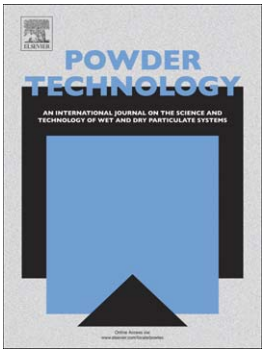
Uniformly dispersion of carbon nanotube in aluminum powders by wet shake-mixing approach

Peng, Tao; Chang, Isaac

DOI:
10.1016/j.powtec.2015.06.039

Document Version
Peer reviewed version

Citation for published version (Harvard):
Peng, T & Chang, I 2015, 'Uniformly dispersion of carbon nanotube in aluminum powders by wet shake-mixing approach', *Powder Technology*, vol. 284, pp. 32-39. https://doi.org/10.1016/j.powtec.2015.06.039



Uniformly Dispersion of Carbon Nanotube in Aluminum Powders by Wet Shake-mixing Approach

Tao Peng, Isaac Chang

PII: S0032-5910(15)00497-0
DOI: doi: 10.1016/j.powtec.2015.06.039
Reference: PTEC 11082

To appear in: *Powder Technology*

Received date: 16 April 2015
Revised date: 26 May 2015
Accepted date: 13 June 2015

Please cite this article as: Tao Peng, Isaac Chang, Uniformly Dispersion of Carbon Nanotube in Aluminum Powders by Wet Shake-mixing Approach, *Powder Technology* (2015), doi: 10.1016/j.powtec.2015.06.039

Uniformly Dispersion of Carbon Nanotube in Aluminum Powders by Wet Shake-mixing Approach

Tao Peng*, Isaac Chang

School of Metallurgy and Materials, University of Birmingham, Edgbaston, Birmingham B15 2TT, UK

A B S T R A C T

Since its discovery, carbon nanotube(CNT) was proposed as an ideal reinforcement material for metal matrix composite for its high strength, excellent electrical and thermal conductivity. CNT reinforced aluminum matrix composite has attracted most attention at the beginning of 21st century due to the need for advanced lightweight alloys for aerospace, automotive and defense industries. However, few researchers have successfully incorporated pristine and undamaged CNT into matrix to enhance the properties of the composite. Both traditional and novel powder metallurgical routes have been explored, nevertheless, challenges like the poor distribution of CNT in Al matrix, the agglomeration of CNTs and the damage of essential CNT tubular structure impeded the full translation of CNT potential into various matrix. To achieve a uniform dispersion of CNT without damaging the CNT structure, the authors have applied a novel wet shake-mixing method which combined the advantages of ultrasonication, turbular mixing and ball milling to fabricate an homogenous Al–0.5 wt. % multi-walled

* Corresponding author.
Tel: +441214145167. *E-mail address*: ptao.mail@gmail.com (T. Peng)

carbon nanotube(MWNT) composite. The original structure and morphology of MWNTs and aluminum powders were well preserved even after all the processing procedures in the as-produced powders. This is confirmed by scanning electron microscopy and X-ray diffraction analysis, particle size distribution and the Raman spectra of the as-produced composite powders.

Keywords:
Composite materials; Metal matrix composites; Raman Spectra; Dispersion; Mixing

1. Introduction

Composite materials consist of a bulk matrix material and one or more reinforcement phases, combining the desired properties of different constituents, which significantly break the performance limitation of traditional monolithic materials systems and remarkably expand the horizon of signal transmission, biomedicine, aerospace and automobile industries[1]. For these industries, aluminum(Al) is the most widely applied metal due to its abundance, low price, good corrosion resistance and, more importantly, low density, which leads to a combination of good specific strength and light-weighted structure for increased fuel efficiency[2]. There is an increasing demand for Al-based systems with higher specific strength and specific modulus properties to cater for the development of modern aircrafts and vehicles that can be operated at higher speed, temperature and longer distance before maintenance[3]. Carbon nanotube (CNT), by its virtue of extraordinary mechanical properties, thermal stabilities and excellent electrical conductivity [3, 4] was proposed as a promising

reinforcement for aluminum matrix composite for both structural and functional applications. It is reported that multi-walled carbon nanotube(MWNT) possessed the highest specific strength(48000 kN·m/kg) of all materials, exhibiting five times Young's modules (~1TP) and up to one hundred times tensile strength (~150GPa) than the best known steel of the same weight [5]. Also, the superb electrical conductivity[6] of carrying an electric current density of 4×10^9 A/cm^2, which is 1000 times greater than that of copper, makes CNT an ideal material for electrical and signal transmission. Moreover, the excellent chemical[7] and thermal stability (stable up to 2527K in vacuum) of CNT contribute to its prospect application in extreme conditions

Thus, a number of researchers[8-14] had tried to develop various methods of incorporating CNTs into Al matrix to increase specific strength, stiffness, thermal and electrical conductivities[15]. Liao and Tan[16] attempted low energy ball milling to disperse 0.5 wt.% CNT in Al matrix. After continuously milling the mixture for 4h at a speed of 200 rpm (agate ball to powder weight ratio 5:1), micro CNT clusters can still be found among the Al powders. These CNT aggregates are resulted from the strong van der Waals force along the long and thin tube in which the length-to-diameter ratio of CNT is up to 1.32×10^8 : 1[17]. Obviously, in order to achieve the full potential of reinforcements, CNTs need to be uniformly distributed in Al matrix. Otherwise, the existence of agglomerates will lead to lower density, more voids in the bulk materials and finally deteriorate the overall properties as the wetting angel between CNT and aluminium is very big that CNT agglomerates would impede the diffusion between aluminium particles and leave more pores in the bulk composite thus decrease the density. Liu and

colleagues[18] utilized high energy ball milling to blend Al-0.5 wt. % CNT at a speed of 300 rpm (ball to powder weight ratio 8:1) for 8-12h. They did not observe any CNT bundles in the matrix but the existing visible individual CNT was seriously shortened or damaged. It is clear that the seamless cylinder shape of CNT is vital for keeping its exceptional properties. No doubt that losing the structure means losing its strength and stability. Furthermore, the morphology of Al particle plays an important role in the dispersion of CNT and densification process. For example, the anchoring of CNT on to Al metal powder requires a particle morphology with a large surface area according to Jiang's research[19]. Whereas coarse and irregular shape particles have poor compaction and sintering ability which subsequently result in severe porosity, low density and weak bonding.

Apparently, it is critical to obtain a homogeneous mixture of CNT and Al with the structure of individual constituents intact before the production of bulk CNT reinforced Al composite for functional and structural applications [20]. Although high energy mechanical alloying is an effective way to disperse CNT in the metallic matrix, the damage of CNT after processing limits the overall properties of composite to a certain level. Hence, in the current work, a novel approach has been explored to fabricate an Al-0.5 wt. % MWNT composite that can preserve the CNT and Al particles in their original state, which is significantly beneficial to the subsequent powder compaction, consolidation and overall properties of the composite. Scanning electron microscopy (SEM), X-ray diffraction (XRD), particle size analysis and Raman spectroscopy were utilized to track the micro-structural evolution of CNT and Al constituents and validate the uniform distribution and structural retention of CNT in the

as-produced composite.

2. Experimental

2.1. Materials

An argon gas atomized, spherical shape aluminum powder, with a size range of up to 10 um produced by the Aluminum Powder Company Ltd, Alpoco was selected as the matrix constituent in order to increase the surface area to attach more MWNT in the aluminium matrix. Multi-walled carbon nanotube (MWNT), 140 ± 30 nm in diameter and 7 ± 2 um in length synthesized by chemical vapor deposition (CVD) method, were purchased from Materials and Electrochemical Research Corporation (M.E.R. Corporation) as the reinforcements in the composite.

2.2. Materials Processing

It is critical to employ an appropriate mixing technique to obtain homogeneous dispersion of reinforcements in the composite. In the current work, the authors combined ultra-sonication, magnetic stirring and shake-mixing to manufacture an Al-0.5 wt. % MWNT composite, in which MWNT s are evenly distributed in the aluminum matrix. The detailed fabrication procedures are depicted in Fig. 1. Firstly, 0.5 g of MWNT was added into 150 mL of ethanol in a 1L glass beaker, which was then put into an ultrasonic bath (James Products Limited, model: Sonic 4500). The solution was sonicated for 4 h by a Colour Direct ultrasonic machine (model: CD-M06 031) to separate pristine CNT bundles into individual tubes by high frequency agitation(power: 180 watt, frequency: 42,000Hz). Meanwhile, 0.5 g of polyvinyl

butyral (PVB) was dissolved in 150mL of ethanol and then 99.5 g of Al powders were added into the as-produced transparent PVB-ethanol slurry. After magnetic stirred the Al-PVB-ethanol mixture for 2 h at a speed of 400 rpm, ideally the surface of Al particles was evenly covered by a thin layer of PVB molecules, which reduced the surface tension of Al and help to absorb MWNT on Al particles[22].

Fig. 1. Schematic illustration of the processing procedures for MWNT/Al composites.

Afterwards, the as-prepared 150 ml PVB coated Al-ethanol suspension was poured into the 150 ml MWNT-ethanol solution for further magnetic stirring for another 4 h at a speed of 500 rpm to homogenously disperse MWNTs in Al matrix. In case of any MWNTs agglomerating together in the stirring process, the as-produced Al-0.5 wt.% MWNT slurry mixture together with 15 of stainless steel ball bearings (diameter: 10 mm, ball to powder ratio: 3:5) was transferred into a plastic bottle. A TURBULA Shake-Mixer (Model: T2F) was used to mix this mixture for 10 min at a speed of 101 rpm. As the shake-mixing is more powerful than simple magnetic stirring but less violent than high energy ball milling, it promotes the uniformity of the composite mixture while keeping the MWNT and Al particles in the original state. Eventually, the as-blended slurry mixture was dried at 140 °C for 12h and shake-mixed for another 10 minutes to break down the powder lumps into uniform composite powders. Subsequently, the MWNT/Al composite powders were compacted into cylinders at 475 MPa and were sintered at 630°C for 1h in argon atmosphere.

2.3. Materials Characterization

A Philips XL-30 and a JEOL 7000 field emission scanning electron microscope (FESEM) were employed to track the size, shape and morphology of Al and MWNT during the processing as well as the distribution of MWNT in Al composite. X-ray diffractometer (Inel EQUINOX 3000) and Raman spectroscopy (Renishaw inVia confocal Raman microscope) were employed to characterize the crystal structure and bonding nature of the constituents respectively. The particle size distribution of MWNT reinforced Al matrix was assessed HELOS/KR- VIBRI/L particle size analyzer.

---

3. Result and discussion

3.1. Materials

Fig. 2. (a) SEM image of raw MWNT clusters; (b) high magnification SEM image of pristine MWNTs and (c). high resolution TEM image of MWNT[1].

The morphology of pristine multi-walled carbon nanotube is showed in the secondary electron images Fig. 2(a) and Fig. 2(b). It can be seen from Fig. 2(a) that the MWNTs are randomly woven together to give ball-shaped bundles which have diameters of around 25 um. These clusters are the biggest challenges of manufacturing effective MWNT-Al composites that many researchers found MWNTs agglomerates[7, 16, 23-25] of different sizes in the metal matrix after various of processing including hot extrusion[7], mechanical alloying[11], hot

[1] The TEM image of MWNT is provided by Materials and Electrochemical Research Corporation.

pressing[15], plasma spray[26] and hot rolling[23]. The higher magnification image of MWNTs is showed in Fig. 2(b). It is obvious that MWNTs were in straight and long tubular shape which has fewer tendencies to agglomerate than curved and entangled MWNTs according to Esawi and his colleagues research[8]. A high resolution transmission electron microscope (HRTEM) image of MWNT showing the multi-layer microstructure is presented in Fig. 2(c).

Fig. 3. (a) SEM micrograph of raw aluminum powders; (b) SEM micrograph of an individual aluminum particle under higher magnification.

Fig. 3(a) and Fig. 3(b) illustrate the size, shape and topography of the pristine aluminum powders. It is evident from Fig. 3(a) that the sizes of raw aluminum particles are under 10 um and the shapes of these particles are irregular which is good for the anchoring of MWNTs onto the Al powder surface. Also it can be seen from Fig. 3(b) that the pristine Al powders have smooth surfaces which is beneficial for compaction due to the smoother surface has a lower surface friction coefficient that ease the friction between particles as well as the friction between particles and die walls. This will consequently lead to the increase of density, reduce of porosity and finally improve the overall properties of the composite[27].

3.2. Mixtures

Fig. 4(a) and Fig. 4(b) show the SEM image of the 4h-ultrasonicated MWNT and 2h-magnetic-stirred Al-0.5 wt. % PVB-0.5 wt. % MWNT mixture respectively. As can be seen from Fig. 4(a), after being ultra-sonicated in ethanol for 4h, MWNTs were almost

disassembled but due to their own electrical charges MWNT have a strong tendency to agglomerate together and some small nodes were formed . The individual MWNT, having no distinct structure damage, is significantly beneficial to the following processing with Al powders and achieving better properties in the final composite. Some researchers failed to disperse MWNT agglomerates in metal matrix because the van der Waals force between the long and thin tubes is too strong to be separated. Besides, the mobility of MWNTs in matrix is decreased by relatively large Al particles which block the free moving paths of MWNTs and became an obstacle to the disassembling and uniform dispersion of MWNT. In contrast, the authors of this paper utilized the small aluminium particles which had relatively less resistance to the unfastening and unlocking of MWNT bundles. As can be seen in Fig. 4(b), some small MWNT clusters were found in Al matrix of the 2h-magnetic-stirred MWNT and PVB-coated Al powders. This is because the suspended individual MWNT was bound by the PVB in the solution thus the small MWNT agglomerates were formed. To solve this problem, further action was taken to unlock and disperse the MWNT bundles.

