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Comment

Graphene Oxide: The Schrödinger's Cat of Nanomaterials

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The controversy surrounding the existence of "graphene oxide" as a well-defined material is explored. While the term "graphene oxide" suggests a specific and uniform structure, the reality points to a mixture of oxidized carbon materials with variability in oxygen functionalities and crystallinity. This discussion challenges the conventional understanding, proposing that graphene oxide may be more accurately described as a spectrum of oxidized carbon structures rather than a singular material. This Comment encourages a re-evaluation of how we define and study these complex nanomaterials.

■ GRAPHENE OXIDE: MYTH OR MISCONCEPTION?

Concerns have been raised about the potential nanotoxicity of "graphene oxide", particularly regarding its effect on cellular systems, which may include cytotoxicity, oxidative stress, and inflammatory responses.[1] These health risks are further complicated by the significant variability in the structure, composition, and particle size of "graphene oxide" across different samples. Therefore, it becomes crucial to define and standardize what is meant by "graphene oxide" as the term is often used loosely, referring to materials with diverse chemical compositions, degrees of oxidation, and physical characteristics. Establishing a clear definition is essential for reproducibility in research and for accurately assessing the biological impacts.

If the reader performs a quick online search, they may find that graphene oxide is also commonly understood as graphite oxide—previously known as "graphitic oxide"—and is frequently described in nonacademic contexts as "a compound composed of carbon, oxygen, and hydrogen in variable ratios, produced by treating graphite with strong oxidizing agents and acids to eliminate extraneous metals. The resulting maximally oxidized material is a yellow solid with a carbon-to-oxygen ratio ranging from 2.1 to 2.9, retaining the layered structure of graphite but exhibiting significantly larger and more irregular interlayer spacing." While this popular definition draws upon several scientific references, it primarily reflects a generalized understanding outside the rigorous framework of academic research, further emphasizing the material's ambiguity and the need for standardized definitions within the scientific community.

The commercialization of "graphene oxide" has grown with dozens of companies offering what are claimed to be "purified" forms of the material. Notably, some companies list the chemical formula for "graphene oxide" as CxOyHz, further emphasizing the variability in its composition. Adding to the confusion, there is a school of thought[2] suggesting that "...If the exfoliated sheets contain only one or few layers of carbon atoms like graphene, these sheets are named graphene oxide (GO)...."

The Web of Science is now cataloging thousands of research articles with "graphene oxide" in the title. The concept of "graphene oxide"—in some cases called the "oxidized form of graphene"[3]—has captivated the scientific community for over a decade, with the material being heralded for its potential in various fields, from electronics to biomedicine. However, a fundamental question remains: does "graphene oxide" truly exist as a distinct material, or is it merely an umbrella term used to describe variably oxidized carbon-based materials with inconsistent and ill-defined structures?

According to the International Union of Pure and Applied Chemistry (IUPAC), an oxide network is defined as a network comprising only metal−oxygen linkages.[3] In the case of "graphene oxide", the term implies a specific structural form of graphene sheets functionalized with oxygen-containing groups such as epoxides, hydroxyls, and carbonyls. However, unlike conventional oxides (e.g., SiO_2 or Al_2O_3), which exhibit well-characterized stoichiometry and crystal structures, "graphene oxide" lacks a consistent formula or structure across samples. If "graphene oxide" were to exist as a distinct material, it would need to be a monolayer graphene structure "uniformly" functionalized with oxygen atoms in a well-defined arrangement. Furthermore, the size of graphene flakes plays a crucial role, as larger sheets often exhibit uneven oxidation, resulting in regions that remain graphitic or partially oxidized. These issues collectively hinder the formation of a consistent, homogeneous "graphene oxide" material, instead producing a heterogeneous mixture of oxidized domains and structural defects that vary between samples.

■ GRAPHENE OXIDE OR OXIDIZED CARBON-BASED MATERIAL?

The synthesis of so-called "graphene oxide" typically involves the oxidation of graphite using methods such as Hummers' method or its variations.[4] These processes result in a material composed of sheets with varying degrees of oxidation, lattice distortion, and defect density. While some oxygen function-

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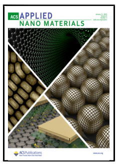

alities are indeed introduced, the resulting materials cannot be consistently described using a simple oxide framework. Instead, what is frequently referred to as "graphene oxide" may more accurately be described as a complex mixture of oxidized carbon structures, including amorphous regions, partially oxidized graphitic domains, and defects. The variability in oxidation and crystallinity suggests that these materials represent a spectrum of oxidized solid carbon rather than a single well-defined compound.

