

Crumpling of a pyrolytic graphite sheet

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Crumpled graphite thin film balls were fabricated with Panasonic Pyrolytic Graphite Sheets (PGS). The fractal dimension, mechanical properties, and electrical conductivity of the crumpled PGS balls have been investigated. The universal local fractal dimension of the PGS balls is found to be 2.58, which is consistent with that of paper balls. The crumpled PGS balls show good mechanical property with Young's Modulus of 16–17 N, which is about the same as that of paper balls and elastoplastic paper balls, but with much smaller sizes, thinner film thicknesses, and less weight. In addition, the crumpled PGS balls show good conductivity, slightly higher than that of the PGS film before crumpling. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4827842>]

I. INTRODUCTION

Studies on thin sheets and their related materials are of both fundamental and practical importance in materials science. In some cases, thin films are found useful in a more compact, crumpled, ball-like form; for example, the crumpled paper are used for the fillers for moving or shipping boxes. The dimensional transition from flat sheets to three-dimensional (3D) crumpled balls renders the thin films good compression and mechanical properties.^{1–3} In modern research, random folding of thin-film materials is of noteworthy importance to many branches of science and industry,⁴ such as nanomechanical architecture of solid nano-membranes and polymerized membranes,^{5–8} folded engineering materials,⁹ geological formations,¹⁰ or even for aggregation-resistant nanosheets.¹¹ And the crumpled materials used in experiments have been made of paper,^{12–15} mylar,^{2,16} aluminum foil,¹⁷ and layers of cream.¹⁸

Graphite films are layer-structured materials formed by stacking two-dimensional single-atomic-layer graphene sheets weakly coupled by van der Waals interaction. Graphene is known as one of the strongest 2D material in nature,^{19,20} which also exhibits excellent electrical and thermal conductivities,^{21–23} makes graphene promising for versatile applications in the fields of solar cells,²⁴ conductive films,²⁵ or even microwave adsorptions.^{26,27} Therefore, it will be very interesting to find out what if one makes graphene into crumpled balls.

In this article, we report on a novel crumpled graphite thin film balls fabricated with Panasonic Pyrolytic Graphite Sheets (PGS). The universal local fractal dimension, mechanical and compression properties, and electrical conductivities of the crumpled PGS balls have been investigated. The crumpled PGS balls show a Young's Modulus of 16–17 N which is comparable with that of paper and elastoplastic crumpled balls, but with much smaller sizes, thinner film thicknesses, and less weight. The crumpled PGS balls

also show good conductivity similar to the PGS film before crumpling.

II. EXPERIMENTAL

A. Materials

EYGS1218 PGS films were supplied by Panasonic Electronic Components. The PGS films we used were 180 ± 5 mm long, 115 ± 5 mm wide, and about 17 ± 5 μ m thick. The density of EYGS1218 PGS films was about 2.10 g/cm³, which was consistent with the density of regular graphite (2.09–2.23 g/cm³). The PGS have good electrical and thermal conductivity, which are 1000–1750 W/(m·K) and 10 000–20 000 S/cm, respectively.^{28,29}

B. Fabrication of PGS crumpled balls

To make the crumpled PGS balls, the PGS films are randomly folded by hand to ball-shape. We have chosen three different sizes of PGS films, which are the original 18 mm \times 11.5 mm films, half of the original (11.5 mm \times 9 mm) and a quarter of the original film (9 mm \times 5.75 mm), to make three different sizes of the crumpled PGS balls, having the diameters of \sim 16.0 mm, 12.7 mm, and 8.3 mm, respectively, as measured by averaging the diameters of three orthogonal directions of the crumpled PGS balls using a digital caliper. The typical PGS films before and after crumpling are shown in Fig. 1.

C. Characterization

To study the mechanical behavior of the crumpled PGS balls, we used a Instron Model 4206 Tensile Tester spherical indentation (with a force limitation of 1000 N) to measure the force (F) versus compression ($\lambda = H/R$ see Fig. 2) curves under uniaxial compression with a constant displacement rate $v = 0.1$ mm/s. The setup of the uniaxial compression test is shown in Fig. 2, for both the loading and unloading test of the crumpled PGS balls. Furthermore, to characterize the electrical properties of the PGS balls, we used a Keithley 2420 Sourceter to measure the I-V curves in a voltage range of -0.10 V to 0.10 V.

