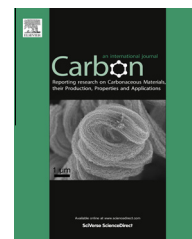


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Editorial

All in the graphene family – A recommended nomenclature for two-dimensional carbon materials

ABSTRACT

Interest in two-dimensional, sheet-like or flake-like carbon forms has expanded beyond monolayer graphene to include related materials with significant variations in layer number, lateral dimension, rotational faulting, and chemical modification. Describing this family of “graphene materials” has been causing confusion in the *Carbon* journal and in the scientific literature as a whole. The international editorial team for *Carbon* believes that the time has come for a discussion on a rational naming system for two-dimensional carbon forms. We propose here a first nomenclature for two-dimensional carbons that could guide authors toward a more precise description of their subject materials, and could allow the field to move forward with a higher degree of common understanding.

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1. Introduction

Graphene research continues to fill the pages of scientific journals, including *Carbon*, and one cannot help but notice some confusion and inconsistency in naming. The confusion does not arise in connection with the isolated, single-atom-thick sheet [1], now universally referred to as “graphene”, but rather with related two-dimensional (2D) sheet-like or flake-like carbon forms. In addition to (monolayer) graphene, we have ultrathin multilayer materials made by graphite exfoliation, which are of significant commercial interest as composite fillers; we have few-layer materials with either ABA stacking or rotational faulting that gives rise to electronic decoupling of the individual layers [2]. We also have graphene oxide and other chemically modified graphenes [3], as well as carbon materials made from graphene or graphene oxide as atomically-thin precursors, which can be stacked, folded, crumpled, or pillared into a myriad of three-dimensional (3D) architectures [4,5]. Many of these materials are new, and are either scientifically or technologically interesting in their own right. Together with graphene they form a family of ultrathin, two-dimensional carbon materials, much in the way that the various types of carbon nanotubes and nanofibers form a family of one-dimensional carbons that defined a new field in the 1990s.

The literature on 2D carbons has grown organically with authors defining terms as needed to describe their products. We see, however, that many authors refer to “graphene” when a quick read through the *Methods* section clearly reveals the paper is entirely about graphene oxide, or about exfoliated graphite flakes. This is so common that authors may now write “monolayer” graphene or “single-layer” graphene [6] just to emphasize that the idea being communicated is about the true single-layer material, graphene, and not one of these related materials.

Some of the terms used by researchers and manufacturers seem contradictory or unscientific. Materials are referred to as “graphene”, even when they contain hundreds of layers, or are particles that lack the basic sheet-like structure of a 2D material. Some authors use “graphene” because they observe 002 lattice fringes by TEM, and these are classically associated with “graphene layers” as the fundamental building blocks of all sp^2 -based carbons. A recent submission to this journal referred to a spherical carbon particle with a crude concentric crystalline order as a “graphene ball”! Is a coal char particle a type of “multilayer graphene”, because it has internal structures with visible 002 lattice fringes? We do not believe this very broad use of “graphene” is desirable for the health of the new field of 2D carbon forms.

To build a new scientific field, one needs a rational scientific nomenclature. We believe the time has come to open a

discussion on nomenclature for two-dimensional carbon forms. What better place than the journal *Carbon*, whose scope is focused on these very materials? So we, the international editorial team for *Carbon*, have made a first attempt to recommend rational names.

Some of the definitions and concepts we propose below will be obvious to established carbon scientists, but we hope that compiling them here in print may be useful to younger scientists and people entering the field. Other definitions raise deep issues in carbon science, and we hope the discussion of those points will be of interest even to experienced researchers. Finally, we want to recommend definitions that are logical and useful for all of the disciplines and subfields in the carbon science community, rather than narrow definitions that serve only a single field or are useful in only one set of applications.

2. Basic principles

Before we present a set of recommended definitions, it will be useful to discuss the basic principles that led us to these definitions.

Principle 1: Replace the single-word term “graphene” with more precise terms as needed to distinguish the various members of the graphene material family. The single-word term “graphene” is now being used in the literature to refer to many different materials, and this casual overuse can cause readers to miss important scientific distinctions. Our first principle is to discourage this and reserve “graphene” for the isolated monolayer. For other materials we want other, specific terms that distinguish the material from the pristine, isolated monolayer, such as “bilayer graphene” or “reduced graphene oxide”.

