

Carbon Nanotube Reinforced Aluminium Matrix Composites – A Review

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Abstract: The present paper illustrates mixing procedures for Al-CNTs powder preparation alongside depiction of the CNTs dispersion results from the different mixing techniques. XRD analysis showed that the mean grain size of powders milled for 6 h was found to be 48.4 nm and for extrudates of CNT-Al, it is 56.6 nm. Based on the geometry and physical properties of multiwalled nanotubes, three strengthening mechanisms were considered for CNT/Al composite system. It was evident from the testing that as the content of nanotubes in the matrix increased, the micro-hardness measured on the Vickers scale also increased. The investigation of the damping behaviour of 2024Al-CNT composite showed that the damping capacity of the composite with a frequency of 0.5 Hz reaches 975×10^{-3} , and the storage modulus is 82.3 GPa when the temperature is 400° C, which shows that CNTs are a promising reinforcement for metal matrix composites to obtain high damping capabilities at an elevated temperature without sacrificing the mechanical strength and stiffness of a metal matrix.

Key words: Composites, differential scanning calorimetry (DSC), microstructure. Carbon nanotube

I. INTRODUCTION

Single and multi-wall carbon nanotubes have created tremendous expectations as strengthening additives for metallic, ceramic and polymer composites due to their high strength and stiffness. Metal matrix/CNT composites (MM/CNT) have a great potential in load bearing applications and electronic packaging due to their high specific strength, high thermal conductivity and low coefficient of thermal expansion. These properties are advantageous in advanced applications like aerospace and automotive structural members where a lower weight leads to savings in energy. In recent years, much research has been focused on the development of CNT reinforced Al matrix composites, because Al matrix composites have been wide prospects of applications in aviation, spaceflight and automobile industries[1]. CNTs have a Young's modulus of 1TPa, making them ideal reinforcements for composite materials. It is important to understand the relevant strengthening mechanisms involved in CNT/Al composites, in order to produce optimized composites [1]. The carbon nanotubes having excellent chemical stability due to their seamless cylindrical graphite structure are an exceptional candidate for the reinforcement in aluminum matrix[2]. The quality of dispersion, however, is a crucial factor which determines the homogeneity and final mechanical properties of these composites[3].

II. Al-CNTs Powder Preparation

0.5% of CNTs was added to the Al powders (Al-0.5 wt% CNT) and three different mixing methods, namely high energy and low energy ball millings, and polyester binder-assisting (PBA) technique were used[3,4,5,6,].

A. High energy ball milling

Al-0.5 wt% CNT powders were mixed by stirring before being transferred into the ball milled jar. Mechanical alloying was performed on a Retch PM400, Germany planetary ball mill machine. To prevent severe cold rolling, stearic acid (2 wt %) was added to the mixture during the ball milling. Ball (agate type) to powder weight ratio was 5:1 with a revolution speed of 200 RPM. Ball milling time was 4 h, with an interim period of 30 min for every one hour, in order to prevent over-heating.

B. Low energy ball milling

The same amount of Al-CNTs powders and balls as used in high energy ball milling were loaded into a plastic container then blended by a horizontal rolling machine. The mixture was rolled for 4 h continuously with a speed of 200 rpm.

C. Polyester binder-assisted (PBA) mixing

A polyester binder assisted (PBA) method for coating CNTs on Al powder was used. Firstly, polyethylene glycol (PEG) with a molecular weight of 20,000 (MW. 20,000) in a form of flake was hand mixed with CNTs in a mortar. The mixture was

transferred into a Haake twin screw mixer. To effectively disperse the CNTs within the PEG, the mixture was melt blended for 20 min, at the desired melting point of PEG, i.e. 70° C, using a speed of 60 rpm. Subsequently the Al powder was fed into the viscous PEG-CNTs mixture. The compound was continuously blended for another 30 min with the same rotational speed to obtain a good dispersion of CNTs in the mixture. The volume fraction of PEG to Al was 4:6. The Al-CNTs-PEG precursor was cooled cured and heated at 400° C in an argon atmosphere for 3 h to thoroughly decompose the PEG. The schematic of the processing procedure is presented in Fig.1a and the schematic of the twin screwers is shown in Fig1b.

