



*Review*

## **A comparative review of Mg/CNTs and Al/CNTs composite to explore the prospect of bimetallic Mg–Al/CNTs composites**

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**Abstract:** Lightweight materials characterized by low density, high strength to weight ratio, low porosity, high corrosion resistance with improved mechanical, thermal, electrical properties are now extensively used in many engineering applications ranging from the deep sea to aerospace. Besides, the materials must be multifunctional comprises a fast and economic manufacturing technique. Metallic matrix CNTs composites such as Mg/CNTs and Al/CNTs have such the aforementioned quality. However, Mg/CNTs and Al/CNTs has some specific advantages and disadvantages that restrict their applicability. To harvest the dual benefit of Mg/CNTs and Al/CNTs, bimetallic matrix Mg–Al/CNTs may be prospected. Mg–Al bimetallic combination for alloying has substantial mechanical properties with heavy amalgamation. Therefore, interdisciplinary research on reinforcement, compacting, bonding, dispersion of CNTs in the bimetallic matrix, microstructure and defect analysis will open the door for producing new class of composite materials. In this work, various Mg/CNTs, Al/CNTs, Mg–Al composites have been studied to find the prospect of Mg–Al/CNTs composites. The worthwhile accomplishment of the reviews will provide the knowledge of fabrication for CNTs reinforced bimetallic Mg–Al based lightweight composites and understand its mechanical behaviors.

**Keywords:** carbon nanotubes; bimetallic matrix; lightweight composites; fabrication method; SWCNTs/MWCNTs

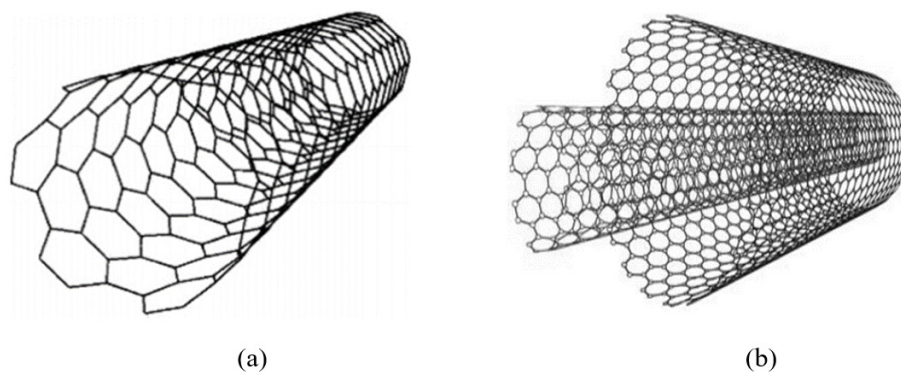
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## 1. Introduction

Material development needs continuous progress to satisfy the expansion of new technology and versatile use of material [1]. A pure and single material hardly satisfies the increasing demands of improved mechanical, chemical, electrical properties [2]. The lightweight composites materials are becoming essential for high-speed trains, automotive vehicles, structures, and prominently in any space applications [3–5]. High strength metallic composites have been used for the last few decades to come across the solution [6].

The most significant metals and alloying elements on earth are Al, Mg, Ni, W, Sn, Cu, Ag, Ti and their various range of blends for different applications [7,8]. The major shortcomings of these composite combinations are strength to weight ratio. Some alloys are very difficult to manufacture and even costly. Few metals are rare and barely imaginable to apply in general engineering problems. Thus, lightweight, and low-cost alloys or composites are demands of the future without any sacrifice of mechanical prospects. A flexible, easier and fast manufacturing or developing process must be available to produce such light-weight composite with superb mechanical properties.

To overcome the problem, composites of metal, fiber, polymer, plastic has served a lot till today. However, the coolest thing ever is the development of carbon nanotubes (CNTs) [9] and its various features and advantages [10] as a reinforcing element to combine it to form reinforced composite with other elements. CNTs looks like a tube in the molecular structure shown in Figure 1 and possess various features [11,12] like high thermal (approximately 2000 W/m/K) and electrical conductivity, large elasticity (~18% elongation) to failure and high Young's modulus of 1.28 TPa. The tensile strength of CNTs are approximately 100 GPa and can be bent considerably without damage. All variety/types of CNTs depicted in Figure 2 [13,14] have a low thermal expansion coefficient but can act as a good electron field emitter with a current density of  $10^{13}$  A/m<sup>2</sup> [11,12].

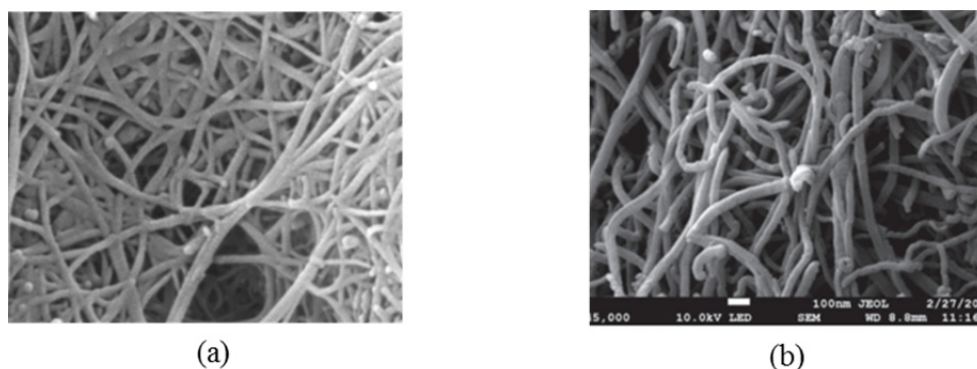


**Figure 1.** Molecular structure of (a) single-walled carbon nanotubes, and (b) multiwall carbon nanotubes.

Besides this physical property, the use of CNTs has some advantages as a reinforcing element. Such as it is extremely small and lightweight, and the resources required to produce CNTs are plentiful and can be made with only a small amount of material. Homogeneous dispersion of CNTs improves the conductive mechanical properties of composites with no apparent agglomeration [10].

There are several categories of matrix available for CNTs reinforcing components. Metallic, polymer and ceramic are the three major matrix categories. Among these matrixes, polymer matrix

and ceramic matrix have used widely as a primary matrix for CNTs reinforcement despite the good matrix properties of metals. The methods used for preparing the CNTs–polymer matrix is the Coagulation Precipitation Technique [15], Solution Mixing [16,17], Melt Mixing [16,18,19], and In-situ Polymerization [2,20–23] with a particular emphasis on evaluating the dispersion state of the nanotubes. The method involved for CNTs reinforced ceramic composites are powder processing follow by sintering, spark plasma sintering, extrusions etc. [24,25]. CNTs reinforcement in polymer and ceramics shows a good mechanical benefit but not competitive with metallic matrix CNTs reinforced composites [26–31].



**Figure 2.** Field Emission Scanning Electron Microscope (FESEM) image of (a) single-walled carbon nanotubes [13], and (b) Multiwall carbon nanotubes [14].

