The Challenge of Contamination in Atomically Precise Manipulation and Processing of Graphene and 2D Materials

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Atomically precise manipulation of graphene and other two-dimensional materials¹ holds promise for fields as diverse as quantum electronics² and nanopore sensing³. However, manipulation of these unique materials often assumes a 'pristine' system largely free of contaminants and other detrimental constituents. Recently, several studies have begun to characterize and describe both the pervasiveness and implications of these contaminants on graphene and other emerging 2D materials, which are typically adsorbed hydrocarbons⁴. These studies indicate that contamination often occludes the majority of exposed surfaces⁵ of the materials of interest, even after cleaning treatments (figure 1) and that contaminants have high surface mobility and low volatility – a combination that causes it to be challenging to remove or control in many beam-based modifications, such as scanning probe, ion beam, and TEM.

Although present on virtually all graphene devices with an exposed surface, only recently have the implications of the contamination begun to be explicitly acknowledged in the literature. With a growing body of research characterizing the contamination with Raman, XPS, AFM, SIMS, and especially STEM, an understanding of the challenge is emerging. Here we provide an overview of sources of contamination, characterization of the properties and composition, an assessment of the implications for utilizing these materials, along with strategies for mitigation.

^{1.} Susi, T. *et al.* Towards atomically precise manipulation of 2D nanostructures in the electron microscope. *2D Mater.* **4**, 42004 (2017).

^{2.} Mol, J. A. *et al.* Graphene-porphyrin single-molecule transistors. *Nanoscale* **7**, 13181–13185 (2015).

^{3.} Branton, D. *et al.* The potential and challenges of nanopore sequencing. *Nat. Biotechnol.* **26**, 1146–1153 (2008).

^{4.} Li, Z. *et al.* Effect of airborne contaminants on the wettability of supported graphene and graphite. *Nat. Mater.* **12**, (2013).

^{5.} Tripathi, M. *et al.* Cleaning graphene: Comparing heat treatments in air and in vacuum. *Phys. status solidi - Rapid Res. Lett.* **11**, 1700124 (2017).



Figure 1: Example micrographs of chemical vapor deposition synthesized single layer graphene imaged in MAADF-STEM at 60kV a) as transferred graphene without special cleaning treatment demonstrating hydrocarbon and other contaminants present on the graphene surface. b) current annealed graphene showing substantially lower prevalence of adsorbed hydrocarbons, however still with a high-degree of surface covered, despite cleaning.