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# Production methods of CNT-reinforced Al matrix composites: A review

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A B S T R A C T

Carbon nanotubes (CNTs)-reinforced Al composites have attracted attention due to their high specific strength and low density, which makes them suitable for the use in aerospace and automobile industries. In this review, preparation methods of Al/CNTs composites for achieving a homogeneous desperation of the CNT in the Al matrix are summarized. In addition, the effect of processing methods on carbon nanotube distribution and enhancement of mechanical properties such as toughness, wear behavior and hardness of the nanocomposites are reviewed. Improvement of mechanical characteristics was observed by the incorporation of carbon nanotubes into aluminum matrix. The strengthening factors gained by the carbon nanotubes addition are the interface of metal and CNTs and the chemical and structural stability of CNTs.


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## 1. Introduction

As composite materials have the capability of developing lightweight materials with tunable properties [1-3] for high-performance applications, they have attracted attention of researchers in recent decades [4-7]. Composite materials are defined as two or more components that are chemically distinct and their interfaces are clearly separated. The matrix can be either metals, ceramics, or polymers [8-11]. The composite materials show high strength and stiffness [14, 15]. The invention of the airplane and the corresponding industry have increased the demand for the development of high strength and lightweight materials [17]. By the enhancement of the stiffness and strength of materials, it is possible to use reduced the dimensions and mass for a particular load bearing application. In aircraft and automobile industry, the reduction of the required size can provide some advantages, including the fuel efficiency improvement and an increase in payload. The increase in the efficiency of fuel consumption of engines is highly desirable due to global oil re-

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source depletion [19]. As most light metallic materials and alloys cannot offer both high stiffness and strength to a structure, metal matrix composites (MMCs) have been developed in which the metal matrix provides the ductility and strength while the reinforcing component provides the strength and/or stiffness. The reinforcing material can be either particles, whiskers, or fibers of a high stiffness metal or ceramic. Some properties of metal matrix composites such as low thermal expansion coefficient and high thermal conductivity can be achieved, which make them good candidates for applications in the electronic packaging field. Nowadays, MMCs are extensively used in the aerospace and automobile industry [22].

Carbon nanotubes (CNTs) are composed of rolled-up sheets of graphene in cylindrical form often in the nanometer range, which shows remarkable thermal, electrical, and mechanical, characteristics. CNTs exhibit almost one hundred times tensile strength (~150 GPa) and approximately six times elastic modulus (~1 TPa) than do high strength steels depending on their diameter, length, orientation, and chirality [25]. The use of lightweight CNTs as nano-reinforcement for composite materials is highly promising. The aim of using carbon nanotubes is to incorporate their remarkable physical and mechanical properties into the bulk of engineering materials [22, 28]. Carbon nanotubes have been used for the reinforcement of metal, ceramics, and polymer matrices. Among these materials, polymer/CNTs composites have been extensively prepared via interfacial covalent functionalization, solution evaporation having significant energy sonication, repeated stirring, and surfactant-assisted processing. On the other hand, metal/ceramic reinforced with CNT has not been investigated widely. To disperse carbon nanotubes in the matrix uniformly, several synthesis methods have been employed for the production of carbon nanotube/ceramic or metal matrix composites such as hot extrusion, spark plasma hot pressing, sintering (SPS), and in situ synthesis [31].

Carbon nanotubes are also promising reinforcements for aluminum that has recently emerged. The poor dispersion of carbon nanotubes and agglomeration in metallic matrices are major difficulties in achieving the reinforcing effect of CNTs in metallic matrices successfully [23, 32]. Different processing methods have been investigated to solve this problem. In this paper, various preparation methods for CNT-reinforced Al composites have been reviewed.

## 2. Carbon nanotubes (CNTs)

The most important and abundant element in nature is carbon, and its pure forms are diamond and graphite. CNTs were first discovered by Iijima in 1991 [33], and by the discovery of the fullerenes and graphene, these materials have attracted attention from researchers. The CNT is in the form of a cylinder consisting of rolled-up graphene sheets with a regular hexagon structure, which can have a diameter of several folds smaller than its length. In the structure of an ideal CNT, graphene sheets consisting of hexagonal-structured carbon atoms are rolled up and create a hollow, tube-like structure. Carbon nanotubes are divided into two categories of single-walled carbon nanotube (SWCNT) and multi-walled carbon nanotube (MWCNT) that is based on the number of graphite sheets incorporating in their structure. In the MWCNTs structure, there are a number of concentric tubes of graphene being fitted on each other [34]. The range of MWNTs and SWNTs diameters is between 2 nm to 100 nm and 0.7nm to 2 nm, respectively, but the length of these nanotubes can vary from several millimeters to micrometers [35]. As a result of properties such as remarkable chemical stability, good absorbability, specific surface area, and unique electronic structure, carbon nanotubes are considered as promising materials as catalyst carriers. Moreover, CNT s are reported to have the capability to be utilized as a support for dispersion of functional materials for the improvement of some properties like structure, activity, surface area, and conductivity [36].

