

## Core @ double shell structured PBO composite with excellent dielectric properties and high heat resistance

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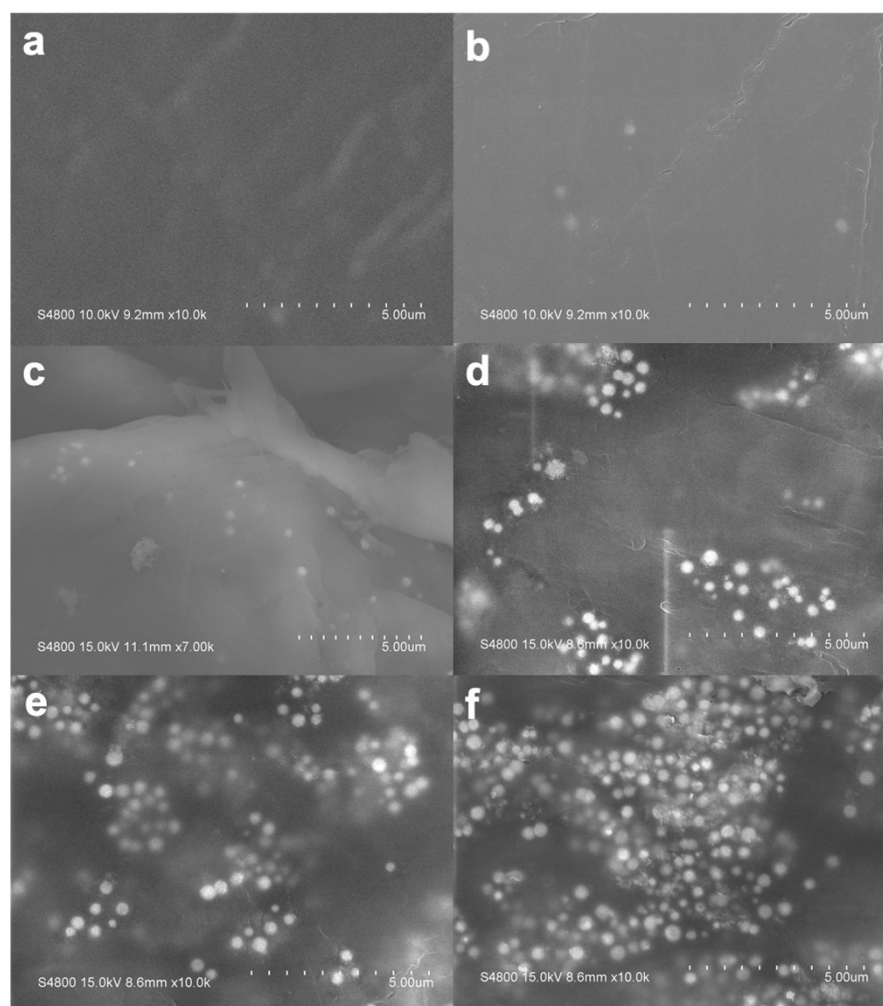


Fig. S1 SEM images of 0wt%, 3wt%, 6wt%, 9wt%, 12wt%, and 15wt%- $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$  /PBO. The distribution of the filler throughout the material is very uniform. when the mass fraction at 3wt%, 6wt% and 9wt%, We found almost every filler particle is dispersed; Almost all the fillers in Fig. 3e (at 1wt%) are also dispersed (Refer to the enlarged

figure 5d in the original text); But, When the mass fraction reaches 15%, part of the fillers began to aggregate (in Fig. s1f).

The distribution of  $\text{Fe}_3\text{O}_4@\text{C}$  is similar to the  $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$ . The distribution of the  $\text{Fe}_3\text{O}_4@\text{C}$  is uniform, but there are also subtle differences in concentration in the smaller area. Compare with  $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$ , the difference is bigger. This phenomenon is obvious in the instrument screen.

