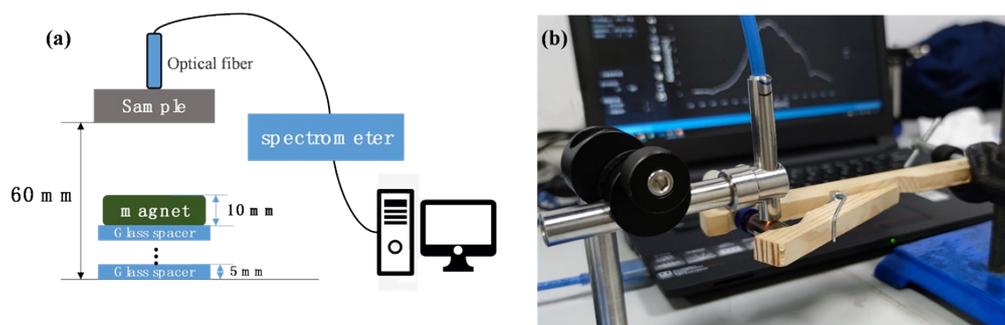


Fig. S3 (a) The schematic illustration and (b) the actual photo showing the reflectance spectra measurement set-up detail. The measurements were conducted in a dark room actually.

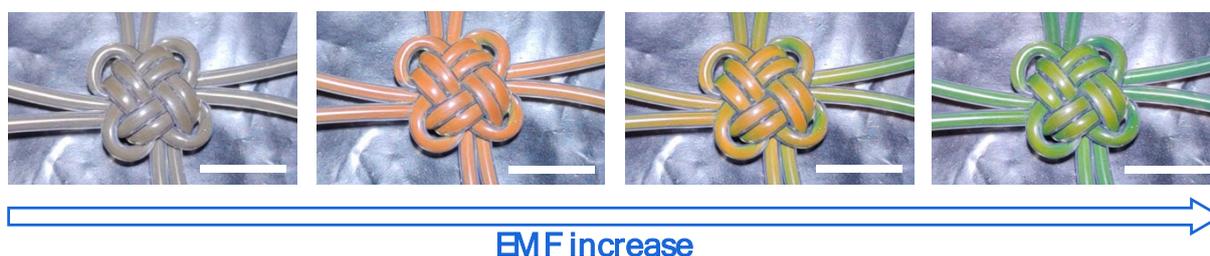


Fig. S4 Lucky knot woven using silicone capillary tube fulfilled with MNCs-400 aqueous dispersion (10 mg/ml) showing brilliant changing color under increasing EMF strength. (Scale bar: 1 cm)

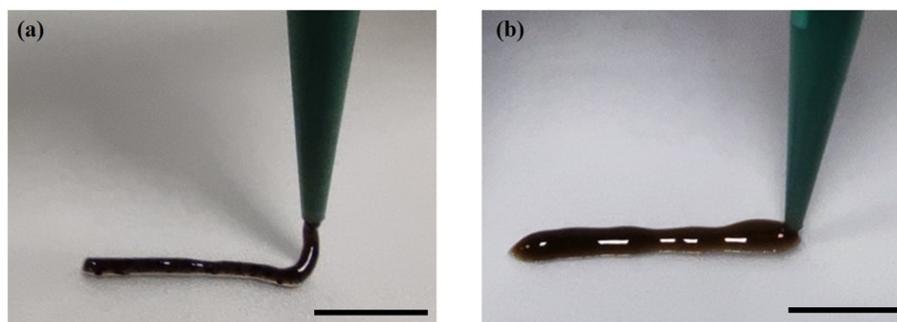


Fig. S5 (a) Photos of printing with MNCs-400 emulsion ink and (b) MNCs-400 emulsion ink without fumed silica. (Scale bar: 5 mm)

