

Comprehensive review on carbon nanotubes embedded in different metal and polymer matrix: fabrications and applications

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Abstract

Carbonaceous material, especially carbon nanotubes (CNTs), have incredible exceptional properties, such as high thermal and mechanical stabilities, and good catalytic and adsorption capabilities. In recent years, hybrid nanocomposites have attained considerable attention due to the combination of unique organic and inorganic elements in a single material. Hence, these nanocomposites have been employed for various applications such as drug delivery, sensors, corrosion protection materials, flame retardant additives, and pollutant adsorbent. These nanocomposites can be fabricated through various approaches that include powder metallurgy, solution processing, reaction processing, melt processing, electrochemical, and many more. This present review mainly summarizes the various techniques for the fabrication, separation, and purification of CNTs and their nanocomposites, especially CNTs-based polymer and CNTs-based metals/ metal oxides nanocomposites. Besides, effects of CNTs embedded with

polymers (such as polypyrrole (PPy), poly-aniline (PANI) and poly-thiophene (PTh), etc.) and metals/ metal oxides (such as manganese oxide (MnO₂), copper (Cu), gold (Au), platinum (Pt), etc.) and how they can be employed toward innovative devices with fascinating properties for a broad range of applications. Moreover, industrial applications of CNTs-based polymer/ metal/ metal oxides nanocomposites have been discussed.

Keyword: *Nanocomposites, Carbon Nanotubes, incorporation, nanocomposites applications*

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1. Introduction

Nanostructured materials such as nanoparticles, nanocomposites, and nanocrystals have attained significant interest due to their exceptional chemical and physical features [1]. In general, carbon-based nanomaterials such as graphene and carbon nanotubes have offered new exciting opportunities for chemists, biologists, material scientists, and physicists owing to their fascinating combinations of chemical and physical properties [2]. Appraising nanomaterials' potential in terms of future applications in nanotechnology, has been the primary concern for researchers [3]; [4]. The initial consideration was to increase the surface to volume ratio to lower the size of the material in nano-scale. It directs to various chemical and physical properties from those of their bulk, mostly measured irrespective of their size. In this manner, many researchers working in nanosciences have been keen to understand the essential physical properties that include exciton generation, exciton life span, Auger recombination, electron injection, and inter-system crossing organic metallic and semiconducting nanomaterials as a function of their particle size.

Regarding carbon-based nanomaterials, especially the size-dependent features, including an excellent combination of mechanical, thermal, optical, electronic behaviour, and

exceptional material properties such as ballistic electronic conduction, catalytic, high current densities, and transparent conduction films. Nanomaterials' size-dependent physical properties have been established to be related to (a) ratio of surface atoms to total atoms in their volume and (b) intrinsic properties of the interior of nanomaterials maintained and modified through alignment of surface atoms. Hence, nanomaterials' physical properties are ruled through their quantum size. More remarkably, controlled-size nanomaterials have displayed attractive physical properties and have been the most technologically vital type of materials [5]; [6].

Nanomaterials have been used as building blocks for devices; for instance, energy conversion systems of the third generation photo-voltaic offer not only down-scaling conventional technologies via no less than an order of magnitude but also provide economical and eco-friendly fabrication pathway because of extreme reduction in the required amount of raw materials [7]. In this case, outstanding electronic and electrical properties, a broad range of electrochemical stability window, and considerable surface area carbon-based nanomaterials have been engineered to produce donor-bridge-acceptor molecular devices, mimicking the competent route of photo-synthesis in nature [8]. Furthermore, carbon nanomaterials' chemical and physical properties can be incredibly fine-tuned and altered and regarding particle size, which is influential first hand and intriguing parameters [9]-as aforementioned, reducing the size to a nanometer of bulk material will increase the nanomaterials' specific area. Therefore, applications like reaction at the liquid-solid or gas-solid interface will also help from this 'striving to smaller' problem. Typical applications related to natural processes include energy conversion, environmental chemistry, energy conversion, and catalysis, where the use of nanomaterials inclines the efficiency, response time, sensitivity and surface area [10].

Apart from the abovementioned nanomaterials, functional carbon-based nanomaterials have fascinated extensive attention, i.e. fullerene (0-D), carbon nanotubes (1-D), graphene (2-D single sheet), graphite (3-D), amorphous carbon, and diamond. Compared to all mentioned

carbon-based nanomaterials, carbon nanotubes that provide a strong robust, economical and exciting platform for developing several types of nanoscale architectures, have remarkably paved the research fields of nanotechnology like none other kinds of materials [11]. These materials have been researched intensively due to the unique properties that predestine them for several potential applications. Several publications have also been devoted to CNTs' unique properties. Most researches reported CNTs having a tubular structure formed entirely of rolled-up layers of interconnected atoms of carbon with diameters range from ten nanometers, whereas length up to centimetres. In terms of structural perspective, CNTs are the third allotropic forms of carbon atoms, with the first and second being diamond and graphite, respectively. Concerning its layers, CNTs are categorized as single-walled carbon nanotubes (SWCNTs), multi-walled carbon nanotubes (MWCNTs), and double-walled carbon nanotubes (DWCNTs). Generally, CNTs holds high specific areas because of hollow geometry, whereas, chemical inertness support and structural integrity relatively high oxidation-stability [12].

Carbon nanotubes can behave either as semiconductors or metals; thus, they can be suitable for several applications such as chemical sensors, field-effect transistors, artificial muscles, space elevators, solar cells, memory devices, integrated circuits, and detectors. Several other unique properties have also been described in text-books and research reviews. Moreover, the nanotechnology and nanoscience field move forward for further investigation to fabricate CNTs with the appropriate parameter for near-future applications. Interestingly, CNTs' electronic and optical features are size-dependent, where the bandgap of semiconducting CNTs is inversely proportional to the tube diameter. One more essential issue, mainly with MWCNTs, is carbon layers consisting of CNTs [13]. Due to the interesting size-dependent properties, CNTs' applications with heterogeneous morphology can be constrained until mono-disperse CNTs in their properties and structure can be developed. Nanomaterials

fabrication with mono-disperse sizes, uniform morphologies, and modified surfaces has been the chief challenges [14].

Several approaches have been designed to synthesis mono-disperse CNTs, aiding their future applications. Bottom-up and top-down are two of the initial approaches that have designed for mono-disperse CNTs fabrication. Typically, CNTs can be produced using different routes, such as arc discharge, physical/ chemical vapour deposition, solvothermal, electrochemical deposition, microwave method, and several others [15]. Every synthesis route comes with merits and demerits, which briefly explained in Table 1.

For the last few years, there have been extensive investigations conducted using CNTs nanocomposites in different materials, such as polymer, metals, and ceramics. Most of the researches has been conducted on the incorporation of ceramics via CNTs. It can be accredited mainly to the relative ease of ceramic fabrication compared to metal and polymer. Researches on CNT incorporated with the ceramics matrix are less than those on polymer and metal matrix.

This review presents recent developments in carbon nanotube-based nanocomposite synthesis, properties, and various applications. This review aims to increase the visibility of CNTs embedded polymers, metals, and metal oxides. Moreover, it also to provide an extensive overview of its recent advances in both laboratory and industry. This review addresses in details various fabrication and processing techniques of CNTs, carbon- and CNTs-based polymer, and metal/metal oxides nanocomposites. Various possible applications of carbon- and CNTs-based polymers/metals/metal oxides nanocomposites have been discussed in detail. As it is multi-disciplinary research, further enhancement and development are urgently needed. Hence, at the end, some crucial prospects have been suggested based on the findings.

2. Synthesis of Carbon Nanotubes

Hollow microtubules of graphitic carbon with around 4-30 nm outer diameter and 1 μm length are recognized as carbon nanotubes following their discovery in 1991 by Sumio Iijima. They were identified to be a concentric assembly of two/ multi seamless graphite layers cylinder and were termed as multi-walled carbon nanotubes (MWCNTs) [16]; [17]. In 1993, graphitic cylindrical multi-layers were stripped off successfully by Tsang et al. [18] through oxidation using carbon dioxide and reached the edge attaining nanotubes comprising of single graphitic layer, consequently revealing the presence of single-walled carbon nanotubes (SWCNTs). Since then, several headways have been proposed producing of MWCNTs and SWCNTs over a range of substrates and in the soot form. Among the various synthesis methods, chemical vapour deposition, template methods, laser ablation, and electric arc discharge are most recognized in the literature. While several reports of producing CNTs using the solar furnace, solid pyrolysis, electrolysis, synthesis through the bulk polymer, diffusion flame, etc., are also observed with attention; therefore, CNTs of different morphologies and dimension are formed [19]; [20]; [21]; [22]; [23]. Figure 1 illustrates various synthesis approaches of CNTs that have been used at the lab and/ or industrial scale.