Fig. 4. (a) SEM image of 4 h-ultrasonicated MWNT; (b) SEM image of 2 h-magnetic stirred Al-0.5 wt.% PVB-0.5 wt.% MWNT mixture; (c) SEM image of 4h magnetic stirred Al-0.5 wt.% PVB-0.5 wt.% MWNT mixture; (d) SEM image of 30 min-shake-mixed Al-0.5 wt.% PVB-0.5 wt.% MWNT mixture.

Fig. 4(c) and Fig. 4(d) show the 4h-magnetic stirred and further 30min-shake-mixed composite powders, separately. As shown in Fig. 4(c), even after 4h magnetic stirring, MWNT agglomerates were still remained but in smaller proportion and still visible in the Al matrix. This suggests the magnetic stirring is not efficient in completely de-agglomerating the

MWNT bundles. To thoroughly resolve the problem of MWNT clusters, the mixture along with 15 stainless ball bearings was transferred to a TURBULA shake-mixer for 30min blending and the as-blended powders is showed in Fig. 4(d). It is apparent that no MWNT agglomerates were found in the Al matrix, which indicates the homogenous distribution of MWNTs. Furthermore, the long tubular MWNTs were visibly intact and the smooth surfaces of Al particles were retained without any change of the particle size, which is good for the following compacting, sintering and good final mechanical properties.

Fig. 5. (a) SEM image of dried Al-0.5 wt.% PVB-0.5 wt.% MWNT mixture; (b) SEM micrograph of dried Al-0.5 wt.% PVB-0.5 wt.% MWNT mixture under higher magnification; (c) SEM micrograph of the as-produced Al-0.5 wt.% MWNT composite powders.

Fig. 5(a) and Fig. 5(b) show the morphology of the dried Al-0.5 wt. % PVB-0.5 wt. % MWNT mixture. It is distinctly seen that micro-sized (around 500um in Fig. 5. (a)) aluminum powders were agglomerated under the binding impact of PVB and the nature of fine powders. The as-blended mixture formed irregular shaped granules(or secondary particles[28]) in Fig. 5(b) are detrimental to die-filling, compaction and pore-filling process during sintering. To solve this problem, the dried composite mixture with 15 stainless steel ball bearings was blended again in a dry argon atmosphere on the TURBULAR shake-mixer for 10 min. The final as-blended composite powders were displayed in Fig. 5(c). It is clear that MWNTs(as indicated by white arrows), of the same shape and length as pristine MWNTs, were individually dispersed among undeformed Al powders. This demonstrated that MWNT and Al kept their original nature (size, shape and morphology) in the as-produced Al-0.5 wt. %

MWNT composite powders. What's also worth noticing is that no welded Al particles were found in the matrix which is a serious a topic for Al powders when high energy ball milling is employed. In the authors' previous work[29], high energy mechanical alloying was used to disperse MWNT in pure Al matrix for various time length. The 1h-ball-milled and 5h-ball-milled Al-1 wt. % MWNT were severely flattened and welded, the particle size increased 10 times and the flake shape powders is useless for industries production due to its poor flowability, compactability and sintering ability.

Fig. 6. Particle size distribution of the pristine aluminum powders and the as-produced Al-0.5 wt. % MWNT powders. (a) and (c) are the cumulative distribution and frequency distribution of the raw aluminium respectively; (b) and (d) are the cumulative distribution and frequency distribution of the as-produced Al-0.5 wt. % MWNT individually.

Fig. 6. presents the particle size data of the pristine Al and the as-produced Al-0.5 wt.% MWNT composite powers. In powder metallurgy industry, particle size distribution and mean particle size are two critical factors that affect the structure and the final properties of products. As can be seen that the size of as-produced composite particles (Fig 6.b) increased as compared to the as-supplied Al powders(Fig. 6a). This is due to relative low stiffness and strength of pure aluminium powders and the plastic deformation induced by the collision impact of ball bearing in the TURBULAR shake-mixer flattened the aluminium particles. The binding effect of PVB may also contribute to the increase of particle size.

According to F. Thümmler and R. Oberacker[28], when the cumulative distribution of powders reaches 50%, the corresponding median particle size Z_{50} is defined as the particle size of the sample. The line graph Figs. 6. (c) and 6. (d) show the cumulative distribution of raw aluminium and as-produced Al-0.5 wt.% powders respectively. It is obvious that after processing, the median particles size[27] Z_{50} for as-produced composite powders reached 15.10um, as compared to that of pristine Aluminum(9.92um). This proves the fine particle size of aluminium is preserved after the processing but slightly shifted to larger size.The increase of the particle size is because aluminium particles are very light and soft, under the collision of relatively heavier stainless steel ball bearings, the aluminium particles are prone to deform. After certain period of milling, the Al particles are deformed and flattened, in which the dimension increase is along one direction and this of the other direction is relatively reduced.

Fig. 7. XRD spectra of pristine aluminum powder, raw MWNT and as-produced Al-0.5 wt. % MWNT powder.

Fig. 7 shows the XRD spectra of pristine aluminum powders, raw MWNT and as-produced Al-0.5 wt. % MWNT powder. As can be seen from the XRD spectra (Fig. 7) that multi-walled carbon nanotube showed a strong characteristic peak at around 26° while the as-produced Al-0.5 wt. % MWNT powders showed a similar XRD peaks to that of the pure Al powder. It is worth noticing that no carbon peak was found in the as-produced Al-MWNT composite powder which may due to the limited detective range of XRD and the small amount (0.5

wt. %) of MWNT in the composite. Also, XRD peaks of aluminium(marked by black triangles in Fig. 7) were picked in the pure MWNT group which is due to the low weight of carbon atom that X-ray penetrated the MWNT layer and hit the aluminium sample holder thus released some diffraction signal of aluminum.

The grain size of powders was measured by Williamson-Hall equation[30, 31]:

$$\beta * \cos\theta = \frac{K\lambda}{D} + 4\varepsilon * \sin\theta \quad (1)$$

where K is the shape factor, λ is the wavelength of Cu Kα radiation, β is the full width at half maximum (FWHM) , θ is the diffraction angle and ε is the strain. The calculated grain size of the raw Al and as-produced composite powder is 1.26um and 1.30um respectively. This suggests the grain size did not change significantly which is beneficial for achieving relatively high strength and stiffness in the bulk materials.

3.3. Compacted composite

The compressiblity of powders is a critical parameter for large scale industrial manufacture. Generally, powders with good compressiblity yields products with good density and requires less processing after compaction which saves the cost of production as well as shorten the manufacture cycle. In the current paper, both raw aluminium and as-produced composite powders was cold compacted by a 30 tones hydraulic press. The green density(measured by Achimedes method), which is the density of the green compact or preform, and the relative density were shown in Table 1. As can be seen from Table 1 that despite the change of mean particle size the density of Al-0.5 wt. % composite powders, comparing to that of pristine

aluminium powders, did not deteriorate after processing and addition of MWNT. In contrast, the green density and relative density of the compacted as-produced composite slightly increased due to the lubrication effect of PVB.

Table 1 The density and Raman shift ratio of pure aluminum and MWNT/Al composite

3.4. Sintered composite

Fig. 8. (a) SEM image of MWNTs homogenously dispersed in the sintered Al composite; (b) SEM image of individual MWNT in the sintered Al composite under high magnification.

Fig. 8(a) shows the SEM image of MWNTs (marked by white arrows) found in the compacted and sintered Al-0.5 wt. % MWNT matrix. As can be seen, MWNTs were evenly distributed in the Al matrix after mixing, compaction and even sintering. According to other researchers work, if the cylinder structure of CNT is damaged or CNT is deformed into amorphous carbon they are vulnerable to react with aluminum forming Al_4C_3 and subsequently no CNT can be observed. Evidently in Fig. 8(a), several MWNTs were uniformly dispersed in the metal matrix which proves both the homogeneity and the undamaged, pristine structure of MWNT. More direct evidence of the retention of CNT structure after processing is shown in Fig. 8(b) in which an individual MWNT, of the same diameter as the pristine MWNTs, was found in the sintered Al matrix after grinding and polishing. This proves that MWNT survived the

processing.

Fig. 9. Raman spectra of the pristine MWNTs and the as-produced Al-0.5 wt. % MWNT composite powder.

The Raman spectra of raw MWNTs and as-produced Al-0.5 wt. % MWNT composite powders are shown in Fig. 9. Apparently, there are two strong peaks in the spectra namely D band and G band. The merge of peak at around 1335 cm^{-1} is due to the vibration of carbon atoms with disordered structure (vacancies, dopants, defects or amorphous carbon) which is called D band[32], while G band at 1570 cm^{-1} represent the vibration of graphite carbon atoms [33]. Hence, the increase of structure damage will lead to the intensity increase of D band. From Fig. 9, the Raman spectrum profile of the as-produced Al-0.5 wt. % MWNT powders showed almost the same shape as that of the pristine MWNT which proves MWNTs preserved the micro tubular structure. Furthermore, the structure change of MWNT can be quantitative analyzed with the intensity ratio: I_D/I_G where I_D and I_G represent the peak intensity of D band and G band respectively. If the defects in MWNT increases, the peak intensity of D band will increase, therefore the I_D/I_G increases. The calculated I_D/I_G of raw aluminium powders is 0.34 while this of the as-produced composite powders is 0.38, which is much better than high energy ball milling that increase the I_D/I_G from 1.17 to 1.30 and 1.61 after 4h and 8h milling[18] respectively. Raman spectra indicated no additional defects were introduced by the processing method based on similar I_D/I_G ratio before and after mixing

which meansthe MWNT structure is preserved after processing as ultra-sonication, magnetic stirring and shake-mixing did not introduce direct impact of high energy into MWNT or aluminium matrix. The slightly increase of I_D/I_G may be due to the collision of ball bearings in the shake-mixer that moderately damage some MWNTs. Also, the reason for the broadening of the Raman peak is due to the attachment of PVB molecules on the surface of both MWNT and aluminium particles. The bonds in the PVB especially carbon-carbon bond would produce some signals and make some noise for the Raman spectra. In addition, the polymerization degree and the length of each PVB molecule is different which leads to the difference of scattering level. The detailed enhancing effect of MWNT in the aluminium matrix is currently under investigation by the authors.

3.5. Processing Comparison

Attracted by CNT's extraordinary properties, a wide range of techniques were attempted by researchers to incorporate different percentage of CNTs into metal matrix. Table 2 lists several typical experiments of previous studies as well as the processing results of present author. J.Z. Liao et al.[16] tested the low energy ball milling of 0.5 wt. % CNT with Al powders for 4h but CNT agglomerates were clearly observed in the Al matrix, which is due to the lack of sufficient energy to untie CNT bundles. A. M. K Esawi et al.[23] added 0.5 wt. % CNT into aluminium powders and blended by no milling media mixer-shaking at 46rpm and no milling media planetary milling at 300 rpm. However, CNT agglomerates were still found in the as-produced powders, which is because no milling media was added leading to the absence of collision and shear force to impact the CNTs. R. Pérez-Bustamante et al.[12] and Z.Y. Liu et

al. [18] both chose high energy ball milling with ball bearings as milling media to disperse 0.5 wt. % CNT into aluminium matrix for long periods of time(5-30h and 8-12h respectively). Although, CNTs were found homogenously distributed in the matrix, the secondary electron images in Pérez-Bustamante's study showed some CNTs were shortened and uncapped while Liu calculated and noticed the I_D/I_G ratio of CNT Rama spectra was increased after ball milling which is also an indication of CNT structure damage as explained before.

To thoroughly solve this problem, the present authors combined the advantages of ultra-sonication, magnetic stirring and mixer-shaking, using PVB to bind the separated MWNT on to aluminum particles and further homogenized by the medium energy striking of ball bearing. 0.5 wt. % MWNT was successfully dispersed into fine aluminum powder matrix without modifying the structure and morphology of MWNT and aluminum. It is worth noticing that T. Kuzumaki et al.[7] stirred 3.0 and 6.0 vol. % in ethanol at 300rpm for 0.5h and observed some CNT bundles. This is due to the lack of binder which adhere CNT to aluminum surface and reduces the surface tension between CNT and aluminum particle. Thus the dispersed CNT tended to agglomerate together once the stirring stopped. In the study of J.Z. Liao et al. [16], although binder(PEG) was employed, the CNT agglomerates was not dissembled before mixing with PEG and Al, which leading to the attachment of CNT bundles instead of individual CNT to aluminum surface. Also, it should be aware that according to J.Z. Liao et al.[24]'s another experiment and other researchers' experience, high loading of CNT leaded to the formation of agglomerates and impeded densification process. The uniform distribution of high concentration of MWNT into metal matrix is necessary to be explored in

the future.

Table 2 Various experiment methods and corresponding results in the present paper and other literature

4. Conclusions

0.5 wt. % multi-walled carbon nanotubes reinforced Al composite powders were successfully fabricated by a novel wet shake-mixing technique without introducing significant deformation or defects to the MWNT. SEM analysis showed that, in the as-produced composite powder, the Al particles retain its fine size and smooth surface while MWNTs were evenly dispersed in the Al powders. Also, the as-produced Al – 0.5wt.% MWNT composite powder kept the same compressibility as pure aluminium powders. More importantly, intact individual MWNT is observed in the sintered bulk Al – 0.5wt.% MWNT composite and the I_D/I_G ratio of Raman spectra of MWNT kept constant which indicated that MWNT preserved the cylinder structure which is essential for the improvement of overall properties of the composite.

Acknowledgements

The authors acknowledge the financial support from the University of Birmingham and China Scholarship Council. Tao Peng would also like to thank the assistance from Dr. Constantin Lucian Falticeanu, Dr. Su Sei, Dr. Nicolas Harrison and Mr. Andrew Ward.

REFERENCES

[1] A.B. Agarwal, S. R. Lahiri, D., Carbon Nanotubes Reinforced Metal Matrix Composites, CRC Press, 2010.

[2] S.R. Bakshi, A.K. Keshri, V. Singh, S. Seal, A. Agarwal, Interface in carbon nanotube reinforced aluminum silicon composites: Thermodynamic analysis and experimental verification, J Alloy Compd, 481 (2009) 207-213.