As a result of the practical lack of scattering mechanisms, which increase the mobility of the carrier along the tubes, carbon nanotubes exhibit a very high electrical conductivity [37]. Furthermore, through changing the nanotube diameter or helicity, the electronic characteristics of single-walled nanotubes can be altered from semiconductor to metallic behavior or vice versa [38]. Therefore, they are being studied for high-speed electronic applications. In nanocomputing and nanoelectronics usage, CNTs can facilitate heat dissipation due to the high thermal conductivity (about 3000 W/ mK) and are comparable to diamond [39]. Through increasing mechanical integrity and electrical connectivity, CNTs would improve the cycle life in lithium-ion batteries for cellphones and computers. An interesting application of carbon nanotubes is producing light. This is due to the recombination of holes and electrons across the semiconductor gap, resulting in the emission of infrared radiation, which is near to the optical windows related to optical fibers [40].

## 3. Metal matrix composites (MMCs)

Metal matrix composites (MMC) are composed of a metal matrix and reinforcing components [41, 42]. Reinforcing components are incorporated into the matrix in the form of small fibers, continuous fibers, whiskers, and particles. In the case of MMCs, the reinforcement can be used in the form of particles, small fibers or whiskers, and continuous fibers or sheets [43, 44].

Metal matrix composites that are discontinuously reinforced with particle, whiskers, or short fibers, are of great importance. One reason is the cost of production, which is an essential factor for large volume production. MMCs can be prepared by conventional metallurgical processes, including powder metallurgy [45] or casting [46], and they can be processed by conventional secondary techniques such as extrusion forging [47], and rolling [48]. They can be used in higher temperatures compared to unreinforced metals, and they exhibit improved strength, modulus wear resistance, and thermal stability. They possess relatively isotropic characteristics in comparison with the composites reinforced by fibers [49].

The metal matrix can be different metals such as Cu [50, 51], Ti, Al, Mg, their alloys, and intermetallic compounds. Due to excellent strength, low density, high toughness, and resistance to corrosion, Al alloys, are considered as important materials for aerospace applications [35, 52]. In the automotive, aerospace, and sports industries in where both lightweight and mechanical properties such as high strength and stiffness are desired, carbon nanotube reinforced metals are capable of revolutionizing these industries [53].

## 4. Aluminum matrix composites (AMCs)

In aluminum Matrix Composites, the matrix is Al or Al alloy, and the reinforcing component is embedded in this metal matrix [54]. This reinforcement is commonly non-metallic and usually a ceramic material like $Al_2O_3$, SiC, CNTs, and etc. [55-60]. The type of reinforcements and their volume fraction in the matrix alters the properties of AMCs. AMCs have some advantages in comparison with unreinforced materials, including improved stiffness, higher strength, controlled coefficient of thermal expansion, enhanced high-temperature behavior, reduced density, higher abrasion and wear resistance, enhanced electrical properties, and enhanced damping capabilities [61-63].

A significant increase in the elastic modulus of unreinforced Al from 70 GPa to 240GPa can be observed by adding 60 vol. % continuous Al fiber. Moreover, reinforcing pure Al with 60 vol. % alumina fiber can reduce the expansion coefficient (24 ppm/°C to 7 ppm/°C. Also, it has been shown that adding 9 vol% Si and 20 vol.% SiCp to Al has the potential of having wear resistance comparable to that of grey cast iron [64]. Overall, through the incorporation of suitable reinforcing components with appropriate volume fraction, it is possible to enhance the technological properties of Al and its alloys greatly. Aluminum matrix composites present such superior combination of characteristics that common monolithic materials are not able to compete [65]. Aluminum matrix composites have been utilized in various functional, structural, and non-structural engineering applications owing to their good performance, and environmental and economic advantages [66]. Lower airborne emissions, lower fuel consumption, and less noise is the key advantages of aluminum matrix composites for utilization in the transportation sector. AMCs are becoming inevitable material in the transport industry because of ever-increasing environmental concerns and improved fuel consumption efficiency [67].

