Graphene synthesis using alcohol precursors

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Although the production of graphene through the mechanical exfoliation of graphite allowed the availability of graphene for fundamental studies, its applications where limited by the small flake sizes and low yield. Alternative routes to the production of graphene were imminent.

The most popular alternative route saw the light in 2009 when the chemical vapor deposition (CVD) synthesis of graphene was reported using polycrystalline nickel as catalyst and methane as carbon source at low pressures [1-2]. Later that year the growth using commercial Cu foils was reported [3]. The synthesis technique relies on the decomposition of methane at ~1000°C at low pressure and its deposition on the metal catalysts. While on Ni the solubility of C is high, the growth mechanism is through the segregation and precipitation of carbon once the Ni is cooled down; in contrast, Cu has a low C solubility and the growth is mediated by surface adsorption [4].

Besides methane (CH_4) other carbon precursors have been used in the synthesis of graphene. Reports of solid and liquid sources appear in the literature [5-6]. The above listed precursors are used in experiments at low pressure conditions (mTorr) and generally pure Ar and H_2 or Ar-H_2 mixtures are involved in the synthesis, which require a sophisticated set-up with pumping system, 2-3 gas lines available, flow meters, and proper piping for contents higher that 5% of H_2 because of safety issues.

Here we describe the synthesis of graphene via the thermal decomposition of alcohols at atmospheric pressure, using copper foils as catalyst and a single gas inlet consisting of a gas mixture of Ar-H_2 with a safe concentration of H_2 (5%).

We have investigated the growth of graphene using two liquid precursors: 2-phenylethanol and ethanol. Commercial copper foils of 25 μm-thick have been purchased; after mild cleaning such foils are placed in the growth chamber set to temperatures of 950°C-980°C, the decomposition of the alcohols at this temperature leads to the growth of graphene on copper. We have studied the as-produced samples by means of scanning electron and optical microscopy. To perform further studies we transferred the samples to Si/SiO_2 substrates using $FeCl_3$ as copper etchant and PMMA as support/protection layer. The transferred films have been characterized using optical microscopy, Raman spectroscopy, and atomic force microscopy.

In Figure 1A, we show a scanning electron microscopy image of a film of graphene on copper grown from ethanol. The typical wrinkles originating from the mismatch in thermal expansion coefficients of both materials are evident. Etching of copper and transference of the graphene film have been carried out. An optical image of a transferred film depicted in Figure 1B shows uniformity at a large scale.

In order to verify the sp2 nature and quality of the produced material we perform Raman spectroscopy measurements on the transferred films (Figure 1C). The high intensity of the 2D (G') band reveals the presence of monolayer graphene.

In this work, we will expose in detail the synthesis and transfer procedures, our results obtained by several characterization techniques, and we will compare the quality of the synthesized materials depending on the alcohol used as precursor.

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Figures

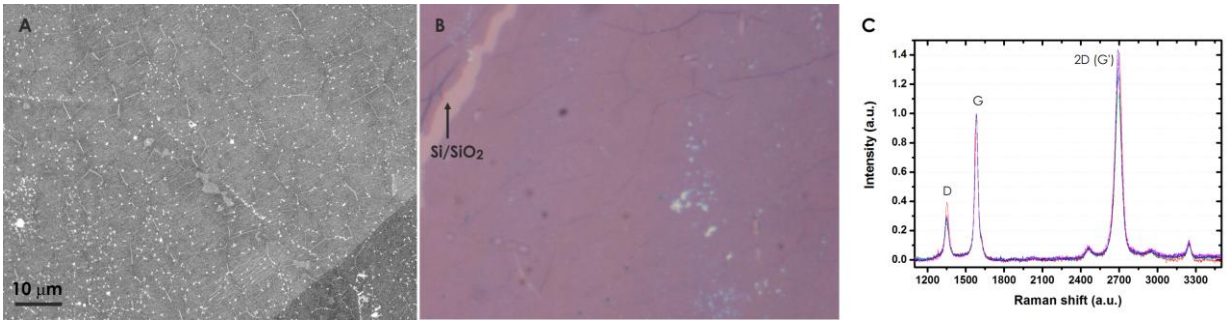

Figure 1 Characterization of the produced graphene films. A) Scanning electron micrograph of the graphene film on copper right after synthesis. B) Optical microscopy image at 100x of the transferred graphene film (in light purple color) on a Si/SiO_2 substrate, a crack at the left up corner leaves visible the substrate. Bluish traces correspond to PMMA residues. C) Raman spectroscopy spectra recorded at different spots of the transferred film.