The influence of the type of graphite on the size of reduced graphene oxide

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Received May 16, 2022; accepted June 30, 2022; published June 30, 2022

Abstract—Reduced graphene oxide is a very attractive material for sensor applications. It exhibits high conductivity at room temperature and a high specific surface area. Since it can be produced in many ways, its properties can be influenced by the fabrication method. In this paper, we investigated the influence of graphite precursors (flake, scalar and synthetic) on the size of reduced graphene oxide. We have shown that the size of the precursor determines the size of the obtained rGO. We have noted that the larger the graphite size, the larger the rGO size.

Reduced graphene oxide (rGO) is a p-type semiconductor that consists of carbon and oxygen atoms [1–2]. rGO has a similar configuration and properties to prostine graphene [3–4], but unlike those of graphene, its carbon layers are decorated by residual oxygen-containing functional groups [2, 5], and it has structural defects [2], [6]. Reduced graphene oxide exhibits a huge surface area [7] and conductivity at room temperature. Therefore, potentially, it has many uses, e.g., in electronic devices [3] or as a gas sensing layer [6].

The method of production of rGO includes three steps: (i) oxidation, (ii) exfoliation, and (iii) reduction [8]. During the first step (oxidation), bulk graphite is oxidized to the form of graphite oxide (GO). In such a process, the functional groups are introduced between graphite layers [9, 10]. Next, GO is exfoliated into monolayers or a few layered stacks of GO by, e.g., sonication in water [11]. The final product, reduced graphene oxide, is obtained by the reduction of GO. The reduction can be carried out, among others, by chemical, photocatalyst, or thermal methods [2]. The thermal reduction is especially interesting because in such a process, the reduction is performed simultaneously with the exfoliation process, and, additionally, the degree of reduction can be easily controlled by temperature [12].

As can be seen, rGO can be produced in various ways. The selection of the production method may affect its properties. Therefore, in this paper, we investigated whether the use of different graphite precursors influences the size of rGO oxidated by the thermal method. For this purpose, we used three types of graphite precursors and one oxidation and reduction method. To measure the size of rGO flakes, we used the standard Atomic Force Microscopy (AFM) system.

We have prepared three reduced graphene oxides by using three different graphite precursors, two natural graphites: flake (Sinograf), scalar (AMG Graphite), and one synthetic (SGL Carbon). The diameters of graphite grains were: 82, 59, and 50 nm for flake, scalar, and synthetic graphite, respectively. The scheme of samples preparation is shown in Fig. 1.

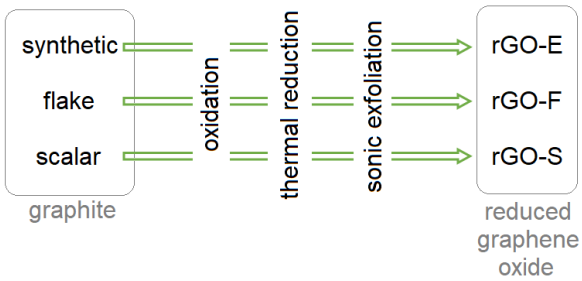

Fig. 1. Scheme of reduced graphene oxides preparation.

In the first step, all graphite precursors were oxidized according to Tour's modified method (graphite (1 g); H_2SO_4 (45 ml); H_3PO_4 (5 ml); KNO_3 (1.5g); $KMnO_4$ (5 g, time 5h). Next, the obtained graphite oxides were thermally reduced in 900°C and nitrogen atmosphere. After such a process, the products are partially reduced and partially exfoliated. Finally, the exfoliation process was performed. For this purpose, the samples were sonicated in a mixture of water/DMF for 30 minutes. The final products were only partially reduced and relatively well exfoliated (not completely). The obtained samples were named: rGO-F, rGO-S and rGO-E depending on the graphite precursor (flake: F, scalar: S and synthetic: E, respectively). To make AFM measurements, we deposited samples on mica substrates using a drop coating method (Fig. 2).

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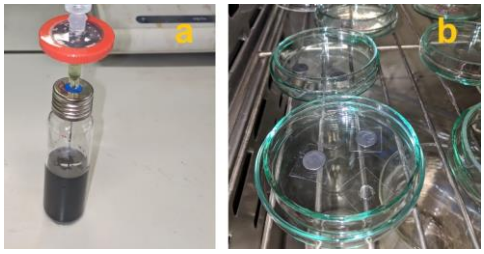

Fig. 2. (a) container with a sonicated solution before filtration through a syringe filter, (b) rGO solution after placing on a mica surface.

