

Hybrid nanodiamond nanoadditives in polymers—Contemporary marks and fruitions

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Abstract: The current manuscript overviews the potential of inimitable zero dimensional carbon nanoentities, i.e., nanodiamonds, in the form of hybrid nanostructures with allied nanocarbons such as graphene and carbon nanotube. Accordingly, two major categories of hybrid nanodiamond nanoadditives have been examined for nanocompositing, including nanodiamond-graphene or nanodiamond/graphene oxide and nanodiamond/carbon nanotubes. These exceptional nanodiamond derived bifunctional nanocarbon nanostructures depicted valuable structural and physical attributes (morphology, electrical, mechanical, thermal, etc.) owing to the combination of intrinsic features of nanodiamonds with other nanocarbons. Consequently, as per literature reported so far, noteworthy multifunctional hybrid nanodiamond-graphene, nanodiamond/graphene oxide, and nanodiamond/carbon nanotube nanoadditives have been argued for characteristics and potential advantages. Particularly, these nanodiamond derived hybrid nanoparticles based nanomaterials seem deployable in the fields of electromagnetic radiation shielding, electronic devices like field effect transistors, energy storing maneuvers namely supercapacitors, and biomedical utilizations for wound healing, tissue engineering, biosensing, etc. Nonetheless, restricted research traced up till now on hybrid nanodiamond-graphene and nanodiamond/carbon nanotube based nanocomposites, therefore, future research appears necessary for further precise design varieties, large scale processing, and advanced technological progresses.

Keywords: nanodiamond; hybrid nanoadditives; graphene; carbon nanotube; properties; radiation shielding; electronics; energy devices

1. Introduction

Advancements in carbon based nanotechnologies led to the discovery of numerous exceptional nanocarbon forms having different dimensionalities, like nanodiamonds, graphene, carbon nanotubes, fullerene, and many more [1,2]. Out of these, nanodiamonds attained a unique technical stance owing to nanosizes, of 2–8 nm, high aspect ratio, mechanical robustness (hardness/stiffness), heat stability, and allied beneficial physical/chemical properties [3]. These unique nanocarbon nanoentities were discovered in 1960's, and later studies focused their mass production, structural and physical details [4,5]. Among prominent applications, nanodiamonds seemed to be worthwhile for lubricants, coatings, sensors/biosensors, tissue scaffolding, bioimplants, and drug delivery systems [6–8]. The surface defects and functionalities (hydroxyl, carbonyl, carboxylic acid, etc.) on nanodiamond surfaces, induced during synthesis or modification routes, further enhanced the structural features of these nanoparticles [9,10]. Consequently, owing to zero dimensionality and surface functional characters, nanodiamond has been reported technically beneficial in the literature, relative to other nanocarbons [11,12]. Furthermore, nanodiamonds revealed fine tendencies to form nanocomposites with

organic or inorganic materials [13]. Herin, synergistic effects between the matrix and nanodiamond nanoparticles were detected responsible for enhancing the characteristics of ensuing nanocomposites [14]. Similarly, an important aspect of nanodiamond nanoparticles has been examined as the formation of hybrid nanostructures with other nanocarbons and their subsequent use as nanofillers [15,16]. Consequent, hybrid nanodiamond nanoadditives have been noticed as a new and limitedly explored research area, demanding comprehensive future investigations in possible technological directions.

This state-of-the-art review article, principally, highlights the potency of nanodiamond nanoparticles to form hybrid nanodiamond nanostructures with graphene and carbon nanotubes. The resulting nanodiamond-graphene or nanodiamond/graphene oxide and nanodiamond/carbon nanotubes were further applicable as nanoadditives for the formation of nanocomposites. Unique combinations of nanodiamonds with allied nanocarbons in turn depicted sole morphological, electrical, optical, thermal, mechanical, and related physical property profiles. Subsequently, according to literature up till now, hybrid nanodiamond nanoadditives based nanomaterials had momentous applications for the fields of electromagnetic radiation defense, electronics/energy devices, and biomedical arenas. Noticeably, design limitations, processing, compatibility, and dispersion challenges need to be resolved for future high tech hybrid nanodiamond nanostructures aiming commercial level deployments. As per our analysis so far, it is important to mention here, this manuscript is altogether an original attempt to review the contemporary state of hybrid nanodiamond nanoadditives derived nanomaterials; therefore, it will be definitely helpful for field scientists struggling for potential opportunities of multifunctional nanocarbon hybrid nanostructures.

2. Nanodiamonds

Nanodiamonds are spherical carbon nanostructures having very small sizes, even down to 2–8 nm in diameters [17]. Distinguished features of nanodiamonds can be listed as high surface area, surface functionalities, crystallinity, hardness, non-toxicity, and structural/chemical stabilities [18]. Looking into the structure of nanodiamonds, a layered nanostructure can be perceived having inner most diamond core, middle layer of sp^2 hybridized carbon atoms, and outer carbon shell of sp^2 and sp^3 hybrid atoms, as illustrated in **Figure 1**. Variety of synthesis techniques have been outlined in the literature for nanodiamonds, namely chemical vapor deposition [19], laser/plasma techniques [20], and the most popular one, i.e., detonation method [21]. Detonation technique, employing an oxygen deficient explosive 2-methyl-1,3,5-trinitrobenzene, has been widely practiced for bulk nanodiamond synthesis having sizes, of 5 nm [22].

