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Formation of graphene aerosol micro-flakes by thermal shock followed by shock wave

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Abstract

Graphene and its derivatives are the promising additives in the composition of energetic materials [1-4]. A high-performance method of generation of few-layer crumpled graphene flakes in the aerosol phase is proposed. The range of sizes – from one to tens of micrometers, the number of layers in flake is about 3-10. The characteristics of the aerosol consisting of graphene microflakes were studied. Due to shock wave action, the fraction of particles acquires electrostatic charges that can be used for targeted deposition on the substrate surface.

1. Experiment

The starting material was an intercalated compound of fluorinated graphite of the composition $C_2F{\bullet}0.13ClF_3$, synthesized according to the method described in [5-7]. At the first stage, the powder sample of this substance was heated up to the temperature of 700-800°C. Decomposition proceeding in the regime of ‘‘thermal shock’’ (accompanied by flash, sound, significant volume increase and blackening) resulted in the formation of highly exfoliated graphite.

At the second stage, a sample of exfoliated graphite was placed in a pneumatic shock wave tube and, under the action of a shock wave, it was dispersed to form an aerosol consist of few-layer crumpled graphene microparticles. The pressure drop realized in the shock wave was 2-4 MPa.

The aerosol jet from the shock tube was directed into 8 liter cylindrical plastic vessel with an end wall from a plastic bag. After deposition of the particles on the substrates, the samples were prepared for optical and electron microscopy (Fig. 1).

2. Microflakes properties

It should be noted that precipitation of graphene particles inside the vessel was ubnormally rapid (time less than one hour). Fig. 2 shows the calculated dependence of the settling velocity of graphene microflakes on the equivalent

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"sedimentation" size. As follows from this calculation, a time interval of about 24 hours is required to sediment the representative fraction of the particles in the distribution.

Another feature is that the noticeable part of the sediment was deposited not only on the lower, but also on the side and top walls of the vessel (Fig. 3). This indicates that electrostatic interaction between aerosol microparticles and wall of plastic vessel take place. Probably this corresponds to the local sites of the wall where triboelectrization occurred. The formation of charges on graphene flakes is presumably due to the nonequilibrium process of breaking bonds between the layers of expanded graphite under the action of a shock wave. The range of sizes of microparticles extends from tenths to tens of microns; the main part has sizes 5 – 50 microns (Fig. 4). About 30% of the particles in the deposit were translucent, that is composed of a limited number of graphene layers. Basing on the optical absorption coefficient of graphene monolayer (2.3%), it was estimated that the thickness of the flakes is about 3 – 10 monolayers.

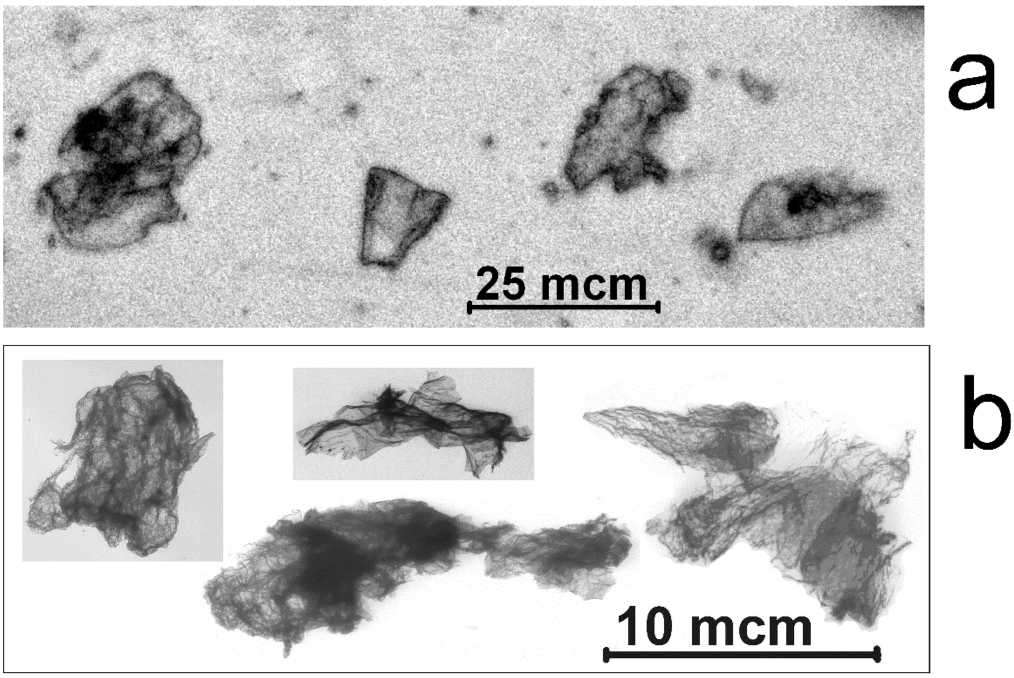

Fig. 1. Images of crumpled graphene flakes obtained by dispersing exfoliated graphite in a pneumatic shock wave: a) under optical microscope, b) under transmitting electron microscope JEM-100SX.

