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Influence of the Interface of Carbon Nanotube-Reinforced Aluminum Matrix Composites on the Mechanical Properties – a Review

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Abstract

Carbon nanotubes (CNTs) are a good reinforcement for metal matrix composite materials; they can significantly improve the mechanical, wear-resistant, and heat-resistant properties of the materials. Due to the differences in the atomic structure and surface energy between CNTs and aluminum-based materials, the bonding interface effect that occurs when nanoscale CNTs are added to the aluminum alloy system as a reinforcement becomes more pronounced, and the bonding interface is important for the material mechanical performance. Firstly, a comparative analysis of the interface connection methods of four CNT-reinforced aluminum matrix composites is provided, and the combination mechanisms of various interface connection methods are explained. Secondly, the influence of several factors, including the preparation method and process as well as the state of the material, on the material bonding interface during the composite preparation process is analyzed. Furthermore, it is explained how the state of the bonding interface can be optimized by adopting appropriate technical and technological means. Through the study of the interface of CNT-reinforced aluminum-based composite materials, the influence of the interface on the overall performance of the composite material is determined, which provides directions and ideas for the preparation of future high-performance CNT-reinforced aluminum-based composite materials.

Keywords: Aluminum matrix composites, Carbon nanotubes, Bonding interface, Mechanical properties

1. Introduction

Continuous industrial development is strictly linked with material advancement. High-performance composite materials can retain the characteristics of the matrix material but also exhibit additional properties, providing support for the development of various types of products. Aluminum-based composite materials are among the most widely used metal matrix composite materials as they have a low density, a high strength-to-weight ratio, good electrical and thermal conductivities, a low cost, and a stable performance. They are widely used in the aerospace and automotive industries [1–5]. Modern carbon materials, such as carbon nanotubes (CNTs) and graphene, have a two-dimensional nanometer-size structure, with high strength, a high elastic modulus, and a good thermal conductivity [6–9]. They are widely used as reinforcements in the preparation of various high-performance composite materials to improve material performance. CNTs exploit sp^2 electrons, which endow them with a high modulus and a high strength. They are often added to metal substrates, such as aluminum and magnesium [10, 11]. The prepared composite materials show good performance. Strength, plasticity, and fatigue resistance [12–15] improve the composite material performance and broaden the range of material applications.

The connection region between the matrix and the reinforcement of the composite material is characterized by an interface, and the shape of this interface directly affects the performance of the composite material. After specific processes, the type of the reinforcement in the matrix changes, the distribution becomes more uniform [16–18], the dislocation density increases, and combines with the matrix to form a uniform interface, which improves the bonding of the hybrid interface, bridging the microcrack interface, etc [19-22]. Thus, by adding CNTs, the performance of the metal matrix composite can be effectively enhanced.

However, the current preparation methods of CNT–Al composites are often affected by numerous factors, such as the properties of CNTs and the preparation process parameters, which results in the poor dispersion and wettability of CNTs in the aluminum matrix, thereby weakening the aluminum matrix and the CNTs. The interfacial connection between the CNTs and the Al matrix leads to a degradation in the mechanical properties of the composite.

This review mainly focuses on the interface state that is obtained upon adding CNTs as a reinforcement to the aluminum matrix, and the influence of the interface state on the mechanical properties of the composite material is discussed. In particular, this review analyzes the state of the anisotropic composite interface that is obtained when using different types of preparation methods, discusses the characteristics of these different interfaces, and analyzes the factors that affect the state of the bonding interface. It is envisaged that the discussion proposed in this work regarding the micro interface and macro effects of material bonding will provide an in-depth understanding of the bonding mechanism of CNT-modified aluminum-based composites as well as directions for the preparation of CNT-reinforced aluminum-based composites with excellent mechanical properties.

2. Classification of the bonding interface of CNT-reinforced aluminum matrix composites

According to the literature, bonding interfaces can be divided into four types, as shown in Figure 1, namely free diffusion, mechanical bonding, reaction bonding, and mixed bonding interfaces. Different types of bonding interfaces have different bonding strengths. In general, the bonding strength of the interface follows the order: reaction bonding interface > hybrid bonding interface > free diffusion interface > mechanical bonding interface.

a) Free diffusion interface b) Mechanical bonding interface

c) Reaction bonding interface d) Hybrid bonding interface

Fig. 1. Main types of bonding interfaces

The free diffusion interface is formed through the random diffusion between the reinforcement and the matrix, as shown in Figure 1a. The bonding strength of this type of interface is mainly affected by the intermolecular interaction force. As CNTs and aluminum-based materials have huge differences in their properties, the two can only be combined under certain conditions. Therefore, it is necessary to increase the interference for this type of interface to promote the fluidity and wettability between the matrix and the reinforcement, thereby promoting the combination between the two. The diffusion degree of the internal elements of the composite material is determined by the fluidity of the matrix and the reinforcement. For this type of interface reinforcement and matrix, a chemical reaction does not generally occur at the interface, the reinforcement is randomly distributed at the interface, and a pull-out phenomenon occurs at the interface when failure fracture occurs [23].

