

Carbon nano-onions based polymeric nanocomposites: Advancements in designs—to—practical applications

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Abstract: Carbon nano-onions are unique, tiny, multilayered carbonaceous nanoparticles having distinct structural, microstructural, and a range of physical characteristics and high-tech uses. Similar to other carbonaceous nanoparticles (graphene, carbon nanotubes), carbon nano-onions, as well as modified carbon nano-onions, have been considered as valuable nanofillers for macromolecules. Along these lines, the present overview refers to the scientific impact of reinforcing carbon nano-onions in polymeric matrices. For this purpose, this manuscript has been systematically categorized into an initial section addressing fundamentals of carbon nano-onions, followed by a major section regarding design, structure-property, and performance aspects of carbon nano-onion nano-reinforced polymeric matrices (conducting, thermoplastics, thermosets). Subsequent review sections unfold promising application zones, challenges, and the future of polymer/carbon nano-onions hybrids. According to the literature, numerous facile and efficient routes (in situ, electrochemical, solution, solvent casting, curing) have been used for fabricating these nanomaterials. Herein, adding carbon nano-onions was noticed to valuably enhance the morphological, physical, and applied scope of the ensuing nanocomposites. Owing to design diversities, a myriad of technological deployments have been noticed for polymer/carbon nano-onions, including space engineering-to-energy production/storage/sensing devices and catalysis. Nevertheless, delimited systematic scientific studies have been performed so far regarding different types of polymer/carbon nano-onions and their applied specifications. Henceforth, focused and continuous research efforts by the field scientists/engineers seemed to be indispensable for overcoming the underlying challenges to attain next-generation industrial-scale carbon nano-onions derivative nanomaterials.

Keywords: carbon nano-onions; polymers; nano-reinforcement; nano-composites; space; energy devices; sensors; solar cells

1. Introduction

Carbon nano-onions appeared as a unique discovery in the widespread family of carbon nanoparticles, owing to their unique structural, property, and performance attributes [1]. Not unlike other carbon nanoparticles (fullerene, nanodiamond, graphene, carbon nanotube), carbon nano-onions have non-reinforcement capabilities towards inorganic and polymeric matrices [2–4]. In this concern, pristine as well as physically/covalently nanomodified carbon nano-onions have been used as nanofillers for a variety of polymeric matrices [5]. The functionalization processes were noticed to further enhance the dispersion and characteristics of carbon nano-onions towards the macromolecules [6]. Numerous fabrication routes have been employed so far for the synthesis of carbon nano-onions and polymer/carbon nano-onions nanocomposites [7]. Herein, the choice of synthesis technique and adjustment of related parameters seem to greatly affect the structure-property-performance profiles of carbon nano-onions-based nanomaterials [8]. Since discovery, innumerable experimental

endeavors and a few theoretical studies have been carried out to unfold the design, synthesis, and essential features (morphology, thermal, electrical, optical, fluorescence, mechanical, etc.) of carbon nano-onions and related nanocomposites [9,10]. Consequently, polymer/carbon nano-onions nanocomposites have exposed technical implications in several sectors, like space structural engineering, electronics, energy devices, photocatalysis, and more [11–14].

This state-of-the-art manuscript on polymer/carbon nano-onion nanocomposites is planned due to a literature discrepancy regarding the limited comprehensive and up-to-date review reports in this field. However, few past review reports were observed for carbon nano-onion and carbon nano-onion-based hybrids; however, these reviews involved narrowed and limited insights on design, properties, and actual technical worth of these high-end nanomaterials [15]. To the best of the literature knowledge to date, this all-inclusive review for the first time sheds light on almost every aspect of the structure, property, and performance of polymer/carbon nano-onion nanocomposites. Accordingly, this article aims to systematically disclose the fundamentals, specifications, probable applications, and underlying field challenges of essential categories of polymer/carbon nano-onions nanocomposites. The planned sections of this manuscript initially state novel recent insights on fundamentals of carbon nano-onions (physical/chemical modifications), followed by nano-reinforcement effects for (conducting, thermoplastics, thermosets, green polymers) matrices, then today's application zones, and lastly future/challenging prospects. Hitherto, presenting a broad coverage of a multitude of scenarios of carbon nano-onions for polymeric materials will definitely lead to industrial-level future advancements in this field. Besides, this novel review on polymer/carbon nano-onion nanocomposites is planned as a useful guide for the field scientists.

2. Carbon nano-onions

Carbon nano-onions are zero-dimensional, multi-layered carbonaceous nanoparticles [15]. Like other carbon nanoparticles, carbon nano-onions have been considered as competent nano-reinforcements for polymeric matrices [16,17]. To outperform the characteristics of carbon nanofillers towards polymers, carbon nano-onions have several valuable structural and physical characteristics [18]. For synthesizing these nanoparticles, several efficient practices have been considered in the literature hitherto, including chemical vapor deposition [19], arc discharge process [20], electron beam irradiation [21], ion implantation [22], annealing [23], pyrolysis [24], and the like (**Figure 1**). Using these tactics, carbon nano-onions have been developed both as hollow and dense core nanoparticles [25]. Accordingly, the quality and yield of the finally formed carbon nano-onions rely upon the choice of manufacturing method, related parameters, and their optimization [26]. An important feature of carbon nano-onions can be seen as their susceptibility towards functionalization or modification [27]. The carbon nano-onions surfaces can be altered using simple physical or noncovalent means (**Figure 2**) or covalent routes (**Figure 3**).

Carbon Nano-onions— Manufacture & Applications

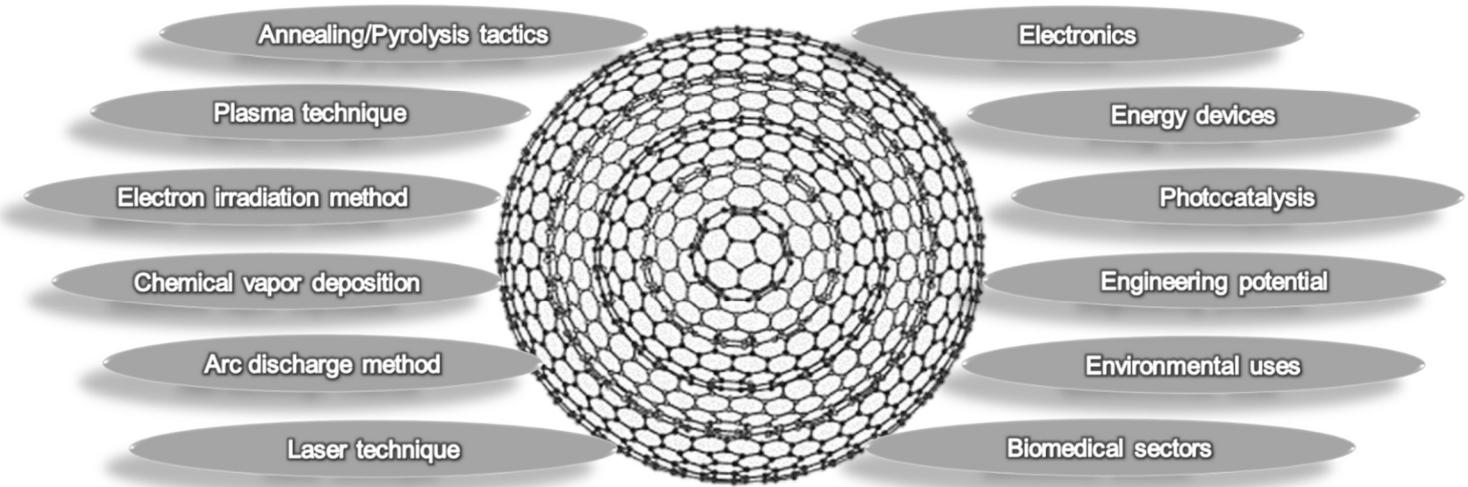


Figure 1. Common manufacturing routes and applications of carbon nano-onions.

