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# Progress on CNT/Al Composites

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**Abstract:** Carbon nanotube reinforced aluminum matrix composites have the advantages of light weight and high specific strength, and have broad application prospects in aerospace, automotive lightweight and other fields. In this paper, the preparation process of CNT/Al composites, the surface modification of CNT and the alloying of aluminum matrix are systematically reviewed. The mechanism and research progress of each method are analyzed. The key properties of CNT/Al composites, such as mechanics, thermal expansion, electrical conductivity and wear resistance, are summarized. The development trend and key research directions in this field are prospected.

**Keywords:** Carbon Nanotube; Aluminum Matrix Composites; Preparation Technology; Mechanical Properties

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## 1. Introduction

As a commonly used lightweight metal material, aluminum alloy is widely used in various fields due to its low density, excellent processing performance and controllable cost. However, the strength, hardness and wear resistance of a single aluminum alloy are difficult to meet the needs of high-end scenarios. Researchers have long been committed to improving the properties of aluminum alloys by adjusting the thermal deformation parameters and optimizing the heat treatment process. Although the mechanical and physical and chemical properties of aluminum alloys can be improved to a certain extent, due to the structural characteristics of the material itself, it has failed to achieve a leap-forward breakthrough in performance. In this context, aluminum matrix composites have become an important research direction to solve this bottleneck problem due to their advantages of strong designability and excellent comprehensive performance[1].

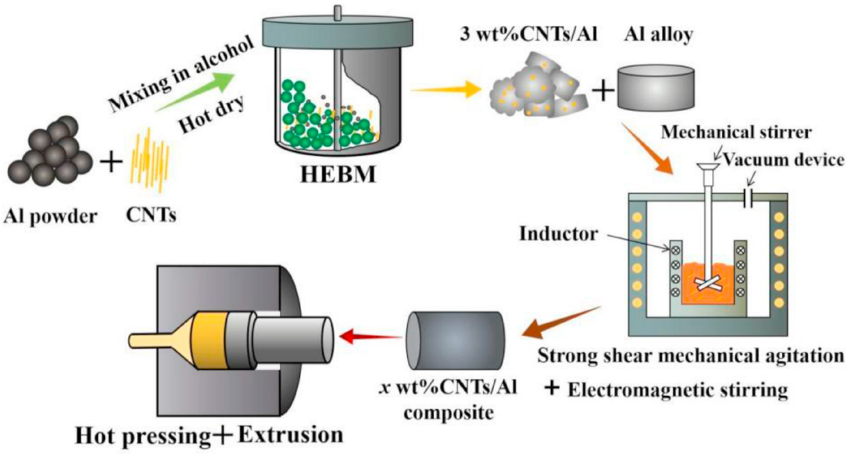
As a new type of nano-reinforcement, carbon nanotube (CNT) has extremely high elastic modulus, tensile strength, excellent thermal conductivity and electrical conductivity. The CNT/Al composites prepared by using CNT as reinforcement can effectively combine the lightweight characteristics of aluminum matrix with the excellent reinforcement effect of CNT, and show broad application potential in high-end fields such as aerospace parts and automobile lightweight structural parts. However, due to the strong surface inertia of CNT and the large difference in physical and chemical properties between CNT and aluminum matrix, there is a problem of weak interface bonding between them, which seriously restricts the full play of CNT's excellent performance. As a connecting bridge between the matrix and the reinforcement in the composite, the structural characteristics and formation mechanism of the interface directly determine the comprehensive properties of the composite, such as mechanics, thermal conductivity and electrical conductivity. Therefore, interface regulation has become the core key to the performance optimization of CNT/Al composites. At present, the research on the interface control of CNT/Al composites mainly focuses on three directions: optimization of composite preparation process, CNT surface modification and aluminum matrix alloying. This paper will systematically review the above research directions and the properties of composites.

## 2. Preparation Process of CNT/Al Composites

### 2.1 Liquid Phase Method

The liquid phase method is to directly add the reinforcement particles or first make the composite preform into the metal melt, and then stir and finally cool to obtain the composite material. However, the interfacial reaction during the preparation process is not easy to control, especially the reaction between the reinforcing particles and the aluminum matrix. In the liquid phase method, the liquid metal can completely encapsulate the carbon nanotubes, and there is enough surface contact to form a strong interfacial bonding force. However, at higher temperatures, serious interfacial reactions may occur, and the formation of brittle interfacial phases affects the properties of the composites. Zhang et al.[2] used high-energy ball milling (HEBM) -semi-solid stirring casting (SSC) process to prepare CNT reinforced Al-Cu-Mg composites, and explored the effect of CNT on the microstructure and properties after hot extrusion, as shown in Fig.1. With the multiple strengthening effect induced by CNT, when the CNT content increased from 0 to 1.5 wt. %, the tensile strength and yield strength of the composites increased from 319 MPa and 261 MPa to 523 MPa and 476 MPa, respectively.

