Electronic supplementary information

Facile Preparation of Graphite Particles Fully Coated with Thin Ag Shell Layers for High Performance Conducting and Electromagnetic Shielding Composite Materials

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In Figure S2, it is seen that the initial pH value of the mixture of NGPs and oxalic acid is higher than that of oxalic acid solution, which is attributed to the base characteristic of NGPs just like active carbon or multiwalled carbon nanotubes. There exists some base groups on the surface of NGPs, such as ether-like, carbonyl-type and pyrone-type groups. These base groups on NGPs may consume a large proportion of $H^+$ from the oxalic acid solution before titration, and thus the concentration of base groups can be derived according to pH value difference between the initial points of two curves. Before their overlapping each other at point C, the red curve shows the relatively slow increase in pH value to the black one, due to buffering effect of acid group on NGPs and its conjugated base. According to carboxyl group ($pK_a < 5$) and phenol-type group ($pK_a = 8-9$), buffer zone of titration curve (red line) ranges pH value from 8 to 9, indicating that acid group on the surface of NGPs is dominated by phenol-type one at this time, not carboxyl one. Similar results have been previously reported on carbon nanofibers. The concentration of phenol-type ($C_{\text{phenol-type}}$) is given by equation (1):

$$C_{\text{phenol-type}} = \frac{C_{\text{NaOH}} V_{\text{NaOH(A)}}}{m}$$  \hspace{1cm} (1)

Where, $V_{\text{NaOH(A)}}$ and $m$ represent the abscissas of point A (that is to say, the volume of NaOH solution) and mass of NGPs, respectively. This result shows $C_{\text{phenol-type}} = 1.1 \times 10^{-5}$ mol/g. Associated with the specific surface area of NGPs (7.07 m$^2$/g), we managed to calculate the density of phenol-type group on NGPs, i.e. 0.94 groups per nm$^2$. 


Table S1. Density of hydroxyl group or silane on the different inner-core particles modified by trimethoxysilane.

<table>
<thead>
<tr>
<th>Cores</th>
<th>Hydroxyl density (nm$^{-2}$)</th>
<th>Silane density (nm$^{-2}$)</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZrP</td>
<td>4.2</td>
<td>2.4</td>
<td>5</td>
</tr>
<tr>
<td>SiO$_2$</td>
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<td>2.9</td>
<td>6</td>
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<tr>
<td>ZnO</td>
<td>11.3</td>
<td>11.3</td>
<td>8</td>
</tr>
<tr>
<td>NG</td>
<td>0.94</td>
<td>51.62</td>
<td>our work</td>
</tr>
</tbody>
</table>
**Figure S1.** Size distribution of NGPs.

**Figure S2.** Titration curves of oxalic acid solution (black line), oxalic acid containing NGPs mixture (red line). The pH value at point A is equal to that at the initial point of red line. Point C is the start of the overlapping point of these two titration curves.
Figure S3. Nitrogen adsorption-desorption isotherms of NGPs, MNGPs-3, MNGPs-6 and MNGPs-8.

Figure S4. SEM images of (a) NGPs and (b) MNGPs synthesized at $C_{\text{MPTMS}} = 52.0$ mmol/L.
Figure S5. (a) Low magnification and (b) high magnification SEM images of MNGPs synthesized at $C_{H_2O} = 37.07$ mol/L.

Figure S6. Influence of MNGPs with various $N_{SH}$ of (a) 0.80, (b) 1.06, (c) 10.11, (d) 17.56 and (e) 28.74 nm$^{-2}$ on the morphology of the as-synthesized NG@Ag CSCPs, other conditions as the typical experiment except for functionalization of NGPs. The insets show the corresponding high magnification images of these composite particles.
**Figure S7.** Influence of $N_{\text{SH}}$ of MNGPs on $N_{\text{Ag}}$ of NG@Ag CSCPs.

**Figure S8.** Cross-section morphology of NG@Ag CSCPs synthesized at respective $V_A$ of (a) 34.0, (b) 50.0 and (c) 64.0 mL, other conditions as the typical experiment except for $V_A$. Scale bars are all 100 nm.
Figure S9. Photographs of ESCMs containing 25 vol.% of (a, c) NG@Ag CSCPs and (b, d) flaky silver powders, respectively. (e) Photograph to show the coaxial fixture of shielding effectiveness test. As is shown in Figure S9, two pieces of ESCMs (180×180 mm) containing (a) NG@Ag CSCPs synthesized in the typical experiment and (b) flaky silver powders are prepared, respectively. Two pieces of ESCMs (annular wafer with outer radius of 55.00 mm, inner radius of 6.25 mm and thickness about 150 μm) containing (c) NG@Ag CSCPs and (d) flaky silver powders were patterned by an engraving machine, respectively. The scale bars in (a) and (b) are 100 mm; the scale bars in (c) and (d) are 25 mm.
Figure S10. Photographs of ECAs determination for flexibility as GB/T 1731-1993 standards, the volume fraction of as-synthesized composite particles as Figure S9. ECAs were coated on tinplate panels of 15 cm × 2.5 cm, its thickness is 200 μm.

Figure S11. Photograph of ECAs determination for impact resistance as GB/T 1732-1993 standards, the volume fraction of as-synthesized composite particles as Figure S9. The direct and indirect impact resistance are indicated by the region A and region B, respectively. ECAs were coated on tinplate panels of 15 cm × 5 cm, its thickness is 200 μm. The scale bar is 20 mm.
Figure S12. Photograph of ECAs determination for cross-hatch adhesion as GB/T 9286-1998 standards, the volume fraction of as-synthesized composite particles as Figure S9. ECAs were coated on FR-4 panels of 18 cm × 7 cm, its thickness is 200 μm. The scale bar is 5 mm.

Figure S13. Effect of aging time on the electrical conductivity of ECAs, the volume fraction of as-synthesized composite particles as Figure S9.
REFERENCES