Based on the form of reinforcing components aluminum matrix composites can be categorized into four different types: (I) Mono filament-reinforced AMCs (MFAMCs) (II) Continuous fiber-reinforced AMCs (CFAMCs) (III) Whisker-or short fiber-reinforced AMCs (SFAMCs) (IV) (PAMCs) [68]. Particle incorporating in AMCs are generally equiaxed ceramic materials having an aspect ratio smaller than 5 [69]. For structural and wear resistance applications, the amount of ceramic components is less than 30 vol. %, while the volume fraction can reach to 70% for applications in electronic packaging fields. In comparison with other types of AMCs, PAMCs exhibit lower mechanical properties; however, compared to pure Al or Al alloys have better properties. The aspect ratio of reinforcing components is greater than five in SFAMCs, but they are non-continuous. AMCs, including short alumina fibers, is one of the most and first popular Aluminum composites developed to be used in pistons. In CFAMCs, the reinforcing components can be alumina, SiC or carbon continuous fibers having a diameter lower than 20 μm. These continuous fibers can either be braided, woven, or parallel before undergoing the production process of composites [70]. MFAMCs are composites consisting of fibers with a large diameter of 100 to 150 μm that are prepared via chemical vapor deposition (CVD) of B or SiC into a W wire core or carbon fiber. In comparison with multifilaments, monofilaments have lower bending flexibility [71].

Production of aluminum matrix composites at an industrial scale is carried out through two main processes. (1) Liquid state processes, (II) Solid-state processes. Solid-state processes include physical vapor deposition, diffusion bonding, and powder mixing and consolidation (PM processing). In-situ processing, spray deposition, infiltration process, and stir casting are categorized under liquid state processes [64].

A 100% increase in the tensile strength with the incorporation of 10 volume percent of carbon nanotube was first reported by Kuzumaki et al. [72]. Researchers have tried to increase the amount of carbon nanotube up to 6.5 vol.% in Al/CNT composite using the powder metallurgy route [73] and secondary processes such as hot deformation and SPS [74]. The incorporation of 5 vol.% CNT was shown to increase the tensile strength to 129% [75]. In contrast, a reduction of hardness by the incorporation of 5 volume percent was observed by Salas et al. [76] using in a shock-wave consolidation. The reduction of properties is a result of the agglomeration of carbon nanotubes in the Al phase and weak interfacial adhesion of the two phases. Laha et al. [77, 78] studied the effect of adding 10 wt% CNT to aluminum coating through thermal spraying methods, and their results showed that CNTs improved the elastic modulus by 78%, hardness by 72%, and decreased ductility by 46%. The elastic modulus of the sprayed Al/CNT composite coating sintered at 673 K was proposed to increase by 80%, which was reported to be a result of a decrease in porosity and residual stress [79].

## 5. Strengthening mechanisms in carbon nanotube-reinforced composites

Fibrous reinforcing components such as carbon nanotubes are used to enhance the tensile strength elastic modulus of the matrix. These improvements are a result of the higher strength and stiffness of carbon nanotubes in comparison with the metal matrix. Researchers have tried to understand the mechanisms incorporating in the strengthening of fiber-reinforced composites. The model applied to study CNT composites is the shear lag models [80] related to all conventional composites reinforced with fibers and is presented below:

$$\frac{l_f}{D_f} = \frac{\sigma_f}{2\tau_{mf}} \tag{1}$$

where $\sigma_f$ is the stress transferred to fibers through the interface of the matrix and the reinforcement and has a relation with the shear stress ($\tau_{mf}$) between the two phases. $D_f$ and $l_f$ are the diameter and length of carbon nanotubes, respectively. The larger aspect ratio of CNTs leads to the larger load transfer to the reinforcement and hence higher reinforcing efficiency is achieved. $\sigma_f$ equals to the fracture strength of carbon nanotubes for a critical length $lc$. The fracture strength of the composite when $l<l_c$ is calculated by:

$$\sigma_c^{Frac} = V_f \sigma_f^{Frac} \left(\frac{l}{2l_c}\right) + V_m \sigma_m^{Frac} \tag{2}$$

