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*Recent Progress in Materials*

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Short Review

# Versatile Surface Chemistry of Carbon-Based Nanoplatforms by Covalent Bonding, Non-Covalent Linking, Crosslinking, and Self-Assembling

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**Academic Editor:** Chih-Ching Huang

**Special Issue**: Hybrid Graphene-based Materials: Synthesis, Characterization, Properties, and Applications

*Recent Progress in Materials*
2023, volume 5, issue 4
doi:10.21926/rpm.2304034

**Received:** July 28, 2023
**Accepted:** November 09, 2023
**Published:** November 15, 2023

**Abstract**

This mini-review discussed the best-known chemical and synthetic methodologies used in the last years to modify carbon allotropes, with an interest in nanotechnology. In this perspective, chemistry with optoelectronics applications and recent trends within bio-applications focusing mainly on graphene and its derivatives were considered. So, the mini-review intended to focus on methodologies to add functional groups with varied reactivities, polymer chemistry, and nanoscale control. These methodologies provide insight for further developments. In this manner, traditional methods using strong acid media to convert simple carbon bonds into carboxylic acid and aldehydes organic functional groups were shown and discussed. Hence, chemical modifications in a variety of solvents could be developed. Notably, many organic chemical reactions, such as bimolecular nucleophilic substitution (SN2), click chemistry, and photochemical reactions, showed essential insights in designing the carbon-based material modifications and the bottom-up method. Moreover, incorporating atomic entities within graphene material defects led to interesting spectroscopic and quantum

properties. The atomic change added blemishes to this homogeneous structure, which was tuned to modify optical properties. In addition, the review was also oriented towards the discussion on incorporating polymeric films, such as boron- and silicon-based monomers, to form polymeric-modified carbon-based slides. In this way, organoboranes and organosilanes permitted chemical functionalization because their chemical modification was more accessible on nanosurfaces. Moreover, emphasis was placed on exploiting non-covalent bonding with ions and polarized molecules with the highly electronic densities of graphene and its derivatives. In this manner, the manuscript intends to summarize the main types of reactions and synthetic pathways reported until today. Therefore, particular focus was given to chemical composition, 2D and 3D chemical structures, and their properties related to non-covalent interactions. Thus, based on the properties and reactivity of carbon allotropes, the review was intended to open the analysis and discussion, considering the design of new carbon-based materials, hybrid nanocomposites, and metamaterials.

**Keywords**
Covalent bonding; non-covalent interactions; derivatives; chemical modifications; material conjugation; carbon allotropes

## 1. Introduction

Carbon-based materials, such as allotropes, are of high interest and broadly used in research and the industry [1] for different reasons. However, new nanomaterials with incorporations of graphene [2], carbon nanotubes [3], fullerenes [4], and derivatives are still the focus of further modifications and studies. In particular, the chemistry of carbon-based materials is not as easy when it is considered within a large group of synthetic methods of organic compounds [5]. The main reason is that they were considered from the beginning to be low-reactive chemical compounds with very low solubility in many solvents that generally require targeted chemical modifications depending on the media [6]. In these perspectives, a revision of the reactivity of carbon-based materials and main chemical reactions was carried out, developed with further perspectives aimed at designing new nanomaterials and hybrid nanocomposites.

Thus, wet chemistry [7], used mainly within clean rooms that allowed fast chemical modifications, could not be implemented, and there is still an interest in developing new, speedier chemical reactions and methodologies. Moreover, graphene and its derivatives have shown exciting and high-impact perspectives within nano-optics and its applications in recent years [8]. Wet chemistry is of high interest to the Laboratory of Optics in its ability to modify surfaces and generate new optical properties. Chemists are looking to incorporate different visually active materials into new visual composites.

As it is known, graphene is a carbon-based chemical structure with a homogeneous and infinite electronic density delocalized wave through a surface, which generates a variety of particular properties within different energy levels [9]. It could be mentioned that it also has semi-metallic and semi-conductive properties that allow unusual electronic behaviors and quantum phenomena [10]. Thus, Graphene Quantum Dots with particular confined electronic properties could provide

engaging Optical active platforms to tune emissions by controlling their tiny chemical surface modifications [11]. Improved electronic conduction in the presence of one of the most common conductors, such as copper, is one of the best-known insights within current materials [12]. Moreover, due to their high sensitivity against low opto-electro-stimulations, graphene-based materials allowed themselves to be transferred towards reduced-size devices and flexible wearables [13]. In addition, quantum properties from varied levels of energy and phenomena produced particular interferences and modifications, too, such as within the Fermi level with anomalous improved quantum signal generation in the presence of twisted structures [14] and pseudo-electromagnetic fields [15].

Moreover, proper polymeric modifications on Quantum surfaces could provide exciting applications such as UV-protective bio-based epoxy/carbon quantum dots nanocomposite coatings [16]. , polyurethane [17], and polyester hybrid composites [18]. So, controlling graphene's chemistry and its combination with other opto-active materials is highly interesting to chemists and physicists [19]. However, intense media modifications should be used to tune their low reactivity and chemical functionalization, such as concentrated inorganic acid additions [20], providing high energies associated with forming new covalent bond formations. In this state of affairs, this mini-review aims to show the best-known and most recently applied chemical modifications of graphene and its derivatives to functionalize and tune chemical structures for specific studies and targeted applications. Besides, as could be noted, the chemical reactivity associated with each shown material highlighted their previously mentioned properties associated with particular electronic configurations and 2D and 3D chemical structures. This way, the versatility of carbon allotropes to tune optoelectronics by covalent and non-covalent interactions was discussed. So, considering their highly electronic densities, how Vander Walls and polar interactions could join the ions was shown [21]. In this manner, multi-charged ionic layers were formed and allowed for the creation of detection systems of ions, such as heavy metal ions and charged chemical species [22]. The chemical surfaces of carbon allotropes permitted the exploitation of electronic waves and their variability to sense and chemically modify their characters.

Moreover, π-π stacking [23] bonding could provide alternative non-covalent interactions to join organic molecules and linkers [24]. Furthermore, atom incorporation within interstitial spaces and into atomic defects within the structure [25] could be achieved as well. Thus, 2D graphene surfaces modified their homogeneity and the chemical properties associated with electronic and energy variations [26].