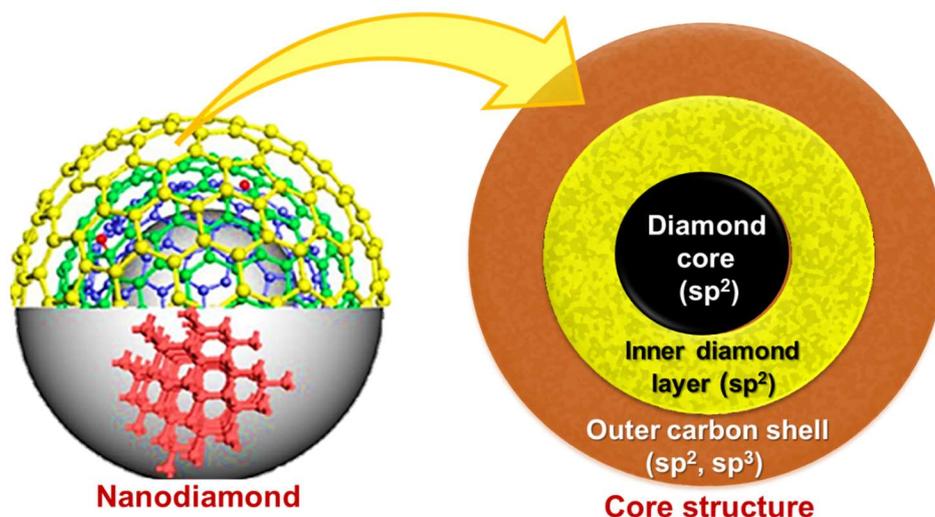


Figure 1. Basic nanodiamond nanostructure.

This process usually involves extreme temperature and pressure conditions for synthesis and collection of resulting nanodiamonds from the detonation soot. Myriad of application areas of nanodiamonds have been reported ranging from nanocomposites to sophisticated devices, vehicles, and medical sides [23–25]. Further inclusion of nanodiamond nanoparticles in organic/inorganic matrices seem to enhance the adhesion and tribological features of the resulting nanomaterials [26,27]. Facile fabrication methods, such as solution/melt techniques and in situ methods, have been studied for the synthesis of nanodiamond nanocomposites [28,29]. Accordingly, nanodiamonds have been reinforced in thermosetting as well as thermoplastic matrices for resultant property and performance evaluations [30]. Today’s research on nanodiamonds seems trending towards the design of hybrid nanodiamond nanostructures, like nanodiamond-carbon nanotube, nanodiamond-graphene, nanodiamond-fullerene, etc., as presented in **Figure 2** [31–33].

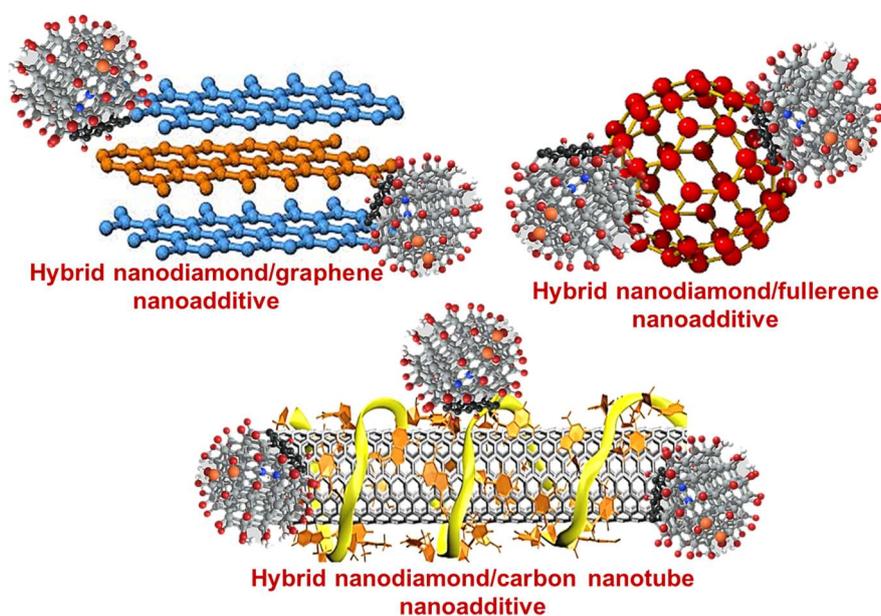


Figure 2. Hybrid nanodiamond nanoadditives with graphene, fullerene and carbon nanotubes.

3. Hybrid nanodiamond nanoadditives filled polymeric nanocomposites

3.1. Nanodiamond-graphene hybrid nanoadditives

For nanocompositing, carbon nanoadditives have attained noteworthy research curiosities to attain high performance nanocomposites for technical purposes [34]. In this concern, innumerable combinations of carbon nanoadditives and polymeric matrices have been scrutinized towards high end applications in high strength/heat defiance engineering structures, aerospace/defense, solar cells, fuel cells, capacitors/batteries, civil, environmental membranes, and biomedical pastures, to name a few [35].

Graphene has been ranked as one of the most peerless discoveries in the world of carbon nanostructures [36]. Initially reported by Novoselov and Geim (2004), graphene has intrinsic zero dimensional one atom thick carbon nanostructure with sp^2 hybridized carbons in hexagonal arrangements [37]. It can be imagined as a single layer out of a stacked graphite structure, which is further prone to surface functionalization (**Figure 3**).

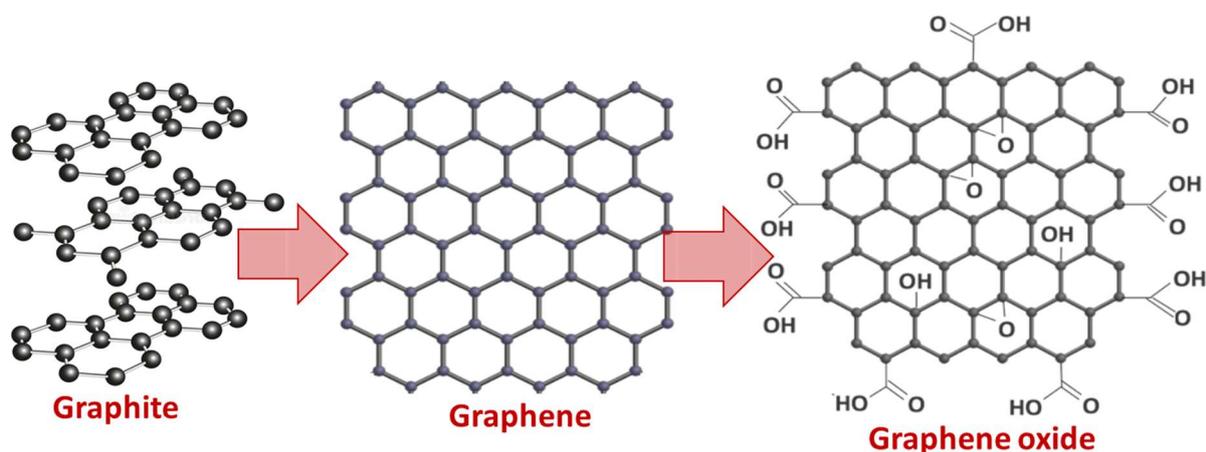


Figure 3. Graphite to graphene and graphene oxide.

