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Smart Mechanical Powder Processing for Producing Carbon Nanotube Reinforced Aluminum Matrix Composites[†]

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Abstract

The central concern in the fabrication of carbon nanotube (CNT) reinforced metal composites is the well balance between uniform dispersion and structural integrity of CNTs. Rapid and uniform self-assembly of CNTs and spherical Aluminum (Al) particles into a core-shell structure is realized by a smart mechanical powder processing. The factors influencing the dispersion uniformity and structural integrity of CNTs during the processing are studied, including the size of Al particles, mixing speed and mixing time. It is revealed that a size of 35 μm is preferred for the Al particles to tear apart the CNT clusters and obtain a uniform dispersion of CNTs on Al surface. Different composite states, CNTs are singly dispersed, thickly wrapped, or embedded in the Al particles, can be obtained by changing the mixing speed. Well coordination between the CNT dispersion homogeneity and structural integrity could be achieved under suitable processing condition. Therefore, it can be adopted as an efficient and intelligent technology to achieve the desired performance in CNT/Al composites.

Keywords: dispersion, structural integrity, high-shear pre-dispersion, CNTs reinforced metal matrix composite, smart powder processing

1. Introduction

During the past two decades, significant progress has been achieved in the fabrication of carbon nanotubes reinforced metal matrix composites (MMCs). Regardless of the material type and fabrication technique, the balance between the uniform dispersion of CNTs in the Al matrix and its structural integrity is vital prerequisite that needs to be considered (Fan G.L. et al., 2014; Wei H. et al., 2014). How to realize the rapid, efficient, and uniform dispersion of CNTs in the Al matrix is one of the key factors restricting the industrial application of CNT/Al and alloy composite materials. It is well known that fine powders are not able to flow freely due to the strong inter-particle cohesion, mainly ascribed to the Van der Waals forces for dry, neutral, and inert particles (Deng X. and Davé R.N., 2017b; Eggersdorfer M.L. et al., 2010), dominating the particle weight. The situation can be more serious when the reinforcement is one-dimensional, namely tubular and flexible such as CNTs (Han Y. et al., 2015; Narh K.A. et al., 2008; Zhang K. and Choi H.J., 2014). The unique geometry, along with large specific surface area and significant Van der Waals force, strongly affects the dispersion status of CNTs.

In addition, the structural integrity of CNTs is considered to be another key factor affecting the strengthening effect by interfacial reaction and the formation of harmful products. Therefore, the MMCs community, especially those CNTs reinforced Aluminum composites, have significant concerns in overcoming the dilemmas associated with dispersion and structural integrity. From this point of view, these factors should be taken into account when designing appropriate preparation processes.

Presently, many CNT/Al composite preparation methods can meet the requirements of CNTs reinforced Al Matrix Composite, but each has its shortcomings. Melt casting and thermal spraying can realize the rapid preparation of large block materials with a high content of CNTs. Still, it is difficult to solve the dispersion of CNTs and the control of interface reaction (Baig Z. et al., 2018). Great efforts have also been made in the field of both large plastic deformation technologies such as accumulative roll bonding (ARB) (Salimi S. et al., 2010), high-pressure torsion (HPT) (Asgharzadeh H. et al., 2014; Tokunaga T. et al., 2008), friction stir processing (FSP) (Johannes L.B. et al., 2006; Sadeghi B. et al., 2018b; Sadeghi B. et al., 2018d) and solid-phase powder metallurgy (PM) techniques (Sadeghi B. et al., 2018a). One could be found that PM based techniques are effective and low cost, so that bring into a clear

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insight to solve the dilemma mentioned above. The PM based methods include high-energy ball milling (Esawi A.M.K. et al., 2009; Sadeghi B. et al., 2020; Sadeghi B. et al., 2019), flake powder metallurgy (Flake PM) (Fan G.L. et al., 2014; Jiang L. et al., 2012), liquid phase ball milling (Chen B. et al., 2015; Jiang L. et al., 2012), nanoscale dispersion method (Kwon H. et al., 2009; Noguchi T. et al., 2004), molecular level mixing (Cha S.I. et al., 2005; Nam D.H. et al., 2012), in-situ growth of CNTs combined with low-energy ball milling (He C., 2007; Cao L.L. et al., 2012; Yang X. et al., 2013) and so on (Baig Z. et al., 2018). However, whether wet ball milling or in-situ growth is used as the initial dispersion method of CNTs, it is a time-consuming and high energy consumption method, which is challenging to match with industrial production. Therefore, it is of great significance to study an effective pre-dispersion way for the preparation of CNT/Al composite with high performance. Precisely, despite recent advances in powder-based processing techniques such as Flake PM (Fan G.L. et al., 2014; Jiang L. et al., 2011; Jiang Y. et al., 2019; Tan Z. et al., 2011), there is still a challenging effort to achieve the extraordinary balance between dispersion and structural integrity of CNTs through Al matrix. In sum, it seems that PM is still the best macro quantitative preparation method for controllability and operability, but there is still much room for improvement.

Recently, various dry particle coating technologies have been developed for advanced material production through covering the surface of larger core (host) particles with fine shell (guest) particles by mechanical forces, which were induced through four different mechanical devices (Foppoli A.A. et al., 2017; Sharma R. and Setia G., 2019). All of those are well designed to prompt de-agglomeration of cohesive nanopowders (NP) to facilitate the NP dispersion (Bhaumik S., 2015; Naito M. et al., 2009; Wei D. et al., 2002). Among these technologies, Mechano Fusion® (MF) system as a smart powder process (Naito M. et al., 2009) has attained a considerable amount of attention to produce the composite powders with desired tailor-made properties for advanced MMCs (Chen M. et al., 2018; Ghoroi C. et al., 2013; Naito M. et al., 2009). Comparing to the wet-based (Jiang L. et al., 2012; Sadeghi B. et al., 2012a; Sadeghi B. et al., 2012b; Zhou S.-M. et al., 2007) and gas-based (He C. et al., 2007; Yang X. et al., 2016) coating techniques, the smart powder process can produce advanced composite materials with minimum energy consumption and environmental impact in a dry environment. In principle, the MF system is expected to achieve the dry and uniform composite of nano-sized CNTs and micron-sized metal powders. Still, CNTs are different from ordinary nanoparticles because of their complex structure and could be easily intertwined and clustering. In such circumstances, opening the clusters and wrapping on the surface of metal powder by mechanofusion is a challenging issue. Very recently, Chen et al. (Chen M. et al., 2018) demonstrated that dry particle coating (also termed as high-shear pre-dispersion) combined Flake PM route not only improves the dispersion efficiency of CNTs in the ball milling process but also well-preserves the structural integrity of CNTs comparing to other PM routes (Chen M. et al., 2018; Chen M. et al., 2019). Nevertheless, the effect of host particle size and the operation conditions on the dispersion and structural integrity of CNTs were not demonstrated. Considering the low cost, simplicity, and low energy consumption of the processing, there is a great potential to transit it from laboratory to large-scale commercial applications. Therefore, there is a pressing need to develop a better understanding of the factors that affect the coating effectiveness of the high-shear pre-dispersion process.

