Catalyst Engineering for Scalable 2D Film Control: The Dark Secrets of Bulk Oxygen and Integrated Pathways for Single-Crystal Growth

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The controllable, reproducible and scalable growth of graphene and related 2D materials remains the foremost challenge for both research and any technologies exploiting their unique properties. While chemical vapour deposition (CVD) has become the most widespread method for 2D material film growth, even basic process parameters remain not well understood due to the manifold, complex parameter space. Here we focus on widely used Cu as process catalyst and reveal the role and control of residual bulk oxygen, as well as showing a novel integrated process pathway for achieving uniform single crystal Cu epitaxial layers of a chosen orientation starting with (111), (110) and (100).

While oxidation is widely used to remove impurities in metal catalysts and to control the nucleation density of graphene[1], we show that minute concentrations of residual bulk oxygen can significantly deteriorate the quality of as-grown graphene highlighted by an increased Raman D/G ratio, increased propensity to post-growth etching and increased fraction of multi-layer graphene nucleation[2]. Starting from commercial Cu foils, we show that a simple hydrogen annealing step after the initial oxidation allows us to lower the residual oxygen level as measured by time-of-flight secondary ion mass spectrometry to produce graphene of significantly higher quality. This can be effectively combined with a short hydrocarbon exposure time of 10 min to achieve near full mono-layer graphene coverage, suitable for emerging industrial applications. We show that residual oxygen can have an equally significant impact on Fe catalysed h-BN CVD[2,3], and discuss the underlying mechanisms with parallels to well-known processes in metallurgy, catalysis and vacuum science. We further demonstrate the scalable deposition of ultra-low roughness single crystal Cu(111) and 2D material growth within a single synthesis step. The total synthesis from catalyst to 2D film takes less than 2 hours, during which there is no exposure to atmospheric conditions, circumventing the most common source of contaminants.

References

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