[3] C.R. Bradbury, J.K. Gomon, L. Kollo, H. Kwon, M. Leparoux, Hardness of Multi Wall Carbon Nanotubes reinforced aluminium matrix composites, J Alloy Compd, 585 (2014) 362-367.

[4] R. Perez-Bustamante, I. Estrada-Guel, P. Amezaga-Madrid, M. Miki-Yoshida, J.M. Herrera-Ramirez, R. Martinez-Sanchez, Microstructural characterization of Al-MWCNT composites produced by mechanical milling and hot extrusion, J Alloy Compd, 495 (2010) 399-402.

[5] E.T. Thostenson, Z.F. Ren, T.W. Chou, Advances in the science and technology of carbon nanotubes and their composites: a review, Compos Sci Technol, 61 (2001) 1899-1912.

[6] S. Hong, S. Myung, Nanotube electronics - A flexible approach to mobility, Nat Nanotechnol, 2 (2007) 207-208.

[7] T. Kuzumaki, K. Miyazawa, H. Ichinose, K. Ito, Processing of carbon nanotube reinforced aluminum composite, J Mater Res, 13 (1998) 2445-2449.

[8] A.M.K. Esawi, K. Morsi, A. Sayed, M. Taher, S. Lanka, The influence of carbon nanotube (CNT) morphology and diameter on the processing and properties of CNT-reinforced aluminium composites, Compos Part a-Appl S, 42 (2011) 234-243.

[9] K. Morsi, A.M.K. Esawi, S. Lanka, A. Sayed, M. Taher, Spark plasma extrusion (SPE) of ball-milled aluminum and carbon nanotube reinforced aluminum composite powders, Compos Part a-Appl S, 41 (2010) 322-326.

[10] D. Poirier, R. Gauvin, R.A.L. Drew, Structural characterization of a mechanically milled carbon nanotube/aluminum mixture, Compos Part a-Appl S, 40 (2009) 1482-1489.

[11] A. Esawi, K. Morsi, Dispersion of carbon nanotubes (CNTs) in aluminum powder, Compos Part a-Appl S, 38 (2007) 646-650.

[12] R. Perez-Bustamante, F. Perez-Bustamante, I. Estrada-Guel, C.R. Santillan-Rodriguez, J.A. Matutes-Aquino, J.M. Herrera-Ramirez, M. Miki-Yoshida, R. Martinez-Sanchez, Characterization of Al-2024-CNTs composites produced by mechanical alloying, Powder Technol, 212 (2011) 390-396.

[13] D. Jeyasimman, K. Sivaprasad, S. Sivasankaran, R. Narayanasamy, Fabrication and consolidation behavior of Al 6061 nanocomposite powders reinforced by multi-walled carbon nanotubes, Powder Technol, 258 (2014) 189-197.

[14] S.K. Singhal, R. Pasricha, M. Jangra, R. Chahal, S. Teotia, R.B. Mathur, Carbon nanotubes: Amino functionalization and its application in the fabrication of Al-matrix composites, Powder Technol, 215-16 (2012) 254-263.

[15] C.L. Xu, B.Q. Wei, R.Z. Ma, J. Liang, X.K. Ma, D.H. Wu, Fabrication of aluminum-carbon nanotube composites and their electrical properties, Carbon, 37 (1999) 855-858.

[16] J.Z. Liao, M.J. Tan, Mixing of carbon nanotubes (CNTs) and aluminum powder for powder metallurgy use, Powder Technol, 208 (2011) 42-48.

[17] X.S. Wang, Q.Q. Li, J. Xie, Z. Jin, J.Y. Wang, Y. Li, K.L. Jiang, S.S. Fan, Fabrication of Ultralong and Electrically Uniform Single-Walled Carbon Nanotubes on Clean Substrates, Nano Lett, 9 (2009) 3137-3141.

[18] Z.Y. Liu, S.J. Xu, B.L. Xiao, P. Xue, W.G. Wang, Z.Y. Ma, Effect of ball-milling time on mechanical properties of carbon nanotubes reinforced aluminum matrix composites, Compos Part a-Appl S, 43 (2012) 2161-2168.

[19] L. Jiang, Z.Q. Li, G.L. Fan, L.L. Cao, D. Zhang, Strong and ductile carbon nanotube/aluminum bulk nanolaminated composites with two-dimensional alignment of carbon nanotubes, Scripta Mater, 66 (2012) 331-334.

[20] X.D. Yang, E.Z. Liu, C.S. Shi, C.N. He, J.J. Li, N.Q. Zhao, K. Kondoh, Fabrication of carbon nanotube reinforced Al composites with well-balanced strength and ductility, J Alloy Compd, 563 (2013) 216-220.
[21] Z.Q. Li, L. Jiang, G.L. Fan, Y. Xu, D. Zhang, Z.X. Chen, S. Humphries, High volume fraction and uniform dispersion of carbon nanotubes in aluminium powders, Micro Nano Lett, 5 (2010) 379-381.
[22] L. Jiang, G.L. Fan, Z.Q. Li, X.Z. Kai, D. Zhang, Z.X. Chen, S. Humphries, G. Heness, W.Y. Yeung, An approach to the uniform dispersion of a high volume fraction of carbon nanotubes in aluminum powder, Carbon, 49 (2011) 1965-1971.
[23] A.M.K. Esawi, M.A. El Borady, Carbon nanotube-reinforced aluminium strips, Compos Sci Technol, 68 (2008) 486-492.
[24] J.Z. Liao, M.J. Tan, I. Sridhar, Spark plasma sintered multi-wall carbon nanotube reinforced aluminum matrix composites, Mater Design, 31 (2010) S96-S100.
[25] M. Jafari, M.H. Abbasi, M.H. Enayati, F. Karimzadeh, Mechanical properties of nanostructured Al2024-MWCNT composite prepared by optimized mechanical milling and hot pressing methods, Adv Powder Technol, 23 (2012) 205-210.
[26] T. Laha, A. Agarwal, T. McKechnie, S. Seal, Synthesis and characterization of plasma spray formed carbon nanotube reinforced aluminum composite, Mat Sci Eng a-Struct, 381 (2004) 249-258.
[27] G.R. M., Powder Metallurgy Science, 1994.
[28] T. F., O. R., Introduction to powder metallurgy, Institute of Materials, London, 1993.
[29] T. Peng, I. Chang, Mechanical alloying of multi-walled carbon nanotubes reinforced aluminum composite powder, Powder Technol, 266 (2014) 7-15.
[30] P. Scherrer, Bestimmung der Grösse und der inneren Struktur von Kolloidteilchen mittels Röntgenstrahlen, Nachrichten von der Gesellschaft der Wissenschaften zu Göttingen, Mathematisch-Physikalische Klasse, 26 (1918).
[31] K. Ada, M. Goekgoez, M. Oenal, Y. Sankaya, Preparation and characterization of a ZnO powder with the hexagonal plate particles, Powder Technol, 181 (2008) 285-291.
[32] Jorio Ado , Gene Dresselhaus, M.S. Dresselhaus, Carbon Nanotubes: Advanced Topics in the Synthesis, Structure, Properties and Applications, Springer Science & Business Media2008.
[33] M.S. Dresselhaus, G. Dresselhaus, R. Saito, A. Jorio, Raman spectroscopy of carbon nanotubes, Physics Reports, 409 (2005) 47-99.

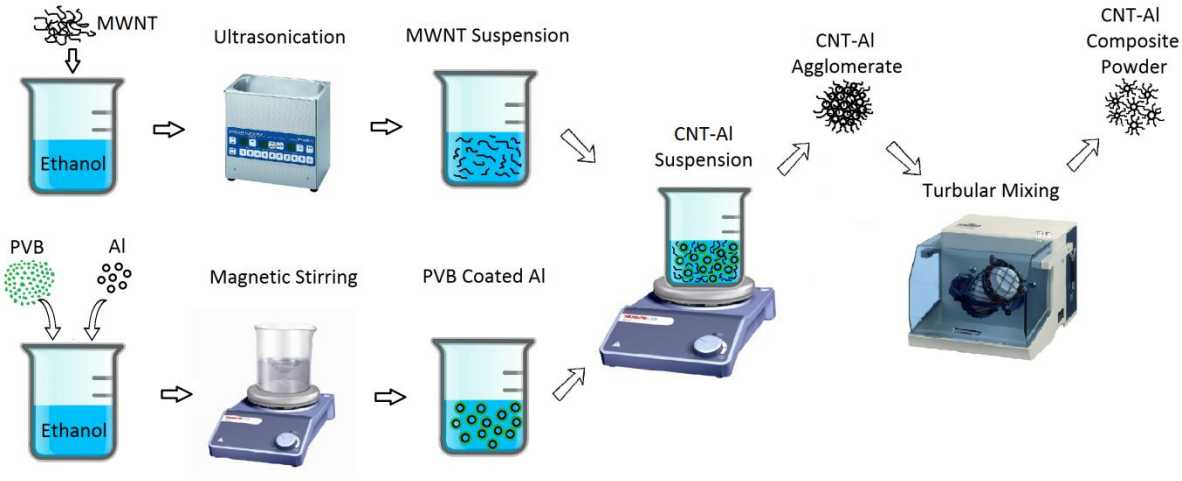


Figure 1

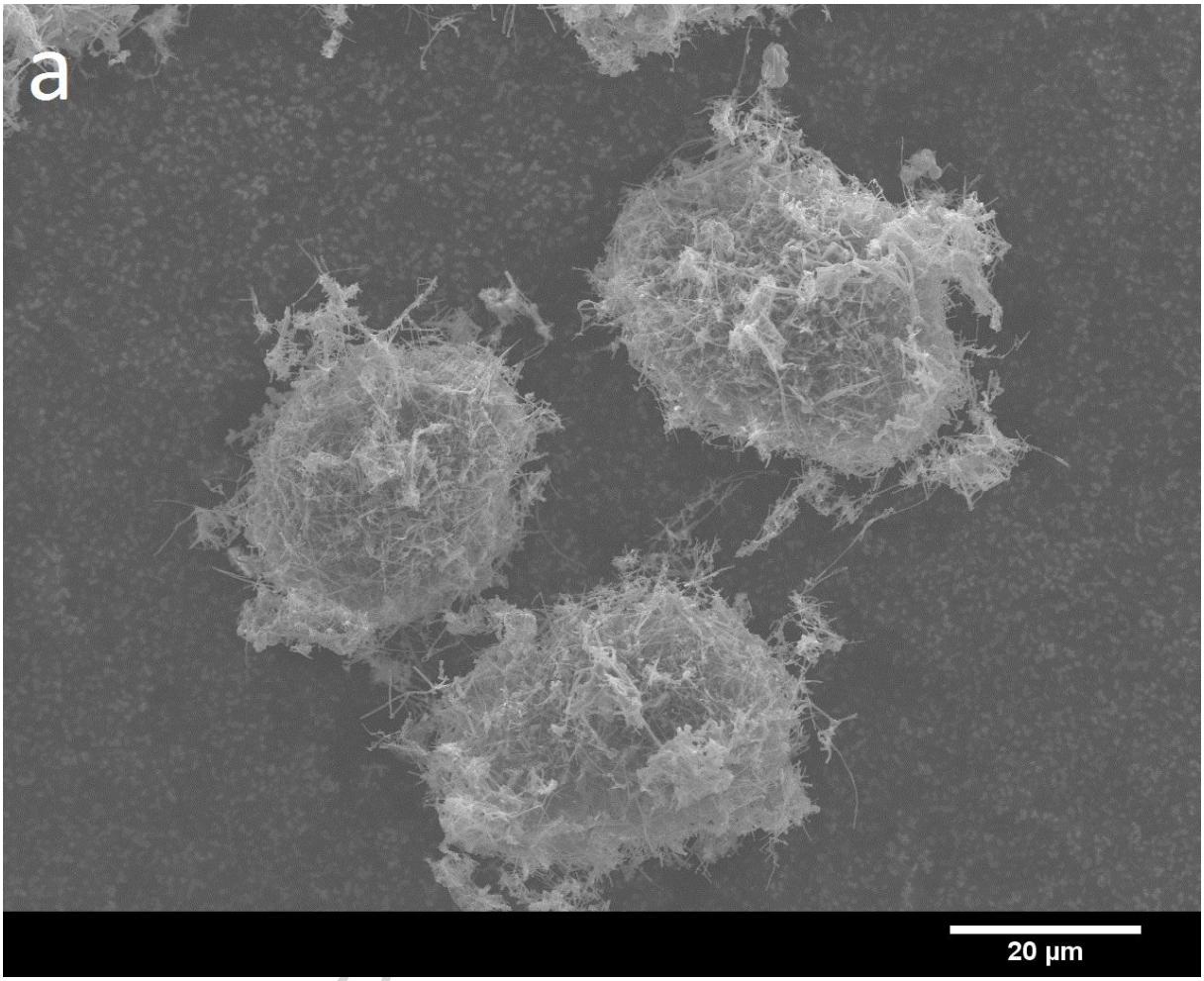


Figure 2 (a)