Many factors may affect the composite effects of particles, and subsequently, dry coating effectiveness, including host particle size, mixing speed and time. Very recently, Zheng et al. (Zheng K. et al., 2020) studied the effects of particle properties such as their sizes, stiffness and surface energies on the coating effectiveness and demonstrated that the host particle size has no significant effect on the coating quality when it is orders of magnitude larger than that of guest particles. They also reported that increasing processing intensity led to guest particles bouncing off of host particles, indicating poor coating performance. It was reported that the discharge efficiency of $LiNi_{0.5}Mn_{1.5}O_4$ cathode could enhance by the CNT coat provided by the mechanofusion method (Hwang T. et al., 2016). Narh et al. (Narh K.A. et al., 2008) studied dry coating of polymer powder particles with de-agglomerated CNTs and reported an acceptable extent of the CNT dispersity. Chen et al. (Chen M. et al., 2019) could effectively tailor the pre-dispersion state of CNT/6061Al composite powders by controlling the high-shear process speed. No comprehensive research has mentioned that host particle size and operating conditions (mixing time and speed of high-shear pre-dispersion process) simultaneously affect the dispersion and structural integrity degree in CNT/Al composite powders. To this end, the current study is trying to uncover the various aspects of the pre-dispersion and structural integrity degree of CNTs in the Al matrix composite. It should be interesting to note that the dispersion and binding mechanisms involved in the high-shear pre-dispersion process depend on the initial size of host particles and guest particles (Ötles S. et al., 2009; Wei D. et al., 2002; Zheng K. et al., 2020). However, it is well known that the contact surface between powder materials is subjected to extremely high local temperature and pressure, resulting in unique phenomena between host and guest or among fine particles or even generating mechanical stress (Naito M. et al., 2009).

In this study, an environmentally friendly and smart powder processing technology, the namely high-shear pre-dispersion process is successfully adopted to fabricate

1.5 wt.% CNT/Al composite powders. Specifically, the effects of host particle size, mixing time, and rotational speed of powder mixing process on the balance of dispersity and structural integrity of CNTs in final composite powders and consequently composite effects are discussed.

2. Experimental

2.1 Raw materials

Atomized near-spherical Al powders with a various average diameter of 2, 5, 10, 15, 35, 70 μm, and industrial multi-walled CNTs with a diameter of about 10–20 nm, length of about 0.5–2 μm and purity of more than 95 % are chosen to explore the size dependence in the high-shear pre-dispersion process. **Fig. 1a** shows the original shape of 35 μm-Al powder have a round surface. A small number of tiny Al particles are attached on the surface, and size distribution is relatively narrow. **Fig. 1b–c** show the initial CNTs, which are in the form of particles with a size of tens of microns. Long and entangled CNTs can be seen at higher magnifications. **Fig. 1d** shows the Raman spectrum of raw CNTs. It is well-known that there are two characteristic peaks at 1346 cm^{-1} and 1570 cm^{-1}, which are called D peak and G peak, respectively (Cavaliere P. et al., 2019; Kwon H. and Leparoux M., 2012; Sadeghi B. et al., 2018c; Zhao Q. and Wagner H.D., 2004). Former is attributed to defects in the hexagonal graphite layer structure, resulting in the in-plane vibration. The latter is related to a single-photon emission generated by the tangential tensile mode of the graphite layer plane (Loa I., 2003; Zhao Q. and Wagner H.D., 2004). The ratio of two peaks (I_D/I_G) is usually used to characterize the graphitization degree of CNTs. The lower the ratio, the fewer the defects of CNTs and the higher graphitization degree is. So it is useful to track the changing trend of the structural integrity of CNTs (Chen M. et al., 2019). The I_D/I_G value of raw CNTs is about 0.95.

2.2 Preparation of CNT/Al composite powder

1.5 wt.% of CNTs were processed with Al powder of different diameters in a prototype of MF system (MECHANO FUSION®) from Hosokawa Micron Corporation, Japan (Naito M. et al., 1993). As illustrated in **Fig. 2a**, it has a container diameter of 80 mm and the clearance/gap between the rotor and container is of 2 mm. The powder mixture was first processed at the speed of 300 rpm for 2 min to homogenize and then processed at the designated speed (1000–4000 rpm) for different times (5–15 min) to obtain composite powders.

2.3 Characterization

The morphology of the powder is observed by Sirion 200 field emission scanning electron microscope (FESEM) at the working voltage of 20 kV. The influence of the process condition on the structural integrity of CNTs is characterized by Raman spectroscopy (SENTERRA R200-L), by using the 532 nm line of an Ar+ laser as the excitation source.