AFM measurements were performed on the NT-MDT, NTEGRA Prima operating in the intermittent contact mode. 256 points per line were recorded at a scan rate of 1 Hz by using the HA_NC probe. To ensure reliability, images were taken in multiple areas on the samples. The obtained data were analyzed using Nova Software.

The investigation carried out in various areas of the samples showed that regardless of the graphite precursor used, rGO has the shape of agglomerates (it is not a single layer) (*Fig. 3*).

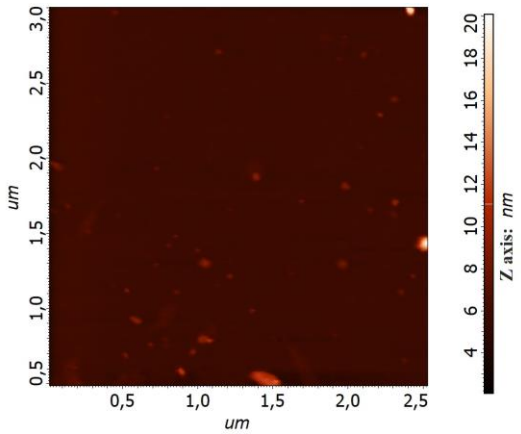

Fig. 3. AFM image of reduced graphene oxide obtained from scalar graphite.

Based on 2D AFM images, we determined the length and width of rGO grains, as shown in Fig. 5.

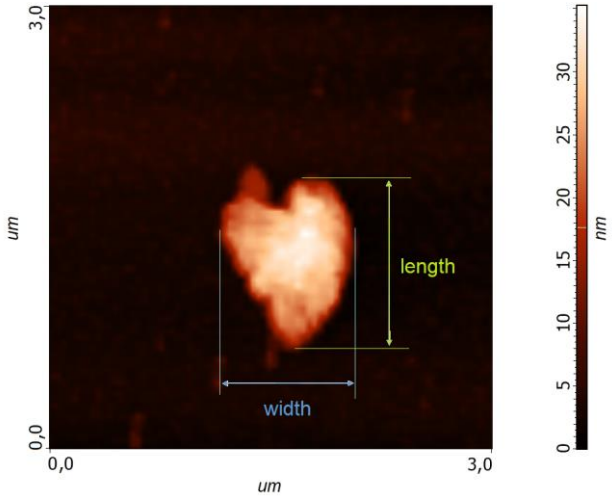

Fig. 4. Method of measuring the length and width of reduced graphene oxide agglomerates.

To determine the influence of the graphite precursor on the size of rGO, we compared the obtained data, as shown in Fig. 5.

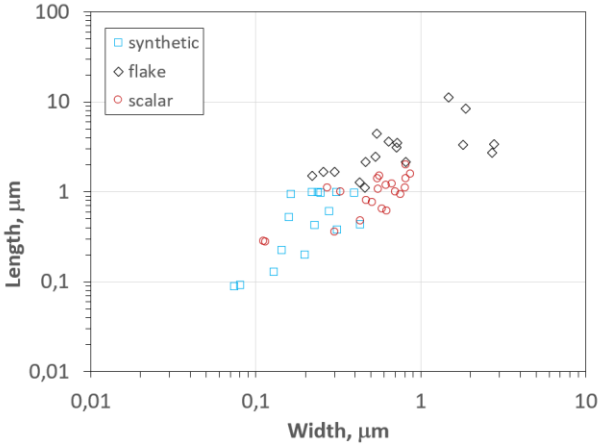

Fig. 5. Comparison of size of reduced graphene oxides obtained from various graphite precursors.

All measured agglomerates range in size from a part of a micrometer to (at most) a few nanometers. Among the tested oxides, the largest sizes were measured for rGO obtained from flake graphite (0.8 × 2÷3 µm). This oxide is also characterized by the greatest ratio of length to width (the most significant variation in the mentioned parameters). The most comparable length and width sizes are observed for rGO obtained from scalar graphite. Their size is 0.7 × 1 µm. The smallest size of rGO is measured for rGO obtained from synthetic graphite and equals 0.2 × 0.8 µm. The performed analysis showed that there is a correlation between the size of the graphite precursor and the size of rGO. Generally, the larger the diameter of the precursor, the greater the rGO.

This paper investigates the effect of various graphite precursors (flake, scalar, synthetic) on the size of reduced and exfoliated graphene oxide. We have shown that the size of the precursor determines the size of the obtained rGO agglomerates. Generally, the larger the graphite size, the larger the rGO size. The size of rGO agglomerates affects their properties and thus their possible application. Therefore, this should be considered when fabricating rGO for specific applications.

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