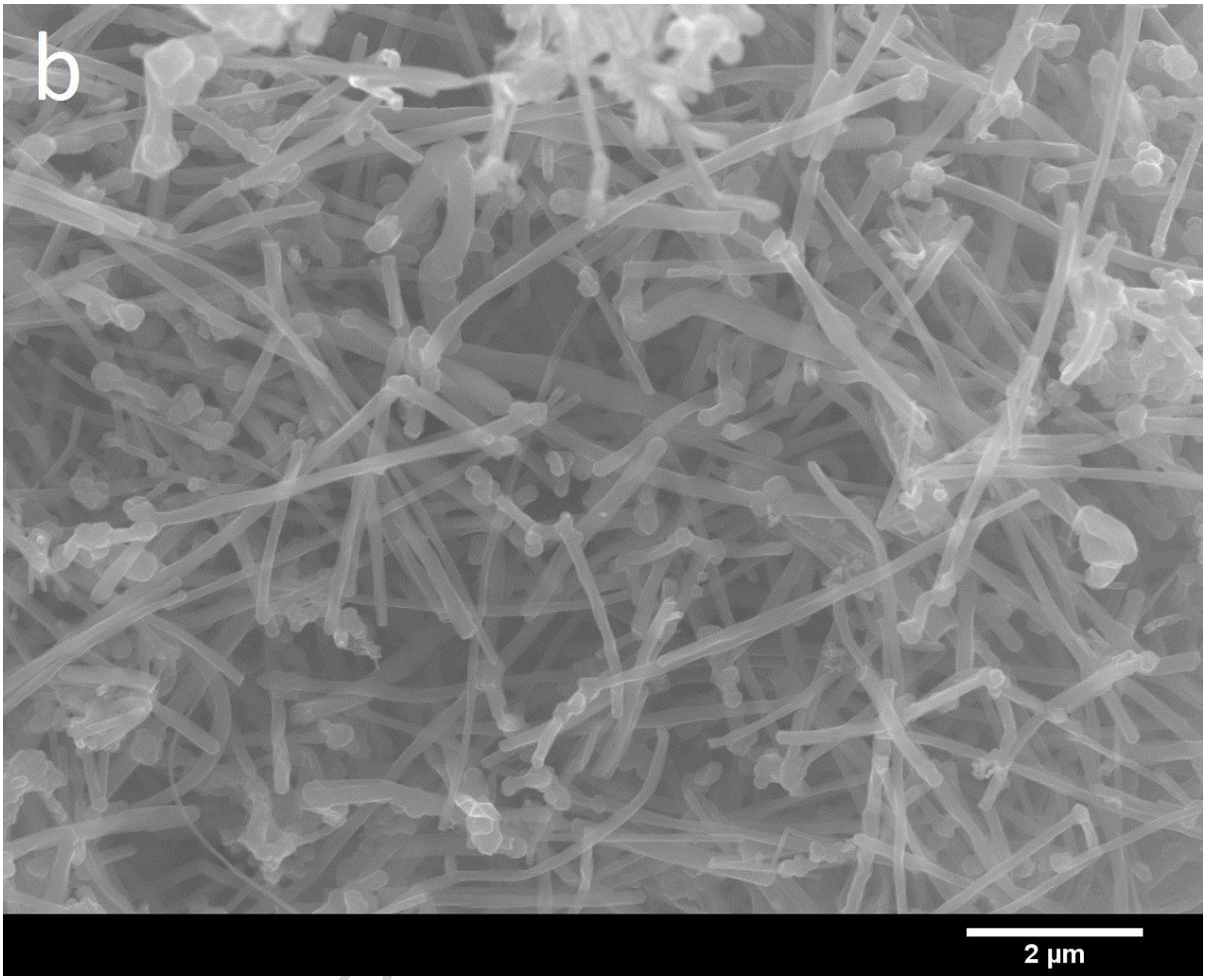


Figure 2 (b)

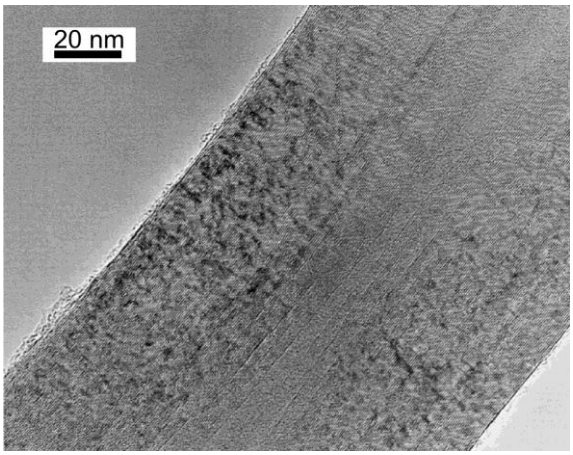


Figure 2 (c)

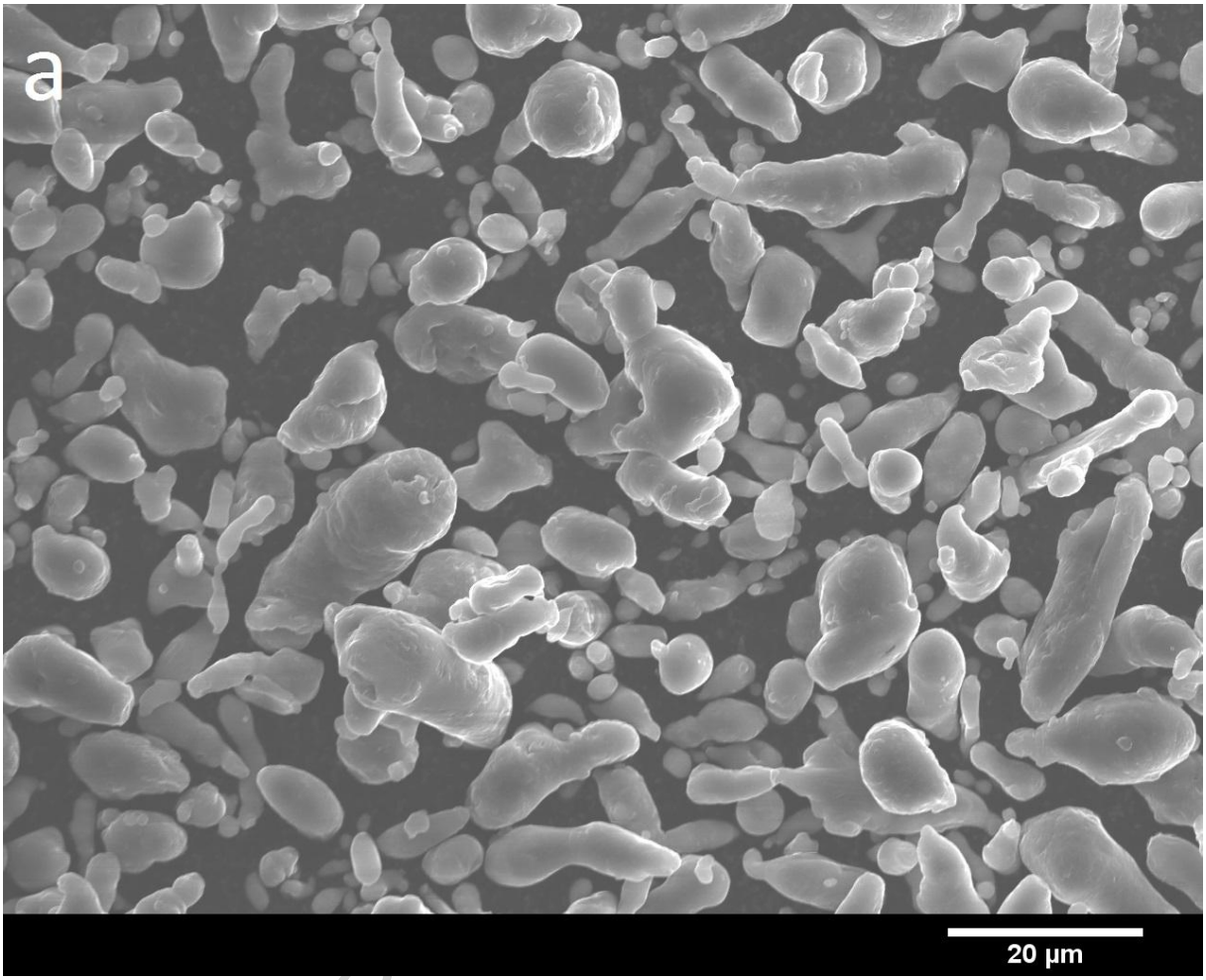


Figure 3 (a)

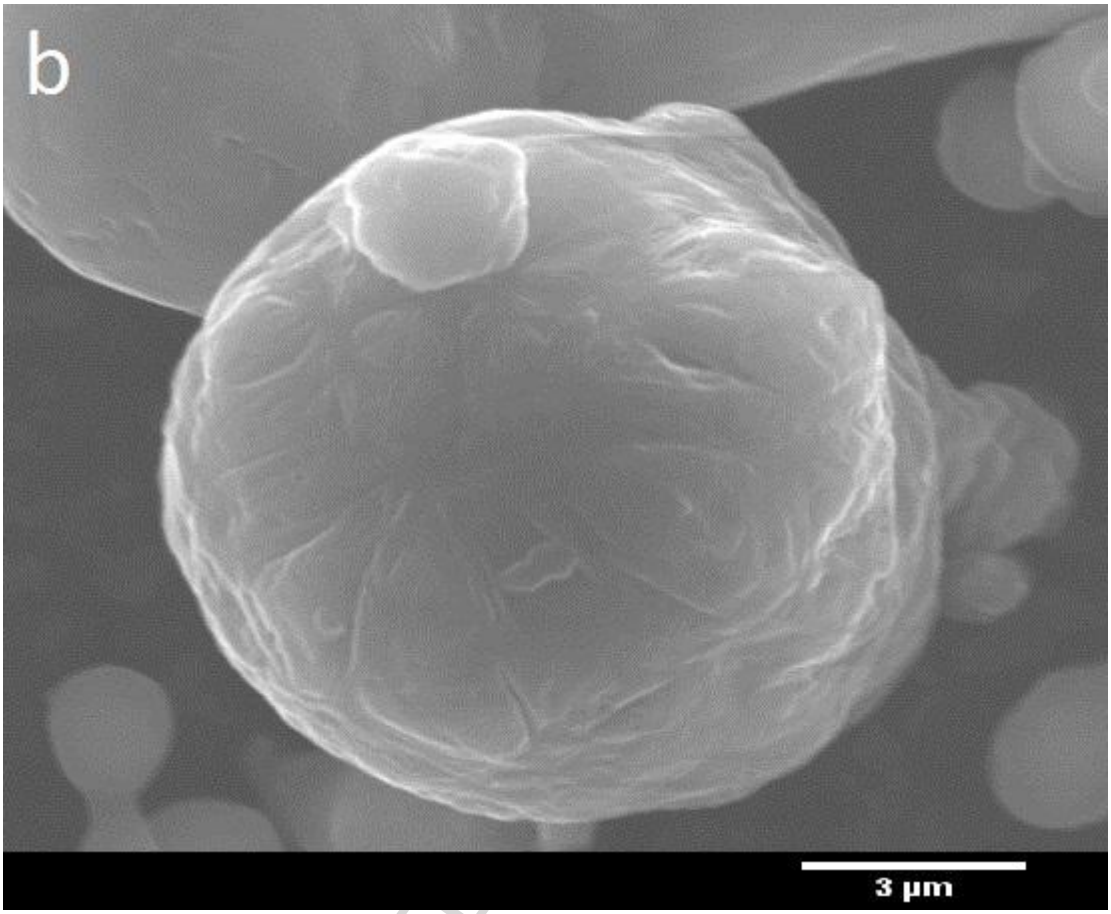


Figure 3 (b)

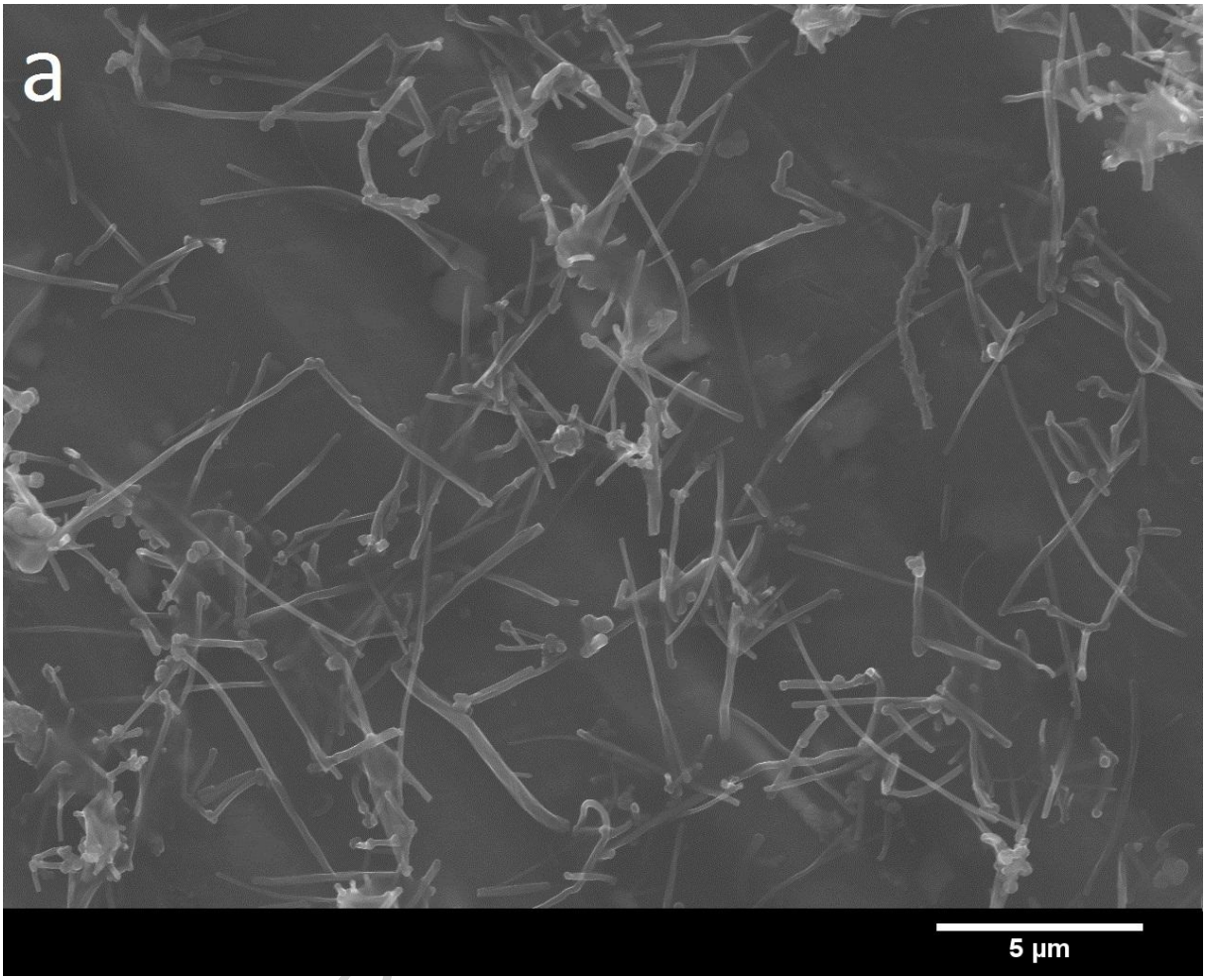


Figure 4 (a)