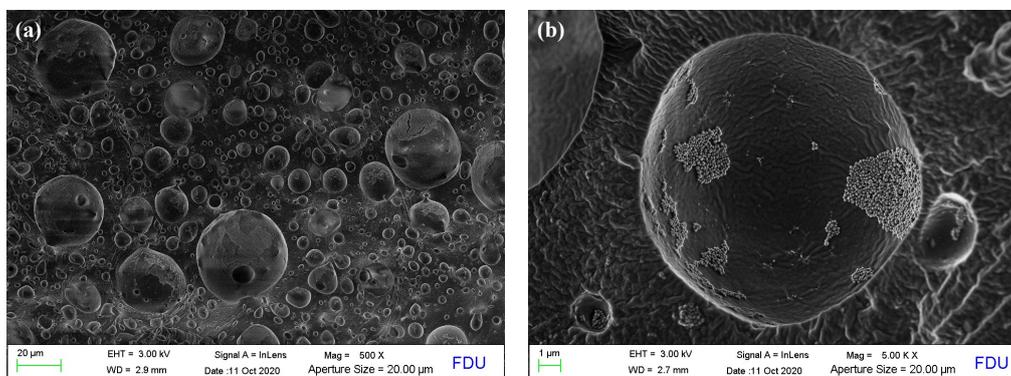


Fig. S6 The cross-section SEM images of the cured MRPC ink made with MNCs-400. (a) The distribution of the pores (b) where the MNCs-400 nanoparticles lie, which derives from the inner glycol phase of the PDMS water-in-oil emulsion.

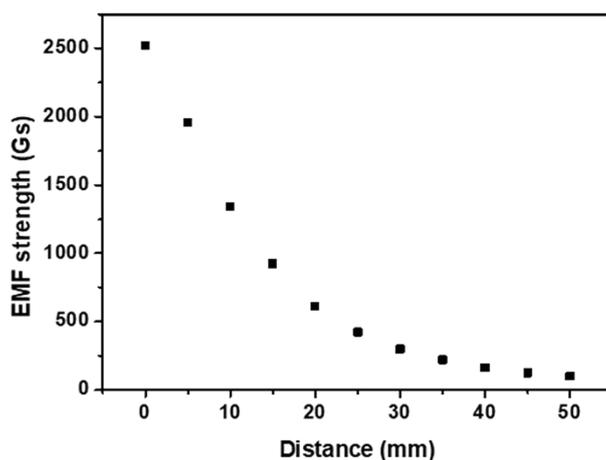


Fig. S7 The relationship between the actual EMF strength and distance away from the magnet surface.

Study on the influence of composition on the properties of the MRPC inks

In our experiments, aqueous dispersion of MNCs-400 was firstly used to be emulsified in PDMS prepolymer mixture. It was found that the performance of the ink could be directly judged by its optimal structural color under EMF (around 300 Gs). In the mixture of 2 g PDMS prepolymer, 0.2 g curing agent, 0.17 g fumed silica and about 0.04 g ethoxylated silicone surfactant as outer phase, the aqueous MNCs-400 dispersion was gradually added into above mixture and blended to form a viscous emulsion. When the addition amount of aqueous dispersion is less than 1 mL, the homogenous emulsion could be obtained and tended to display dark green under EMF (Fig. S8 a₁). When the amount of aqueous dispersion is over 1.2 mL, the homogenous emulsion could not be obtained, indicating the maximum value for emulsification of the dispersion (Fig. S8 b₁).