*Figure 1: The schematic diagram of the preparation process of CNT/Al copper magnesium composite material[2].*



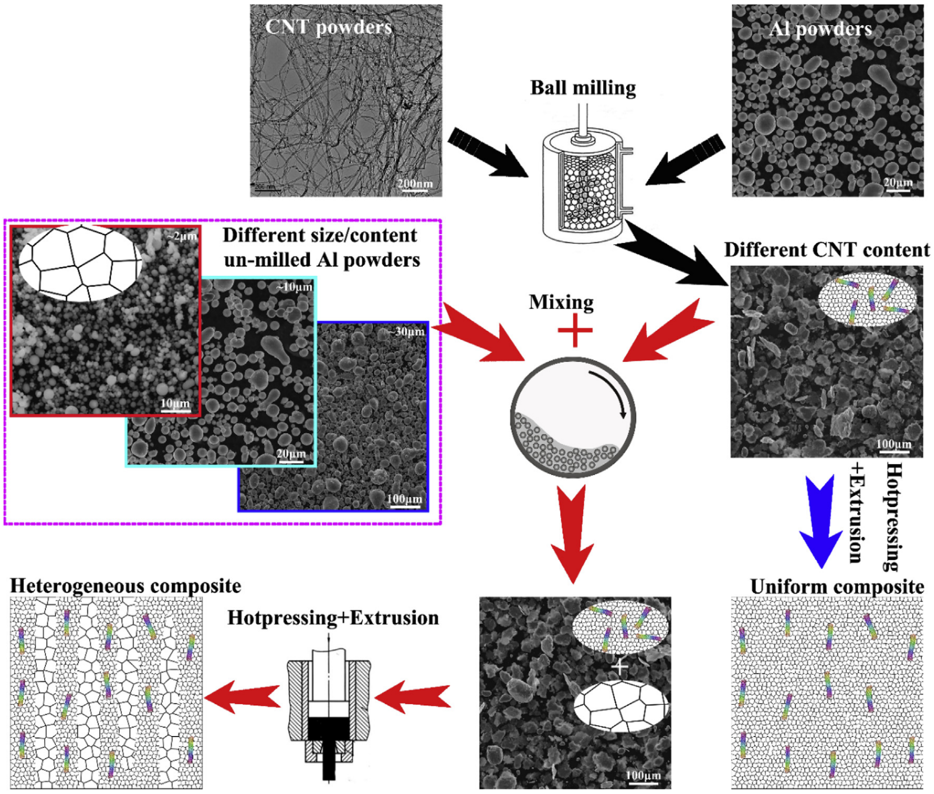
### 2.2 Solid-state Method

The solid phase method mainly includes powder metallurgy, hot extrusion, cold pressing sintering and spark plasma sintering (SPS). In order to improve the density, carbon nanotube dispersion and mechanical properties of materials, it is often supplemented by single or composite secondary processing processes such as extrusion, rolling, friction stir welding and forging.

The powder metallurgy method achieves uniform dispersion of the reinforcing phase by mixing the matrix powder with the reinforcing phase particles. After mixing, the green body is obtained by cold pressing, and the density can reach 80% of the theoretical density. Subsequently, the green body was sealed and degassed to remove the water adsorbed on the surface of the powder ; finally, a high-density composite material was prepared by isostatic pressing or uniaxial sintering[3]. Choi et al.[4] found that when preparing aluminum matrix composites by powder metallurgy method, with the increase of ball milling strength, carbon nanotubes and aluminum matrix formed a good interface bonding through mechanical occlusion. Singhal et al.[5] prepared aminated carbon nanotubes reinforced aluminum matrix composites by powder metallurgy method. The results show that the dispersion of aminated carbon nanotubes in aluminum matrix is significantly better than that of unmodified carbon nanotubes, which can be uniformly dispersed in aluminum matrix without obvious agglomeration.

