Electronic Supplementary Information

The activation temperature and oxidation time are two main factors which influence the exfoliation process. And these two parameters were also studied in this work. The samples activated at various temperatures were named as "temperature-A" (eg. GOC activated at 300 °C was signed as "300-A"). When these samples were further oxidized for various times, they were marked as "temperature-A-time-O" (eg. GOC activated at 300 °C and further oxidized for 96 h was named as "300-A-96 h-O"). And after the rapid thermal expansion process, each of the product was signed as "temperature-A-time-O-E", respectively (eg. GOC activated at 300 °C, oxidized for 96 h and expanded at 1050 °C for 30 s was named as "300-A-96 h-O-E").



Figure S1 XRD patterns and Raman spectra of (a) and (b) the derivations from various temperature activations of GOC and further 96 h oxidation; (c) and (d) products after rapid thermal expansion of the derivations mentioned in (a) and (b).

In our previous works on KOH activation of GOC, it is found that the introduced defect density of GOC is much dependent on the activation temperature, and at the reaction temperature of 600 °C, the maximum defect density was introduced into GOC¹. Here, the influence of defect density

on the oxidation and thermal expansion of activated GOC was investigated and the resulting XRD patterns and Raman spectra were shown in Figure S1. On the XRD curve of 300-A-96h-O (Figure S1a), although the graphitic stacking peak became broad and weakened compared with that of GOC, the peak position (at ca. 23 °) almost did not changed, which indicated that, with a low defect density after 300 °C activation, GOC structure still remained inert for the oxidation and intercalation and few functional groups were decorated on the interlayer space. However, after activation under 400 °C, the stacking peak (ca. 11.2 °) shift could be clearly observed in the pattern of 400-A-96h-O despite a much weaker and broader graphitic stacking peak still remained at ca. 24°. Then, the graphitic stacking peak totally disappeared on the pattern of 600-A-96h-O and reappeared on that of 700-A-96h-O. All those indicated that the sample of 600-A-96h-O has the highest oxidation degree. Based on XRD patterns presented above, we deduced that the opening up of the activated GOC is much dependent on the defect density, that is, higher defect densities on GOC facilitate the oxidation process, leading GOC to a deeper oxidation degree. However, according to the Raman spectra shown in Figure S1b, the defect densities on all products are approximately equal to each other. It is because that, after 96 h oxidation, most of the defect areas (which could also be seen as the active sites) was totally etched.

On the structural transformation of GOC shown in Figure 1a, we mentioned that after rapid thermal expansion, the additional functional groups were removed and the interlayer space of 600-A-96h-O-E (PGNs) was reduced compared with that of 600-A-96h-O (TGO). Here in Figure S1c, the stacking peak diffraction intensity of 300-A-96h-O-E is much higher than those of other expansion products. Since the 300-A-96h-O exhibited the lowest oxidation degree, it could be easily understand that 300-A-96h-O-E still remained in a pristine graphitic stacking structure so that expansion could barely occur in this material. The exfoliation degree of the expansion products could be illustrated by the different diffraction intensity of the stacking peaks at around 24.5 °. When the obtained oxidized product performed a higher oxidation degree, after thermal treatment, the expansion product would show lower stacking peak intensity. Therefore, the sample 600-A-96h-O-E, prepared from the precursor with the lowest peak intensity at ca. 24.5 °, can be considered as owning the highest exfoliation state. This is the reason why we choose the series of 600 °C activated GOC to investigate other impact factors on exfoliation phenomena. The Raman spectra of the all the expansion samples (Figure S1d) are also nearly the same.



Figure S2 XRD patterns and Raman spectra of (a) and (b) the derivations from 600 °C activation of GOC and further different time oxidation, (c) the ID/IG ratios of Figure S1 (b); (d) and (e) products after rapid thermal expansion of the derivations mentioned in (a) and (b), (f) the ID/IG ratios of Figure S1 (e).

The reaction time of oxidation is another important factor that largely influences the exfoliation and expansion degree. To further understand the relationships, we select the GOC derivation 600-A and its oxidation, expansion products as samples for the investigation. On the XRD pattern of 600-A-48h-O (Figure S2 a), the stacking peak remained at ca. 23.5 ° even though it became weak and broad. This is quite similar to that of 300-A-96h-O. 600-A-48h-O also exhibited low oxidation degree which is ascribed to the lacking of oxidation time. For the samples of 600-A-96h-O, 600-A-168h-O and 600-A-240h-O, they showed the similar peak shifting on their XRD curves from 23.5 ° to about 10 °. Raman spectroscopy was further taken to examine the transformation of carbon structures and the results are shown in Figure S2 b. As we proposed, the defect areas introduced by activation could be etched during the oxidation process. Here, the defect densities of the products were reduced with the elongation of the oxidation time, which could be seen from the ratio of I_D to I_G given in Figure S2 c. This evidence reconfirms the significant effect of defect densities on the oxidation-exfoliation processes as we suggested. As 600-A-48h-O was not totally exfoliated, after expansion, it showed high stacking peak intensity, which is also similar to that of 300-A-96h-O-E (Figure S2 d). Besides, with the elongation of oxidation time, after expansion, a shoulder peak at ca. 22 $^{\circ}$ appeared on the XRD curve of 600-A-168h-O-E and intensified on that of 600-A-240h-O-E. As discussed before, this peak characteristically disclosed the disorder turbulent carbon structure. Here, the XRD analysis results indicated that the long-time oxidation seriously destroyed the graphitic structure and some amorphous regions among deeply oxidized products couldn't be totally repaired by rapid thermal treatment. This phenomenon could also be proven by the Raman spectra given in Figure S2 e and f, where the ratio of I_D/I_G was increased from 600-A-96h-O-E to 600-A-240h-O-E.

References

1. S. Zhang, H. Song, J. Zhang, L. Zhu, X. Chen, H. Zhang and J. Zhou, RSC Adv., 2013, 3, 6117-6122.