Influence of polycrystalline Ni films structure on thickness uniformity of the graphene films grown by CVD

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Many potential applications of graphene have been proposed [1–3]. Therefore, it is highly desirable to develop reliable synthesis techniques to fabricate the graphene films. Recently we have developed a method to grow graphene films by low-pressure no flow chemical vapor deposition (CVD) using Ni thin films deposited on SiO2/Si substrates. As it was shown earlier, the quality and uniformity of graphene films strongly depend on the structure of Ni films [4, 5]. Numerous defects in Ni films (grain boundaries, triple junctions and dislocations) lead to the formation of multilayer graphene. Strong (111) texture and large grain size in Ni films promote growth of mono- and bi-layer graphene [5].

The Ni films were deposited by the self-ion assisted deposition technique [6] on oxidized Si substrates at 0 and -5 kV biases. Substrates were placed in a quartz tube reactor, which was pumped down to a pressure about 10^{-6} torr and then inserted into a furnace preheated up to 950°C. When the samples were heated to the reaction temperature, acetylene was admitted into the quartz tube up to a pressure of 0.4 torr for 5 s and then pumped out and the quartz tube reactor was extracted from the furnace. All graphene films were grown at the same conditions.

We examined the structure of the Ni films after the graphene growth, using electron backscattering diffraction (EBSD) in the scanning electron microscope (SEM). We observed that the Ni films deposited at -5 kV bias were strongly (111) textured. The average grain size was about 1 μm. Ni films deposited at 0 kV bias had (111) oriented grains and noticeable fraction of (100) oriented grains. Bimodal grain size distribution was observed in the films. In the matrix of rather small grains (about 1.5 μm) large enough grains (4-6 μm) were found. That is beginning stage of abnormal grain growth occurred.

Transfer of the resulting graphene was done with aid of poly(methyl methacrylate) (PMMA) that was spincoated on the surface of the graphene film to serve as a support. PMMA/graphene layer was detached from the substrate by wet-etching of the Ni film with a 1 wt% aqueous solution of hydrochloric acid and then manually laid on the target substrate (SiO2/Si). The PMMA was finally removed by exposure to acetone in vapor and then liquid form.

Raman spectra and mapping images of graphene films were measured with Renishaw Raman microscope using 633 nm excitation wavelength. We collected Raman spectra over 2000 μm^2 area. The I2D/IG values were then extracted from the spectra. Figure 1 show the I2D/IG contour maps of graphene grown on 0 and -5 kV biased Ni films (a and b, respectively), on an oxidized silicon substrate. Nearly 75% of Raman spectra collected from the graphene grown on 0 kV bias Ni films shows the hallmark of monolayer/bilayer graphene and about 25% - few-layer graphene. Graphene films grown on –5 kV bias Ni films demonstrated only about 33% of monolayer/bilayer and 67% of few-layer graphene.

Thus, the graphene grown on Ni films with weaker (111) texture had higher thickness uniformity and lower number of graphene layers.

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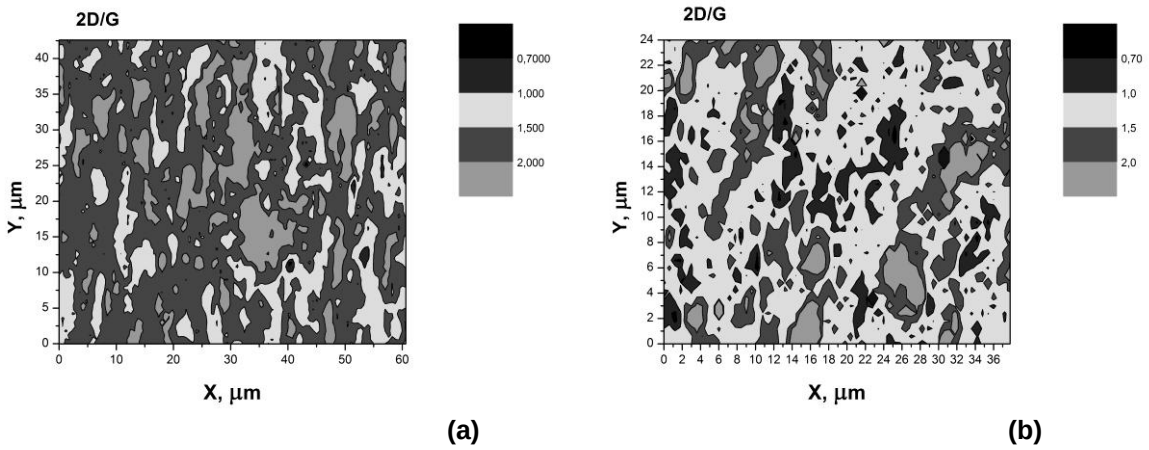

Figure 1: I_{2D}/I_G contour maps of graphene grown on 0 (a) and -5 kV (b) biased Ni films.