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Prussian Blue-Assisted One-Pot Synthesis of Nitrogen-Doped Mesoporous Graphitic Carbon Spheres for Supercapacitors

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Materials

Resorcinol (98%), phenol (99%), iron (III) chloride and potassium hexacyanoferrate were obtained from Acros Organics, NJ, USA. Formaldehyde (37wt%) was purchased from Fisher Scientific, USA. Pluronic F127 and polyvinylpyrrolidone (PVP, MW 40000) were purchased from Sigma-Aldrich. Deionized water (DW) was obtained from in-house Ion pure Plus 150 Service Deionization ion-exchange purification system and Aqua One (Amarillo, TX). All reagents (analytical grade) were used without further purification.

Characterization

Nitrogen adsorption isotherms were measured for all samples at -196 ºC using ASAP 2020 volumetric analyzer (Micromeritics, Inc., Norcross, GA) and nitrogen of 99.998% purity. All samples were out gassed under vacuum at 200 ºC for 2 h prior to adsorption measurements. The specific surface area of the samples was calculated using the Brunauer−Emmett−Teller (BET) method within the relative pressure range of 0.05−0.20. Pore size distributions were obtained from the nitrogen adsorption isotherms by the DFT (Density Functional Theory) method provided by Micromeritics.

Thermogravimetric (TG) profiles were recorded using high resolution thermogravimetric mode on TGA Q-500 analyzer (TA Instruments, Inc., New Castle, DE), from 25 to 900 ºC in flowing air with a heating rate of 10 ºC / min. The initial weight of each sample was between 5-15 mg. The TG profiles were used to obtain information about the stability of the carbon samples studied.

Scanning electron microscopy (SEM) images were obtained using SEM-Quanta-450 scanning electron microscope. Prior to SEM analysis all samples were dispersed in ethanol followed by sonication at concentrations of ~5-10 wt. %. Sonicated samples were deposited on SEM Hitachi specimen holder and dried under vacuum at 100 ºC for 1h.

Transmission electron microscopy (TEM) images were obtained on a FEI Tecnai G2 F20 microscope. Prior to TEM analysis, the samples were dispersed in ethanol by moderate sonication at concentrations of ~5-10 wt. %, followed by deposition of these samples on a Lacey carbon coated, 200-mesh, copper TEM grid by dipping into the sample suspension followed by drying under vacuum at 100 ºC for 1 h.

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Raman Spectra were collected on a Horiba LabRam HR-800 Raman Spectrometer equipped with an asymmetric Czerny Turner spectrometer (1200 g/mm grating) and CCD detector (1024×256 pixels, each pixel being 26 μm x 26 μm) as reported elsewhere (D.J. Deka et al., Applied Catalysis B: Environmental 2019, **248**, 487–503).

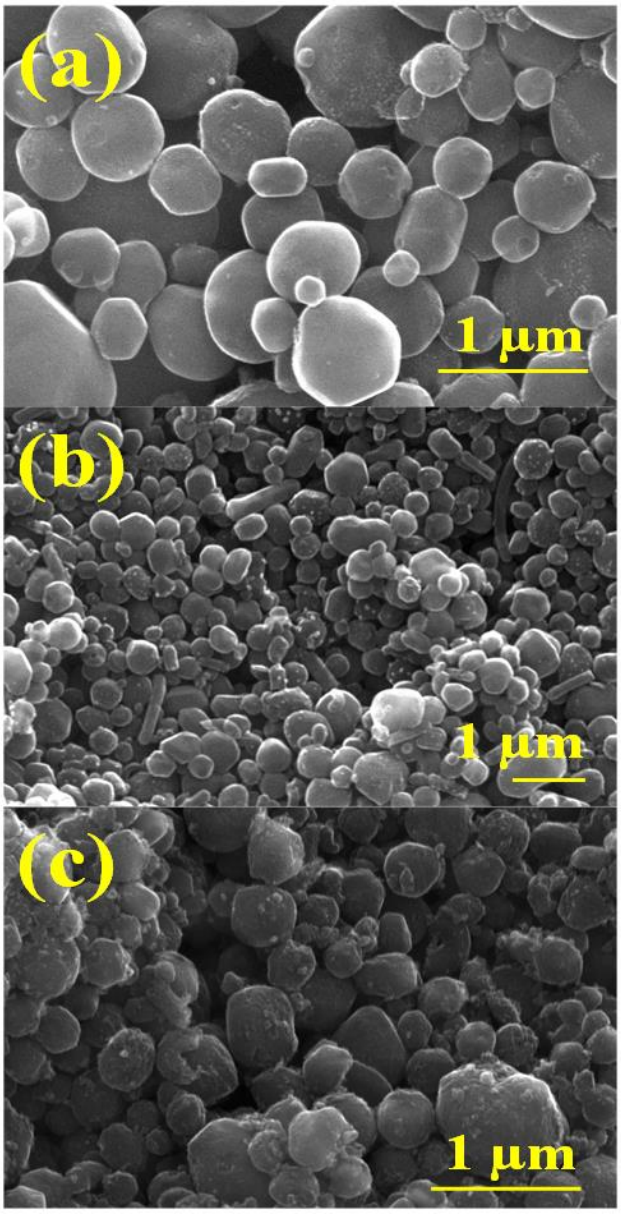

Figure S1. Scanning electron microscopy (SEM) images of (a) OMCS-F1.11, (b) OMCS-F1.37 and (c) OMGCS-9 samples (F1.11 and F1.37 denote 1.11 g and 1.37 g of F127 used in the synthesis, respectively).