Al/CNT composites synthesized via ball milling and extrusion were shown to follow this relation well [18]. An undesirable reaction that can occur in metal matrix nanotube composites is the formation of carbide at the interface of metal and CNTs. Thus, the shear strength of the formed phase affects transferring the stress to carbon nanotubes. If the applied stress is higher than the shear strength, fiber pulls out occurs due to the carbide layer fracture [81]. The relation derived for the strength in the presence of a carbide interfacial layer by Coleman et al. [82] is given as:

$$\sigma_c = \left(1 + {}^{2b}/_{D}\right)\left[\sigma_{shear}\, {}^{l}/_{D} - \left(1 + {}^{2b}/_{D}\right)\sigma_m\right] V_f + \sigma_m \tag{3}$$

where the interface shear strength is denoted by $\sigma_{shear}$. D and b are the diameter of carbon nanotubes and the width of the carbide layer, respectively. It has been reported that the strength measured experimentally (83.1 MPa) is much less than that calculated using this relation (226 MPa). This is a result of different factors that are not taken into account in this model, including the clustering of carbon nanotubes, uniformity of the interfacial carbide phase, and porosity [77]. For example, the carbon nanotube elongated clusters has been observed through the microstructural study of Cu/carbon nanotubes composites synthesized via spark plasma sintering of mechanically alloyed powders post-processed by cold rolling [83].

In the case of Al/CNT, the strengthening mechanisms are proposed to be the generation of dislocations due to the mismatch between thermal expansion of the matrix and reinforcement as well as precipitation hardening through the Orowan looping mechanism. However, these mechanisms have not been observed yet. To achieve the strengths near the theoretical strength, it is important to disperse CNTs uniformly in the matrix [12].

Elastic modulus enhancement is due to the high tensile modulus of carbon nanotubes (350–970 GPa). Most of the models that have also been developed for polymer/carbon nanotubes are applicable for metal matrices [84].

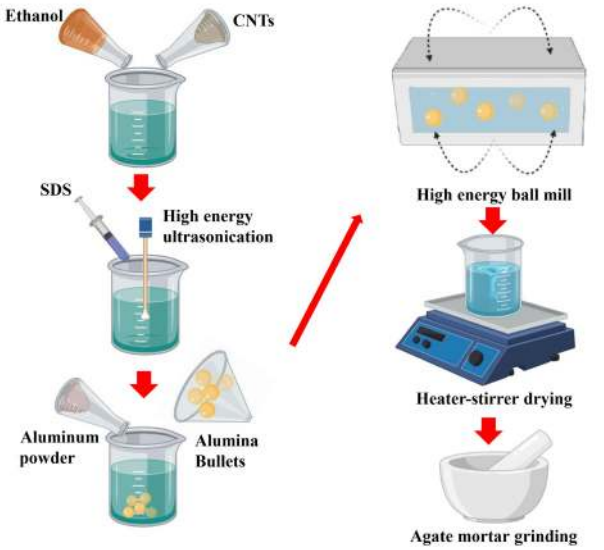


**Fig. 1.** Dispersion of CNTs in Al through ultrasonication.

## 6. CNT-reinforced AMCs processing techniques

*6.1. Powder metallurgy*

One of the widely-used synthesis methods of MMCs is powder metallurgy, which involves cold pressing of powders and subsequent sintering, or hot pressing [85]. Various techniques are used for the preparation of the metal matrix composites in the form of particles or whiskers. The first stage in the preparation of MMCs via powder metallurgy is the blending of particles and the matrix powder to achieve a homogeneous distribution. After blending, the powder mixture is cold-pressed (is called green body) and reaches to 80% of theoretical density. In order to remove the moisture of the powder surface, the prepared green body is degassed in a sealed container. Finally, the compacted powder undergoes sintering process under isostatic or uniaxial pressure to prepare fully compact composites [86]. One of the challenges in preparation of these composites is the dispersion of CNTs in the metal matrix. The improvement of properties could be achieved only when there is a uniformly distribution of CNTs. If such dispersion is not obtained, agglomerated particles and the micro-pores will be formed throughout the microstructure. In order to alleviate this issue, a variety of methods has been developed for the effective dispersion of CNTs in the matrix including ball milling, ultrasonication, application of surfactants, and metallization [24, 87].

*6.1.1. Ball milling*

In ball milling process, metal powders undergo repeated fracture and welding by several hard balls in the milling container. This method can be used for the dispersion of CNTs in the metallic matrix [88]. The metal powders and CNTs are entrapped between the container wall and balls or balls themselves resulting in the separation of the nanotubes, destruction of the agglomerates, and dispersion of CNTs among the metal particles. The dispersion degree depends significantly on milling time. By the increase of the milling time, the agglomeration is reduced and the metal particles change from round shape to flattened shape in which CNTs are embedded [89].