Since discovery, numerous efficient methods have been practiced for graphene synthesis, namely mechanical/chemical exfoliation, chemical vapor deposition, hydrothermal, laser ablation, arc discharge, plasma tactics, lab scale organic synthesis, so on and so forth [38]. Few of the worth mentioning properties of graphene, include optical transparency, electron conductivity ($\sim 200,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$), thermal conductivity ($\sim 5000 \text{ W/mK}$), Young's modulus ($\sim 1 \text{ TPa}$), and all that. Owing to inherent wrinkling tendencies, graphene has been surface modified to introduce functional groups, like hydroxyl, carbonyl, carboxylic acid, ketone, and more [39]. For this purpose, Hummer's route and Brodie methods have been commonly used for the conversion of graphene to functionalized graphene, commonly named as graphene oxide [40]. As per literature, graphene oxide reveal superior electron/thermal conduction and mechanical stability, relative to neat graphene nanosheets [41]. As an important application, graphene and graphene oxide have been used as efficient nanofillers to form high tech polymeric nanocomposites

aiming robust space structures, automotive parts, defense materials, energy/electronic devices, textiles, drug delivery and tissue scaffolds, to name a few [42–44].

Hybrid nanodiamond nanoadditives of graphene and graphene oxide have been successfully designed and documented [45,46]. As per literature, hybrid nanodiamond-graphene nanoparticles depicted sizes of 4–5 nm and 5–9 times superior surface area ($49.573 \text{ m}^2 \text{ g}^{-1}$), relative to neat graphene oxide [47]. Consequently, such hybrid nanodiamond-graphene oxide nanoparticles were suggested for applications, like dehydrogenation and degradation of ethylbenzene [48]. According, research endeavors also focused on introducing these graphene based hybrid nanodiamond nanoadditives in matrices for designing multifunctional composites, nanocomposites, and nanomaterials [49–51]. Herein, literature reports were detected on detonated nanodiamond-graphene oxide nanomaterials hybrid nanodiamond-graphene nanoparticles [52,53]. As effective nanocomposite matrices for nanodiamonds, block copolymers of polystyrene, poly (vinyl butyral-*co*-vinyl alcohol-*co*-vinyl acetate), etc. have been investigated [54,55]. In this regard, Kausar [56] fabricated blends of poly (vinyl butyral-*co*-vinyl alcohol-*co*-vinyl acetate) and polystyrene matrices. Using graphene oxide obtained by Hummer's method, hybrid nanodiamond-graphene oxide nanoparticles were formed by simple solution sonication route. Afterwards, the as prepared hybrid nanodiamond-graphene oxide nanoparticles were introduced to the poly (vinyl butyral-*co*-vinyl alcohol-*co*-vinyl acetate)/polystyrene blend via solution technique. For these nanocomposites, hybrid nanodiamond-graphene oxide based nanocomposite had higher tensile strength, of 32–35 MPa, relative to pristine graphene oxide based system (22–28 MPa). Adding 10 wt.% hybrid nanodiamond-graphene oxide nanoparticles to the blend matrix also depicted 10% thermal degradation temperature, of 532 °C–554 °C, and electrical conductivity, of 2.5 Scm^{-1} , as compared with the pristine graphene oxide system (i.e., 489–522 °C and 1.3 Scm^{-1} , respectively). Wang et al. [57] proposed a design of hybrid nanodiamond-reduced graphene oxide nanoparticles using simple solution/heating methods. Initially, graphene oxide was formed by Hummer's method and the converted to reduced graphene oxide along with the surface attachment of nanodiamond nanoparticles (**Figure 4A**). **Figures 4B–D** show high resolution transmission electron microscopy and scanning electron microscopy micrographs of hybrid nanodiamond-reduced graphene oxide nanoparticles. Tiny nanodiamond nanoparticles can be seen scattered on the surface of reduce graphene oxide nanosheets.