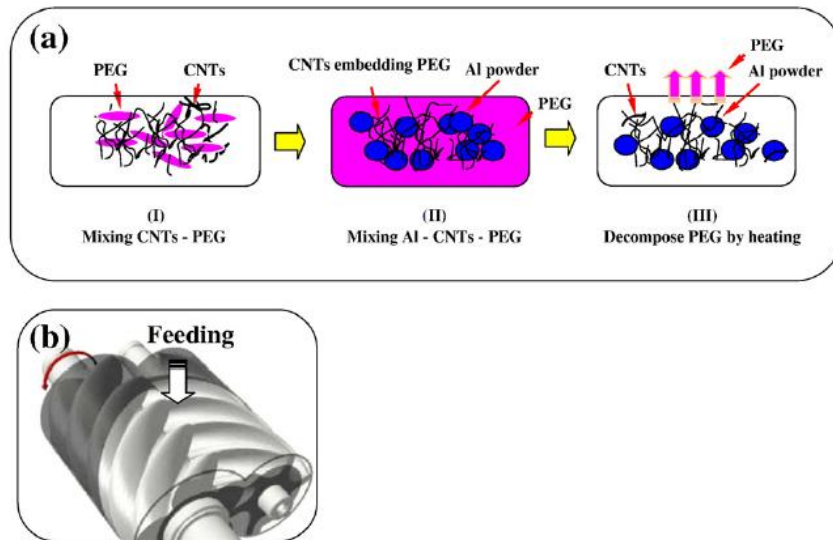


Fig.1a Illustration of the mixing procedures of Al-CNTs powders by polyester binder-assisted (PBA) technique; and the (b) schematic of the twin-screw mixer [3]

The summary of the CNTs dispersion results from the different mixing techniques is schematically presented in fig 2 and Table 1[3].

TABLE 1
THE SUMMARY OF THE CNTS DISPERSION RESULTS FROM THE DIFFERENT MIXING TECHNIQUES [3]

Mixing technique	Al powder morphology	C N T morphology	Dispersion effect
P B A	Round shape	Negligible change	Coated with Al powder through agglomerates still existed
Low energy ball milling	Round shape with minor change	Medium change	Dispersed within Al powder though agglomerates still existed
High energy ball milling	Round shape with severe change	Severe damage	Effectively dispersed though insufficiently distributed

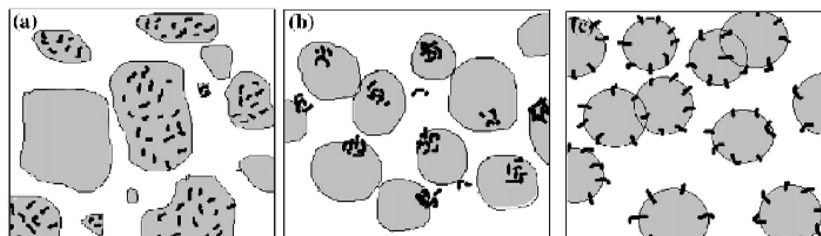


Fig2 Schematic depictions of CNTs and Al powder after different mixing techniques. In (a) high energy ball milling, CNTs were effectively dispersed though insufficiently distributed within the Al powders; and in (b) low energy ball milling, CNTs dispersed within Al powder though agglomerates still existed; and in (c) the PBA method, CNTs were coated with Al powders though agglomerates still existed. [3]

Ball milled samples containing CNTs exhibited high notch sensitivity and consistently fractured outside the gauge length, which was not the case for pure milled aluminium samples. Fig.3 shows nanoindentation and Vickers hardness results for extruded samples of un-milled and milled Al, in addition to the milled Al-2 wt% CNT composites. The hardness is seen to increase significantly (approx. 3 times) for extrusions of ball milled powders, due to excessive strain hardening. XRD analysis showed that the mean grain size of powders milled for 6 h was found to be 48.4 nm and for extrudates of CNT-Al, it is 56.6 nm which is only slightly greater than the grain size of ball milled powders, thus suggesting that extrusion at 500° C retains the nanostructure[5].

