$\ln d_g$. These findings offer strategies to maximize the *ER* of graphite in the microwave-assisted exfoliation. First of all, natural, flake-type graphite having large particle size (the larger, the better) is preferred to achieve the industrial-scale production of defect-free graphene through *ER* maximization. Secondly, there is a flooring value for graphite size (less than 4.7 to 9.4 μm depending on the form and type of graphite) that can be exfoliated by microwave irradiation (Figs 4a-d).

E. Reduction Process Using Benzyl Alcohol

Together with Raman spectrometer and XPS, the changes in the extent of oxygen through reduction process with benzyl alcohol were measured for raw graphite, EG with reduction process, and EG without reduction process, thus examining the effect of the reduction process on C:O ratio. For 2 kinds of samples (i.e., EG with reduction process and EG without reduction process), their Raman spectra were recorded, as shown in Fig. 5a. The Raman spectra of EG with reduction process and EG without reduction process showed G band at 1584 cm^{-1}, 2D band at 2673 cm^{-1}, and almost no line at 1370 cm^{-1}, which were well matched with the reported fundamental Raman modes for graphite [15]. This shows qualitatively that the EGs prepared by microwave irradiation have C:O ratio similar to raw graphite. We also performed a quantitative analysis on C:O ratio in EG with reduction process and EG without reduction process using XPS. The C 1s core level spectra of the samples (especially, EG with reduction process) were deconvoluted into 3 peaks of 284.95, 285.90, and 292.77 eV corresponding to C-C, C-O, and pi-pi moieties, respectively. The chemical fractions of C-C and C-O existed in each sample were calculated on the basis of the area of the individual peaks and the atomic sensitivity factor (Fig. 5b). The C:O ratios measured from EG with reduction process and EG without reduction process were 6.41:1 (13.5%) and 10.49:1 (8.7%), respectively. This conforms that the microwave-assisted exfoliation method employed in this study produces EG having a relatively low oxygen content of 13.5% and the oxygen content of the EG decreases slightly but not significantly (8.7%) with the reduction process.

IV. Conclusions

The expanding nature of graphite in the microwave-assisted exfoliation has been understood to build the backbone for the industrial-scale production of defect-free graphene and oil adsorption material. In detail, the dependence of the *ER* of graphite on mechanophysical parameters (e.g., mixing ratio, mixing time, microwave irradiation time, and graphite properties) can be summarized as follows: among the combination of graphite, $KMnO_4$, and HNO_3 amounts, a mixing ratio of 1:1:2 is desirable to achieve the maximum *ER*; 5 minutes of agitation produce the maximum *ER* of graphite; a microwave irradiation time of 60 or larger seconds is enough to fully expand graphite; natural, flake-type graphite having large particle size is preferred. Noticeably, the last phenomenon can be predicted from our fluid-dynamics-based model. Extrapolation of this study to other exfoliation method might help us to investigate the critical expanding nature of graphite, therefore mass-producing high-quality graphene.

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References

[1] R.K. Flitney, "Soft packings," Tribology International, vol. 19, pp. 181, 1986.

[2] W.W. Focke, H. Badenhorst, W. Mhike, H.J. Kruger, and D. Lombaard, "Characterization of commercial expandable graphite fire retardants," Thermochimica Acta, vol. 584, pp. 8-16, 2014.

[3] H. Fukushima, L. Drzal, B. Rook, and M. Rich, "Thermal conductivity of exfoliated graphite nanocomposites," Journal of Thermal Analysis and Calorimetry, vol. 85, pp. 235-238, 2006.

[4] S.D. Chakarova-Käck, E. Schröder, B.I. Lundqvist, and D.C. Langreth, "Application of van der Waals density functional to an extended system: adsorption of benzene and naphthalene on graphite," Physical Review Letters, vol. 96, pp. 146107, 2006.

[5] K.R. Paton, E. Varrla, C. Backes, R.J. Smith, U. Khan, A. O'Neill, et al., "Scalable production of large quantities of defect-free few-layer graphene by shear exfoliation in liquids," Nat Mater., vol. 13, pp. 624-630, 2014.

[6] F. Bonaccorso, A. Lombardo, T. Hasan, Z.P. Sun, L. Colombo, and A.C. Ferrari, "Production and processing of graphene and 2d crystals," Mater Today, vol. 15, pp. 564-589, 2012.

[7] A.M. Dimiev and J.M. Tour, "Mechanism of graphene oxide formation," Acs Nano, vol. 8, pp. 3060-3068, 2014.

[8] T. Wei, Z.J. Fan, G.L. Luo, C. Zheng, and D.S. Xie, "A rapid and efficient method to prepare exfoliated graphite by microwave irradiation," Carbon, vol. 47, pp. 337-339, 2009.

[9] Y.W. Zhu, S. Murali, M.D. Stoller, A. Velamakanni, R.D. Piner, and R.S. Ruoff, "Microwave assisted exfoliation and reduction of graphite oxide for ultracapacitors," Carbon, vol. 48, pp. 2118-2122, 2010.

[10] D.R. Dreyer, S. Murali, Y.W. Zhu, R.S. Ruoff, and C.W. Bielawski, "Reduction of graphite oxide using alcohols," J Mater Chem., vol. 21, pp. 3443-3447, 2011.

[11] E.H.L. Falcao, R.G. Blair, J.J. Mack, L.M. Viculis, C.W. Kwon, M. Bendikov et al., "Microwave exfoliation of a graphite intercalation compound," Carbon, vol. 45, pp. 1367-1369, 2007.

[12] O.Y. Kwon, S.W. Choi, K.W. Park, and Y.B. Kwon, "The preparation of exfoliated graphite by using microwave," J Ind Eng Chem., vol. 9, pp. 743-747, 2003.

[13] S.H. Yoon and Y.H. Cho, "High-precision digital microflow controllers using fluidic digital-to-analog converters composed of binary-weighted flow resistors," J Microelectromech S., vol. 15, pp. 967-975, 2006.

[14] S. Iijima, "Helical microtubules of graphitic carbon," Nature, vol. 354, pp. 56-58, 1991.

[15] A.C. Ferrari, J.C. Meyer, V. Scardaci, C. Casiraghi, M. Lazzeri, F. Mauri, et al., "Raman spectrum of graphene and graphene layers," Physical Review Letters, vol. 97, pp. 187401, 2006.