Highly efficient growth conditions for monolayer graphene films by ethanol chemical vapor deposition: observation of the initial growth stages

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The growth of crystalline graphene on copper by the chemical vapour deposition (CVD) of ethanol (C_2H_5OH) presents advantages over methane-based CVD in terms of safety, cost, and ease of processing [1,2]. It is believed that the oxygen atoms in the ethanol make the catalytic growth process more efficient than with methane, and the faster growth kinetic allows the growth of continuous graphene films in seconds rather than in minutes. However, this kind of growth can result to a less ordered graphene lattice, as evidenced by larger D bands observed by Raman spectroscopy [3]. Large hexagonal single-crystal graphene was successfully grown using methane, and more recently ethanol, inside copper "enclosures" [4, 5], where the copper surface was not directly exposed to the growth atmosphere.
In this work, when the CVD growth of graphene was done at 1000°C with an ethanol partial pressure as low as 0.05 Pa and keeping the total process pressure lower than 50 Pa, it was possible for the first time to observe the initial growth stages of graphene, where individual islands still have to merge into the continuous film (Figure 1). The effect of the CVD process temperature was also investigated and the graphene quality was assessed by Raman spectroscopy (Figure 2). At 1070°C the growing graphene film was both less defective and more developed (almost continuous) than at 1000°C. This study implies that the quick graphene growth on a copper surface is due to the fast growth of the individual islands and not only to an increase in nucleation density, making it possible to grow high quality graphene films at high temperature in few seconds.

References

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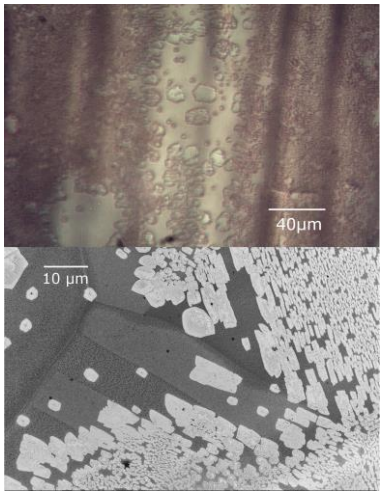
Figures

(1)



(2)

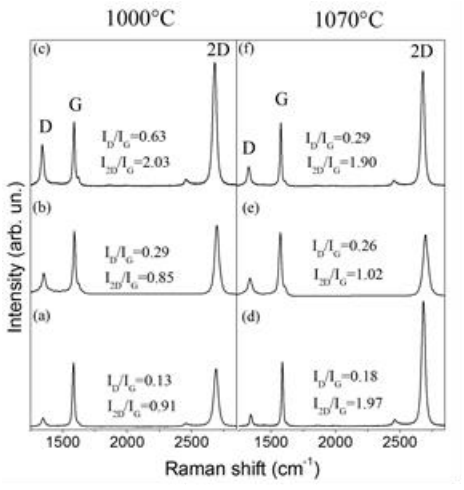


Figure 1. Optical and SEM micrographs of the growing graphene islands on the copper surface (CVD at 1000°C for 30 s). Figure 2. Raman spectra of graphene films transferred on Si/SiO_2 (300nm); the films are grown at 1000°C (a-c) and 1070°C (d-f), for 30 min (a,d) and 30 s (b, c, e, f); the total pressure is 40Pa with 2Pa Ethanol (a,b,d,e) and 10Pa with 0.05 Pa Ethanol (c,f).