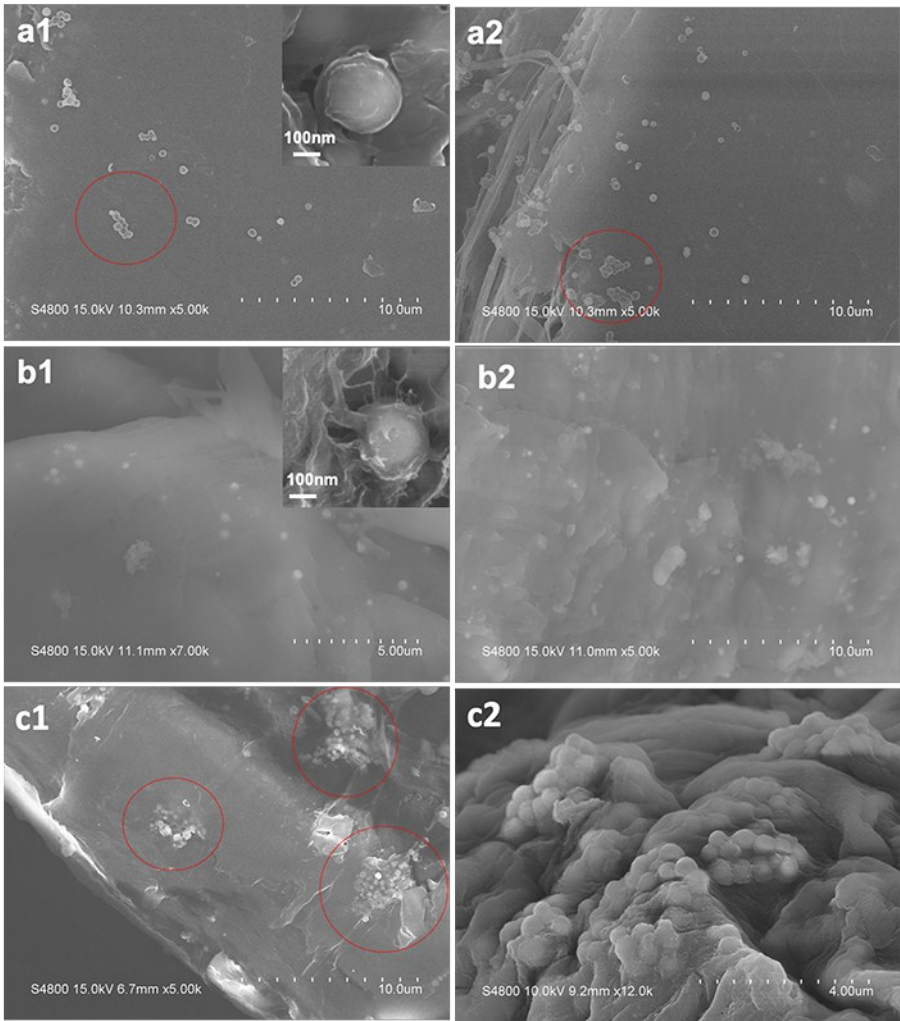


Fig. s2 SEM images of 6wt%- $\text{Fe}_3\text{O}_4@\text{C}$  /PBO(a1/a2), 6wt%- $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$  /PBO(b1/ b2); Mix-6wt%- $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$  /PBO(c1/c2)

Table s1 dispersion of the filler in the matrix

| film                                                        | Preparation            | Distribution throughout the material | Distribution in small area | Figure |
|-------------------------------------------------------------|------------------------|--------------------------------------|----------------------------|--------|
| 6wt%- $\text{Fe}_3\text{O}_4@\text{C}$ /PBO                 | In-Situ Polymerization | good                                 | medium                     | a1/a2  |
| 6wt%- $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$ /PBO     | In-Situ Polymerization | good                                 | good                       | b1/b2  |
| Mix-6wt%- $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$ /PBO | Physical mixing        | bad                                  | bad                        | c1/c2  |

(a1/a2) and (b1/b2) are prepared by in-situ polymerization, but (c1/c2) is prepared by physical mixing method. First, from the perspective of the entire material area, the filler distribution of (a1/a2) and (b1/b2) are uniform, but the distribution of (c1/c2) is not uniform. We found that the fillers of (c1/c2) were all piled up in several areas, while the other areas had almost no filler. (as shown in Fig. c1, c2) Second, from the perspective of small area, the uniformity of (b1/b2) is better than (a1/a2)'s. As shown in (a1/a2), We can often observe a small group of  $\text{Fe}_3\text{O}_4@\text{C}$  nanoparticles in a small group, But this phenomenon is rarely seen in (b1/b2), and Almost all  $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$  in (b1/b2) are individually separated.