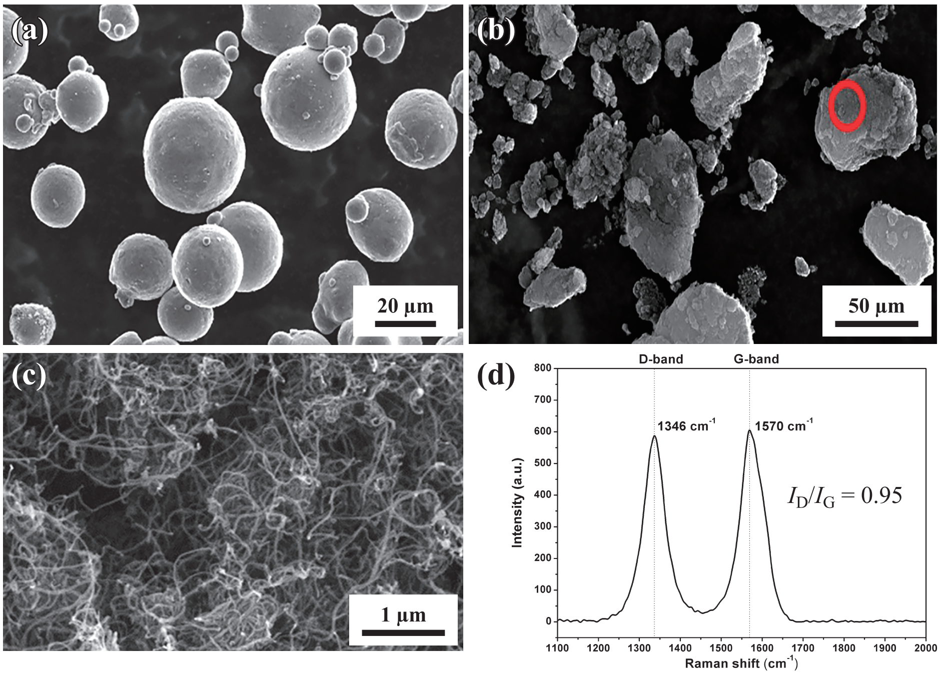

Fig. 1 FESEM of **(a)** spherical 35 μm-Al powders, **(b)** raw CNTs, **(c)** magnification of CNTs in **(b)**, **(d)** Raman spectrum of raw CNTs.

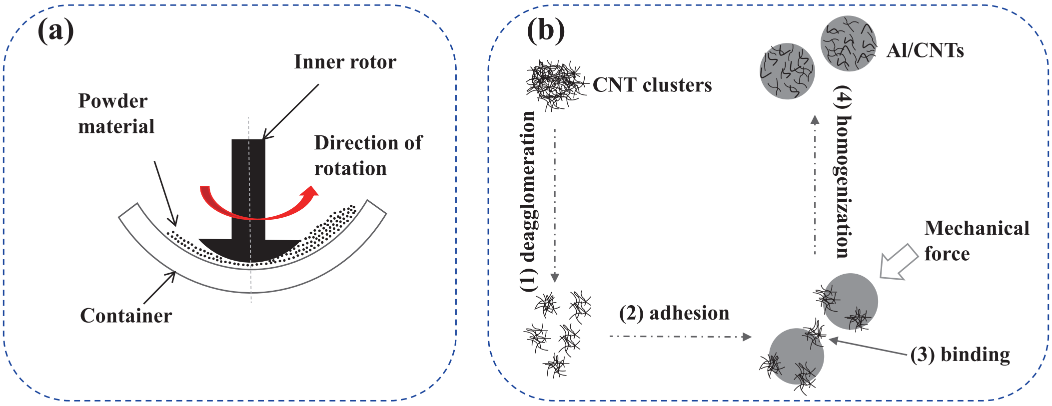

Fig. 2 Illustrations of **(a)** prototype of Mechano Fusion® system (Naito M. et al., 1993), **(b)** the working principle of CNTs dispersion on Al powder in MF system.

3. Result and discussion

The main working principle of the MF system is to use the centrifugal force generated by the high-speed rotation of the rotor and the gravity effect of the material itself to make the material pass through the clearance between the rotor and the container at high speed. The mutual shear force and impact force between the materials make the smaller particles adhered onto the more considerable materials, to realize the core-shell structured composite particles (Naito M. et al., 2009; Qu L. et al., 2015). In the case of CNTs and Al particles, as illustrated in **Fig. 2b**, the CNTs as the guest particles should be first torn apart by the shear forces of Al particles (host particles) into smaller particles or even individual CNTs, and then adhered onto the surface of Al particles. The repeating collision between Al particles made CNTs uniformly dispersed on the surface of Al particles, forming a Al/CNTs core-shell structured composite powder. Therefore, the dispersion status is mostly affected by the particle size or mass ratio between host/guest particles, the processing speed and time.

3.1 Effect of Al particle size

The average particle size of the initial Al powders not only determines the surface area of the powders but also determines the mass ratio between the particles and CNTs. On the one hand, the smaller the average particle size of Al powder is, the larger the corresponding surface area is, which is more conducive to the uniform distribution of CNTs; on the other hand, the smaller the mass of a single Al particle is, the weaker its impact energy on CNTs clusters will be. Precisely, smaller Al particles provide lower collision energy but higher surface area, which in turn controls the type of contacts and interparticle adhesion force (Deng X. and Davé R.N., 2017a) and subsequent dispersion of CNTs.

Fig. 3 shows FESEM of CNT/Al composite powder obtained by mixing 1.5 wt.% CNTs with Al powder of different particle sizes at 2500 rpm for 10 min. As one can see, the shape of Al particle is not significantly changed during the high-shear pre-mixing process as reported (Chen M. et al., 2018). The main reason for this was due to the fact that the processing condition applied was still below the limit of Al particle deformation. For the smaller Al particle size, namely 2, 5 µm, the de-agglomeration and dispersion of CNTs hardly occurred. Most CNTs remained in clusters (indicates by yellow arrows in **Fig. 3a–b)**; only very few individual CNTs were attached on the surface of Al particles (**Fig. 3d–e**). When the particle size increased to 10, 15 µm, more individual CNTs were attached on the surface of Al particles, while a small number of clusters still existed. Further increasing the particle size to 35, 70 µm, only tiny flattened CNT clusters can be occasionally found, indicating almost all the CNTs were torn apart and attached on the surface of Al particles. That is, the larger the particle size of the Al particle is, the more CNTs are attached to the surface. The larger the mass or volume of Al particles is, the easier it is to wrap CNTs on the surface. The initial size of the CNTs clusters is of tens of microns, and so the collision force exerted from those small Al particles (smaller than 10 µm) are not high enough to tear the CNT clusters apart. Still, on the contrary, these collisions made the CNT clusters more solid than initial CNTs. Therefore, there is a size limit that the pre-dispersion of CNTs on the Al surface can be applied.