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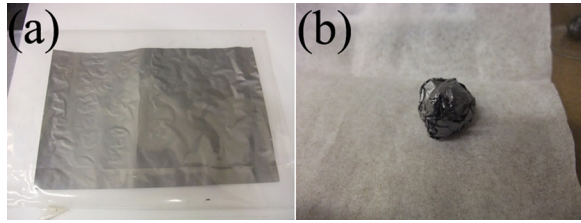


FIG. 1. Panasonic PGS, (a) and crumpled PGS balls (b).

III. RESULTS AND DISCUSSION

The dimensions of crumpled balls are different from regular 2-D or 3-D structures. Fractal dimension (D) is used to describe the crumpled-ball structure, which characterizes the typical relation between its diameter (R) and mass (M) using the following equation:¹²

$$R = kM^{1/D}. \quad (1)$$

Figure 3 shows a log-log plot of the R versus M for our crumpled PGS balls with diameters of ~ 16.0 mm, 12.7 mm, and 8.3 mm. From the plot, we extract the fractal dimension of the crumpled PGS balls to be $D \sim 2.58$. The result is consistent with the universal local fractal dimension of 2.51 or 2.64 of the crumpled paper balls.^{12,30} This means the folding characteristics of the PGS films are same as paper.

Figures 4(a) and 4(b) show typical force (F)-compression ($\lambda = H/R$) behaviors of crumpled PGS ball with diameters of 12.7 mm and 15.8 mm under uniaxial compression. The deformation of the crumpled PGS balls is essentially irreversible (see the image in Fig. 2(c) and unloading curve in Fig. 4(a)). The inset in Fig. 4(b) shows the force-compression curve $F(\lambda = H/R)$ in a log-log plot, which does not obey the normal power-law scaling for $F \propto \lambda^{-2}$.³¹ Edwards and co-workers have worked on the granular materials and attempted to develop a statistical mechanics approach for the inherent states of crumpling networks.^{32,33} Balankin and Huerta, who have followed Edwards' work on the crumpling networks, suggested that the loading part of experimental force-compression curve might be precisely fitted by the following relationship:⁴

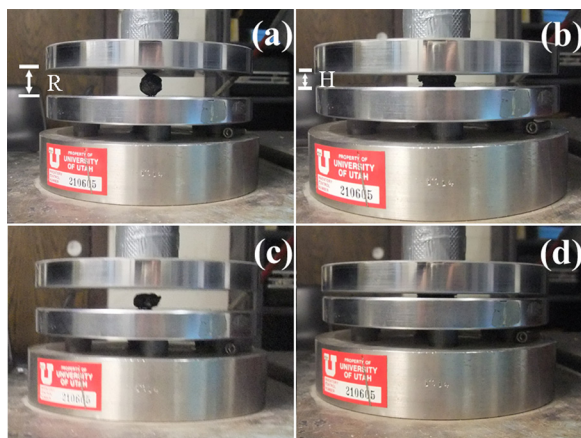
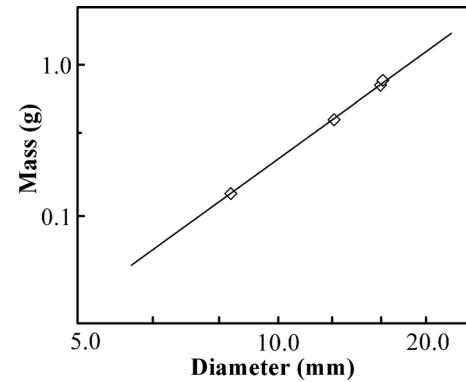


FIG. 2. Setup of axial compression test of PGS balls: (a)-(b) loading, (c) unloading, and (d) loading to the indentation instrument limit (1000 N).