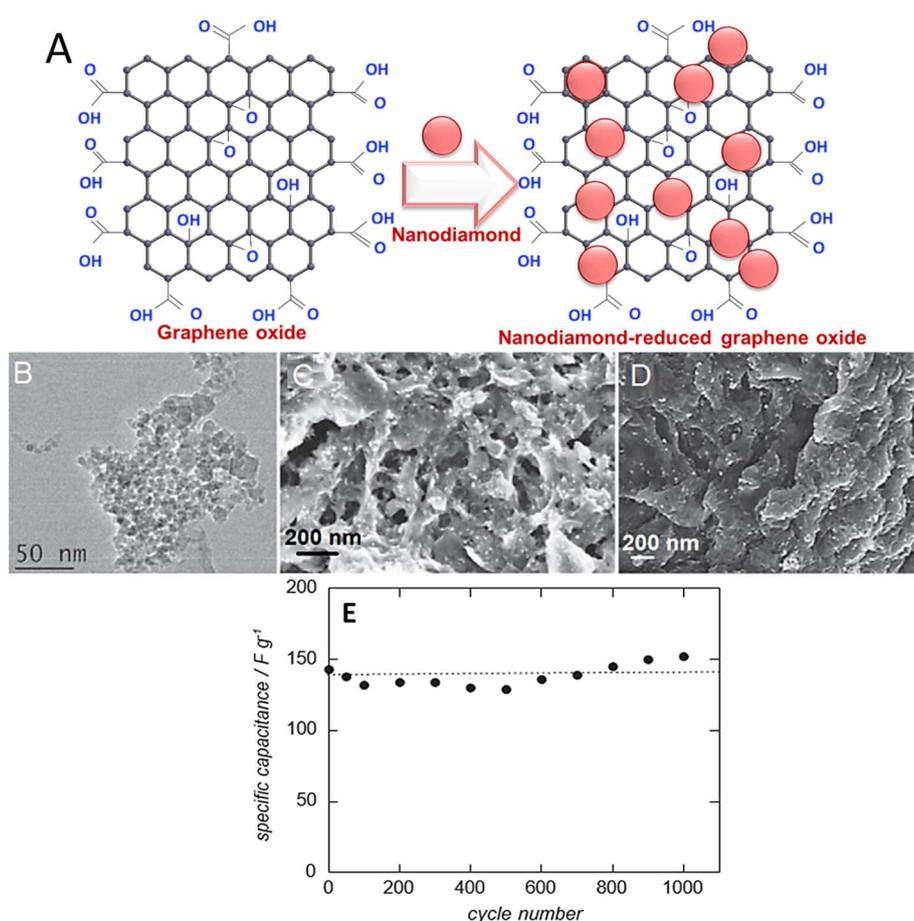


Figure 4. (A) Synthesis route to hybrid nanodiamond-reduced graphene oxide nanoparticles; (B) high-resolution transmission electron microscopy (HRTEM) image of hydroxyl functional nanodiamond (ND-OH); (C and D) scanning electron microscopy (SEM) images of nanodiamond-reduced graphene oxide (ND-rGO) matrices; (E) specific capacitance of nanodiamond-reduced graphene oxide (ND-rGO) (10:1) vs. cycle number (2 A g⁻¹) [57]. Reproduced with permission from Elsevier.

Moreover, **Figure 4E** presents specific capacitance vs. cyclic number plot of hybrid nanodiamond-reduced graphene oxide. Accordingly, these hybrids had optimum specific capacitance of $\sim 186 \text{ F g}^{-1}$ over 1000 cycles at current density of 2 A g⁻¹. This performance was attributed to the formation of compatible electron transport phases between the two nanocarbon nanostructures. Consequently, the hybrid nanodiamond-reduced graphene oxide hybrid designed via facile routes were suggested for next generation supercapacitor electrode materials. Furthermore, Yu et al. [58] used solution approach for the fabrication of hybrid nanodiamond-functional graphene nanoparticles filled poly (vinylidene fluoride) nanocomposites. Adding 45 wt.% hybrid nanodiamond nanoadditives revealed thermal conductivity, of around 0.66 W/mK, i.e., twice higher than unfilled poly (vinylidene fluoride) [59]. Moreover, hybrid poly (vinylidene fluoride)/nanodiamond-functional graphene nanocomposite with 10 wt.% nanofiller had superior electrical conductivity ($7.1 \times 10^{-7} \text{ S cm}^{-1}$), than the unfilled polymer ($5.1 \times 10^{-15} \text{ S cm}^{-1}$). Similar research extensions were noticed on valuable application of hybrid nanodiamond-graphene and hybrid nanodiamond-graphene oxide nanoadditives for high performance nanocomposites [60–62]. Besides, theoretical studies on hybrid nanodiamond-

graphene nanoparticles were also performed. For example, De Vita and group [63] investigated the structure of nanodiamond-graphite via atomistic modeling method. **Figure 5A** presents nanodiamond-graphite interfaces formed while graphitization process on nanodiamond surfaces. Moreover, electronic states localization between nanodiamond and graphite phases were explored via isovalue surfaces based electron density method, as given in **Figure 5B**. According to results, graphitization processes seem to occur at extremely high temperatures and thermal fluctuations were suggested for changes in the surface binding energies of nanodiamond-graphite nanostructures criterion [64]. In this way, successful experimental and theoretical researches were observed on nanodiamond-graphene nanostructures, therefore further predicting applications towards high performance engineering nanostructures, supercapacitors, field emission diodes, and allied high tech applications [65–67].

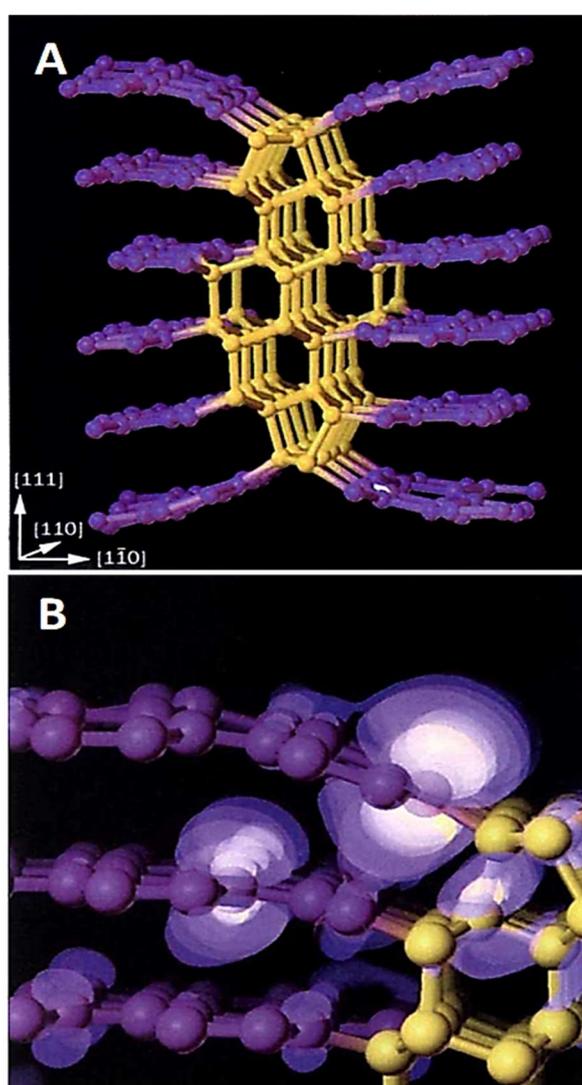


Figure 5. (A) Atomistic model of a diamond-graphite interface formed during graphitization of diamond surface; (B) Isovalue surfaces of electron density showing an electronic state localized at interface between diamond and graphite phases [63]. Reproduced with permission from Nature.