In the case of metallic matrix CNTs reinforced composites, incorporation of CNTs into the metallic matrix, improves the mechanical properties such as microhardness, fracture toughness, electrical and thermal properties [26]. The major metallic matrix so far found to be used are Al and Mg matrices. The common use of CNTs in the metallic matrix is done by powder metallurgy processes. The material scientist reveals that augmentation of CNTs via powder metallurgy technology or mechanical mixing has successfully enhanced the strength of Al/CNTs and Mg/CNTs based alloys [27,28].

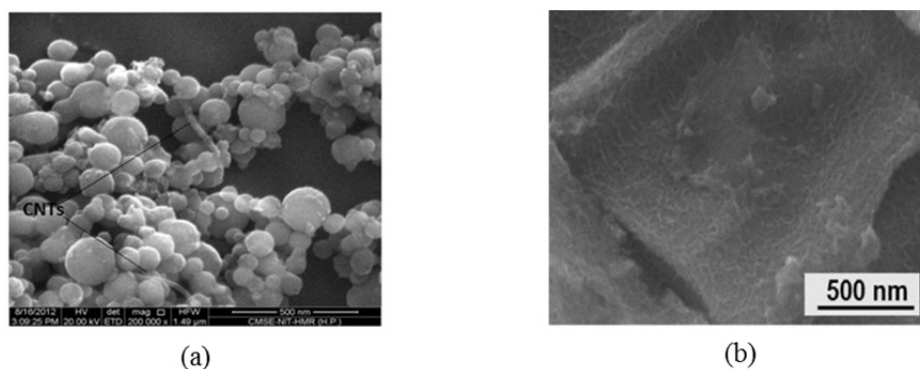
However, CNTs may damage in mechanical powder mixing operation and during secondary processing [29,30] though the processes do not affect the graphical structure. Generally, Al and Mg is not easily wetted by Carbon [32], but the Nano Scale Dispersion (NSD) method [33,34] and Spread Dispersion Method [35] to form CNTs/Al or CNTs/Mg composite has succeeded in producing uniform dispersion of CNTs in Al or Mg matrices. The dispersion of CNTs may be promoted by the wet shake-mixing approach [36]. In the consolidation stage of CNTs/powder matrix composite, Hot extruded CNTs dispersed well at the boundaries [37–39]. The CNTs become aligned with the extrusion direction in the composites obtained by this process [37].

Al-Aqeeli [40] used ball milling as mixing and dispersion method followed by Sintering, Spark Plasma Sintering (SPS), Microwave Sintering ( $\mu$ WS), and Hot Isostatic Press Sintering (HIPS) for the consolidation of ball mill Al powder with CNTs with a variation in temperatures of 400, 450, and 500 °C. With the use of ball milling as a mixing and dispersion method, the researcher recommended the SPS consolidation to produce such composites to achieve the highest hardness values and around 100% densification.

On the other side, CNTs reinforced with Mg has also attracted the materials field for its good properties [41–44]. The hot extrusion process has also regarded as a novel process for Mg/CNTs composites. Kim et al. [38] and Mindivan et al. [39] explain the properties of hot extruded Mg/CNTs alloy in their work. Shimizu et al. [45] work on the Mg alloy for improved properties by adding some crystalline CNTs. Muhammad et al. [46] and Straffelini et al. [47] recommended Spark Plasma Sintering (SPS) technology as appropriate for Mg alloy. In some cases, ultrasonic melting may be used for further dispersing CNTs in the molten Mg [48].

For achieving more strength-to-weight ratio for light-weight applications such as car and aerospace, the bimetallic matrix carries a good prospect to add value in the composites. Mg/CNTs exhibited better tensile, compressive strength but corrosive properties are not so good [49]. Though Al/CNTs show extended quality in corrosion resistance but weight and toughness are not desirable. Since Al-Mg composite is good bimetallic composites, the addition of CNTs in Al–Mg bimetallic matrix will harvest both quality of Al/CNTs and Mg/CNTs and will make it to useable for more rigorous conditions.

We noticed that the bimetallic matrix to CNTs reinforced composites have rarely been tried previously due to a lack of information on the uniform dispersion and easier method to compact the constituents together followed by reinforcement. Figure 3 shows the Scanning Electron Microscope (SEM) images of Al/CNTs and CNTs/AZ91D composites respectively and illustrated the dispersion and consolidation defects. Few authors like Yan et al. [50] used Spark Plasma Sintering (SPS) method followed by hot extrusion to produce Mg–Al/CNTs composites with better tensile and compressive properties. Their analyses revealed that the Mg–Al/CNTs composite demonstrated an enhancement in mechanical properties like elastic modulus, yield strength, ultimate tensile strength and failure strain compared to pure Mg/CNTs or Al/CNTs [50,51].



**Figure 3.** (a) SEM image of Al/CNTs composite with 2 wt% of CNTs [29], and (b) higher magnification and 2.0 vol% CNTs/AZ91D nanocomposites [52].

Therefore, the main challenge in the production of this bimetallic matrix composites reinforced by CNTs is the development of a manufacturing process. The chosen process should ensure the dispersion of nanoparticles without damaging CNTs, and the formation of a strong bond with the metallic matrix during consolidation. Therefore, this review study focuses on the properties of Mg/CNTs and Al/CNTs composites experimented by the research, preparation methods and post-treatment to enhance mechanical properties. It will help to fabricate the Mg–Al/CNTs in future.

## 2. Detailed review results

### 2.1. Mg/CNTs composites

#### 2.1.1. Fabrication method of Mg/CNTs composites

Being lightweight, Mg is thought to be a good composite forming element with CNTs and has a good composite forming properties though little work has found. Sun et al. [53] were first to claim the fabrication of Mg/CNTs composites using in situ synthesized CNTs–Mg powders by the powder metallurgy process. Li et al. [54] developed a two-stage improved processing method of Mg/CNTs, where the first stage deals with pre-dispersed CNTs on Mg alloy chips and the second stage comprises melting of Mg chips and CNTs in a stirring machine. In another study, melt stirring techniques are applied successfully in which galvanic coupling takes place at increased rates as CNTs bundles are broken up and homogeneously dispersed in the matrix [55].

A modified powder metallurgy technique followed by hot pressing and hot extrusion respectively could successfully fabricate AZ31/CNTs composites with a homogeneous distribution of the CNTs at proper content [56]. The wettability issue between CNTs and A356 alloy has been enhanced by using Mg surfactant [32]. In addition, Mg surfactant can help to reinforce CNTs uniformly as well as the highest mechanical strength is achieved. The crystallographic texture of the composites reveals the exalted dispersion of the above reinforcing method. Cold pressing followed by hot extrusion without sintering steps is another emerging way for Mg/CNTs composite fabrication with uniform distribution of CNTs [57].

Recently Akinwekomi et al. [58] have proposed a powder metallurgy processing technique combined with rapid microwave sintering to fabricate Mg/CNTs composites. The researchers also suggested the unique rapid sintering method without recourse to any secondary processing. The technique provides a significant improvement of mechanical properties for bimetallic matrix–CNTs reinforced composite. In another work of Akinwekomi et al. [59], they have used the same process for Mg alloy–CNTs composite foams with improved compression and energy absorption properties. The conclusion of the work outlined the sintering process scheme as a rapid and energy saving efficient technique for metal matrix/CNTs composites.