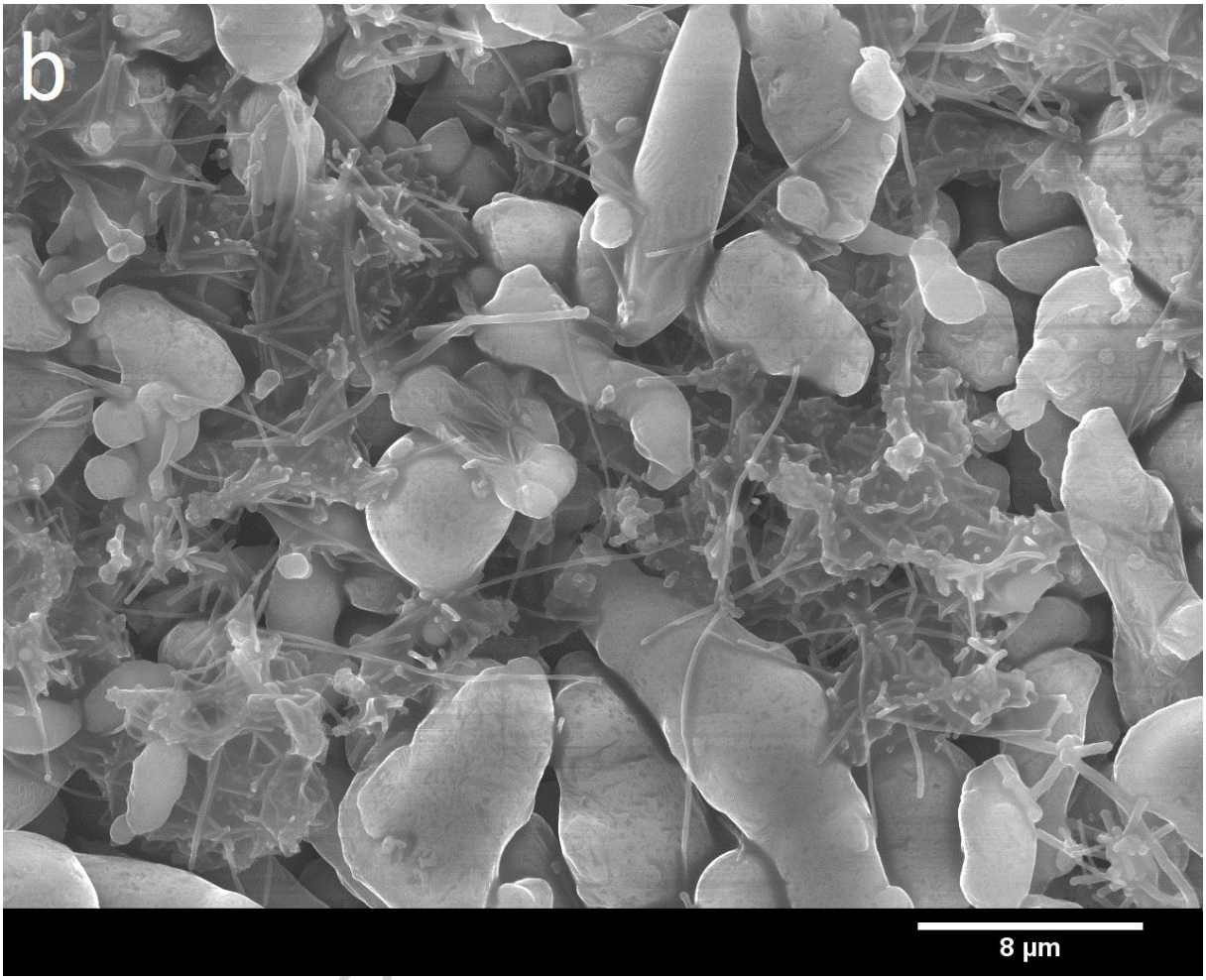


Figure 4 (b)

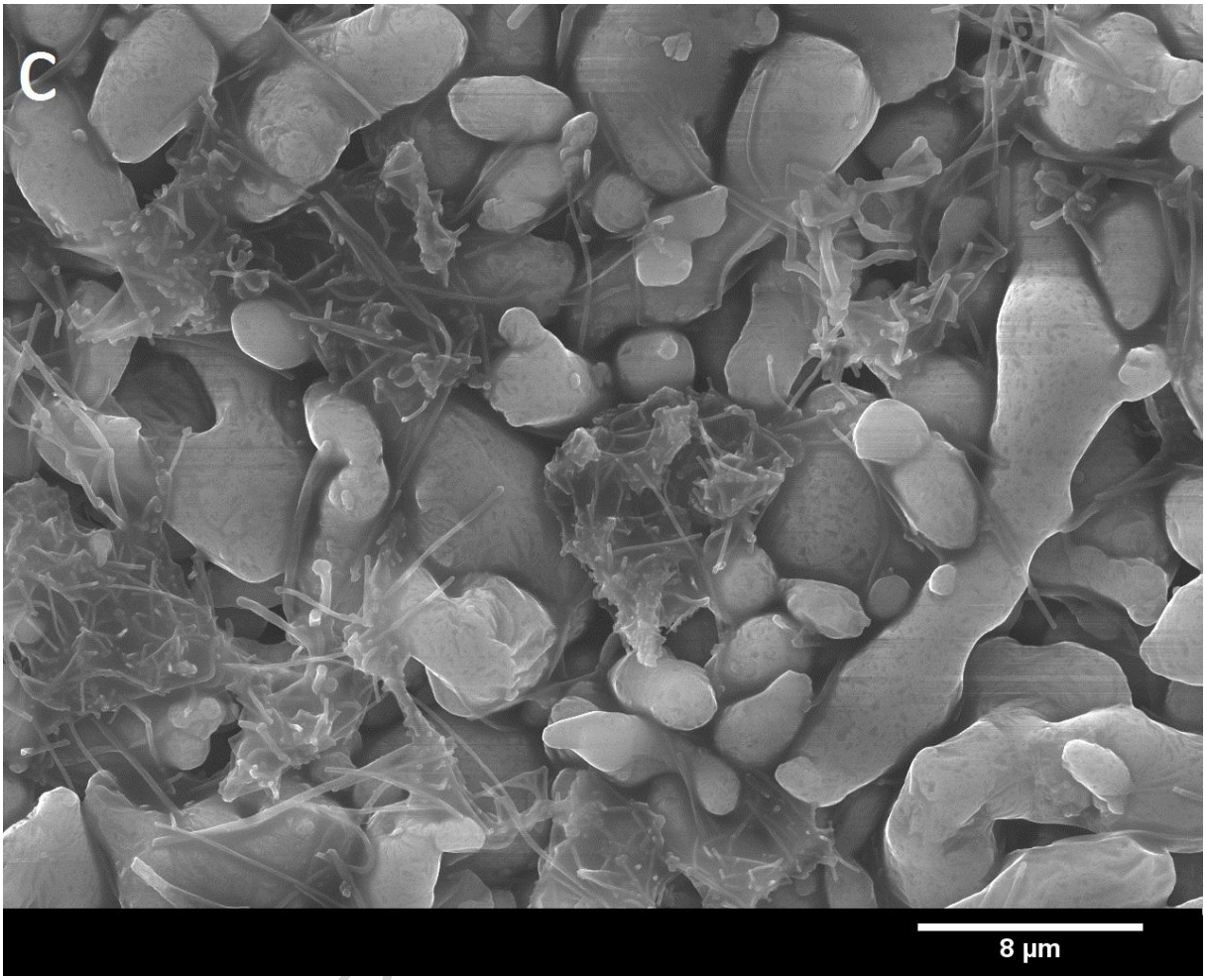


Figure 4 (c)

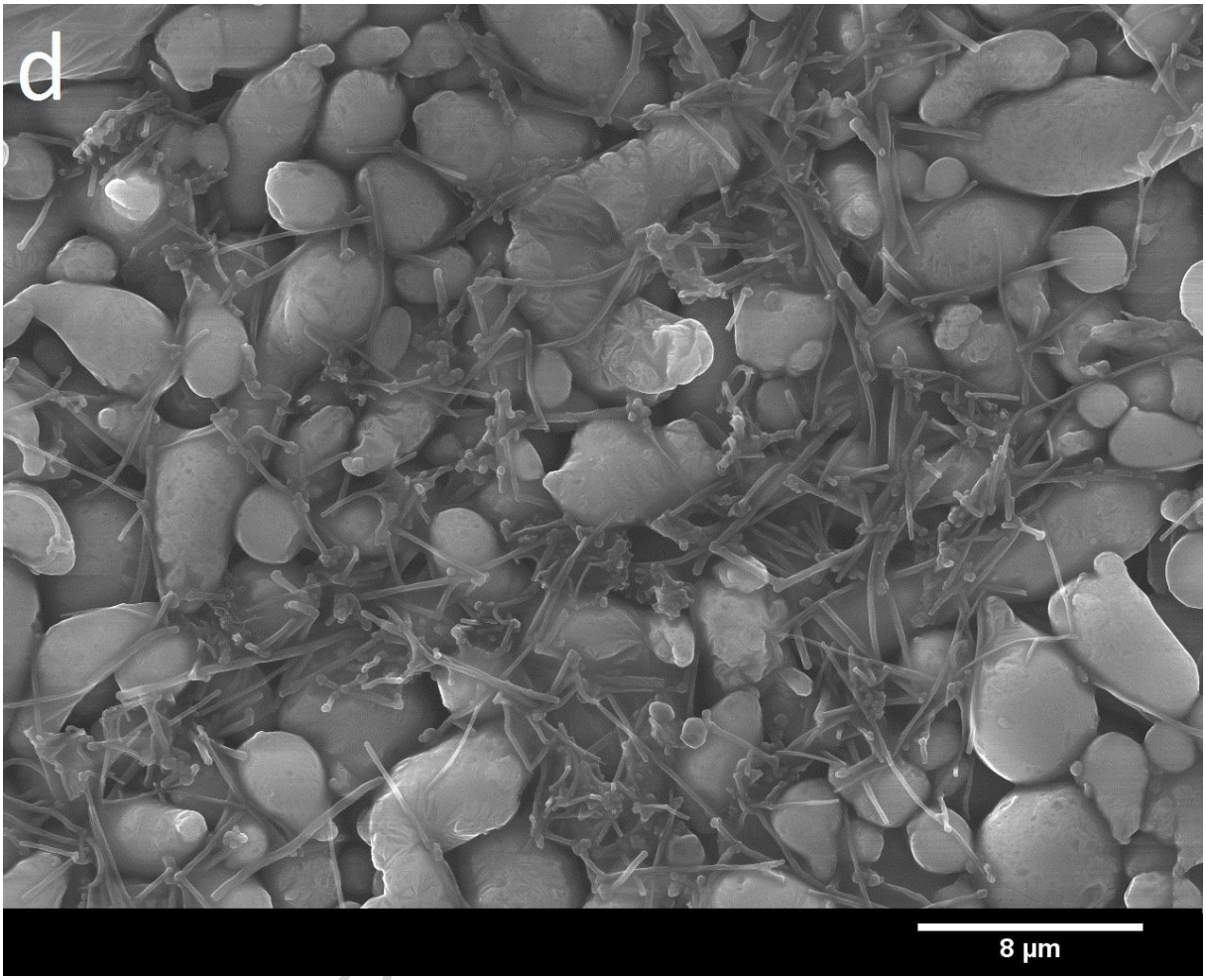


Figure 4 (d)