The mechanical bonding interface is formed when the reinforcement and the matrix material are bonded under the action of an external force, and almost no chemical reaction occurs between the reinforcement and the matrix, as shown in Figure 1b. The bonding strength of this interface is lower than that of the free diffusion interface. When subjected to a large external force, a poorly bonded interface may debond and fail, so that the force cannot be continuously transmitted to the reinforcement. Due to the forced bonding, several post-preparation treatments, such as the aging treatment and the solution treatment, must be conducted to remove the internal stress due to the bonding between the reinforcement and the matrix material and increase the strength of the composite material. After the CNTs in this interface are subjected to a mechanical force during preparation, the interface arrangement becomes generally more regular and ordered in the direction of the force, and the interface appears smooth and flat [24].

The reaction bonding interface is generally grown in situ on the matrix, and the typical preparation method consists of growing CNTs on the aluminum matrix via an in situ synthesis. An atypical preparation method involves the reaction of the additional products produced during the preparation process. And the performance of ball milling time and heat treatment temperature in the preparation process also influences the reaction-bonded interface. The schematic diagram of the bonding is shown in Figure 1c. Since this type of bonding interface is directly grown on the aluminum substrate, the distribution of the CNTs will be more uniform than that of the other type bonding interface, and the bonding force is higher than that of the free diffusion and mechanical bonding interfaces [25].

The hybrid bonding interface is the most commonly obtained composite material interface when using the existing preparation methods, as shown in Figure 1d. Due to the high extrusion force, high temperature, and other technological means adopted in the preparation process of the composite material, the interface bonding method is complicated and often includes two or three of the above interface bonding types at the same time, but the specific preparation process will focus on a specific interface type.

3. Interfacial bonding mechanism under different preparation methods

Nowadays, the most commonly used preparation methods of CNT-reinforced aluminum-based composites mainly include the casting method [6], powder metallurgy method [26], and in situ synthesis method [27]. There are several differences in the morphology of the interface obtained using these different preparation methods, and the bonding mechanism is also different.

3.1. Free diffusion interface

The free diffusion interface is generally an interface obtained under the application of external factors to promote the effective movement of particles toward the junction between the two materials and their mutual penetration. The free diffusion interface is generally obtained when using the casting method and the spraying method. In CNT-reinforced aluminum matrix composites, the interface obtained through casting is induced by the free diffusion of CNTs in the liquid. Typical preparation methods are the induction melting method, vacuum-assisted investment method, and molten-state stirring casting method. Regarding the induction melting method, CNTs have been precoated with titanium by Muhammad Mansoor et al. [28], which increased the dispersibility of the CNTs and the wettability of the CNTs and the aluminum matrix. The results showed that titanium-plated CNTs are completely and freely attached to the aluminum matrix, and the two are connected naturally, as shown in Figure 2b. In addition, a high content of CNTs (0.4%) results in composites with finer grains than those prepared with a low content of CNTs (0.1%); the CNTs are well dispersed in the ultrafine crystal region of the Al matrix and have a larger interface, as shown in Figures 2c and 2d. Bedri et al. [29] used a vacuum-assisted investment casting process to prepare CNT/6063Al composites; through this method, uniform dispersion of the CNTs into the aluminum alloy matrix was attained at high temperatures. As shown in the fracture scan of Figure 3, the CNTs are tightly connected to the broken part of the composite material, can bridge and pull out, and transfer stress to the composite material, so that the interface strength of the composite material can be increased. The mechanical experiment results show that the compressive strength and hardness of the material have been greatly improved.

The interface obtained through the spraying method is induced by the random dispersion of the CNTs in the solid. The typical preparation methods include thermal spraying and plasma spraying [23]. However, in this case, the connection between the CNTs and the aluminum matrix only relies on the weak van der Waals force between them; thus, debonding and curling can occur near the CNTs. Furthermore, stress is concentrated in this area, which results in delamination, and the CNTs are prone to peel off from the aluminum particles, so the obtained interface bonding strength is very weak. At the same time, during the spraying process, a gasification reaction occurs at the defect locations on the CNTs, generating carbon monoxide gas (which in turn generates bubbles and reduces the strength of the interface bonding), and the thermal activation reaction weakens its strengthening ability. These reactions not only hinder the formation of the interface but also reduce the strength of the interface bond [30].

Researchers added different CNT contents, used different aspect ratios in the molten state of the ZL105 aluminum alloy, and prepared CNT–Al materials using the mechanical stirring casting method to obtain composite materials with different mechanical properties. The preparation process is shown in Figure 4a. After the heat treatment, the strength of the different materials was tested, and it was found that the CNT–Al composite material with a CNT content of 1.25% and a small aspect ratio had the best strength, which was 10.53% higher than that of the ZL105 aluminum alloy. This is mainly because the addition of an adequate CNT amount can reduce the occurrence of agglomeration and promote the improvement of the material properties. At the same time, it was found from the microstructure (Figure 4b) that the different material structures are excessively smooth and flat, and the bonding state of the interface is good. This may be because CNTs with a small aspect ratio have a larger interface bonding area with the aluminum matrix, which improves the interface bonding. Moreover, this method is convenient, suitable for actual production, and can be easily adopted to realize mass production.