Principle 2: Distinguish “graphene”, the two-dimensional material, from “graphene layer” the structural element in graphite and other 3D carbons. Authors who wish to discuss sheets of sp^2 -bonded carbon atoms within 3D carbons can use the term “graphene layer”, which has been widely used in the carbon science literature for decades prior to Geim and Novoselov’s discovery in 2004 [1]. In *Carbon* this usage began to appear in titles and abstracts in the late 1980’s, and became quite common in the 1990s. It is usually attributed to Hans Peter Boehm (Carbon Honorary Advisory Board member) and coworkers [7] who in 1986 defined graphene in an Editorial contribution on the pages of *Carbon* as a hypothetical final member of infinite size of the polycyclic aromatic hydrocarbon series naphthalene, anthracene, phenanthrene, tetracene, coronene, ovalene, etc. [7]. The substances in this series have the common ending “ene” for organic compounds with carbon–carbon double bonds and its last member contains “graph” from graphite, a root that derived from the Greek word for drawing, which is an early use of graphite. For the final committee recommendations from the 1986 proposals, see Boehm et al. [8]. At that time, “graphene” had not been isolated from 3D carbon bodies, but the hypothetical “graphene layer” became a useful concept for thinking about and describing the structure of carbon materials. Carbons were regarded as assemblies of these “graphene layers” whose size, orientation, and degree of perfection defined a carbon material’s properties.

When graphene layers stack, they may have the Bernal ABA structure or the rhombohedral ABCA structure, as in the different crystalline phases of graphite. Alternatively there may be no defined positional relation between atomic positions in one plane and those in other planes, as if wire mesh screens were randomly stacked one upon the other. This latter structure is referred to as “turbostratic” in the older literature, and is sometimes referred to as “rotationally faulted” in the recent literature. The distinction between Bernal stacking and rotational faulting can be very important, as the lack of three-dimensional order in rotationally faulted structures leads to electronic decoupling of the layers, which allows multilayer structures to exhibit massless Dirac fermions and high carrier mobilities that are characteristic of the isolated graphene layer [2].

Regardless of the stacking arrangement, the structural unit in all of these materials is the “graphene layer”. Graphite, multilayer graphene, and even most disordered carbons with sp^2 -bonding are said to be composed of graphene layers regardless of spatial relation among neighboring layers (Bernal stacking or rotational faulting/turbostraticity). Graphene layers in carbon materials may be extended flat planes, or may have limited lateral dimensions of only several nanometers. The “graphene layer” is a useful theoretical construct for discussing and visualizing the fine structure of carbon materials, and for describing the “texture”, which refers to the orientational patterns among the layers that determines anisotropic properties in carbons. This is classic carbon science terminology from the 1980’s, and we feel it should not be abandoned just because we have now found a way to isolate those layers to make new free-standing two-dimensional materials.

Principle 3: Consider the lateral dimensions. Lateral dimensions or widths of graphene materials range from tens of nanometers to micrometers to macroscopic dimensions, and these lateral dimensions may affect percolation thresholds, band gaps, cell interactions, and many other properties and behaviors. A nomenclature should provide some guidance for describing 2D materials in ways that capture not just thickness and layer registry, but also in-plane sizes – the “lateral dimension” as in “microsheets” or “nanosheets”.

Principle 4: Base names on crystallography and morphology. One might be tempted to develop a nomenclature based primarily on material features or properties that are the most interesting or most important for device performance. Unfortunately the material features that govern performance in one application (e.g., carrier mobility in nanoelectronics) are not the same as in another application (e.g., aspect ratio in composite materials with percolating filler phases). We believe a better approach is to rely on crystallography, because atomic arrangements define the phases of matter and are more fundamental than any particular property or set of properties. Classification of nanoscale materials, however, cannot be based on crystallography (phase) alone, but must also include morphological descriptors for shape and size. For graphene materials shape and size are most conveniently expressed by the number of layers (or thickness), lateral dimension, and in-plane shape. In-plane shapes may be anisotropic (ribbon-like) or roughly isotropic (equi-axial), and may take on a myriad of polygonal shapes often with

ragged edges. We are unaware of efforts to systematically describe or name the many in-plane shapes seen in graphene materials, except for the zig-zag and armchair varieties of graphene ribbons, and will for now focus here on the number of layers (thickness) and the nominal lateral size.