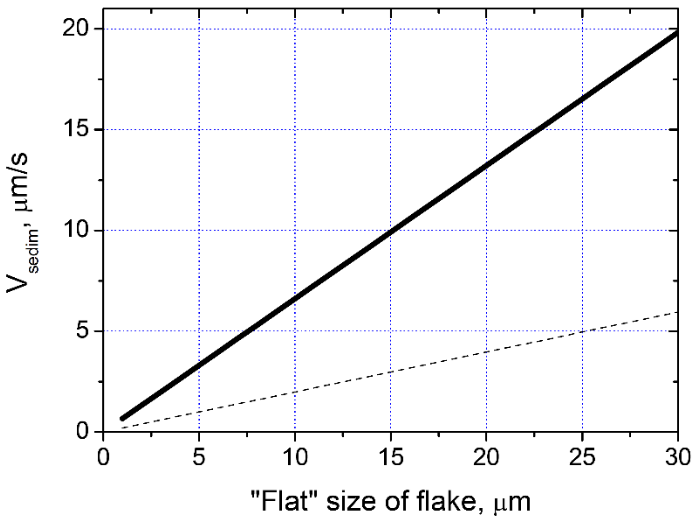

Fig. 2. The calculated dependence of the settling velocity of graphene microparticles on the equivalent "sedimentation" size: the solid line – ten-layer particle, the dash line – three-layer particle.

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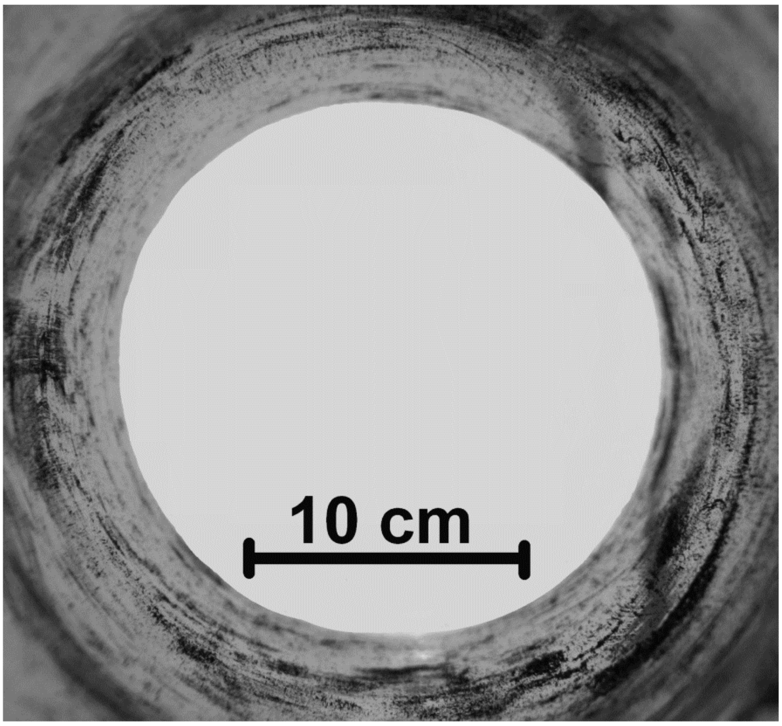

Fig. 3. View of non-uniform sediment of particles on the inner surface of the cylindrical vessel