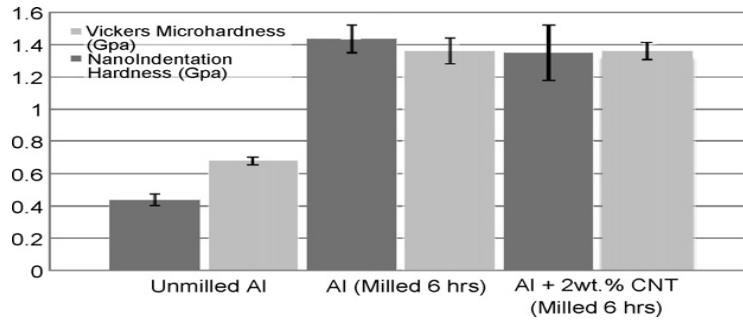


Fig 3 Vickers micro-indentation and nano-indentation hardness of un-milled and milled pure Al and Al-2 wt% CNT extruded samples [5]

III. Fabrication Techniques

A. Powder Metallurgy

The aluminum powder (purity of 99.0%, grain size of 70µm) and CNTs were homogeneously mixed by hand grinding for 30min, and ethanol was added to avoid possible oxidation. The mixture of CNTs and aluminum powder was uniaxially pressed in steel dies under a pressure of 300MPa for 2 min. The specimens were isothermally sintered at 100°C for 1 h and 600°C for 2 h in a pure argon atmosphere. The specimens for testing, containing 0 to 3% of CNTs respectively, were 60 mm X 10 mm X 2 mm in size [1].

Multiwalled carbon nanotubes (MWNTs) were treated with the reflux within the concentrated nitric acid for 0-25 h to purify and disperse the tangled MWNTs. The results show that the reflux time markedly affects the morphology of MWNTs [7].the weight loss of MWNTs increase as the reflux time increases. Meanwhile, the dispersion of MWNTs with 0-2.0 wt. % in 2024Al powders using mechanical stirring with an assisting ultrasonic shaker in ethanol was also studied. When the content of MWNTs is less than 1.0 wt. %, MWNTs can uniformly distribute on the surface of 2024Al powders; however, when the content of MWNTs is 2.0 wt. %, MWNTs entangled with each other on the surface of 2024Al powders[7].

B. Compcasting

A356 aluminium alloys reinforced with CNTs were produced by stir casting and Compcasting routes. In order to alleviate the problems associated with poor wettability, agglomeration and gravity segregation of CNTs in the melt, CNTs were introduced into the melts by injection of CNT deposited aluminum particles instead of raw CNTs. Aluminum particles with a mean diameter of less than 100µm were first deposited by CNTs using Ni-P electro less plating technique and then injected into the melt agitated by a mechanical stirrer. The slurry was subsequently cast at a temperature corresponding to full liquid as 0.15 and 0.30 solid fractions

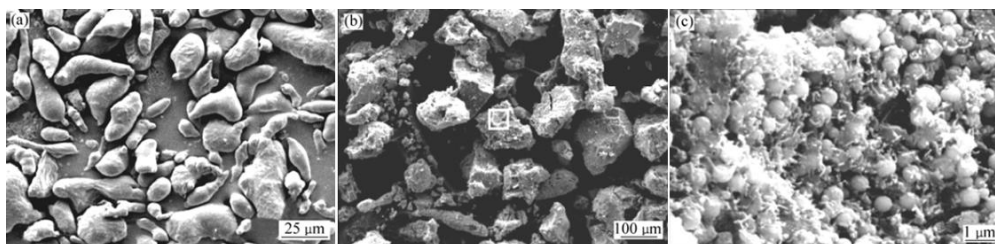


Fig4 SEM images of aluminum particles: (a) Before coating; (b) After coating; (c) Higher magnification of the area marked by rectangle in (b) [8]

Fig.4 Illustrates the SEM images of aluminum particles before and after coating. It is shown that a proper mechanical interaction between aluminum particles and Ni-P-CNT composite coating has been achieved. It is also evident that the coating has increased the average size of the aluminum particles. The higher magnification image of the area marked by the rectangle in Fig.4b is shown in Fig.4c and reveals the characteristic morphology of the Ni-P electroless coating on the aluminum particle and, more importantly, a very uniform distribution of CNTs in the Ni-P-CNT composite coating. Due to gradual co-deposition of Ni-P and CNTs on the aluminum particles, no CNT agglomerates can be observed in the composite coating. The results show that the addition of CNTs to A356 matrix can significantly refine both full liquid and semi-solid cast microstructures. Hardness of the samples is also significantly increased by the addition of CNTs and A356-CNT composite cast at 0.3 solid fractions produces highest hardness [8].

C. High Pressure Torsion (HPT)

Fig.5 shows severe plastic deformation by high pressure torsion (HPT) of powders at elevated temperature (473 K) was employed to achieve both powder consolidation and grain refinement of aluminum matrix nanocomposites reinforced with 5 Vol% CNTs. Before the HPT, the powders were ball milled using a planetary ball mill in order to achieve a molecular level mixing. Aluminum was treated by the same process for a reference. The HPT processed disk was composed of considerably equilibrium grain boundaries with high misorientation angles. The CNT-reinforced ultrafine grained microstructural features resulted in high strength and ductility [9].