3.2. Nanodiamond-carbon nanotube hybrid nanoadditives

Carbon nanotubes, initially discovered in 1990s, are hollow cylindrical carbon nanostructures made up of hexagonally aligned sp^2 hybrid carbon atoms [68]. Depending upon the number of overlapping cylinders, carbon nanotubes can be single walled, double walled, or multi walled nanostructures [69–71]. Single walled carbon nanotube usually own smaller diameters, of 1–2 nm, while multi walled carbon nanotubes can have larger diameters, of up to 150 nm [72]. Amid prominent features, carbon nanotubes seem to display high surface area, electronic, optical, electrical, magnetic, mechanical, and thermal properties [73,74]. Consequently, carbon nanotubes have been effectively used as nanofillers for polymers to form high performing nanocomposites [75]. Applications of carbon nanotube can be itemized for sensors, capacitors, batteries, solar/fuel cells, aerospace/automotive, defence/engineering, textiles, biomedical, and further technical fields [76–78].

Into the bargain, previous as well as few recent research attempts have been noticed regarding hybrid nanodiamond-carbon nanotube nanoadditives [79]. For example, Chatterjee et al. [80] reported on epoxy nanocomposites with hybrid nanodiamond-carbon nanotube nanoadditives via solution technique and studied the physical properties. Farooq et al. [81] also premeditated the epoxy/hybrid nanodiamond-carbon nanotube nanomaterials obtained by solution technique. Here, 0.1 wt.% hybrid nanofiller remarkably enhanced the tensile strength of epoxy matrix, from 895 to 1187 MPa. These results were credited to nanofiller dispersion, matrix-nanofiller interactions, and so efficient load transfer properties through the matrix [82]. Khabashesku et al. [83] studied covalently linked hybrid nanodiamond-carbon nanotube nanostructures using urea/ethylene diamine based linkers. The resulting hybrid nanodiamond-carbon nanotube nanoadditives were studied for nanocompositing with polyurea/polyurethane matrices using solution method. According to scanning electron, transmission electron, and atomic force microscopy analysis, polyurea/polyurethane/nanodiamond-carbon nanotube nanocomposite had pearl-necklace morphology. In addition, including 0.2 wt.% nanofiller significantly enhanced the tensile strength, by 64%, relative to the unfilled polymers, due to matrix-nanofiller compatibility. Fedoseeva et al. [84] studied hybrid nanodiamond-carbon nanotube nanostructures prepared by simple dispersion and sonication technique in dimethyl sulfoxide and water mixture. Accordingly, **Figures 6A,B** show transmission electron microscopy images of nanodiamond-carbon nanotube nanostructures with carbon nanotubes having diameters of 50 nm and nanodiamonds of 5 nm. However, mostly nanodiamonds were observed in the form of small aggregated, of 30 nm and larger aggregates of 200 nm, between the carbon nanotube domains.

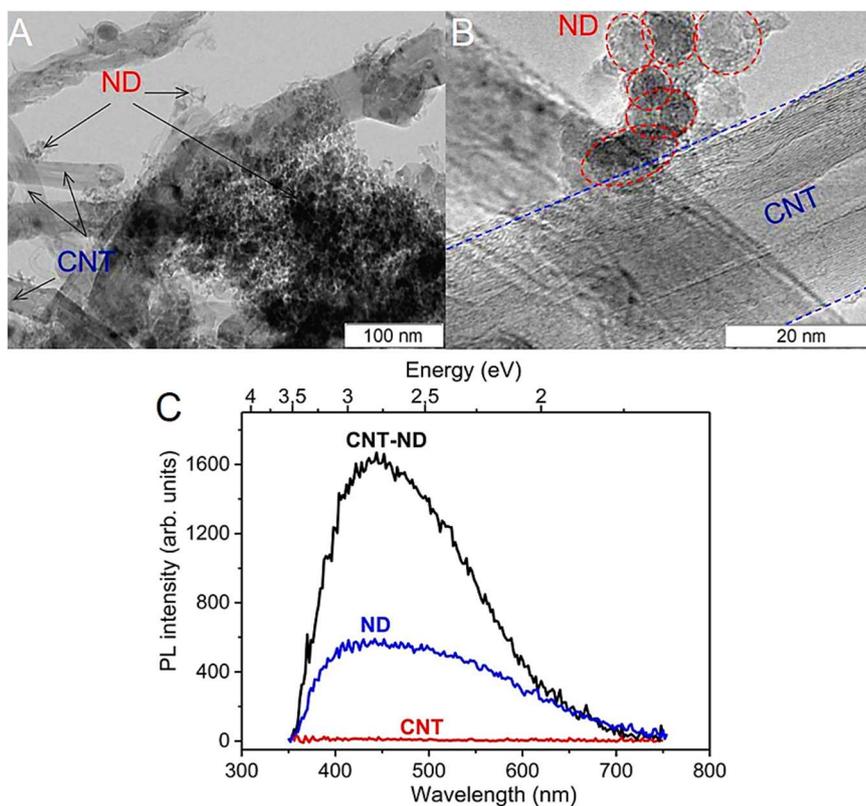


Figure 6. (A and B) Transmission electron microscopy images of carbon nanotube-nanodiamond (CNT-ND) hybrid nanomaterial; (C) photoluminescence spectra of CNT-ND hybrid material, carbon nanotube (CNT) and nanodiamond (ND) [84]. Reproduced with permission from Nature.

Figure 6C shows photoluminescence spectra for pristine nanodiamond, carbon nanotube, and hybrid nanodiamond-carbon nanotube nanostructure. Owing to metallic characters, carbon nanotube had no peak in 300–800 nm. Similarly, intrinsic nature of nanodiamonds depicted their characteristic blue-green photoluminescence peak between 400 to 600 nm. On the other hand, hybrid nanodiamond-carbon nanotube nanostructure depicted a broad peak ranging from 380 to 700 nm, having an intense blue photoluminescence peak at 440 nm due to the presence of nanodiamonds. However, photoluminescence peak intensity of hybrid can be seen double than pristine nanodiamond, probably due to interlinking of nanodiamond-carbon nanotube via carboxyl and hydroxyl group condensations, and so enhance the overall fluorescence properties. Hence, the literature well reported the microstructural and structural confirmations of hybrid nanodiamond-carbon nanotube nanostructures.