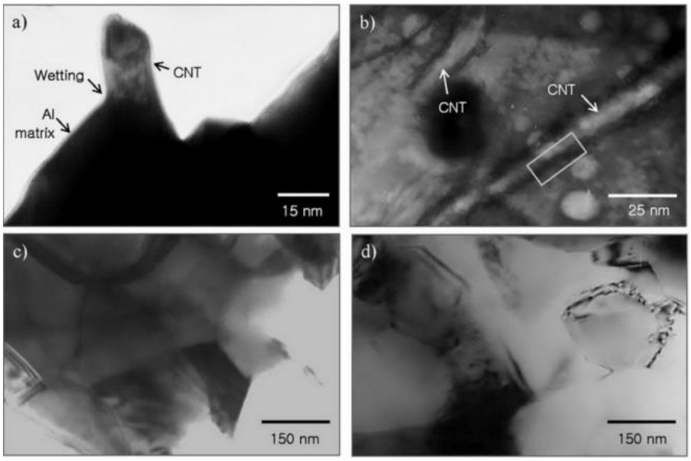


Fig. 2. TEM micrographs of the composite specimens: (a) a single nanotube protruding out of the etched matrix, (b) CNTs dispersed in the matrix, and (c) and (d) micrographs showing the grain sizes of the Al–0.1MWCNTs and Al–0.4MWCNTs specimens, respectively [28]

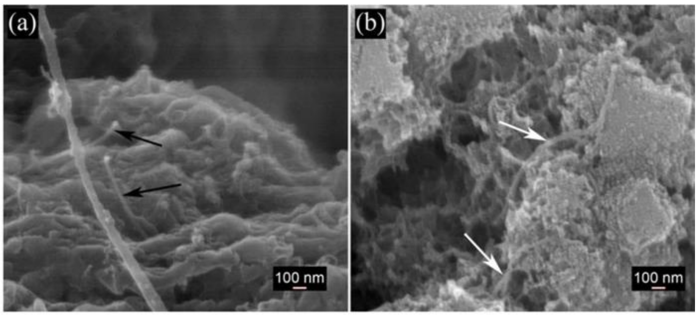


Fig. 3. SEM images of the fractured surfaces of the composites: (a) the black arrows show the pulling-out of the CNTs between fractured surfaces; (b) the white arrows show the bridging of the CNTs between different sides of the matrix material [29]

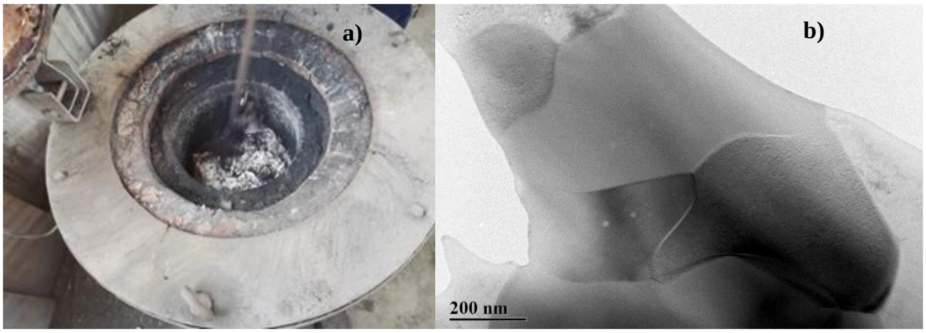


Fig. 4. (a) Aluminum alloy in the molten state and (b) TEM microscopic morphology of the composite sample

3.2. Mechanical bonding interface

The mechanical bonding interface is mainly formed by forcibly combining the reinforcement and the matrix during the preparation of the composite material. The bonding strength of this interface is relatively weak; therefore, it is necessary to perform a heat treatment or other related processes to disperse and embed the reinforcement at the interface into the matrix as much as possible to strengthen the bonding between the two. The most representative preparation methods for this type of interface are the sandwich technology method, friction stir method, and hot rolling composite method. Cesar A Isaza M and others used the sandwich technology to roll the composite material, as shown in Figure 5a. The results show that the CNTs and the aluminum matrix have a distinct layer, and the combination interface is flat and regular. No agglomeration or damage occurs, but there are several pores between the interlayers, as shown in Figures 5b and 5c [31]. However, Liu et al. [24] used the friction stir method in combination with the hot rolling process to mix and stir the CNTs and the aluminum powder into a billet and then performed friction stir and hot rolling, as shown in Figure 6. Their results indicate that the CNTs are arranged in the aluminum matrix in an orderly manner along the hot rolling direction and do not completely react with the aluminum matrix. However, the CNTs react at the ends to form Al_4C_3, as shown in Figure 7a. A small amount of Al_4C_3 and CNTs are arranged in an orderly manner in the aluminum matrix to form a relatively flat, smooth, and tightly bonded interface, which improves the tensile strength and increases the elongation of the composite material.