*6.1.2. Ultrasonication*

Ultrasonication is a technique in which high frequency ultrasound waves are applied to disperse CNTs in an organic solvents or aqueous surfactants. To reduce the hydrophobicity of CNTs, their surfaces are modified by polymer adsorbates and surfactants to enhance their solubilization in water. Through ultrasonication, the collapse of micro-bubbles produced during the process creates a high local shear stress at the CNT bundles end and when a gap or a gas bubble is formed, surfactants are absorbed on CNTs. The surfactant-coated carbon nanotubes is produced by unzipping process along the longitudinal axis [90]. Fig. 1 illustrates dispersion of CNTs in Al through sonication followed by ball milling process.

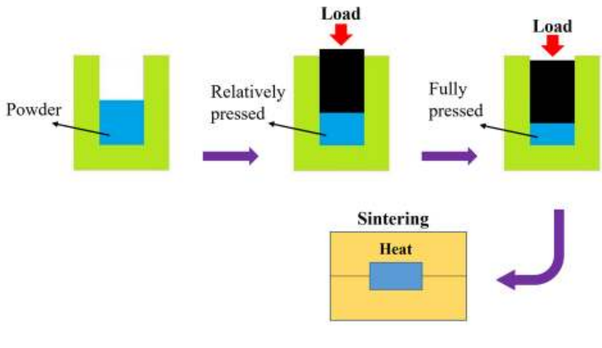


**Fig. 2.** Schematic illustration of cold pressing and sintering.

*6.1.3. Application of surfactants*

A way of the surface modification and prevention of agglomeration and rebinding of CNTs is using surfactants. Generally, the surfactants have two parts in their structure: a hydrophobic and a hydrophilic part.

The reactive agents produce steric or electrostatic repulsion between CNTs particles and reduce their surface energy leading to the improvement of the suspensions metastability. According to the head group charges, the surfactants are anionic, cationic, and nonionic or zwitterionic [91].

*6.1.4. Metallization*

The CNTs surface can be coated with some metals including W, Ni, Mo, Co, and Cu in order to prevent the agglomeration of the reinforcement, enhance the interfacial adhesion between the matrix and the reinforcement, and inhibit the reaction between them [92].

The other important factor affecting the properties of the composites produced by powder metallurgy is the sintering process. The samples can be sintered either above or below the matrix solidus temperature [93]. Different sintering processes employed for the preparation of Al/CNT nanocomposites are described below:

*6.1.5. Cold press and sintering*

The schematic illustration of cold pressing and sintering is shown in Fig. 2. George et al. [12] prepared Al and CNT powder mixture via ball milling followed by the compaction of the milled powder in a circular die applying120 KN load. The sintering process of the billets was carried out in an $N_2$ environment. According to the results, the mechanical properties of carbon nanotube reinforced aluminum, e.g. Young's modulus were enhanced [94].

Sivananthan et al. [13] prepared the powder mixture of MWCNT and Al via ball milling with 10 steel balls with a 15 mm diameter for 10 h at room temperature. Then, by applying a pressure under increased temperature, the powder mixture was converted to the green body and sintered for 1 h at 600 °C in an Ar atmosphere. They indicated that the

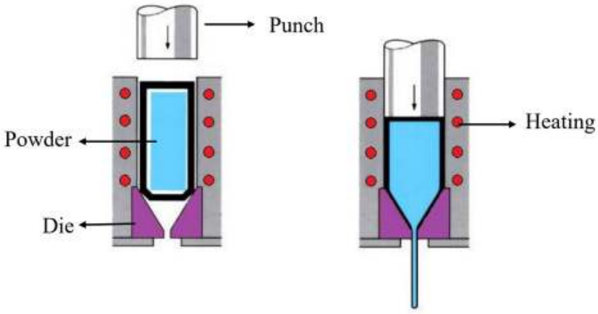


**Fig. 3.** Schematic illustration of the hot extrusion process.

Al physical properties were improved by the addition of carbon nanotubes. However, the thermal and electrical conductivities decreased by the addition of 0.5 to 3 wt. % carbon nanotubes. The results showed that the incorporation of CNTs does not improve the electrical and thermal conductivities of the Al matrix. Therefore, Al/CNT composites may not be suitable for applications requiring conductive properties, and they might be employed for thermal and electrical resistive applications.

*6.1.6. Hot extrusion*

Esawi et al. [16] mixed up 5 wt.% carbon nanotubes filler using a ball mill. To consolidate the ball-milled powder mixture, the mixture was cold-compacted and sintered by hot extrusion. The hot extrusion process is illustrated in Fig. 3. They investigated the effect of the carbon nanotubes content on the mechanical characteristics of the nanocomposites. The results revealed that stiffness enhanced up to 23% and tensile strength enhancements up to 50% in comparison with pure Al. In the nanocomposites with 5wt. % carbon nanotube, the formation of the carbide phase was recorded. In the composites with CNT content higher than 2 wt. %, the dispersion of the reinforcement was difficult, and hence, the expected enhancement in mechanical characteristics with increasing of the carbon nanotubes content in the matrix was not fully understood.