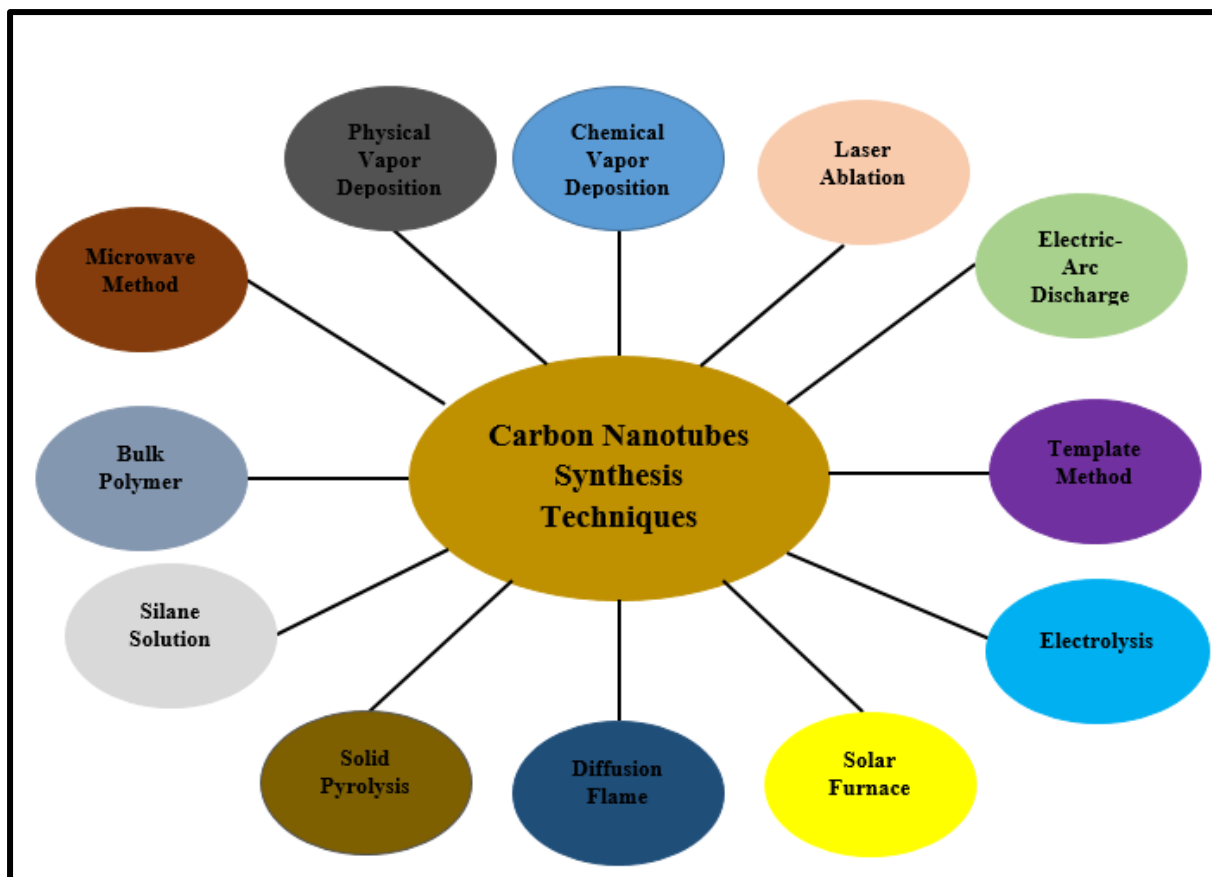


Figure 1: Carbon Nanotubes synthesis techniques

Table 1: Overview of carbon nanotubes synthesis techniques

| Synthesis Technique | Discovery | Principle | Experimental Description | Merits | Demerits | References |
|------------------------|----------------------|--|---|--|---|-----------------------|
| Electric-arc discharge | 1991 by Sumio Iijima | Two graphite rods acting as electrodes are placed in an enclosure, filled with some inert gas at low pressure. The anode is moved closer to the cathode until an arc appears. After a specific arc discharge duration, depressurization, and cooling of the chamber, CNTs, and by-products are collected. Without a catalyst, CNTs are fullerene, MWCNTs, and graphite carbon. However, in | <ul style="list-style-type: none"> ● Inert gas: Helium, Argon. ● Pressure: 50 to 700 mbar. ● Power: Between 50 to 100A ● Catalyst: Iron, yttrium, nickel, sulfur, and molybdenum. | <ul style="list-style-type: none"> ● Form better quality nanotubes. ● Form slight defects on the nanotubes. ● Cost-effective technique ● A simple and convenient technique | <ul style="list-style-type: none"> ● Energy consumption is higher. ● Insignificant for industrial up-scaling. ● Relatively high temperature is needed for operation ● Compared to other techniques, the | [24]; [25]; [26]; [7] |

| | | | | | | |
|----------------|------------------------------------|--|---|---|---|------------------|
| | | the presence of a catalyst, SWCNTs are formed. | | | length of CNTs produced are shorter | |
| Laser-ablation | 1996 by Smalley and his co-workers | <p>In this technique, intense laser pulses ablate which a carbon target, usually containing transition metals as a catalyst, are placed in a tube furnace to heat. During the process, the inert gas is flowed via the chamber to carry the formed nanotubes to the copper collector. The CNTs and by-products are collected once the laser beam switch-off and the chamber is cooled.</p> | <ul style="list-style-type: none"> ● Temperature: 1200 °C ● Inert gas: Argon ● Light intensity: 12 W/cm². ● Catalyst: Copper, cobalt, nickel, lead, cobalt/ nickel, cobalt/ lead | <ul style="list-style-type: none"> ● The technique is applicable at room temperature. ● Production and yield rate are comparatively higher. ● Nanotubes display a better-graphitized structure. ● Less defect compared to the | <ul style="list-style-type: none"> ● Small carbon deposit ● Expensive technique ● Suitable for laboratory production only. | [27]; [28]; [29] |

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|---------------------------|------------------------------------|--|---|--|--|-------------------------|
| | | | | <p>electric arc technique.</p> <ul style="list-style-type: none"> • Nanotube offers high electrical conductivity. | | |
| Chemical vapor deposition | 1993 by Yacaman and his co-workers | <p>In this technique, carbon vapour is formed through hydrocarbons cracking or carbon monoxide as feedstock with supported metal as a catalyst. The process usually carried out at atmospheric pressure in a flow furnace. However, the substrate generally surrounded by catalysts, and heated up to 700 °C. CNTs start</p> | <ul style="list-style-type: none"> • Inert gas: Argon, hydrogen, nitrogen, hydrocarbons (methane, acetylene) • Temperature: 700 °C. | <ul style="list-style-type: none"> • Scalability • Continuous operation • Moderate temperature range. • Structure control technique. | <ul style="list-style-type: none"> • Comparatively more defects • High carbonaceous impurities deposits. | <p>[30]; [31]; [32]</p> |

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|---------------------------------|--|---|---|--|--|---------------------|
| | | to form after inert gas pass through the chamber | | | | |
| Sono-chemical/ Hydro-thermal | 2000 by Yoshimura and his co- workers | The techniques employ synthesis fluid and a tube. Yoshimura and his co-workers used commercial grade SWCNTs in Tuttle-type autoclaves filled with doubled distilled water. Several researchers suggested that various carbon sources can be used to process the techniques. | <ul style="list-style-type: none"> • Temperature: 200 to 800 °C • Pressure: 100 MPa • Duration: 30 min to 48 hr. | <ul style="list-style-type: none"> • Form high-quality CNTs. • The diameter and structure of the tube can be controlled. • Ease operation compared to gaseous processes. • Recycling of solutions is possible. | <ul style="list-style-type: none"> • The synthesis duration is longer. • However, various reactors can be used but it will slow the production. • External energy is required to attain fine particles. | [33]; [34]; [35] |
| Electrolysis | 1995 by WK Hsu | In this technique, alkali/ alkaline earth metals from their chloride | <ul style="list-style-type: none"> • Alkali: Lithium, Potassium, Sodium | <ul style="list-style-type: none"> • Simple apparatus | <ul style="list-style-type: none"> • Produce nanotubes are | [36]; [37]; [38] |

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|---------------|----------------------------------|--|--|--|--|------------|
| | and his co-workers | salts are placed on graphite cathode. Interaction among the metal found with cathode follows the CNTs formation. | <ul style="list-style-type: none"> ● Alkaline Earth: Magnesium, Calcium ● Temperature: 600 °C ● Inert gas: Argon ● Power: 3-20 A° and 20 V | <ul style="list-style-type: none"> ● Economical raw materials ● Consume low energy for electrolysis ● The structure and morphologies of the product can be controlled | curled irregular as well as in bunch. | |
| Solar furnace | 1996 by Heben and his co-workers | This technique focused on sublimating a mixture of catalyst and graphite powder in a vessel in inert gas. | <ul style="list-style-type: none"> ● Inert gas: Argon, Helium ● Temperature: 2627 to 2727 °C | <ul style="list-style-type: none"> ● Solar energy is used as a light. Save power cost ● Potential to maintain each | <ul style="list-style-type: none"> ● Argon as inert gas display poor quality SWCNTs | [39]; [40] |

| | | | | parameter individually | | |
|-----------------|---|---|---|---|--|------------|
| Template method | - | Deposit solid carbon coating achieved from chemical vapour deposition techniques onto porous substrate walls whose pores are parallelly arranged. | <ul style="list-style-type: none"> ● Feedstock: Hydrocarbon ● Substrate: Zeolite, Alumina | <ul style="list-style-type: none"> ● High purity nanotubes produce ● No catalyst remnant ● Produce few carbon phases ● Produce nanotubes structure is open at ends. | <ul style="list-style-type: none"> ● Chemical treatment is a must to recover the nanotubes. ● Produce nanotubes are sensitive due to the open ends ● This technique is not suggested for nanotubes mass production. | [41]; [42] |

As above-mentioned, various approaches can be utilized through which CNTs can be fabricated, but among all laser ablation, chemical vapour deposition and electric-arc discharge techniques are the most popular one, and broadly employed for CNTs' fabrications. One of the most important factor of their popularity is high quality, continuous operation, and scalability of CNTs.

3. Nanocomposites

To take full advantage of CNTs as effective reinforcement for extreme strength composite, CNTs must not create aggregates and completely disperse to improve the interfacial contact with the matrix. In literature, the term nanocomposite was first appeared in 1961 by Blumstein [67]. The term defined as multi-phase solid material where at least one dimension of the incorporation phase is in nano-level (<100 nm) [68]. Presently, polymer composites are known as the prime application field for CNTs. These nanocomposites are considered in a range of fields such as aerospace, sporting goods, automotive, and infrastructure division. Such a wide-broad range of applications of such materials is because of their significant strength and durability, light-weight, and process flexibility. Moreover, CNT/ polymer nanocomposites are considered as electro-magnetic interference protecting material due to this material's high electrical conductivity. Though, effective exploitation of CNTs for producing nanocomposites primarily depends on CNTs' homogenous dispersion throughout the matrix instead of damaging their integrity. Besides, good interfacial bonding is vital to attain extensive load transfer across the CNT matrix interface, a must requirement for enhancing the composites' mechanical properties. Therefore, it is essential to reach a high degree of CNT dispersion throughout processing without causing its property [69].

Nowadays, fabrication techniques of metal/ metal oxides with carbonaceous materials have gained significant attention from many scientific researchers. However, there is minimal information available for metal nanocomposites incorporated with CNTs [70]; [71].

Incorporating carbonaceous material into the metal matrix is relatively complex because of the severe production conditions employed for producing composite materials [72]. The primary challenges to produce metal matrix nanocomposites for commercial applications are the fulfilment of homogeneous dispersion of carbonaceous incorporating materials in a metal matrix, the development of high interfacial bonding, and the retention of carbonaceous materials structure solidity [73]. The following section discusses the various fabrication approaches, properties, and applications of CNTs/ metal nanocomposite and CNTs/ polymer nanocomposite, respectively.

3.1 Fabrication techniques of carbon nanotubes/ polymer nanocomposites

As abovementioned, CNTs have a significant tendency to create aggregates because of their extensive surface area. These aggregates continue until the extreme shear force is provided, e.g., vigorous mixing of the polymer. However, such mixing may cause nanotubes structures, conceding their properties. Thus, the main challenge is to completely disperse individual nanotubes in the matrices to realize the full potential of CNTs. It has proved that CNTs surface modification assists in CNTs dispersion but not recommended for long terms regarding stability. Nonetheless, many techniques have been positively implemented to attain intimate combining of nanotubes with polymer matrices, and solution blending; surface assisted mixing, dry powder mixing, in-situ polymerization, and melt mixing.

