Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information for

Enhanced magnetorheological performance of highly uniform magnetic carbon nanoparticles

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1. Reaction mechanism at each step

The magnetic carbon nanoparticles (MC NPs) were fabricated through the polymerization of polypyrrole nanoparticles (PPy NPs) and the carbonization of PPy NPs. First of all, polypyrrole nanoparticles were fabricated by chemical oxidation polymerization.[1] The mechanism for chemical oxidation polymerization of polypyrrole is described in **Fig. S1**. In the initiation step, radical cations are generated by the oxidation of pyrrole monomer. The oxidation of pyrrole monomer takes place simultaneously with the reduction of iron (III) ion which is a spontaneous reaction. Subsequently, a radical-radical coupling happens between two radical cations with forming a dimer, a bipyrrole, through deprotonation. In chain propagation step, the bipyrrole is oxidized and turns into radical cation. The bipyrrole radical cation couples with other radical cations, which is consecutively repeated during the propagation step. By the attack of water molecules or impurities in polymer chains, the termination occurs.

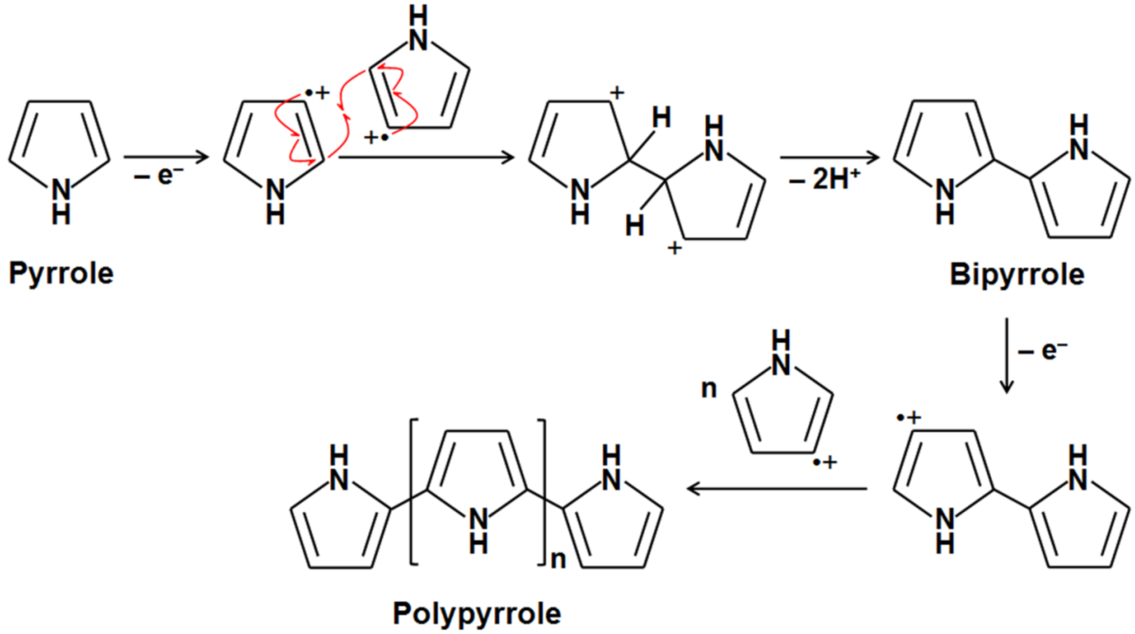

Fig. S1 Polymerization mechanism of polypyrrole.

In the carbonization process of the PPy NPs, the PPy NPs were placed in a quartz tubular furnace, and heated up to 600 °C in air atmosphere. Some of iron (III) chloride which was utilized as an oxidant formed iron-based complexes (*e.g.*, $FeCl_4^-$) and embedded in the PPy NPs. At the carbonization temperature (600 °C), the iron-based complexes lost their halogen atoms and formed iron oxides. During the carbonization process, the PPy NPs turned into carbon NPs through dehydrogenation and aromatization.

2. High-resolution TEM and SAED pattern analysis

The composition and crystallographic phase of the magnetic carbon nanoparticles by taking high-resolution TEM (HRTEM) image and selected area electron diffraction (SAED) pattern (**Fig. S2 and S3**). As shown in HRTEM image, several dark regions shaped like a spot were observed at the particles. To confirm that the dark regions in the particles were γ-Fe_2O_3, SAED patterns were taken at both the dark regions and other part in the particle. From the SAED patterns, the d-spacing values were calculated as described in **Fig. S3**. The d-spacing values calculated from the SAED pattern taken at dark regions correspond to the standard atomic spacing for γ-Fe_2O_3 from the JCPDS No. 39-1346. In addition, diffuse rings were observed in SAED patterns taken at carbon nanoparticle regions. The appearance of the (002) and (100) reflections indicates that the particles are composed of amorphous carbon. Judging from the SAED pattern analysis, the dark regions in the particles indicates γ-Fe_2O_3, and the particles consist of amorphous carbon.

