

## EFFECT OF ULTRASONICATION DISPERSION TECHNIQUE ON SINTERING PROPERTIES OF CNT REINFORCED Al-Zn-Mg-Cu POWDER

CNT has been researched significantly due to its properties especially mechanical and conductivity properties. Due to strong affinity between particles, dispersion has been remained a problem for its applications. Ultrasonication technique was used to disperse CNT on aluminum powder. Ultrasonication has successfully dispersed CNT on aluminum powder in ethanol. Low energy ball milling for 1h under argon gas with 5:1 ball to powder ratio was used to optimize dispersion of CNT on aluminum. CNT concentration in this research is ranged from 0.1 up to 3wt%. Premix Al-5.5Zn-2.5Mg-0.5Cu powder was used as matrix for this research. Sintering at 580°C for 1h under argon gas was carried to produce high density materials. Pores are still remained on material and it is possibility that pores were caused by agglomeration of CNT. Dispersion high content of CNT is still remained obstacle on this research. Aluminum carbide, Al<sub>4</sub>C<sub>3</sub> is recognized as a 'bridge between aluminum and CNT for stress transfer was investigated in this research. Raman Spectroscopy, XRD and SEM-EDS were carried out to characterize materials.

*Keywords:* Powder Metallurgy, Nanocomposites, Sintering, Aluminum Alloys

### 1. Introduction

Aluminum-CNT composite has been researched with several researchers because of high mechanical properties of CNT. High energy ball milling is commonly used to produce Aluminum-CNT composite with providing good dispersion and mechanical properties. Interface between Aluminum and CNT has been improved by this method. High energy ball milling which includes continuously fracturing and cold welding, it may damage CNT structure and reduce its optimal properties. Impurities may also come from ball mill due to impact of high energy. Iron as impurity to the powder was found as result from continuous contact between Aluminum powder and balls [1-2].

Ultrasonication technique to disperse CNT was used by S. Simoes et al. CNT was well dispersed by this technique. But interface between Aluminum and CNT from this technique was not as strong compared to high energy ball milling as it shows by their mechanical properties behavior [3-4]. Formation of aluminum carbide, Al<sub>4</sub>C<sub>3</sub> seems to be important factor to have better interface and mechanical properties. Aluminum carbide is able to transfer stress from matrix into reinforcement particle which leads to higher mechanical properties. Formation of aluminum carbide mainly depends on temperature process. On some cases, aluminum carbide was found after high energy ball milling [1-3].

Alumix 431D has shown high sinterability and mechanical properties by reaching sintering density for over than 97% relative. Alumix 431D is premix powder with aluminum as base powder with additional Zn, Mg and Cu. This powder has similarity of chemical composition with AA7075. This powder is also heat treatable, increasing on mechanical properties was shown after T6 heat treatment. Intermetallic phases which responsible for mechanical properties of this powder are MgZn<sub>2</sub> and Al-CuMg [5-6]. The purpose of this research to investigate effect of ultrasonication dispersion methods of CNT to Alumix 431D.

### 2. Experimental method

Premix Al-5.5Zn-2.5Mg-0.5Cu powder was used as matrix in this research. This powder was named ALUMIX431D and produced by Ecka Granules, Germany. Multi walled CNT was used as reinforcement particles and produced by Sigma Aldrich. This MW CNT has diameter of 5.5nm and length of 6-9 nm. Ultrasonication was done with using Ultrasonication equipment with 0.5 wave length for 30 min under ethanol for 100 ml. After ultrasonication, to maximize dispersion of MW CNT on aluminum powder, milling was done with 10mm hardened steel ball with 5:1 ball to powder ratio under argon gas and 300 rpm for 1h.