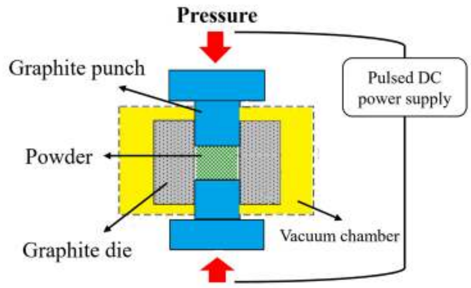


**Fig. 4.** Schematic illustration of the SPS process.

Choi et al. [18] produced Al/MWNTs composites via ball-milling and hot extrusion. The efficiency of reinforcing by carbon nanotubes follows the discontinuous fibers volume fraction rule in the grain size less than 70 nm. The research asserted that it is possible to produce large-scale Al/CNTs composites with uniaxially aligned carbon nanotubes via a conventional powder metallurgy method. The strengthening efficiency follows the discontinuous fibers volume fraction rule for composites possessing grain sizes of 200 nm and 72 nm, and nanotubes are able to effectively transfer loads.

*6.1.7. Spark plasma sintering (SPS)*

Kwon et al. [20] synthesized Al/CNTs composites using SPS, followed by hot extrusion. The SPS process is shown schematically in Fig. 4. The results indicated that the tensile strength improved by adding carbon nanotubes to Al without reducing the elongation. They proposed that the existence of carbon nanotubes in the boundary layer influences the mechanical properties, which results in effective stress transfer among CNTs and Al matrix due to the aluminum carbide formation and

**Table 1.**
Synthesis methods of aluminium/CNTs composites and their properties

| Method | CNT content | Density (g/cm$^3$) | Mechanical properties* | Ref. |
|---|---|---|---|---|
| Cold press and sintering | 2 vol. % | - | **TS:** 138 MPa | [12] |
| Cold press and sintering | 3 wt. % | 2.6 | **H:** 75.5 HV | [13] |
| Ball milling and hot extrusion | 5 wt. % | - | **TS:** 250 MPa<br>**Indentation modulus:** 74 GPa | [16] |
| Ball milling and hot extrusion | 4 vol. % | - | **TS:** 440 MPa<br>**YS:** 300 MPa | [18] |
| SPS and hot extrusion | 1 vol. % | 2.642 | **TS:** 207.5 MPa<br>**El:** 21.4 % | [20] |
| SPS and hot extrusion | 5 wt. % | - | **TS:** 174 MPa<br>**YS:** 96 MPa<br>**H:** 50 HV | [21] |
| SPS and extrusion | 2.5 wt. % | 2.59 | **CS:** 415.3 MPa<br>**H:** 99.1 HV5 | [23] |
| SPS | 5 wt. % | 2.58 | **TS:** 130 MPa | [24] |
| Flake PM and hot extrusion | 2 vol. % | - | **TS:** 440 MPa<br>**E:** 90 GPa | [26] |
| FSP (4-pass FSP) | 1 wt. % | 2.733 | **TS:** 477 MPa<br>**YS:** 385 MPa<br>**El:** 8% | [27] |
| FSP | 6 vol. % | - | **TS:** 190 MPa<br>**El:** 10 % | [29] |
| SD | 5 wt. % | - | **TS:** 300 MPa | [30] |

*TS: Tensile Strength, YS: Yield Strength, CS: Compressive Strength, El: Elongation, E: Young's modulus, H: Hardness