The hot extrusion method refers to the processing technology of extruding the material powder from the mold hole by high pressure under the heating state. Wu et al.[6] used semi-solid powder metallurgy process to achieve partial liquid phase sintering by raising the temperature near the melting point of aluminum powder during hot pressing. It was found that the increase of liquid phase composition could significantly improve the density and mechanical properties of the material. The hardness of CNT/6061Al alloy composite reached 87.5 HV after sintering at 620 °C, which was nearly twice as high as that of the matrix alloy. Esawi et al.[7] mixed 5wt. % CNT with aluminum powder by ball milling, cold pressing and hot extrusion sintering. The results showed that the stiffness of the composite was 23% higher than that of pure aluminum, and the tensile strength could be increased by 50%. When the CNT content exceeded 2 wt. %, the dispersion difficulty increased, and the mechanical properties did not meet the expectations. Liu et al.[8] used a bimodal structure design (Fig. 2), and added unmilled aluminum powder to the CNT/Al composite powder, so that the elongation of the composite was 100% higher than that of the uniform CNT/Al composite, and the tensile strength remained basically unchanged. Bakshi et al.[9] found that in the densification process of composite powder, when the extrusion ratio is greater than 20 and the pressing force reaches the order of GPa, the composite material has better performance ; the greater the extrusion ratio and pressing force, the higher the deformation degree of the material, and the hot extrusion rate and temperature need to be reasonably controlled, otherwise it is easy to cause processing instability and surface cracking.

*Figure 2: A schematic diagram of the preparation process of CNT/2009Al composites with heterogeneous and uniform structure*[8].

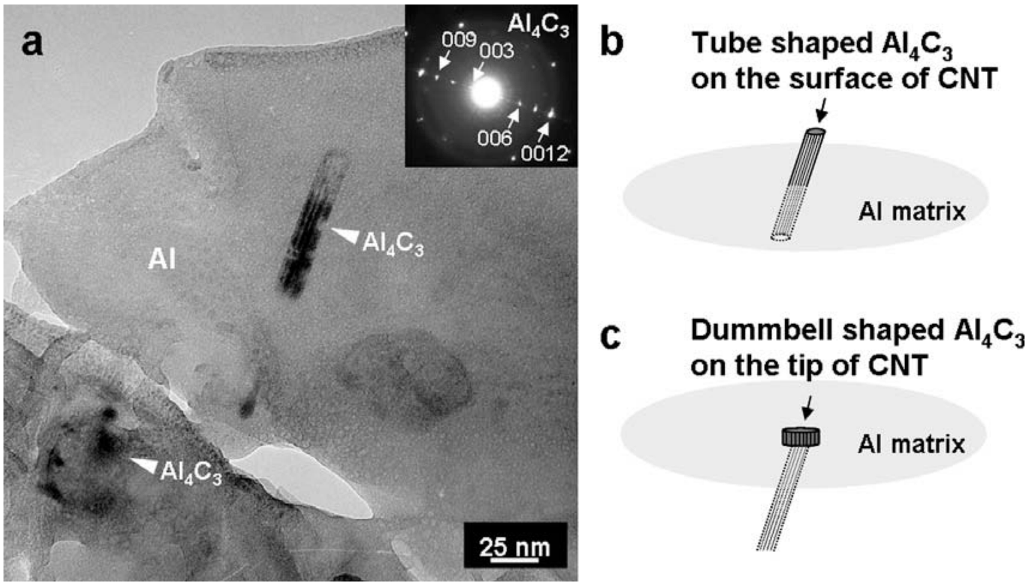


George et al.[10] prepared CNT/Al powder mixture by ball milling, and sintered in nitrogen atmosphere after 120 kN pressure cold pressing. The results showed that the mechanical properties such as Young 's modulus of the composite were significantly improved. Xu et al.[11] found that the higher the input energy of ball milling and the longer the time, the more serious the damage of CNT, and the defects will promote the formation of $Al_4C_3$ in the subsequent sintering or heat treatment process. The interfacial reaction rate of high-speed ball milling composites is 61.85%, which is much higher than 2.42% of low-speed ball milling. Although high-speed ball milling can make CNT embedded in Al matrix to achieve good bonding, it will seriously damage the crystal structure of CNT. Chen et al.[12] showed that amorphous natural $Al_2O_3$ could be transformed into $\gamma$-$Al_2O_3$ nanoparticles under the suitable SPS conditions of 540~635°C, 30~300 MPa, and 60 min, thereby improving the strain hardening ability of the composites. Chen et al.[13] used the cumulative extrusion bonding (AEB) process to process ultrafine-grained aluminum. The microstructure characterization showed that a good interface bonding was obtained after 6 passes of processing, and the average grain size was ≤440 nm. Compared with the traditional accumulative rolling bonding process, this process can refine the grains more efficiently and significantly improve the mechanical properties of the samples.