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To produce high density materials, compaction followed by sintering under argon gas were carried out. Compaction was done at 700 MPa to produce green body materials with 20 mm diameter. Vickers hardness was used to measure mechanical properties of sintered materials with 5lb load and 10 second holding time. SEM-EDS and X-ray Diffraction analysis were used to characterize the structural analysis of the samples. The diffraction patterns of samples were achieved by X-ray diffraction analysis. XRD patterns were obtained using Rigaku Diffractometer with Cu K $\alpha$  radiation ( $\lambda = 0.154$  nm) in the range of  $2\theta$  from 20-80° by the step and scanning speed for 0.02° and 5°C/min. To characterize thermal properties, differential scanning calorimetry (DSC) was carried out. DSC was measured using SDT-Q600 DSC manufactured by TA instruments. Powder was heated at a rate of 10°C/min under flowing nitrogen (100ml/min).

### 3. Results and Discussions

Morphology of MWCNT powder is shown by Fig. 1. Needle-like morphology is expected for CNT powder. Raman spectroscopy of CNT before and after ultrasonication and low energy ball milling are shown on Fig. 2. There is no big difference on raman shifts on both cases, this indicates that ultrasonication and low energy ball milling did not damage much on carbon structures on MW CNT. Ratio of  $I_D$  and  $I_G$  of non-damaged Al-CNT composite will be approximately less than 1 [4]. In case of high energy ball milling which is common method to disperse CNT on aluminum powder will give different on raman shift, because high energy ball milling for long time is able to damage

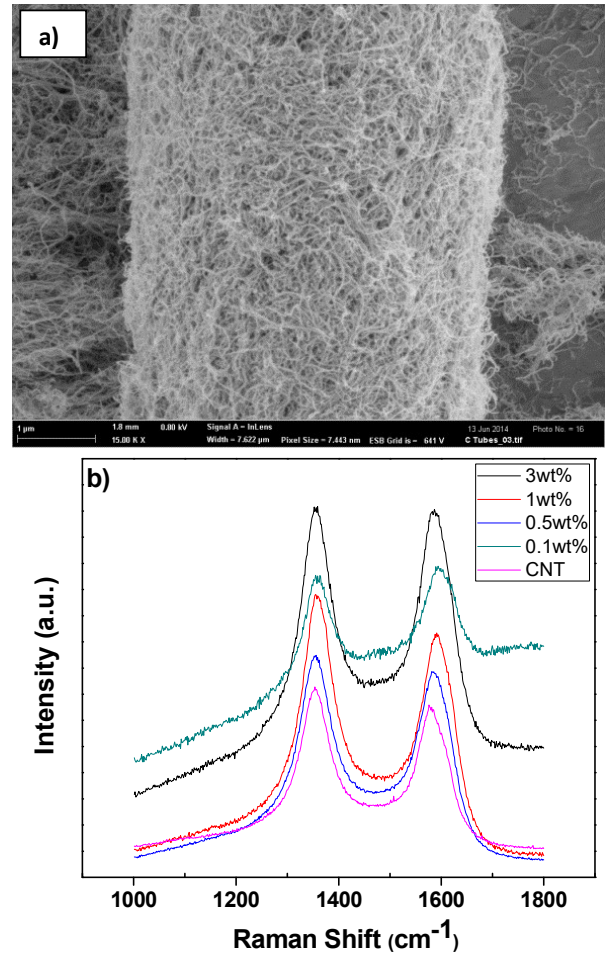


Fig. 1. a) SEM figure of MWCNT powder and b) Raman spectroscopy of as received CNT and after ultrasonication