Thickness (layer number) is a key variable in graphene science and technology and should be specified accurately wherever possible. In practice, some threshold layer number is needed to distinguish materials in the graphene family from conventional carbon forms. Unfortunately, the choice of such a threshold value is not fully clear. The interest in the graphene field has compelled researchers to develop new ways of milling or chemically exfoliating graphite into ever smaller layer packets, and there is now a continuum of thickness values in 2D (high-aspect-ratio) carbons from 0.34 nm (monolayer graphene) up to micrometers. The electronic structure of these materials becomes indistinguishable from graphite when layer numbers exceed about ten [9], and this layer number is now sometimes cited as the boundary between graphene materials and graphite materials. This is not entirely satisfactory for a general nomenclature, since it is based on electronic properties, which are only one aspect of graphene science and technology. Nevertheless, this observation about electronic property transitions provides at least some physical basis for choosing a threshold layer number, and will therefore be used in our recommended nomenclature below.

Principle 5: Make use of established definitions in nanoscale science. At the nanoscale, material size and shape modulate the bulk properties, and this size dependence is central to the nanotechnology movement of the past several decades. The US federal government has defined nanotechnology as “the control and restructuring of matter at the nanoscale in the size range of about 1–100 nm, in order to create materials, devices, and systems with fundamentally new properties and functions due to their small structure”. Similarly, a nanomaterial is a material with “one or more dimensions between 1 and 100 nm”. Interestingly, if one adopts this definition literally, single or bilayer graphene with lateral dimension >100 nm would not be a nanomaterial, since its thickness is less than 1 nm! Neither would C₆₀ at 0.7 nm diameter, or some single-walled carbon nanotubes of diameter <1 nm. The use of “about 1–100 nm” would allow most people to include SWCNTs, C₆₀, and graphene in the definition of nanomaterials. The European Commission also defines nanomaterials as those having “one or more external dimensions in the size range 1–100 nm”, but to avoid the problem of excluding these well-known nanomaterials included a clause: “fullerenes, graphene flakes and single wall carbon nanotubes with one or more external dimensions below 1 nm should [also] be considered as nanomaterials”. This clause is potentially important for the legal definition of graphene, and its potential regulation for health and safety along with other nanoscale materials.

We believe these established definitions from nanoscale science are useful for rationalizing graphene nomenclature.

The prefix “nano” can be used to describe the lateral dimensions, as in “graphene oxide nanosheets”, which are monolayer materials with lateral dimensions below 100 nm. Such nanoscale sheets are of interest for increased dispersibility, enhanced uptake by living cells uptake, or size-dependent band gap. The prefix “nano” can also be used logically to distinguish ultrafine graphite forms (thickness <100 nm) from thicker flakes, such as those produced by traditional graphite milling operations. There is no need, however, to include “nano” to describe the thickness of a material, since the presence of the word “graphene” itself implies a 2D material whose thickness is always much less than 100 nm.

Principle 6: Promote continuity in the literature. Finally, we want to adopt many of the terms now in use, where possible. We prefer to clarify rather than replace existing terms that have grown organically, if we can do so within our rational naming system, because recommending a complete abandonment of terms used over a decade of graphene-related research would be disruptive and impractical.

3. Definitions

With the above principles in mind, we propose the following terms for graphene and related graphene-based materials.

Graphene – a single-atom-thick sheet of hexagonally arranged, sp²-bonded carbon atoms that is not an integral part of a carbon material, but is freely suspended or adhered on a foreign substrate. The lateral dimensions of graphene can vary from several nanometers to the macroscale. Note with this definition, other members of graphene family of 2D materials cannot be simply called “graphene” but must be named using a unique multi-word term that distinguishes them from the isolated monolayer (see below).

Graphene layer – a single-atom-thick sheet of hexagonally arranged, sp²-bonded carbon atoms occurring within a carbon material structure, regardless of whether that material structure has 3D order (graphitic) or not (turbostratic or rotationally faulted). The “graphene layer” is a conceptual structural unit that has been used for many years to describe the structure and texture of 3D carbon materials with primary sp²-hybridized bonding.

Turbostratic carbon – three-dimensional sp²-bonded carbon material in which there is no defined registry of the layers, meaning there is no spatial relationship between the positions of the carbon atoms in one graphene layer with those in adjacent layers. The name derives from “turbo” (rotated) and “strata” (layer) and can also be called rotationally faulted. This is a common structure in carbon materials prepared at lower temperatures or in “hard carbons” that do not pass through a fluid phase during carbonization and resist the development of 3D crystalline order even upon very high-temperature heat treatment

Bilayer graphene, trilayer graphene¹ – 2D (sheet-like) materials, either as free-standing films or flakes, or as a

¹ An alternative and general method for describing the materials listed above (bilayer graphene, trilayer graphene, few-layer graphene, multi-layer graphene) is to identify the form (flake, film, coating) modified by a compound adjective based on the “graphene layer” concept as a structural component. For example, one could describe a “four-graphene-layer flake”, a “bi-graphene-layer coating”, or a “multi-graphene-layer shell”. This is a logical usage and is flexible enough to describe a wide variety of carbon architectures.

substrate-bound coating, consisting of 2 or 3 well-defined, countable, stacked graphene layers of extended lateral dimension. If the stacking registry is known it can be specified separately, such as “AB-stacked bilayer graphene”, or “rotationally faulted trilayer graphene”.