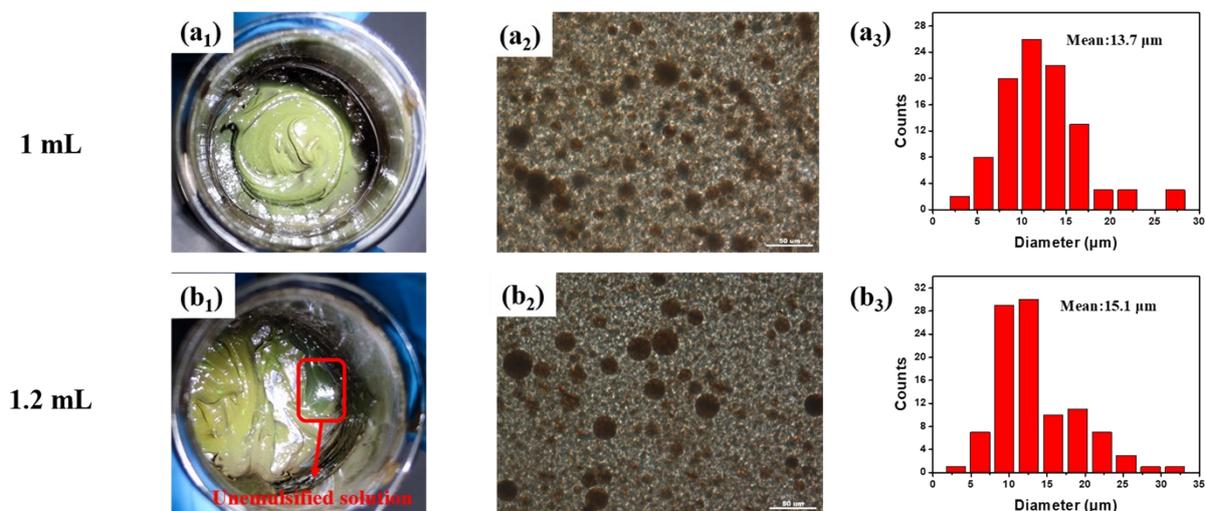


Fig. S8 (a₁, b₁) The color performance, (a₂, b₂) optical microscope images and (a₃, b₃) emulsified droplet size distribution histogram of MRPC emulsion ink using (a) 1 mL and (b) 1.2 mL of MNCs-400 aqueous dispersion.

As a magneto-responsive ink, the MNCs need to assemble themselves inside the tiny droplets to form numerous reflective units, so the volume and density of droplets is supposed to be very important related to the macroscopic color. The emulsified droplet size distribution could be observed under the optical microscope (Fig. S8 a₂ and b₂), the mean droplets size only increase from 13.7 μm to 15.1 μm (Fig. S8 a₃ and b₃), which was consistent with the unobvious change of the color performance (Fig. S8 a₁ and b₁). In this part, it should be mentioned that it is hard to verify the border of the emulsified droplets under 5 μm or less, which are believed to have little contribution to the reflective color based on PC assembly for such tiny volume.

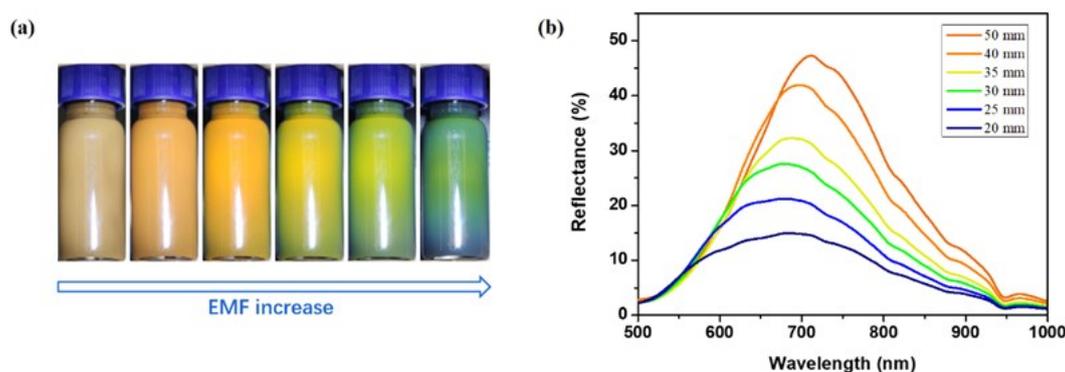


Fig. S9 (a) The photos of the colorful MNCs-400 EG dispersion (10 mg/mL) in 2 mL glass vial and (b) its reflection spectra under different EMF strength.

In our experiments, we also found that the MNCs-400 could well-dispersed in EG by solvent replacement and maintaining its good magnetic response property (Fig. S9), then we tried to emulsify the EG dispersion in the PDMS prepolymer mixture (the amount is same as above recipe). The loading amount less than 0.5 mL brought too weak structural color to be distinguished. When 0.5 mL EG dispersion was added and emulsified, there were similar appearance and mean droplets size as the emulsion did using aqueous dispersion (Fig. S10 a₁-a₃). Surprisingly, when the amount of EG was increased to 1.0 mL, the emulsion ink showed brighter color of grass green (Fig. S10 b₁). Meanwhile, it is noteworthy that the droplets were bigger and denser, with the mean droplets size growing to 27.4 μm (Fig. S10 b₂ and b₃). These significant difference implied that EG dispersion was more suitable than aqueous dispersion for emulsification in our system. Similarly, the mixture then started to show phase separation after exceeding the maximum value for emulsification at 1.5 mL (Fig. S10 c₁). The mean droplet size also got slightly smaller to 23.7 μm (Fig. S10 c₂ and c₃), which might be attributed to affected the emulsion stability by the excessive EG dispersion.