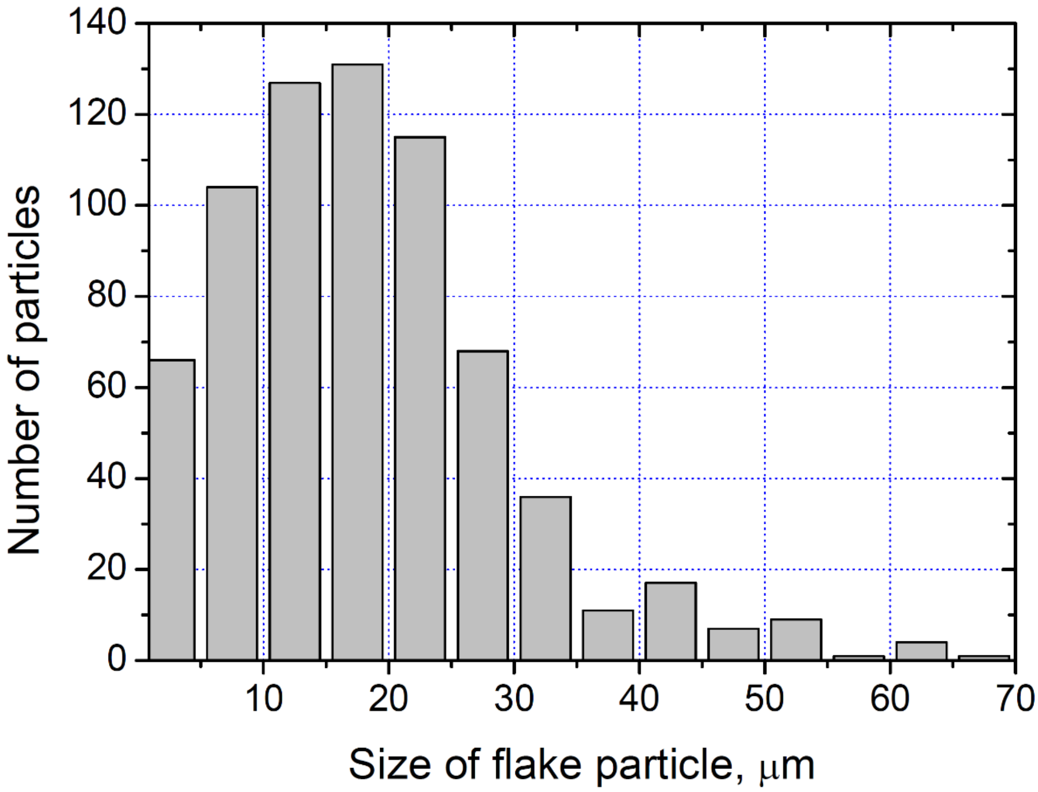

Fig. 4. Histogram of the size distribution of crumpled graphene flakes.

Conclusion

To produce the graphene particles, a method based on sonication of graphite (or graphene precursor) in a suspension is often applied. The paper [8] contains a summary table with comparative characteristics (yield and efficiency) from 15 publications on the production of graphene based on ultrasonic exfoliation in a liquid. Duration of this procedure is from tens of minutes to tens of hours.

So, high productivity of the generation of graphene microparticles is a significant advantage of the proposed shock wave method. Technically, this method can be realized in a unified cartridge, where the thermal shock and dispersing by a shock wave performed in series.

Acknowledgements

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References

[1] Li, R., Wang, J., Shen, J. P., Hua, C., & Yang, G. C. (2013). Preparation and characterization of insensitive HMX/graphene oxide composites. Propellants, Explosives, Pyrotechnics, 38(6), 798-804.
[2] Liu, T., Geng, C., Zheng, B., Li, S., & Luo, G. (2017). Encapsulation of Cyclotetramethylenetetranitramine (HMX) by Electrostatically Self-Assembled Graphene Oxide for Desensitization. Propellants, Explosives, Pyrotechnics, 42(9), 1057-1065.
[3] Sabourin, J. L., Dabbs, D. M., Yetter, R. A., Dryer, F. L., & Aksay, I. A. (2009). Functionalized graphene sheet colloids for enhanced fuel/propellant combustion. ACS nano, 3(12), 3945-3954.
[4] Isert, S., Xin, L., Xie, J., & Son, S. F. (2017). The effect of decorated graphene addition on the burning rate of ammonium perchlorate composite propellants. Combustion and Flame, 183, 322-329.
[5] Lee, J. H., Shin, D. W., Makotchenko, V. G., Nazarov, A. S., Fedorov, V. E., Kim, Y. H., ... & Yoo, J. B. (2009). One-step exfoliation synthesis of easily soluble graphite and transparent conducting graphene sheets. Advanced materials, 21(43), 4383-4387.
[6] Grayfer, E. D., Nazarov, A. S., Makotchenko, V. G., Kim, S. J., & Fedorov, V. E. (2011). Chemically modified graphene sheets by functionalization of highly exfoliated graphite. Journal of Materials Chemistry, 21(10), 3410-3414.
[7] Makotchenko, V. G., & Nazarov, A. S. (2009). Structural characteristics of dicarbon polyfluoride. Journal of Structural Chemistry, 50(6), 1088-1095.
[8] Yi, M., & Shen, Z. (2014). Kitchen blender for producing high-quality few-layer graphene. Carbon, 78, 622-626.