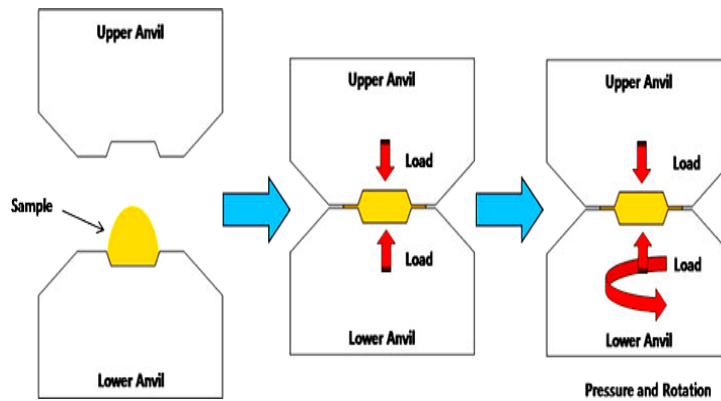


Fig 5 Schematic illustration of HPT facility and operation with a pressure showing the first compressive stage and the next torsional stage [9]

IV. Chemical stability of CNTs in Al matrix

For CNTs / Al composites, an important issue of the chemical stability of the CNTs in the Al matrix, is whether or not CNTs can be applied to the Al matrix as reinforcement. The DSC pattern of the mixture of CNTs and 2024 Al powders is shown in Fig.6. It was distinctly seen that a sharp exothermic peak appeared beyond the Al melting peak, which could be Al₄C₃ carbides formed in the reactions of CNTs – Al [10, 11].

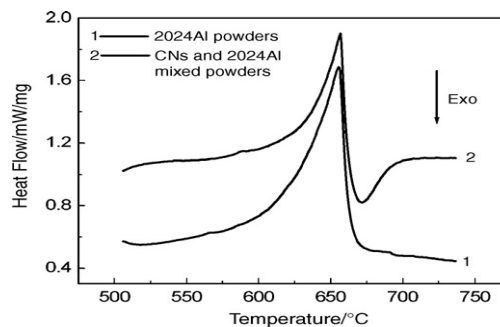


Fig6 DSC of a mixture of CNTs and 2024Al alloy powders. [10]

V. Strengthening Mechanisms

Based on the geometry and physical properties of multiwalled nanotubes, three strengthening mechanisms were considered for CNT/Al composite system, namely thermal mismatch, Orowan looping and shear lag models.

A. Thermal mismatch

There exists a significant coefficient of thermal expansion mismatch between the matrix and the carbon nanotubes, which would result in the prismatic punching of dislocations at the interface leading to work hardening of the matrix. The dislocation density generation is likely to be higher, which in turn would result in increased strengthening. Incremental strength, $\Delta\sigma = \alpha\mu\rho^{(1/2)}$ Where $\alpha = \text{constant} = 1.25$, μ is the modulus of rigidity of the matrix (Al), b is Burger’s vector. Dislocation density (ρ) is given by $\rho = 10A\varepsilon/(bt(1-A))$, where A is the reinforcement (carbon nanotube) volume fraction, ε is thermal strain, b is Burgers vector, and t is the dimensions of the reinforcement.

B. Orowan looping

In this mechanism the motion of the dislocation is inhibited by nanometer sized carbon nano tubes, leading to bending of these dislocations between the carbon nanotubes. This produces a back stress, which will prevent further dislocation migration and results in an increase in yield stress. The Orowan looping mechanism is important in aluminum alloys, which are strengthened by fine precipitates. Since carbon nanotubes effectively represent very fine particles, perpendicular to the tube axis, in the order of a few nanometers, it strengthens the aluminum matrix. Incremental shear strength of the composite $\Delta\tau = \text{Constant} * \mu b A^{(1/2)} / r * \ln(2r/r_0)$ where constant = 0.093 for edge dislocation and 0.014 for screw dislocation, r_0 is the core radius of dislocation = 3.5×10^{-9} m, b is the Burger’s vector, μ is the modulus of rigidity of matrix (Al) = 2.64×10^{11} N/m², and r is the volume equivalent radius = 1.593×10^{-7} m for MWNT and 7.087×10^{-9} m for SWNT.

