Thermal Stability and sp^2 Clustering in Tetrahedral Amorphous Carbon: a Raman and Atomic Force Microscopy Study.

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We report a comparative study of the thermal stability and sp^2 clustering of tetrahedral amorphous carbon (ta-C) films using Raman spectroscopy and atomic force microscopy (AFM).

Ta-C films with different sp^3 fractions (70, 75, 80, and 88%) deposited on silicon by a filtered cathodic vacuum arc (FCVA) technique have been annealed in argon in steps of 100^o from 100 to 1100 ^oC. The subsequent structural and electronic changes in the films have been monitored using visible Raman spectroscopy and AFM in contact and current imaging mode with an ultra-sharp conductive cantilever.

Raman spectra excited by 514.5 nm argon laser light have been used for analysis of the bonding in ta-C films [1,2]. The asymmetry and intensity of the peaks near 1560 cm^{-1} (so-called G-peak), 1350 cm^{-1} (D or disorder peak) and 960 cm^{-1} (second order silicon peak from the underlying Si substrate) have been correlated with the presence and growth of nm-size graphitic domains in the films upon annealing. The film with the highest sp^3 content (88%) appeared to be very resistant to sp^2 transformation compared to other films.

AFM current imaging [3] allowed us for the first time to observe directly formation of the graphitic nanoclusters at the surface of the 88% sp^3 film and to estimate their size (Figure 1). After annealing at T<500 ^oC the film is not conductive. After annealing at 550 ^oC conductive spots 1-2 nm in size appear at the surface. Annealing at higher temperature results in the growth of the conducting islands, the islands coalescing into a near continuous conducting layer for annealing temperatures of about 900 ^oC.

Large scale topographic and current AFM scans revealed a correlation between surface roughness and conductivity: the higher the roughness, the higher the current. The AFM results also correlate with the Raman measurements which reveal that sp^2 clustering in the ta-C films begins in the temperature range of 400-550 ^oC. At higher temperature the clustering continues but it does not lead to macroscopic graphitization of the films.

The results provide important insights into the transformation and structure of ta-C with implications for understanding of some practical issues such as stress relief in ta-C films and their application as protective coatings for electronic devices and magnetic disks.

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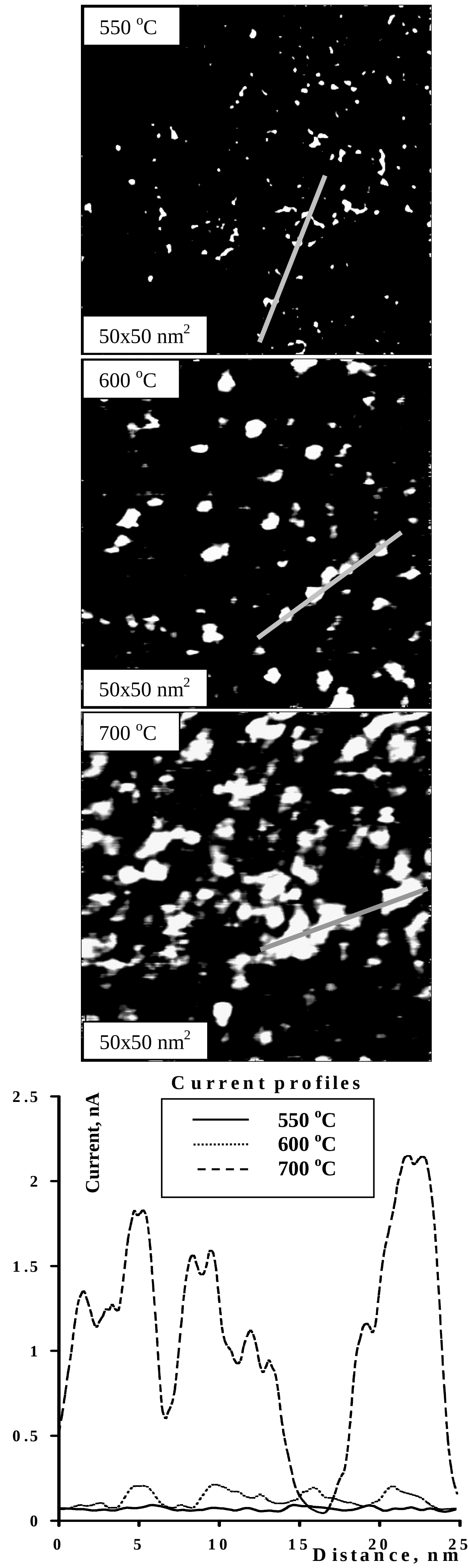

Figure1. AFM current images and profiles (along grey lines) showing formation and growth of conducting graphitic nanoclusters in ta-C film.