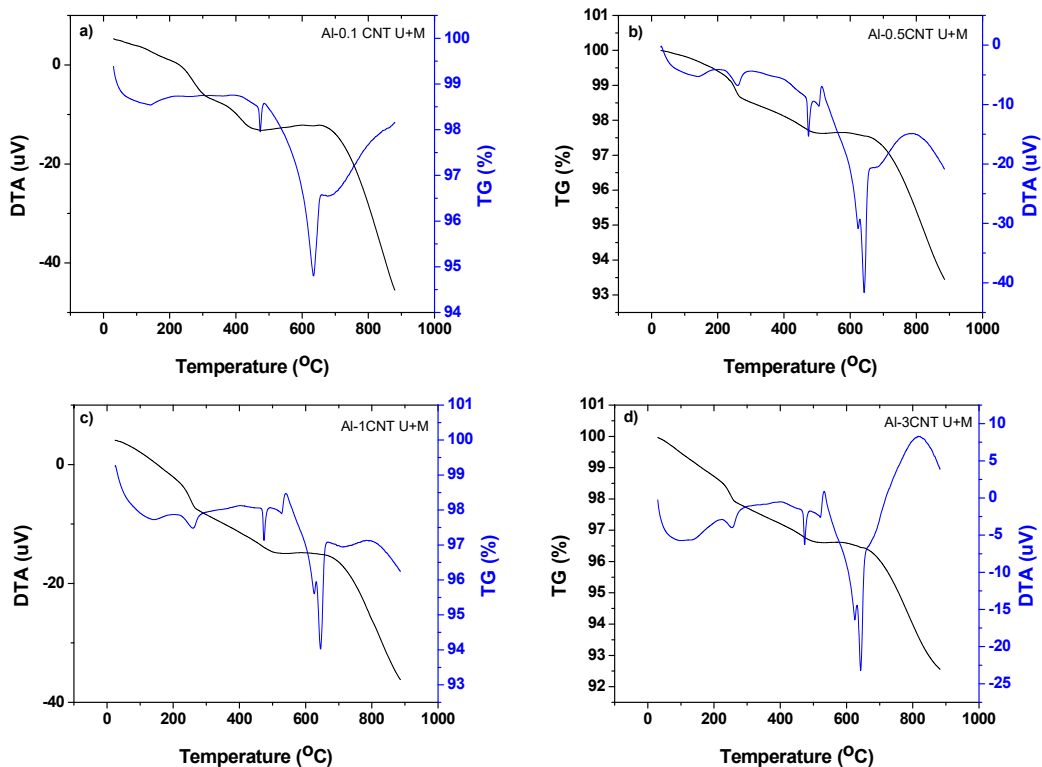


Fig. 2. DSC-TGA of Alumix 431D with a) 0.1wt.%, b) 0.5wt.%, c) 1wt.% and d) 3wt.% CNT after ultrasonication

carbon structure on CNT. But ultrasonication technique is also able to damage CNT structure if done for longer time with high frequency which leads to shortened CNT. After ultrasonication, CNT is able to deagglomerate if the processing time is not sufficient. At least 15 mins ultrasonication for CNT dispersion is required according to Simoes et al results [3].

To estimate sintering temperature, DSC-TGA was carried out as shown at Fig. 2. DSC line for all powders show similar trend with having 2 endothermic peaks. These peaks represent eutectic point for certain materials. First endothermic peak at 450°C indicates eutectic point of Al-Mg followed by peak at 630°C which is eutectic point of Al-Zn. Due to small amount of Cu content, it is supposed to have eutectic point at 520°C. From this data of view, sintering temperature is able to estimate. To be able to have liquid phase, sintering temperature must be higher than certain eutectic point. And according to thermogravimetric line (black line), all composites have gone through mass reduction with increasing temperature. The first thermogravimetric line is around 450°C, it is supposed to be lubricant removal. Lubricant is supposed to be removed before reaching sintering temperature otherwise, it can hinder diffusion process between elements during sintering. 580°C sintering temperature was chosen because at that temperature, this alloy powder is able to form optimum liquid phase to be able to achieve optimum sintering density without formation of particle coarsening as shown by previous results [7].

Fig. 3 shows microstructures of Alumix 431D with MW CNT after sintering. Pores are larger with increasing content of MW CNT. This is assumed to agglomeration of MW CNT on grain boundary. High content of agglomeration means lower sintering density and mechanical properties. It seems that 0.1 up to 0.5wt% of MWCNT are suitable to have acceptable sintering properties. Intermetallic phases from other elements were shown inside and on grain boundary. This powder has possibility to form some intermetallic phases such as  $MgZn_2$ ,  $Al_2Cu$ , and  $AlMgCu$ . Aluminum, Zinc, and Copper are homogeneously dispersed throughout the surface. Magnesium tends to be near grain boundary and pores, it is expected as spinel,  $MgAl_2O_4$  from reaction between Magnesium and Aluminum Oxide,  $Al_2O_3$  will decompose close to grain boundary or pore. From EDS analysis as well, there are impurities from this composite such Fe and Sn. Fe is a common impurity on Aluminum alloys or it might from ball milling process. Sn seems to be impurity which added by manufacturer, as it also appears from another results Fig. 4 [7].