3.1.1 Solution processing

This technique is also referred to as solution mixing and often used for CNT/polymer nanocomposites preparation [74]. This technique usually involves the following steps (a). CNTs dispersion in an appropriate solvent (b). Polymer solution mixing (c). recovering the composite through film casting or precipitating [75]. Organic and aqueous medium have been used to fabricate CNT/polymer nanocomposites [76]; [77]. The complications in diffusing uncontaminated CNTs insolvent through simple stirring may need the use of high-power ultra-

sonication to offer metastable CNTs suspension [78]. Apart from using high-power ultra-sonication, several other approaches for CNTs dispersion, such as magnetic stirring, reflux, and shear mixing. To improve CNTs dispersion, acid-treated, modified CNTs, and heat-treated techniques are usually used [79]; [80]; [81].

The extended use of high-power ultra-sonication may affect the CNTs' length, i.e. lower the aspect ratio, which is unfavourable to composite properties [82]. To reduce this issue, surfactants have been considered to disperse extensive loadings of CNTs [83]; [84]. Whereas in solvent blending, the slow evaporation stage often followed by nanotubes aggregation. It can be overcome through CNT/ polymer suspension on a rotating substrate or fall on a hot substrate to accelerate the evaporation process [85]. Moreover, coagulation is another technique involving CNT/ polymer suspension pouring into an additional non-solvent [86]. It followed SWCNTs entrapment through precipitating polymers chains which in-turn protects the bundling of SWCNTs. This approach is practical in terms of poly-ethylene and PMMA nanocomposites [87]. Several CNT/ polymer nanocomposites have been produced successfully through solution mixing techniques, including MWCNT/poly-ethylene oxide, SWCNT/poly-methyl-methacrylate, and MWCNT/poly-Lactic acid [88]; [89]. Thus, the solution mixing technique is restricted to polymers that readily dissolve in solvents appropriate and followed to CNTs stable suspension.

3.1.2 Reaction processing

This technique is also known as in-situ polymerization and is the most efficient approach to improve CNTs dispersion and interaction among polymer matrix and CNTs, significantly [90]; [91]. The technique pact with the dispersion of CNTs into monomers in the absence/ presence of a solvent leading standard techniques of polymerization [92]. Compared to other techniques, this technique used monomers as initial materials, not polymers. However, reaction processing generally involves polymers that are insoluble, thermally unstable, and cannot be treated

through solution mixing and melts processing. Various researches have been conducted on MWCNT/ poly-urethane, MWCNTs/nylon, MWCNT/polystyrene, etc. [93]; [94]. Moreover, in-situ polymerization techniques aid covalent bonding among polymer matrix and functionalized CNTs through various chemical reactions. A few examples include MWCNTS/ polyimides composites prepared through the reaction of pyro-mellitic dianhydride and 4,4'-oxydianiline; MWCNTs/polyaniline composites fabricated through chemical oxidative polymerization and MWCNTs/poly-pyrrole were synthesized through in-situ chemical polymerization and in-situ inverse microemulsion [95]; [96]. Furthermore, many kinds of researches have also been carried out on epoxy-based nanocomposites [97]; [98].

3.1.3 Melt processing

This technique is also known as melt blending and is considered the most convenient, economic, solvent-free, rapid production, and environmentally friendly technique to produce CNT/ polymer nanocomposite [99]. Melt blending deal with thermoplastic polymers, such as polycarbonate, polystyrene, polypropylene, etc. [100]; [101]; [87] In this technique, polymer melt to produce viscous liquid led through mixing with CNTs. However, CNTs dispersion can be upgraded through shear mixing, which can be attained through injection molding and extrusion approach [102]; [74]. Compared to the solution mixing technique, melt blending is measured as less efficient due to thermoplastic polymers high viscosity, which hindrance the attaining uniform CNTs dispersion, and its use restricted to low filler concentrations in thermoplastic matrices. Several studies used fiber-spinning, which creates extended fiber specimens [103]; [104]. Regarding industrial application, the melt blending technique is recommended due to its economical cost and ease of operation for higher scale production for commercial use.

Although the melt processing technique is convenient, high temperature and shear force can weaken nanocomposite property, as the high shear force needed to attain CNT dispersion

may follow to fragmentation of CNT. So high shear stress is needed to reach desired dispersion at the lowermost possible CNTs damage. Furthermore, high-temperature use is also severe, as high temperature develops CNT dispersion through lowering the viscosity; thus, extreme high temperature can damage the polymer un-contaminated properties. Therefore, temperature optimization is essential. Several modifications in melt compounding have been developed to overcome these issues, for instance, combining the solution and melt processing by subjecting solvent cast SWCNT/ polymer film to numerous sequences of melt processing [105]. Nevertheless, optimization of processing states is a pressing issue, for nanotubes types and the wide range of nanotube-polymer combination [106]. MWCNT/ poly-carbonate, SWCNT/ poly-imide, and MWCNT/ nylon-6 nanocomposite are a few of the successful instances of melt processing [92].

3.1.4 Latex technology

Comparatively, there is a new technique to reinforce CNTs into the polymer matrix, and the technology is well-known as latex technology [74]. The technique is colloidal distinct polymer particle dispersion, generally in the aqueous phase. This technology is convenient for CNTs to disperse in almost all the polymers generated via emulsion polymerization or taken into the form of an emulsion. Compared to in-situ polymerization, the CNTs addition in this technology is conducted after the synthesis of polymers. The technology involves the following steps (a) Dispersion (MWCNTs entanglements) or exfoliation (SWCNTs bundles) of CNTs in aqueous surfactant solution, (b) Stir the stable dispersion of surfactant-treated CNTs with polymer latex, and (c) Freeze-drying and followed by melt blending approach to attain nanocomposite [107]. The operation ease, reliable, permit to reinforce individual CNTs into a very viscous polymer matrix, reproducible and versatile are a few of the merits of latex technology. Water is used as a solvent for CNTs dispersion; therefore, the technology is eco-friendly, safe, and economical. Currently, polymer latex is commercially generated on a large scale, and this manufacturing is

matured [108]; [109]. Apart from all other techniques, this technique is easy; therefore, the vision for scale-up production of CNTs/ polymer nanocomposite is bright through this technology.

3.1.5 Miscellaneous processing

In recent years, several new processing techniques have been designed to produce CNT/polymer nanocomposite with a substantial CNT content or for specific applications, such as the spinning of coagulant, pulverization, densification, and layer by layer deposition. All these techniques along with their features are summarized in Table 2. Without any doubt, nanocomposites components are developing field, numerous researches are being conducted to plan new processing techniques that can fabricate nanocomposites with distinct properties and structure for specialty end applications.

Table 2: Miscellaneous processes for CNT/polymer nanocomposite production [110]; [111]; [112]; [113]

| Processing Technique | Methodology | Carbon Nanotubes | Merits |
|-----------------------------|---|--|---|
| Spinning of coagulant | CNT coagulation into mesh through wet spinning and changing the mesh into solid fiber via slow draw method. | Preliminary dispersion of CNT with a surfactant solution | Active to produce CNT/polymer fiber |
| Pulverization | CNTs and polymer are combined and pulverized using twin-screw/ pan mill | CNTs (As-received) | Polymers possible grafting on CNTs, high scale-up, solvent-free |

| | | | |
|---------------------------|--|---|--|
| Densification | CNT forest is synthesized and carried to an uncured epoxy pool. The matrix is infused into the CNT forest and later cured. | CNT forest | Vol.% of CNT can be maintained through changing CNT forest densification, aligned CNTs in nanocomposites |
| Layer by layer deposition | Solid substrate i.e. silicon wafers, glass slides, are dipped into CNT/polymer solutions and subsequent curing. | Preliminary dispersion of CNT insolvent | Significant loading of CNT (max. 50 wt.%), structural defects made from CNT/polymer phase segregation can be lessened. |

Up to date scientific community has explored several techniques for the development of CNT/ polymer nanocomposites, but mainly employed techniques with industrial feasibility are melt and solution processing. The primary reason of their fame is the cost and simplicity comparing to others designed techniques. Due to these fact, most of the researches have been performed using these techniques.

3.2 Properties of CNTs/polymer nanocomposites

Generally, substantial increases in features like modulus and strength can be attained via CNTs reinforcement in the polymer matrix. For instance, 1 wt. % of MWCNTs addition to polystyrene/ MWCNTs nanocomposite films results in an enhancement in break stress and tensile strength, i.e. 25% and 40-42%, respectively [114]. Nevertheless, modulus and strength were improved as a result of incorporation with CNTs, the impact toughness of nanocomposites was decreased, although few studies reflected a noticeable enhancement in toughness. Another study in which 1 wt.% CNTs were incorporated with polyethylene and increase ductility and toughness of 104 and 150%, respectively [115].

The enhancement of the thermal strength of polymeric nanocomposites is very desirable. The CNTs addition allows an upgrade in the glass transition temperature (T_g) and melting and thermal decomposition temperatures of the polymeric matrix because of the constraint effect on polymer segments as well as chains. Furthermore, surfactant addition also helps to increase the T_g , for instance, 1 wt.% CNTs to epoxy. Epoxy thermal conductivity upgraded up to 300% with 3 wt.% SWCTNs addition, reported [116].

Furthermore, electrical conduction in polymer nanocomposites has also been studied, extensively, particularly for industrial applications. Thus, their consideration in polymer/ CNTs nanocomposites, supercapacitors have several potential applications in electro-magnetic interference shielding (EMI), electronic packaging, and wave adsorption. Also, CNTs/ polymer nanocomposites also can be used for organic photovoltaic devices, optical sensors, optical elements, and optical switching [117]. Tables 3 and 4 listed the mechanical and electrical strength of various CNTs/ polymers nanocomposites that have been studied recently.