Further observation of the micromorphology of the Al particle surface shows that the degree of the surface smoothness is changed in larger sizes of Al particle. The surface of the Al particle is no longer smooth, and a certain number of pits appear, which should be related to the significant improvement of the impact force between the larger Al particles. The small Al particles that initially attached on the surface of big particles were flattened and stuck on the Al surface, making a rougher surface (**Fig. 3i**) with CNTs embedded in the surface pits. By and large,

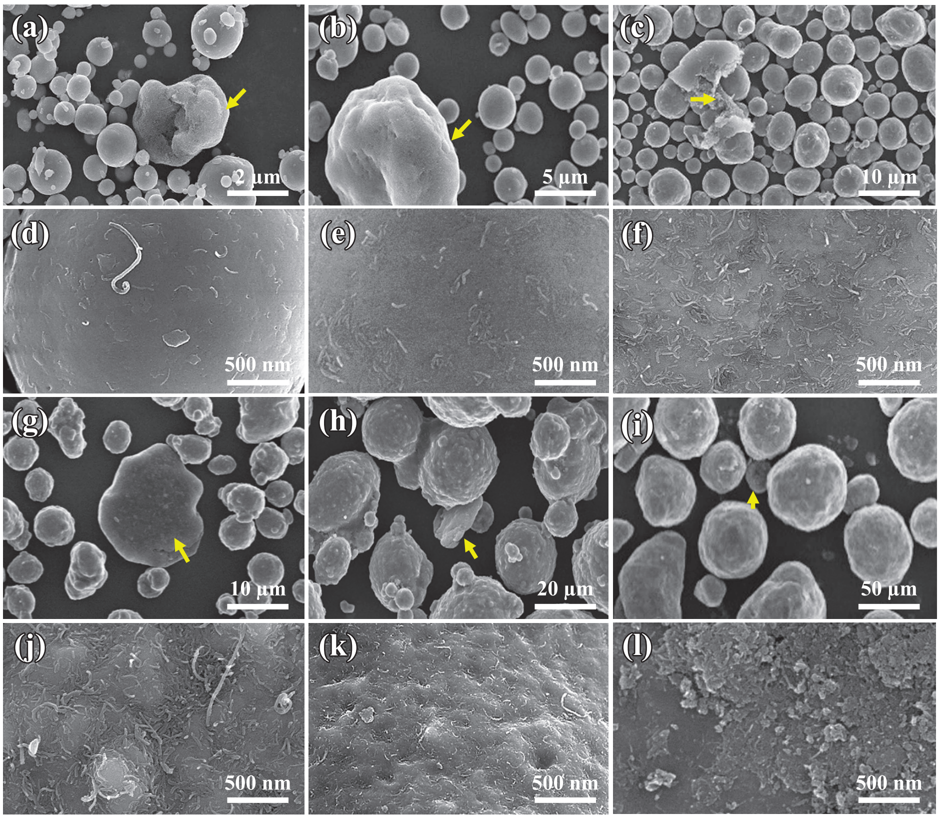

Fig. 3 FESEM of pre-mixed CNT/6061Al composite powders with different particle sizes processed at 2500 rpm for 10 min, **(a, d)** 2 µm, **(b, e)** 5 µm, **(c, f)** 10 µm, **(g, j)** 15 µm, **(h, k)** 35 µm, **(i, l)** 70 µm. Arrows indicate the presence of the CNT clusters.

under the current mixing conditions, Al particles with the average size above 35 µm can provide the appropriate level of pre-dispersion of CNTs and complete the pre-dispersion process well. Considering that the essential purpose of the pre-dispersion process is to reduce the pressure of subsequent powder milling, to shorten the time required for CNT dispersion, as well as keeping the structural integrity of CNTs, the Al powders with 35 µm particle size is preferred.

3.2 Effect of pre-dispersion speed (rotational speed) on CNT dispersion

It is well known that the force acting on the powder blend generally increases with rotational speed (Naito M. et al., 1993). The higher the speed, the more robust and useful the interaction force can be produced between the materials and subsequently affect the flowability of the processed powder. As described in the previous section, the 35 µm Al powder succeeds in completing the task of opening the CNT clusters completely at 2500 rpm. Therefore, it is considered to alter the pre-dispersion speed further and observe its effect on the dispersion status.

Fig. 4 shows the FESEM of CNT/Al composite powder after mixing 35 µm Al powder with CNTs for 10 min at different rotational speeds. Big and flattened CNT clusters can be easily observed at low rotational speed (1000 rpm, **Fig. 4a**), but the amount and size of CNT clusters progressively decreased with the increasing rotational speed up to 2000 rpm. A thick layer of dispersed CNTs can be observed on Al particle surface at 2000 rpm, as shown in **Fig. 4f**. With the increase of mixing speed to 2500, 3000, 4000 rpm, small Al chips were attached on the surface of Al particles, and only a small number of CNT ends can be observed on the surface of Al particles, as shown in **Fig. 4j–l**. It can be deduced that with the increase of speed, CNTs show a gradual "in-depth" change process from the surface to the interior of the Al particle, which is completed by the embedding of CNTs into Al chips and the mutual grinding of Al powders.

The above results depict the trend that de-agglomeration and dispersion of CNTs increase as a function of increasing rotational speed. This is because as the rotational speed increases, the intensity of collisions between Al particles and CNT clusters violently increases when passing through the clearance between the rotor and container. Because of severe shear, friction, and compression forces, big CNT clusters are quickly torn apart and broken into the smaller ones. And then the small CNT clusters adhered on the surfaces of Al particles were further de-agglomerated and uniformly dispersed throughout the surface of Al particles by the repeating collision between Al particles throughout the processing time.

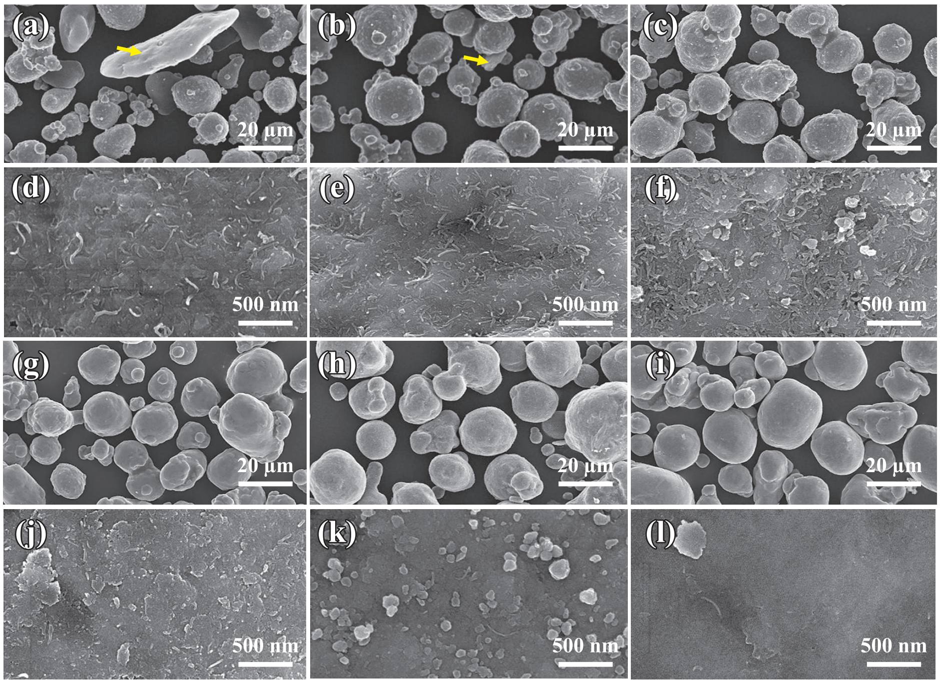

Fig. 4 FESEM images of Al at CNT composite powders processed by high-shear pre-dispersion process with various rotational speed, **(a, d)** 1000 rpm; **(b, e)** 1500 rpm; **(c, f)** 2000 rpm; **(g, j)** 2500 rpm, **(h, k)** 3000 rpm, **(i, l)** 4000 rpm. The yellow arrows indicate the CNT clusters.