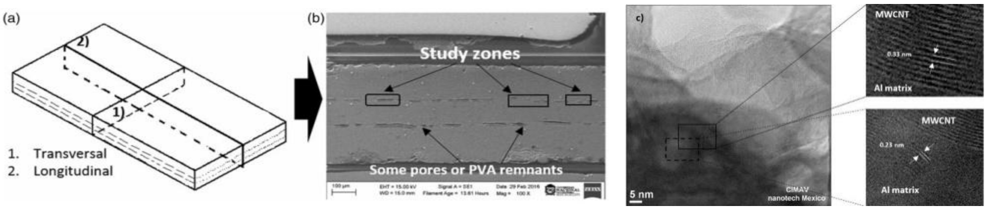


Fig. 5. (a) Outline of the composite sections studied, (b) study zone close to the interface (SEM-SE image), and (c) HRTEM images of the composite interface showing details of the MWCNTs and the Al matrix [31]

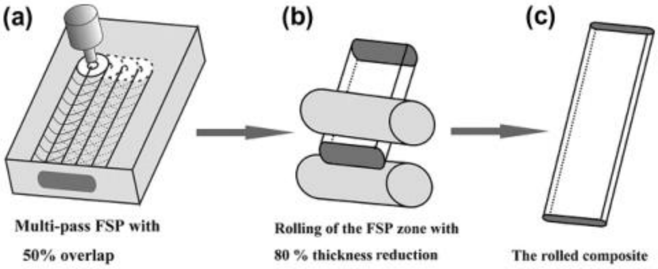


Fig. 6. Schematic of the CNT/2009Al composite fabrication flow [24]

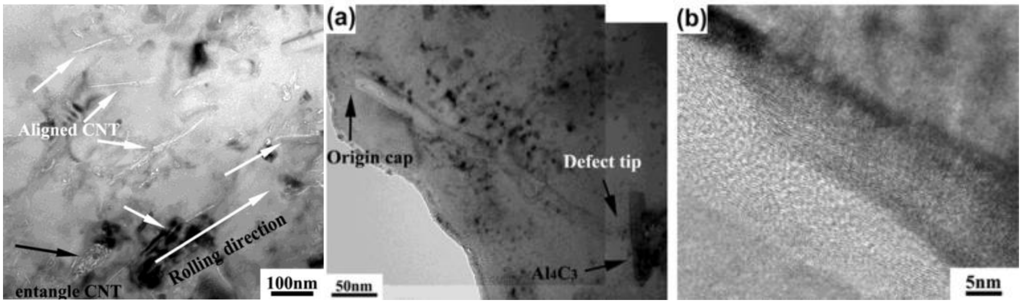
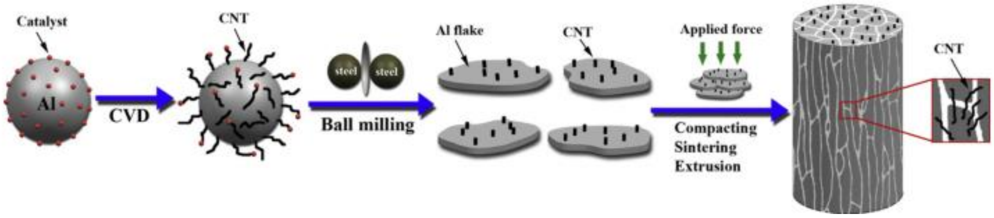


Fig. 7. (a) TEM image showing a CNT–Al interface exhibiting good bonding and (b) HRTEM image showing the structure of a CNT [24]

3.3. Reaction bonding interface

The reaction-bonded interface is the interface obtained through the chemical reaction between the internal compounds of the composite material under certain conditions. The chemical interaction between the metal matrix and the CNTs gives rise to a strengthening effect. However, it is difficult to control the interface formed through the reaction combination. The in situ synthesis method is typically used to obtain a reaction-bonded interface. The bonding interface is generated at the same time that a chemical reaction occurs on the matrix to grow the reinforcement, which can ensure a good bonding performance between the reinforcement and the matrix. Reaction-bonded interfaces can also be obtained using atypical methods, but these interface reactions are often accompanied by the production of Al_4C_3. Several studies have found that a small amount of Al_4C_3 can enhance the interface connection [32]. However, if the Al_4C_3 content is too high since the Al_4C_3 performance is worse than that of the CNTs, the interface between the CNTs and the Al matrix becomes a non-coherent interface between carbide and the matrix [33], which will result in side effects. Yang et al. [25] used the in situ synthesis method shown in Figure 8 to uniformly deposit a Co catalyst on the surface of Al powder. Then, CNTs were grown on the Al powder via chemical vapor deposition and subsequently dried. After ball milling for a short time, compaction, sintering, and hot extrusion of the CNT–Al composite powder, the CNT–Al composite material was obtained. In the preparation flow chart, the CNTs grew in situ and the aluminum matrix is tightly and firmly bonded. As shown in Figure 9a, the surface of the CNTs synthesized in situ is smooth and clean; there is no amorphous carbon on the surface of the aluminum matrix and the CNTs are not agglomerated. In addition, the CNTs are tightly surrounded by the aluminum matrix, and the two are fused to form the largest tight interface bonding area; this results in an ideal carbon layer spacing of 0.34 nm, as shown in Figure 9b. Therefore, the strength and elongation of the composite material are greatly improved, which is due to the in situ growth process on the aluminum matrix. Compared with the interfaces obtained through many preparation methods, the reaction bonding interface obtained using the in situ synthesis method is the strongest [25], and the dynamics enhancement effect is the best.



a) b) c) d)

