Influence of different copper treatment on the formation of single-layer graphene by CVD method

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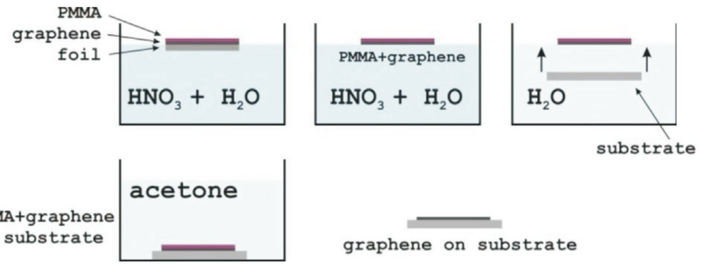
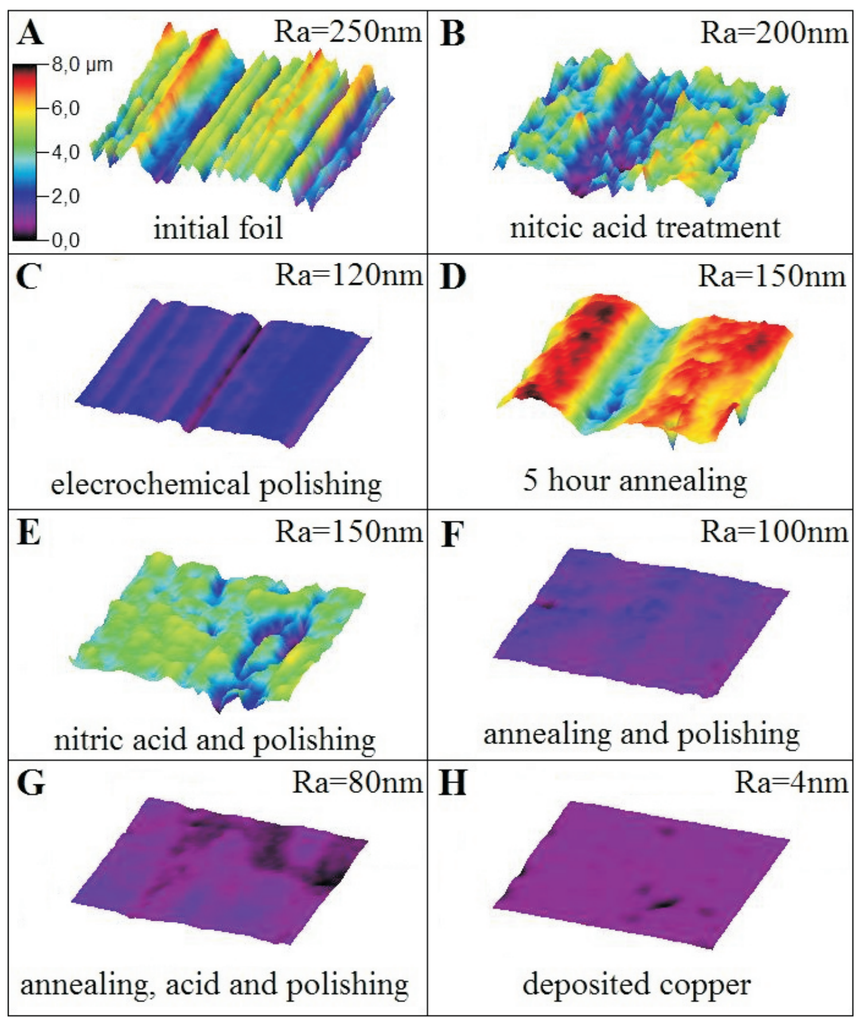
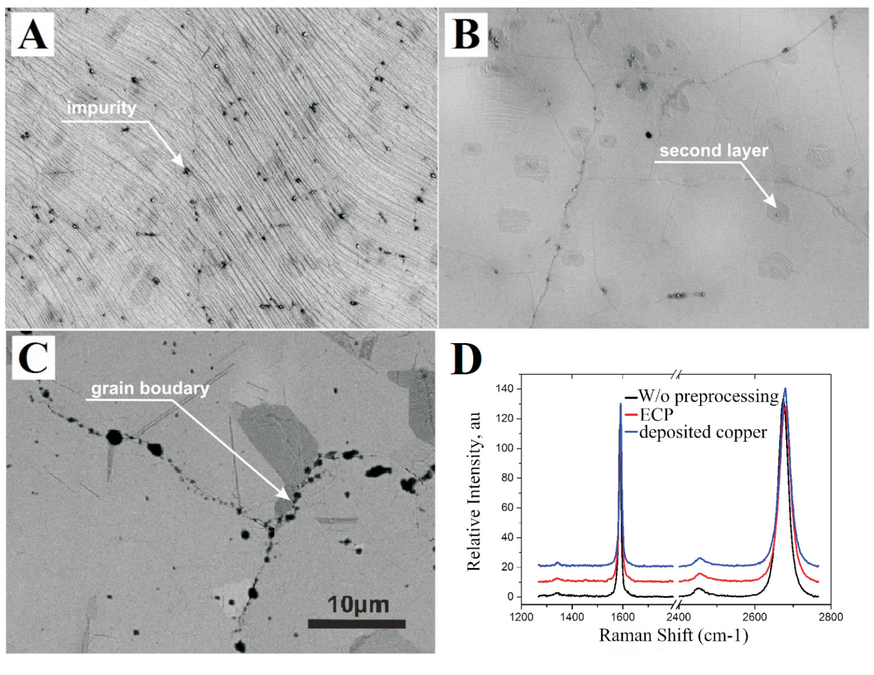
Chemical vapor deposition synthesis of graphene on copper foil from methane is the most promising technology for industrial production. However, an important problem of the formation of the second and subsequent graphene layers during synthesis arises due to the strong roughness of the initial copper foil. Here we demonstrate the various approaches to prepare a smooth copper surface before graphene synthesis to reduce the formation of multi-layer graphene islands. Six methods of surface processing of copper foils are studied, and the decrease of the roughness from 250 to as low as 80 nm is achieved. The correlation between roughness and the formation of multi-layer graphene is demonstrated. Under optimized conditions of surface treatment, the content of the multi-layer graphene islands drops from 9 to 2.1%. The quality and the number of layers of synthesized graphene are analyzed by Raman spectroscopy, scanning electron microscopy, and measurements of charge mobility.