C. Shear lag

This model involves the transfer of load from the matrix to the reinforcement by interfacial shear stress. Thus the stiffness of the carbon nanotubes is directly utilized. The Young’s modulus (E_c) of the composite is $E_c = A E_f (1 - (\tan h (ns))/ (ns)) + (1 - A) E_m$ where E_f is the Ypung’s modulus of the reinforcement (CNT), $n = (2 E_m / (E_f (1 + \gamma_m) \ln (1 / A)))^{(1/2)}$, E_m is the Young’s modulus of matrix (Al), s is the aspect ratio of the reinforcement (CNT), A reinforcement (CNT) volume fraction, $\gamma_m =$ Poisson’s ratio of Al matrix, and aspect ratio $s = 100$ for MWNT and 500 for SWNT. The strengthening of the composite could be due to the synergetic effect of these mechanisms[12].

VI. Properties

The effects of CNT content on the relative density, the hardness, and the friction and wear behaviour of the CNT/Al composites under dry sliding conditions were investigated [1].

A. Micro-hardness

The investigations have been extended to carry out the micro-hardness measurements on composite samples containing 1, 2, 4 and 10 wt% of carbon nanotubes reinforced aluminium matrix. It was evident from the testing that as the content of nanotubes in the matrix increased, the micro-hardness measured on the Vickers scale also increased. Fig.7 Illustrates the different values of micro-hardness with the quantity of nanotubes reinforced in the matrix [2].

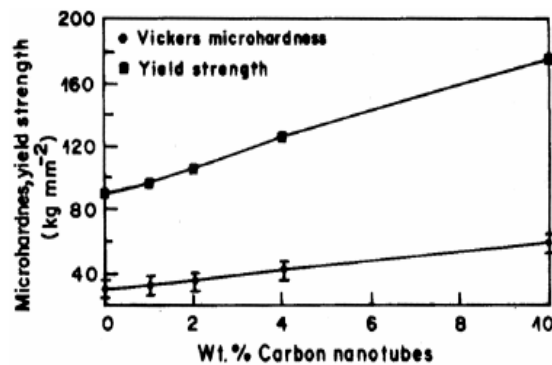


Fig.7 Change in micro hardness and yield strength with respect to increase in content of the CNT in aluminum matrix [2]

B. Electrical properties

There is a slight increase in electrical resistivity with the addition of carbon nanotubes. The carbon nanotubes have, in any case, a lower electrical conductivity than aluminium. In addition, if the carbonnanotubes are agglomerated at the Al grain boundaries, they can form a kind of grain boundary phase which will increase the scattering of the charge carrier at grain boundaries, hence reducing the conductivity. The carbide phases, which are also poorly conducting, and the porosity, which is larger than pure Al, can also influence the electrical resistivity of the composites.

From room temperature on, the electrical resistivities decreased linearly as the temperature decreased, which is typical metallic behaviour. At about 80 K there is an abrupt drop (>90%) in the electrical resistivity of the Al-carbon nanotube composites. Below this transition temperature the resistivity remains at the level of “zero resistance” down to the helium temperature. Such

a phenomenon resembles a superconducting transition. There has been no explanation for these observations [13]. The tensile yield and ultimate strength of Al-MWCNT increased by 90% with 2 wt% addition of MWCNTs. [4]

C. Wear

The results indicate that the addition to the aluminium matrix of 2.0% (mass fraction) carbon nanotube causes the increase in the Vickers hardness of about 80%. Within the range of carbon nanotubes content from 1.0% to 2.0% both the friction coefficient and wear rate composites decrease with the increase of carbon nanotube content. The delamination wear is the main wear mechanism for the composites [1].

D. Damping Characteristics

The damping behaviour of 2024Al-CNT composite was investigated with a frequency of 0.5, 1.0, 5.0, 10, 30 Hz at a temperature of 25-400°C. The experimental results show that the frequency significantly affects the damping capacity of the composite when the temperature is above 230° C; meanwhile, the damping capacity of the composite with a frequency of 0.5 Hz reaches 975×10^{-3} , and the storage modulus is 82.3 GPa when the temperature is 400° C, which shows that CNTs are a promising reinforcement for metal matrix composites to obtain high damping capabilities at an elevated temperature without sacrificing the mechanical strength and stiffness of a metal matrix. There are three possible mechanisms that could be responsible for the observed increase in mechanical damping: (1) high inherent damping of carbon nanotubes with graphite structure, (2) energy dissipation caused by interfacial sliding at the nanotube-matrix interface at an elevated temperature and (3) energy dissipation caused by a small quantity of micro voids that exists in the composites fabricated by the powder metallurgy technique [14].