Fig. 8. Schematic illustration of the procedure used to fabricate the CNT–Al composites. (a) Preparation of the Co catalyst spread homogeneously on the surface of the Al powder, (b) in situ synthesis of the CNTs on the Al powder via CVD, (c) ball milling of the in situ synthesized CNT–Al powders, and (d) fabrication of the CNT–Al composites through compacting, sintering, and hot extrusion [25]

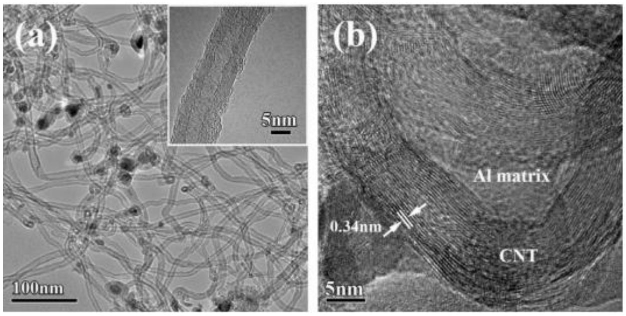


Fig. 9. TEM images of (a) the CNTs obtained via in situ synthesis and (b) the 2.5 wt.%-CNT–Al composites obtained after 90 min of milling and hot extrusion. The inset in (a) shows the magnified image of a CNT [25]

3.4. Hybrid bonding interface

A hybrid interface is an interface that combines the aforementioned interface types. There exist many preparation methods to obtain a hybrid interface, mainly including the spark plasma sintering (SPS) method and powder metallurgy in combination with the friction stir method. The interface formation is affected by different preparation methods and process parameters. During SPS, a reaction bonding interface is easily obtained under a high sintering temperature and a high pressure, and a mechanical bonding interface is also formed in the composite powder block during hot extrusion. In the actual preparation process, for most of the preparation methods of CNT-reinforced aluminum matrix composite materials, the obtained interfaces are almost all hybrid and bonded interfaces. Only with different interface types can there be the abovementioned three interface bonding types. Zhou et al. [34] reported a procedure consisting of ultrasonic vibration and dispersion drying of the CNTs and aluminum powder, which had been acidified in advance in an ethanol solution, followed by hot extrusion into massive objects. Using the SPS technology, Al_4C_3, which is beneficial for interface bonding, was grown at the interface between the CNTs and the aluminum matrix. It was found that the low refractive indexes of MWCNT (002), Al (111), Al (220), and Al (002) give rise to a relatively stable coherent interface on the aluminum matrix. Therefore, the interface between the CNTs and the aluminum matrix was close and clean, as shown in Figure 10. At the same time, the measurement showed that Al_4C_3 has a larger diameter than CNTs (Figure 11c), and the coarse grains of Al_4C_3 will affect the interface bonding effect, resulting in a decrease in the interface force of the reaction bonding.

The composite material interface obtained using powder metallurgy in combination with the friction stir method also exhibits many forms. The mixed ball milling, briquetting, and friction stir processes can be easily used to obtain mechanical bonding interfaces. In the later high-temperature treatment, reaction bonding interfaces may appear. At the same time, an interface will form due to free diffusion, so there is no specific bonding interface in the composite preparation method. Liu et al. [35] reported a CNT–Al composite interface obtained using powder metallurgy in combination with the friction stir method; the interface exhibited a zigzag shape, and there is only the connection interface between CNTs and aluminum. The interface was clean, and no reaction occurred. In the powder metallurgy process, the CNTs are embedded in the aluminum matrix through the hot pressing and hot forging processes, which give rise to a mechanical bonding interface, as shown in Figure 12a. However, metal carbide Al_4C_3 was found elsewhere in the sample (Figure 12b, c), and Al_4C_3

was attached to the CNTs. This is because parts of the CNTs are damaged by friction stirring, which results in defect sites and gives rise to a weak interface reaction between the CNTs and the aluminum matrix. Overall, the use of powder metallurgy in combination with the friction stir method results in two interfaces, namely a mechanical bonding interface and a reaction bonding interface. The strength of this material is maximized under the combined action of the two interfaces.

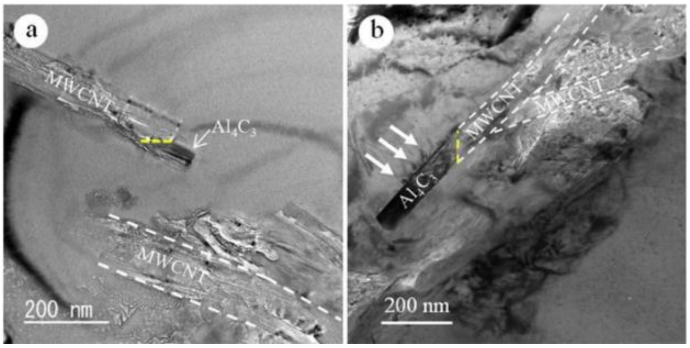


Fig. 10. HRTEM images of the 5 vol.% MWCNT–Al composite after heat treatment (a) at 873 K for 0.1 h and (b) at 883 K for 1 h; the white arrows in (b) indicate the stress contrast [34]