Hybrid composites of AZ91 magnesium alloy were found to be fabricated by semisolid stirring assisted ultrasonic cavitation [60]. They found the optimum hybrid ratios of CNTs and silicon carbide (SiC) nano particulates is 7:3, where tensile mechanical properties were improved. A comparative study was done for Magnesium (Mg) composite reinforced with CNTs in pure Mg and AZ61 Mg alloy matrix via powder metallurgy route containing wet process using isopropyl alcohol (IPA) based zwitterionic surfactant solution with unbundled CNTs [61]. This study revealed that only the AZ61 Mg alloy matrix showed tensile strength improvement and clarified that the addition of CNTs has no effects on microstructures and grain orientations of the composite.

Ultrasonic melt processing in Mg/CNTs composite showed that no reaction product was found between the interface of Mg matrix and CNTs which in turn leads to good interfacial bonding [62]. With the application of microwave sintering followed by hot extrusion Mg/CNTs composites are obtained with minimal porosity [63]. Accumulative roll bonding is mentioned as a new method for Mg/CNTs composite fabrication which leads to improving strength [64].

### 2.1.2. Surface and microstructural characteristics of Mg/CNTs composites

Funatsu et al. [65] studied the initial galvanic corrosion behavior of MWCNTs reinforced AZ61B alloy composites after and before the composites heat treatment. They presented that a corrosion rate of about 30% was reduced after the heat treatment of the composites compared to non-treated composites. Another corrosion behavior investigation presented that good dispersion of MWCNTs leads to higher corrosion rates on the composite surface [55]. They concluded two main points; one is by adding MWCNTs on the Mg composites. Its corrosion rate was drastically improved and the other is homogeneous dispersion of MWCNTs results to lower corrosive resistivity. The impact of CNTs on Mg-based amorphous composites corrosion behavior was investigated by Lou et al. [66]. Corrosion resistance is enhanced up to 30% and the bridging effect is found between the matrix and corrosive oxide film. Also, the addition of CNTs has a positive impact on wear rate and friction coefficient [57].

Microstructural characterization provides a better understanding of the interfacial bonding with failure mechanisms. Poor compositing effect exposed that a loose and dispersive distribution has occurred between the matrix and CNTs [67]. SEM micrographs showed that poor interfacial bonding contains numerous macroscopic cracks and fine microscopic cracks were observed in the matrix [68]. SEM image of tensile fracture has dimples and tear ridges whereas compressive fracture has microcavities and clusters [56]. These micro-cracks and cavities lead to high stress concentration which results in lower strength of the composites. In another result found that compressive failure contains macroscopic crack with mixed mode of shear and brittle failure [58].

However, the failed surface examined under SEM shows several pores have collapsed and extends from the pore walls to the matrix [59]. Under high magnifications interfacial reactions did not exist between the CNTs and matrix due to the surface pretreatment [60]. Although CNTs addition might affect the microstructure and grain orientation of the Mg matrix, FESEM observation and Electron Backscatter Diffraction (EBSD) analysis clarified that neither microstructure nor texture was significantly influenced by CNTs addition [61]. The failure mode of Mg/CNTs composites was brittle with the presence of cleavage steps due to the poor interfacial integrity [63].

### 2.1.3. Effect of SWCNTs/MWCNTs on Mg/CNTs composites

Effect of SWCNTs/MWCNTs plays an important role in strengthening mechanisms, load transfer, density, porosity etc. Strengthening due to the load transfer of the MWCNTs increases linearly and becomes more important than other strengthening effects at higher MWCNTs amounts according to the present model [54]. At a threshold amount of about 0.3 wt%, the load transfer effect exceeds the Orowan strengthening and the thermal mismatch effect. Without the pre dispersion of MWCNTs, the coupling effect takes place with MWCNTs agglomerates [55]. The load transfer effect is insignificant if the MWCNTs volume fraction is too low which in turn poor strengthening effect in the composites [64].

The addition of MWCNTs more than 1 wt% increases the porosity and decreases the density and electrical conductivity [57]. Another study reveals that the addition of MWCNTs has not any significant impact on porosity and density [59]. With the addition of MWCNTs porosity and density are increased on the other side grain size is reduced [63]. Goh et al. [69] found that porosity has an incremental trend with the addition of MWCNTs whereas density is decreased. Grain refining

strengthening was caused due to the addition of SWCNTs and this strengthening effect of SWCNTs works as a reinforcement [62]. This grain refinement effect works well on AZ91D when the SWCNTs are uniformly dispersed. The analysis presented if MWCNTs are added more than 1 wt% in melt stirring technique a larger amount of MWCNTs are separated to the top or bottom of the melt and did not reside in the melt [70].

#### 2.1.4. Coefficient of Thermal Expansion (CTE) and thermal behaviors of Mg/CNTs composites

CTE has a positive impact on the strength enhancement of Mg/CNTs reinforced composites. It has been reported that improvement in tensile and compressive strength of Mg/CNTs composites is characterized by dislocation generation due to the mismatch in CTE elastic modulus between Mg matrix and CNTs [51]. However, mismatch in CTE between CNTs and Mg matrix and extensive dislocation nucleation around the CNTs leads to the hardening of the metal matrix [54]. Mismatch of CTE between Mg matrix and CNTs could also lead to the increase in yield strength, the higher the difference of CTE between Mg matrix and CNTs the higher the increase of the yield strength [56]. Zhou et al. [60] presented CTE is responsible for the room temperature strength improvement of Mg/CNTs reinforced composites. Improvement in thermal conductivity occurs with the addition of CNTs which caused an increasing undercooling as well as grain refinement of the composites [62]. Another investigation shows that with the addition of CNTs lowered the CTE of the Mg matrix [63]. CTE results indicate that Mg/CNTs nanocomposites are thermally more stable than monolithic pure Mg [69]. Table 1 below illustrates the fabrication methods and its various constituents of Mg/CNTs composites.

**Table 1.** Summary of Mg/CNTs composites fabrication method.

Powder preparation	Fabrication method	CNTs content (vol%/wt%)	Remarks	Refs.
Ball milling	Melt processing	0.5 wt%	Melt processing further dispersed tiny CNTs clusters; CNTs were mainly located along matrix grain boundaries because of push effect during solidification	[48]
Ultrasonication	Semi-powder metallurgy with vacuum sintering followed by hot extrusion	0.1 wt%	Semi powder metallurgy can successfully synthesize Mg based CNTs reinforced composites; synergetic effect revealed uniform dispersion of CNTs; efficient load transfer	[51]
Chemical vapor deposition and Ball milling	Sintering followed by hot extrusion	1.8, 2.4, and 3.0 wt%	Homogeneous dispersion of CNTs on Mg powders; tensile strength was enhanced to 45% compared to pure Mg	[53]
Mechanical stirring	Melt stirring	0.1 wt%	Good dispersion of MWCNTs in Mg matrix; MWCNTs do not act as a grain refiner in the matrix	[54]
Mechanical stirring	Melt stirring	0.1, 1, and 5 wt%	Good dispersion of MWCNTs leads to even higher corrosion rates; corrosion rate increases with the increasing amount of MWCNTs	[55]