Multi-layer graphene (MLG)¹ – a 2D (sheet-like) material, either as a free-standing flake or substrate-bound coating, consisting of a small number (between 2 and about 10) of well-defined, countable, stacked graphene layers of extended lateral dimension. If the stacking registry is known it can be specified separately, such as “ABA-stacked multi-layer graphene”, “Bernal-stacked multi-layer graphene” or “rotationally faulted multi-layer graphene”. Carbon films containing discontinuous or fragmented graphene layers of very small lateral dimension should be called “carbon thin films” rather than “multi-layer graphene”, since they do not consist of a defined number of countable graphene layers of extended lateral dimension.

Few-layer graphene (FLG)¹ – a subset of multi-layer graphene (defined as above) with layer numbers from 2 to about 5.

Graphite nanoplates; graphite nanosheets; graphite nanoflakes; 2D graphite materials with ABA or ABCA stacking, and having a thickness and/or lateral dimension less than 100 nm. The use of nanoscale terminology here can be used to help distinguish these new ultrathin forms from conventional finely milled graphite powders, whose thickness is typically >100 nm. An acceptable alternative term is ‘ultrathin graphite’, though “ultra” is less specific than “nano” in describing the maximum thickness.

Exfoliated graphite – a multilayer made by partial exfoliation (thermal, chemical, or mechanical) of graphite into thin multilayer packets that retain the 3D crystal stacking of graphite. This is an operational definition – one based on the fabrication process rather than the resulting material – and as such can overlap with other definitions such as graphite nanoplates (above).

Graphene nanosheet – a single-atom-thick sheet of hexagonally arranged, sp²-bonded carbon atoms that is not an integral part of a carbon material, but is freely suspended or adhered on a foreign substrate and has a lateral dimension less than 100 nm. “Graphene nanosheet” is now commonly used in the literature to refer to all graphene materials, but “nano” is not needed here as all “graphene” samples are very thin. That use of “graphene nanosheet” is not recommended as it interferes with its more logical use to describe the important subset of graphene materials with lateral dimension in the nanoscale (<100 nm).

Graphene microsheet – a single-atom-thick sheet of hexagonally arranged, sp²-bonded carbon atoms that is not an integral part of a carbon material, but is freely suspended or adhered on a foreign substrate and has a lateral dimension between 100 nm and 100 μm. This term is recommended over the more general “graphene”, when one wants to emphasize the micrometer scale of the lateral dimension in cases where it is key to properties or behaviors.

Graphene nanoribbon – a single-atom-thick strip of hexagonally arranged, sp²-bonded carbon atoms that is not an integral part of a carbon material, but is freely suspended or adhered on a foreign substrate. The longer lateral dimension

should exceed the shorter lateral dimension by at least an order of magnitude to be considered a ribbon, and the shorter lateral dimension (width) should be less than 100 nm to carry the prefix “nano”.

Graphene quantum dots (GQD) – An alternative term for graphene nanosheets or few-layer graphene nanosheets, which is used particularly in studies where photoluminescence is the target property. Generally, GQDs have very small lateral dimensions <10 nm (average ~5 nm) at the lower end of the range for graphene nanosheets, which is <100 nm lateral dimension [10]. Some GQDs may be few-layer materials.

Graphene oxide (GO) – chemically modified graphene prepared by oxidation and exfoliation that is accompanied by extensive oxidative modification of the basal plane. Graphene oxide is a monolayer material with a high oxygen content, typically characterized by C/O atomic ratios less than 3.0 and typically closer to 2.0.

Graphite oxide – a bulk solid made by oxidation of graphite through processes that functionalize the basal planes and increase the interlayer spacing. Graphite oxide can be exfoliated in solution to form (monolayer) graphene oxide or partially exfoliated to form few-layer graphene oxide.

Reduced graphene oxide (rGO) – graphene oxide (as above) that has been reductively processed by chemical, thermal, microwave, photo-chemical, photo-thermal or microbial/bacterial methods to reduce its oxygen content.