The tentative conclusion to be drawn is that by changing the mixing speed (rotational speed), the type of contacts between CNTs (as guest) and Al particles (as host) can be changed to strongly affect the dispersity status. On the one hand, at too slow rotational speeds, guest-guest (CNT clusters) and host-host (Al-Al) contacts remain due to insufficient mechanical forces. And on the other hand, at high rotational speeds, CNTs both in the form of cluster and individual are embedded in Al particles (host-guest contact). The former results in the remaining of CNT cluster, and the latter causes heterogeneity dispersion of CNTs in Al particles. Therefore, it can be inferred that the best condition for uniform dispersion of CNTs is that CNTs are singly dispersed throughout the surface of Al particles at a moderate rotational speed.

Additionally, it is shown that when rotational speed increases from 2500 rpm to 4000 rpm, small Al particles attached on the surface of bigger ones were gradually stuck and probably embedded in Al particles, generating Al particles with a bigger size and smoother surface (**Fig. 4h–i**), compared to lower rotational speeds (**Fig. 4a–b**).

3.3 Effect of pre-dispersion time on CNTs dispersion

The mixing process of CNTs and Al powder is the process of gradually opening and coating CNTs clusters on the surface of Al powder. The efficiency of the dry particle coating process is determined by the total mass, mass ratio, mixing speed and mixing time. Among these affecting parameters, mixing time not only has a significant influence on dispersity status and structural integrity of CNTs but also leads to alteration of the appearance of Al powder.

To study the effect of mixing time on the dispersion status of CNTs during the high-shear pre-dispersion process, the Al powder and CNTs with a medium particle size of 35 µm are selected as the primary materials. According to the tailored objectives, two different rotational speeds of 1500 rpm and 2500 rpm are chosen to do the mixing process at different mixing times of 5, 10 and 15 min. **Fig. 5a–c** show that the CNTs on the surface of Al particles can be observed under 1500 rpm for different mixing times. After a short mixing time (5–10 min, **Fig. 5a–b**), CNTs coated onto the Al surfaces are discrete, and by increasing the mixing time, a continuous film-like CNT coating is gradually formed (15 min, **Fig. 5c**). Also, the length of CNTs decreased from 0.5–2 µm to 0.2–0.5 µm, indicating that CNTs clusters were cut short during the opening process due to shear and compression stresses. For the mixing time of 15 min, no CNT clusters can be observed, which indicates that CNTs have been completely attached on the surface of Al particles. **Fig. 5d–f** show the CNT dispersion status at 2500 rpm for various mixing times. After 5 min processing, we can see that more CNTs are attached on the surface than mixed at the speed of 1500 rpm for the same time. With the increase of processing time, most CNTs are embedded underneath the Al chips, which should be attributed to the presence of the enormous impact, compression, and shear forces between the Al particles when they passed through the clearance between the rotor

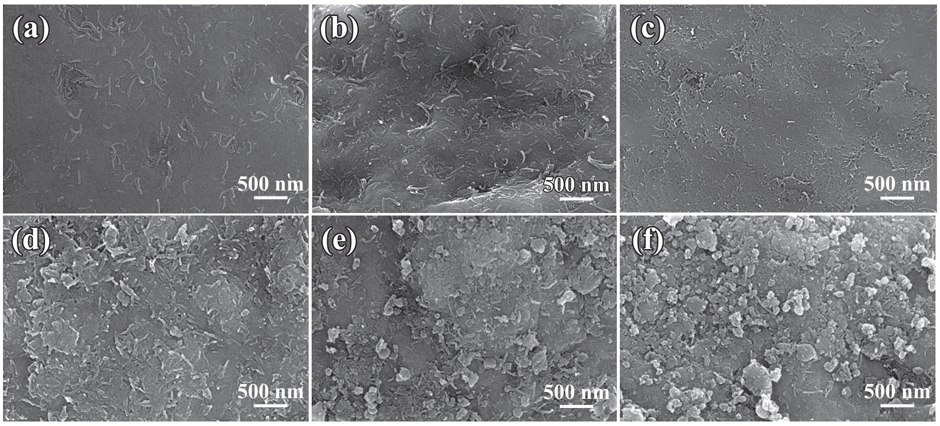

Fig. 5 FESEM images of CNT/Al composite powders premixed at **(a–c)** 1500 rpm and **(d–f)** 2500 rpm for different time: **(a, d)** 5 min, **(b, e)** 10 min, **(c, f)** 15 min.

and container. Due to such severe shear and compression stress, a long-time mixing may result in the embedding of some CNT clusters underneath Al chips before they were fully de-agglomerated and spread over the surfaces. It could increase the responsibility of subsequent ball milling dispersion at the expense of more structure damage of CNTs under high energy ball milling dispersion. It can, therefore, be used to suggest an optimum mixing time to achieve uniform distribution of individual CNTs onto the surface of Al particles without embedding them and thus, the improvement of pre-dispersion efficiency.

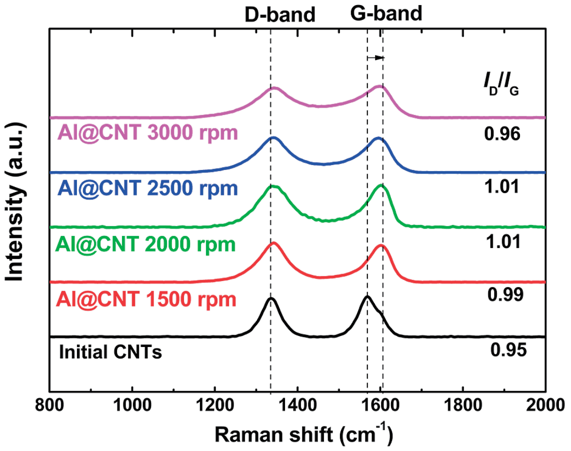

Fig. 6 Raman spectrum of pre-dispersed CNT/Al composite powder at different rotational speeds.