In addition, covalent modifications by carbon modifications and functionalization were achieved. Thus, oxidation was produced in carboxylic and aldehyde groups [27] in drastic and mild conditions, which allowed for the covalent linking of varied materials. Other modifications were discussed based on incorporating polymeric films, such as boron- and silicon-based monomers. In this manner, organoboranes [28] and organosilanes [29] permitted chemical functionalization as well (**Figure 1**). The versatility is achieved by summarizing the reported strategies; in this manner, from an overview standpoint, it checks possibilities and evaluates new ones in the context of the design of the generation of hybrid optical active nanocomposites, where carbon-based materials could play essential functions.

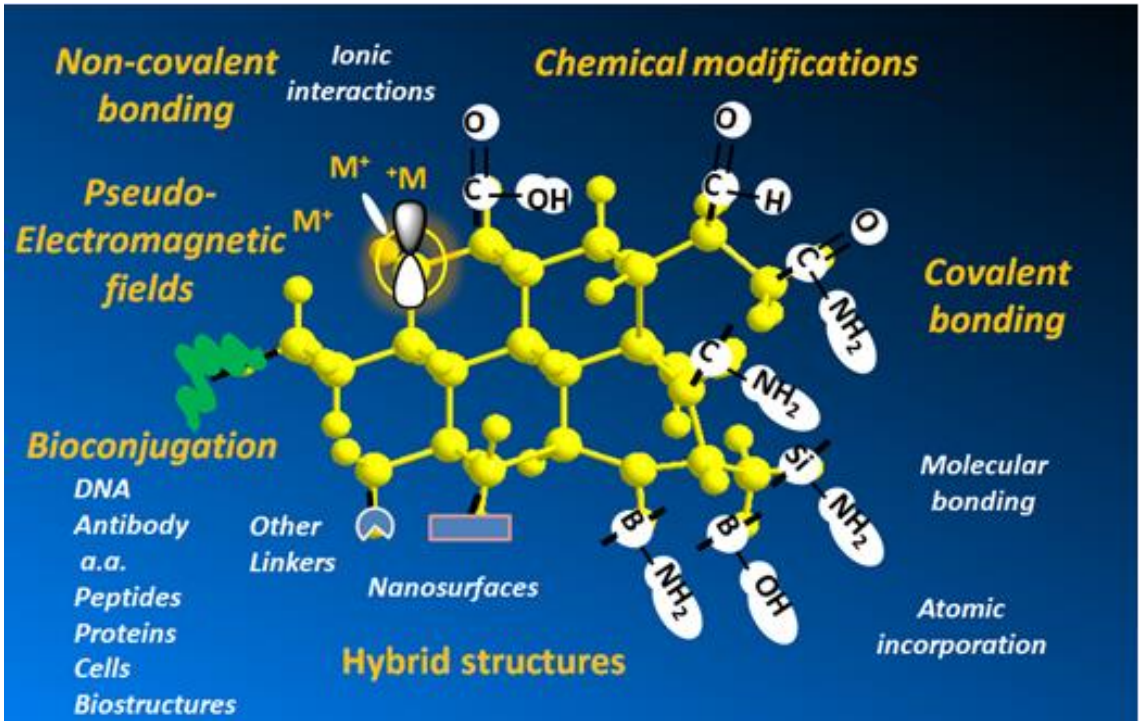


**Figure 1** Schema of chemical modifications on highly conjugated carbon-based materials. Reprinted with permission from A. G. Bracamonte et al. Copyright 2023.

By following these different strategy pathways, it is possible to generate varied new linking sites based on i) covalent bonding, ii) non-covalent linking, ii) ionic interactions, iii) π-π stacking, iv) Vander Walls interactions, v) polar interactions, vi) electromagnetic fields, magnetic and electrostatic interactions, etc. These variable linking forces should be done by tuning the material with chemical or physical modifications. Therefore, different types of molecules and materials could be added. In addition, incorporating hetero-atoms within graphene material defects led to interesting spectroscopic and quantum properties. The atomic modification added blemishes to these homogeneous structures, which were tuned to modify optical properties [30]. In a similar manner to new Carbon chemical structures such as graphitic carbon nitride/carbon quantum dots (GCN/CQD) using simple sources of Carbon and Nitrogen from starch and urea, respectively (**Figure 2**), developed different chemical structures and Opto-electronic perspectives [31].

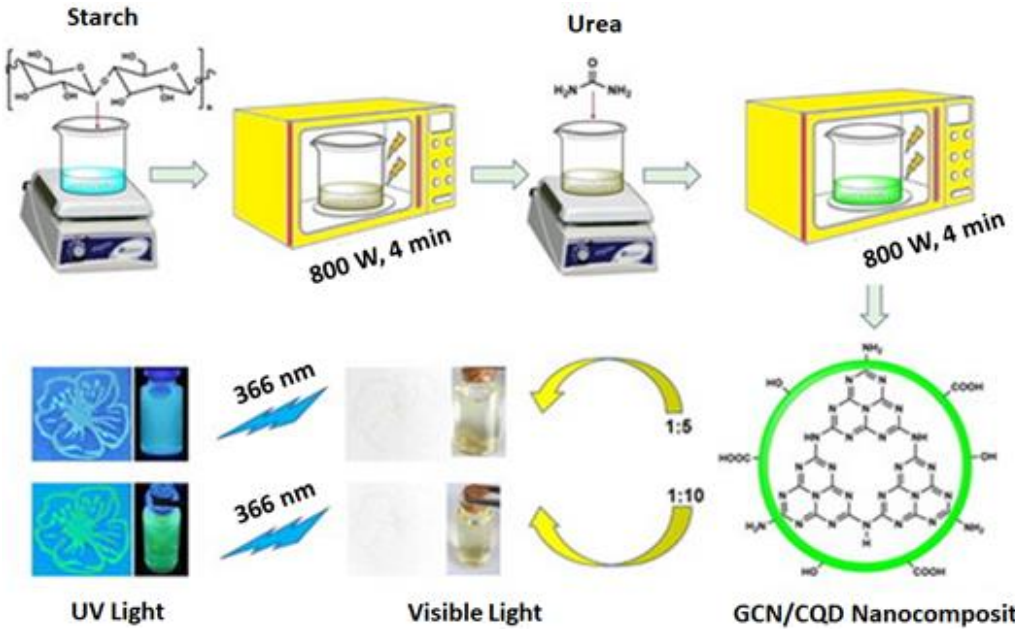


**Figure 2** Schema of Synthesis of graphitic carbon nitride/carbon quantum dots (GCN/CQD) using starch and urea. Reprinted with permission from Madhi, A. et al. Copyright 2023. Fullerenes, Nanotubes, and Carbon Nanostructures [31].