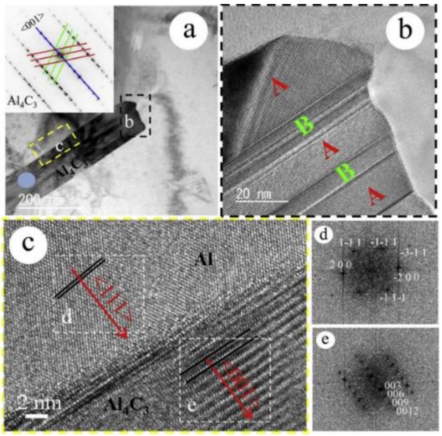


Fig. 11. Morphology of the 3.0 vol.% MWCNT–Al composite after heat treatment at 913 K for 2 h: (a) HRTEM image of a twined Al_4C_3 with its SAED pattern (inset) taken at the blue spot; (b) HRTEM image of the magnified twinning in (a); (c) HRTEM image showing the Al_4C_3–Al interface acquired from the marked area in (a); (d) and (e) FFT patterns of Al and Al_4C_3 at the clean interface in (c), respectively [34]

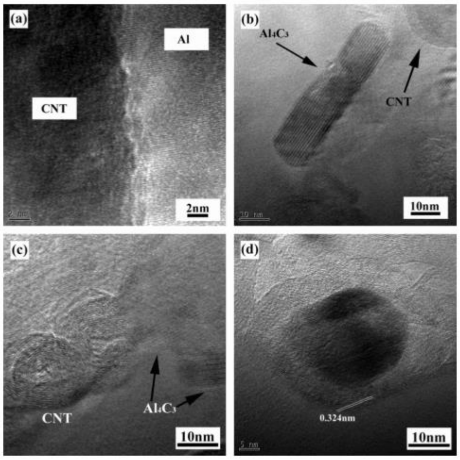


Fig. 12. HRTEM images showing (a) the interface between Al and a CNT, (b) Al_4C_3 in the Al matrix near a CNT, (c) Al_4C_3 attached to a CNT, and (d) a layered wall structure of the CNTs in a four-pass FSP 3 wt.% CNT/2009 Al composite [35]

The formation of a free diffusion interface is mainly affected by the mutual diffusion of the liquid metal solution or solid powder. The use of appropriate ball milling or stirring vibration methods can improve the bonding effect. The formation of a mechanical bonding interface is mainly affected by the magnitude of the mechanical force and the subsequent heat treatment process, and using an adequate mechanical force is crucial. The reaction bonding interface is mainly affected by the reaction degree; the most important aspect is to use an appropriate reaction time to obtain the most ideal bonding interface. However, the reaction degree is not convenient for the qualitative expression; thus, the reaction degree can be characterized using the electrochemical dissolution–gas chromatography calibration method combined with the calculation expression [36]. The formation of the hybrid bonding interface is mainly affected by the preparation process and its parameters. In order to obtain the optimal bonding interface, several experiments have been carried out to summarize the best combination parameters.

4. Factors influencing the interfacial bonding of CNT–Al composites

The interface state of the metal matrix composite material is greatly affected by the preparation process and parameters, and the interfacial bonding effect of CNT-reinforced aluminum matrix composites will be affected by key factors, such as the preparation method, process parameters, and material component state.

4.1. Influence of the preparation methods and processes on the bonding interface

Different preparation methods will result in significantly different types of interface bonding, and different types of interface bonding will exhibit different interface bonding forces, which will directly lead to considerable differences in the material properties. For example, the aforementioned vacuum-assisted investment method can be used to obtain a free diffusion interface, the friction stir method in combination with the hot rolling process can be used to obtain a mechanical bonding interface, the in situ synthesis method can be used to obtain a reaction bonding interface, and the powder metallurgy method can be used to obtain a mixed bonding interface.

Different bonding interface states are also obtained using different preparation process parameters. Zhang [37] prepared SiCw + CNTs/6061Al hybrid composites using the squeeze casting method. Treating the material in three different process states: as-cast state, hot extrusion and post-hot extrusion heat treatment. The interface bonding state obtained after hot extrusion is the best. This is because the heat of the reaction and other processes can inhibit the formation of inadequate interfaces [38]. In powder metallurgy [39], the degree of the interface reaction between CNTs and aluminum can be directly affected by controlling the sintering temperature. The interface reaction becomes strong when the annealing temperature is higher than the melting point of aluminum. However, when the annealing temperature is lower than the melting point of aluminum, a small amount of Al_4C_3, which is beneficial for interface bonding, is generated at the location of the amorphous carbon coating, at defect sites, and at the open end of the CNTs [40]. Therefore, the interface phase can be controlled by precisely tuning the process temperature. Jiang et al. [41] modified the traditional powder metallurgy process and used sheet metallurgy to obtain nanocomposite materials with a double size particle, and the CNTs transitioned from an approximately one-dimensional state to a two-dimensional array. As a result, the contact area between the CNTs and the aluminum matrix increased, and the bonding stability increased so that the tensile strength and ductility of the composite material also increased. The use of mechanical impact methods, such as ball milling, can have a shearing effect on CNTs with relatively large length and diameter. This is a method commonly used to obtain short-fiber CNTs. This method can effectively promote the shortening of CNTs and refine the grains. Furthermore, it can enhance the dispersibility of the CNTs and promote their interfacial reaction [42–45]. However, a longer ball milling time does not necessarily lead to a better composite; a too long ball milling time will cause damage to the CNTs [46], and interface reactions are prone to consume the added CNTs at the damage site, resulting in carbides that reduce the quality of interface bonding [47]. In the actual process, smelting aids, process control agents, and nSiC particles can be added as dispersants during ball milling, which can not only avoid damage to the CNTs but also eliminate the oxide film of the aluminum particles, reduce the occurrence of cold welding [48–51], reduce the shear hysteresis effect of the ball milling process, and promote interface bonding.