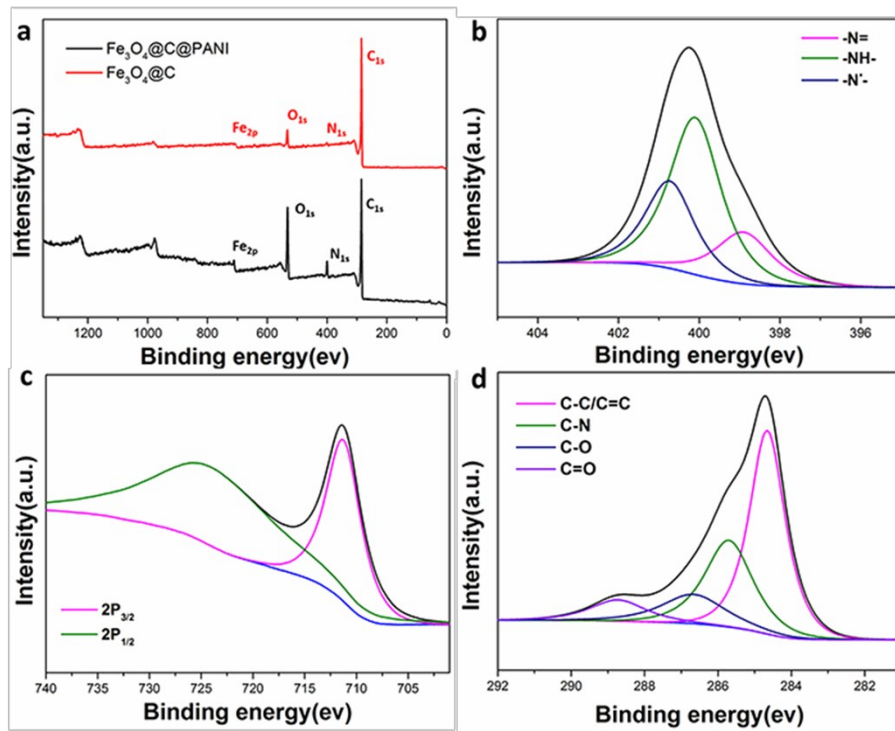


Fig. S3 XPS spectrum of (a)  $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$  and  $\text{Fe}_3\text{O}_4@\text{C}$ ; (b)/c)/ (d) XPS data of the  $\text{N}_{1s}$ ,  $\text{Fe}_{2p}$  and  $\text{C}_{1s}$  regions of the  $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$ , respectively

Compared with  $\text{Fe}_3\text{O}_4@\text{C}$ ,  $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$  has a distinct  $\text{N}_{1s}$  peak around 400, Which indicate that the N element is mainly from PANI. Fig. 8b is the spectrum of the  $\text{N}_{1s}$  of  $\text{Fe}_3\text{O}_4@\text{C}@\text{PANI}$ , which shows three different peaks at 398.9, 399.9 and 400.3 eV, corresponding to the  $-\text{N}=$ ,  $-\text{NH}-$ , and  $-\text{N}^+$  bonds, respectively.<sup>1,2,3</sup> The region of the C 1s spectrum can be decomposed into four components corresponding to C-C/C=C (284.6 eV), C-N (285.7 eV), C-O (286.6 eV) and C=O (288.7 eV), respectively.<sup>1</sup> C-N bonds come from PANI molecular chain. The appearance of C-O (286.6 eV) and C=O (288.7 eV) are due to the subsequent acidification and polymerization process. Two

broad peaks centered at 723.9 eV and 711.2 eV are assigned to the spin-orbit split doublet of  $\text{Fe}_{2p1/2}$  and  $\text{Fe}_{2p3/2}$ , in accordance with the reported values for  $\text{Fe}_3\text{O}_4$ .<sup>4</sup> “The broadness of  $\text{Fe}_{2p}$  peaks indicates the presence of dual iron oxidation states ( $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ )”<sup>5, 6</sup>

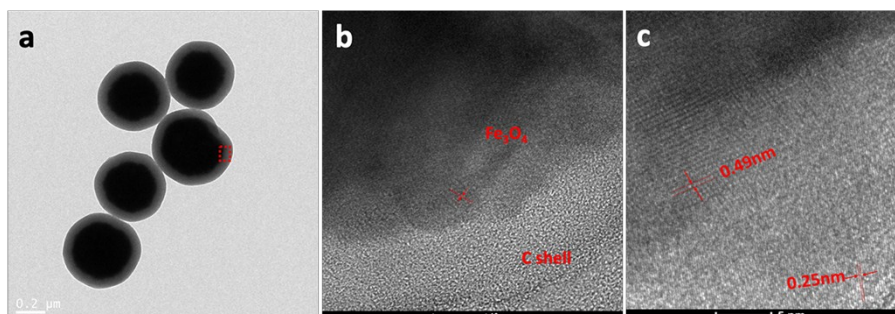


Fig.s4 (a-b) HRTEM of  $\text{Fe}_3\text{O}_4@\text{C}$ ; (c) HRTEM of  $\text{Fe}_3\text{O}_4$  (the the core part of  $\text{Fe}_3\text{O}_4@\text{C}$ )