Non-covalent Functionalization of Carbon Nano-onions

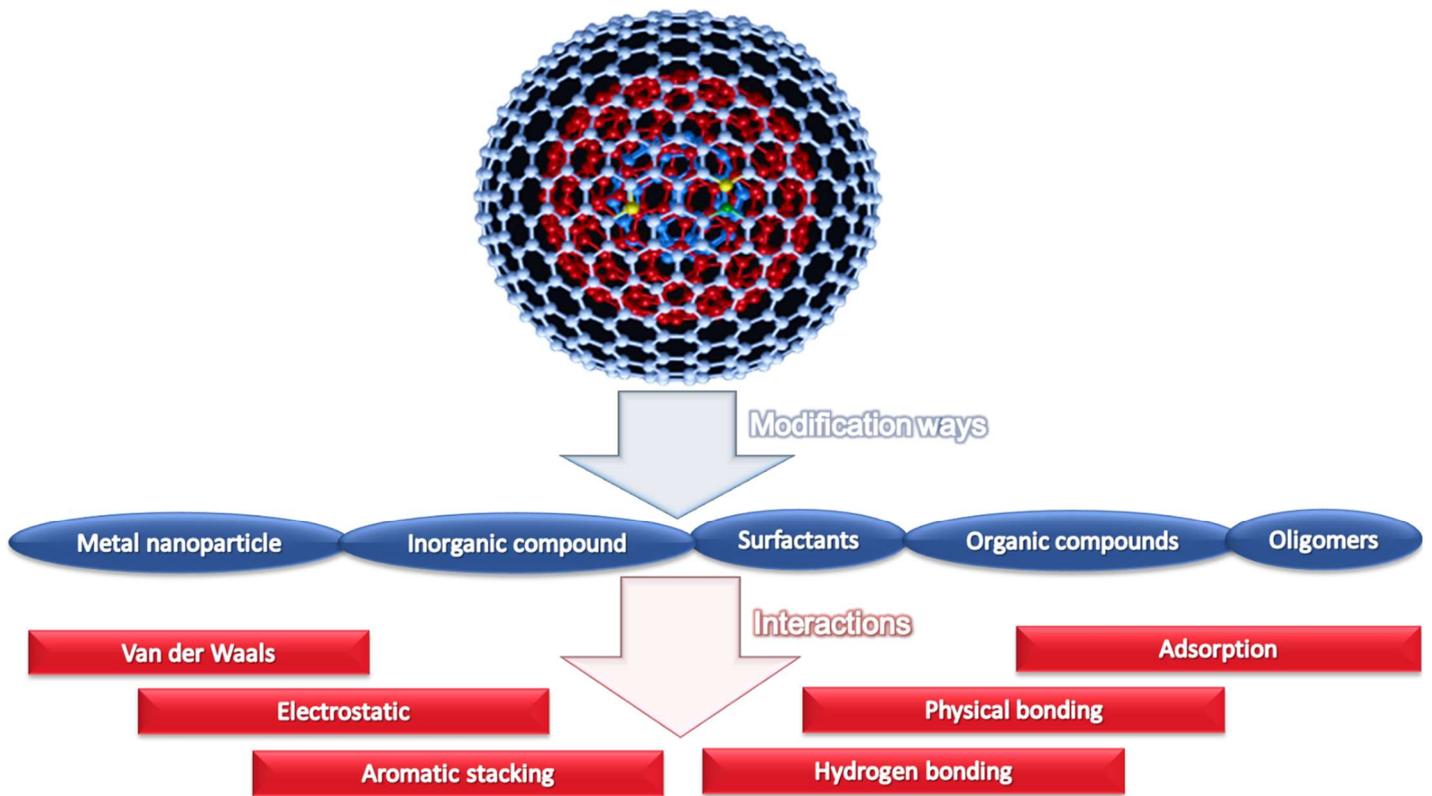


Figure 2. Functionalization routes towards carbon nano-onions.

Physical or noncovalent surface functionalization of carbon nano-onions may involve electrostatic, van der Waals, π - π stacking, hydrogen bonding, and other surface interactions [28]. On the other hand, several covalent routes, such as oxidation, amidation, reduction, fluorination, sulfonation, and so on, have been used for covalent alterations of carbon nano-onions [29]. The purpose of both the physical and chemical

modification of carbon nano-onions seemed to be the development of fine interfacial links with polymers, thereby enhancing the performance of the resulting nanocomposites [30,31]. The practical applications of carbon nano-onions may involve countless zones of electronics (sensors, smart devices) [32], energy conversion devices (photovoltaics, fuel cells) [33], energy storage devices (supercapacitor, batteries) [34,35], catalysis [36], engineering structures [37], and biomedical areas [38].

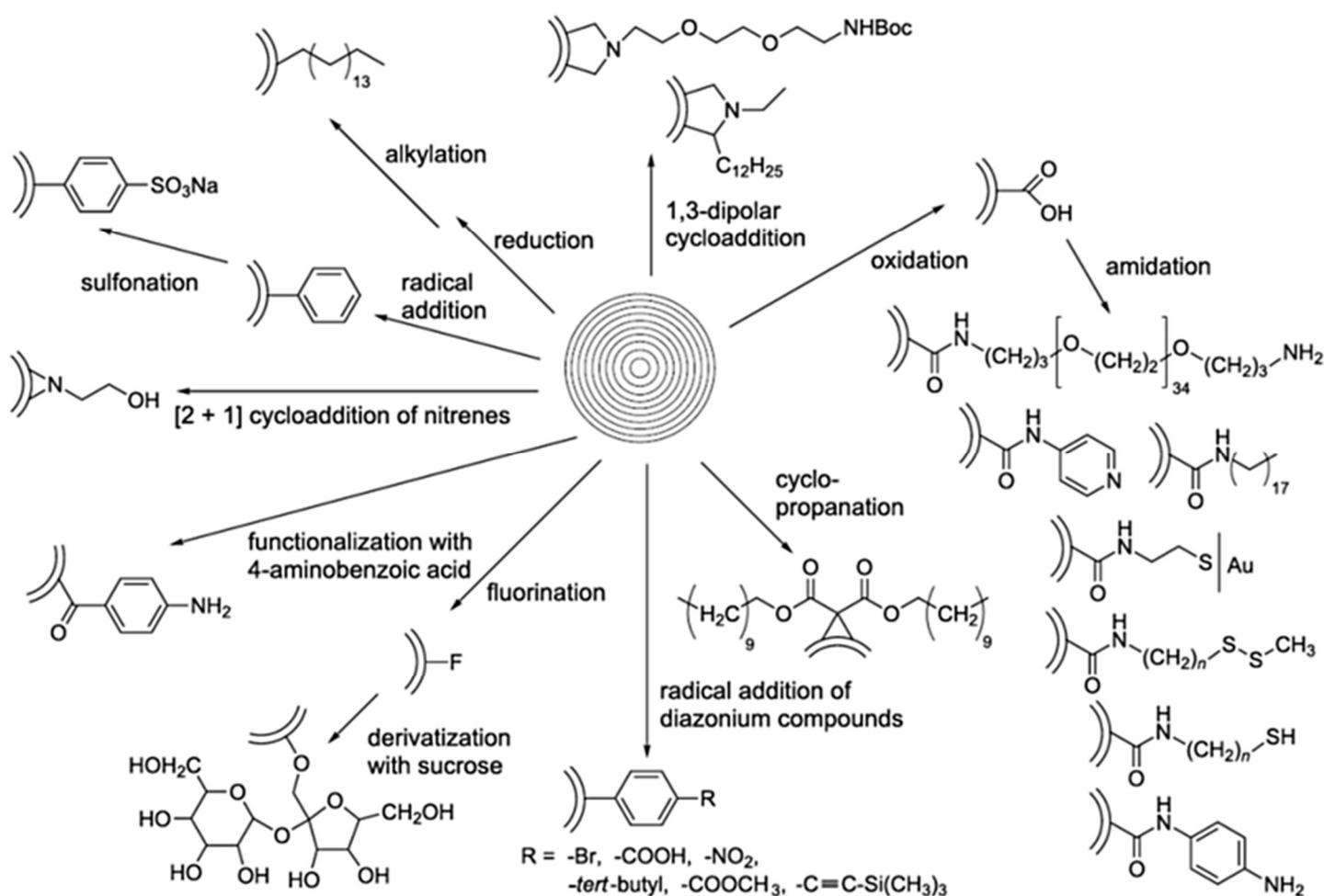


Figure 3. Covalent modification paths for carbon nano-onions [29]. Reproduced with permission from beilstein-journals.org (Open access).