Graphenization – the development, growth, or perfection of graphene layers during the processing of disordered carbonaceous solids. The graphene layers may occur within a 2D (sheet-like) or 3D carbon material. A related term is “carbonization”, which refers to the primary conversion of organic material into a carbonaceous solid - one consisting primarily of elemental carbon - regardless of structure. Also related is “graphitization”, which refers to the development of 3D crystalline order including defined ABA or ABCA graphene layer registry, which typically appears only in the later stages of order development. In some cases, graphenization may occur after carbonization as a distinct process of solid-state rearrangement leading to the loss of amorphous material and appearance of well-defined graphene layers. In other cases, graphene layers may grow directly from the gas or melt phase without undergoing separate identifiable stages of carbonization and graphenization.

Graphene materials (also graphene-based materials, graphene nanomaterials, graphene-family nanomaterials) – overarching terms for the collection of 2D materials defined above that contain the word “graphene”, including multilayered materials (*N* less than about 10), chemically modified forms (GO, rGO), and materials made using graphene, graphene oxide, or another graphene material as a precursor.

Graphenic carbon materials – the broadest class of carbonaceous solids that consist primarily of elemental carbon bonded through sp²-hybridization. Graphenic carbon materials, or “graphenic materials”, include the 2D forms described in this article (graphene materials, as above), as well as 3D forms whose structures are based on the “graphene layer”

as the conceptual structural unit, including activated carbons, carbon fibers, chars, cokes, and bulk graphite.

4. Derivative terms

There are numerous ways in which these terms can and should be combined to describe new materials more accurately than we do presently. A few examples are:

Graphene oxide nanosheets – graphene oxide (monolayer) with lateral dimension less than 100 nm. These materials form a subclass of graphene oxides that are more readily dispersible and are of interest for biomedical delivery applications.

Few-layer graphene nanoribbons – a 2D material with 2 to about 5 layers, an aspect ratio in the lateral plane greater than about 10 and a width less than 100 nm.

Multilayer graphene oxide film – a multilayer ($N \geq 2$) structure typically made by restacking of graphene oxide monolayer sheets.

Graphene materials can also be processed or hybridized in a myriad of ways, leading to additional modifying terms that include “folded”, “wrinkled”, “activated”, “decorated” or “functionalized”, and the resulting product names can and should reflect this processing, such as titania-decorated reduced graphene oxide microsheets, or activated microwave-exfoliated graphene oxide [11]. These terms are useful and offer precise description of new materials.

5. Additional examples of terms or usages that are not recommended

Below are a few more example terms and usages we see that do not follow the principles that guided this nomenclature:

Graphite layer – this term is not recommended since “graphite” refers to the three-dimensional crystal, and a single layer cannot have the minimal structure required to define graphite. The correct term is “graphene layer” regardless of where the layer occurs or its relation to neighboring layers.

Graphene nanosheet – this commonly used term is not recommended when referring to graphene of microscale lateral dimension, or to any multilayer material, or to any type of graphene oxide regardless of size or number of layers.

Graphene nanoplates, graphene nanoplatelets – these terms are used for some industry products with microscale lateral dimension, but are not recommended in our scientific nomenclature for reasons given above. “Graphene” does not need the prefix “nano” to indicate thinness, and instead “nano” used in this way should indicate the lateral dimension.

Graphitic – this term is not recommended when the only information available is the appearance of 002 lattice fringes by TEM. Graphitic requires the existence of 3D order or layer registry, which is not directly observable by conventional TEM fringe imaging. This is a common misuse among authors and applies not only to the 2D materials of primary interest in this article, but also to all graphenic carbon materials.

6. Summary

We hope our readers will agree that referring to a material structure as a “reduced few-layer graphene oxide film” is much more descriptive and scientifically accurate than referring to the same structure as just “graphene”, as many authors do now in their articles. We hope our *Carbon* contributors will adopt such a practice. Use of rational names only achieves so much in science, however. We recommend that all authors working on 2D materials provide characterization wherever possible in their research articles. Chemical composition, crystal structure, thickness, lateral dimension, and their distributions help tremendously in defining a new material.

Finally, we anticipate there will continue to be some overusing of the word “graphene” as a catch-all for many materials, both because it is easy to fit into a paper title, and because researchers and companies want to be associated with this exciting field. So do we. Partly for this reason are we recommending “graphene materials” or “graphene-based materials” as overarching phrases to be used to describe a range of emerging 2D materials in this field. As we build this new area of science and technology, we encourage our *Carbon* authors and the international carbon community to be more precise in the description of its products, and we hope this article can help guide authors and allow the field to move forward with a higher degree of common understanding.

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