Catalyst size effects on multilayer graphene growth on molybdenum

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Chemical vapor deposition (CVD) on metal catalysts is a widely adopted route for scalable graphene synthesis compatible with microelectronic integration, however, controlling the thickness of multilayer graphene remains a key challenge [1]. While copper has been extensively employed, its limited thermal stability and diffusion issues motivate the exploration of alternative catalysts such as molybdenum [2]. In this work, we investigate the role of catalyst size and lateral confinement in governing multilayer graphene growth during CVD on molybdenum supported on tetraethyl orthosilicate (TEOS), a technologically relevant substrate for microelectronic applications [3]. Graphene is grown on a 35 nm thick continuous sputtered molybdenum film and on micro- and nanostructured molybdenum patterns defined by electron beam lithography, both supported on a 1.6 µm thick TEOS layer. Catalyst geometries ranging from planar films to laterally confined structures with dimensions from 100–500 nm up to 6 µm enable a direct comparison between flat and confined growth. Structural and chemical properties of the two systems are investigated by Rutherford backscattering spectrometry, secondary ion mass spectrometry, and scanning and transmission electron microscopy. On flat molybdenum films, CVD at 1000 °C and 0.1 mbar promotes unidirectional diffusion between molybdenum, silicon, and carbon, leading to the formation of interfacial Mo–Si and Mo–C phases and the growth of multilayer graphene with approximately fifteen layers. In contrast, nanostructured molybdenum undergoes solid-state dewetting and multidirectional diffusion resulting in morphological instability and a reduction of graphene thickness, with only about five layers, as directly observed by cross-section STEM (figure 1). These results show that catalyst size plays a key role in multilayer graphene growth, enabling thickness control on molybdenum catalysts.

References

[1] Zhang et al., Sci. Rep. 2014, 4, 4431.
[2] Y. Grachova et al., Procedia Eng. 2014, 87, 1501-1504.
[3] A. M. Mahajan et al., Surface and Coatings Technology 2004,183(2-3), 295-300.

Figures

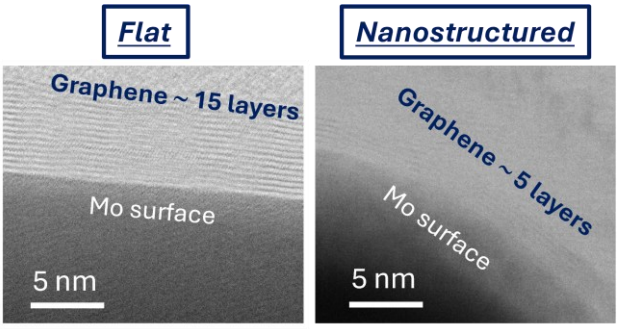

Figure 1: Cross-sectional STEM images of graphene grown on flat and nanostructured Mo catalysts supported on TEOS/Si, showing a different number of graphene layers as a function of the catalyst lateral size.