FIG. 3. Diameter (R) versus mass (M) curve of the crumpled PGS balls fabricated by different PGS sizes. The fractal dimension is about 2.58.

$$F = -Y \left(\frac{1-c}{\lambda-c} - 1 \right). \quad (2)$$

Using Eq. (2), we can fit nicely the force-compression curves of the crumpled PGS balls under axial loading, as shown in Figs. 4(c) and 4(d), respectively. This indicates that the randomly folded PGS balls also obey the same force-compression relation under uniaxial loading as was found for randomly folded elastoplastic paper or hyperelastic sheets in literature.⁴ From the fitting, we extract the Young's Modulus for the 12.7 mm and 15.8 mm crumpled-ball samples to be 16.69 N and 16.30 N, respectively, which are comparable with that of ~ 400 mm paper balls in literature.⁴ We have also repeated and tested some ~ 16 mm PGS crumpled-ball samples. The samples show Young's Modulus around 16–17, which are similar to the 15.8 mm sample (as shown in the inset of Fig. 4(d)). Furthermore, in order to study the limitation of compression for the PGS balls, we have loaded with a 15 000 lb force to press the PGS ball with a diameter of 15.8 mm by a laboratory press carver. The height of the PGS ball is about 0.73 mm after the 15 000 lb compression, for which λ is about 0.046.

Beside the material properties, the Young's modulus is also highly dependent on the crumpled-ball sizes and thin film thicknesses as a result of Balankin-Huerta's research.⁴ In order to compare the crumpled PGS balls more intuitively with other crumpled materials, we have also used copy papers with the same size of EYGS12128 PGS (about 18 mm \times 11.5 mm) and with a thickness of about 0.060 mm to fabricate crumpled paper balls with a diameter of around 20 mm. The compression properties of the paper balls were also measured by the spherical indentation and one typical force-compression curve is shown in Fig. 4(e). The crumpled paper balls behave similar compression properties as crumpled PGS balls, and the Young's Modulus can also be fitted and calculated by Eq. (2), which is shown in Fig. 4(f). These results indicated that our novel crumpled PGS balls have comparable Young's Modulus (16–17 N) with that of 18.18 N of copy paper, and 13.67 N of elastoplastic paper in the literature,⁴ but with much smaller sizes, thinner film thicknesses, and less weight.

The electrical properties of the PGS barely changed after crumpling. The I-V curves of a whole piece of PGS film

