Large-area graphene from catalytic metals to arbitrary substrates by electrochemical delamination and transfer printing

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Graphene growth on catalytic metals (Cu, Ni, Pt, Ru,…) by CVD followed by transfer currently represents one of the most viable roots to large area graphene-based electronics. Beyond catalyst and growth condition optimisation, transfer represents a critical step of this approach, with a strong impact on the final quality and uniformity of graphene on the target substrate. The typically adopted method to separate graphene from the growth substrate, i.e. chemical etching of the metal (typically in $FeCl_3$ for Ni or Cu) while using a sacrificial polymer film (typically PMMA) as the graphene support, presents several drawbacks (metal contaminations remaining from the metal substrate and/or from the etchant, polymer residues on graphene) and limitations (difficult or not possible etching in the case of noble metal substrates). Furthermore, metal waste by etching raises the process cost in view of an industrial scale-up, so that approaches for reusing the metal substrate in several growth cycles are highly desirable.
Recently, graphene has been effectively separated from the growth substrate without metal etching, using the mechanical action of hydrogen bubbles generated at graphene/metal interface [1] in an electrochemical process using electrolytes like NaOH and KOH [2]. This approach potentially solves the metal contaminations issues, allows reusing the substrate for unlimited number of growth, without limitations to the kind of catalytic metal.
In this work, the influence of the critical parameters involved in the electrochemical delamination (such as the electrolyte molar concentration, the cell overvoltage,..) on the structural and electrical quality of transferred graphene was investigated. An optimised transfer-printing method, allowing a fine control of pressure and temperature ramps applied between the polymer/graphene stack and the target substrate, was developed for graphene transfer on different substrates, such as SiO_2, Si, GaN, AlGaN, SiC or soft materials (PEN). Furthermore, specific functionalization/derivatization treatments [3] have been developed to optimise the graphene adhesion on each kind of substrate. Different combinations of support polymers and solvents (for final graphene cleaning) have been considered. The nanoscale morphological and electrical properties of the transferred graphene will be investigated by advanced scanning probe techniques [4], the structural and chemical properties by atomic resolution STEM and EELS [5]. Finally, the average electrical properties on large area will be tested by electrical measurements on proper device structures (FETs, TLM and VdP structures).

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