3. Carbon nano-onions nano-reinforcement effects in polymeric nanocomposites

Various types of carbon nano-onions-based nanocomposites have been designed and investigated, including inorganic nanomaterials (like metal nanoparticles) based hybrids [39,40]. These inorganic carbon nano-onions nanomaterials have gained worth for energy, optical, and sensing devices, and biological fields [41]. Nevertheless, design, processing, and ecological limitations seemed to be hindering high-end progressions in the fields of inorganic carbon nano-onions hybrids. Consequently, research advancements have led to the remarkable innovations in the arenas of organic carbon nano-onions nanocomposites [42]. In this regard, polymer/carbon nano-onions

nanocomposites have been essentially explored with major classes of polymers, like conducting, thermoplastics, and thermosetting polymers [43].

Conducting or conjugated polymers (often termed as synthetic metals) constitute a distinct category of macromolecules, owing to semiconducting backbones [44,45]. Some commonly named conductive polymers in the literature to date include polycarbazole, polyacetylene, polyphenylene, polyaniline, polypyrrole, polythiophene, and numerous polythiophene-derived forms [46]. These polymers have been frequently manufactured by using facile electrochemical, in situ, solution, melt, and chemical methods [47]. Like other polymers, conducting polymers have been studied as valuable matrices for a variety of carbonaceous nanoparticles (including carbon nano-onions) [48,49]. Subsequently, for the past few decades, research trends have been noticed for designing high-end conducting polymer/carbon nano-onions nanocomposites [50]. In initial attempts, like Kovalenko et al. [51] reported, in situ electrochemical synthesis of polyaniline and carbon nano-onions derived nanocomposites for supercapacitor electrodes. As per results, adding up to 28 wt.% nanofiller revealed a specific capacitance of 640 Fg^{-1} in 10 000 galvanostatic cycles. Similarly, Plonska-Brzezinska et al. [52] fabricated polyaniline/carbon nano-onions nanocomposites by using the in situ polymerization method for supercapacitor electrodes. As per results, the as-designed nanocomposite had 17 times higher specific capacitance ($\sim 207 \text{ F g}^{-1}$), relative to pristine carbon nano-onions ($\sim 12 \text{ F g}^{-1}$). Correspondingly, initial discoveries have been observed for carbon nano-onions filled polypyrrole, polythiophene, and other conjugated macromolecular systems [53]. For example, Rettenbacher et al. [54] adopted the solution refluxing technique for reinforcing nano-onions in poly(bis-o-diynyl arene) matrix, as mentioned in **Figure 4A**. **Figure 4B** displays a high-resolution transmission electron microscopy micrograph of carbon nano-onions nanoparticles in the conjugated matrix. The nanoparticle can be observed with twenty concentric sp^2 hybrid carbon rings, owing to the efficiency of the synthesis method used for the formation and dispersion of the nanofiller. Moreover, **Figure 4C(a-c)** presents thermogravimetric analysis thermograms for neat carbon nano-onions, unfilled polymer, and the hybrid, respectively. As per results, poly(bis-o-diynyl arene)/carbon nano-onions hybrid had superior initial degradation temperature and char yield of $500 \text{ }^\circ\text{C}$ and 80%, respectively, due to the formation of compatibilized nanocomposites, as compared to unfilled polymer ($400 \text{ }^\circ\text{C}$ and 60%, respectively).

In later attempts, Olejnik et al. [55] formed polyaniline nanotubes by the template method and polyaniline nanotube/oxidized carbon nano-onions hybrids using ultrasonication/reflux in aqueous nitric acid. Commercially available nanodiamonds (sizes $\sim 6 \text{ nm}$) were used to form pristine carbon nano-onions in a yield of $>95\%$ by using carbonization and annealing techniques at $1650 \text{ }^\circ\text{C}$ (pressure 1.1 MPa).

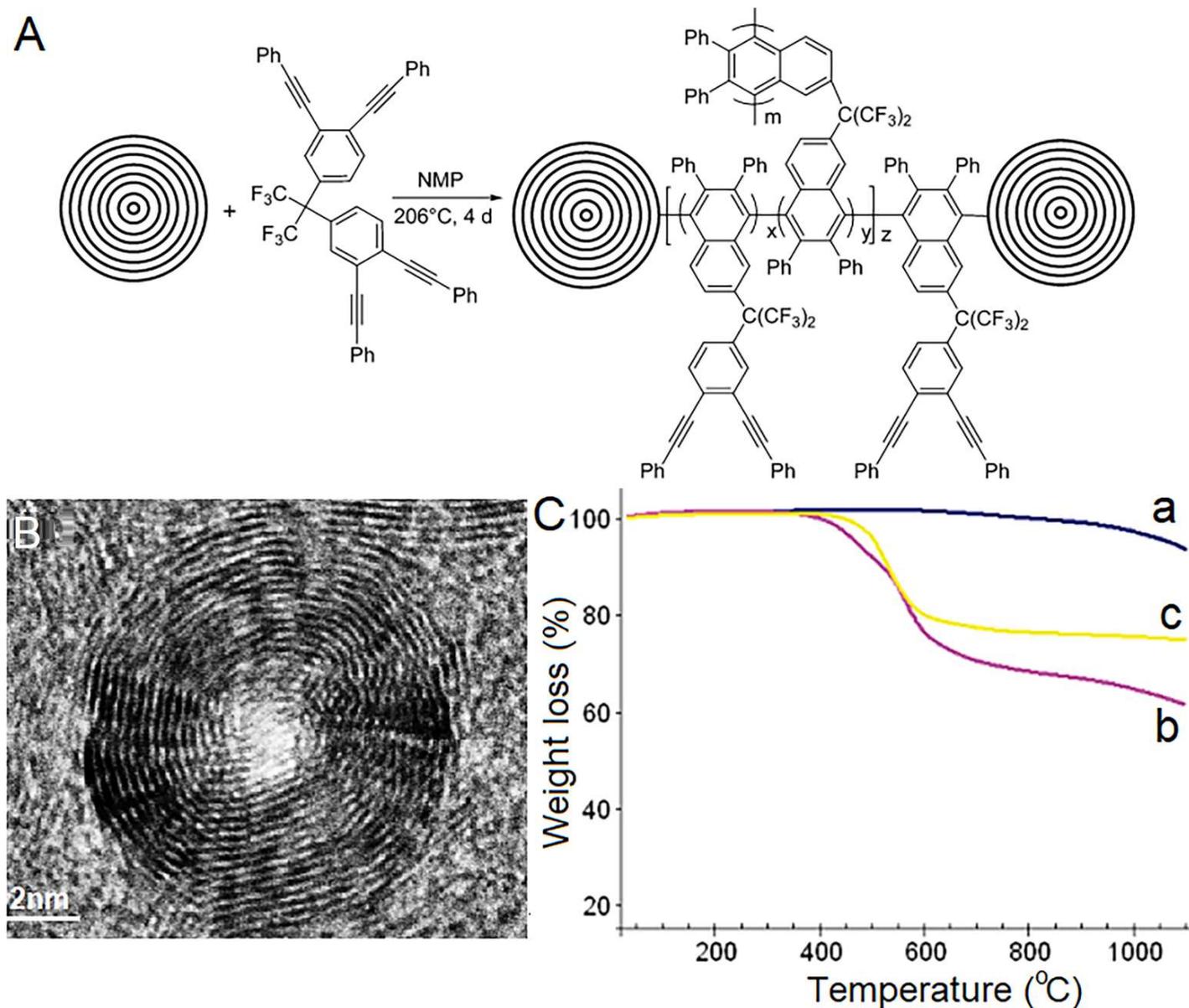


Figure 4. (A) Formation of carbon nano-onions reinforced poly(bis-o-diynyl arene) hybrid; (B) high resolution transmission electron microscopy micrograph of the nano-onion in hybrid matrix; (C) thermogravimetric analysis of (a) carbon nano-onions; (b) poly(bis-o-diynyl arene); and (c) poly(bis-o-diynyl arene)/carbon nano-onions hybrid [54]. NMP = N-Methyl-2-pyrrolidone (NMP). Reproduced with permission from ACS.