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Powder preparation	Fabrication method	CNTs content (vol%/wt%)	Remarks	Refs.
Ball milling	Compaction followed by sintering and hot extrusion respectively	0.5, 1.0, 2.0, and 4.0 wt%	Weaken the basal plane texture of composites; poor wettability of CNTs with matrix; mechanical strength was attributed due to the efficient load transfer with the involvement of Orowan mechanism and thermal mismatch	[56]
Ball milling	Cold pressing followed by hot extrusion	0.5, 1, 2, and 4 wt%	Cold pressing followed by hot extrusion without sintering provides a unique opportunity for Mg based light weight composites; uniform distribution of CNTs at the chip surface decreased with increase in the CNTs content	[57]
Ball milling	Rapid microwave sintering	1–3 vol%	Composites exhibited a mixed mode of shear and brittle fracture as a failure mechanism; microwave sintered composites showed significant improvement in hardness and compressive properties	[58]
Ball milling	Rapid microwave sintering	1, 2, and 3 vol%	Significant improvements in the compressive strength and energy absorption properties; failure of composites are the initiation of cracks and their propagation along the loading direction, as well as a mixed mode of shear and brittle failure of the pore walls and matrix	[59]
Ultrasonication	Semisolid stirring assisted ultrasonic cavitation	0.7 wt%	The grains of the matrix in the AZ91 was obviously refined; tensile and yield strength was increased	[60]
Mechanical stirring	Spark plasma sintering followed by hot extrusion	0.26 vol%	Microstructure nor texture was significantly influenced by CNTs addition; elongation was improved due to the elimination of excess amount of MgO formation	[61]
Ultrasonication	Melt processing	0.5 wt%	Grain refinement with the addition of CNTs; yield strength, ultimate tensile strength, and elongation were improved significantly but afterward further improvement is restricted due to poor interface bonding between CNTs and AZ91D matrix	[62]
Ball milling	microwave sintering followed by hot extrusion	0.3, 0.5, 0.7, and 1 wt%	CNTs reinforcements lowered the coefficient of thermal expansion; brittle fracture is evidenced by the presence of cleavage steps as failure mode	[63]
Ball milling	Accumulative roll bonding	3 vol%	New method for fabricating Mg/CNTs composites with good mechanical properties; homogeneous dispersion of CNTs	[64]
Blending	Sintering followed by hot extrusion	1.5 vol%	Mg/CNTs nanocomposites are thermally more stable than monolithic pure Mg; ductility was reduced due to the activation of non-basal cross slip with the addition of CNTs	[69]



## 2.2. Al/CNTs composites

### 2.2.1. Fabrication method of Al/CNTs composites

Al as a readily available material always on demand to add for composite formation. Various forming processes like in situ chemical vapor deposition applied for Al/CNTs composite formation and better dispersion of CNTs in the Al matrix [71]. The better dispersion of CNTs in the Al matrix can also be obtained by Ultrasonication held for a longer time [72]. Such as Simões et al. [73] focused on powder metallurgy of Al–Ni matrix composites reinforced by CNTs by ultra-sonication to get good dispersion quality. Liao et al. [74] applied a hot extrusion process for Al–3 wt% MWCNTs composite. The results are simultaneous improvements in density, hardness and tensile strength [73–75]. Another forming technique namely sintering has employed for novel Al-matrix nanocomposites reinforced with CNTs [76]. Various sintering techniques like spark plasma sintering, Microwave sintering, Hot isostatic press sintering etc. have also been incorporated in Al/CNTs reinforced composite forming processes [40]. More specifically powder metallurgy as a preprocessing or post processing method with the sintering technique has proven effective for CNTs reinforced Al matrix composite [77].

In another study, Choi et al. [78] fabricated Al composites with MWCNTs as reinforcement by hot extrusion of ball-milled powders. They suggested that MWCNTs could be an outstanding reinforcing agent. At the same time, mechanical strengths were enhanced with MWCNTs uniformly dispersed throughout the composites. Some other studies which are fabricated CNTs reinforced Al matrix composites by hot extrusion process with subsequent preprocessing or post-processing techniques [79–81]. Friction stir processing is also one of the best popular methods for Al/CNTs composites. In the stir casting process, CNTs disperse in the Al matrix individually. However, some CNTs become shortened and some  $Al_4C_3$  are formed in the matrix along with finer grain boundaries.

The hot-pressing method results in the clustering of CNTs, therefore inappropriate for fabricating Al/CNTs composites [82]. Hot extrusion provides alignment of CNTs in extrusion direction, but inhomogeneous poor dispersion of CNTs in the Al matrix. A novel fabrication method of Al/CNTs composites was developed named Nano-Scale Dispersion (NSD) where carbon nanotubes were dispersed uniformly in nanoscale in Al matrices [83]. With the incorporation of MWCNTs in a very small amount, the mechanical properties of Al/CNTs were greatly enhanced. In some cases, spark plasma sintering has been carried out. Nevertheless, the choice is limited mostly for Cu/CNTs and in Al/CNTs systems.

For Al/CNTs composites in the high-pressure die casting process, elongation at fracture and tensile strength could be promoted to 27% and 8% respectively. Fracture toughness has been investigated by Sun et al. [84], where Al/CNTs are fabricated by simple colloidal processing. They found with the addition of 0.1 wt% of CNTs increased the fracture toughness. A combination of Rheo-casting and Squeeze casting techniques have been developed for Al/MWCNTs nanocomposites [26]. A comparison has been made among high energy ball milling, low energy ball milling and polyester binder-assisted (PBA) method in context to the dispersion of CNTs in Al powder [85].

### 2.2.2. Surface and microstructural properties of Al/CNTs composites

SEM image provides laminar types of fracture and fracture surfaces have fine dimples (<10  $\mu\text{m}$ )

with CNTs aligned in the fracture direction [19]. Dimples found in the microstructure which suggests strong interfacial bonding between the CNTs and the matrix [26]. SEM micrographs present poor dispersion of CNTs that leads to non-uniform fracture and CNTs were not pulled out but broken when the composites were fractured [33]. Brittle fracture is found on the microstructural analysis with different direction of slip plane in one composite specimen [86]. Many broken CNTs were found on the fracture surface and among them only a few CNTs were pulled out [71]. However under loading, agglomerated CNTs were responsible for fracture initiation and failed to share the stress effectively [74]. Fracture surface under tensile test shows a lot of dimples associated with ductile fracture [75].

Kumar et al. [82] analyzed the corrosion behavior of Al nano metal matrix composite (ANMMC) prepared with 98% purity of MWCNTs. Hardness is improved with the addition of 1.75 wt% of MWCNTs in the composite and showed better corrosion resistance. The dispersion time of CNTs in the Al matrix has been inquired by Simões et al. [87] for microstructural evolution and found clusters of CNTs at the grain junctions mainly but CNTs dispersed well in Al matrix. Zhou et al. [88] observed the interface and interfacial reactions of MWCNTs reinforced Al composites below the melting temperatures of Al. Microstructure views MWCNTs is dispersed with Al matrix in close interfaces and aligned in the direction of the forming technique used hot extrusion.