Furthermore, an oxidized monolayer of graphene does not qualify as "graphene oxide" in the traditional sense of an oxide because the oxidation does not produce a uniform, stoichiometric compound with a consistent structure. In classical chemistry, an oxide is defined as a compound formed by the bonding of oxygen atoms with another element in a specific ratio, leading to a well-defined and predictable crystalline structure. In the case of oxidized graphene monolayers, the oxidation introduces various functional groups (e.g., hydroxyls, epoxides, and carbonyls) scattered across the carbon lattice rather than creating a uniform oxide phase. This variability in the type, location, and density of oxygen functionalities, along with the lack of a consistent structural arrangement, means that oxidized graphene remains a modified carbon material rather than forming a distinct chemical entity that would be considered a true "oxide" under chemistry definitions.

Unlike crystalline oxides, which possess a uniform structure repeated throughout the material, the "graphene oxide" exhibits wide variations in its degree of crystallinity. Different preparation methods, precursor quality, and processing conditions result in significant differences in the size and distribution of oxidized regions as well as the concentration and types of oxygen functionalities present. This lack of structural consistency calls into question the validity of classifying these materials under a single term. The variability in crystallinity further complicates the characterization of "graphene oxide". In some cases, samples may exhibit graphitic domains interspersed with highly amorphous and disordered regions, making it challenging to identify any "oxide" structure using standard spectroscopic techniques. Therefore, instead of a distinct form of carbon-based solids modified with oxygen, it may be more appropriate to describe these materials as oxidized carbon solids with variable crystallinity. "Graphene oxide", as described in numerous studies,[1,2,4] often exhibits significant variability in its oxidation level, functional group distribution, and crystallinity. This heterogeneity, which results from diverse synthesis methods and precursor qualities, raises questions about the validity of classifying these materials under a single term. Most samples analyzed consisted of multilayer agglomerates with amorphous and graphitic domains, further complicating their characterization and comparison.

RECONSIDERING THE DEFINITION: OXIDIZED CARBON-BASED MATERIALS

Given the lack of a consistent structural and compositional definition, it may be time to reconsider the nomenclature surrounding "graphene oxide." The term "graphene oxide" has been widely used in the literature; however, its application often lacks adherence to precise definitions. This inconsistency complicates the reproducibility and hinders the development of standardized methodologies. The term implies a specific chemistry that does not accurately reflect the diversity of samples encountered in different studies. Describing these materials as "oxidized carbon-based materials (OCM)" or "variably oxidized graphitic solids (VO-GS)" would better capture the range of structural characteristics observed. This would also promote a more rigorous approach to characterizing these materials, focusing on the degree of oxidation, crystallinity, and defect density rather than assuming a "graphene oxide" identity. The proposed nomenclature alternatives are not intended as definitive replacements. Rather, they are a call to action for the scientific community to adopt descriptors that better reflect the material's variability. Such a reclassification could enhance our understanding of these materials and foster more precise communication within the field.

In summary, the idea of "graphene oxide" as a distinct material with well-defined properties appears to be a misconception. What is often termed "graphene oxide" encompasses a variety of oxidized carbon materials with varying levels of crystallinity and oxygen content. As such, it would be more scientifically accurate to classify these materials according to their actual structural and compositional features rather than adhering to a term that lacks a precise meaning. Establishing a more rigorous and standardized approach to defining "graphene oxide" is not merely an academic exercise; it has practical implications for industries ranging from electronics to biomedicine. Clear definitions would improve reproducibility, enable more accurate comparisons across studies, and facilitate the development of applications that leverage the unique properties of these materials. Just as graphene has faced challenges not in its definition but in its production quality (*The war on fake graphene*),[5] "graphene oxide" lags even further behind, still grappling with inconsistencies that hinder its standardization and reliable application. Perhaps it is time to accept that "graphene oxide" is less a material in the conventional sense and more a reflection of our collective struggle to pin down the true nature of an ever-elusive nanoworld.

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