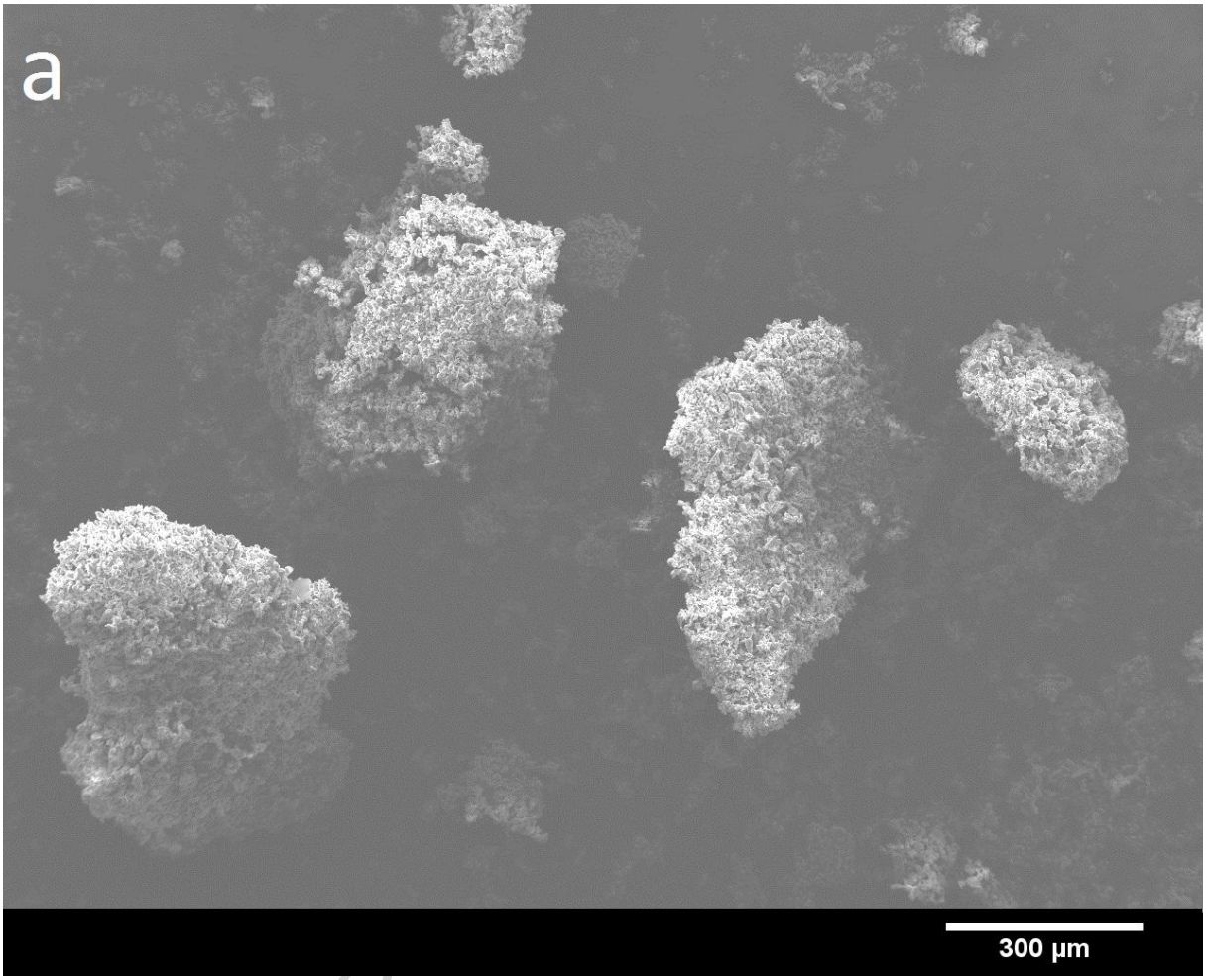


Figure 5 (a)

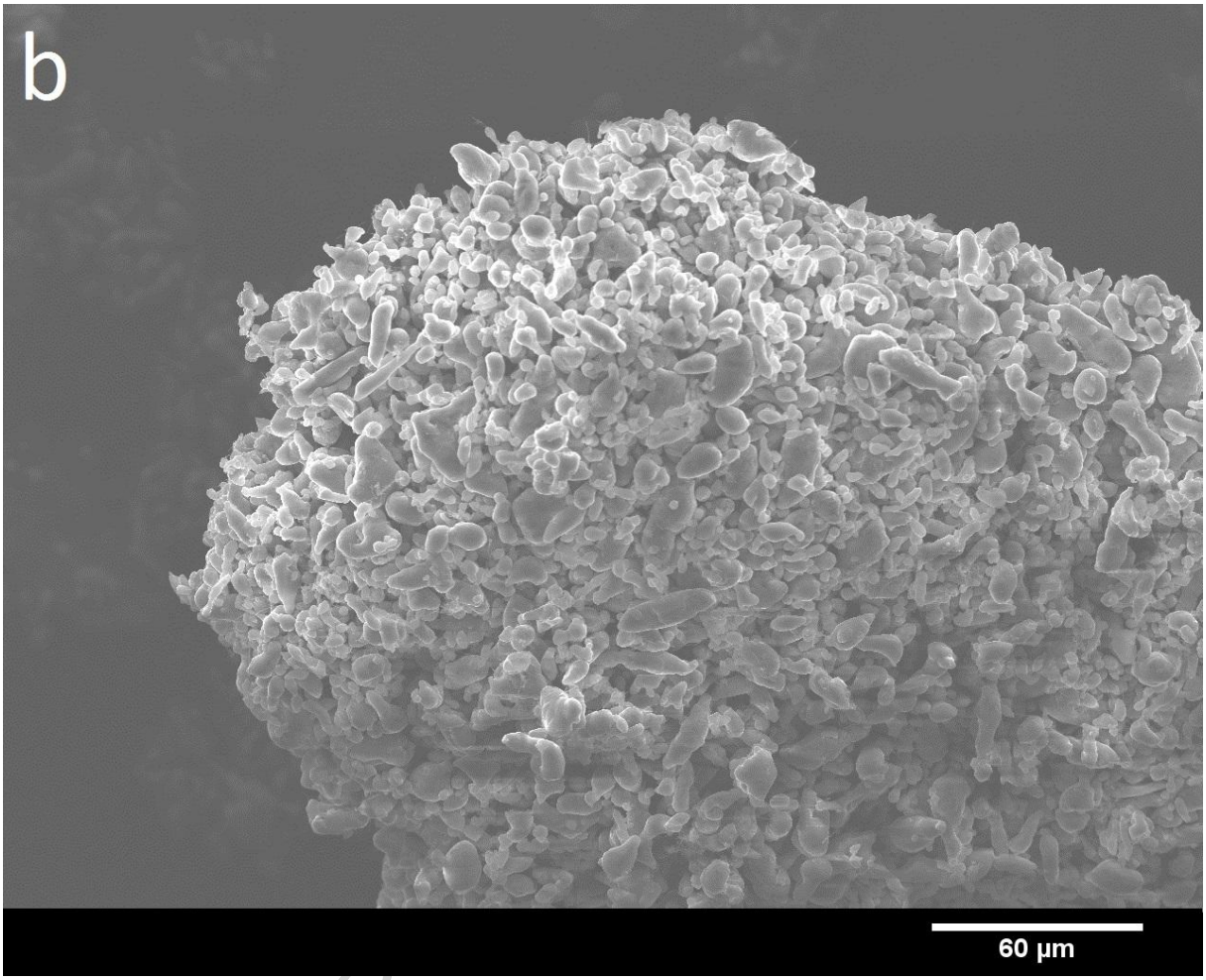


Figure 5 (b)

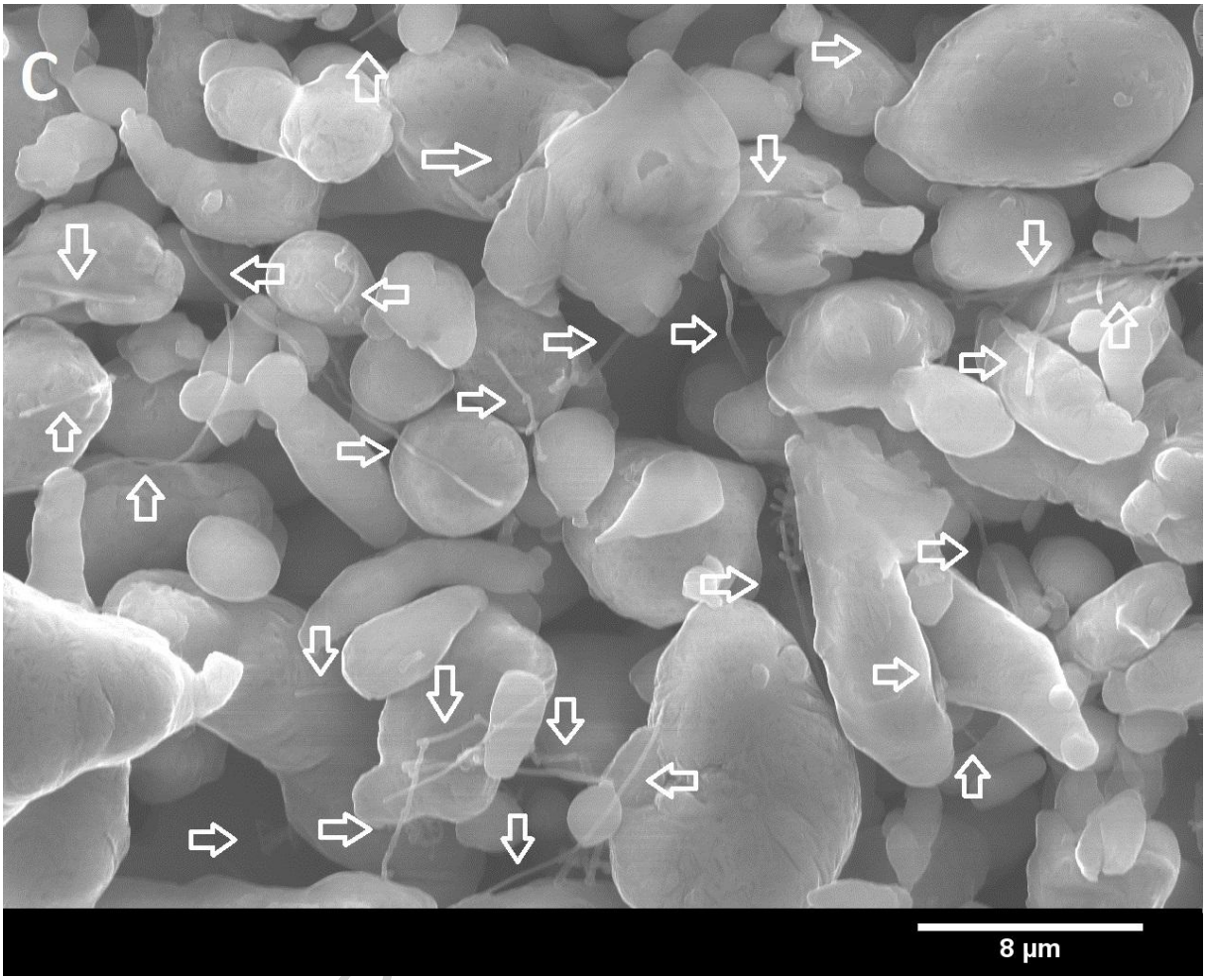


Figure 5 (c)

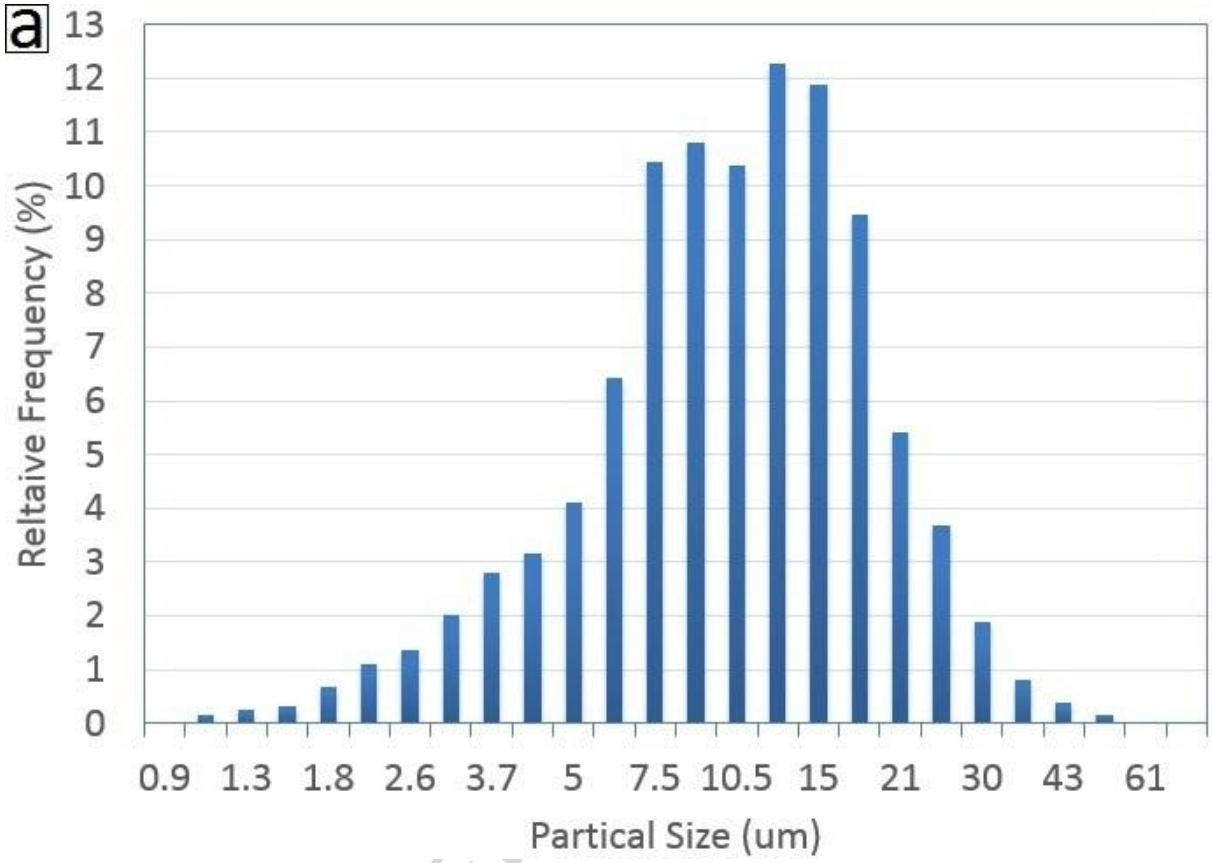


Figure 6 (a)