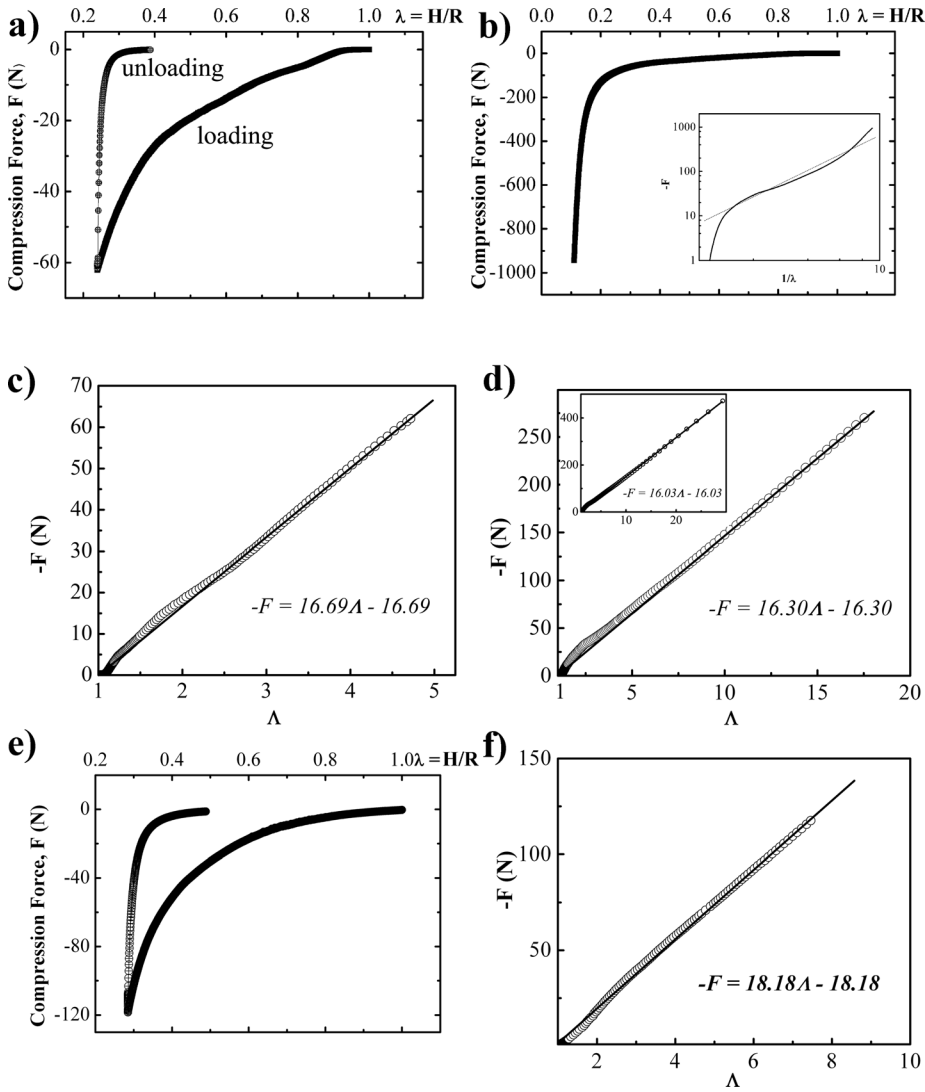


FIG. 4. (a) and (b) The force (F) versus compression ($\lambda = H/R$) curve of the crumpled PGS balls with diameters (R) of 12.7 mm (a) and 15.8 mm (b) under uniaxial compression. The inset in (b) shows the force-compression curve $F(\lambda = H/R)$ in log-log coordinates, which does not obey the normal power-law scaling for $F \propto \lambda^{-2.31}$. (c) and (d) The fitted curves of (a) and (b) by Eq. (2), which show Young's Modulus of about 16.69 and 16.30, respectively. $\Lambda = (1-c)/(\lambda-c)$, c as fitting parameter. The circles show the experimental data and the straight lines show the fitted linear correlation. The inset in (d) shows the fitted force-compression curve of one of the repeated sample, showing a Young's Modulus of 16.03, which is similar with the 15.8 mm sample. (e) The F versus λ curve of a crumpled copy paper ball, and the diameter (R) is about 20 mm. (f) The fitted curve of (e).

before and after crumpling were measured in a voltage range of -0.10 V to 0.10 V, which are shown in Fig. 5. The linear behavior of the I-V curves indicated that the PGS film and its crumpled PGS ball can be considered as resistors under low voltage. In Fig. 5, the crumpled PGS ball shows good conductivity with a resistance of a few ohms, which is even slightly higher than that of PGS film. This may come from the crosslink of the graphite sheets in the crumpled ball structure.

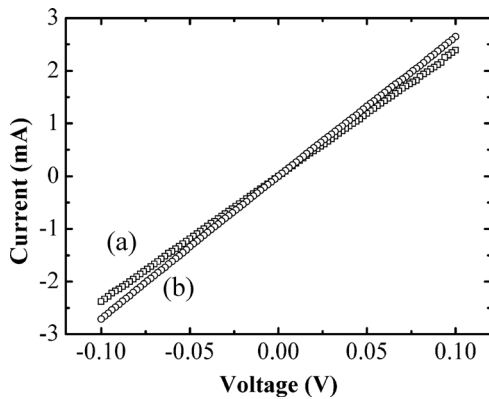


FIG. 5. I-V curve of the PGS sheets before (a) and after crumpling (b).

IV. CONCLUSIONS

Novel crumpled graphite thin film balls were fabricated with Panasonic PGS. The universal dimension of these crumpled PGS balls is about 2.58, which is consistent with that of crumpled paper balls. The crumpled PGS balls show good mechanical property with Young's Modulus of 16–17 N which is comparable with that of paper balls and elastoplastic paper balls, but with much smaller sizes, thinner film thicknesses, and less weight. In addition, the crumpled PGS balls show good conductivity, slightly higher than that of the PGS film before crumpling. The conductive crumpled PGS balls with good mechanical properties show good potentials for mechanical and electrical applications.