E. Toughness of aluminium-carbon nanotube composites

Fig.8 shows the effect of CNT addition on the ratio of the failure strain of Al/CNT composite to that of un-reinforced Al sample prepared by the same route. It is observed that except one study [4], all studies observe a reduction in the ductility of the composite due to CNT addition. There is a large scatter in the data for similar compositions showing that ductility is sensitive to processing. Two trends can be observed from fig 8 the first trend is obtained from the tensile data on the samples prepared by Deng et al. and Esawi and Borady[15]. Which indicates that poor dispersion and poor CNT-matrix bonding leads to low strengthening as well as a decrease in the ductility. A poor CNT-matrix bonding can be susceptible to cracking nucleation under tension. Due to the small size of the CNTs, the cracks may be smaller and below the critical flaw size to create any damage. However overlapping CNTs could lead to interconnected cracks which will cause premature failure of the composite. The second trend is obtained from data from Choi et al. And He et al[15], which show that strengthening is achieved at the expense of ductility. This is a commonly observed observation in fibre reinforced composites resulting due to the smaller ductility of the fibre. So there is a trade-off that needs to be made between strength and ductility.

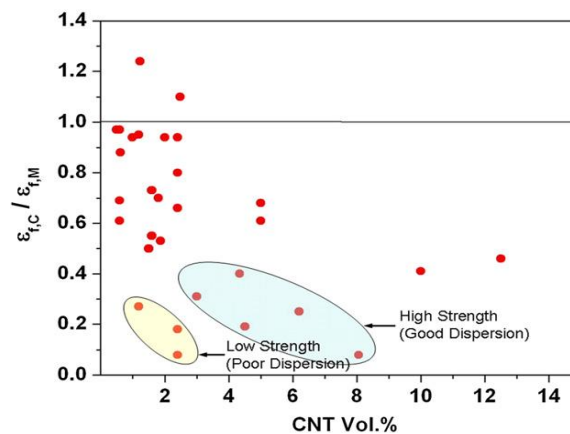


Fig 8 Effect of CNT content on the ratio of the failure strain of Al/CNT composite ($\epsilon_{f,C}$) to un-reinforced Al sample ($\epsilon_{f,M}$) [15]

Fig.9 shows the percentage change in the toughness of Al/CNT composites over Al sample prepared by the same route. The measure of toughness is obtained by the product $\sigma UTS \epsilon_f$. It is observed that there is a lot of scatter in the data. A decrease in the toughness is observed when CNT dispersion is poor as shown by the data by Deng et al. and Esawi and Borady[15]. At high CNT concentrations above 5 vol.% CNTs, there is a decrease in the toughness of the composite. The effect of the increase in the strength due to good dispersion is suppressed by the decrease in ductility, even for good dispersion.

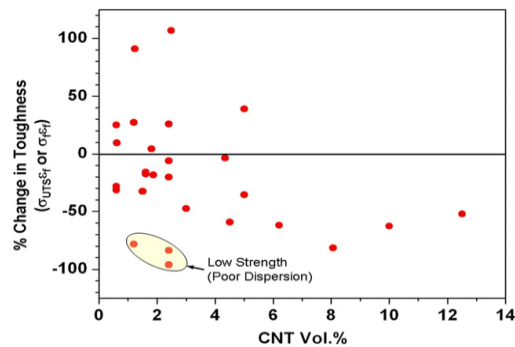


Fig.9 Effect of CNT content on change in the toughness of Al/CNT composites over Al sample[15]

VII.CONCLUSION

Ball milled samples containing CNTs exhibited high notch sensitivity and consistently fractured outside the gauge length. It is shown that a proper mechanical interaction between aluminum particles and Ni-P-CNT composite coating has been achieved. It is also evident that the coating has increased the average size of the aluminum particles. The HPT processed disk was composed of considerably equilibrium grain boundaries with high misorientation angles. The CNT-reinforced ultrafine grained microstructural features resulted in high strength and ductility. The strengthening of the composite could be due to the synergistic effect of mechanisms thermal mismatch, Orowan looping and shear lag models. CNTs are a promising reinforcement for metal matrix composites to obtain high damping capabilities at an elevated temperature without sacrificing the mechanical strength and stiffness.

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