3.4 Pre-mixing speed dependence of structural integrity

The good maintenance of structural integrity of CNTs is considered to be one of the main factors that should be considered during the processing, although some results showed that in the absence of a significant interface reaction, the failure of the tubular structure on the surface of CNTs can enhance the interface bonding between CNTs and Al matrix, its bearing capacity and strengthening effect (Chen B. et al., 2014; Ci L.J. et al., 2006; Kwon H. et al., 2013).

As shown in **Fig. 6**, compared with the I_D/I_G value (0.95) of the initial CNTs powder, it is found that the mechanofusion pre-dispersion process had little damage to the structural integrity of CNTs. With the increase of rotational speed, the I_D/I_G value first increased to about 1.01 and then decreased to about 0.96 at 2800 rpm. The decrement of the I_D/I_G value indicates less damage. This is very likely that, the pre-dispersed CNTs are rapidly embedded underneath the surface of the Al chips at high speed, resulting in a better protection of CNTs (Chen M. et al., 2019). Therefore, it can be concluded that CNTs are just cut short efficiently without much structural integrity damage during the pre-dispersion process. The structural integrity is much better preserved than those traditional ball milling dispersion process.

To sum up, for the raw CNT clusters with dozens of microns, the size of Al particles should over 15 µm, as to provide sufficient impact energy to tear apart the clusters via mechanical forces under the MF system. Changing the rotational speed can obtain different composite states. With the increase of speed, CNTs show a gradual "in-depth" change from the surface to the interior of the Al particles, which is completed by the embedding of CNTs into Al chips and the mutual grinding of Al powders. However, the effect of their combination with the subsequent powder milling process needs to be considered to select the best pre-dispersion process parameters. Chen's work (Chen M. et al., 2019) shows that only by pre-dispersion at moderate speed, in which CNTs are well de-agglomerated and partially embedded in Al matrix, can provide a controllable balance between the CNT dispersity and structural integrity protection compared with the other processing conditions.

4. Conclusion

The smart powder processing based on the Mechano Fusion® system can realize the rapid composite of CNTs and Al powders. The particle size of Al powders, the mixing speed and time have a significant influence on the dispersion status. For the raw CNTs with dozens of microns, there is a size and speed limit to provide sufficient impact to tear apart the clusters and coat them onto the surface of Al particles. Considering the dispersion uniformity and surface area of Al powders to bear CNTs, the size of 35 μm Al powder and the speed around 2000–2500 rpm are preferred. The efficiency of processing is very high and under suitable speed, 15 min is sufficient to obtain a uniform dispersion of CNTs all over the surfaces of Al particles. The CNTs were cut short, but structural integrity was not severely damaged. Therefore, a high-shear pre-dispersion process as a smart mechanical powder processing is a relatively simple and environmentally friendly approach to produce CNT/Al composite powder with the desired balance between dispersion and structural integrity of CNTs in metallic matrix.

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References

Asgharzadeh H., Joo S.-H., Kim H.S., Consolidation of carbon nanotube reinforced aluminium matrix composites by high-pressure torsion, Metallurgical and Materials Transactions A, 45 (2014) 4129–4137. DOI: 10.1007/s11661-014-2354-6

Baig Z., Mamat O., Mustapha M., Recent progress on the dispersion and the strengthening effect of carbon nanotubes and graphene-reinforced metal nanocomposites: a review, Critical Reviews in Solid State and Materials Sciences, 43 (2018) 1–46. DOI: 10.1080/10408436.2016.1243089

Bhaumik S., Principles and applications of mechanical dry coating—review and state-of-the-art, 5, Master of Science, New Jersey Institute of Technology, 2015. https://digitalcommons.njit.edu/theses/223/

Cao L.L., Li Z.Q., Fan G.L., Jiang L., Zhang D., Moon W.J., Kim Y.S., The growth of carbon nanotubes in aluminum powders by the catalytic pyrolysis of polyethylene glycol, Carbon, 50 (2012) 1057–1062. DOI: 10.1016/j.carbon.2011.10.011

Cavaliere P., Sadeghi B., Shamanian M., Ashrafizadeh F., Al-based nanocomposites produced via spark plasma sintering: effect of processing route and reinforcing phases, in: Cavaliere P. (Ed.) Spark Plasma Sintering of Materials: Advances in Processing and Applications, Springer International Publishing, Cham, Switzerland, 2019, pp. 161–190. ISBN: 978-3-030-05327-7. DOI: 10.1007/978-3-030-05327-7

Cha S.I., Kim K.T., Arshad S.N., Mo C.B., Hong S.H., Extraordinary strengthening effect of carbon nanotubes in metal-matrix nanocomposites processed by molecular-level mixing, Advanced Materials, 17 (2005) 1377–1381.

Chen B., Jia L., Li S., Imai H., Takahashi M., Kondoh K., In situ synthesized Al_4C_3 nanorods with excellent strengthening effect in aluminum matrix composites, Advanced Engineering Materials, 16 (2014) 972–975. DOI: 10.1002/adem.201400232

Chen B., Li S., Imai H., Jia L., Umeda J., Takahashi M., Kondoh K., An approach for homogeneous carbon nanotube dispersion in Al matrix composites, Materials & Design, 72 (2015) 1–8. DOI: 10.1016/j.matdes.2015.02.003

Chen M., Fan G., Tan Z., Xiong D., Guo Q., Su Y., Zhang J., Li Z., Naito M., Zhang D., Design of an efficient flake powder metallurgy route to fabricate CNT/6061Al composites, Materials & Design, 142 (2018) 288–296. DOI: 10.1016/j.matdes.2018.01.044

Chen M., Fan G., Tan Z., Yuan C., Xiong D., Guo Q., Su Y., Naito M., Li Z., Tailoring and characterization of carbon nanotube dispersity in CNT/6061Al composites, Materials Science and Engineering: A, 757 (2019) 172–181. DOI: 10.1016/j.msea.2019.04.093

Ci L.J., Ryu Z.Y., Jin-Phillipp N.Y., Ruhle M., Investigation of the interfacial reaction between multi-walled carbon nanotubes and aluminum, Acta Materialia, 54 (2006) 5367–5375. DOI: 10.1016/j.actamat.2006.06.031