4. Technical significance of hybrid nanodiamond nanoadditives based nanocomposites

Among technological applications, hybrid nanodiamond-nanocarbon nanomaterials seemed valuable for electromagnetic interfering radiation protection, similar to pristine nanodiamonds, due to high magnetic permeability [85]. To safeguard the electronics/energy devices, living beings, and environment from hazardous electromagnetic radiations, polymeric nanocomposites based shields have

been investigated in the literature [86]. Particularly, polymeric nanomaterials with carbon nanoadditives, like carbon nanotube, graphene, nanodiamond, etc., had superior electrical conductivity and magnetic permittivity for electromagnetic interference radiation shielding [87,88]. In this concern, radiation shielding performances of the polymeric nanomaterials have been analyzed for adding nanocarbon contents as well as mechanisms (reflections, multi reflections, and internal reflections) involved in radiation protection [89]. Nanodiamond or onion like carbon nanospheres have been reported for worthwhile surface properties, electron conduction, and electromagnetic absorption features [90]. For instance, poly(dimethyl sulfoxide)/onion like carbon nanospheres nanocomposites have been designed and studied for electromagnetic interference shielding [91]. Including 2–4 wt.% nanofiller seemed to enhance the dielectric constant, electrical conductivity, dielectric permittivity, and overall electromagnetic shielding properties of these nanomaterials, owing to fine dispersion and matrix-nanofiller compatibility effects. Song et al. [92] examined polydopamine/nanodiamond-hollow carbon nanosphere nanocomposites for electromagnetic interference shielding application. Herein, **Figure 7A** shows transmission electron microscopy images of nanodiamond-hollow carbon nanosphere nanoparticles. As per micrographs, hollow carbon nanosphere visibly developed an inner core layer of 11 nm in the hybrid nanoparticle nanostructure, whereas nanodiamonds formed an outer layer of 17 nm. To analyze the electromagnetic interference shielding, **Figure 7B** shows plots of radiation shielding vs. changing thickness and frequencies. As per results, it appears that 5 wt.% hybrid nanodiamond nanostructure loading caused reasonable reflection loss of about -47.1 dB at 2.15 mm thickness. In addition, radiation absorption performance seemed to be connected to the variations in the shield thicknesses. Such high end electromagnetic shielding performance of hybrid nanodiamond nanostructure was attributed to the outstanding matrix-nanofiller compatibility as well as formation of multiple interlinked interfaces for better radiation absorption characters.

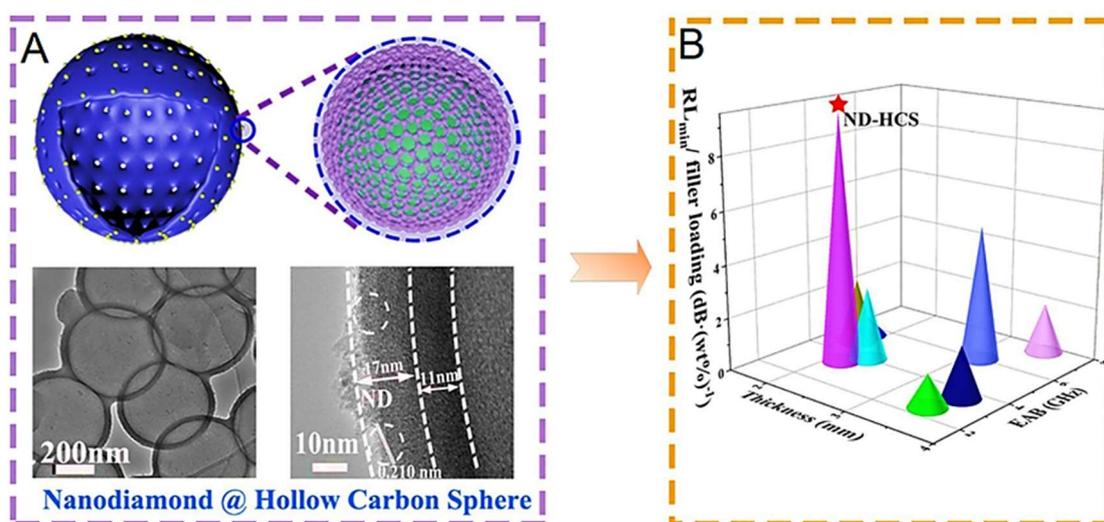


Figure 7. (A) Transmission electron microscopy of nanodiamond@hollow carbon sphere (ND-HCS); (B) plots of radiation shielding vs. changing thickness and frequencies for ND-HCS nanocomposite [92]. Reproduced with permission from Elsevier.

Another important utilization of nanodiamond derived hybrids can be seen for semiconducting electronic devices [93]. For example, transistor is a common semiconducting device which can boost power/electrical signals of the electronic devices [94]. Pristine nanodiamonds have been explored for field effect transistor applications [95]. For instance, nanodiamond and silicon based ion sensitive field effect transistors have been reported [96]. Such transistors had high responses near the Nernst limit. Hsu et al. [97] reported on nanodiamond based field effect transistor. They reported superior emission current, DC voltage gain, and low gate turn-on voltage of 160 μA , 1000 and 25 V, respectively. Similarly, nanodiamond hybrids have been applied in the field effect transistors [98]. Since, pristine graphene has a drawback of zero bandgap, therefore nanodiamonds-graphene hybrids have been designed for effective electronic devices applications. After hybrid formation, beneficial band gap properties can be induced to support the desirable carrier transport features. Zhao et al. [99] comparatively investigated the hydrogen terminated monolayer nanodiamond and hydrogen terminated single crystal diamond and subsequent hybrid graphene-hydrogen terminated monolayer nanodiamond and graphene-hydrogen terminated single crystal diamond nanomaterials. Ultimately, the hybrid nanodiamond-graphene nanomaterial was explored for field effect transistor application. Accordingly, **Figure 8a** shows Van der Pauw based Hall effect analysis for Hall mobility, properties of hybrid graphene-hydrogen terminated monolayer nanodiamond and graphene-hydrogen terminated single crystal diamond nanostructures. The results revealed superior carrier transport and Hall effects of hybrid hydrogenated nanodiamond-graphene, relative to that of single crystal diamond. Similarly, **Figure 8b** depicts variations in carrier density and sheet resistivity analysis vs. temperature of the samples using same method. Herein, carrier density of hydrogenated nanodiamond-graphene was higher than single crystal diamond-graphene, probably due to better interactions and alignment between the hydrogenated nanodiamond nanoparticles and graphene. The sheet resistance of both the samples were almost comparable with in similar temperature ranges. **Figure 8c** shows behavior of hybrid hydrogenated nanodiamond-graphene for contact with water and carbon dioxide in air. Owing to hydrogen termination of nanodiamonds, a hole layer was developed on the surface, which was responsible for the superior Hall properties. **Figure 8d** reveals a surface structure of hybrid hydrogenated nanodiamond-graphene with hydrogen termination. Due to electron transfer from graphene to nanodiamond surface, holes and p-type characters were observed for hybrid hydrogenated nanodiamond-graphene accountable for Hall effect for high end field effect transistor application. Further comprehensive attempts are need to design high end nanodiamond based field effect transistors.