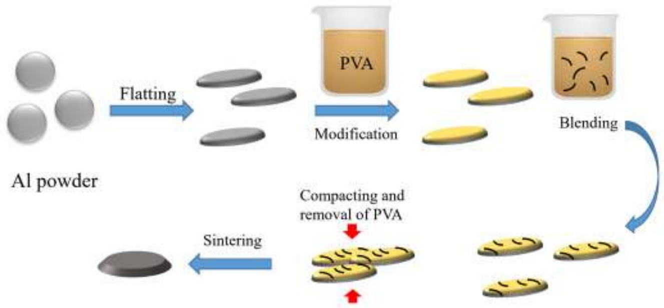


**Fig. 5.** Schematic illustration of flake PM.

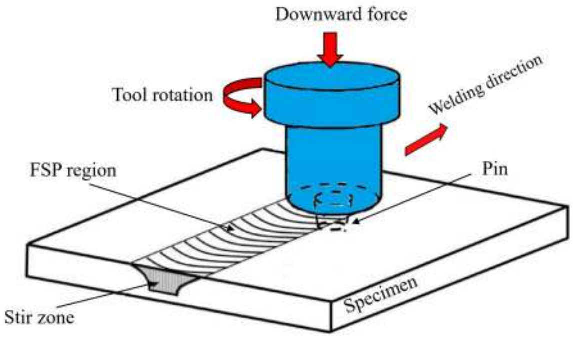


**Fig. 6.** Schematic illustration of FSP.

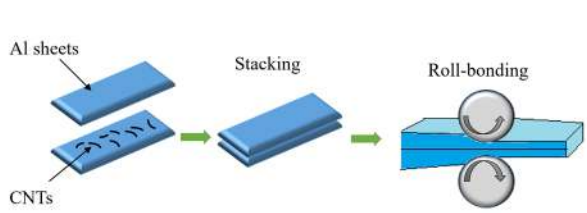


**Fig. 7.** Schematic illustration of the rolling process.

well-aligned carbon nanotubes along the direction of extrusion.

In another research, 0 to 2 wt. % MWCNTs were added to the Al matrix through the mixing of the reinforcement and matrix powders in a roller mill. The mixture was then sintered by SPS, and subsequent hot extrusion and a fully compact carbon nanotube-reinforced aluminum were achieved. The composite mechanical properties with a low amount of CNTs (0.5 wt. %) at room temperature showed a significant increase in comparison with pure Al without increasing the cost. With increasing the amount of CNTs in the composite, the mechanical properties reduced as a result of the agglomeration of carbon nanotubes and the lack of sufficient bonding at the interface [21].

Wu et al. [24] also employed spark plasma sintering for the preparation of MWNTs- reinforced Al composites containing 0 to 5 wt.% carbon nanotubes. According to the results, thermal conductivity increased by the addition of up to 1 wt. % CNTs compared to the pure Al. Thermal conductivity reached a maximum of 199 W/m/K for the composites containing 0.5 wt.% CNTs. This composition of the Al composites also showed the maximum tensile strength of 130 MPa. They summarized that CNT-reinforced Al composites prepared by SPS method are promising materials for applications requiring high thermal conductivity.

CNTs (2.5 wt.%) and Al powders were ball-milled and consolidated through a spark plasma extrusion (SPE) process by Morsi et al. [23]. The advantages of SPE compared to SPS is the possibility of preparing materials with extended geometries and providing bulk deformation influenced by an electric current which provides unique properties in materials. Because CNTs have strengthening effects in Al and decrease the Al crystal size, aluminum/CNT composites had enhanced compressive strength (10%) and hardness (33%).

#### *6.1.8. Flake powder metallurgy (flake PM)*

The steps of flake powder metallurgy route are illustrated in Fig. 5. To disperse carbon nanotubes uniformly in aluminum matrix, the flake powder metallurgy (flake PM) strategy was used by Jiang et al. [26]. To disperse the reinforcement in the matrix uniformly, CNTs were adsorbed onto the surface of aluminum nanoflake through slurry blending and the prepared composite powders were consolidated via hot extrusion. In flake processing, the Al spherical powders changes to nanoflakes, and their surface is modified by polyvinyl alcohol hydrosol; thus, high surface and geometrical compatibilities between aluminum and CNTs powders are achieved. Therefore, the achievement of a homogeneous and less-agglomerated distribution of carbon nanotubes is possible via direct slurry blending. As carbon nanotubes are not exposed to high-energy physical forces such as ball milling, the structural integrity of carbon nanotubes is well maintained in the composite. Therefore, in this study, ductile CNT-reinforced composites with the plasticity of 6% and tensile strength of 435 MPa was produced, which is greater than values obtained from other conventional routes.

Rikhtegar et al. [95] also used the flake PM route for the modification of CNTs dispersion in Al powder. They used short fibers and long fibers of CNTs-COOH with 1.5wt. %CNTs to reinforce and strengthen the Al powders with particle sizes of <20 µm and <45 µm and with a large aspect ratio of 125 and 50, respectively. The influence of different variables of the process, including speed and time of rotation in ball milling, Al nanoflake production, the carboxyl agent, and chemical modification by polyvinyl alcohol on the wall of carbon nanotubes as well as morphological changes of the components were studied.