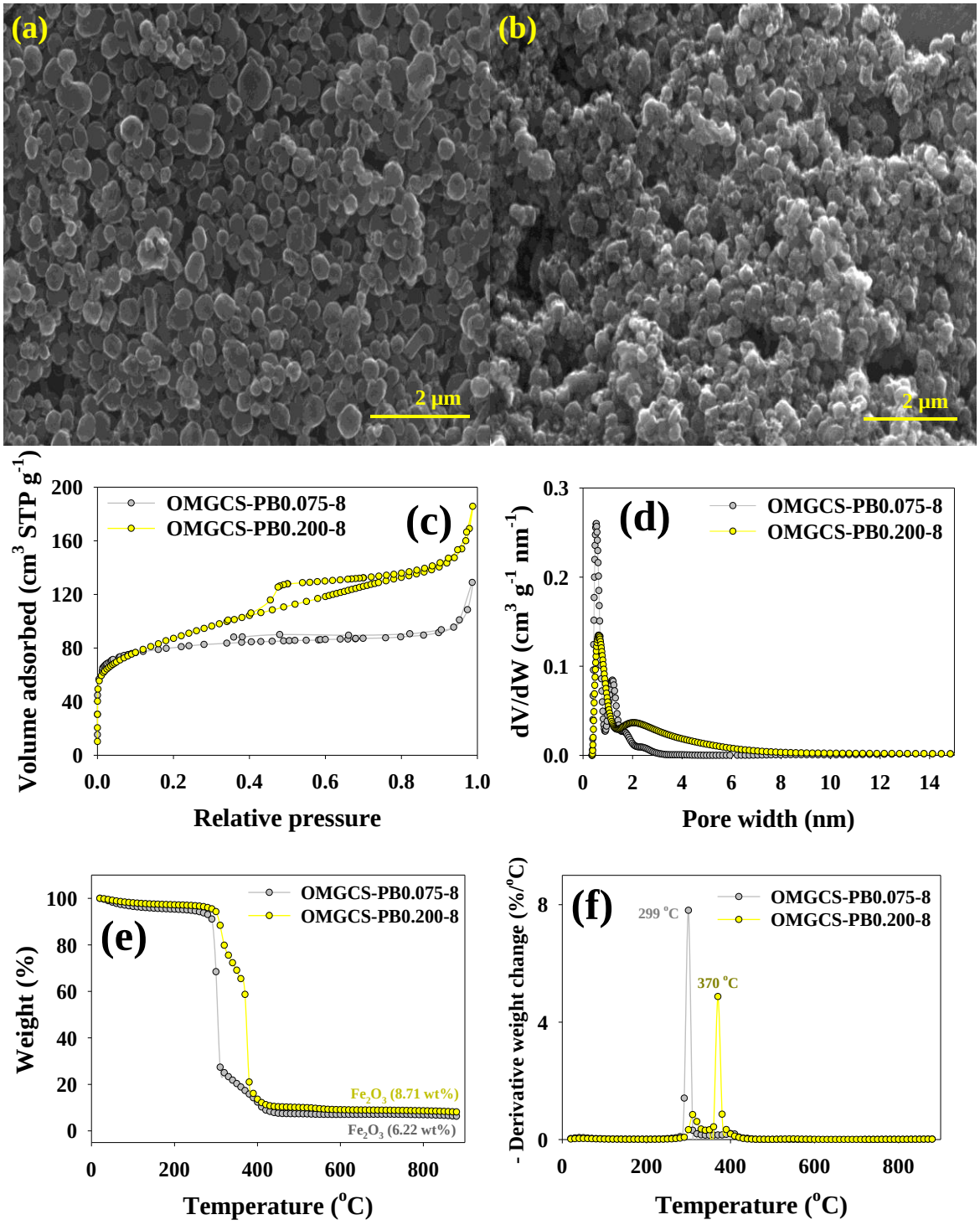

Figure S2. Scanning electron microscopy (SEM) images of (a) OMGCS-PB0.075-8 (b) OMGCS-PB0.200-8, (c) N_2 adsorption isotherms, (d) pore size distributions (PSDs), (e) thermogravimetric (TG) profiles and (f) differential thermogravimetric (DTG) profiles of the samples studied (PB0.075 and PB0.200 denote 0.075 g and 0.200 g of PB nanoparticles used in the synthesis, respectively).