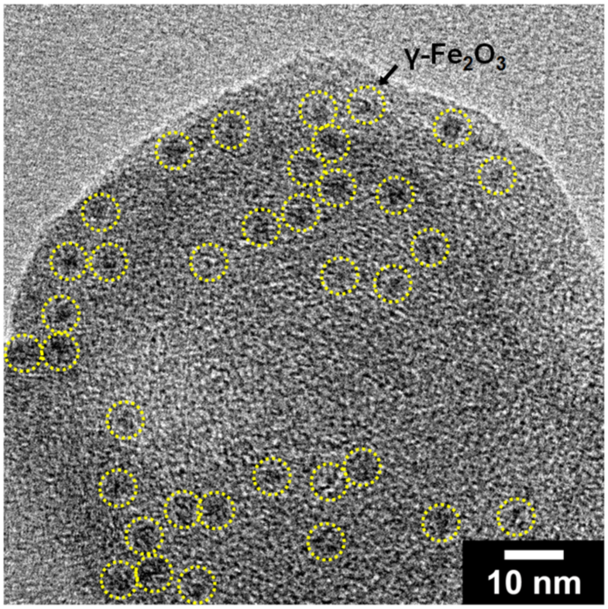

Fig. S2 High-resolution TEM image of magnetic carbon nanoparticle.

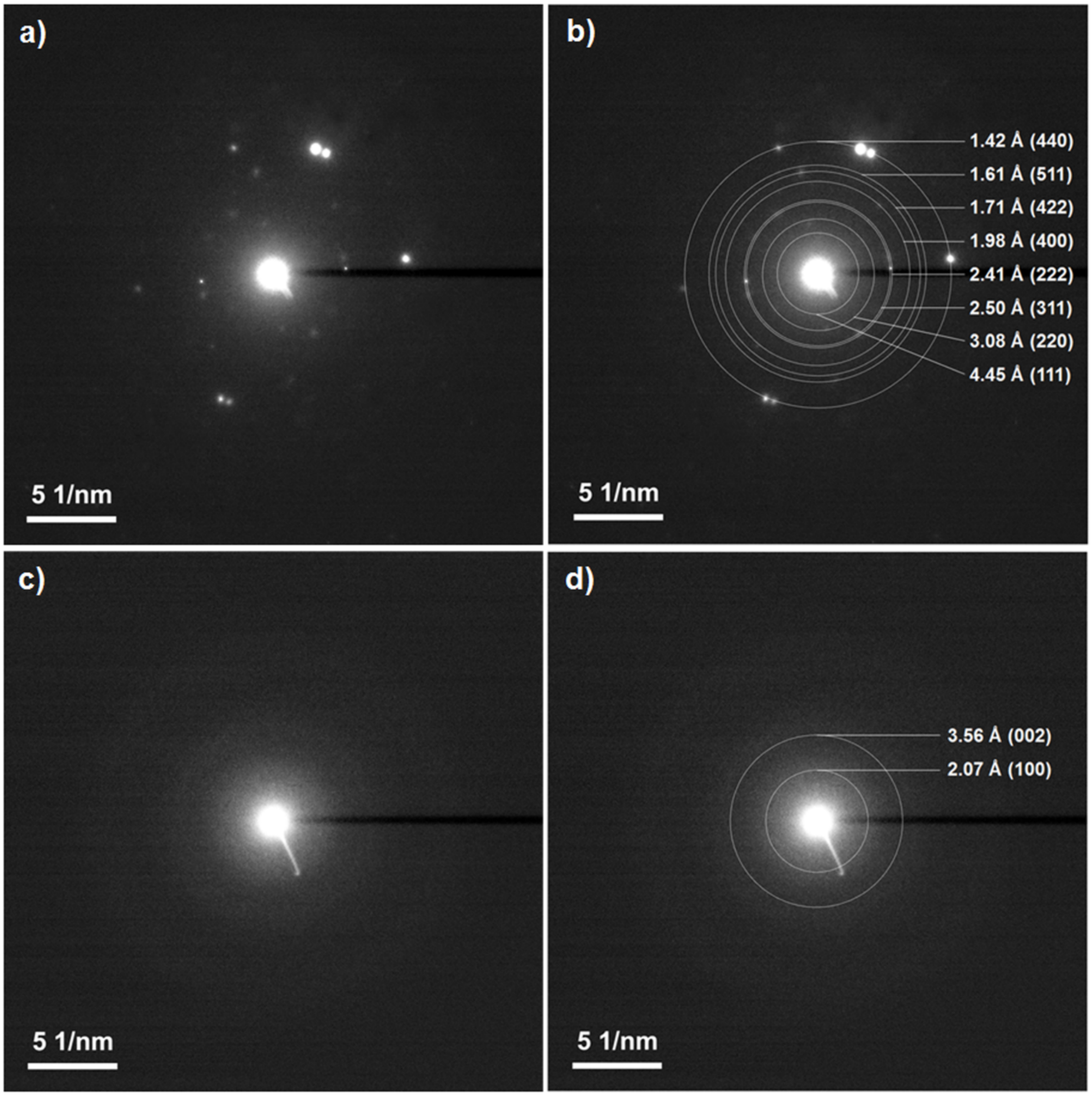

Fig. S3 Selected area electron diffraction (SAED) patterns taken at a), b) γ-Fe_2O_3 part of magnetic carbon nanoparticles (MC NPs), and c), d) carbon part of MC NPs. To make it easy to recognize, the rings were inserted at the electron diffraction patterns in b) and d).

3. Reference

1 J. Jang, *Adv. Polym. Sci.*, 2006, **199**, 189.