

Growing perfect monolayer of graphene from nickel surfaces

C. Bichara,¹ H. Amara,² A. Zappelli¹ and F. Ducastelle²

¹ Centre Interdisciplinaire de Nanoscience de Marseille, CNRS, 13288 Marseille, France

² Laboratoire d'Etude des Microstructures, ONERA-CNRS, BP 72, 92322 Châtillon Cedex, France
hakim.amara@onera.fr

Abstract

Growing graphene on a metal surface is one possible way to obtain a high quality graphene, with a controllable number of layers. The synthesis usually relies on a chemical vapor deposition of a carbon bearing gas on the surface of a metal such as Ir, Cu, or Ni [1]. In the present work, we investigate the latter case of graphene on Ni that is of particular interest because the role of carbon solubility in subsurface layers is both difficult to investigate experimentally and important to understand for the production of high quality graphene [2].

To understand the interaction of carbon with nickel at the atomic level, we have developed a tight binding model [3] implemented in a Grand Canonical Monte Carlo (GCMC) code. It has been used to investigate the nucleation and growth of carbon nanotubes in CVD processes [4, 5]. In this latter case, we study the chemical and physical states of the metal catalyst as a function of size, temperature and carbon chemical potential conditions corresponding to growth of SWNTs. We also study the interfacial properties of the NPs with respect to sp^2 carbon walls, show that they strongly depend on the amount of carbon dissolved, and emphasize their role in the growth of tubes [6].

With the same approach, we investigate the CVD synthesis of graphene on Ni (111). We identify thermodynamic conditions (temperature and carbon chemical potential) to obtain a graphene monolayer. Owing to the significant technical improvements of our grand canonical Monte Carlo code (CPU time speedup by two or three orders of magnitude, as explained in [7]), we can extend the range of our previous calculations [8] to present carbon adsorption isotherms to slab of nickel containing 1000 atoms, and temperatures ranging from 800 to 1400 K. A possible scenario is the following : during the course of the GCMC simulations, the adsorption of C atoms begins at the surface while some atoms are incorporated in the interstitial sites between the Ni layers. When the solubility limits is reached, a surface C structure begins to develop in the form of chains creeping on the surface and eventually crossing each other. At their intersections, threefold coordinated C atoms act as nucleation centers for C sp^2 structures that develop on the surface. These sp^2 C atoms interact weakly with the underlying Ni atoms and can detach from the surface to form graphene (see Figure 1).

Moreover, depending on the growth conditions, we show that variable amounts of carbon atoms can be found in the subsurface layers, while the first subsurface layer shows a tendency for carbon depletion when graphene covers the Ni surface. This result is surprising since the subsurface interstitial sites have been identified as most favorable for carbon incorporation on Ni(111) surfaces. To confirm this effect, we have performed static TB and DFT calculations to predict the stability of C atom located at different depths from the Ni slab surface covered or not with a graphene overlayer.

To conclude, such depletion is probably at the root of interesting catalytic property of Ni. Indeed, this lower stability of carbon close to the surface can be used to control the number of layers formed. This lack of C atoms in subsurface sites can play the role of a barrier limiting the presence of C atoms on the surface and suggesting that the control of one layer is possible [9].

References

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Figures

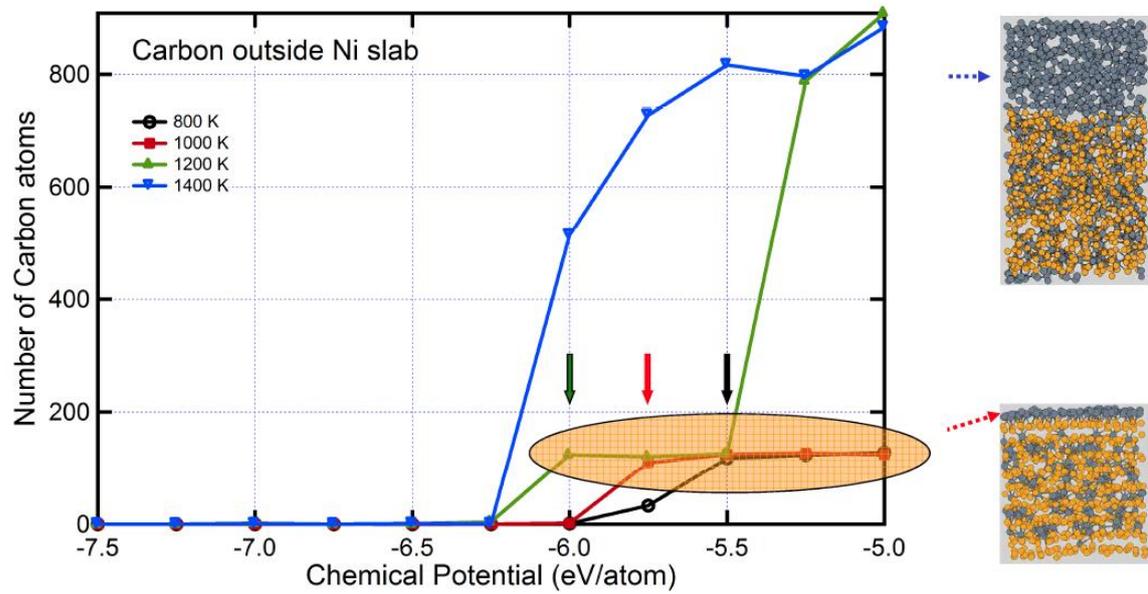


Fig. 1. Adsorption isotherms of Carbon atoms on a Ni(111) slab at different temperatures