Figure 5A illustrates possible structures of pristine polyaniline nanotubes and polyaniline nanotube/oxidized carbon nano-onions. **Figure 5B,C** show scanning electron microscopy images of pristine polyaniline nanotubes and oxidized carbon nano-onions functional polyaniline nanotubes, respectively. Pristine polyaniline nanotubes reveal open-ended smooth surfaces. On the other hand, deposition of carbon nano-onion nanoparticles of ~ 5 nm on polyaniline nanotube walls and nonuniform surfaces can be observed in the micrograph. **Figure 5D** depicts capacitive current vs. sweep rate plots for polyaniline nanotube/oxidized carbon nano-onions hybrid. The capacitive current was linearly dependent on the sweep rate due to fine electrolyte diffusion. Finally, polyaniline nanotube/oxidized carbon nano-onions hybrids had optimally high specific capacitance of >940 F g^{-1} , relative to the nanocomposite with

pristine carbon nano-onions ($\sim 270 \text{ F g}^{-1}$). A few recent attempts have also been noticed for conducting polymeric hybrids with carbon nano-onions, such as Kumari et al. [56], who explored the potential of these hybrids for environmental purposes. Nevertheless, recent research efforts seem limited in the field of conducting polymer/carbon nano-onions hybrids.

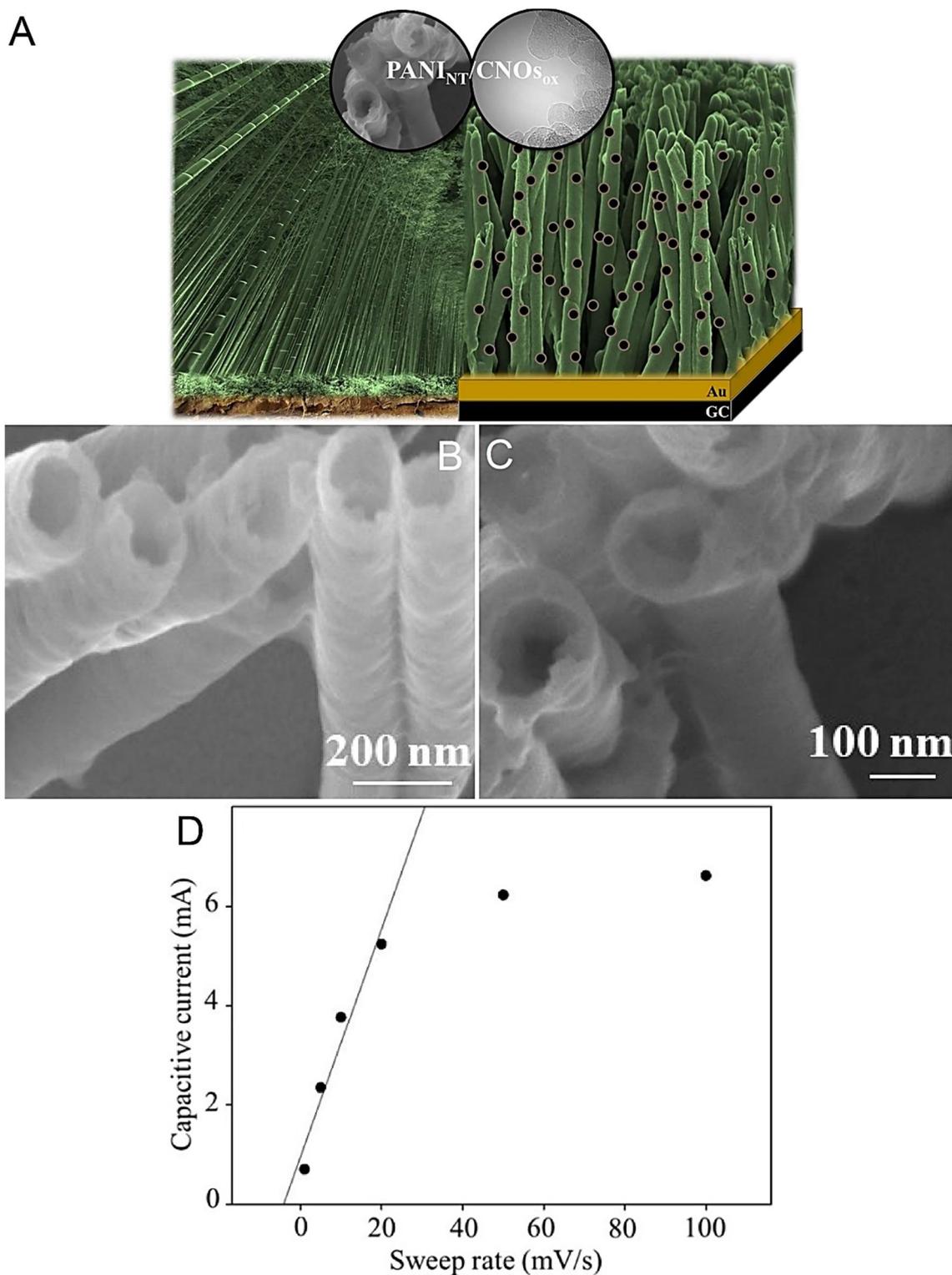


Figure 5. (A) An illustration of (left) pristine polyaniline nanotubes and (right) polyaniline nanotube/oxidized carbon nano-onions; scanning electron microscopy images of (B) pristine polyaniline nanotubes; (C) oxidized carbon nano-

onions functional polyaniline nanotubes; **(D)** The cyclic voltammograms for capacitive current vs. sweep rate dependence [55]. PANI_{NT} = polyaniline nanotubes; CNOs ox = oxidized carbon nano-onions. Reproduced with permission from MDPI.

Thermoplastics (a major category of polymers) have been investigated as competent matrices for nanocarbon (like carbon nano-onions) nanoreinforcements to form high-performance nanomaterials [57–59]. In earlier times, Zhou et al. [60] used ring-opening polymerization technique for the formation of poly(ϵ -caprolactone) grafted carbon nano-onions using ϵ -caprolactone monomers. Similarly, the atom transfer radical polymerization strategy was used to graft polystyrene onto the surfaces of carbon nano-onions. The resulting polymer-grafted carbon nano-onions hybrids developed fine interfacial links and compatibility due to covalently grafted nanostructures. Furthermore, the poly(ϵ -caprolactone) and polystyrene grafted carbon nano-onions nanocomposites revealed superior thermal decomposition temperatures, in the range of 400–450 °C (relative to neat polymers or nongrafted hybrid samples). Consequently, as per recent research reports, carbon nano-onions have been reinforced in several thermoplastic polymers, including poly(vinyl alcohol) [61], poly(vinyl chloride) [62], polyethylene [63], polystyrene [64], poly(ϵ -caprolactone) [65], poly(vinylidene fluoride) [66], poly(methyl methacrylate) [67]. Recently, Serban et al. [68] reported carbon nano-onions filled poly(vinyl alcohol) hybrids using drop casting method. According to atomic force microscopy micrograph (**Figure 6A**), poly(vinyl alcohol)/carbon nano-onions nanocomposite had higher roughness parameter (>430 nm), relative to pristine carbon nano-onions (~354 nm). These results can be credited to the change in surface topographical properties of the nanocomposite with the nanoparticle addition. Furthermore, the as designed poly(vinyl alcohol)/carbon nano-onions nanocomposite was used as a humidity sensor. **Figure 6B** shows relative humidity and resistance vs. time plots for poly(vinyl alcohol)/carbon nano-onions nanocomposite. As apparent from fluctuations in the graph, including carbon nano-onions significantly influenced the relative humidity and resistance of the nanomaterial. **Figure 6C** illustrates relative variation in resistance vs. relative humidity of poly(vinyl alcohol)/carbon nano-onions nanocomposite. Herein, resistance variations were particularly observed at the relative humidity of 80–85%. The outcomes seem to be due to water sorption and desorption processes by the nanocomposite sample.

