Thermal oxidation of few-layer graphite plates: an SPM study

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Commercial exfoliated graphite (1 mg material in 20 ml benzene) was treated ultrasonically for 3 hours. Droplets from the suspension were dispersed on Si wafer. Graphite plates of 1-5 μm in diameter were observed by tapping mode atomic force microscopy (AFM) using a Nanoscope IIIa instrument operating in air. After the AFM investigation, the sample was introduced into a quartz reactor of 18 mm inner diameter and oxidized in an electric furnace with an effective heating length of 200 mm [1]. The oxidization was carried out in three steps. In the first step the sample was oxidized at 450 ^{o}C for 10 minutes (step I). In the second and the third steps the sample was heated at 550 ^{o}C for 10 minutes (step II) and 20 minutes (step III), respectively. AFM measurements were performed after each oxidization step and the effect of oxidization was observed on the same graphite platelet.

To study in more detail the effect of oxidization, we selected an area near the right-hand side edge of the plate in Fig. 1. This portion of the graphite plate is shown in Fig. 2 after the oxidization steps I (Fig. 2a), II (Fig. 2b), and III (Fig. 2c), respectively. The label marks *11'* in Fig. 2a) show a height of 0.7 nm, which corresponds to the thickness of two graphene layers. However, we can easily notice that the plate thickness is not uniform. The marks labeled *22'* show thicker parts with 0.37 nm additional height, which is due to the presence of another incomplete graphene layer. One can observe several pits on the surface in Fig. 2a), at the regions where this top graphene layer is missing. These monolayer deep pits appear due to the oxidization step I. Similar oxidization pits were observed by scanning tunneling microscopy on oxidized highly oriented pyrolitic graphite (HOPG) surfaces [2, 3].

Fig. 2b) shows the graphite plate after oxidization step II (550 ^{o}C for 10 minutes). One can observe that in this case the plate edges have also been oxidized and the shape of the analyzed area has changed. The label marks *11'* show a height of 0.39 nm, which means that the thickness of this part has been decreased to one graphene layer. Similarly to Fig. 2a), the plate thickness is not uniform, the label marks *22'* show two-layer thick islands. The effect of the third oxidization step (550 ^{o}C for 20 minutes) is illustrated in Fig. 2c. Notice that the thickness of the plate became uniform, the islands of the second layer were removed completely and eventually we obtained a single graphene layer (label marks *11'*). The oxidization decreased also the horizontal diameters of the plate. As a consequence, the analyzed part detached from the body plate and it can be considered a standalone graphene platelet (Fig. 2c). One can observe that the substrate surface (the native SiO_2 layer) has also been etched, and approx. 0.4 nm deep trenches have formed along the circumference of the graphene platelet (label marks *22'*).

In this work we have also observed the self-assembly of the few-layer graphite plates into a free-standing film. The unused suspension (few-layer graphite plates dispersed in benzene) was deposited and left untouched. After 2 month, it was observed that the solvent evaporated completely from the bottle and the graphite plates assembled into a continuous and cohesive film. Portions of this free-standing graphite film were investigated by SPM techniques (AFM, STM).

References:

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Figures:

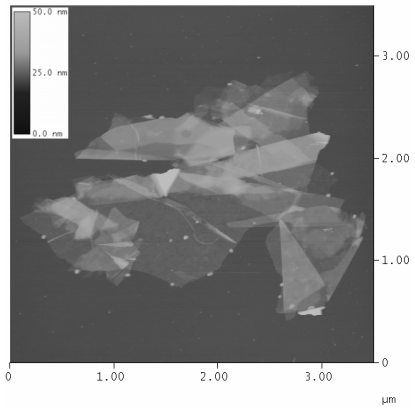

Fig. 1. AFM image of a few-layer graphite plate.

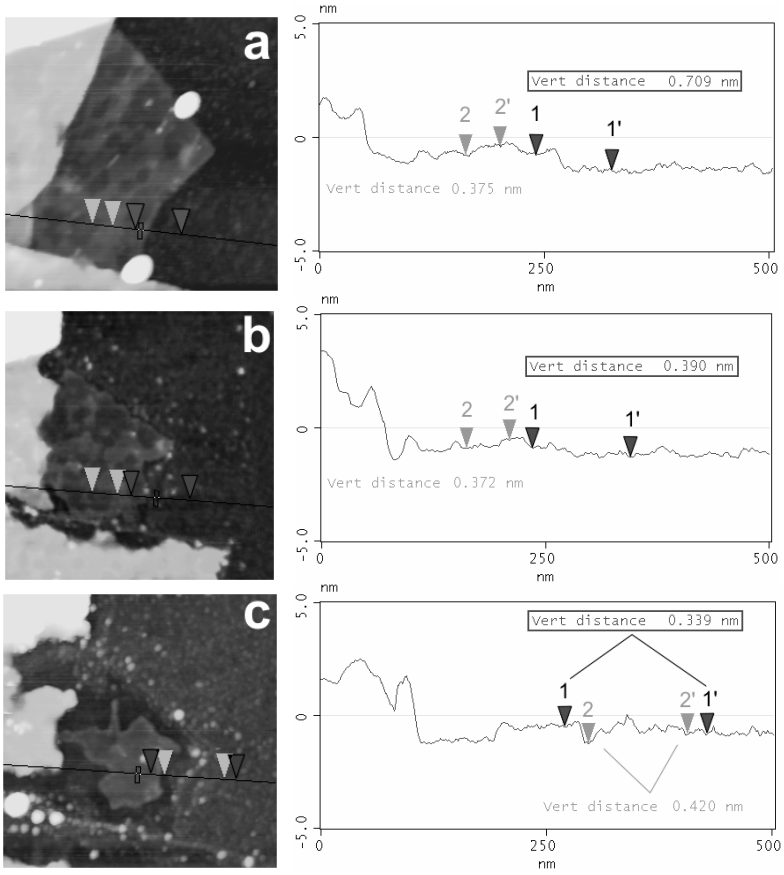

Fig. 2. AFM images of the same area of the few-layer graphite plate: (a) – after oxidization step I (450 ^{o}C for 10 minutes); (b) – after oxidization step II (550 ^{o}C for 10 minutes); (c) – after oxidization step III (550 ^{o}C for 20 minutes).