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- ¹T. Tallinen, J. A. Astrom, and J. Timonen, *Nature Mater.* **8**, 25 (2009).
- ²K. Matan, R. B. Williams, T. A. Witten, and S. R. Nagel, *Phys. Rev. Lett.* **88**, 076101 (2002).
- ³A. Lobkovsky, S. Gentges, H. Li, D. Morse, and T. A. Witten, *Science* **270**, 1482 (1995).
- ⁴A. S. Balankin and O. S. Huerta, *Phys. Rev. E* **77**, 051124 (2008).
- ⁵R. Lipowsky, *Nature* **349**, 475 (1991).
- ⁶M. J. Bowick and A. Travesset, *Phys. Rep.* **344**, 255 (2001).
- ⁷M. Huang, C. S. Ritz, B. Novakovic, D. Yu, Y. Zhang, F. Flack, D. E. Savage, P. G. Evans, I. Knezevic, F. Liu, and M. G. Lagally, *ACS Nano* **3**, 721 (2009).
- ⁸M. Huang, T. Boone, M. Roberts, D. E. S. M. G. Lagally, N. Shaji, H. Qin, R. Blick, J. A. Naim, and F. Liu, *Adv. Mater.* **17**, 2860 (2005).
- ⁹T. A. Witten, *Rev. Mod. Phys.* **79**, 643 (2007).
- ¹⁰A. J. Wood, *Physica A* **313**, 83 (2002).
- ¹¹J. Luo, H. D. Jang, T. Sun, L. Xiao, Z. He, A. P. Katsoulidis, M. G. Kanatzidis, J. M. Gibson, and J. Huang, *ACS Nano* **5**, 8943 (2011).
- ¹²M. A. F. Gomes, *J. Phys. A* **20**, L283 (1987).
- ¹³D. L. Blair and A. Kudrolli, *Phys. Rev. Lett.* **94**, 166107 (2005).
- ¹⁴A. S. Balankin, O. S. Huerta, R. C. M. de Oca, D. S. Ochoa, J. M. Trinidad, and M. A. Mendoza, *Phys. Rev. E* **74**, 061602 (2006).
- ¹⁵C. A. Andresen, A. Hansen, and J. Schmittbuhl, *Phys. Rev. E* **76**, 026108 (2007).
- ¹⁶A. Boudaoud, P. Patricio, Y. Couder, and M. B. Amar, *Nature* **407**, 718 (2000).
- ¹⁷A. S. Balankin, I. C. Silva, O. A. Martinez, and O. S. Huerta, *Phys. Rev. E* **75**, 051117 (2007).
- ¹⁸M. A. F. Gomes, C. C. Donato, S. L. Campello, R. E. de Souza, and R. Cassia-Moura, *J. Phys. D* **40**, 3665 (2007).
- ¹⁹C. Lee, X. Wei, J. W. Kysar, and J. Hone, *Science* **321**, 385 (2008).
- ²⁰C. Si, W. Duan, Z. Liu, and F. Liu, *Phys. Rev. Lett.* **109**, 226802 (2012).
- ²¹K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov, *Science* **306**, 666 (2004).
- ²²S. Yamanaka, T. Nishino, T. Fujimoto, and K. Yoshikazu, *Carbon* **50**, 5027 (2012).
- ²³Y. Pan, H. Zhang, D. Shi, J. Sun, S. Du, F. Liu, and H.-j. Gao, *Adv. Mater.* **21**, 2777 (2009).
- ²⁴M. Lagally and F. Liu, "Graphite-based photovoltaic cells" (Wisconsin Alumni Research Foundation, University of Utah Research Foundation, United States, 2010). Available at <http://www.google.com/patents/CA2680025A1>.
- ²⁵S. Stankovich, D. A. Dikin, G. H. Dommett, K. M. Kohlhaas, E. J. Zimney, E. A. Stach, R. D. Piner, S. T. Nguyen, and R. S. Ruoff, *Nature* **442**, 282 (2006).
- ²⁶Z. Liu, G. Bai, Y. Huang, F. Li, Y. Ma, T. Guo, X. He, X. Lin, H. Gao, and Y. Chen, *J. Phys. Chem. C* **111**, 13696 (2007).
- ²⁷Z. Ye, W. D. Deering, A. Krokhin, and J. A. Roberts, *Phys. Rev. B* **74**, 075425 (2006).
- ²⁸C.-Y. Wen, Y.-S. Lin, and C.-H. Lu, *J. Power Sources* **189**, 1100 (2009).
- ²⁹C.-Y. Wen and G.-W. Huang, *J. Power Sources* **178**, 132 (2008).
- ³⁰A. S. Balankin, R. C. M. de Oca, and D. S. Ochoa, *Phys. Rev. E* **76**, 032101 (2007).
- ³¹G. A. Vliegenthart and G. Gompper, *Nature Mater.* **5**, 216 (2006).
- ³²S. F. Edwards and R. B. S. Oakeshott, *Physica A* **157**, 1080 (1989).
- ³³R. Blumenfeld and S. F. Edwards, *Phys. Rev. Lett.* **90**, 114303 (2003).