Deng X., Davé R.N., Adhesion and friction of dry-coated nano-rough particles, Powder Technology, 314 (2017a) 20–27. DOI: 10.1016/j.powtec.2016.07.035

Deng X., Davé R.N., Breakage of fractal agglomerates, Chemical Engineering Science, 161 (2017b) 117–126. DOI: 10.1016/j.ces.2016.12.018

Eggersdorfer M.L., Kadau D., Herrmann H.J., Pratsinis S.E., Fragmentation and restructuring of soft-agglomerates under shear, Journal of Colloid and Interface Science, 342 (2010) 261–268. DOI: 10.1016/j.jcis.2009.10.062

Esawi A.M.K., Morsi K., Sayed A., Gawad A.A., Borah P., Fabrication and properties of dispersed carbon nanotube–aluminum composites, Materials Science and Engineering: A, 508 (2009) 167–173. DOI: 10.1016/j.msea.2009.01.002

Fan G., Jiang Y., Tan Z., Guo Q., Xiong D.-B., Su Y., Lin R., Hu L., Li Z., Zhang D., Enhanced interfacial bonding and mechanical properties in CNT/Al composites fabricated by flake powder metallurgy, Carbon, 130 (2018) 333–339. DOI: 10.1016/j.carbon.2018.01.037

Fan G.L., Xu R., Tan Z.Q., Zhang D., Li Z.Q., Development of flake powder metallurgy in fabricating metal matrix composites: A review, Acta Metallurgica Sinica-English Letters, 27 (2014) 806–815. DOI: 10.1007/s40195-014-0148-x

Foppoli A.A., Maroni A., Cerea M., Zema L., Gazzaniga A., Dry coating of solid dosage forms: an overview of processes and applications, Drug Development and Industrial Pharmacy, 43 (2017) 1919–1931. DOI: 10.1080/03639045.2017.1355923

Ghoroi C., Han X., To D., Jallo L., Gurumurthy L., Davé R.N., Dispersion of fine and ultrafine powders through surface modification and rapid expansion, Chemical Engineering Science, 85 (2013) 11–24. DOI: 10.1016/j.ces.2012.02.038

Han Y., Zhang X., Yu X., Zhao J., Li S., Liu F., Gao P., Zhang Y., Zhao T., Li Q., Bio-inspired aggregation control of carbon nanotubes for ultra-strong composites, Scientific Reports, 5 (2015) 11533. DOI: 10.1038/srep11533

He C., Zhao N., Shi C., Du X., Li J., Li H., Cui Q., An approach to obtaining homogeneously dispersed carbon nanotubes in Al powders for preparing reinforced Al-matrix composites, Advanced Materials, 19 (2007) 1128–1132. DOI: 10.1002/adma.200601381

Hwang T., Lee J.K., Mun J., Choi W., Surface-modified carbon nanotube coating on high-voltage $LiNi_{0.5}Mn_{1.5}O_4$ cathodes for lithium ion batteries, Journal of Power Sources, 322 (2016) 40–48. DOI: 10.1016/j.jpowsour.2016.04.118

Jiang L., Li Z., Fan G., Cao L., Zhang D., The use of flake powder metallurgy to produce carbon nanotube (CNT)/aluminum composites with a homogenous CNT distribution, Carbon, 50 (2012) 1993–1998. DOI: 10.1016/j.carbon.2011.12.057

Jiang L., Li Z.Q., Fan G.L., Zhang D., A flake powder metallurgy approach to Al_2O_3/Al biomimetic nanolaminated composites with enhanced ductility, Scripta Materialia, 65 (2011) 412–415. DOI: 10.1016/j.scriptamat.2011.05.022

Jiang Y., Wang D., Liang S., Cao F., Zou J., Xiao P., Effect of local alloying on interfacial bonding in laminated copper matrix composites reinforced by carbon nanotubes, Materials Science and Engineering: A, 748 (2019) 173–179. DOI: 10.1016/j.msea.2019.01.013

Johannes L.B., Yowell L.L., Sosa E., Arepalli S., Mishra R.S., Survivability of single-walled carbon nanotubes during friction stir processing, Nanotechnology, 17 (2006) 3081–3084. DOI: 10.1088/0957-4484/17/12/044

Kwon H., Estili M., Takagi K., Miyazaki T., Kawasaki A., Combination of hot extrusion and spark plasma sintering for producing carbon nanotube reinforced aluminum matrix composites, Carbon, 47 (2009) 570–577. DOI: 10.1016/j.carbon.2008.10.041

Kwon H., Leparoux M., Hot extruded carbon nanotube reinforced aluminum matrix composite materials, Nanotechnology, 23 (2012) 415701. DOI: 10.1088/0957-4484/23/41/415701

Kwon H., Takamichi M., Kawasaki A., Leparoux M., Investigation of the interfacial phases formed between carbon nanotubes and aluminum in a bulk material, Materials Chemistry and Physics, 138 (2013) 787–793. DOI: 10.1016/j.matchemphys.2012.12.062

Loa I., Raman spectroscopy on carbon nanotubes at high pressure, Journal of Raman Spectroscopy, 34 (2003) 611–627. DOI: 10.1002/jrs.1035

Naito M., Abe H., Kondo A., Yokoyama T., Huang C.C., Smart powder processing for advanced materials, KONA Powder and Particle Journal, 27 (2009) 130–143. DOI: 10.14356/kona.2009013

Naito M., Kondo A., Yokoyama T., Applications of Comminution Techniques for the Surface Modification of Powder Materials, ISIJ International, 33 (1993) 915–924. DOI: 10.2355/isijinternational.33.915

Nam D.H., Cha S.I., Lim B.K., Park H.M., Han D.S., Hong S.H., Synergistic strengthening by load transfer mechanism and grain refinement of CNT/Al–Cu composites, Carbon, 50 (2012) 2417–2423. DOI: 10.1016/j.carbon.2012.01.058

Narh K.A., Agwedicham A.-T., Jallo L., Dry coating polymer powder particles with deagglomerated carbon nanotubes to improve their dispersion in nanocomposites, Powder Technology, 186 (2008) 206–212. DOI: 10.1016/j.powtec.2007.12.004

Noguchi T., Magario A., Fukazawa S., Shimizu S., Beppu J., Seki M., Carbon nanotube/aluminium composites with uniform dispersion, Materials Transactions, 45 (2004) 602–604. DOI: 10.2320/matertrans.45.602