In this manner, it was possible to discuss the design of nanomaterials, hybrid, and functional materials to lead fundamental research focused on photo-physics, quantum materials, and metamaterials [32]. All the discussion was centered on the chemical modifications, advantages, and disadvantages, tuning properties according to needs, and considering chemical changes and targeted applications. In this context, the meaning of tuning properties becomes relevant when it is shown that it is possible to tune highly dense electronic orbitals and, consequently, further electronic waveguiding [33] through and across both sides of the planes of graphene-based materials and their derivatives. In these perspectives, the chemical vapor deposition (CVD) [34] on wafers and other substrates to functionalize materials as platforms to propose new optical active media was also discussed [35].

Thus, an overview of the multidisciplinary research field was shown from the point of view of chemical modifications and functionalization to tune chemistry, functionalization, and susceptible electronic and quantum phenomena. An emphasis was placed on a broad spectrum of possibilities, from optoelectronics applied on micro-devices to bio-applications. In this way, the novelty of this manuscript stems from the fact that it focuses on the leading chemical and physical properties that could be used to tune carbon-based materials by using different covalent and non-covalent synthetic pathways.

## 2. Versatile Surface Chemistry Modifications by Covalent Linking, Crosslinking, and Polymeric Assembling

This subsection is oriented towards the chemical modification of graphene and its derivatives and shows strategies used on other carbon allotropes that could be useful. Thus, as mentioned previously, it is essential to highlight that even if these compounds are considered inert and very stable chemical compounds, with time, it was shown that they are versatile chemical structures

when it comes to tuning their surfaces. This dual characteristic could be explained by considering the carbon electronic configuration and malleable covalent bonds. Therefore, different synthetic strategies and chemical methodologies were used to modify them from single carbon atoms into larger surfaces. The best-known and applied chemical surface modification was achieved by chemical oxidation in drastic conditions, such as solid acid media [36]. From the beginning, the question of controlling this reaction was posed; however, it was not as easy as expected. Even the manipulation of these compounds at the laboratory was not evident due to the low solubility in the best-known organic solvents and the insolubility in aqueous media. But, even in that case, the methodologies developed converted the highly conjugated chemical structure related to highly dense electronic orbitals and apolar characteristics to polar surfaces. These modifications allowed modifying the electronic waves moving on both sides of planes and the interactions between them.

In this context, the reports could be mentioned by modifying oxidant agent ratios. For example, varying mass proportions of $KMnO_4$ to graphite led to different colloidal graphene oxide dispersions. The graphene obtained presented between 1-3 and 4-10 layers, up to even more than ten. This control in the chemical synthesis and variable graphene multi-layer assembly could be detected by ultraviolet-visible (UV-vis) measurements of varied multi-band thick layer dispersions, highlighting an intense peak at 230 nm.

Due to their improved dispersibility and solubility in varied solvents, further chemical modifications are possible after the obtention of controlled graphene oxide nanoassemblies at the multi-layer level. As it is known, the carboxylic groups of graphene oxides could be modified by varied wet chemical methods that, from organic chemistry to optic laboratories and Life Sciences by bioconjugation techniques, showed high-impact insights and applications. It highlights applied developments such as bio-based surface modification of wool fibers by Chitosan-Graphene Quantum Dots Nanocomposites [37]. In addition, it should be noted that, in particular, in 2022, the Nobel Chemistry Prize was awarded to the United States and Denmark for developing click chemistry within biological media for cell modifications [38]. Organic chemistry reactions within cells allowed for targeted molecular changes and labeling. This versatility of small molecular linkers is applied to bioconjugation several types of materials, where carbon-based ones, such as graphene, carbon nanotubes, and derivatives, are also involved due to their fast kinetic and stable thermodynamic properties. Thus, many molecular linkers could be used. For example, non-dispersible carbon nanotubes were modified to increase their dispersibility in water and polar solvents by adding polar organic groups in their structure, such as (-COOH), ($-NH_2$) as well as other spacers [39] (**Figure 3**).

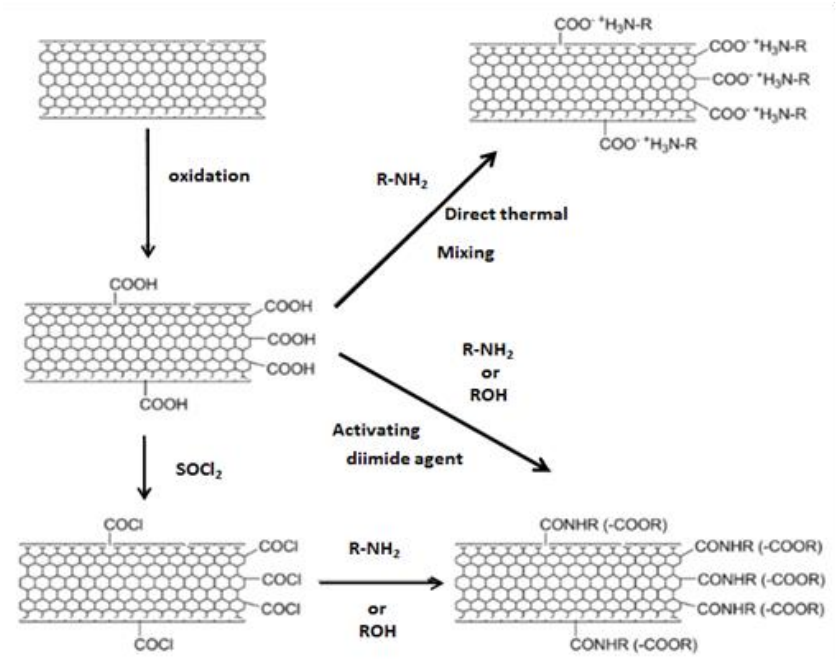


**Figure 3** Chemical modification of carbon nanotubes (CNTs) with amines or alcohols. Reprinted with permission from C. Galiotis et al. Copyright 2010 Progress in Polymer Science [39].