After ultrasonication and low energy ball milling, aluminum is still dominant peak as shown by XRD pattern at Fig. 5a. Aluminum carbide,  $Al_4C_3$  which is usually formed after high energy ball milling is not detected with this XRD. At some cases, even after high energy ball milling, aluminum carbide did not form. It is assumed that formation of aluminum carbide after mechanical milling depends on high the content of CNT and how long the process. Formation of  $Al_4C_3$  is important as bridge between

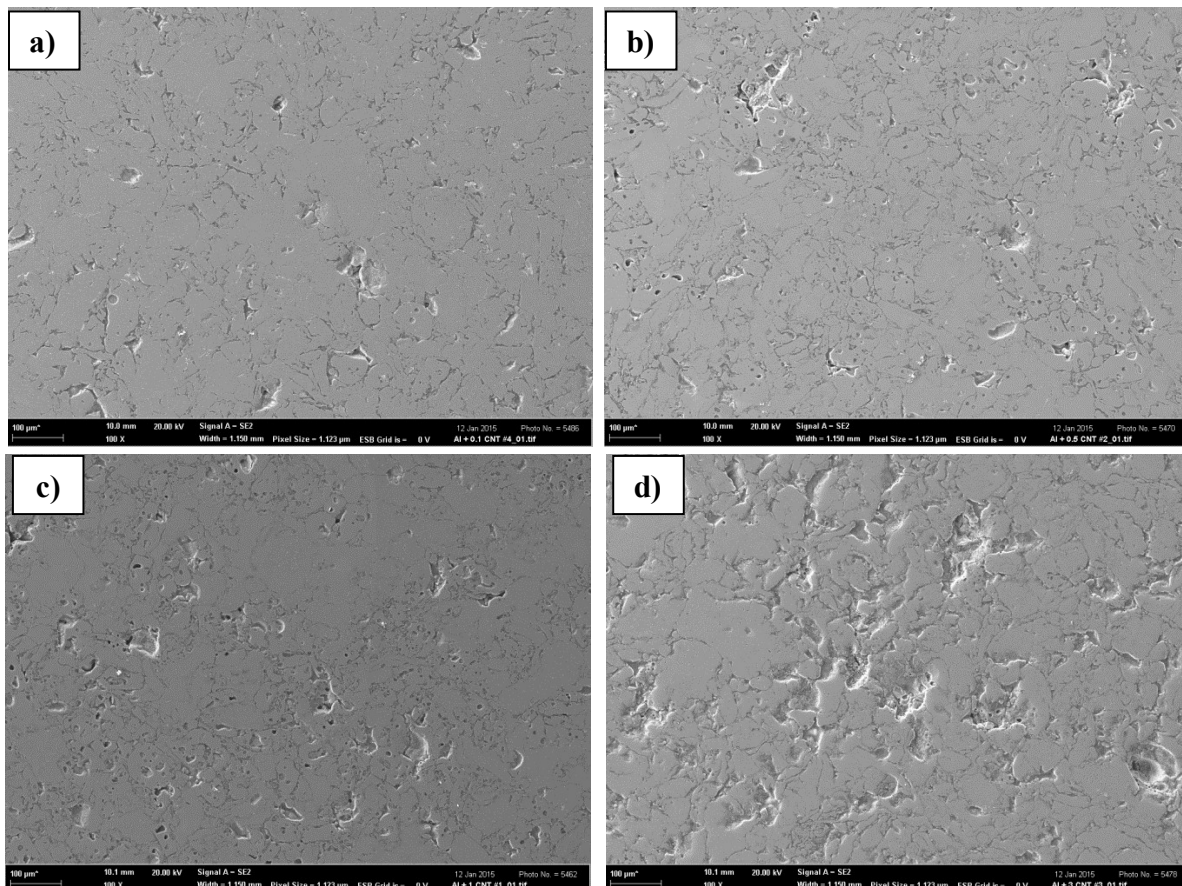


Fig. 3. Microstructures after Sintering a) 0.1wt.% CNT b) 0.5wt.% CNT c) 1wt.% CNT d) 3wt.% CNT