### 2.2.3. Effect of SWCNTs/MWCNTs on Al/CNTs composites

SWCNTs/MWCNTs wt% or vol% have a significant effect on Al reinforced composites. Research exhibited that with the increases of MWCNTs, homogeneous distribution decreases and at the same time agglomeration increases with excessive plastic deformation [89]. Deterioration of the composite's property could be occurred due to the agglomeration of MWCNTs beyond 2 wt% in the Al matrix [90]. Effects of vol% in conductive properties of Al/MWCNTs reinforced composites were examined and till 0.75 vol% of MWCNTs effectively dispersed with alignment in Al matrix [91]. In another study, the effect of aspects ratio was presented in Al/MWCNTs composite [92]. The effects of MWCNTs morphology and diameter on the processing parameters and composites properties were carried out [93]. Al/MWCNTs based composites were produced by using a combined method of cryomilling and spark plasma sintering [94]. It has been carried out nanoindentation, microhardness, and tribological studies to understand the effect of MWCNTs content on mechanical properties. Characterization results confirm that retention of MWCNTs improved the mechanical properties and MWCNTs were partially damaged during cryomilling.

A literature analysis of factors affecting strengthening like CNTs dispersion, processing technique, degree of deformation and CNTs–matrix interface on the elastic modulus, strength and toughness of composites were analyzed by Bakshi and Agarwal [95]. The evaluation presented in the article showed that strengthening is not workable when chemical interaction between the metal matrix and MWCNTs, and intermediate phases may grow between MWCNTs and Al [96].

Effects of SWCNTs direction alignment on the composite property explored by Liu et al. [97]. Coatings on the SWCNTs and their effects on the molecular level of composite property have been assessed [98]. Coatings on MWCNTs have a positive effect on the homogeneous dispersion of MWCNTs compared to the uncoated MWCNTs [99].

#### 2.2.4. Coefficient of Thermal Expansion (CTE) and thermal behaviors of Al/CNTs composites

CTE plays an important role in the hardness improvement of Al/CNTs composites. Hardness is enhanced due to the dislocation density during cooling and the difference of CTE between the Mg and CNTs [26]. However, CNTs addition strengthen the matrix effects due to the difference in the CTE between the CNTs and Al matrix which increased the hardness of the composites [86]. A reaction takes place between Al and CNTs at 835–1085 °C and Differential Scanning Calorimetry (DSC) at about 835–1085 °C reveals that the reaction occurs between Al and CNTs is very gentle [71]. Moreover, very high temperature during the fabrication in Al/CNTs composites causes the Al in contact to vaporize and creates cavities [72]. Another study reports, even after the heat treatment the volume contraction of Al may increase to the mechanical adhesion of MWCNTs to the matrix [76]. The transition layer between Al matrix and MWCNTs should be considered because this layer lost the periodicity and became amorphous after the heat treatment. Below, Table 2 illustrates the fabrication methods of Al/CNTs composites for some readily adopted methods.

**Table 2.** Summary of Al/CNTs composites fabrication method.

Powder preparation	Fabrication method	CNTs content (vol%/wt%)	Remarks	Refs.
Ball milling	Spark plasma sintering followed by hot extrusion	1, 2, 4, and 6 wt%	Homogenous distribution of CNTs in the composites; dimpled fracture is observed which helps in dispersion strengthening and bridging mechanism is found	[1]
Chemical vapor deposition	Induction melting	0, 0.1, and 0.2 vol%	Homogeneous distribution of the nanotubes and no evidence of their segregation was found; induction melting holds strong promises for the fabrication of Al/CNTs composites	[19]
Ball milling	Combination of Rheocasting and Squeeze casting	0.5, 1.0, 1.5, 2.0, and 2.5 wt%	Well uniformly dispersed MWCNTs into the melt resulted in good distribution and less agglomeration of CNTs in the matrices of the produced A356/MWCNTs; combination of Rheocasting and Squeeze casting techniques is a novel processing technique for A356/MWCNTs composites	[26]
Ball milling	Cold compaction followed by hot extrusion	0.5, 1, 2, and 5 wt%	Carbide formation was observed in the composites; large aspect ratio of CNTs tends to agglomeration	[27]
Mechanical Stirring	Hot pressing followed by hot extrusion	5.0 and 10.0 vol%	No carbide formation is observed at the interface between CNTs and Al; mechanical properties is slightly affected by the annealing time	[31]
Ball milling	Nano scale dispersion followed by heating and cooling	0.8, and 1.6 vol%	Uniform dispersion of MWCNTs with improving the wetting property; composites obtained were highly reinforced and not to melt at a temperature far above the melting point of Al	[33]

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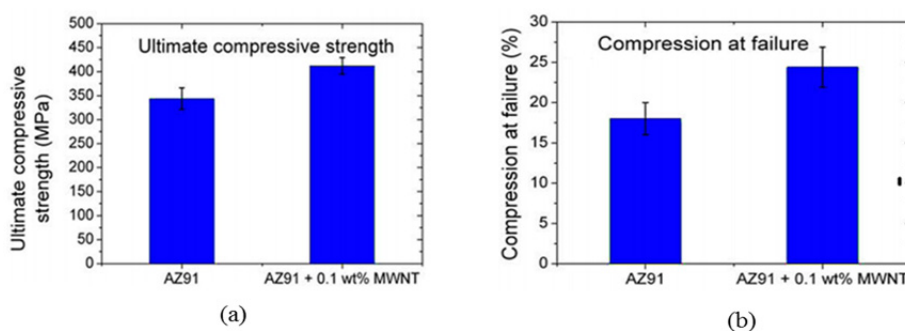
Powder preparation	Fabrication method	CNTs content (vol%/wt%)	Remarks	Refs.
-	Spread dispersion	0.5 wt%	Composites with fine structure can be produced by Spread dispersion method; tensile strength was improved up to 66%	[35]
Combination of ultra-sonication, magnetic stirring and shake-mixing	Compaction followed by sintering	0.5 wt%	Evenly dispersion of MWCNTs without the significant deformation or defects to the MWCNTs	[36]
Ball milling	Spark plasma sintering, Microwave sintering, Hot isostatic press sintering	0.5–2.0 wt%	The highest hardness values with 100% densification was obtained for spark plasma sintering; most suitable temperature for sintering was found 500°C	[40]
In situ chemical vapor deposition	Cold pressing followed by sintering and hot pressing	0, 1.5, 3.5, 5, and 6.5 wt%	Homogeneous dispersion of CNTs in the matrix; strong interfacial bonding between CNTs and matrix	[71]
-	Friction stir processing	0.5 wt%	Improved hardness and yield strength in friction stir processing; due to the presence of cavities in the material, mechanical properties were not enhanced in selective laser melted	[72]
Magnetic stirring	Selective laser melted			
Ball milling	Cold compaction followed by vacuum sintering and hot extrusion	0–3 wt%	A good agreement and optimized content of MWCNTs for density optimized MWCNTs content is suggested to be 2 wt%	[74]
Nano scale dispersion	Spark plasma sintering followed by hot extrusion	1 vol%	Enhancement in tensile strength with no degradation of elongation; effective load transfer from the matrix to the CNTs	[75]
Blending and mechanical milling	Compaction followed by pressure less sintering	0.25, 0.50, and 0.75 wt%	Yield stress and maximum strength were considerably increasing as the volume fraction of MWCNTs increases; milling time has an important effect on the composite properties	[76]
Ultrasonication	Infiltration method	1, 2, and 3 vol%	The compressive strengths were not necessarily proportional to the CNTs fraction rather it increased with the Al <sub>2</sub> O <sub>3</sub> fraction; an excessive amount of MWCNTs results to agglomeration	[86]