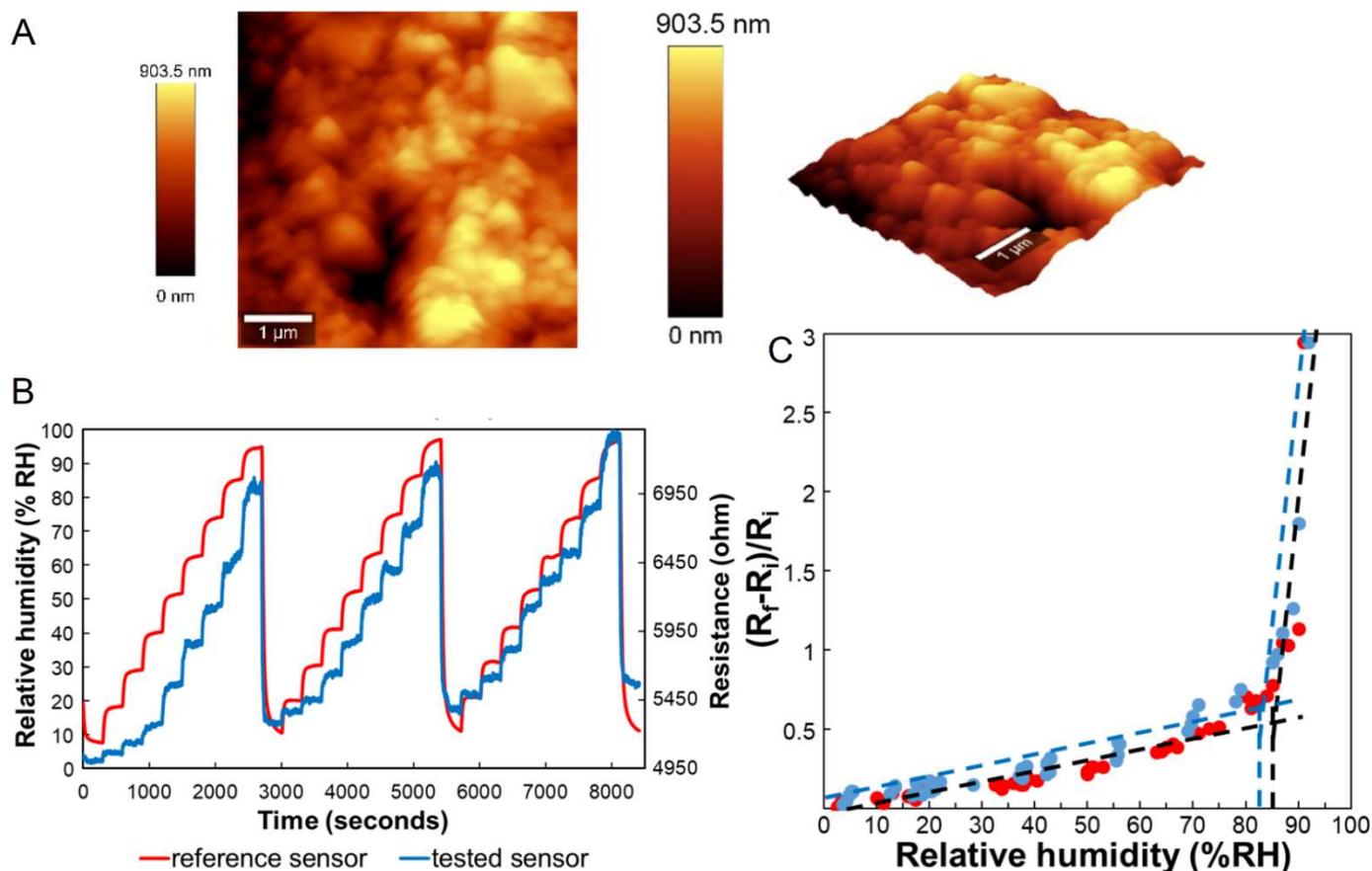


Figure 6. (A) Atomic force microscopy image of poly(vinyl alcohol)/carbon nano-onions nanocomposite (1:1 wt.%); (B) relative humidity and resistance variation vs. time plots for poly(vinyl alcohol)/carbon nano-onions nanocomposite (1:1 wt.%); (C) relative variation in resistance $(R_f - R_i)/R_i$ vs. relative humidity (RH) of poly(vinyl alcohol)/carbon nano-onions nanocomposite (where red dots = sorption; blue dots = desorption; black dashed line = inflection point during adsorption at RH of 50.5%; blue dashed line = inflection point during desorption at RH of 56.5%) [68]. Reproduced with permission from MDPI.

Besides, green thermoplastic polymeric matrix, like chitosan, has also been studied for carbon nano-onions based nanocomposites [69–72]. Grande Tovar et al. [73] industrialized chitosan grafted carbon nano-onions hybrids, using solution reflux and centrifugation methods. **Figure 7A** demonstrates a route for the formation of chitosan grafted carbon nano-onions by the conversion of purified carbon nano-onions into oxidized carbon nano-onions and acyl chloride carbon nano-onions. **Figure 7B** shows thermogravimetric analysis thermograms for pristine chitosan, oxidized carbon nano-onions, and finally formed chitosan grafted carbon nano-onions hybrid. The major degradation for nongrafted chitosan was found at <400 °C, due to the breakage of amine and acetyl chains. On the other hand, two-step decomposition was observed, due to hydroxyl/carboxylic groups on nano-onion surfaces (170–260 °C) and because of breakdown of chitosan backbone and chitosan-carbon nano-onions links (600–700 °C). It was observed that including nanoparticles considerably enhanced the main chain degradation temperature of the polymer backbone. Nevertheless, focused future scientific efforts must be performed for further progresses in the arenas of thermoplastic matrix carbon nano-onions based hybrids.

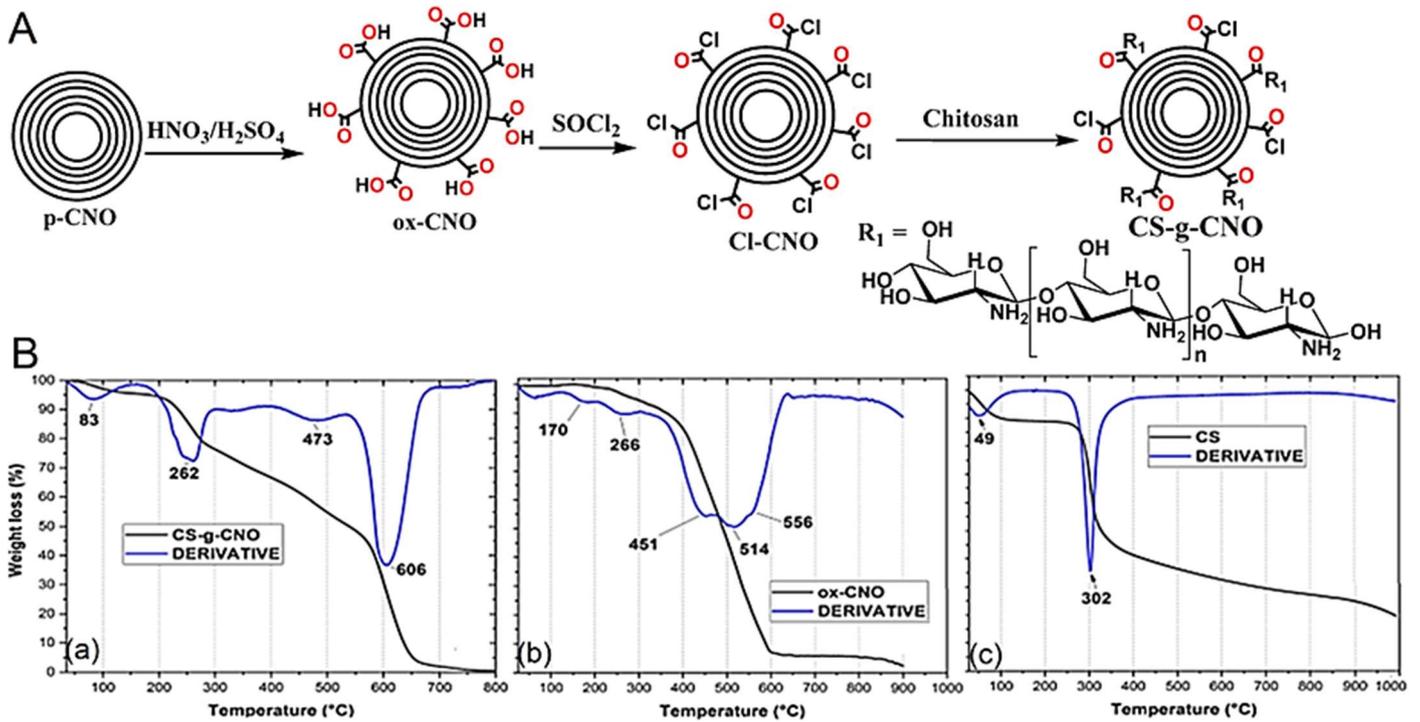


Figure 7. (A) Schematic for the formation of chitosan grafted carbon nano-onion hybrid; (B) thermogravimetric analysis thermograms for: (a) chitosan grafted carbon nano-onion; (b) oxidized carbon nano-onions; (c) pristine chitosan [73]. CS = chitosan; p-CNO = purified carbon nano-onions; ox-CNO = oxidized carbon nano-onions; Cl-CNO = acyl chloride functional carbon nano-onions; CS-g-CNO = chitosan grafted carbon nano-onion. Reproduced with permission from MDPI.