Table 3: Mechanical properties of CNT/polymer nanocomposites

| Carbon Nanotube | Polymer | Fabrication Technique | Testing | Toughness (% rise) | Young's Modulus (% rise) | Storage Modulus (% rise) | Tensile Strength (% rise) | References |
|-------------------------|-----------------------------|------------------------------|----------------|---------------------------|---------------------------------|---------------------------------|----------------------------------|-------------------|
| MWCNTs (Refined) | Poly-styrene | Solution Processing | Tensile | - | 42 | - | 25 | [104] |
| | | Melt Processing | | - | 100 | 100 | 10 | [118] |
| SWCNTs (grafting to) | | Solution Processing | | - | 78 | - | 82 | [119] |
| MWCNTs (Uncontaminated) | Poly-ethylene (Low density) | Melt Processing | | - | 89 | - | 56 | [120] |
| MWCNTs (Refined) | | High Energy Ball Milling | | 100 | 30 | - | 150 | [121] |

| | | | | | | | | |
|--------------------------------|--------------------------|-------------------------|--------------|----|-----|----|-------|-------|
| MWCNTs (Poly-ethylene grafted) | | Melt Processing | | 60 | 65 | - | 30 | [122] |
| MWCNTs | Poly-Propylene | Pulverization Technique | | - | 15 | - | 20 | [112] |
| SWCNTs (Refined) | | Solution Processing | DMA | - | - | 15 | - | [123] |
| | | | Tensile | - | 200 | - | 150 | [124] |
| SWCNTs (Uncontaminated) | | Melt Processing | DMA | - | 40 | 60 | 15 | [125] |
| | | - | | - | 75 | - | [126] | |
| SWCNTs (Functionalized) | Parmax | Solution Processing | Tensile | - | 28 | - | 72 | [127] |
| MWCNTs (Functionalized) | Poly-methyl-methacrylate | Reaction Processing | Tensile, DMA | 75 | - | 88 | 75 | [128] |
| MWCNTs (Uncontaminated) | Poly-carbonate | Solution Processing | Tensile | - | 46 | - | 47 | [129] |
| SWCNTs (Functionalized) | Poly-vinyl-alcohol | | | - | 79 | - | 47 | [130] |

| | | | | | | | | |
|----------------------------|-------------------------|------------------------|-----|---|---|----|---|-------|
| MWCNTs (Uncontaminated) | Poly-hexyl- thiopene | Reaction Processing | DMA | - | - | 47 | - | [131] |
|----------------------------|-------------------------|------------------------|-----|---|---|----|---|-------|

Table 4: Electrical properties of CNT/polymer nanocomposites

| Carbon Nanotubes | Polymer | Fabrication Technique | Percolation Threshold Φ_c (wt.%) | Polymer Electric Conductivity (Sm^{-1}) | Carbon Nanotubes Electric Conductivity (Sm^{-1}) | Nanocomposite Electric Conductivity (Sm^{-1}) | References |
|----------------------------|--------------------------|------------------------------|---|--|---|--|-------------------|
| SWCNTs | Poly-styrene | Solution Processing | 0.27 | $\sim 10^{-14}$ | - | $\sim 10^{-3}$ | [80] |
| SWCNTs (Uncontaminated) | | | 0.45 | $\sim 10^{-17}$ | - | $\sim 10^{-1}$ | [132] |
| MWCNTs | Poly-ethylene | Melt Processing | ~ 0.75 | $\sim 10^{-18}$ | - | $\sim 10^{-3}$ | [133] |
| | Poly-methyl-methacrylate | Solution Processing | 0.003 | $< 10^{-7}$ | - | $\sim 3 \times 10^3$ | [134] |

| | | | | | | | |
|----------------------------|-----------------------------|------------------------|----------|--------------------------|----------------------|-------------------|-------|
| SWCNTs (Functionalized) | Poly-carbonate | | 0.11 | 10^{-13} | 5×10^4 | 4.8×10^2 | [135] |
| MWCNTs | Poly-vinyl alcohol | Melt Processing | 1-1.5 | 5×10^{-14} | - | 10 | [136] |
| | | Solution Mixing | 0.72 | $\sim 10^{-10}$ | - | 0.27 | [137] |
| SWCNTs | Poly-ethylene- imine | | - | - | - | 2×10^4 | [138] |
| MWCNTs | Polyaniline | Reaction Processing | - | 2.6×10^{-1} | 23 | 6.6 | [139] |
| SWCNTs | | Solution | - | - | - | 2×10^3 | [140] |
| MWCNTs (Uncontaminated) | Poly-3- hexylthiopene | Processing | 10.62 | 10^{-4} | - | 7×10^1 | [141] |
| SWCNTs | Poly-ethyl- methacrylate | Reaction Processing | ~ 3 | $\sim 2 \times 10^{-10}$ | $\sim 3 \times 10^4$ | ~ 0.4 | [142] |

| | | | | | | | |
|--------|---------------------------|---------------------|-------|-----------|---|-----------|-------|
| MWCNTs | Nylon 6 | Melt Processing | 2~2.5 | - | - | 0.1 | [143] |
| | Poly-m-phenylene-vinylene | Solution Processing | - | 10^{-1} | - | 10^{-2} | [144] |

3.3 Processing techniques of carbon nanotubes/metal nanocomposites

Carbon nanotubes incorporated in a metal matrix are fabricated by several approaches that have been employed for the fabrication of CNTs/ metal nanocomposites. Powder metallurgy is well-known and broadly used technique for the development of CNTs/ metal nanocomposites. Apart from the powder metallurgy process, electroless and electro-deposition are also known and used techniques for the deposition of thin-coating of metal matrix/ CNTs, moreover, metals deposition onto CNTs. However, regarding low-melting-point metals, for instance, magnesium and bulk metallic glasses, solidification, and melting is a practicable path. Furthermore, scattered efforts have also been done on indigenous techniques for the processing of CNTs/ metal nanocomposites. In the section below, briefly described various processing techniques of CNTs/ metal nanocomposites:

3.3.1 Thermal spray

The term thermal spray is described as spraying of molten/ semi-molten particles onto the substrate to form a deposit or coating through solidification and impact pathway. The thermal spray approach comes with numerous benefits such as a large cooling rate during solidification i.e. 10^8 K/s, which usually results in the retention or production of the nanocrystalline matrix in the coating [145]. In terms of heat source, thermal spray methods can be categorized into plasma spraying, flame spraying, cold spraying, or high-velocity oxy-fuel spraying. In plasma spraying, the source of heat is plasma that is produced through inert gas ionization through an arc hit between the copper cathode and tungsten cathode. Powders withdrew into the plasma, absorb the heat, and gain kinetic energy as well as projected at considerable velocities onto the substrate to create the coating. The velocities of the particle can be high as 100 m/s [146]; [147]. Whereas in high-velocity oxy-fuel spraying, the heat source is high-pressure fuel oxygen mixture combustion. The fuel can be liquid or gas, for instance, propane, hydrogen, and kerosene. The particle velocities are also extremely higher, up to 1500 m/s, following the

production of dense coating [148]; [148]. Another merit of thermal spraying is that it can be used for near-net shaping of majority nanocrystalline elements.

Based on the literature, very limited studies on thermal spraying using cold spraying, high-velocity oxy-fuel spraying and plasma spraying of carbon nanotubes incorporated with metal have been conducted.

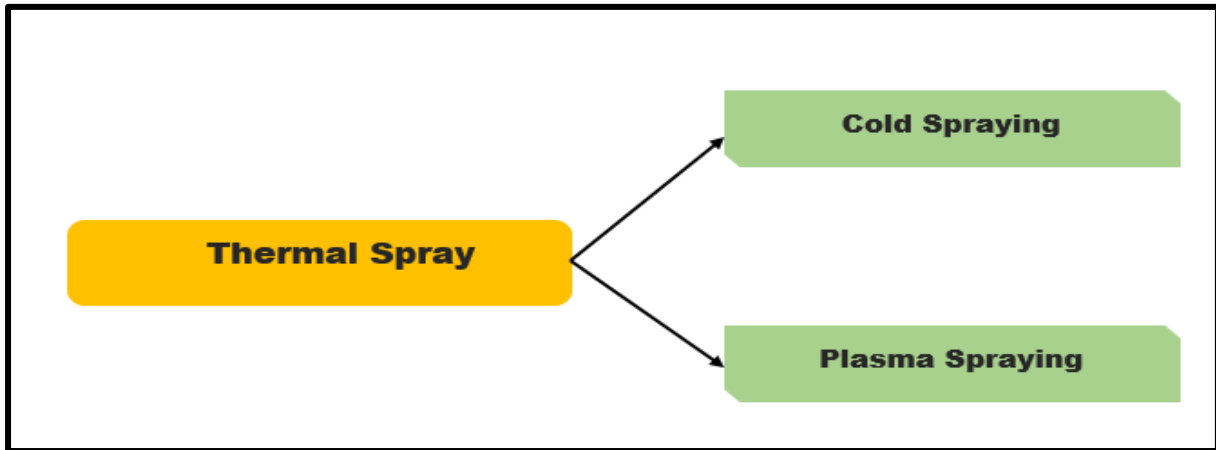


Figure 2: Carbon nanotubes/metal nanocomposites processing routes using thermal spray

3.3.2 Powder metallurgy

Studies related to aluminum-carbon nanotubes and around half of the research study on CNTs incorporated with copper have been conducted through the powder metallurgy technique. However, several studies have also used CNTs with magnesium (Mg), silver (Ag), titanium (Ti), nickel (Ni), and inter-metallics via this technique [149]. The general technique steps include CNTs mixing with metal powder using mechanical or grinding alloying, led by consolidation through sintering and compaction, cold and hot isostatic pressing, or spark plasma sintering. Particularly, composite compacts are exposed to post sintering deformation methods, such as equi-channel angular processing and rolling [150]. Regardless of process stages, the primary focus is on attaining better reinforcement through CNTs' homogeneous dispersion in metal matrix and bonding at CNT/ metal interface.

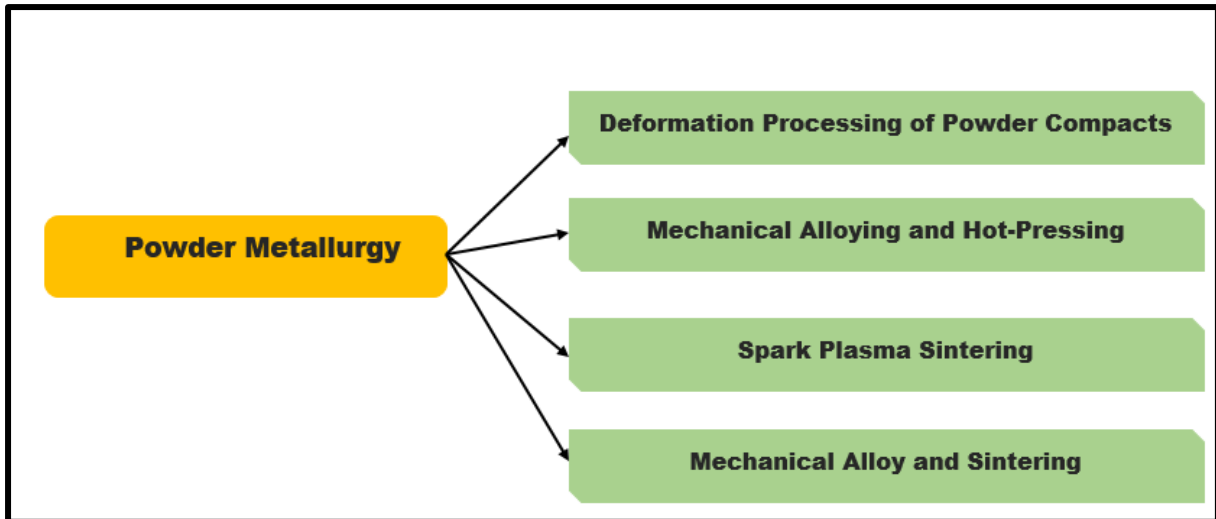


Figure 3: Carbon nanotubes/metal nanocomposites processing routes using powder metallurgy

3.3.3 Electro-chemical deposition

After the powder metallurgy technique, electrochemical deposition is the second well-known path for CNTs-metals, based on literature. The primary difference among the two methods is that the electrochemical deposition method is mainly used for the development of thin composite coatings with a thickness of around 20~180 μm , however, some studies do not state the thickness of coating [151]. Furthermore, this method has been applied for coating CNTs with metals to form 1-D composites, for applications such as electrodes, magnetic recorders, and nano-sensors. Electrochemical deposition is categorized into electroless deposition and electro-deposition, both have been used for the fabrication of CNTs/ metal matrix. Electro-deposition follows the conventional electrochemical cells in which composite film is placed through current-flow between anode and cathode [152]. However, the electroless technique does not need any external source of energy, it is generally a chemical process in which thermo-chemical decomposition of metallic salts is carried out in a bath to exit metallic ions that produce a composite with CNTs.