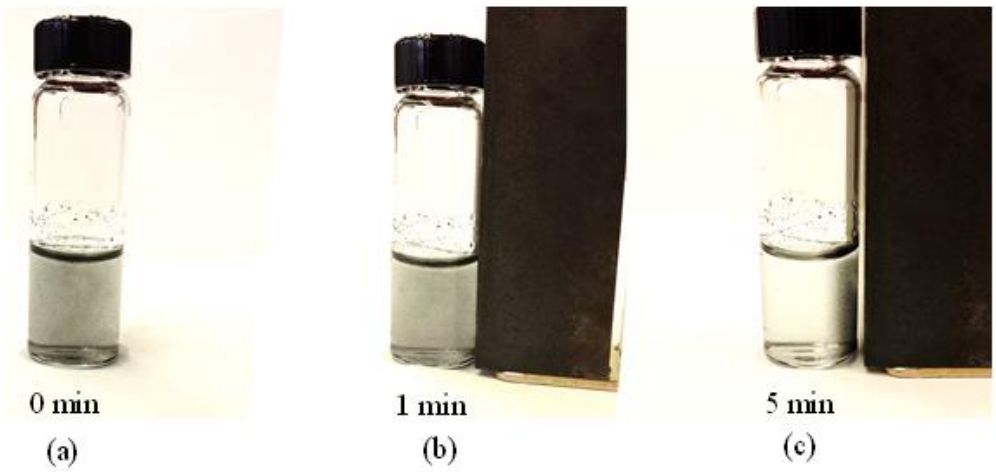

Figure S3. Magnetic separation of graphene nanoballs (CS-CPB0.1-6) from an aqueous solution by applying external magnetic field; (a) No magnetic field, (b) 1 min after placing the magnet, (c) 5 min after placing magnet; complete separation

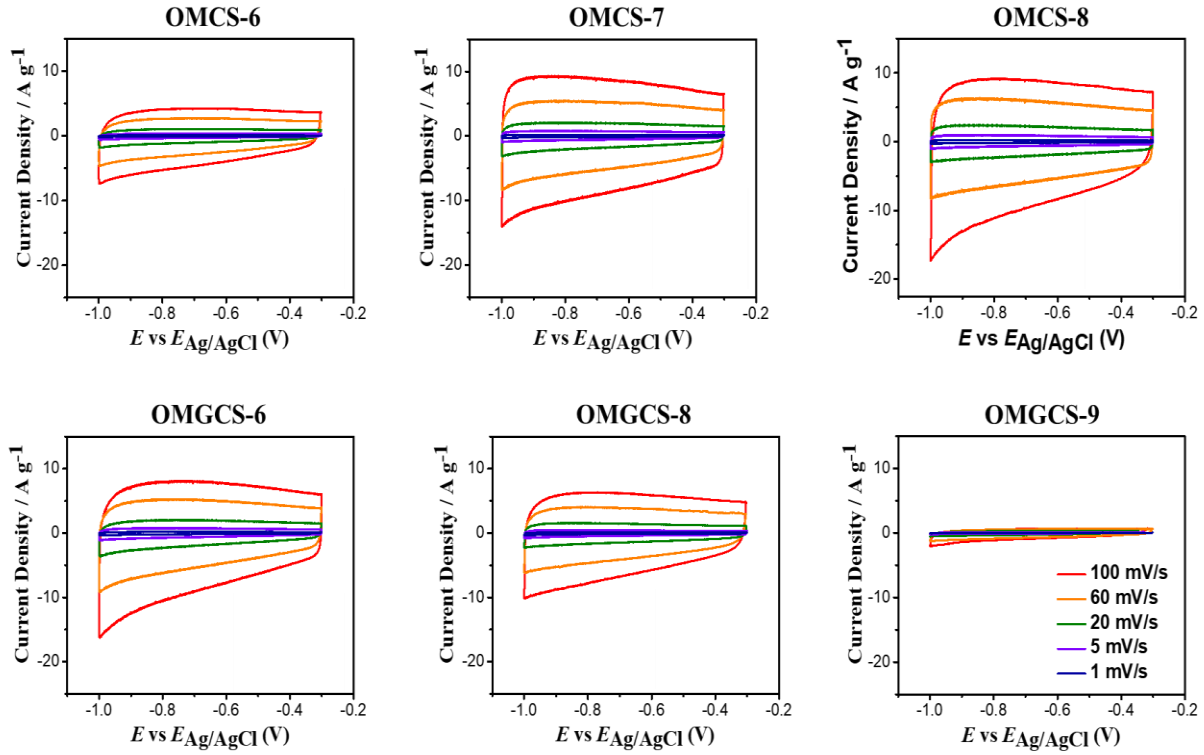

Figure S4. Cyclic voltammograms of the carbon samples studied in 6M KOH using scan rates of 100 mV/s (red), 60 mV/s (orange), 20 mV/s (green), 5 mV/s (purple), and 1 mV/s (blue).

Table S1. Specific capacitances for various carbon samples measured in 6M KOH electrolyte under 1 mVs^{-1} scan rate.

Sample	Specific Capacitance (Fg^{-1})
OMCS-6	83.0
OMCS-7	156
OMCS-8	176
OMGCS-6	166
OMGCS-7	247
OMGCS-8	116
OMGCS-9	26.7