The chemical modification on CNT surfaces with variable chemistries permitted the incorporation of their excellent properties within polymeric nanocomposites; electrical conductivity applications within polymeric conductive substrates based on this material concept were noted. Moreover, due to their small size, these nanostructures' excellent optical active properties can be homogeneously embedded into lightweight matrices, like those offered by engineering polymers incorporated within wearables and conductive films from electronic films and devices. In this manner, the variable chemical strategies for grafting polymers onto carbon nanotubes or their incorporation, such as carbon nanotubes within polymer substrates, could provide different optoelectronic properties. Thus, by embedding other intercalant agents, electron shuttle molecules, and semi-conductive materials, new properties that could be considered in many cases, such as metamaterials, could be tuned [40].

In these regards, grafting conductive polymers on graphene oxide through cross-linkers could be mentioned in developing different synthetic strategies, such as a stepwise approach [41]. It is just an example, but in this case, and for the current discussion focused on synthesis and chemical modification, to modify carbon-based materials, the challenge in the manipulation of these types of materials should be highlighted in particular with the incorporation of polymeric chains, polymeric layers, and shells [42]. An emphasis was placed on a three-step reaction to furnish a composite of graphene and a conductive polymer. In brief, i) firstly, graphene oxide was modified with a diamine, which acted as a linker for polymer attachment; then, ii) by an activation site, the free amine was attached, added as a linker; in this way, in a third step, the polymeric growth was initiated and, at the same time, that graphene oxide was reduced. For this methodology, no catalyst, acid, or reducing agent was required to synthesize a graphene-polymer composite directly. As for the characterization, various instrumental techniques were used, including step-by-step AFM analysis, to characterize the structure of the products in each step and confirm the covalent functionalization

among graphene oxide, cross-linker, and polymer. The average surface height was sequentially increased after each step, indicating the success of the sequential reactions. Thus, a solid and stable composite was obtained, with excellent electrochemical performances compared to mechanical mixes that produced improved properties in each case. The importance of the order and chemicals involved showed the impact on the final properties. The graphene-polymer composite showed excellent electrochemical performance and stability compared with a composite prepared by the physical mixing of graphene and polymer. This example opened the discussion to develop new approaches incorporating other optoelectronic active molecules attached to carbon-based materials. It is a mention to be made to mark potential current trends in this high-impact research field. Other surface modifications, such as molecular spacers, optical active molecules, pharmacophores, and additional functional molecules, could be added. This could be the case of the synthesis of resorcinol-functionalized, multi-walled carbon nanotubes as a nano-adsorbent for the solid-phase extraction and determination of diclofenac in human plasma and aqueous samples [43]. To achieve this functional carbon-based material, three simple chemical steps were proposed: i) drastic oxidation in the presence of concentrated nitric acid to form carboxylic acid groups, ii) the addition of cyanuric chloride in dioxane/xylene media, to incorporate the resorcinol in soft primary media then. In this manner, cyanuric chloride and resorcinol were incorporated into the aromatic hydroxyl functionalization of the surface of multi-walled carbon nanotubes (**Figure 4**).

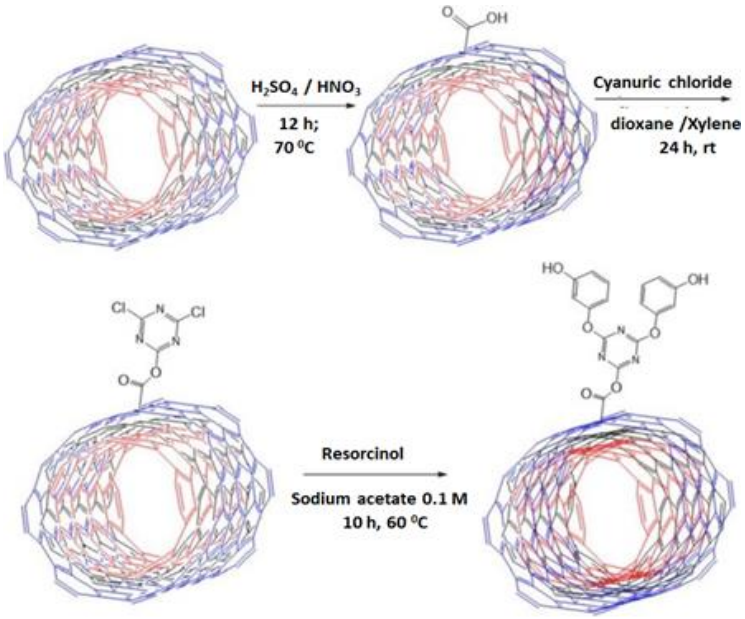


**Figure 4** Synthesis of resorcinol-functionalized multi-walled carbon nanotubes as a nano-adsorbent for the solid-phase extraction and determination of diclofenac; chemical steps; i) drastic oxidation in presence of concentrated nitric acid to form carboxylic acid groups, ii) addition of cyanuric chloride in dioxane/xylene media, to then iii) incorporate the resorcinol in soft primary media. Reprinted with permission from S. H. Khoshravesh et al. Copyright 2022 Colloid and Interface Science Communications [43].

The carbon-based nanomaterial could be considered a versatile chemical surface platform where multi-functions could be tuned by incorporating different molecules with targeted functionalities. Therefore, in controlled conditions, the bifunctionalization of graphene oxide, graphene as a

promising tool for broadening applications, was obtained. The multi-function was associated with the grafting of accurate linking of neighbor molecules regarding site-specific reactions. Thus, mono- and bifunctionalized graphene oxides containing thiol and imidazole groups were reported. Here, anchoring was achieved on the carboxylic acid sites of graphene via amidation reactions. The concentration ratio that led to the consecutive addition of the reagents (containing imidazole and thiol groups) was varied (**Figure 5**). The functionalized materials were evaluated as nanocatalysts in the neutralization reaction of a toxic organophosphate [44].

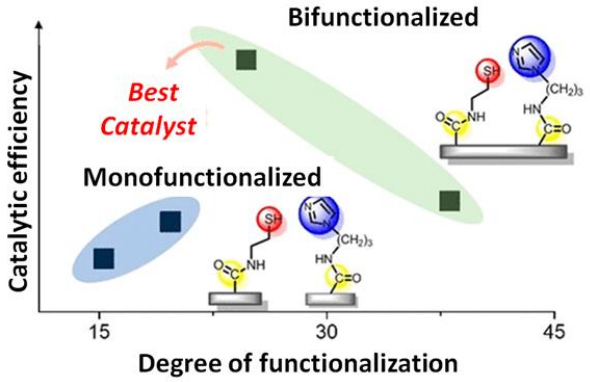


**Figure 5** Distribution of catalytic properties of bifunctionalized graphene oxides containing thiol and imidazole groups as nanocatalysts in the neutralization reaction of a toxic organophosphate. Reprinted with permission from Y. H. Santos et al. Copyright 2022 Colloid and Interface Science Communications [44].