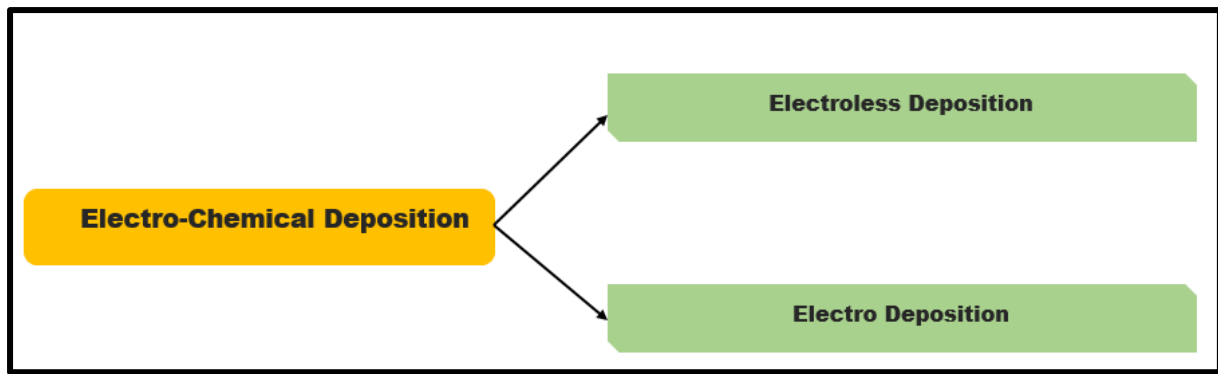


Figure 4: Carbon nanotubes/metal nanocomposites processing routes using electrochemical deposition

3.3.4 Melting and solidification

It is one of the most conventional fabrication methods for metal matrix composites, but have also been used for the CNTs incorporated composites production too. Several studies have been carried out that use the melting and solidification pathway for CNTs/ metal composites preparation due to the need for extreme temperature for melting. The method may show few disadvantages such as CNTs' damage and development of chemical reaction products at the interface of CNTs/ metal [153]; [154]. Thus, this pathway is recommended for composites with a low melting-point matrix. Another demerit is that suspended CNTs may form a cluster because of surface tension forces.

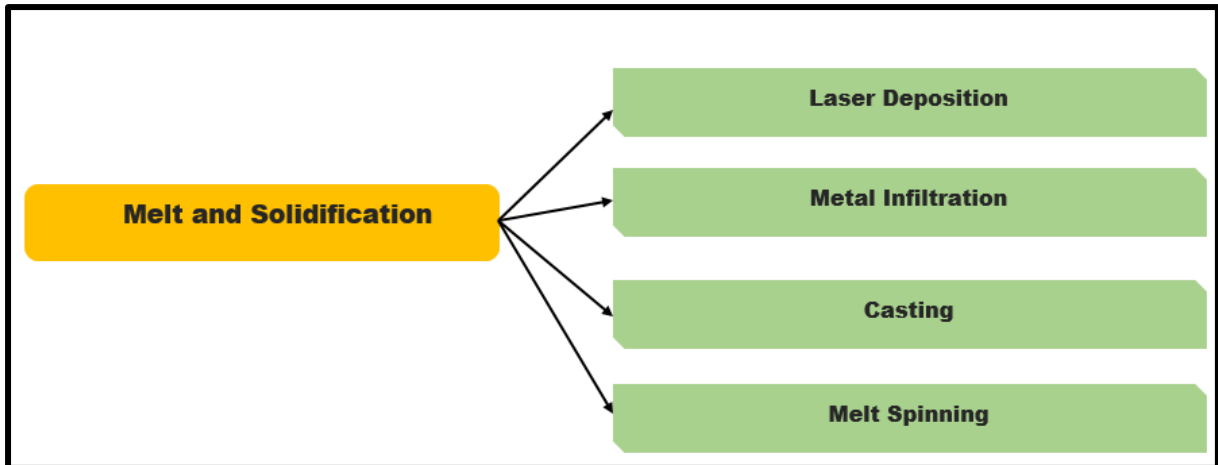


Figure 5: Carbon nanotubes/ metal nanocomposites processing routes using melt and solidification

3.3.5 Other methods

Many researchers have discovered exceptional fabrication pathways. Few of these pathways are upgraded from the traditional methods, however, others are original and novel methods that produce CNTs/ metal nanocomposites.

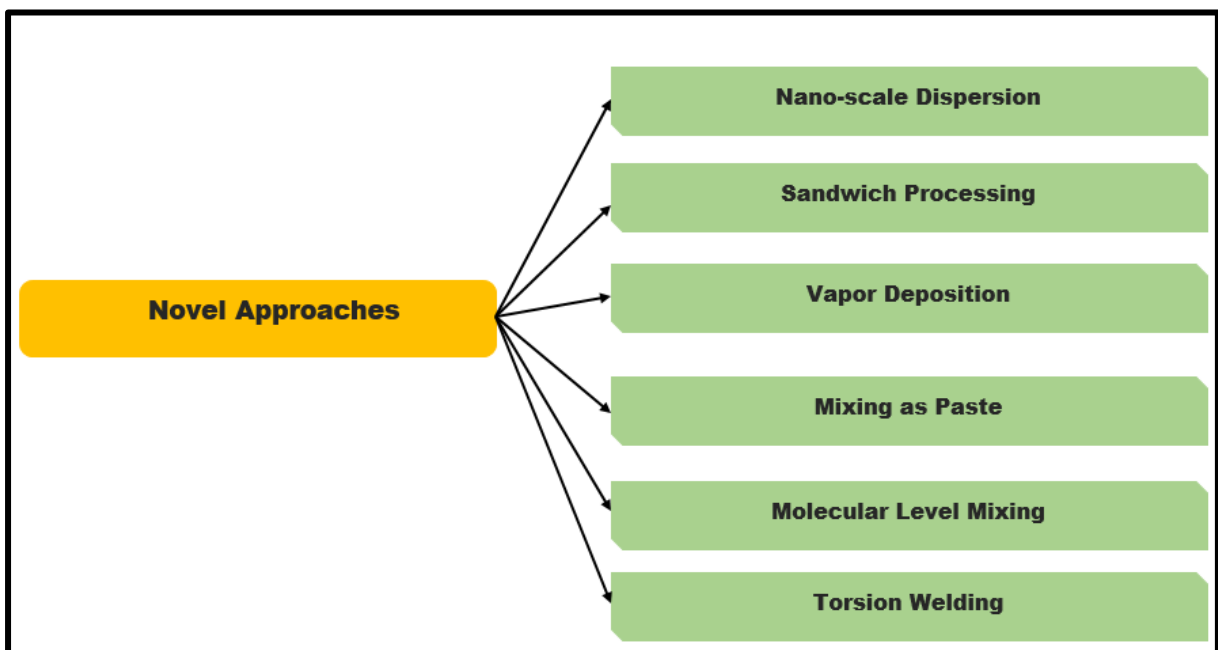


Figure 6: Carbon nanotubes/metal nanocomposites processing routes using novel approaches

Table 5: Advantages and disadvantages of fabrication techniques of CNTs/metal nanocomposites

| Processing techniques | Advantages | Disadvantages | Other routes | References |
|-----------------------|--|--|---|--|
| Thermal spray | <ul style="list-style-type: none"> ● The technique is applicable for a broad range of materials that can be deposited as coating, such as metals, oxides, alloys, refractory metals, plastics, and ceramics, moreover, the abovementioned materials in combine form. ● Coating deposition and spray rates are comparatively cheaper in terms of processing costs than other available techniques. ● Coating thickness can be varied and controlled (5 μm to 6.5 mm). ● An appropriate range of applications, such as electrical conductivity and resistance, metal composite structures, net, and near-net-shape | <ul style="list-style-type: none"> ● In contrast to other coating techniques, tensile bond strength is low for the thermal spray technique. ● Coating through the thermal spray technique is anisotropic. ● The technique is typically not considered as structural members as well as do not perform satisfactory line or under point loading. | <ul style="list-style-type: none"> ● Plasma spraying ● Cold spraying ● High-velocity Oxy-fuel spraying ● Flame spraying | <p>[155];</p> <p>[156];</p> <p>[157];</p> <p>[158]</p> |

| | | | | |
|-------------------|---|---|--|---------------------------|
| | <p>element production, and abradable coatings for gas turbine engines.</p> <ul style="list-style-type: none"> ● Equipment cost is reasonably low. ● Negligible thermal degradation to a substrate is possible while spraying | | | |
| Powder metallurgy | <ul style="list-style-type: none"> ● One of the oldest techniques started in 2500 B.C. by Egyptians. ● Composite made using this technique required ease processing as well as limited machining ● Materials produced using this technique do not come with any contaminants, gas spots, or internal stresses. ● The cost-effective technique, moreover, product dimensions can be controlled with satisfactory physical and mechanical properties. | <ul style="list-style-type: none"> ● Expensive equipment cost. ● Certain design demerits. ● Higher raw material costs. ● Not appropriate for low melting point metals ● Comprehend uniform dense products cannot be formed. ● Product size is restricted due to press capacity. | <ul style="list-style-type: none"> ● Spark Plasma Sintering ● Deformation processing of powder compacts ● Mechanical alloying and sintering | <p>[159]; [160]; [15]</p> |