## 2.3 CNT In-Situ Generation Method

CNT in-situ formation method refers to the process of directly in-situ growth of CNT on the surface of metal particles. However, the carbon source used in this method to generate CNT is combustible gas, which not only has certain safety hazards, but also has low yield, thus limiting its practical application. Kwon et al.[14] prepared Al/CNTs composites by SPS combined with hot extrusion process. It was found that the addition of CNT can improve the tensile strength without reducing the elongation. The reason is that CNT exists in the grain boundary layer, and the formation of $Al_4C_3$ and the good orientation of CNT along the extrusion direction achieve effective stress transfer (Figure 3). Yang et al.[15] synthesized CNT in situ after depositing 5% (mass fraction) nickel on the surface of aluminum powder by impregnation method. The results show that the high crystallinity CNT synthesized in situ has a bamboo structure, which can avoid the interface reaction with aluminum matrix below 1,000 °C. The yield strength and elastic modulus of the composite with CNT content of 1.5% prepared by hot pressing are 2.2 times and 3 times that of the pure aluminum matrix, respectively.

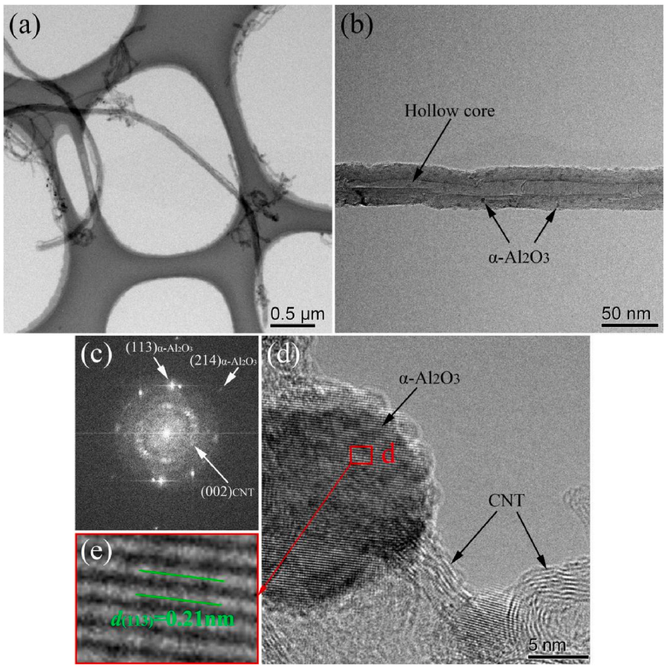
*Figure 3: High resolution transmission electron microscopy images of the microstructure of CNT/Al composites after extrusion[14]. (a) $Al_4C_3$ morphology, and the crystal structure of $Al_4C_3$ was confirmed by illustrations. The surface of (b) and (c) defective carbon nanotubes (CNT) forms tubular $Al_4C_3$, while the CNT tip forms granular $Al_4C_3$*



# 3. Surface Modification of CNT

The core of CNT surface modification is to improve the surface activity by regulating its surface state and microstructure, thereby improving its compatibility with the aluminum matrix and inhibiting the agglomeration of CNT, mainly including surface oxidation modification and surface coating treatment. In order to realize the surface functionalization of the original multi-walled CNT without destroying its structural integrity, Zhou et al.[16] modified the CNT by mild acid treatment. The appropriate amount of acid-treated CNT and Al powder were dispersed in ethanol respectively, and the suspension was mixed after 1 h ultrasonic treatment. The results showed that the modified CNT could be uniformly dispersed in the aluminum matrix. So et al.[17] coated silicon nanoparticles on the surface of CNT and formed a SiC layer after high temperature annealing. The SiC layer not only reduced the wetting angle between CNT and aluminum matrix and improved the interfacial wettability, but also significantly enhanced the interfacial bonding strength by forming SiC covalent bonds. Chen et al.[18] prepared 2195Al/CNT composites by $Al_2O_3$ partially coated modified CNT, combined with melting casting and hot extrusion. After aging treatment, the tensile strength, yield strength and elongation reach 697 MPa, 590 MPa and 11.3%, respectively, and the mechanical properties are significantly improved compared with those of 2195 alloy under the same conditions. CNT is mainly distributed in the grain boundary, and there is a small amount of fluorescent rod-like $Al_4C_3$ phase near it, which is formed by the segregation of the solute element interface of the alloy, as shown in Figure 4.