Thermosets have also been favorably investigated as matrices for nanocarbon nanoparticles [74,75]. Most well known thermosetting polymers for nanocomposite formation are epoxy resins [76,77]. In accordance, Palaimiene et al. [78] reinforced carbon nano-onions in epoxy matrix through the solution casting technique. These nanoparticles had hollow nanostructures with sizes of ~50–200 nm and were consistently dispersed in the epoxy matrix. Later, the epoxy/carbon nano-onions hybrids were studied for electrical conductivity features, owing to the formation of percolation networking of nanoparticles in the matrix. Tretjak et al. [79] developed a system of epoxy resin and carbon nano-onions based nanocomposite using a solution curing technique. **Figure 8A** shows transmission electron microscopy micrograph of neat carbon nano-onions. The nanoparticles of 40 nm sizes appeared as tiny clusters with touching boundaries.

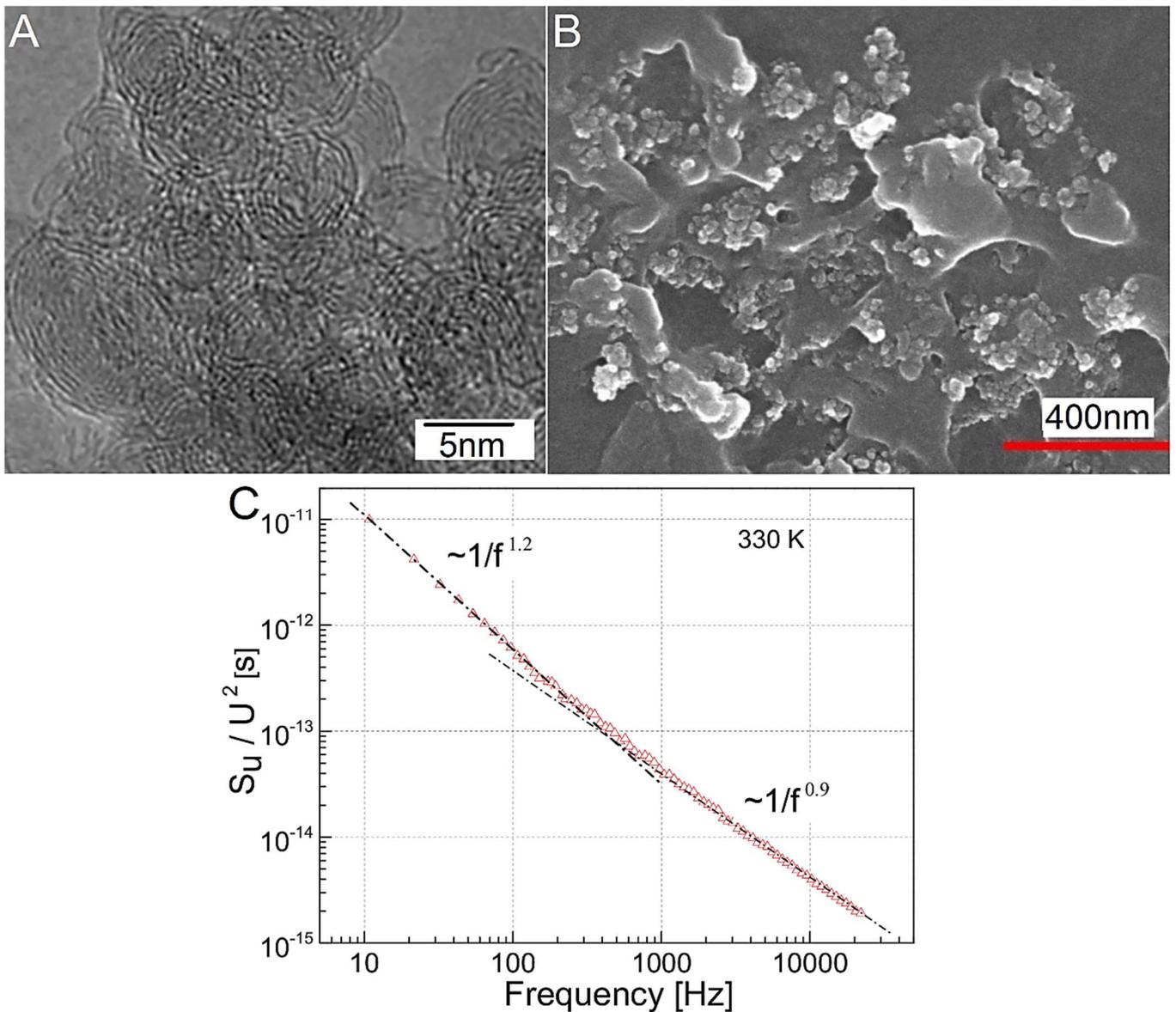


Figure 8. (A) Transmission electron microscopy image of carbon nano-onions (40 nm); (B) scanning electron microscopy image of epoxy/carbon nano-onions nanocomposite (10 wt.%); (C) voltage noise spectrum of epoxy/carbon nano-onions nanocomposite at 330 K (red triangle); lower temperature (black dashed lines) [79]. Reproduced with permission from MDPI.

Figure 8B illustrates scanning electron microscopy micrograph of epoxy/carbon nano-onions nanocomposite with small aggregated nanoparticles dispersed throughout the epoxy matrix. As per voltage noise spectrum of epoxy/carbon nano-onions nanocomposite (**Figure 8C**), presence of small nano-onion nanoparticles caused tunnelling and barrier effects; thereby demanding higher voltage noise. However, among all categories of polymers, carbon nano-onions have been most restrictively explored with the epoxy matrices, because just few studies have been conducted in this area.

According to the scientific surveys presented in this section, carbon nano-onions have been applied as remarkable nanofillers for conjugated, thermoplastics, as well as thermosetting matrices. Nevertheless, with all these matrices, sufficient comprehensive literature investigations were not conducted to truly unveil the

potential of carbon nano-onions nanofillers. Thus, systematic future investigations need to be performed to establish multitude of promising polymer/carbon nano-onions nanocomposites and related next-level industrial deployments.

4. Applied zones of polymer/carbon nano-onions nanocomposites

Carbon nano-onions derivated nanocomposites (either physically or covalently linked) have valuable microstructural, physicochemical, and applied features, owing to interfacial compatibility [80,81]. Research progressions in the field of polymer/carbon nano-onions hybrids can be seen in the form of a multitude of applied designs [82,83].

Among applied disciplines, these multifunctional carbon nano-onions based nanomaterials have gained focused for aerospace engineering structures [84,85]. In this concern, carbon nano-onions hybrids have revealed technical worth for engines, wings, and fuselage of spacecrafts [86]. In spite of this technical worth of carbon nano-onions in aerospace zones, confined research endeavors have been performed up till now; thereby demanding future research focus of field scientists and engineers [87].