| | | | | |
|----------------------------|---|---|--|---------------------------|
| | <ul style="list-style-type: none"> • No secondary or finishing steps. • Minimum material loss. • For CNTs/metal nanocomposite, the powder metallurgy technique can be modified such as the rolling technique. | | <ul style="list-style-type: none"> • Mechanical/ mixing alloying and hot pressing | |
| Electrochemical deposition | <ul style="list-style-type: none"> • Can maintain a thickness deposit. • Fair deposit distribution • The processing cost is comparatively economical. • Deposits less porous, at alike thickness. • Various finishes are possible. | <ul style="list-style-type: none"> • Fewer metals can be deposited from the aqueous phase. • Limited deposition rate i.e. 20 to 100 $\mu\text{m/h}$. • Pre-treatment is a must for few substrates • Practically, a limited range of thickness. | <ul style="list-style-type: none"> • Electro-less deposition. • Electro-deposition | [161]; [162]; [163] |
| Melting and solidification | <ul style="list-style-type: none"> • The technique is suggested for a low melting point matrix such as Al, Mg, etc. • The distribution of CNTs can be controlled. | <ul style="list-style-type: none"> • In literature, limited researchers have employed this technique for CNTs/ metal nanocomposites • Melting stages required extremely high temperature | | [164]; [165] |

| | | | | |
|--|--|---|--|--|
| | | <ul style="list-style-type: none">● High-temperature damages CNTs as well as undesired compounds in the nanocomposites.● Maintaining the solidification stage, directly cause the mechanical strength of the nanocomposites.● The technique itself required one or more stages to obtain the final product. | | |
|--|--|---|--|--|

The key to attaining the desired incorporation effect from nanocomposite lies in the satisfactory reinforcement of CNTs into metal matrix. Incorporating CNT in metal matrix is extremely challenging task. Because when the concentration of CNT is greater than a few wt.%, substantial declination in expected material features. Among all the above-mentioned fabrication techniques, powder metallurgy is the well-known and best due to the cost and mass production in comparison to others techniques.

3.4 Properties of CNTs/metal nanocomposites

Composite components generally display considerable benefits and properties in-contrast to monolithic components. Mono-lithic components and their alloys cannot comprehensively maintain the demands of current developed technologies and performance necessities. It means that through incorporating reinforcements into a such as polymer/ metallic/ ceramic matrix, the matrix properties upgrade in terms of significant mechanical strength, temperature stability, and chemical durability. The availability of reinforcing materials can lift the chemical and physical structure extensively.

As stated earlier, CNTs' discovery promotes electrifying openings for emerging applications in the utmost areas of science and engineering. A number of researches have been carried out that emphasize CNT-based composites properties. Carbon nanotubes incorporation with the metal matrix is likely to assist in attaining lightweight structural components with improved thermal, mechanical, and electrical properties.

3.4.1 Thermal properties

Nanotubes are mainly known to hold high thermal conductivity and extremely low coefficient of thermal expansion i.e. 1812 ± 300 W/mK and ~ 0 , respectively [166]. Thus, CNTs/ metal nanocomposites possess significant potential to be cast-off for thermal management. An extensive declined in the coefficient of thermal management (CTE) of 63% was reported by

Tang and his co-workers with the use of 15% CNTs/Al (vol.). Tang's experiment also concluded that an increase in CTE is directly proportional to CNTs content [167]. Another work done by Deng and his co-workers stated a reduction of 12% CTE through 1.28% CNTs/Al (vol.), which has been credited to CNTs large surface area that offers extensive interface, and therefore, limits metal matrix thermal expansion [168]. Studies conducted by Goh and his co-workers have also stated a massive drop of 9% CTE with 0-30 CNTs/Mg (wt.%) [169]. Moreover, 0.7% CNTs/ Ni (wt.%) nanocomposite show an increase in thermal conductivity of 200%, mainly due to electro-deposition technique that offers excellent dispersion as well as CNTs bonding that produce defect-free interface with the matrix.

The enhancement in thermal strength of CNTs/metal nanocomposites significantly depends on CNTs' distribution and its bonding with the matrix. Hence, the content of CNT and processing pathways are essential factors that determine CNTs/metal nanocomposites' thermal properties.

3.4.2 Electrical properties

Owing to remarkable electrical properties, showed by current-carrying density of approximately 4×10^9 A/cm², CNTs have been used as strengthening to metals for improvement of electrical properties [170]. 12.5% CNTs/Al (vol.%) fabricated using powder metallurgy showed increased electrical conductivity by 66%. [171] A study conducted lately reported no change in the electrical resistivity up to the addition of 10% SWCNTs (wt.%) to CNTs/ Cu nanocomposite [172]. Whereas, research conducted by Feng and his co-workers notice a marginal increase in electrical resistivity when 10% CNTs (vol.%) added to CNT/Ag nanocomposite. The sharp increase in electrical strength beyond 10% CNTs (vol.%) is credited to incline in strain and interfacial area in the matrix because of the CNT cluster existence, both of which delays electron transfer via the composite [173].

Several study groups have researched the CNTs/metal nanocomposites suitability in various electrical applications, for instance, CNT/Ni nanocomposites have displayed good potential to be used as field-emission display, screening uniform images with better-quality. Another research work has shown the use of CNT/ Si as anode materials for lithium-ion batteries [174].

3.4.3 Corrosion property

Researches regarding corrosion studies are mostly performed on electro-deposited CNT/ Ni composite coatings with only one study on CNT/Zn composite coating [175]; [176]. Because of the existence of voids and pores, electro-deposited coatings are susceptible to corrosion. Most of the researchers have witnessed an incline in corrosion property of the nanocomposite coatings with the addition of CNTs. An increase of 24% reported by Yang and his co-workers with the addition of 5%CNTs (wt.%), while a 75% increase reported by Chen and his co-workers without CNT content mentioned [177]. Furthermore, his research has also determined the corrosion rate of nanocomposite and concluded 5 times lower than coating with nickel (Ni). All these research studies have shown that enhancement in corrosion resistance mainly from CNTs' chemical inertness that offers a passive layer on the coating surface, and filling of pores and voids of electro-deposited coatings leaving no space for the opening of localized corrosion [178].

3.4.4 Friction property

Wear properties are serious for coating and thus, wear researches are mostly on CNTs/Ni composite coating prepared through electro-deposition methods [179]; [179]. There is limited research on CNTs/Al and CNTs/Cu nanocomposites [180]. The outcomes of most of the research studies have shown a decrease and increase in the coefficient of friction (COF) and wear resistance, respectively, when CNTs are embedded in the metal matrix. The COF decrease has been credited to MWCNTs' lubricating nature produced by convenient descending of their

walls, which are connected through weak van der Waals forces. The better resistance to wear is due to the CNTs role as spacer preventing the matrix rough surface from contacting with wear pin [181].

Studies carried out by Tu and his co-workers have stated extreme enhancement of wear strength in CNT/Cu nanocomposite prepared using powder metallurgy method [182]. Their work has shown a reduction in COF and wears rate of 91% and 140%, respectively, with 16% CNTs (vol.%) [183]. Moreover, the molecular level mixing method has also assisted in upgrading CNTs/Cu wear properties with the addition of 10% CNTs (vol.%) [184]. The primary factor for the enhancement in nanocomposite wear properties is homogeneous CNT dispersion. CNT incorporation plays a key role in the advancement of wear resistance and decline in COF of metal matrix [164]. CNTs effect on wear strength of CNT/Ni has been researched broadly, but still required further studies for other CNT/metal systems to optimize the association between the content of CNT and its wear strength.

3.4.5 Mechanical properties

The serious issues in mechanical properties in CNTs/metal nanocomposites are the homogeneous distribution of CNTs in the metal matrix as well as the bonding and interfacial reaction to the matrix, to act excellently as reinforcement. Due to this fact, various fabrication techniques were abovementioned critically discussed regarding this phenomenon. Table 8 listed the tensile property of various nanocomposites that have been researched by different researchers or group of researchers in the past.

Table 6: Tensile properties of CNTs/metal nanocomposites from experimental studies

| Nanocomposites | Fabrication technique | Technique description | Young's Modulus (GPa) | Tensile Strength (MPa) | Yield Strength (MPa) | Fracture Strain (%) | References |
|----------------|--|-----------------------|-----------------------|------------------------|----------------------|---------------------|------------|
| MWCNTs/Al | Dry powder mixing, hot pressing and hot extrusion | - | - | 89 | - | 41 | [185] |
| | Ball milling, hot pressing | 300 rpm, 6 hr. | - | 170 | 104 | 14 | [186] |
| | High energy milling, vacuum sintering, and hot extrusion | 5 hr. | - | 159 | 105 | - | [187] |
| | Ball milling, hot rolling | 500 rpm | 70.02 | 291 | 262 | 13.5 | [188] |
| | | | 95.15 | 492 | 483 | 4 | |
| MWCNTs/Mg | | - | - | 194 | 128 | 12.7 | [154] |

| | | | | | | | |
|-----------|---|--------|------|-----|-----|------|-------|
| | Melt Stirring, Melt-deposition and hot extrusion | | - | 210 | 140 | 13.5 | |
| | Powder Mixing, cold compaction, sintering, and extrusion | 10 hr. | - | 203 | 133 | 12 | [169] |
| | Ball milling, melt infiltration and casting | - | 60 | 160 | - | 3.74 | [189] |
| MWCNTs/Cu | Spark plasma sintering | - | 51.6 | - | 119 | - | [190] |
| | | | 80 | - | 220 | - | |
| | Wet powder mixing, and hot extrusion | - | - | 206 | - | - | [191] |

3.5 Applications of carbon nanotubes-based nanocomposites

Carbon nanotubes incorporation with polymer/ceramics/metal allows the development of innovative nanocomposites with upgraded or even novel sets of thermal, electrical, electro-chemical, electromagnetic, and mechanical properties. Therefore, they are identified to demonstrate numerous dependent applications, which are discussed in detail in the following section:

3.5.1 Supercapacitors

Recently, supercapacitors have attained significant attention within energy storage devices because of their rapid charging/ discharging, high specific power as well as longer life-span. In terms of a charge storage system, supercapacitors are classified as an electrical double-layer capacitors, hybrid supercapacitors and pseudo-capacitor [192]. MWCNTs, activated carbon, and graphene oxide are well-known carbon-based materials used as an electrode for an electrical double-layer capacitor. Whereas, materials such as conducting polymers and metal oxides are more often used as an electrode for pseudo-capacitors. The electrochemical performance of electrodes can be upgraded through hybrid nanocomposites, where electrodes store the charge, hence developing extensive specific capacitance and specific energy [193]; [194]; [195].