4.2. Influence of the material composition

The bonding interface is the connection between two materials; thus, the properties of the CNT material itself, such as composition and wettability, have a direct impact on the bonding interface.

4.2.1. Wettability

The thermal expansion coefficient, surface tension, modulus, density, and other properties of CNTs are quite different from those of aluminum [52–55], which leads to poor wettability between the two materials. Common ways to improve the wettability between CNTs and aluminum include reducing the contact angle between the two, generating functional bonds (covalent bonds, hydrogen bonds [56], etc.), borrowing transition layers, using mechanical rolling processes, and alloying elements with a high affinity.

Coating carbon materials with nickel or copper will reduce the contact angle between aluminum-based materials and carbon materials and lead to the formation of Al–C covalent bonds [57, 58]. The coating metal tends to dissolve in the matrix and form a stable compound with the matrix [59]. Studies have shown that metal-plated CNT composite materials can exhibit a compact and smooth interface without a noticeable porosity [26]. At the same time, to ensure that the coated metal can be uniformly deposited on the surface of the CNTs, the coating time needs to be controlled. [60]. The carbon-silicon reaction and high-temperature annealing can also be used to introduce a SiC nano transition layer to reduce the contact angle of aluminum and thus increase the wettability between Al and C [61]. Promoting the mismatch between two different materials and realizing interfacial dislocations to form prismatic punching [29] represents another strategy to increase the wettability between Al and C, but CNTs need to be pretreated before metal plating [62]. Using the rolling process can force the fusion of CNTs and aluminum [63], but this method is not as effective as the above wetting method. Adding Li, Mg, Ca, and other alloying elements with a high oxygen affinity to aluminum-based composites can also improve the wettability between metals and reinforcements [64]. In addition, owing to the advancement of simulation techniques, three microscale simulation methods, namely molecular dynamics, first-principles calculations, and Ab-initio molecular dynamics, can be used to characterize the wettability of liquid-solid crystal surfaces [65].

4.2.2. Dispersion

The dispersion degree of CNTs has an important influence on the interface bonding state of composite materials. Due to the high surface energy of CNTs, there is a large van der Waals force between each pair of CNTs [66]. A low dispersion degree can lead to segregation, resulting in an insufficient and uneven interface bonding, which also hinders the strengthening of CNTs. This is also one of the reasons that hinder the development of CNT-reinforced aluminum matrix composites. At present, the most common dispersion method is ball mill vibration. This method can be used to reduce the aspect ratio of carbon fibers [67] and increase the uniform dispersion degree of carbon fibers in the aluminum matrix in the liquid state, thereby effectively avoiding the agglomeration of CNTs at the interface. However, the use of ball milling to promote dispersion has limitations. If the flake aluminum powder is too thick, it cannot be broken by ball milling. At this time, the aluminum powder will fix the CNTs and hinder their dispersion [68]. In order to increase the dispersibility of CNTs in the aluminum matrix, different strategies can be adopted, including the in situ synthesis of SiCp on CNTs, CNT hybridization, and preparation of highly dispersible amino-functionalized CNTs (fCNTs). [69]. In addition, researchers have found that a high temperature and a high pressure can promote the dispersion of atomic activities, but these conditions will cause irreversible damage to CNTs [70], and these consequences need to be considered in practical applications.

4.2.3. Form and content

The morphology of CNTs and the aluminum matrix will have a direct impact on the interface bonding. In particular, the shape, aspect ratio, integrity, CNT content, and the size of the aluminum matrix particles will affect the bonding state of the interface. Compared with the traditional tubular structure of CNTs, chevron-shaped CNTs have more defects and will react weakly with the aluminum matrix to form a thin intermediate layer, preventing the formation of the interface [71]. CNTs with a small aspect ratio have several advantages. When combined with a metal matrix, they exhibit a larger effective contact area and can form a bonding interface more easily [72–74]. At the same time, CNTs with small diameters have a stronger capillary action, which can attract surrounding metal atoms to produce noncovalent bonds, forming a smooth and bubble-free bonding interface [75]. In addition, the Al_4C_3 formed due to the damaged CNTs at the damaged sites can also enhance the interface connection [76], but it is not as effective as the CNTs themselves. Adding an appropriate amount of CNTs to CNT–Al materials can result in finer grains [77] as well as numerous nucleation sites to promote bonding between CNTs and aluminum [46]. However, adding too much CNTs will cause agglomeration, resulting in an insufficient interface bonding of the composite material and degraded mechanical properties [78]. The size of the aluminum powder particles will also affect the formation of the interface. The aluminum powder undergoes deformation, fracture, and cold welding during the ball milling process [79], which causes the aluminum particles to become finer; however, when the grain diameter is less than 70 nm, dislocation annihilation will occur, leading to the interface becoming worse [80]. At the same time, the silicon content in the aluminum alloy will also affect the production of carbides. As the silicon content increases, the quantity of carbides produced will decrease, and carbides have a direct effect on the interface bonding [81].