Another strategy used to modify carbon-based materials is their modification with polymers with variable shell lengths, such as for core-shell nanoarchitectures [45, 46] or multilayered substrates [47, 48]. In this context, the encapsulation and modification of carbon-based nanomaterials to design new approaches and materials were highlighted. Thus, the development of high permittivity properties from polymer composites based on long single-walled carbon nanotube incorporation within silicone was recently reported [49]. In this context, the accessible methodology related to the redispersion of variable nanotube lengths in $CH_2Cl_2$ by sonication to mix it with silicone was noted. Miniaturized disks with a diameter of 29.0 mm and a thickness of 3.5 mm were obtained by stirring the mix and heating it in silicon molds. These miniaturized carbon nanotubes silicone substrates allowed for the study of the permittivity of dielectric composites for numerous applications dealing with matter/electromagnetic radiation interactions.

Therefore, these composites demonstrated record-high dielectric permittivity in the low-frequency range (102-107 Hz) and in the X-band (8.2-12.4 GHz), significantly exceeding the literature data for composite materials with similar CNT content. This particular behavior was explained by the specific 3D-structured material in the form of neuron-shaped clusters, from which a dependency on the nanotube lengths in a complex polymer matrix percolating network was highlighted.

In this manner, many strategies to modify carbon-based materials within the nanoscale and beyond to tune nano-optics, nano-electronics, electromagnetics, non-classical light, and electronic phenomena were shown, and further interest within an open spectrum of research fields, from quantum phenomena to bioconjugation, bioapplications and hybrid synthetic nano-biomaterials and green photonics materials, to mention some of the possible current areas of interest in progress.

## 3. Physical Phenomena to Tune Surface Interactions and Properties: Non-Covalent Interactions and Related Strategies

Carbon-based materials, such as graphene, carbon nanotubes, graphene quantum dots, graphene derivatives, fullerenes, etc., are all associated with homogeneous chemical structures with highly electronic densities moving on different sides. Thus, electromagnetic waves from highly conjugated carbon atoms in a plane could be polarized against external media modifications and stimulated remotely, such as by laser excitation [50, 51]. It should be noted that electromagnetic properties were studied and exploited to develop electronic and quantum signal transmission through space and time. In this context, it should be highlighted that, regarding these physical and chemical properties, there is a vast number of publications on varied research themes and topics, focusing on only one at a time. In this regard, this section is intended to show and discuss the best-known properties from recent reports from the literature and current studies that could provide the basis for intangible interactions and non-covalent bonding. Besides, in this manner, new strategies to join materials, as well as to couple physical properties, could be added. So, the condensed electronic and quantum properties could not be neglected in the design and modification of these types of materials because they strongly affect physical and chemical modification and interference.

Consequently, this means that the reactivity of carbon allotropes is strongly affected by these properties that should be known. For these reasons, they deserve this highlight and are described in the following examples. Therefore, one of the most essential properties generated from carbon allotropes, such as graphene, is the stimulation towards electromagnetic field generation [52]. This way, pseudo-electromagnetic fields generated from single and multilayered graphene could interact in their surroundings. So, plasmonic properties could also be analyzed from the near field, within closer nanometer lengths, towards the far area within longer distances. Moreover, quantum properties could be affected by these new energy modes. Graphene showed interesting properties for light-matter solid interactions (**Figure 6**) [53], such as graphene-based plasmonics for varied signal modulation and sensing [54].

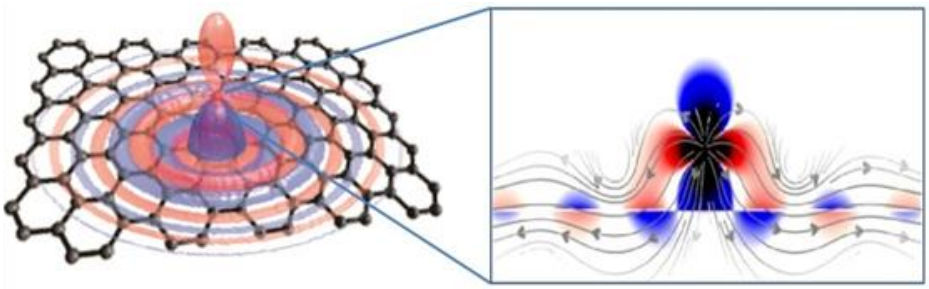

**Figure 6** Scheme of long-distance graphene plasmons generated on single-layered graphene. Reprinted with permissions from Koppens et al. 2011 [53], Nano Lett.

The theoretical calculations and imaging showed that the electronic hotspots and their movement generated potential sites of non-covalent interactions (**Figure 7.i**) with other polarizable and electromagnetically active atoms and materials. This could be the case of multi-layered graphene slides (**Figure 7.ii**) or any other molecules interacting by π-π stacking (**Figure 7.iii**). This molecular polarization capability could be tuned to modify their surface by non-covalent bonding, such as adsorption on grafted graphene surfaces. In this manner, graphene could act as a non-

covalent platform to interact with their close surroundings for different targeted applications such as ion doping of surfaces and targeted ion interactions.

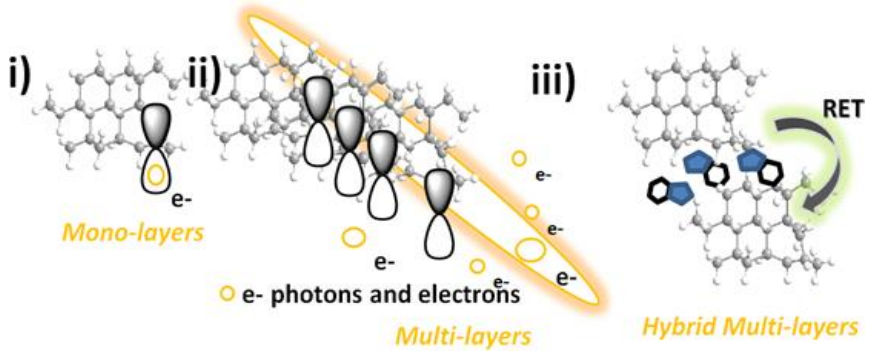


**Figure 7** Schema of highly-conjugated carbon-based materials: i) representation of reduced-sized graphene quantum dots; ii) graphene bi-layers interacting producing electronics and photonics waveguides; iii) molecular intercalating agents incorporated within graphene slides. Reprinted with permission from A. Guillermo Bracamonte et al. Copyright 2023 Current Research in progress.

