Tunable stability of microwave plasma synthesized few-layer graphene dispersion in water

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Formation of a stable dispersion of graphene in water is an important technological issue concerning both fundamental as well as applied research. Hydrophobic nature of graphene surface makes it very difficult to form stable dispersion of pure graphene, with high C/O atomic content ratio, in water without the use of surfactants. Up to now the most often used approach is the utilization of the graphene oxide (GO), oxidized formed of graphene with large quantities of carbon-oxygen/hydrogen related functional groups, which is well soluble in water/(surfactant) mixture. Such dispersion is applied to the selected surface and after liquid evaporation the reduction of the material to reduced graphene oxide is carried out. However, for the future large-scale applications, the use of dangerous chemicals as well as time and energy consuming processing steps should be minimized.

From this point of view, microwave plasma-based gas-phase synthesis of graphene [1] and its derivatives represents a simple, environmentally friendly, and easily scalable production method enabling emerging large-scale applications. Microwave plasma decomposition of organic precursors, hydrocarbons and alcohols, and subsequent formation of carbon nanostructures in the gas-phase is strongly dependent on the temperature and plasma plume stability of the environment formed by the plasma discharge [2]. Structure of the synthesized carbon material changes from graphitic nanoparticles to graphene nanosheets with increasing H/C atomic ratio of the precursor and delivered microwave power.

In our work, gas-phase synthesized few-layer graphene was prepared by high-temperature decomposition of ethanol in dual-channel microwave plasma torch at atmospheric pressure. Samples of few-layer graphene with different amount of structural disorder and oxygen content were prepared using variation of torch nozzle central channel gas flow (Qc = 500 – 920 sccm) and delivered microwave power (100-350 W). Ethanol flowrate was kept constant at 23 sccm carried out by 700 sccm of Ar. Lower power and increased flow rate of Ar in central channel led to the high-temperature environment instability (Figure 1) and structural and chemical changes of the material. More details can be found in our previous publications [2, 3]. After the synthesis, the nanopowder was scraped from the reactor walls and analyzed by electron microscopy, Raman and X-ray photoelectron spectroscopy (XPS). Plasma diagnostic was carried out by optical emission spectroscopy and ICCD camera imaging.

Fig. 1: Instability of torch discharge at high Qc.

Samples of graphene with different amount of disorder and oxygen related functional was used for preparation of dispersions in water. Dispersions were produced using deionized (DI) water containing 7.5 mg of graphene nanopowder in 500 ml of DI water. Three methods of preparation were compared: a) applying standard ultrasound for 10 minutes, b) applying more powerful cavitation technique and c) plasma treatment of nanopowder before ultrasonication. Plasma treatment of nanopowder was carried out in dielectric barrier discharge (DBD) using N_2 or N_2/propane-butane (N_2 – 6 slm /PB – 85 sccm) atmosphere for 120 s followed by ultrasonication in DI water for 10 minutes.

Prepared dispersions were left to settle down and regularly monitored (Figure 2). We can observe that graphene material exhibiting higher disorder – hdG type, as determined by higher D/G Raman band

ratio and lower sp^2/sp^3 bond ratio as well as higher oxygen content, 3 at% compared to 1 at% (G type sample), formed more stable dispersion and after the sedimentation phase of larger graphene clusters, 48 hours, the suspension became stable without any observable changes. We also observe positive influence of plasma treatment of the nanopowder in DBD, however according to the material analysis there were only small changes in the chemical composition of the material after plasma treatment.

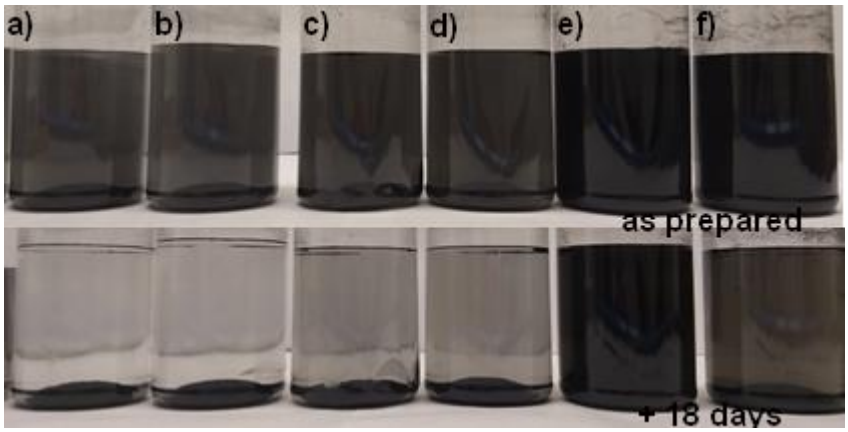

Fig. 2: Stability of few-layer graphene dispersion in water (type, preparation method): a) G, ultrasound b) hdG (ultrasound) c) G DBD N_2, d) G N_2/PB e) hdG N_2, f) hdG N_2/PB

In conclusion, direct control of structural and chemical properties of few-layer graphene during its synthesis using microwave plasma decomposition of ethanol at atmospheric pressure enabled us to control and improve graphene dispersion stability in water providing important insights into the formation mechanism of such dispersions and simplifying preparation process of such dispersions.

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