4.3. Other influencing factors

In addition to the abovementioned influencing factors, other factors, such as the chemical purity of the CNTs and the coating material as well as the functional bonds will also have an impact on the interface bonding state. Huang et al. [82] found that the interface between purified CNTs and the aluminum matrix is free of impurities and is relatively weakly affected by external factors. CNTs were also coated with TiC nanoparticles [83]. The enhancement mechanism consists of the local inhibition of the reaction of the MWCNTs with the matrix alloy, thereby preventing the in situ formation of carbides during the solid-phase consolidation process. The acid treatment of multiwalled CNTs will result in the formation of some defects in the CNTs and introduce more oxygen-containing functional groups [84, 85]. The oxygen in the surface functional groups can enhance the interaction between carbon and metal and provide a strong bond between the CNTs in the composite material and the metal matrix, so that a high-strength interface bonding can be realized [86]. Furthermore, the degree of the interface bonding can be controlled by adjusting the time of the acid treatment. To promote the interface bonding, polyethylene glycol was used as the polyester binder, and the aluminum powder was coated with CNTs during the auxiliary ball milling. The structure and morphology of the CNTs were retained as unchanged as possible, and the damage degree was reduced [87]. Paraffin wax was used to functionally modify the CNT–COOH bond and the pure aluminum powder. The modified CNTs have an increased dispersion and are well integrated with the matrix interface; furthermore, the mechanical properties were found to be significantly improved [88].

In summary, many factors affect the formation of the interface of CNT-reinforced aluminum matrix composites. Among all the existing preparation methods, the in situ synthesis of CNTs on the aluminum matrix results in the best interface [25]. However, this preparation method also has numerous disadvantages, including the difficulty of controlling the reaction time and the fact that it is a complex process. The interface can be optimized by taking appropriate measures. For example, the process parameters should be strictly controlled to avoid CNT damage and introduce functional bonds, the CNTs should be coated with metals to increase their wettability, and the milling time should be controlled to promote the dispersion of the CNTs and ensure the integrity of the structure. Furthermore, appropriate content of CNTs should be used, and their size and structure or the size of the aluminum particles should be adjusted to increase the interface bonding area. Additionally, the CNTs should be pretreated and purified, and modifiers and binders should be used to promote the interface bonding between CNTs and the aluminum matrix. There is more than one way to optimize the interface integration. Therefore, one or several of these methods can be used to obtain a good bonding interface, thereby improving the performance of the composite material. In the future, more advanced preparation technologies may be implemented to overcome the current problems regarding the interface bonding of CNT–Al composites.

4. Conclusion

This review focuses on the interface morphology of CNT-reinforced aluminum matrix composites, explores the influence of different preparation methods and process parameters on the formation of the interface, and inspires the nanomodification of aluminum alloys to understand the relationship between CNTs and aluminum. The following conclusions can be drawn:

1. The interface formed between the two materials can be mainly classified into four types: free diffusion, mechanical bonding, reaction bonding, and mixed bonding interfaces. Different interface types exhibit different bonding strengths, which in turn leads to differences in the mechanical properties of the composite materials. The free diffusion interface is obtained due to molecular dynamics when the materials are mixed, and it can be promoted through the application of external influences, such as stirring vibration. The mechanical bonding interface has a weak bonding strength. To enhance its bonding strength, a heat treatment process must be carried out to allow the reinforcement to diffuse and embed in the matrix to reduce the agglomeration of the reinforcement. The most common interface type is the reaction bonding interface, and the mixed bonding interface is often composed of the reaction bonding interface and other bonding interfaces.
2. Different preparation methods will result in interfaces with different bonding strengths, and different process parameters in the same preparation method will also have a great impact on the formation of the interface. The shape and content of the reinforcement will affect its dispersion in the matrix material, and the wettability of the reinforcement will affect the binding force of the remaining matrix materials. Ball milling, plating, and other effective reinforcement pretreatments can be used to obtain an interface with a higher bonding strength, thereby improving the mechanical properties of the material. Due to the different properties between the matrix and the reinforcement, when preparing CNT–Al composites, the dispersion and wettability of the reinforcement should be increased as much as possible while using the most economical preparation process.

Although there has been considerable progress in the field of CNT-reinforced aluminum matrix composites, due to the issues regarding CNT dispersion and a weak bonding interface, CNT–Al composites are still in the stage of experimental exploration and practical production applications. There is still a long way to go, and CNTs have not reached their full potential. Therefore, in the future, it is envisaged that the preparation of CNT–Al composites will be carried out through new technical means to continuously overcome the current unfavorable interfacial bonding factors, such as the poor wettability and dispersibility, and truly exploit the reinforcing effect of CNTs in the aluminum matrix. Based on the existing products, the CNT reinforcing effect can substantially improve the overall performance of the products.

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