Recently, high-efficiency ion enrichment inside ultra-short carbon nanotubes was reported [55]. This ion enrichment based on ion-polar interactions offered the possibility to assay alternative ways of water purification and develop ion batteries, super-capacitors, and memory devices where controlled doping of ions showed important roles in charge transfer and detection. In this study, the ion-enrichment capacity was evaluated by varying the length of carbon nanotubes. Results showed that the ion-enrichment power of ultra-short CNTs was much higher than that of longer CNTs. Furthermore, a broad range of ions could be enriched in the ultra-short CNTs, including alkali-metal ions (e.g., $K^+$), alkaline-earth-metal ions (e.g., $Ca^{2+}$), and heavy-metal ions (e.g., $Pb^{2+}$). It was concluded that, under the hydrated-cation-π interactions, the ultra-short CNTs with few end caps and blockages could enrich ions highly efficiently, allowing, in this manner, to alternatively modify graphene surfaces, which paves the way to explore other carbon allotropes.

Similarly, this capability could be incorporated within textures and porous materials, such as flexible zeolitic imidazole-based metal-organic frameworks named ZIF-8. In this manner, multi-layer graphene oxides efficiently removed copper ions [56]. The kinetic fitting of interactions by isotherm curve fitting confirmed that the adsorption law of $Cu^{2+}$ by Graphene@ZIF-8 was the pseudo-second-order kinetic model, and the Langmuir isotherm model was applied. Based on these results, the model showed a process of $Cu^{2+}$ adsorption via a monolayer chemisorption. To address the performance, the maximum $Cu^{2+}$ adsorption capacity of Graphene@ZIF-8 reached 431.63 mg/g under the optimal adsorption conditions. As for the methodology applied to modify these types of porous materials, the simple way of knowing the precise and accurate controlled conditions in each step of this chemical material modification could be highlighted. In brief, as it could be observed, by applying ultrasonication for the dispersion of materials, blending mixes, redispersing, and leaving enough time to interact both main components at each step within colloidal dispersion, it was possible to dry modified powder after the application of vacuum and heating in the last stage of synthesis (**Figure 8**). It is highlighted that the modified composite was achieved by close contact with materials and non-covalent interactions within appropriate media use. Potential perspectives

towards removing heavy metals were opened; research in progress of views in this direction could be found.

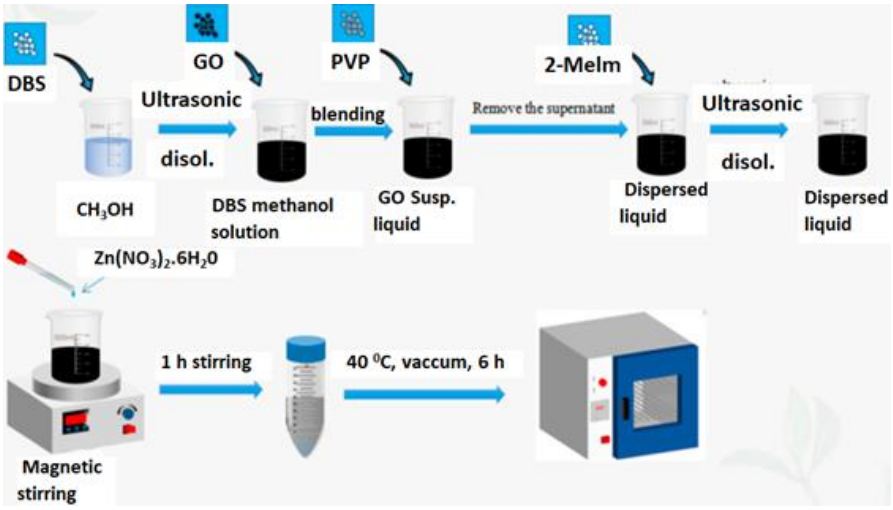


**Figure 8** The preparation process of Graphene@ZIF-8 and assessment for $Cu^{2+}$ adsorbing capacity. Reprinted with permission from X. Lv et al. -C. Shi, Copyright 2022 Nanomaterials [56].

## 4. Other Technological Based Techniques: Vapor Depositions, Laser and Tips Assisted Techniques

This section presents other strategies to modify carbon-based chemical structures to obtain functional materials based on other technological techniques. This way, essential techniques and associated methodologies using physical and chemical phenomena joined to lasers, electromagnetic fields, near-field microscopy, miniaturized tips and devices, etc., are worth noting. In this manner, the technology instrumentation plays the leading role; otherwise, the modified material could not be obtained. However, chemical and physical properties are involved, of course, to be managed by the applied instrumentation. It is a fundamental concept, but it is highlighted as a brief introduction and common factor in all the techniques developed, as well as to be taken into account in new ones. These techniques are considered advanced instrumentation to tune new materials, and they are already incorporated within the manufacturing of advanced materials in the high-tech industry; however, at the same time, they are currently used for research and development due to their versatility in the possible design incorporating different types of materials.

In this regard, there is an emphasis on the techniques and methods where the physical phenomena of material evaporation are implemented and then deposited on surfaces. This is the case of physical vapor deposition (PVD). The PVD is related to many vacuum deposition methods that can produce thin films and coatings on substrates, including metals, ceramics, glass, polymers, and carbon allotropes. Thus, carbon-based materials could be transferred from a condensed phase to a vapor phase to be deposited as a condensed phase in the form of a thin film. The most common methods used to fabricate semiconductor devices and thin films for solar cells, such as sputtering and evaporation, are mentioned [57]. In addition, chemical vapor deposition (CVD) methods where varied chemicals could be deposited on targeted substrates, such as wafers, are mentioned. Wafers are semiconductor thin slices or tiny substrates, such as crystalline silicon (C-Si) pieces, used to design and fabricate optoelectronic chips and devices. It is noted that the molecules involved

undergo chemical modifications in which they react or decompose on the substrate surface. Therefore, the modified substrate is obtained. This methodology allowed for the development of microfabrication processes to achieve monocrystalline deposition, polycrystalline control, and amorphous and epitaxial growth on wafers [58]. Varied materials were used to accurately be deposited on wafers and in a reproducible manner that led to manufacturing the incorporation of silicon (dioxide, carbide, nitride, oxynitride), carbon fiber, nanofibers, carbon nanotubes, diamond and graphene, fluorocarbons, and other types of filaments.