A widely explored research zone for carbon nano-onions hybrids has been observed for energy and electronic systems, like energy production (solar cells), energy storage (supercapacitors, batteries), and electronics (sensors) [88,89]. For energy production devices, earlier attempts involved using inorganic nanomaterials in solar cell devices, which proved to be an expensive and non-ecological approach [90,91]. Afterwards, research trends have been shifted towards polymeric and nanocarbon (carbon nano-onions, fullerene, nanodiamond, graphene, etc.) type nanomaterial designs for solar cells [92,93]. In addition, organic-inorganic hybrids have also been considered for photovoltaics [94–96]. Notably, for commercial-level success, photovoltaic devices must have a power conversion efficiency of 10–20% or higher [97]. Such performance can only be attained by adjusting or optimizing the fabrication and interfacial parameters of carbon nano-onion-based designs [98]. Accordingly, pristine carbon nano-onions as well as related polymeric nanocomposites have been focused for high performance solar cell devices [99]. For instance, Zheng et al. [100] reported poly(3,4-ethylenedioxythiophene): polystyrene sulphonate/carbon nano-onions nanocomposite as a hole transporting layer for perovskite solar cell. This system exhibited a superior power conversion efficiency of >15%. Here again, restricted studies have been seen so far regarding carbon nano-onion-based solar cell architectures.

Concerning energy storage systems, supercapacitors gained scientific curiosity as the most efficient charge-storing and charge-discharging devices [101–103]. Modern supercapacitor designs have been industrialized using carbon nanoparticles, such as graphene, fullerene, nanodiamonds, and carbon nano-onions [104]. For this purpose, various functional forms of carbon nano-onions have been designed. For instance, Pallavolu et al. [105] prepared nitrogen-doped carbon nano-onions by using the flame pyrolysis technique. The modified nanoparticles revealed a specific capacitance of 234 F g⁻¹. Pan et al. [106] prepared nitrogen/oxygen-codoped carbon nano-onions by using carbonization and hydrothermal methods. The modified carbon nano-onions were used for supercapacitor electrodes. Consequently, power density

and energy density of 400 kW kg⁻¹ and 71 Wh kg⁻¹, respectively, were attained. Similarly, Jin et al. [107] and Mohapatra et al. [108] developed modified carbon nano-onions with doped and porous surfaces for supercapacitor electrodes. The as-designed nanomaterials revealed superior surface area, specific capacitance (>350 F g⁻¹), and capacitance retention (~99%) for high-end device applications. Majumder et al. [109] developed polyaniline and carbon nano-onions derived nanocomposite via in situ polymerization technique. The resulting hybrid had a reasonable specific capacitance of ~200 F g⁻¹ and power densities. The polyaniline/carbon nano-onions nanocomposite electrode performance was credited to the superior surface area and wettability towards the electrolyte. However, carbon nano-onions linked nanomaterials need to be further explored due to limited research endeavors regarding their charge-storing competence.

Another essential category of charge-storing devices exploiting nanomaterials can be named as batteries, especially lithium-ion batteries [110,111]. In accordance, Zhou et al. [112] recently used plasma-enhanced chemical vapor deposition technique and nickel nanoparticle templates to form core-shell carbon nano-onions. These nanomaterials depicted superior specific capacity (>660 mAh g⁻¹) and capacity retention (~94%) over repeated cyclic performance (500 cycles). Moreover, Namwong et al. [113] formed successfully formed carbon nano-onions hybrids using the template technique. Considerably higher specific capacity of >1100 mAh g⁻¹ was attained for these nanomaterials. Among a few more attempts, Hryniewicka et al. [114] formed nitrogen and sulfur doped carbon nano-onions and amalgamated with polyacrylonitrile-poly(acrylic esters) block copolymer to form lithium sulfur battery electrodes. The resulting block copolymer/carbon nano-onions nanocomposite-based battery electrode exhibited 20% higher specific capacity, relative to a pristine carbon nano-onions-based electrode. The field of carbon nano-onions hybrids based charge-storing batteries also seemed to be an inhibited area of research demanding future scientific attention for industrial deployments.

Amongst electronic devices, advanced sensors have been developed using carbonaceous nanomaterials (graphene, carbon nanotube) [115]. Hither, carbon nano-onions and related nanocomposites have also been sightseen for smart sensing devices [116,117]. Consequently, Panda et al. [118] formed a dual sensor (fluorescence plus electrochemical) using carbon nano-onions and hydrothermal practice. Amid few polymer/carbon nano-onions designs, Serban et al. [119] anticipated poly(vinyl alcohol) and carbon nano-onions based nanocomposites for humidity sensors. The resulting poly(vinyl alcohol)/carbon nano-onions hybrids were found to have chemiresistive performance desirable for humidity detection of >80%. The efficiency of these systems was credited to the electron acceptor-donor interactions of poly(vinyl alcohol)/carbon nano-onions hybrid sensor towards the water molecules. Nevertheless, more scientific attempts must be performed to expand this essential application of polymer/carbon nano-onions nanocomposites towards molecular sensing and biosensing sectors.

Last but not least, carbon nano-onions and derived carbon nano-onions nanomaterials have been tested for catalysis and photocatalysis potential [120,121].

Hence, the environmental/sustainability impact of carbon nano-onions and derived nanocomposites can be validated by the future expansions in these areas [122,123].

Figure 9 displays an illustration to summarize the applied zones of carbon nano-onion-based nanocomposites, as argued in this section. It can be stated that industrial-level progressions of these nanomaterials completely rely on intense curiosities and collective future endeavors by the field scientists and engineers.

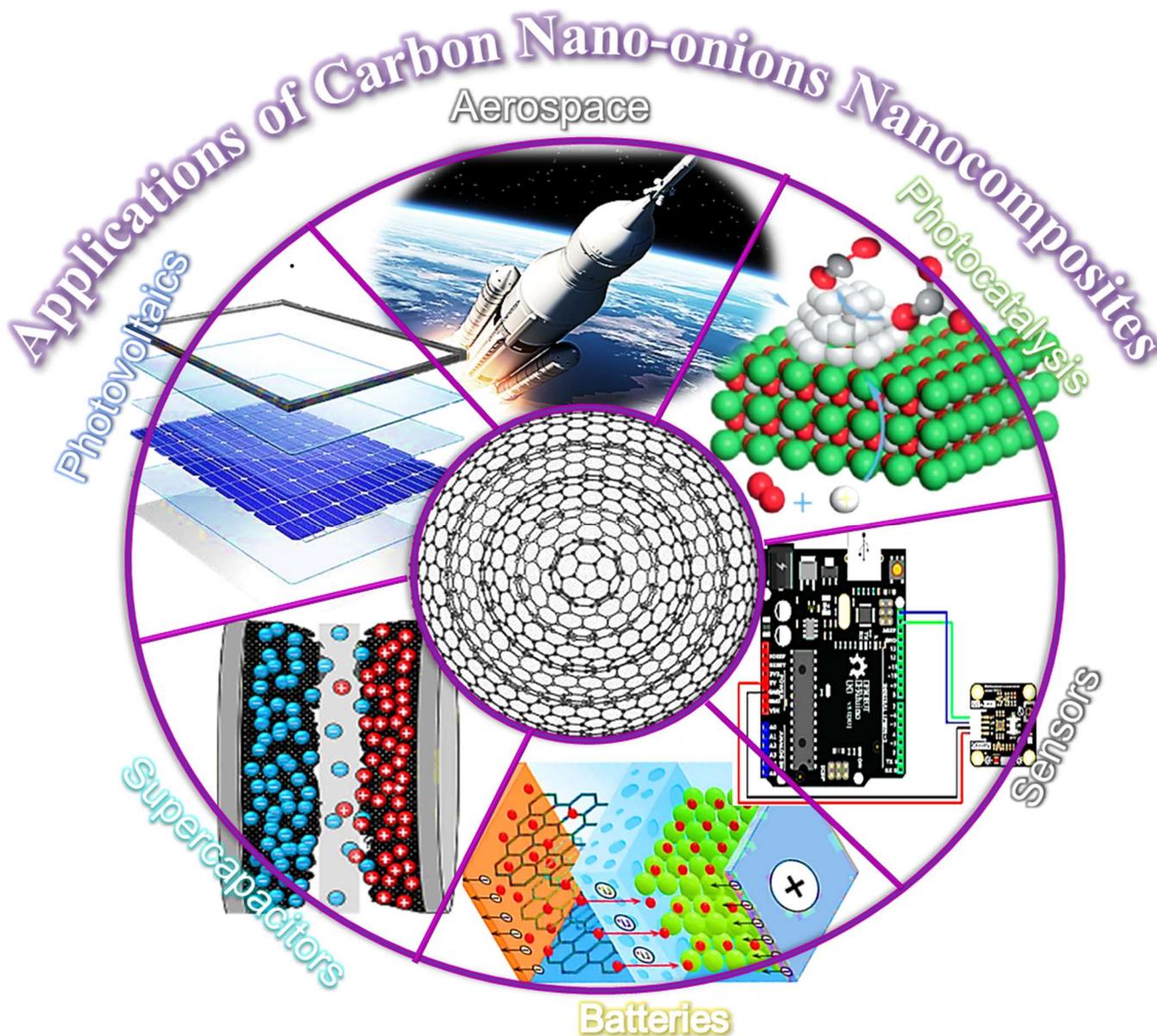


Figure 9. Schematic of applied zones of carbon nano-onion-based nanocomposites.