### 3. Mechanical properties of Mg/CNTs and Al/CNTs composites

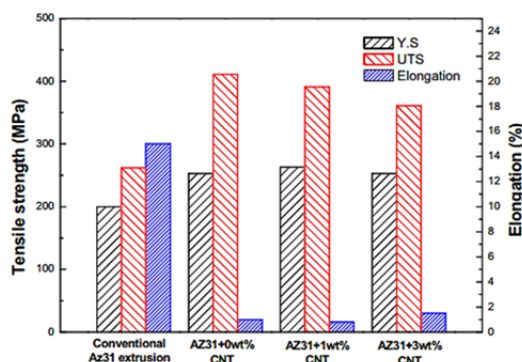
#### 3.1. Mg/CNTs composites

The investigations on mechanical properties of Mg/CNTs composite are very few as compared to other-CNTs reinforced metal matrix composites. Variations in the mechanical properties of Mg/CNTs composites are analyzed here. A maximum increase in tensile strength of 150% was shown in Mg/CNTs composites by Sinian et al. [100], where they used 0.55 vol% of CNTs. Such a

high enhancement in the mechanical property was achieved due to the Ni coating on the CNTs which improve the wetting in the matrix. In another study report presented that 90% increase in hardness where composites prepared by friction-stir processing, but CNTs wt% or vol% had not been indicated [101]. An increase of 20% in ultimate compressive strength and 36% increase in compression at failure was shown by Li et al. [54] with the addition of 0.1 wt% of CNTs. Figure 4 shows the increase in ultimate compressive strength and compression at failure. Very few analyses revealed the enhancement in yield strength. Goh et al. [102] investigated yield strength increase in Mg/CNTs composites and found 11% increase with the addition of 1.3 wt% of CNTs. In another study, Goh et al. [69] analyzed the fatigue behavior of Mg/CNTs composites and reached in the conclusion that the number of cycles of failure is decreased with the addition of CNTs. Kim et al. [38] applied mechanical alloying and hot extrusion process in Mg/CNTs composites fabrication and used 3 wt% of CNTs as a maximum value. The results disclosed that with the addition of CNTs in the composites ultimate tensile strength is reduced and elongation exhibited less than 2%. They claimed this occurs due to the formation of the MgO layer between the Mg and CNTs during fabrication. Figure 5 indicates the reduction of tensile strength and elongation about less than 2% in AZ31/CNTs composites.



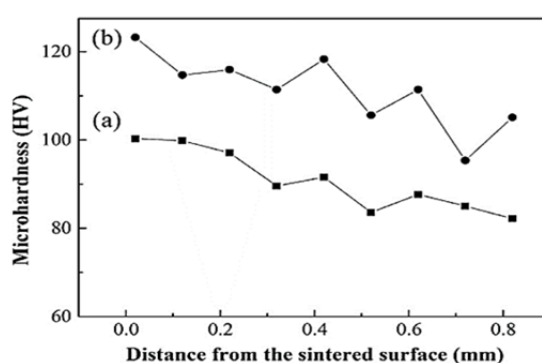
**Figure 4.** Comparison of AZ91 and AZ91–MWCNTs composites (a) ultimate compressive strength, and (b) compression at failure [54].



**Figure 5.** Ultimate tensile strength, yield strength and elongation comparison of AZ31–CNTs composites [38].

Yuan and Huang [103] utilized 1 wt% of CNTs in Mg composites and demonstrated improved

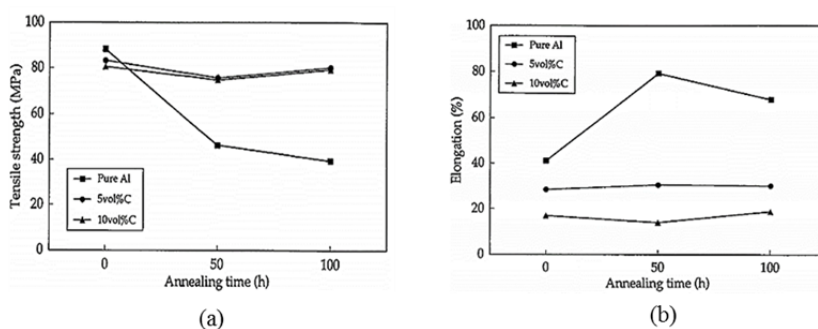
microhardness shown in Figure 6. By adding 1% of relatively short and straight CNTs in Mg composites, tensile strength was improved to 24% and ductility is reduced greatly from 14% to 5% [45]. Ni coated 0.3 wt% Mg/CNTs composites showed improved microhardness and tensile strength by 41% and 39% respectively [104]. Park et al. [105] presented Si coated 5 vol% Mg/CNTs composites tensile strength improvement by 44% and hardness 100%. Improved achievement in elongation and plasticity by 10.8% and 17% respectively by using 1 vol% of CNTs and claimed that it is achieved due to the homogeneous distribution and individual distribution of CNTs [106]. Again using 1 vol% of CNTs as a reinforcement and found 53% increase in yield strength and 18% increase in ultimate tensile strength [107]. Li et al. [108] used 0.5, 1 and 1.5 vol% of CNTs in the Mg–6Zn matrix. They observed the highest yield strength and ultimate tensile strength at 1 vol% and modulus of elasticity at 1.5 vol%. Most of the study concluded that extensive mechanical property is obtained due to the homogeneous distribution of CNTs in the matrix.



**Figure 6.** Microhardness (a) monolithic AZ91D, and (b) AZ91D/MWCNTs composite [103].

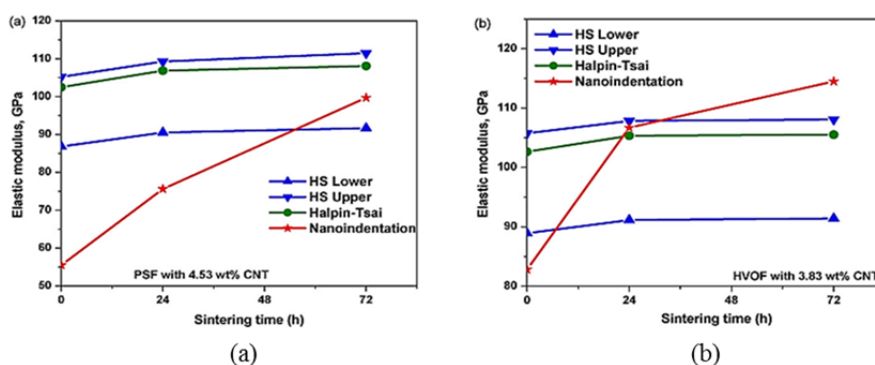
### 3.2. Al/CNTs composites

A lot of investigations on Al/CNTs composites were done. But most of the analysis was on microstructure, XRD pattern, SEM, TEM, grain analysis rather than mechanical property investigations. However, for the first time, 100% increase in tensile strength is reported by Kuzumaki et al. [31]. They also reported a reduction in elongation less than 20% with respect to annealing time with 10 vol% of CNTs addition by using hot-press and hot-extrusion methods. Figure 7 shows the tensile strength improvement and elongation reduced of Al/CNTs composites.