Among recent examples from the literature, several publications focused on graphene CVD can be highlighted. The results showed remarkable material at a large scale without sacrificing the quality, which is very important. In addition, it is not very sensitive to the quality of materials; therefore, it could be developed using materials of different rates depending on the applications. In this manner, varied top-down and bottom-up approaches were found to produce different architectures within heterojunctions and design applications in the energy and electronic devices sectors [59]. Similarly, essential developments were reported by designing and fabricating modified membranes with semiconductor materials. These are possible targeted technological applications in the accurate and current market from where fundamental research always inquires for improvements and innovation [60].

Moreover, expanding strategies incorporating laser-based techniques could be noted, for example, nano-processing of self-suspended monolayer graphene and defect formation by femtosecond-laser irradiation [61]. This technique developed a femtosecond-laser, processing-based methodology of self-suspended monolayer graphene grown by CVD, within holes of a < 100 nm diameter produced in situ by multipoint drilling. Thus, scanning the transmission of electron microscopy revealed the formation of nanopores on the laser-irradiated graphene that augmented the surface of interactions, improved bonding on the characters, and opened new studies, properties, and applications.

In addition, they controlled thin graphene layers without any modification generated by wrapped nanoarchitecture with new and particular electromagnetic and quantum properties. In this regard, it is essential to underline the relation between the electronic confinement and topological spaces for tuning electronic interactions. For example, strain engineering of giant pseudo-magnetic fields, valley polarization, and topological channels was developed in graphene within the nanoscale, influencing electromagnetic properties and quantum energy levels (**Figure 9**) [62]. The wrapped 3D-modified graphene structure was obtained by accurately depositing Pd nanocrystals (NCs). The deformation of the deposited graphene modified the electronic strains, leading to energy variation within pseudo-electromagnetic fields (Please check Figure 9, about the sensed variations within Landau and Fermi levels of energies and electronic conductions).

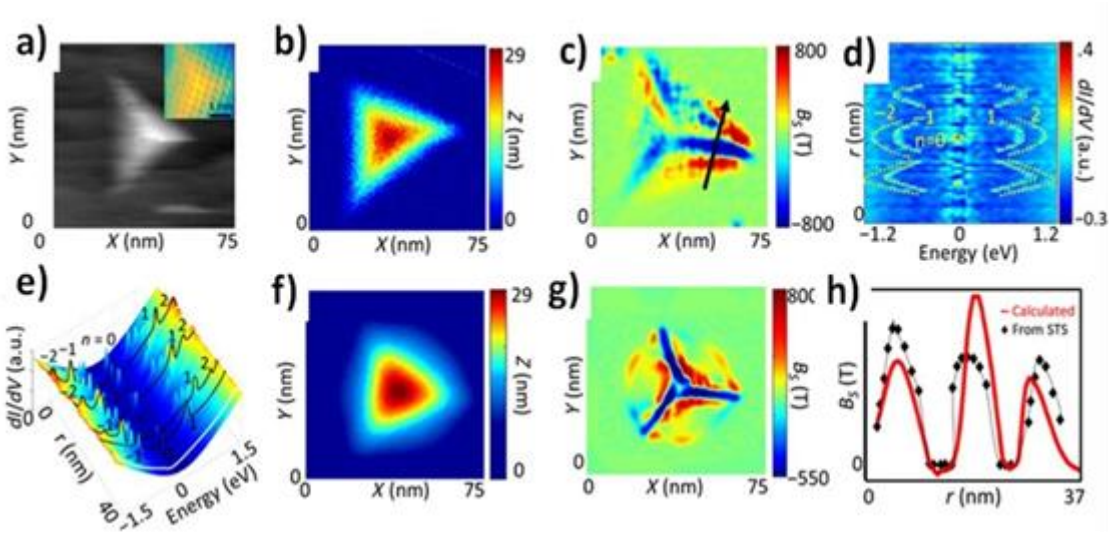


**Figure 9** Topographic and spectroscopic studies of strain-induced effects on graphene at room temperature due to one Pd tetrahedron NC. a) 3D topographic images of the distorted graphene taken by AFM (central panel) and STM (inset, zoom-in image with atomic resolution). b) 3D topographic image of the distorted graphene taken by STM. c) Pseudo-magnetic field map calculated from the topography over the same area as shown in (b). d) Tunneling conductance spectral difference relative to the Dirac spectrum of strain-free graphene shown along the line cut indicated by the black arrow in (c), revealing spatially varying strengths of strain-induced pseudo-magnetic fields as found by the variations in the Landau-level separation. a.u., arbitrary units. e) Representative spectra of tunneling conductance versus energy of strained graphene along the black line cut in (c), showing quantized conductance peaks in strained regions and V-shape Dirac spectrum in strain-free regions, as exemplified by the white curve located at $r$ ~ 36 nm. f) 3D topographic map of graphene/h-BN deformation on an ideal tetrahedron, as computed from MD simulations described in the Supplementary Materials. g) Pseudo-magnetic field map computed from the topographic distortion in (f). h) Comparison of the absolute values of pseudo-magnetic fields $|B_S(r)|$ derived from topographic studies (red line) and Landau-level separations in STS (black diamonds), showing overall satisfactory agreement. Here, $r$ denotes the distance measured from the lower-left end to the upper-left end of the black arrow shown in (c). Reprinted with permissions from N. C. Yeh et al. 2020 [62], Science Advances, Sci.

Regarding the graphene interactions between inter-layers manipulating van der Waals heterostructures, it should be noted that electron-electron interactions [63] could produce strain in graphene lattices by the strain-induced and scalar gauge potentials [64]. These modified gauge potentials resulted from the differences in the distances or angles between the electronic orbitals because structural distortions generated other quantum phenomena, such as the scattering of Dirac fermions and modifications in the local charge densities. In contrast, the excess of gauge potential led to pseudo-magnetic fields.