5. Challenges and future lookouts

Table 1 presents an overview of carbon nano-onion-reinforced matrices studied for different properties and applications. In addition, mechanical, thermal, capacitance, and other physical attributes of polymer/carbon nano-onions nanocomposites systems have been compared with the analogous hybrids with graphene or carbon nanotubes. As per analysis, several studies supported the superior

reinforcing effects of carbon nano-onion nanoparticles, relative to other carbon nanofillers.

Table 1. Design, essential properties, and applications of carbon nano-onions based nanocomposites and comparison with other nanocarbon-based nanomaterials.

Matrix	Nanofiller/loading (wt.%)	Fabrication	Characteristics/applications	Property improvement (%)	Ref
Polyaniline	Carbon nano-onions/ 28 wt.%	In situ electrochemical synthesis	Physical interactions; specific capacitance 640 Fg ⁻¹ ; 10,000 galvanostatic cycles; Supercapacitors	156–180	[51]
Polyaniline	Carbon nano-onions/ 15 wt.%	In situ polymerization	Physical interactions; specific capacitance ~207 F g ⁻¹ ; supercapacitors	1625	[52]
Polyaniline	Carbon nanotubes; graphene 1–5 wt.%	In situ polymerization	Specific capacitance ~230–250 F g ⁻¹ ; Supercapacitors	15–25	[124,125]
Poly(bis-o-diynyl arene)	Carbon nano-onions/ 2 wt.%	Solution refluxing technique	Physical interactions; initial degradation temperature and char yield 500 °C and 80%, respectively; supercapacitors	25–33	[54]
Polyaniline	Carbon nano-onions/ 1 wt.%	Ultrasonication/ reflux method	Physical interactions; specific capacitance >940 F g ⁻¹ ; supercapacitors	248	[55]
Poly(ε-caprolactone)	Carbon nano-onions/ 3 wt.%	Ring opening polymerization technique	Covalent linking; thermal decomposition temperatures ~450 °C	50	[60]
Poly(ε-caprolactone)	Carbon nanotube/ 1 wt.%	Ring opening polymerization technique	Thermal decomposition temperatures ~400 °C	5	[126]
Poly(ε-caprolactone)	Graphene oxide/ 5 wt.%	Ring opening polymerization technique	Thermal decomposition temperatures ~420 °C	5	[127]
Poly(vinyl alcohol)	Carbon nano-onions/ 1–5 wt.%	Drop casting method	Physical linking; roughness parameter >430 nm; relative humidity 80–85%; humidity sensor	50	[68] [119]
Chitosan	Oxidized/acyl chloride carbon nano-onions/ 1–5 wt.%	Solution refluxing; centrifugation method	Covalent grafting; maximum thermal degradation; temperature 600–700 °C	50–70	[73]
Chitosan	Carbon nanotubes/ 5 wt.%	Solution method	Maximum thermal degradation temperature 550 °C	10	[128]
Chitosan	Graphene oxide/ 1–3 wt.%	Solution method	Maximum thermal degradation; temperature ~350–400 °C	10–14	[129]
Poly(3,4-ethylenedioxy-thiophene): polystyrene sulphonate	Carbon nano-onions/ 25 wt.%	Casting technique	Hole transporting layer; power conversion efficiency; perovskite solar cell	>15	[100]
-	Nitrogen/oxygen-codoped carbon nano-onions	Carbonization; hydrothermal method	Power density and energy density 400 kW kg ⁻¹ and 71 Wh kg ⁻¹ , respectively; Supercapacitor	50	[106]

Polyacrylonitrile-poly(acrylic esters) block copolymer	Nitrogen/sulfur doped carbon nano-onions/ 1 wt.%	Casting	Specific capacity; lithium ion batteries	20	[114]
Poly(vinyl alcohol)	Carbon nano-onions/ 1 wt.%	Solution method	Tensile strength; tensile modulus	27–38	[70]
Poly(vinyl alcohol)	Carbon nanotube/ 9 wt.%	Solution method	Tensile strength; tensile modulus	3–5	[130]
Poly(vinyl alcohol)	Graphene/ 0.5–3.5 wt.%	Solution method	Tensile strength 29 MPa; tensile modulus 0.52 GPa	10–15	[131]

Despite the research progress so far, fabrication of carbon nano-onions and polymer/carbon nano-onion nanocomposites seemed to be restricted to small or lab scale R & D. Subsequently, several challenges have been noticed hindering environmentally friendly fabrication of these nanomaterials on a large scale. In addition to commercial level production and scalability, significant research efforts have not been performed yet to overcome the challenges for reproducibility of polymer/carbon nano-onion nanocomposites. In this case, precursor material, material, design, synthesis technique, processing parameters and conditions, material and fabrication expenses, product yield, purity, reproduction, and similar aspects need to be investigated. Herrin, use of expensive hydrocarbons, catalysts, toxic solvents, energy demands, and costs of carbon nano-onion and nanocomposite synthesis seemed to be responsible for economic and environmental challenges for industrial-scale productions [132,133]. For instance, carbon nano-onions had a higher price (>1000 \$/Kg), as compared to graphene (up to 100 \$/Kg) and carbon nanotubes (100–200 \$/Kg) nanoparticles [134]. Similarly, reproducibility and sustainability of these nanomaterials depend upon future studies on their life cycle assessments (LCAs) from green synthesis to end-of-life stages [135]. Hence, economic, ecological, sustainability, and reproducibility challenges have been noticed regarding the commercial-scale production of carbon nano-onion-based nanomaterials. As per analysis, modification of nano-onions can be proved effective to further enhance the physical and physicochemical specs of polymer/carbon nano-onions nanocomposites. In short, by overcoming all these limitations, applications of carbon nano-onions-filled nanomaterials can be further expanded towards aerospace, batteries, capacitors, solar cells, sensors, photocatalysis, and other high-tech fields related to today’s industrial sectors. Nonetheless, limited research efforts so far regarding polymer/carbon nano-onions hybrids seemed to be the major hindrance for their further field progress, large-scale production, reproducibility, and real-world commercial deployments.

6. Conclusions

Briefly, this dynamic review thoroughly discloses the worth of a unique zero-dimensional nanocarbon (carbon nano-onion) for different macromolecular categories. Consequently, different types of polymer/carbon nano-onions nanomaterials have been developed with anomalous microstructures, electrical conductivity, charge/ion transport, heat resistance, mechanical resilience, catalytic, and other notable characteristics. Both pristine and modified carbon nano-onions have been used for the formation of polymeric nanocomposites. Incidentally, synthesis,

feature explorations, and probable applications of polymer/carbon nano-onions hybrids have been discussed in this article. As a distinct category of polymer/carbon nano-onions nanocomposites, conjugated polymer-based hybrids have been designed by mostly opting for in situ, electrochemical, and solution tactics. For manufacturing thermoplastic polymer/carbon nano-onions nanomaterials, solution, sonication, and refluxing techniques have been adopted. On the other hand, thermosetting polymers such as epoxies have been processed via solvent casting and curing strategies. As per analysis, significant structural, physical, and high performance scenarios of polymer/carbon nano-onions nanocomposites were dependent on matrix-nanofiller miscibility and interfacial effects. Remarkable progress has been observed for different types of polymer/carbon nano-onion hybrids in the fields of engineering structure, energy/electronic devices, and catalysis. It has been noted that despite the field's progress so far, future scientific interest may overcome the foremost challenges hindering the scalability and commercialization of these nanomaterials. Conclusively, focused future research endeavors on polymers/carbon nano-onions nanocomposites may open several advanced avenues for worldwide modern scientific communities.

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