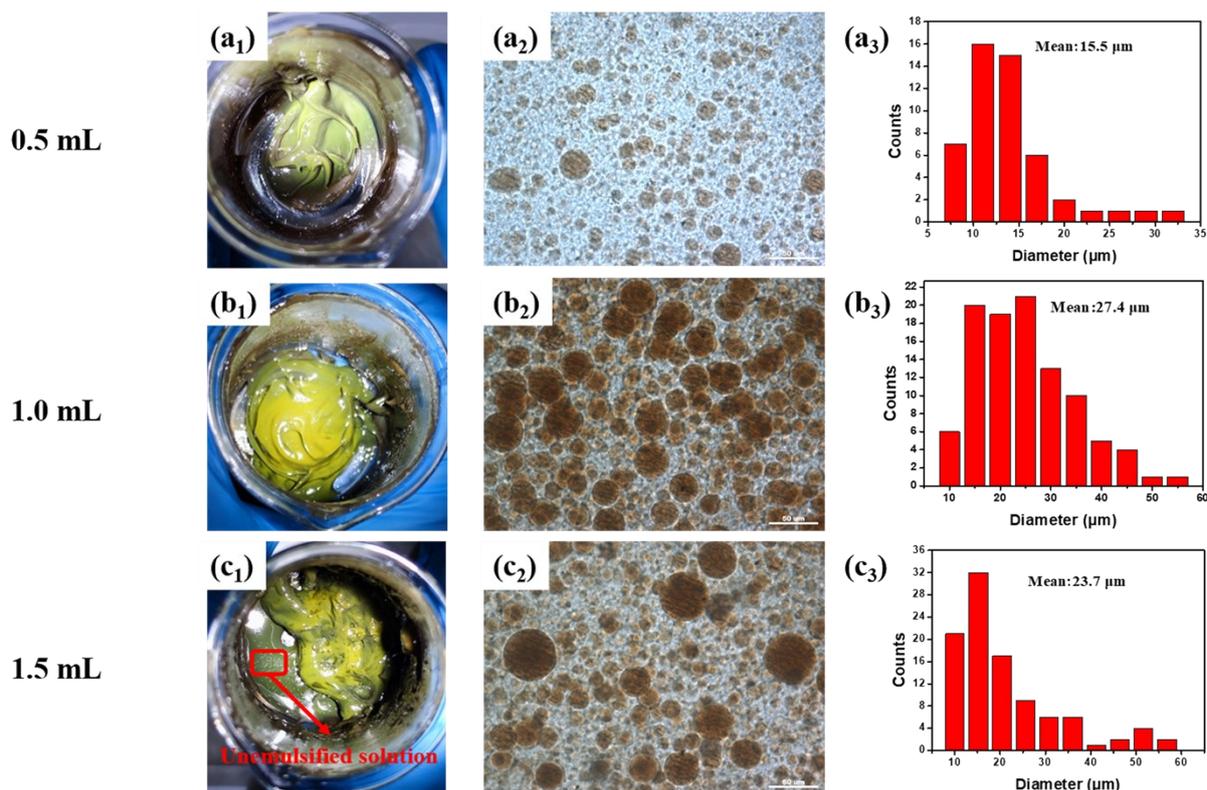


Fig. S10 (a₁, b₁ and c₁) The color performance, (a₂, b₂ and c₂) optical microscope images and (a₃, b₃ and c₃) emulsified droplet size distribution histogram of MRPC emulsion ink using (a) 0.5 mL, (b) 1.0 mL and (c) 1.2 mL of MNCs-400 EG dispersion.

In summary, the suitable droplet size and density could offer effective reflective units to make the structural color looks brighter. As a result, EG solution was finally selected for the preparation of MRPC inks.

Movie S1 Diluted MNCs-400 aqueous dispersion under optical microscope. 1D-nanochain is formed and oriented along the EMF direction when the magnet is approaching and moving. The magnet is taken away at the end of the movie.

Movie S2 The MNCs-400 dispersion (10 mg/ml) under sunlight showing different structural color by changing the distance between the magnet and sample.

Movie S3 Uncured photonic crystal ink made with MNCs-400 under optical microscope. MRPC nanoparticles are distributed in the inner phase of the EG/PDMS emulsion. 1D-nanochain is formed and oriented along the EMF direction when the magnet is approaching and moving. The magnet is taken away at the end of the movie.

Movie S4 Customized butterfly pattern 3D-printed using photonic crystal ink made with MNCs-400 and MNCs-300, showing magnetic responsive structural color with good repeatability in dynamic EMF.