They reported that in this method, hydrogen bonding is formed between –COOH groups of carbon nanotubes and the –OH groups of PVA, and consequently, a good dispersion of reinforcement in the Al matrix was observed for both short and long carbon nanotubes.

### *6.2. Friction stir processing (FSP)*

Liu et al. [27] applied powder metallurgy and subsequent friction stir processing (FSP) (Fig. 6). Investigation of the microstructure indicated that carbon nanotubes were well dispersed in the Al-based composites. The smaller grain size of aluminum was achieved due to the tendency of carbon nanotubes to be located along grain boundaries. The carbon nanotubes retained their layered structure despite shortening and the formation of $Al_4C_3$ in the matrix. It was shown that during the FSP process, there was no severe damage to nanotubes. The composites containing 1 wt. % and 3 wt. % nanotubes showed an increase in the yield strength about 23.9% and 45.0%, respectively, compared to pure Al.

In other work, Al/MWCNTs composites with various contents of CNTs were synthesized using friction stir processing. The investigation showed good dispersion of CNTs in the Al matrix through friction stir processing. Hardness and tensile tests indicated that the hardness and tensile strength of Al/MWCNTs composites slightly increased with the increase of the nanotube content; however, the reduction in elongation was observed. For the specimen containing 6 vol. % CNTs, ultimate tensile strength reached a maximum of 190.2 MPa, which was two times higher than that of pure Al. The ductility of the nanocomposites decreased with the increase of the carbon nanotube content [29].

For homogeneous incorporation of a high volume fraction (>50%) of CNTs in the Al matrix, Izadi et al. [96] used multi-pass FSP. TEM and SEM investigations showed that CNTs were dispersed uniformly after three passes; however, the tubular shape of CNTs was destroyed by the thermo-mechanical cycles. The microhardness of the samples increased significantly compared to non-reinforced samples due to the influence of the reinforcing component on grain refinement and strengthening of the matrix.

Moreover, TEM analysis indicated that after 3 passes, CNTs were mostly transformed into turbostratic and polyaromatic structures along with $Al_4C_3$. They concluded that two passes preserve the structure of multi-walled CNTs while for uniform distribution of the reinforcement, three passes are required, but the reinforcement stability was not preserved.

*6.3. Spread dispersion (SD)/ rolling process*

X Liao et al. [30] prepared Al/CNTs composite using a spread dispersion (SD) technique. The SD procedure was the repeated pressing and rolling for the preparation of aluminum/carbon nanotubes nanocomposites. This technique was first utilized by Yasuna et al. [8–11] in 1997. The rolling process is depicted schematically in Fig. 7. This technique consists of the stacking of several metal sheets and the subsequent pressing and rolling in order to bond the layers and produce a bulk form from multilayers.

In this study, the researchers applied this method for the dispersion of carbon nanotubes in the Al matrix. It was reported that ultra-fine grain size about 20 nm was formed, the ductility decreased and tensile strength of the nanocomposites increased by 66% over pure aluminum. This improvement was due to stronger Al/CNT bonding, eliminated porosity, the disappearance of the CNT-free zones, and segregation of clustered CNTs.

Samadzadeh et al. [97] synthesized Al/ MWCNTs composites using the roll bonding process. According to the results, using the solution dispersion route in comparison with the spread route leads to the decrease of Al sheets bond strength. Furthermore, the sheets bond strength reduced by the incorporation of carbon nanotubes at a specific reduction of thickness. At higher thickness reductions, the bond strength improved in the sheet with and without carbon nanotubes, and the increase in the entry temperature enhanced bond strength; however, the increase in bond strength was higher in pure Al compared to the reinforced composites. Different synthesis methods of aluminum/CNTs composites and the effect on their mechanical properties are summarized in Table 1.

## 7. Conclusions and future insights

In this review, the processing techniques applied for the synthesis of aluminum/CNTs composites and mechanical properties enhancement, including hardness, toughness, and wear resistance, were discussed. According to previous research reports, a very effective factor in the improvement of the mechanical properties of theses Al-based nanocomposites is CNTs distribution in the metal matrix. Therefore, the preparation conditions and methods are required to be optimized in order to have good dispersion of the reinforcement, enhanced interfacial properties between the two phases, reduction of Al matrix cold working, and subsequently less damage to nanotubes. In addition, other existing challenges are carbon nanotube distribution at micro level in the case of bulk manufacturing, the influence of carbon nanotube alignment, uniform dispersion at high concentration of CNTs. Overall, new methods or modifications of conventional methods are required to develop to optimize these factors.

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