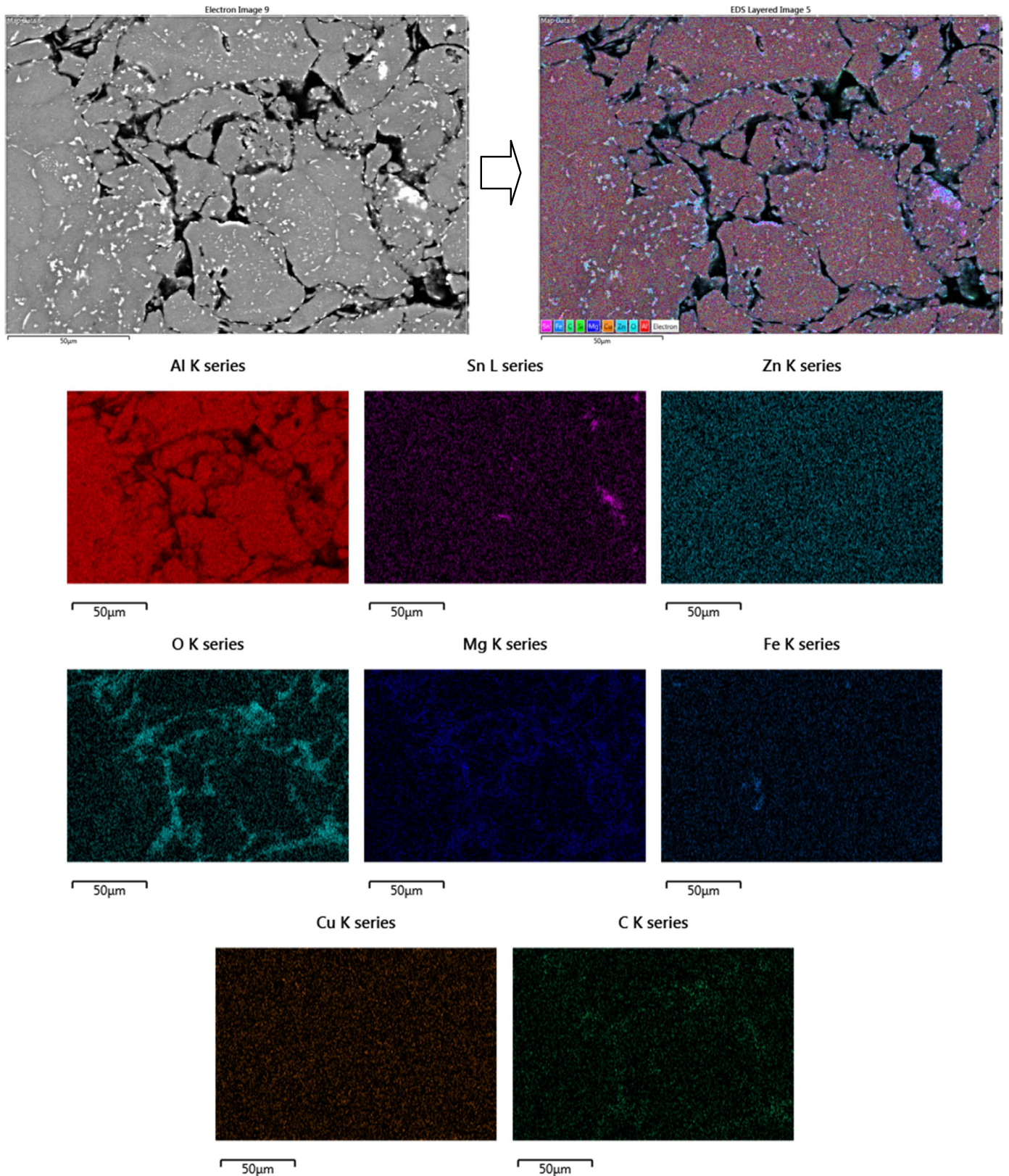


Fig. 4. EDS analysis of sintered Alumix 431D with 0.1wt.% CNT

Aluminum matrix with CNT to be able to transfer the load from matrix to obtain higher mechanical properties. Microstructures also represent mechanical properties of this material as shown by Fig. 5b. Higher content of agglomeration of MWCNT reduced hardness after sintering. And it seems that difficulty may arise

when content of MWCNT increased for more than 0.5wt.%. 49 HV was obtained as highest hardness for this composite powder with 0.5wt.% MWCNT and reduce with increasing amount of MWCNT.