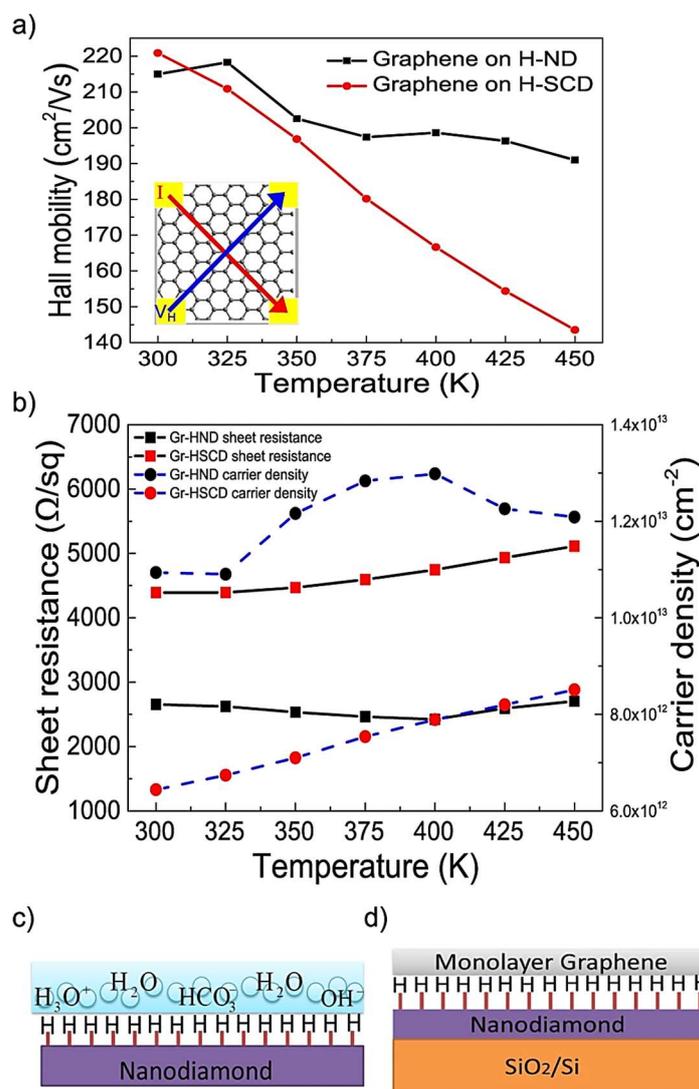


Figure 8. Hall properties of GrHND and GrHSCD: **(a)** Hall mobility for GrHND and GrHSCD, inset is Van der Pauw method for measurement; **(b)** comparison of sheet resistance and carrier density of GrHND and GrHSCD; **(c)** schematic picture of the hydrogenated ND surface in contact with a water layer as it forms in air; **(d)** schematic for GrHND heterostructure [99]. HND = hydrogen terminated monolayer nanodiamond; HSCD = hydrogen terminated single crystal diamond; GrHND = Graphene-hydrogen terminated monolayer nanodiamond; GrHSCD = Graphene-hydrogen terminated single crystal diamond. Reproduced with permission from Nature.

Among energy storage devices, supercapacitors have attained notable place for efficient energy storing/conversions [100]. With nanotechnology progress, innovative nanomaterials have been applied to form high efficiency supercapacitors [101]. In supercapacitors, pristine nanodiamonds have found immense scope [102]. However, using pristine nanodiamonds in supercapacitor electrodes may result in low specific capacitance and cyclic performance [103]. Therefore, nanocomposites of nanodiamond filled conjugated polymers have been designed and studied [104]. Palaniappan et al. [105] prepared polyaniline/nanodiamond nanocomposites for supercapacitor electrodes using emulsion polymerization technique. They found

specific capacitance of the nanocomposite electrode $> 300 \text{ F g}^{-1}$. Kovalenko et al. [106] reported higher values of specific capacitance ($> 600 \text{ F g}^{-1}$) for the designed polyaniline/nanodiamond nanocomposite based electrode. The nanodiamond based nanocomposite electrodes for supercapacitors revealed repeated cyclic performance and long functioning life [107]. Similarly, few nanodiamond hybrids have been advantageously applied for supercapacitor applications [108,109]. For instance, Wang et al. [57] formed hybrid nanodiamond-reduced graphene oxide nanomaterials for supercapacitor electrodes using solution phase method. Consequently, the hybrid nanodiamond-reduced graphene oxide revealed optimal specific capacitance of about 186 F g^{-1} . However, as compared to pristine nanodiamond filled polymers, hybrid nanodiamond based nanocomposites have been less studied for supercapacitor electrodes and so further research needed in this direction.