Ötles S., Lecoq O., Dodds J.A., Dry particle high-impact coating of biopowders: coating strength, Particulate Science and Technology, 27 (2009) 352–361. DOI: 10.1080/02726350902993987

Qu L., Morton D.A., Zhou Q.T., Particle engineering via mechanical dry coating in the design of pharmaceutical solid dosage forms, Current Pharmaceutical Design, 21 (2015) 5802–5814. DOI: 10.2174/1381612821666151008151001

Sadeghi B., Cavaliere P., Nosko M., Trembošová V., Nagy Š., Hot deformation behaviour of bimodal sized Al_2O_3/Al nanocomposites fabricated by spark plasma sintering, Journal of Microscopy, (2020). DOI: 10.1111/jmi.12947

Sadeghi B., Cavaliere P., Roeen G.A., Nosko M., Shamanian M., Trembošová V., Nagy Š., Ebrahimzadeh N., Hot rolling of MWCNTs reinforced Al matrix composites produced via spark plasma sintering, Advanced Composites and Hybrid Materials, 2 (2019) 549–570. DOI: 10.1007/s42114-019-00095-7

Sadeghi B., Sarraf-Mamoory R., Shahverdi H.R., Surface modification of $LiMn_2O_4$ for lithium batteries by nanostructured $LiFePO_4$ phosphate, Journal of Nanomaterials, 2012 (2012a) 743236. DOI: 10.1155/2012/743236

Sadeghi B., Sarraf-Mamoory R., Shahverdi H.R., The effect of $LiFePO_4$ coating on electrochemical performance of $LiMn_2O_4$ cathode material, Iranian Journal of Chemistry and Chemical Engineering (IJCCE), 31 (2012b) 29–34. https://www.sid.ir/en/journal/ViewPaper.aspx?id=276841

Sadeghi B., Shabani A., Cavaliere P., Hot rolling of spark-plasma-sintered pure aluminium, Powder Metallurgy, 61 (2018a) 285–292. DOI: 10.1080/00325899.2018.1478779

Sadeghi B., Shamanian M., Ashrafizadeh F., Cavaliere P., Rizzo A., Friction stir processing of spark plasma sintered aluminum matrix composites with bimodal micro- and nano-sized reinforcing Al_2O_3 particles, Journal of Manufacturing Processes, 32 (2018b) 412–424. DOI: 10.1016/j.jmapro.2018.03.013

Sadeghi B., Shamanian M., Cavaliere P., Ashrafizadeh F., Effect of processing parameters on the microstructural and mechanical properties of aluminum–carbon nanotube composites produced by spark plasma sintering, International Journal of Materials Research, 109 (2018c) 900–909. DOI: 10.3139/146.111686

Sadeghi B., Shamanian M., Cavaliere P., Ashrafizadeh F., Sanayei M., Szpunar J.A., Microstructural and mechanical behavior of bimodal reinforced Al-based composites produced by spark plasma sintering and FSP, International Journal of Advanced Manufacturing Technology, 94 (2018d) 3903–3916. DOI: 10.1007/s00170-017-1144-x

Salimi S., Izadi H., Gerlich A.P., Fabrication of an aluminum–carbon nanotube metal matrix composite by accumulative roll-bonding, Journal of Materials Science, 46 (2010) 409–415. DOI: 10.1007/s10853-010-4855-z

Sharma R., Setia G., Mechanical dry particle coating on cohesive pharmaceutical powders for improving flowability—a review, Powder Technology, 356 (2019) 458–479. DOI: 10.1016/j.powtec.2019.08.009

Tan Z., Li Z., Fan G., Li W., Liu Q., Zhang W., Zhang D., Two-dimensional distribution of carbon nanotubes in copper flake powders, Nanotechnology, 22 (2011) 225603. DOI: 10.1088/0957-4484/22/22/225603

Tokunaga T., Kaneko K., Horita Z., Production of aluminum-matrix carbon nanotube composite using high pressure torsion, Materials Science and Engineering: A, 490 (2008) 300–304. DOI: 10.1016/j.msea.2008.02.022

Wei D., Dave R., Pfeffer R., Mixing and characterization of nanosized powders: an assessment of different techniques, Journal of Nanoparticle Research, 4 (2002) 21–41. DOI:

10.1023/A:1020184524538

Wei H., Li Z.Q., Xiong D.B., Tan Z.Q., Fan G.L., Qin Z., Zhang D., Towards strong and stiff carbon nanotube-reinforced high-strength aluminum alloy composites through a microlaminated architecture design, Scripta Materialia, 75 (2014) 30–33. DOI: 10.1016/j.scriptamat.2013.11.014

Yang X., Liu E., Shi C., He C., Li J., Zhao N., Kondoh K., Fabrication of carbon nanotube reinforced Al composites with well-balanced strength and ductility, Journal of Alloys and Compounds, 563 (2013) 216–220. DOI: 10.1016/j.jallcom.2013.02.066

Yang X., Zou T., Shi C., Liu E., He C., Zhao N., Effect of carbon nanotube (CNT) content on the properties of in-situ synthesis CNT reinforced Al composites, Materials Science and Engineering: A, 660 (2016) 11–18. DOI: 10.1016/j.msea.2016.02.062

Zhang K., Choi H.J., Smart polymer/carbon nanotube nanocomposites and their electrorheological response, Materials, 7 (2014) 3399–3414. DOI: 10.3390/ma7053399

Zhao Q., Wagner H.D., Raman spectroscopy of carbon-nanotube-based composites, Philosophical Transactions of the Royal Society of London Series A: Mathematical, Physical and Engineering Sciences, 362 (2004) 2407–2424. DOI: 10.1098/rsta.2004.1447

Zheng K., Kunnath K., Ling Z., Chen L., Davé R.N., Influence of guest and host particle sizes on dry coating effectiveness: when not to use high mixing intensity, Powder Technology, 366 (2020) 150–163. DOI: 10.1016/j.powtec.2020.02.059

Zhou S.-M., Zhang X.-B., Ding Z.-P., Min C.-Y., Xu G.-L., Zhu W.-M., Fabrication and tribological properties of carbon nanotubes reinforced Al composites prepared by pressureless infiltration technique, Composites Part A: Applied Science and Manufacturing, 38 (2007) 301–306. DOI: 10.1016/j.compositesa.2006.04.004

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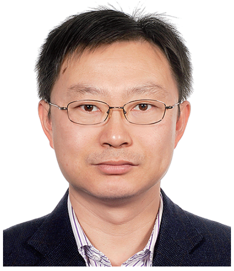
Authors' Short Biographies



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