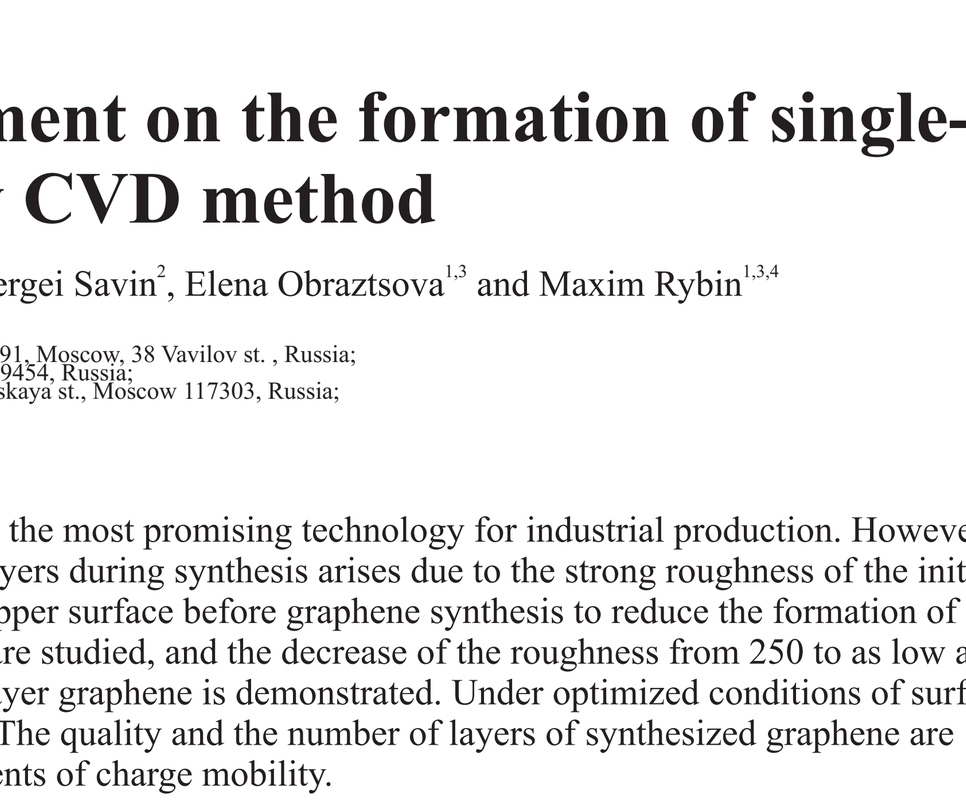
Scheme of the transferring process graphene film to a substrate