In biomedical sector, surface chemistry, non-toxicity, and bio-compatibility properties of nanodiamonds and polymer/nanodiamond nanocomposites revealed important potential applications [110]. In this concern, nanodiamond filled biodegradable polymers, poly (vinyl alcohol), poly (ethylene glycol), etc. have been applied for wound healing, antimicrobials, tissue engineering implants (dental, catheters, skin, etc.), and biosensors [111–113]. For example, nanodiamonds filled ecological polymers, like cellulose/chitosan have been used for wound healing [114]. Ostadhosseini et al. [115] filled 2–3 wt.% nanodiamond in chitosan/bacterial cellulose matrix. Due to superior antibacterial activity, these nanomaterials were found efficient for wound healing application. Similarly, biosensing application of nanodiamonds has explored in the literature [116,117]. Qureshi et al. [118] formed biosensor probes based on fluorescent nanodiamonds. The biosensors were found effective for detection and diagnosis of biomolecular species. Likewise, hybrid nanodiamond nanoparticles have been used in the poly (methyl methacrylate) and poly (iso-butyl methacrylate) matrices for biocompatible applications [119,120]. Accordingly, biomedical applications of biocompatible hybrid nanodiamond nanoparticles filled polycarbonate and poly (lactic acid) nanomaterials were reported [121,122]. For example, Yao et al. [123] designed nanodiamond/graphene oxide based nanocomposite for biosensing application. Efficiency of the nanomaterial was attributed to microporous layered nanostructure of nanodiamond-graphene oxide in the hybrid design. Nevertheless, biomedical applications of polymer/nanodiamond-nanocarbon nanocomposite need to be investigated comprehensively in the literature for future commercial scale utilizations.

5. Future of hybrid nanodiamond nanocomposites

Figure 9 shows overall applications areas of hybrid nanodiamond nanostructures discovered so far in the literature surveys. Regarding practical applications, limited attempts were noticed on field effect transistors and supercapacitors based on hybrid nanodiamond nanostructures and so focused studied seemed necessary to define precise structural designs for next generation advance in electronics and device applications. It appears that, under each category of hybrid nanodiamond-graphene and hybrid nanodiamond-carbon nanotube nanostructures, more research efforts must be reported to find further design and applied possibilities

of these remarkable nanomaterials and future progress in this field. Concisely, the research seen up till now is not sufficient for real world applications of hybrid nanodiamond nanostructures and focused research investigations seem indispensable for future developments in radiation defense, electronics, and biomedical fields.

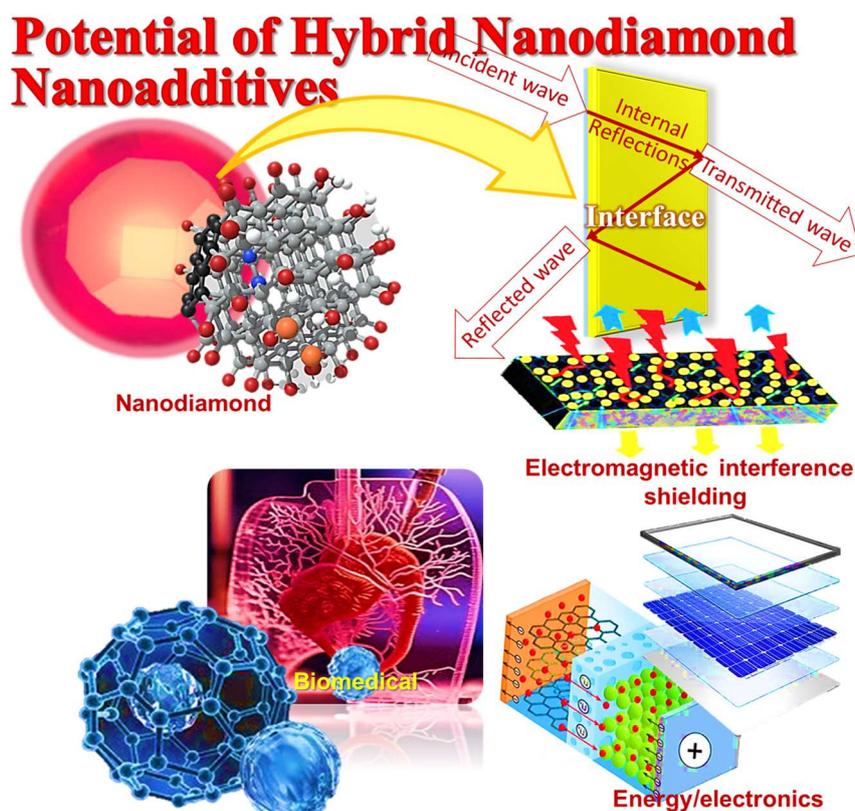


Figure 9. Possible applications of hybrid nanodiamond nanoadditives based nanomaterials.

In future, using hybrid nanodiamond nanofillers in polymers may reveal interesting applications for solar cells, actuators, and similar high end energy and electronic devices, which have not been explored so far. For future industrial applications, efficient synthesis routes, nanodiamond purification, and high quality isolation of nanodiamond nanoparticles need to be focused. In this context, understanding mechanisms and structure-property relationships in hybrid nanodiamond nanocomposites need to be engrossed by the field scientists and researchers. In future, precise control over designs, features, and technical performance of hybrid nanodiamond nanocomposites also seem necessary to discover high-tech applications in unexplored industrial sectors up till now, like space/automotive industries, defense, construction, textiles, microelectronics, and several other uncharted practical fields.

6. Conclusions

This novel article argues the state of hybrid nanodiamond nanoadditives, especially stating hybrid nanodiamond-graphene, nanodiamond/graphene oxide, and nanodiamond/carbon nanotube nanoadditives, according to literature availability. Here, synergistic effects between nanodiamonds and carbon nanoparticles

considerably enhanced the overall physical properties of these nanocomposites, including surface properties, structural varieties, microstructure, electronics, conducting, mechanical/thermal, capacitance, and radiation shielding contour. Ultimately., progress on hybrid nanodiamond nanoadditives led to the formation of high temperature/strength nanocomposites, electromagnetic interference radiation shields, supercapacitors, and tissue engineering applications. Besides the progress seen up to now, hybrid nanodiamond nanoadditives based nanomaterials need to be examined comprehensively in future to uncover the full practical spectrum of these remarkable nanocomposites.

Conflict of interest: The author declares no conflict of interest.

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