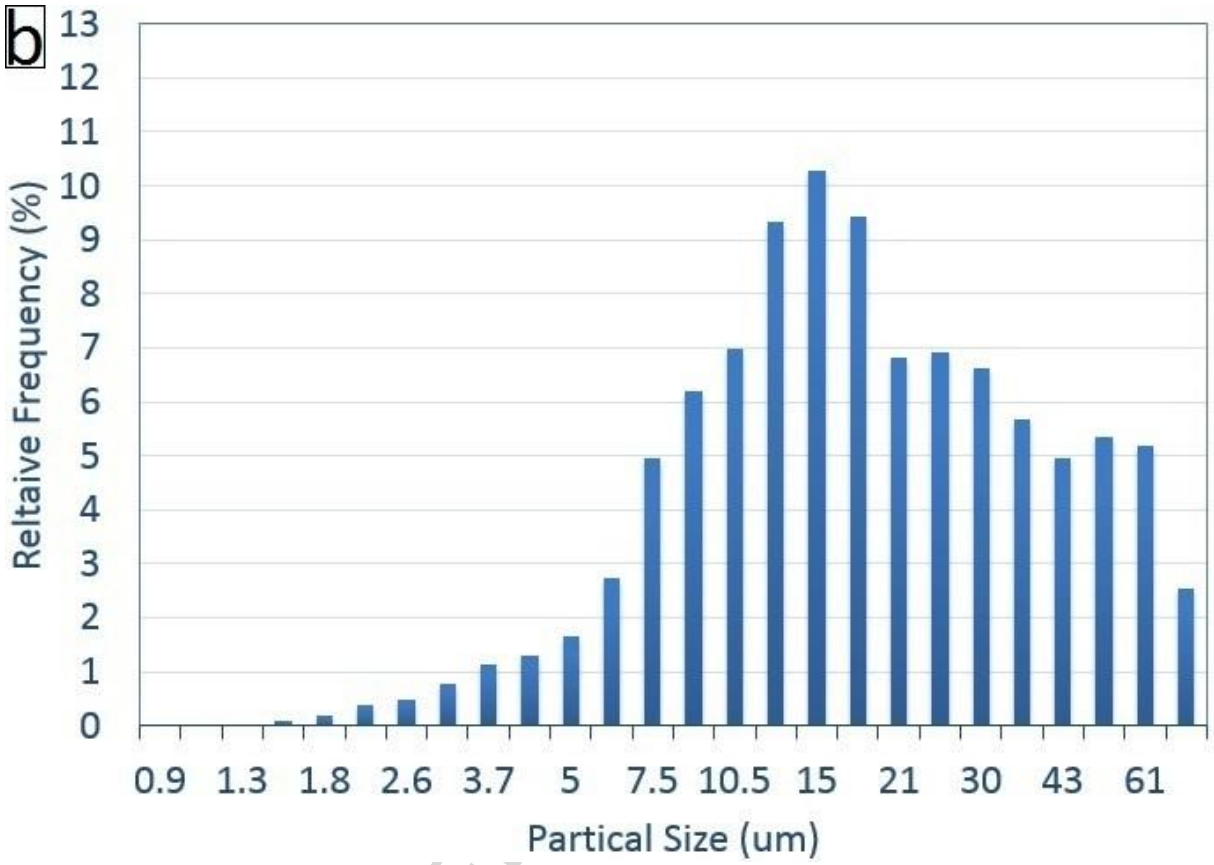


Figure 6 (b)

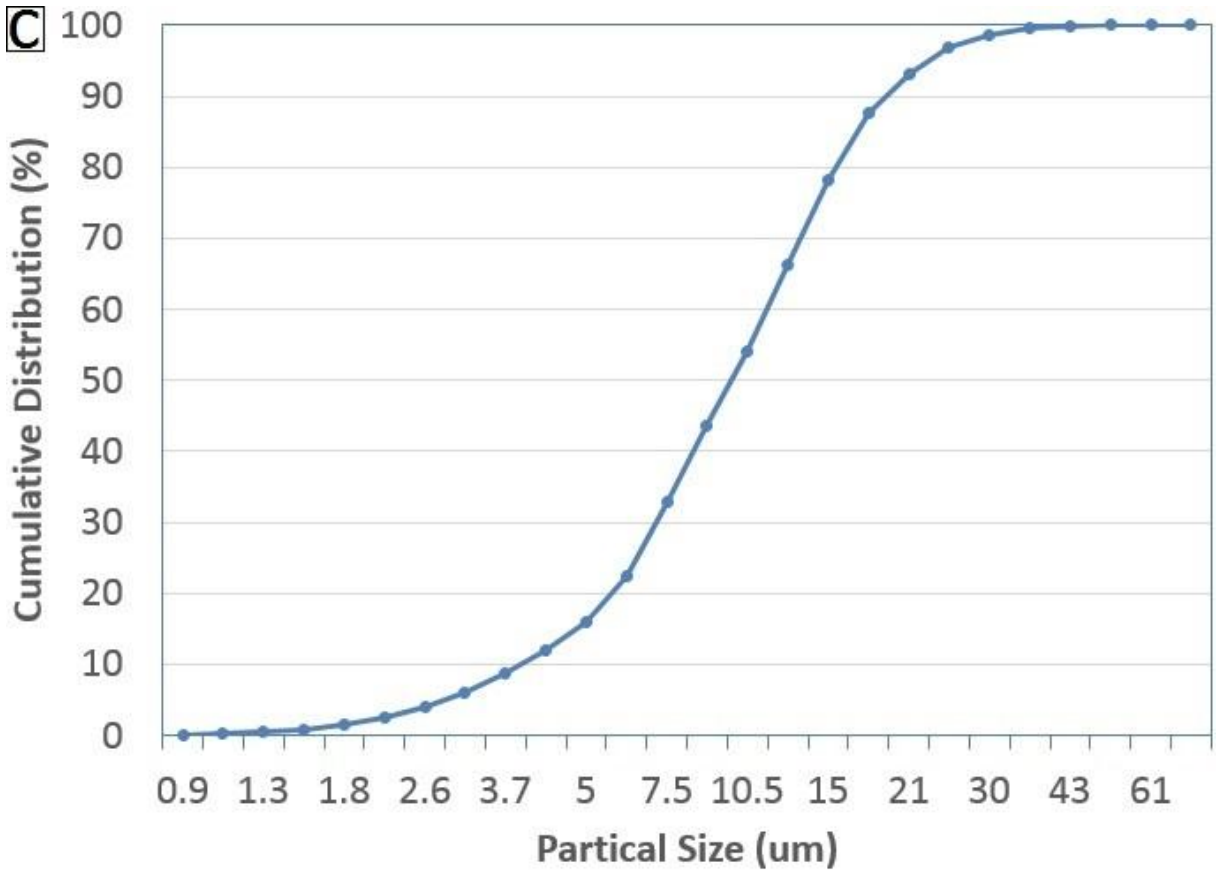


Figure 6 (c)

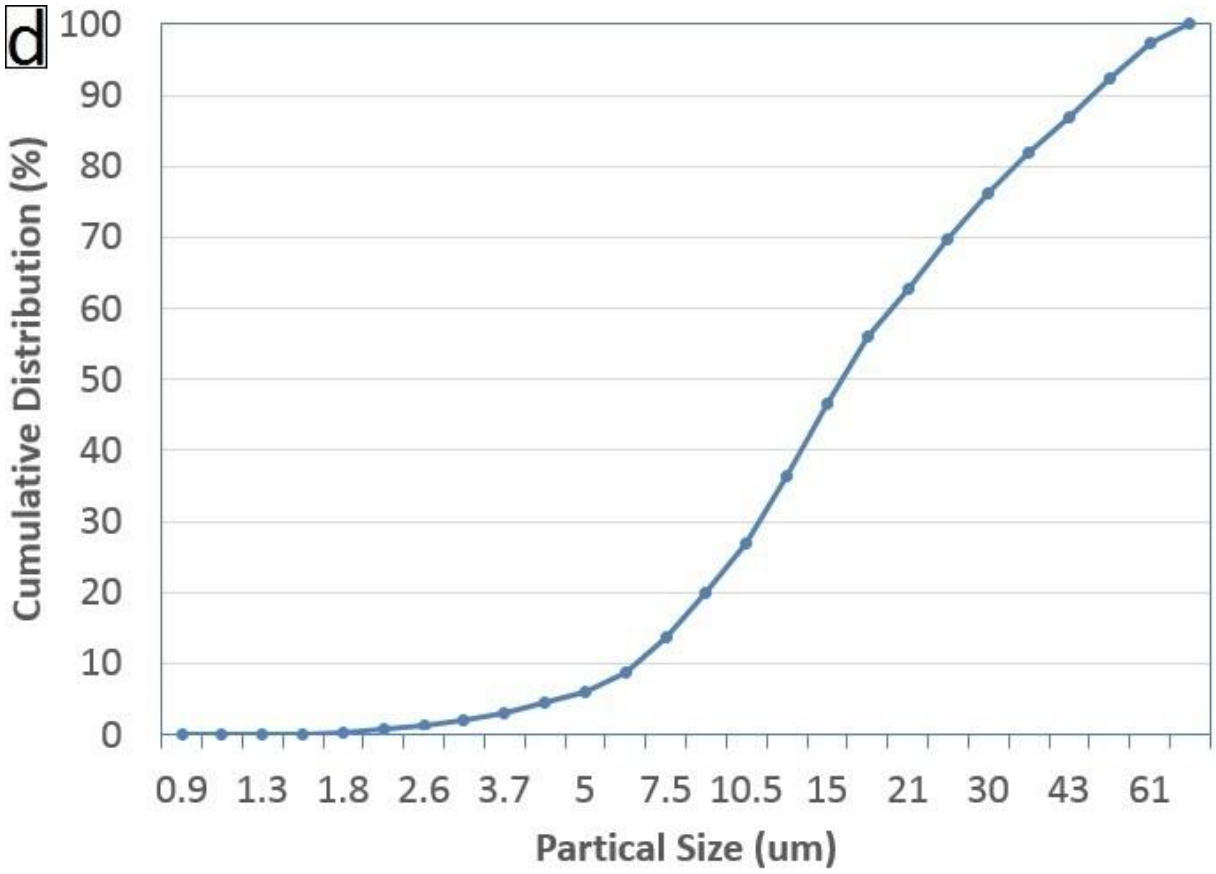


Figure 6 (d)