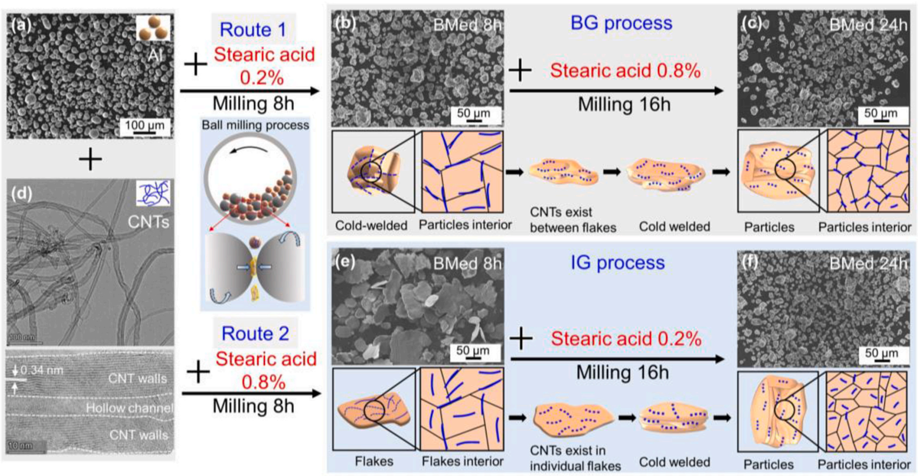
*Figure 4: TEM images[18]: (a-b) TEM images, (c) SAED patterns, (d-e) HRTEM images.*



# 4. Aluminum Matrix Alloying

Matrix alloying refers to the addition of a small amount of alloying elements (Si, Cr, etc.) to the aluminum matrix. Through the interfacial chemical reaction between the alloying elements and CNT, a certain thickness of the interfacial reaction layer is formed, thereby improving the interfacial bonding strength and inhibiting the reaction between CNT and the aluminum matrix to form $Al_4C_3$ phase which is easy to hydrolyze. Yuan et al.[19] introduced magnesium into aluminum powder when preparing CNT reinforced aluminum matrix composites. It was found that micro-nano strengthening phases such as $Al_4C_3$, $MgAl_2O_4$ and $Al_2O_3$ were formed at the bottom of the pits of the composites. At the same time, the $Al_2O_3$ layer is replaced by spinel phase ($MgAl_2O_4$), which is distributed both around the interface and inside the layered grains. Such fine nano-grains can effectively alleviate the stress concentration, eliminate the early crack source during the tensile process, and significantly improve the extensibility of the material. Cao et al.[20] reported a strategy to improve the ductility and tensile strength of CNTs/Al composites by embedding CNT into the matrix grains. The preparation process is shown in Figure 5. Two kinds of composites were prepared by modified ball milling powder metallurgy process, which were CNT mainly dispersed in the grain interior and mainly distributed in the grain boundary. The results show that compared with the composites with CNT distributed at grain boundaries and the reference composites, the composites with CNT dispersed inside the grains have higher tensile strength and ductility at different CNT contents.

*Figure 5: Preparation schematic diagram of CNTs/Al composite powder with different carbon nanotube distribution[20]*



## 5. Properties of CNT Reinforced Aluminum Matrix Composites

CNT reinforced aluminum matrix composites have excellent comprehensive mechanical properties. The uniform dispersion of CNT in aluminum matrix plays an increasingly important role in improving the comprehensive properties of composites. At the same time, the introduction of CNT can significantly reduce the thermal expansion coefficient of the composite material. On the one hand, the thermal expansion coefficient of CNT itself is almost zero, on the other hand, the CNT at the interface has a restraining effect on the aluminum matrix, thus inhibiting the thermal expansion of the composite material [21-23]. Since the conductivity of CNT is lower than that of aluminum matrix, its addition will lead to a decrease in the conductivity of the composites, and the conductivity decreases with the increase of CNT content. The study of wear performance shows that the friction coefficient and wear loss of the composites decrease with the increase of CNT content due to the self-lubricating effect of CNT. At the same time, the introduction of CNT improves the deformation resistance of the aluminum matrix, and the synergistic effect of the two makes the composite obtain excellent wear resistance[24-27].

## Conclusion

In this paper, the interface control and properties of CNT reinforced aluminum matrix composites are systematically reviewed. The three paths of preparation process, surface modification and matrix alloying work together to effectively improve the CNT dispersion and interface bonding strength, and achieve the improvement of material properties. At present, there are still some problems such as uneven dispersion of CNT and insufficient controllability of interface reaction. In the future, it is necessary to focus on process optimization, interface reaction regulation and performance balance to promote its development to engineering scale application.

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## Conflict of Interests

The authors declare that there is no conflict of interest regarding the publication of this paper.

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