**Figure 7.** (a) Tensile strength, and (b) Elongation of Al/CNTs composites [31].

The maximum increase of 129% in tensile strength has been determined by adding 5 vol% of CNTs [109]. But, another study reported that with the addition of 5 vol% of CNTs may cause deterioration in hardness [110]. This is occurred due to the poor interfacial bonding and agglomeration of CNTs in Al matrix. From the past, researchers have tried to embody CNTs in the powder metallurgy route associated with Spark Plasma Sintering and/or hot deformation within the range of 1–6.5 vol% [109,111–113,83]. Another experiment revealed that, tensile strength and hardness were increased by 184% and 333% respectively with the incorporation of 6.5 vol% of CNTs in the chemical vapor deposition method and obtained homogeneous distribution of CNTs and good interfacial bonding [114]. With the addition of 1.6 vol% of CNTs in the nano-scale dispersion method, a very homogeneous distribution is obtained, and compressive yield strength enhanced by 350% [83]. Thermal spraying methods have been adopted in CNTs reinforced composites [111,115,116] and found 72% enhancement in the microhardness, 78% in elastic modulus, a marginal enhancement in tensile strength, and 46% decrease in ductility with the amalgamation of 10 wt% of CNTs in the composites. It has been also reported that elastic modulus is further increased to 80%, after the sintering of the sprayed coatings showed in Figure 8.



**Figure 8.** Elastic modulus with respect to sintering time (a) plasma spray formed with 4.53 wt% of CNTs, and (b) high velocity oxyfuel spraying with 3.83 wt% of CNTs [117].

It is informed that in plasma spray processing with the incorporation of 12 vol% of CNTs improved the elastic modulus up to 40% and elastic recovery could be increased with the further addition of CNTs by Bakshi et al. [118]. In another study of Bakhshi et al. [119] used cold spraying for CNTs reinforced aluminum composites and presented no further quantification on the enhancement in the mechanical properties of the composites due to the heterogeneous distribution of the CNTs in the composites. Therefore, it is suggested that uniform and homogeneous distribution of the CNTs and good interfacial bonding is the prime concern for the mechanical properties enhancement in the Al/CNTs composites.

#### 4. Strengthening mechanisms of Mg/CNTs and Al/CNTs composites

After the consolidation of the metallic matrix and CNTs, the form composites may not show the required level of mechanical properties. Therefore, the consolidated composite requires some further treatment for strengthening at the anticipated level. Various strengthening mechanisms for Mg/CNTs and Al/CNTs reinforced composites have been proposed based on the geometry and physical

properties of CNTs. To predict the strength of Mg–Al/CNTs reinforced composites, it is necessary to understand the strengthening mechanism of CNTs in composites.

Different analysis and mathematical strengthening method show that the average grain sizes of the Mg, the volume fraction of the CNTs, the aspect ratio of the CNTs to Mg, shear modulus and yield stress of the Mg matrix has a significant impact on the strengthening of Mg/CNTs composites [54,108,120,121]. Also, some research has considered the length of the CNTs [78,120,121] for strengthening. The mathematical formula has developed [54,108,78,120,121] to consider the above mention factors to determine the correct amount of CNTs in Mg matrix with the expected level of yield or tensile stress, or even comparing with the shear modulus. Shear-lag model, and the Halpin–Tsai model, linear model, and the root mean square model [122] are used to analysis the Mg/CNTs composites strength. Since the Mg/CNTs composites are still a new grade of materials, the absolute strengthening mechanism is not available so far.

In case of Al/CNTs, CNTs have a CTE is of  $\sim 10^{-6} \text{ K}^{-1}$  where commercial pure Al has CTE of  $23.6 \times 10^{-6} \text{ K}^{-1}$  which shows much greater CTE than CNTs. Therefore, during the fabrication of Al/CNTs composites there exists a significant coefficient of thermal expansion mismatch between the matrix and CNTs, which would punch dislocations at the interface, guided to work hardening of the matrix. In the Orowan looping [123] mechanism of Al/CNTs strengthening, bending of the dislocations between the CNTs is occurred due to the motion of the dislocations, which is obstructed by nanometer sized CNTs. This induces back stress, which prevents further dislocation and increases the yield stress. In Al matrix, fine precipitates of CNTs due to their small diameter leading to a lower density of Griffith flaws. That's why the dislocation density generation is likely to be higher, which in turn would result in an increased strengthening of Al/CNTs [124].

Also, strengthening affected by the load transfer between the interfacial shear stress from the matrix to the reinforcement called shear lag depends on the aspect ratio of CNTs to Al and stiffness of CNTs [125]. Depending on the types of the CNTs, such as for SWCNTs the aspect ratio is around  $\sim 1000$ , and MWCNTs have aspect ratio of in the range of  $\sim 100$ . Hence SWCNTs has good wetting properties in Al matrix for effective interfacial shear stress transfer. Though the bonding between the Al and CNTs is too poor due to the surface tension discrepancy of Al and CNTs, an addition of salt such as  $\text{K}_2\text{ZrF}_6$  salt can be used for the bonding improvement between Al and CNTs.

We know that the CTEs for Mg and Al are close, but its grain size as well as aspect ratio has a variation of compare to CNTs. Therefore, the dislocation of the Mg–Al/CNTs must also consider and view microscopically to identify the good model that best fits with the experimental analysis to describe the strengthening.

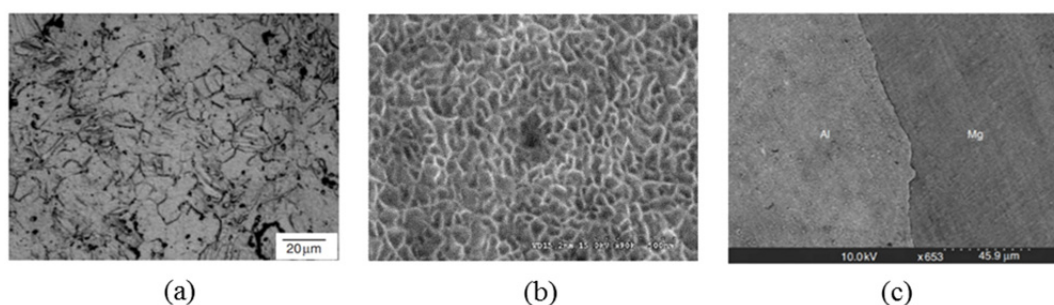
## 5. Summary of findings

Nowadays, various new approaches and methods of bimetallic composite formation are becoming popular among engineers. It is high time; a suitable forming process is developed to industrialize the unique nature of CNTs reinforced materials in Mg–Al bimetallic matrix focusing on mechanical characteristics and financial aspects. A variety of metals or their alloys can be used to prepare a matrix for CNTs/MWCNTs reinforced materials. Each metal matrix offers unique characteristics and undergoes distinct processing techniques. To study the Mg–Al/CNTs bimetallic matrix composites, we need to study the prospect of Mg/CNTs, Al/CNTs, Mg–Al composites separately shown in Figure 9. For this purpose, we have studied various articles. The summary of the



findings is discussed below:

(a) The study tracked that no other alloying is found better than Mg–Al as the bimetallic matrix depicted in Figure 9 [126,127]. Mg–Al combination has good reinforcement properties experienced by many investigators [128,129]. Mg–Al and their alloys possess low weight and excellent properties as structural materials. Again Mg–Al/CNTs possess great potential in the automotive industry because of the prominent combination of high specific strength from Mg alloy and excellent corrosion resistance from Al alloy [130]. The previous works on the Mg–Al and their alloy also depict the suitability to form a bimetallic matrix for CNTs. It can be improved and optimized easily with the addition of CNTs reinforcement and a good fabrication technique. A few years ago, Wong et al. [131] processed Mg–Al based nanocomposites using hybrid microwave sintering. At that work, physical properties were improved close to the melting temperature.