Polypyrrole (PPy), poly-aniline (PANI), and poly-thiophene (PTh) are conducting polymers that are referred to as superior electrode materials. Among different conducting polymers, polypyrrole is more well-known because of its better redox quality, high conductivity, cost-effective, environmental stability, and non-toxicity. However, polypyrrole's cyclic stability is less due to repetitive ionic exchanges in the polymer matrix during electrochemical cycles that occur to amend polypyrrole structural confirmation [196]. Carbon nanotubes, particularly MWCNTs and SWCNTs are widely investigated in supercapacitors application as this electrode possesses excellent chemical stability and conductivity. MWCNTs

has the potential to offer a conducting route during charge and discharge, thus developing a fast-electro-chemical kinetic process [197]. In contrast to MWCNTs, SWCNTs have been recognized as high-end CNTs technology due to its unique properties such as fewer defect density, high surface area, and better electronic features. Due to its outstanding properties, SWCNTs are considered the superior electrode materials for supercapacitors [198]. Currently, the production of polypyrrole/ carbon nanocomposite as supercapacitor electrodes have the potential to improve the electro-capacitive performance. Different studies have been carried out on the production of CNTs nanocomposites as an electrode component through various strategies for supercapacitors are described in Table 9,

Table 7: Fabrication of supercapacitors through various carbon-based nanocomposites

| Carbon Nanotubes | Metal/Metal Oxide | Conducting Polymer | Processing Techniques | Specific Capacitance (Fg⁻¹) | Power Density (kW.kg⁻¹) | Energy Density (Wh. kg⁻¹) | References |
|-------------------------|--------------------------|---------------------------|-----------------------------------|---|---|---|-------------------|
| SWCNTs | MnO ₂ | PPy | Reaction Processing | 351 | 10 | 39.7 | [199] |
| MWCNTs | - | PANI | Polymerization | 1566 | 125 | 217 | [200] |
| f-MWCNTs | - | PANI | Chemical Oxidative Polymerization | 381 | - | - | [201] |
| Carbon nano-foam | - | PEDOT:PSS | Electro-Polymerization | 210.8 | 19.7 | 8.53 | [202] |
| MWCNTs | Au | PANI | Chemical Polymerization | 528 | - | - | [203] |
| MWCNTs | MnO ₂ | PEDOT: PSS | Electro-chemical Deposition | 428.2 | 10 | 63.8 | [204] |
| CNTs-Gr | - | PPy | Reaction Processing | 453 | 0.566 | 62.96 | [205] |
| SWCNTs-Gr | - | PPy | Reaction Processing | 324 | 1.975 | 28.8 | [206] |
| MWCNTs-GO | - | PPy | Electro-Polymerization | 358.69 | 0.441 | 40.45 | [207] |
| MWCNTs | Cu | Poly (An-co-Py) | Chemical Polymerization | 383 | 1.546 | 53.19 | [208] |
| MWCNTs | Ni | - | Electrochemical Deposition | 1643 | - | - | [209] |
| Ex-MWCNTs | - | PANI | Reaction Processing | 809.6 | 4.9 | 72.5 | [210] |
| MWCNTs | MnO ₂ | - | - | 77.83 | 571.3 | 29.84 | [211] |
| MWCNTs | Co-HCF | - | Chemical Precipitation | 648.5 | 0.876 | 140.7 | [212] |
| Mesoporous Carbon | CuO | - | - | 616 | 0.438 | 26.6 | [213] |

3.5.2 Transistors

In modern electronics, field-effect transistors display an important role regarding inherent parts of different devices, for instance, computer chips. It is difficult to design novel device geometries to improve gate electro-statics required for competent On/Off switching for extremely scaled molecular transistors with rapid channel [214]. Nanocomposite materials such as combining conducting organic polymer, abbreviated as COP, and CNTs provide an alternative pathway to introduce electronic properties. Polypyrrole, poly-phenylene-vinylene, poly-thiophene and poly-m-phenylene-vinylene are few of the examples of COPs [215]; [216]; [144];. It was also recommended that in CNTs/ COP nanocomposites, either polymer modifies the CNTs or COPs are doped with CNTs. Carbon nanotubes, especially SWCNTs attained much attention as they exhibit outstanding electronic and mechanical properties, and excellent chemical, electrical and thermal stability. Their potential to be used in different technological applications, like field-effect transistors, memory elements, molecular wires, logic gates, and high strength fibers, is well developed. Based on the literature, SWCNT is appropriate to be used in functional field-effect transistors as semi-conducting channel and proved more compatible than silicon-based devices. Therefore, studies carried out by Qi and co-workers reported that SWNTs with poly-3-hexylthiophene-2,5-diyl influenced significant current modulation than metal contacted devices at similar voltage [217].

In comparison to organic crystalline semiconductors, thin-film transistors developed from COP materials usually suffer from lower performance because of their low carrier mobility [218]. The properties of the single molecules and molecules' structural order are the main factors, defining the macroscopic properties of organic semiconductor materials. Polyaniline is a unique conducting polymer in which the conductivity results through a partial oxidation process. Regarding structural modification or electronic association between two components, CNTs in conducting polymers were displayed to exhibit properties of individual

elements with a synergistic effect [219]. CNTs will not only permit the carriers to be transported through offering percolation routes, thus assist to enhance mobility.

CNTs/polyaniline nanocomposite prepared through reaction technique has shown an increase in polymer properties because of the existence of CNTs [220]. Such combined properties are favorable in organic electronics. DuPont were the first to use CNTs/ polyaniline nanocomposite as printable conductors for application related to organic electronics [221]. Moreover, Ramamurthy and co-workers demonstrated that enhancement in material consistency as well as a decrease in defect densities will build these nanocomposites appropriate for use in producing organic electronic devices. Furthermore, it was recommended that through finely dispersing the CNTs in the polymerization phase, a better level of homogeneity may be attained further improving the material quality [222].

3.5.3 Thermo-electric devices

Lately, a number of attempts have been conducted to associate the remarkable mechanical, thermal stability, electrical conductivity, and tunability of the seebeck coefficient of CNTs with the low thermal conductivity, solution processability, economical and scalable fabrication of conducting polymers. Specifically, nano-scale hetero-structuring is suggested as a decent approach to adjust electrical and thermal conductivity and thermopower. In opening tries, conducting CNTs/ polymer nanocomposites, for instance, CNTs/ PEDO, were fabricated via reaction techniques, to familiarize hetero-junctions and to display the effects of components on thermo-electric properties [223].

The hetero-junctions presence is projected to enhance the thermoelectric transport properties that are obstructing heat flow and favouring electronic conduction. Certainly, it has been detected that CNTs/ poly(3,4-ethylene-dioxy-thiophene)-poly-styrene-sulfonate reveal enhanced electrical conductivity without expressively decreasing/altering thermo-power,

ultimately resulting in upgraded power factor (PF). This behavior consequences from thermally separated, however, electrically connected junctions in the nanotube system, which offers it practicable to tune the properties in favor of significant thermo-electric figure of merit [224]. Correspondingly, when CNTs' bundle coated with PANI, thermally insulating PANI interfacial deposits may act as energy filters, which offer high energy carriers to go through as well as scatter the low energy carriers, thus upgrading the Seebeck coefficient. Moreover, PANI growth over CNTs creates an ordered chain structure that decreases the π - π conjugation defects along the PANI backbone letting incline in carrier mobility [225]. Subsequently, composite holds identical thermal conductivity as that of pristine PANI but significantly enhanced power factor than pristine CNTs or PANI.

Apart from conducting polymers, several studies have been done using conventional insulating polymers with CNTs to enhance thermoelectric power factor. For instance, SWCNTs/ PVDF composite with thin film-based systems [226]. In addition to the thermo-electric properties' enhancement, CNTs are also helpful to upgrade the composite based thermo-electrical components mechanical strength, for both bulk and thin films.

3.5.4 Sensors

Sensor technology is a very wide field, and its upcoming development will include the interaction of almost every technical and scientific discipline. The development of science and technology via new findings is rarely possible without a parallel growth in the field of sensor technology. These developments are the results of a non-stop chain of new sophisticated measurement methods and at the heart of each measuring instrument is its sensor [227]. Regarding sensor classification, sensors are classified as natural sensors and man-made sensors. The natural sensors can be initiated in living beings, where the stimulus signal is processed and conveyed as an electrochemical signal produced because of ion-exchange. Whereas, regarding man-made sensors, the stimulus is conveyed and processed into an electro-

chemical signal because of charge-exchange. The word sensor can be described as the device that changes a physical phenomenon into an electrical signal [228]. Usually, sensors are recognized in a similar way as electric devices. Based on the performance, applicability range, and cost, electronic devices are mostly designed to go along with physical sensor components, which is hence extremely essential to an inclusive electronic circuits. If a sensor is properly designed, then optimum abstraction of information is convenient. Apart from the abovementioned sensors classification, sensors can also be classified in terms of different physical phenomena measured, such as distance, pressure, temperature, magnetoelectric, angular motion, chemical transformation, biochemical process, photo-electric, and many more. Moreover, classification can be carried out based on operation type i.e. digital or analog, whether the amount is calculated directly or indirectly, the material used as sensor applications, application field, the stimulus applied, and material used [229].

Recently, a significant amount of research and development has been carried out on CNTs and its nanocomposites due to remarkable properties it holds, such as chemical and physical. CNTs and its nanocomposites are not broadly used only in nano-electric systems, hydrogen storage materials, catalyst, supercapacitor but also consider for applications regarding sensors [230]; [231]; [232]; [233].

Sensors, for instance, chemical sensor and gas sensing at low temperature are of great attention due to low consumption of energy. Thus, most available sensors nowadays, apart from a few operate at a higher temperatures. Regarding sensitivity improvement, several approaches have been defined. For instance, modify the CNTs with gold (Au) or platinum (Pt) nanoclusters to improve the sensitivity in gas chemi-resistors, reported by Penza and co-workers [234]. Similarly, Maeng and co-workers that had designed an extremely sensitive nitrogen oxide (NO₂) sensor array through an undecorated SWCNT mono-layer junction [235].