#### 4. Conclusions

Ultrasonication technique successfully dispersed MWCNT on Alumix 431D. Ultrasonication and low energy ball milling powders have similar raman characteristics, this indicates that the dispersion process did not affect on carbon structure of MWCNT. According to microstructures after sintering of these nanocomposites, it seems that there is agglomeration especially for material with MWCNT content for more than 0.5wt%. This agglomeration leads to lower sintering density and mechanical properties.

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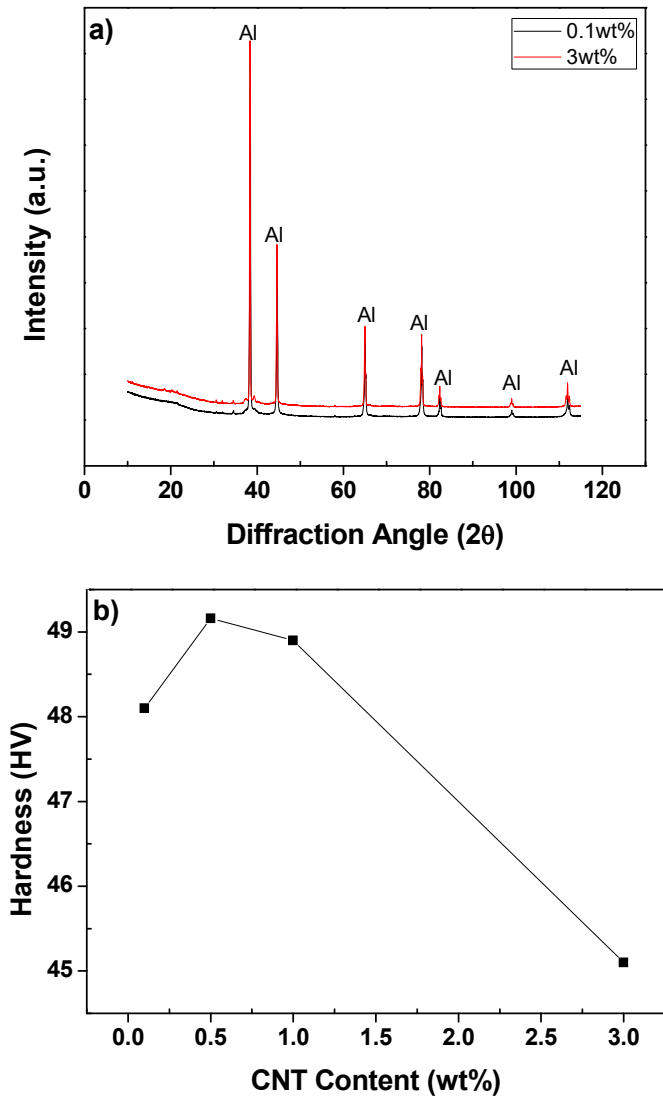


Fig. 5. a) X-Ray Diffraction (XRD) and b) Vickers Hardness of sintered Alumix 431D with MWCNT