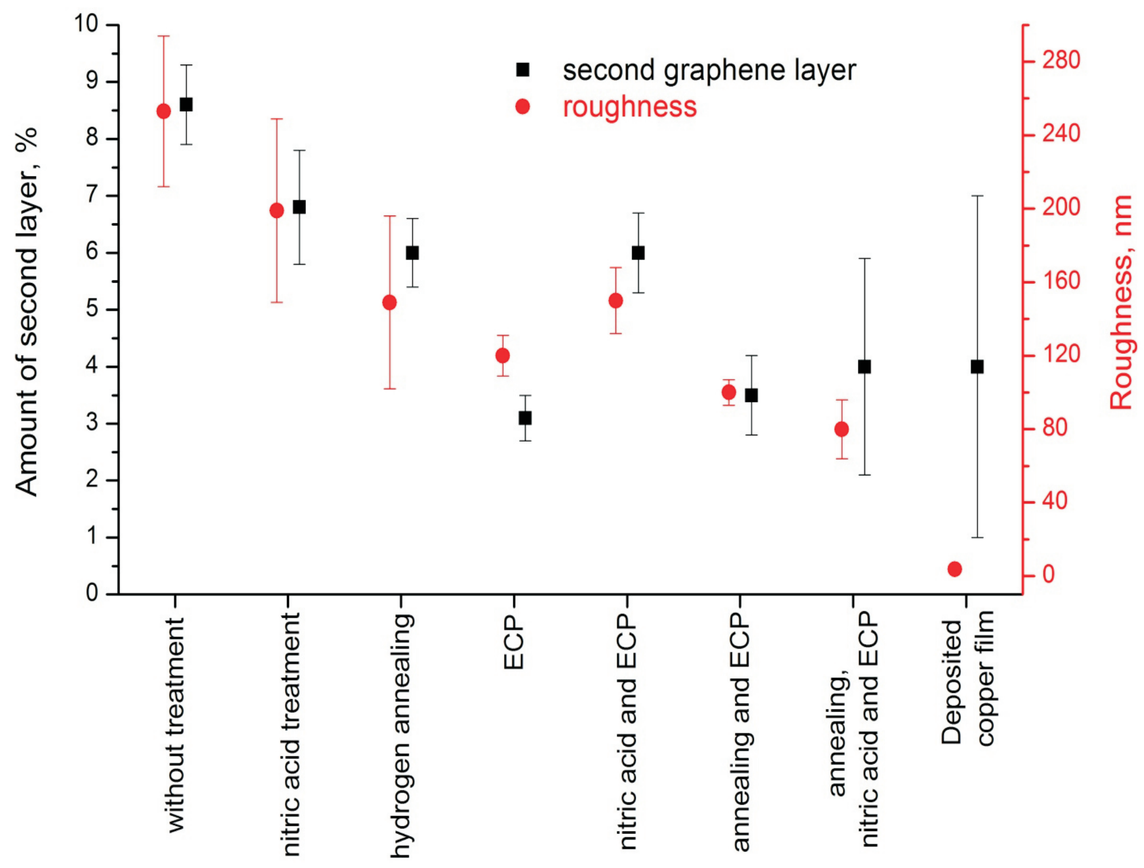
The roughness measurements of copper foil by interference microscope: a - without pre-treatment, b - nitric acid treatment, c - electrochemical polishing, d – hydrogen annealing for 5 hours, e – nitric acid treatment and electrochemical polishing, f – hydrogen annealing and electrochemical polishing, g - hydrogen annealing, nitric acid treatment and electrochemical polishing, h - deposited copper film. All measurements were done from 50x50 micrometers area. All figures have the same scale bar in color grade and it is indicated in figure A.



SEM images and Raman spectra of graphene films synthesized on copper foils with different treatment (a - without pre-treatment, b - annealing and polishing) and c - deposited copper. The scale bar is the same for all SEM images and presented in figure C.



Principle scheme of a change in the copper surface roughness during various treatments (a-e) and formation of graphene layers (f).



The effect of method of copper treatment on average roughness of copper surface (red circles) and the amount of few layer graphene (black squares).

The chemical vapor deposition technique is an efficient approach for the synthesis of high-quality graphene on copper foil over large areas. The roughness of the copper surface is the key parameter for the fabrication of the most uniform single-layer graphene film.

The use of the original commercial copper foil with Ra = 250 nm leads to the formation of a two-layer graphene film on an area of 9% of the total surface. Additional processing of catalytic copper foil causes a decrease in the amount of few layer graphene. The lowest roughness of 80 nm is achieved using a combination of annealing, etching and electrochemical polishing, which results in the synthesis of the film with only 2.1% of the few layer graphene.

It is found that the optimal treatment is simply electrochemical polishing, which gives the same amount of a few layer graphene film, but on a copper surface with Ra = 120 nm. A further decrease in roughness even to 4 nm does not lead to a decrease in the amount of the few layer graphene.