Table 8 listed various studies that have been conducted using carbon nanotubes-based nanocomposites for chemical and biosensors applications:

Table 8: Carbon-based nanocomposites for sensors applications

| Carbon Nanotubes | Polymer | Metal/ Metal Oxide | Sensor | Detection limit (ppm) | Sensitivity (%) | Response/ Recovery Time | References |
|-------------------------|----------------|--------------------------------------|--|--|--|--------------------------------|-------------------|
| MWCNTs | PEDOT:PSS | - | Humidity | - | 10 | - | [236] |
| MWCNTs | PANI | - | Ammonia (NH ₃) | - | 15.5 | 6/35 | [237] |
| MWCNTs | PANI | - | Carbon monoxide (CO) | - | 26.7 | 76/210 | [238] |
| SWCNTs | PANI | - | Hydrogen | - | - | 2.30 | [239] |
| MWCNTs | PANI | - | Ammonia (NH ₃) | 4 | 40 | 18/46 | [240] |
| f- MWCNTs | - | Nafion:Nano-Platinum-Gold | Ascorbic Acid | - | - | - | [241] |
| MWCNTs | - | Tin-dioxide (TiO ₂) | Nitrogen dioxide (NO ₂) | 5 | 39 | - | [242] |
| MWCNTs | - | TiO ₂ : PtO ₂ | Ethanol | 250 | - | 23/46 | [243] |
| MWCNTs | - | Gold (Au) | Nitrogen dioxide (NO ₂) Ammonia (NH ₃) Hydrogen sulfide (H ₂ S) | 0.2 | NO ₂ : 2.4 NH ₃ : 0.07 H ₂ S: 9 | - | [244] |
| MWCNTs | - | Tungsten trioxide (WO ₃) | Hydrogen | - | High | - | [245] |
| SWCNTs | - | Platinum (Pt) | Nitrogen dioxide (NO ₂) Ammonia (NH ₃) | NO ₂ : 1 NH ₃ : 7 | NO ₂ : 1.2 NH ₃ : 1 | 282-294/3786-3894 | [246] |
| SWCNTs | - | Copper (Cu) | Hydrogen sulfide (H ₂ S) | 5 | - | 7/9 | [247] |
| CNT foam | PDMS | - | Pressure | - | High | - | [248] |
| MWCNTs | - | Platinum (Pt) | Carbon dioxide (CO ₂) | 0.05 | - | 300-600/900 | [249] |

3.5.5 Other applications

Due to the CNTs unique range of properties, it has opened a brand new age of enhanced multi-functional materials. CNTs embedded in polymer/ metal matrices offer materials that can be used for several significant performance engineering applications. Recently, the most extensive use of CNT nanocomposites is in electronics. These nanocomposites would be considered to safeguard electromagnetic interference as well as electrostatic discharge materials. Nanotubes' microwave absorbing capability can be exploited to heat impermanent housing structures as well as applications in space exploration. CNTs' thin layer on plastic could also be considered in transparent conducting composites. Carbon nanotubes nanocomposites exceptional mechanical strength might be employed to design some high-end sporting belongings, for instance, baseball bat, badminton racket, and tennis racket, and hence offering higher performance. Briefly, the prime market for CNT nanocomposites will certainly for high-value applications that may absorb the additional costs, that involve commercial divisions, for example, electronic particularly aero-space which needs high strength, light-weight, high-temperature resistant composites.

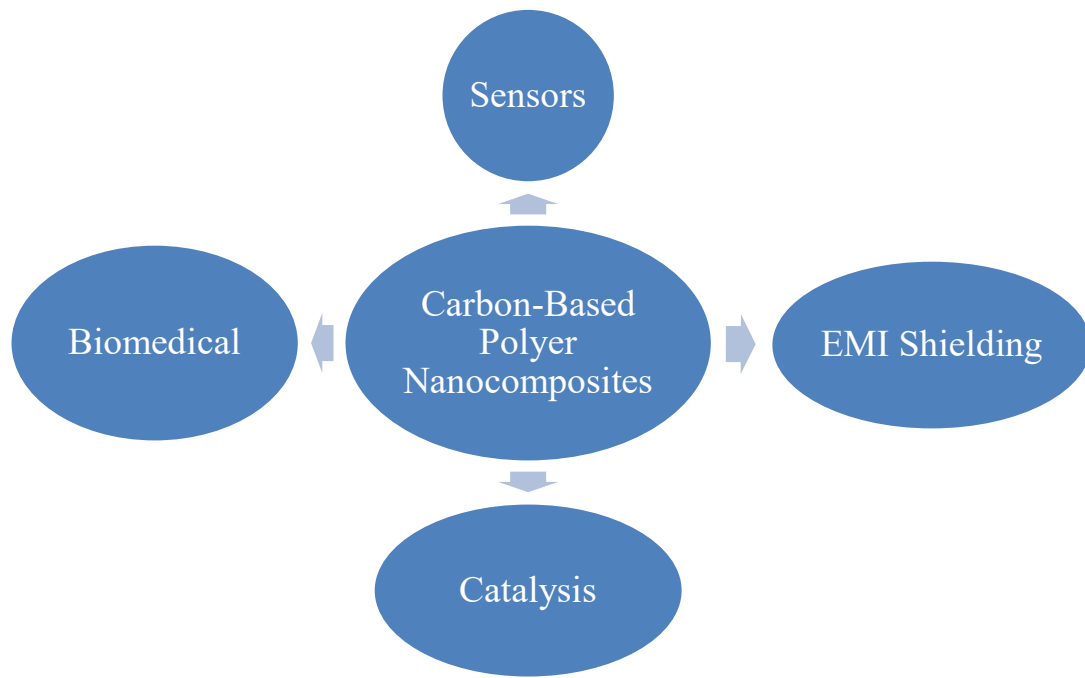


Figure 7: Applications of carbon nanotubes/polymer nanocomposites

Moreover, there is an extensive increase in publication number regarding CNT embedded in polymer/ metal matrices that eventually show its interest is kept on increasing. This might be somewhat accredited to the availability and low cost of good quality of CNTs. CNTs incorporation in metal matrix offered diverse applications in different engineering fields such as tribological coating to functional applications i.e. hydrogen storage, catalysis, and thermal management in electronic packaging. There has been an accomplishment in CNTs use with aluminum to produce micro-electro-mechanical systems (MEMS), in terms of high-frequency resonators for which extensive elastic modulus is needed. Furthermore, stiffness and strength improvement has also been noticed in a number of metal systems, and reduction of COF and wear rates have also been reported through the addition of CNT in copper and nickel. Thus, it also has been reported that CNT higher content, loading strength tends to reduce. This is mainly due to the incapability of the process to homogeneously disperse CNTs or attain dense components at high content of CNT. Novel methods have also been developed for attaining

better CNTs dispersion at the laboratory and large scale. Figure 8 illustrated the potential applications of CNT embedded metal nanocomposites in different industries.

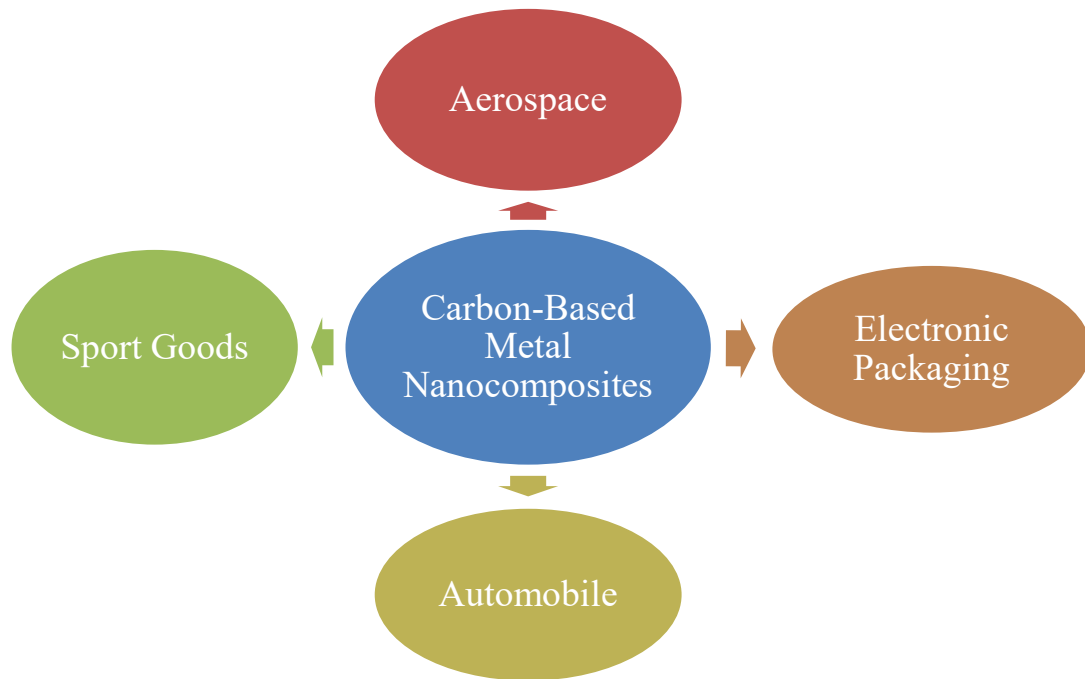


Figure 9: Applications of carbon nanotubes/metal nanocomposites

4. Future prospects

Remarkable progress has been made in functional carbon- and CNT-based nanocomposites synthesis and applications. Owing to the unique properties of CNT, the CNT-based nanocomposites exhibit enhanced properties, such as enhanced charge separation and transportation properties, which greatly improve the overall performance of the nanocomposite. CNT remains a very interesting material for researchers. With advances in both fundamental physics and chemistry of practical techniques of CNT and CNT-based nanocomposite in revolutionary applications, a new human age would be recorded. Followings are a few recommendations for further improvements of the fabrication and applications:

- i.* Developing new nanocomposites using different types of CNTs and graphene as well as graphene oxides.

- ii.* Developing novel nanocomposites using different types of CNTs and mxenes.
- iii.* Developed CNT-based nanocomposites can be further extended to the modelling of nanocomposite systems for a wide range of applications
- iv.* Developed CNT-based nanocomposites can further be employed for different applications due to their properties such as in aerospace, wind blades, and sporting goods

5. Conclusion

In this review studies, fabrications, and applications of carbon nanotube-based nanocomposites have been systematically compiled and discussed. Authors have tried to choose some of the exceptional examples from the literature where the CNTs and CNTs-based nanocomposites properties are comprehensively or partially utilized to produce remarkable effects and applications. The materials offer extensive potential for a broad range of applications such as storage, sensors, environmental, and medical. As referred to as multi-disciplinary research, further enhancement and development is needed strongly. Concerning this review studies, this studies aim to increase the visibility of CNTs embedded polymer and metal, moreover, provide an extensive overview of recent advances in both laboratory and industry.

At last, the successful application of CNTs incorporation with different polymers, metals, metal oxide, and their consideration into the real-world needs a strong enhancement in methodology that guarantees reproducibility as well as a better understanding of the structure-property relationship. Furthermore, it is also essential to address the biocompatibility of CNTs and its nanocomposites materials for their applications related to medical and biological purposes. Health and safety regulations required further detailed studies regarding toxicology, exposure, and studies focused on such issues should be initiated.

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