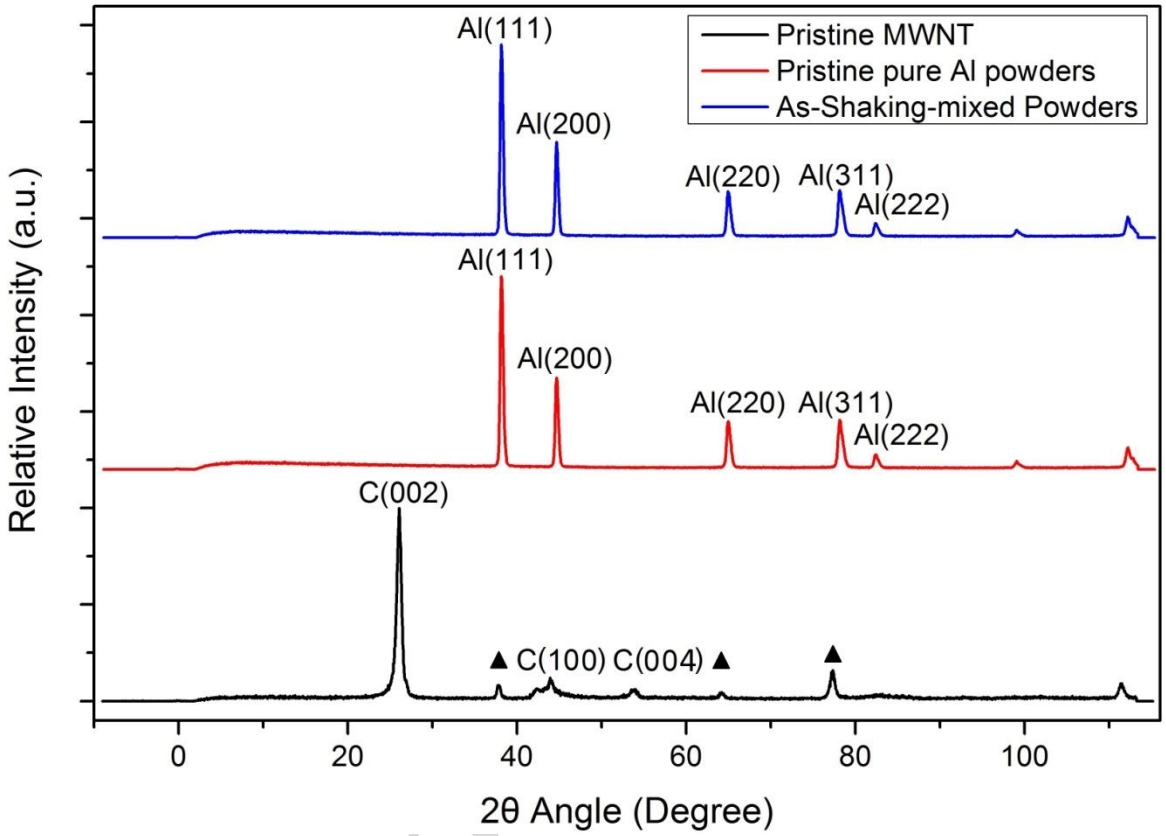


Figure 7

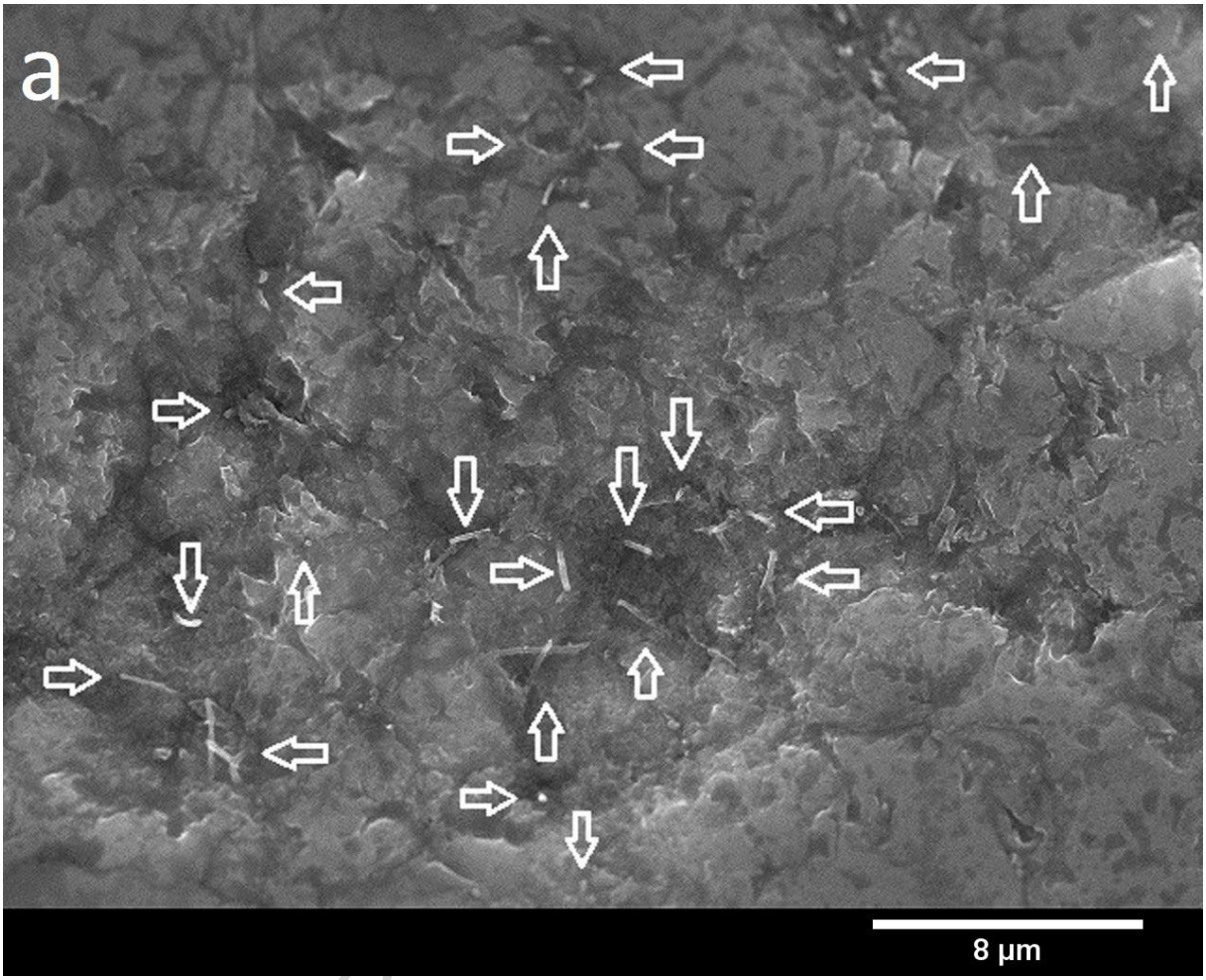


Figure 8 (a)

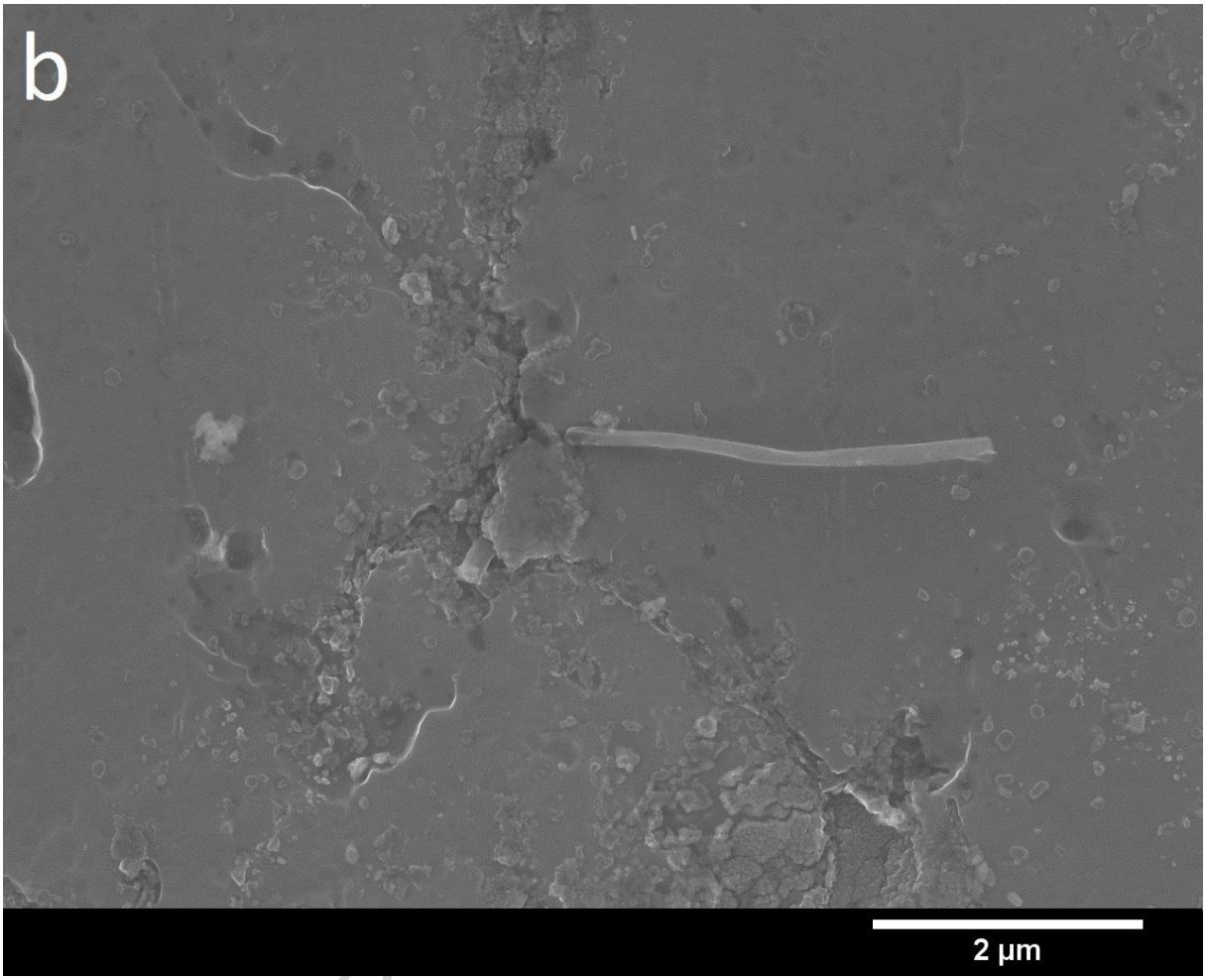


Figure 8 (b)

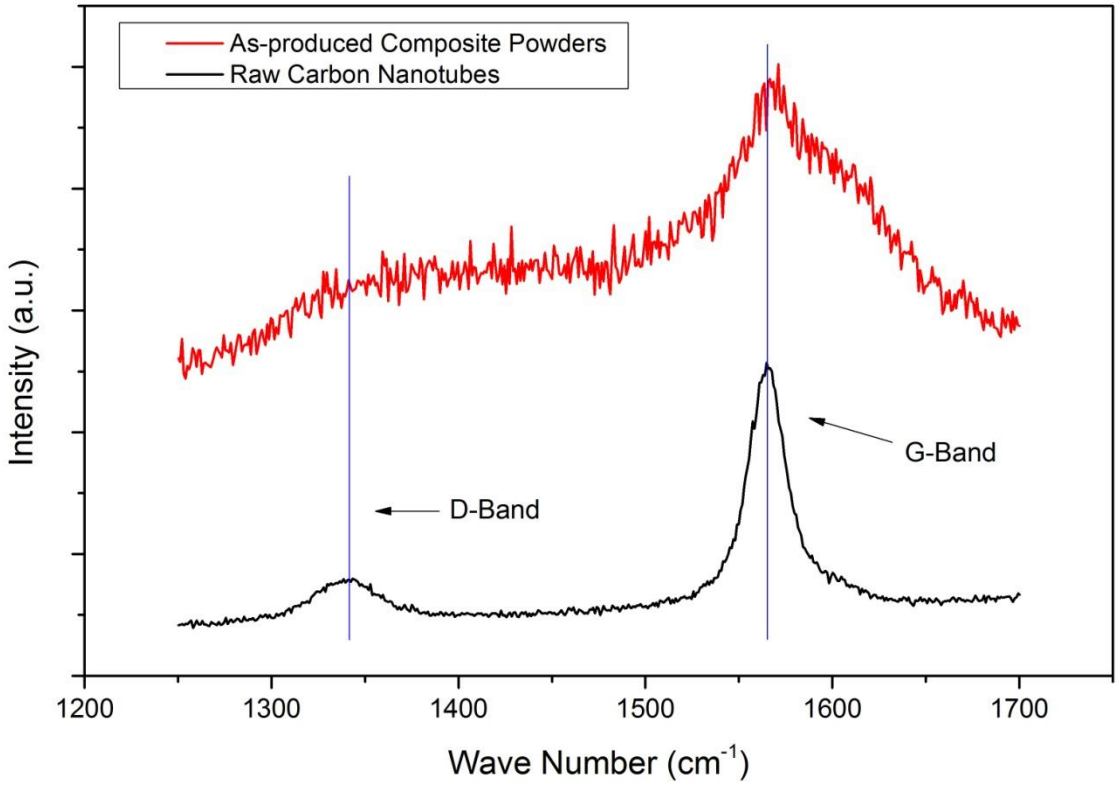


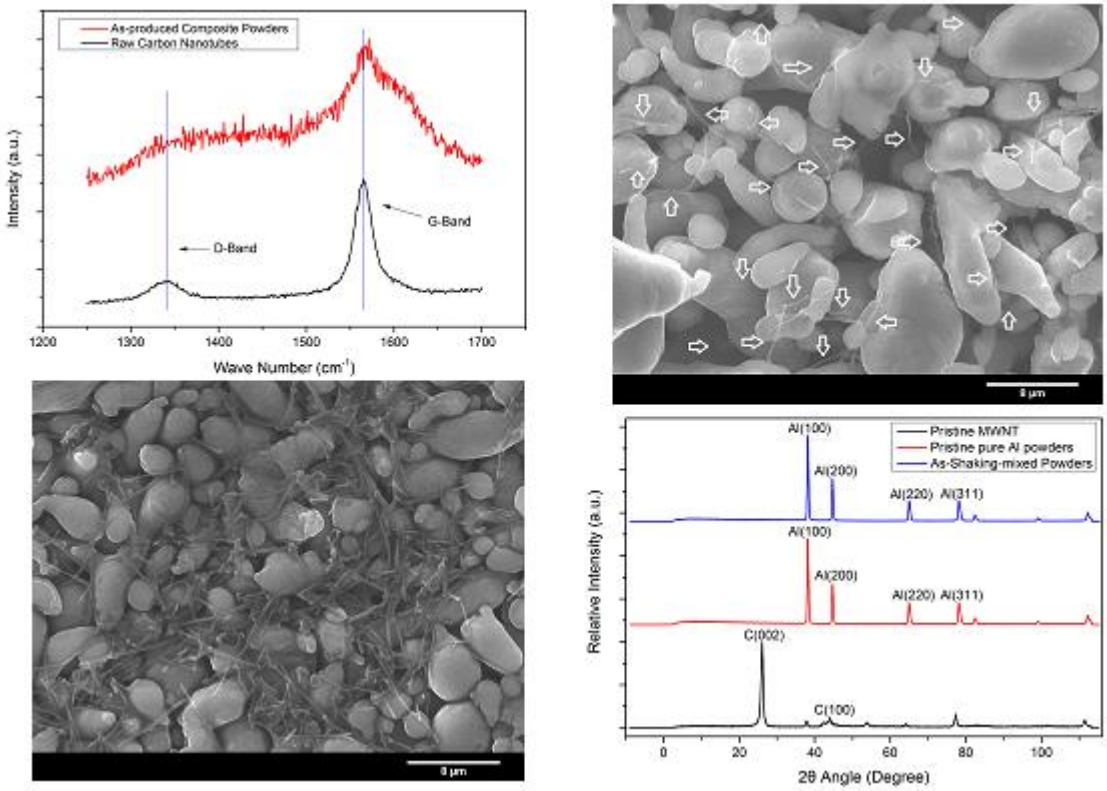
Figure 9

Table 1 The density of pure aluminum and MWNT/Al composite

	Green Density (g/cm^3)	**Theoretical Density (g/cm^3)**	**Relative Density (g/cm^3)**
Raw Al	2.31 ± 0.03	2.70	85.62 ± 1.1%
Al– 0.5 wt.% CNT	2.36 ± 0.05	2.69	87.73 ± 1.9%

Table 2 Various experiment methods and corresponding results in the present paper and other literature

	Matrix	CNT Concentration	Processing	Result
Authors of this paper	Al	0.5 wt. %	Wet-shake mixing	Intact CNT and uniformly dispersion
T. Kuzumaki et al.[7]	Al	3 or 6 vol. % (2.1 or 4.3 wt. %)	Stirred in ethanol at 300 rpm for 0.5h	CNT agglomeration and no obvious reinforcing effect
J.Z. Liao et al.[23]	Al	0.5, 1.0 and 2.0 wt. %	Can rolling	High loading of CNT formed agglomerates and impeded densification
A. M. K. Esawi et al. [22]	Al	0.5, 1.0 and 2.0 wt. %	1. No milling media mixer-shaking at 46rpm	CNT agglomeration
			2. No milling media planetary milling at 300rpm	CNT agglomeration
J.Z. Liao et al.[16]	Al	0.5 wt. %	1. Low energy ball milling for 4h	CNT agglomeration
			2. PEG assist Milling	CNT agglomeration
R. Pérez-Bustamante et al.[12]	Al2024	0.5 - 5.0 wt. %	High energy ball milling for 5, 10, 20 and 30 h	CNT damaged (shortened and uncapped)
Z.Y. Liu et al. [18]	Al	0.5 wt. %	Ball milling at 300 rpm for 8-12h	I_D/I_G ratio increased (damage of CNT structure)



Graphical abstract

HIGHLIGHTS

- Multi-walled carbon nanotube were homogenously dispersed in aluminum powders.
- The structure of MWCNT was intact and well preserved even after mixing.
- The aluminum particles kept original fine size in the as-produced composite powders.
- Wet shake-mixing technique is an effective method to incorporate CNT to Al matrix.