**Figure 9.** Microstructure of (a) Mg Rod, (b) Al rod, and (c) Al–Mg bimetallic composite [126].

(b) As a composite fabrication prediction, the powder obtained by powder metallurgy has sophisticated mechanical properties like toughness, fatigue, fracture, and strength for its uniform particle distribution and chemical homogeneity. In developing prospected Mg–Al/CNTs reinforced composite, research must be focused on developing Mg–Al powder and improving the adding techniques of CNTs in the metallic matrix. This also includes fabricating corrosion-resistant films/coatings, micro/nano dispersion, bonding process etc. This hypothesis must translate into conceptual formation with the buildable forming process. After preparing subsystems, constituents and consolidation assembly, the reinforcement must be treated for practical application. Afterward, the composition, performance data collection, combine property improvements and adjusting composition and processing for best performance are required. Therefore, it seems that the work of CNTs reinforced in the bimetallic matrix will be a new addition to engineering science.

(c) Therefore, the first step to achieving Mg–Al bimetallic CNTs composite forming is a powder metallurgy process. But, numerous powder metallurgy methods are available depending on the particles, size, bonding, and nature of properties required for Mg/CNTs and Al/CNTs separately. Very little work was done on Mg–Al bimetallic CNTs composite. Below we have tried to mention some Mg/CNTs and Al/CNTs productions methods with different approaches of composite forming.

(d) The best anticipated processing technique for forming Mg–Al/CNTs is sintering (Tables 1 and 2). Our survey study (Tables 1 and 2) found that the use of sintering (about 50%) as a consolidation technique is more popular among the researchers and the reported mechanical properties are also good. Sintering offers a reduction in processing time, volumetric and uniform heating, precise selective and controlled heating, improved properties, and environment-friendliness.

Microstructural characterization of Mg/CNTs and Al/CNTs revealed that the use of sintering in nanocomposite production promotes the dispersion and embedding of individual CNTs in the metallic matrices [36,40,51,53,56,58,59,61,63,69,71,74–76]. Also sintering does not suffer from any reaction by products. Even CNTs are not damaged during the preparation and fabrication of the Mg–Al nanocomposite.

(e) During the fabrication process if pre-treatment executes, some CNTs structure in the processed composite may be damaged [62,76]. However, in some cases, they may adversely affect the properties due to the non-uniform dispersion of CNTs in the metallic matrix and poor interfacial adhesion at the CNTs to metal interface [132,133]. The unfavorable chemical reaction of CNTs with the matrix and low compact ability are the most significant challenges, requiring more concentration [134,135]. Besides, for all the mixing stages, the prevention of oxidation is a crucial fact to look up. The addition of chemical catalysts such as alcohol and acid mixture will help homogeneous dispersion of CNTs and enhance adhesive properties. Since the Mg and Al have almost the same melting point, the selection of fixed sintering temperature is enough to add up the constituents. The heating time and the temperature gradient must be set as per the sintering facility and volume of the composition. However, the efficient heating by rapid microwave sintering must be multidirectional in the presence of some good susceptors to shorten the sintering time.

(f) In order to produce optimized composites, strengthening mechanisms involved in Mg–Al based CNTs composites are very important [136,137]. Based on the above-mentioned discussions, any production method must have two focus points. Firstly, manufacturing and experimental facility; secondly, microscopic analysis of Mg–Al based CNTs composites. Microstructural characterization studies to be held at different stages of the forming process is a vital concern to investigate the existence of porosity, reinforcement distribution, matrix reinforced interfacial integrity, grain size, and grain morphology. Since no previous records of Mg–Al bimetallic CNTs are readily available, Therefore, the researcher must suggest a verified strengthening mechanism for this newly developed composite type. However, the shear lag theory and thermal mismatch condition analysis can easily help to understand the strengthening mechanism of the Mg–Al/CNTs. The adopted strengthening mechanism may be Orowan strengthening [138,139], Hall–Petch strengthening, and load transfer effect [140]. The selected secondary processing may be hot extrusion (Tables 1 and 2) to obtain the improve properties by modifying the percentage of reinforcement distribution. Further in-situ observation of Mg–Al to CNTs composites will make a better understanding of the composite behavior and suppress the crack formation.

## 6. Conclusion

In this review, a comparative study has done successfully for Mg/CNTs and Al/CNTs and the prospect of CNTs reinforced Mg–Al bimetallic composites has investigated. The focus of the study was forming technology, and subsequent results presented by the research. Though there is no available direct study of Mg–Al/CNTs composites, Mg/CNTs and Al/CNTs have studied carefully to sort out the resourceful information for future research perspective. According to the findings of the short survey study, the sintering process has appeared effective for Mg–Al/CNTs, Mg/CNTs and Al/CNTs composite formation. Thus, this process is assumed effective for Mg–Al bimetallic matrix with CNTs. The diffusion processes with phase reformation are very prominent for Mg/CNTs, and Al/CNTs. Thus, similar results are anticipated for Mg–Al composites with CNTs. It focuses on

obtaining optimum and good reinforcement by achieving homogeneous dispersion of CNTs in the bimetallic matrix and good interfacial bonding at the metal-CNTs. Thus, excellent mechanical, electrical, and magnetic properties of the developed composites are predicted with the dual benefit of Mg and Al metals. The microstructure evolution of the Mg–Al bimetallic composites and the bonding characteristics of bimetallic matrix-CNTs at the interfaces may be characterized through optical microscopy, XRD, HRTEM, FESEM etc. Later the strengthening mechanism, fatigue and crack growth of the metals-CNTs composite may be analyzed based on the fracture morphologies.

## 7. Research prospect of CNTs reinforced Mg or Al alloys

Future research prospect should be towards the investigations on different series of Mg and Al alloys other than pure metals that will create the potential industrial applications. To produce the mass scale of Mg–Al/CNTs composites, attention should be paid on the homogeneous dispersion method of CNTs in Mg or Al matrix and effective bonding technique between the matrix and CNTs. Special attention should be paid to minimize the CNTs damage during solid state processing. To better understand the strengthening mechanisms and developed high performance, there is still a vast scope of work could be conducted on theoretical studies of CNTs reinforced Mg and Al alloys, involving the relationships between processing parameters, micro and nano-structures, and multi-scale mechanical modelling and simulation.

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## Conflict of interests

The authors declare no conflict of interest.

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