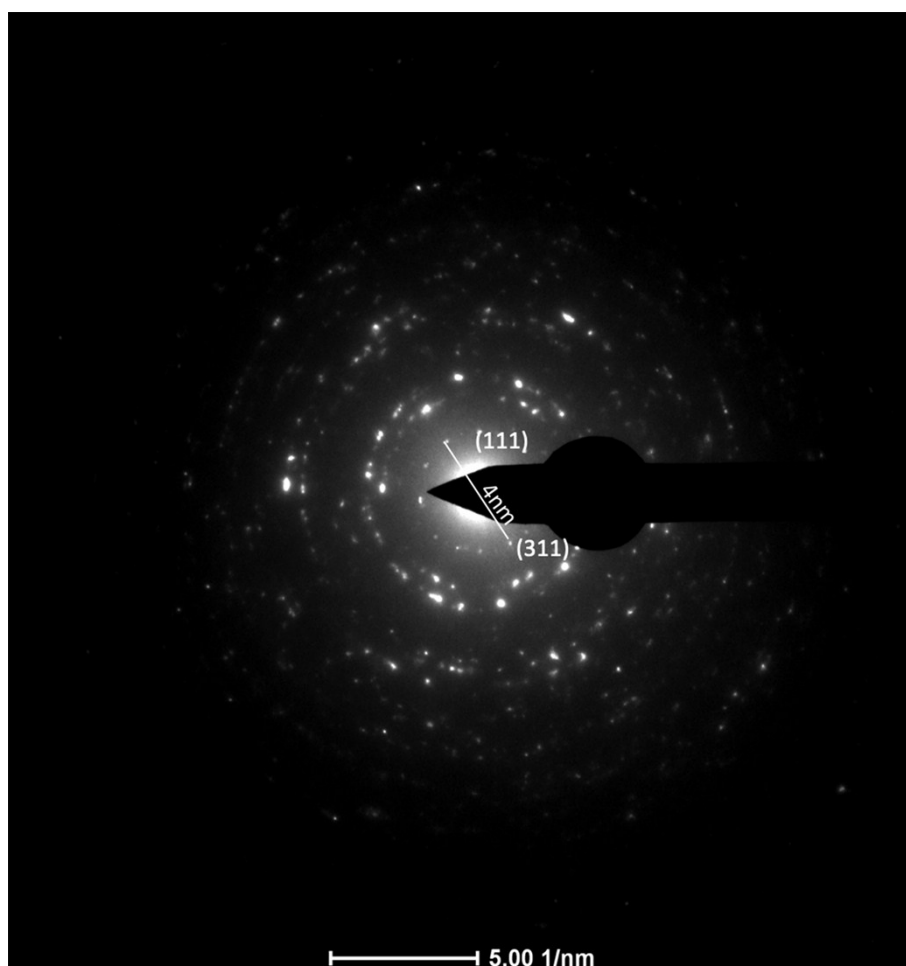


Fig. s5 SAED of  $\text{Fe}_3\text{O}_4$

As is shown in fig. s4, HRTEM may tell the lattice information. I choose the interface of  $\text{Fe}_3\text{O}_4$  and carbon as the observation area. We can see the boundaries of  $\text{Fe}_3\text{O}_4$  and

carbon in Fig. s4b.  $\text{Fe}_3\text{O}_4$  is inverse spinel structure, so it has obvious lattice fringes. But carbon is amorphous carbon and don't have clear lattice fringes. Fig. s4c shows that there are two kinds of lattice fringes of  $\text{Fe}_3\text{O}_4$ , and the corresponding interplanar spacing is 0.49nm (111) and 0.25 nm(311), respectively.

The selected area electron diffraction (SAED) pattern in Fig. s5 is composed of a series of concentric circles of different radii, which are produced by a large number of small grains of  $\text{Fe}_3\text{O}_4$  in the irradiation zone.<sup>6,7,5</sup> The diffracted beam produced by the same crystal face family forms a conical surface with a half-apoint angle of  $2\theta$ , and its line with the ground is a circle, and the radius of the ring is  $R=L\lambda/d=K/d$ , K and L is the instrument parameter. we can speculate that the aperture (radius 2 nm) near the bright spot corresponds to the (111) crystal plane. The aperture (radius 4n m) corresponds to the (311) crystal plane. The other apertures correspond to the other crystal planes in  $\text{Fe}_3\text{O}_4$ .

#### Note and references

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