Therefore, variable electromagnetic solid fields could be tuned by accurately controlling the graphene layer interactions, such as those more prominent than 300 Tesla in graphene nanobubbles [65]. Finally, the capability of these 3D structures to generate particular properties was noted, such

as i) a high electro-active response at different levels, ii) an anomalous quantum hall effect shown by this pseudo-electromagnetic coupling, iii) topological zero modes in molecular graphene assemblies; and iv) topological valley transports and across graphene wall domains [66]. In addition, potential coupling with other molecules and optical active materials was not so broadly studied. Thus, there is still a huge potential for further studies on interactions by conjugation with other materials.

Moreover, focusing on more simple ways to conjugate graphene derivatives and other carbon allotropes, it should be mentioned and highlighted that producing thin multi-layered graphene sheets on compatible substrates is possible. To achieve that, the manipulation of these materials within colloidal dispersions should be taken into account. So, an automated one-drop assembly for facile 2D film deposition was recently developed [67]. A new 2D strategy for the automated manufacturing of high-quality thin films was demonstrated. The methodology is relatively simple and consists of using a computerized pipette, controlling diluted concentrations, adding drops under continuous, homogeneous stirring onto a substrate heated on a hotplate, and controlled convection. In this manner, and slowly eliminating the solvent, the interactions to form multi-layered assemblies of Ti O nanosheets with varied materials, such as metal oxides, graphene oxide, hexagonal boron nitride, etc., were achieved in a few minutes.

## 5. Concluding Remarks and Future Perspectives

The presented research works highlighted that mainly graphene, carbon nanotubes, fullerenes, and derivatives are the main carbon allotropes involved in chemical modifications for targeted fundamental studies and applications. In all developments, well-known methodologies, modified ones, and new strategies were proposed. Thus, chemical modifications were achieved by solid oxidations in highly acidic conditions to obtain carboxylic acid groups or by controlled conditions to get aldehyde groups; then, a versatile point of covalent linking by relatively simple organic reactions was opened. These critical organic groups allowed for modifying the stable and non-reactive carbon-based slides. Different molecular linkers and other organic groups could be added in this manner.

Moreover, the control of the surface chemistry led to attractive interactions with polymeric chains that augmented even more the possibilities to modify chemically the carbon-based structures. In this context, it is noted that, in the case of the use of polymers, it is possible to alter thin slides or surfaces as well as to build further bottom-ups in 3D within the nanoscale. The use of polymers increased drastically the additional synthetic pathways to modify their characters.

In addition, and by highlighting some perspectives, it can be mentioned that the path to entirely modify the chemistry of carbon-based materials, such as graphene and its derivatives, has been paved according to these strategies. It results in a nanomaterial based on a stable carbon-based template with interesting conductive, electronics and emission properties already studied. The interest in incorporating biomolecules, such as amino acids, proteins, carbohydrates, and genomic materials, and looking for developments within life sciences has been noted. The study of Quantum Biology and coupled phenomena within varied, bigger biostructures, such as unicellular microorganisms and related structures containing photo-active receptors, has already been started. This way, the potential participation of carbon-based nanostructures as electron shuttles, electronic conductions, and non-classical light emissions is highlighted.

In addition, the particular properties based on their highly dense electronic conjugation and related pseudo-electromagnetic fields that allowed additional ways of non-covalent interactions and the generation of pseudo-electromagnetic fields with consequent new physical and chemical properties were discussed. These last characteristics are still being studied and incorporated in multiple setups and devices to develop and improve functional materials. Thus, the chemical modification by wet chemical methods, as well as other technological-based methodologies using lasers, modified tips, microscopy techniques, and further analytical instrumentation to tune 3D slides joining similar or completely different materials just by controlling the chemistry of contact and by physical manipulation, such as mentioned before for the graphene slide wrapping, are worth noticing.

Finally, it is mentioned that recent material trends showed varied, exciting insights into where carbon-based nanomaterials could be incorporated. These perspectives showed multidisciplinary aspects that should be mentioned to stimulate researchers toward new prototypes of functional materials. Thus, for example, emphasis was given to varied research fields, such as innovations in synthesizing graphene nanostructures to develop high optoelectronic sensitivity against molecular sensing, electronic conductions, and quantum interferences [68]. However, not only were highly technological perspectives considered, but also high-impact perspectives within Nanomedicine, Pharmaceutics, and uses in Bioassays with promising photocatalytic and antimicrobial activities, from novel conjugated molecules to highly conjugated carbon-based nanoplatforms [69]. In this context, the incorporation of different materials should be considered, as mentioned previously; however, the combination of short molecular spacer with polar, electro-active, and biocompatible properties is highlighted. Thus, combining other inorganic materials and particular properties, such as magnetics, opens multi-functional perspectives in addition to classical optoelectronics [70]. In addition, emphasis was given to the incorporation of new, completely synthetic nanomaterials, such as quantum dots in silico as new nano-laser dyes [71], as well as to the use of biomaterials available in nature, such as cellulose joined to synthetic polymers as poly acrylic [72]. In this manner, the previous properties of carbon-based materials and allotropes should be considered for the conjugation of the different materials. So, due to the importance of the simple and complex chemistry involved, defined by the robust stability of high conjugation, this paper was intended to provide an overview of many strategies, considering the combination with other types of materials.

## Acknowledgments

We are particularly grateful to the University of Akron, Institute of Polymer Science and Engineering, NASA Astrobiology Institute, Ohio, United States, and to the Département de Chimie and Centre d'Optique, Photonique et Laser, Québec, Canada. Moreover, we would like to acknowledge the different grants and funds that have allowed us to do research within the field of Nanomaterials Science. In particular, we would like to thank SECyT (Secretary of Science and Technology from the National University of Cordoba, UNC, Argentina) for the completion of payments of the Grant for Young Researchers awarded to AGB at INFIQC, Dep. of Organic Chemistry, Faculty of Chemical Sciences, UNC, Argentina.

Finally, it is specially acknowledged to the Journal "Recent Progress in Materials" for this opportunity to AGB as Invited Guest Editor in the Special Issue entitled: "Hybrid Graphene based

Materials: Synthesis, Characterization, Properties, and Applications" at Editorial LIDSEN Publishing Inc., Open Access Publisher (USA).

**Author Contributions**

The author did all the research work of this study.

**Competing Interests**

The author has declared that no competing interests exist.

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