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High Pressure Sintering of Al_2O_3-CNT Nanocomposites

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Article Information

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In this study, Al_2O_3-CNT nanocomposites were sintered at 800-1000 °C under 5 GPa for 10 minutes using two different powders as starting material, γ-Al_2O_3 and AlOOH. The effect of transition alumina powders on the properties of the high pressure sintered Al_2O_3-CNT nanocomposites were studied. Al_2O_3 ceramics have been widely used for structural, electrical and optical applications. Carbon nanotubes (CNTs) have recently been considered as reinforcing elements in ceramic matrix composites due to their unique mechanical properties. Reactive pressure sintering at low temperatures took place during the fabrication of CNT reinforced ceramic composites to avoid the agglomeration of carbon nanotubes. The hardness and densities were investigated and correlated with the microstructure of the dense materials. SEM analysis was used to determine the CNT distribution in the matrix, the morphology and size of the particles.

Alumina is one of the most widely used ceramic materials owing to its relatively high hardness (15-22 GPa), good oxidation resistance and chemical stability with iron or steel [1]. However applications have been limited due to its low fracture toughness. In most cases, low addition of carbon nanotubes as reinforcement improves the fracture toughness, on the other hand it degrades the hardness or strength. Carbon nanotubes (CNTs) have recently been considered as reinforcing elements in ceramic matrix composites due to their unique mechanical properties [2-4].

Carbon nanotubes tend to be agglomerated each other owing to a strong van der Waals attractive force. There have been several attempts to fabricate carbon nanotube reinforced ceramic matrix nanocomposites. To create functional CNT-reinforced composites, two main problems have to be overcome:

1. Homogeneous dispersion of nanotubes in the matrix
2. Generation of strong interfacial bonding between CNTs and the matrix[1, 5-7].

Dense and nanostructure alumina ceramics are widely used in practical applications due to its outstanding mechanical, electrical and optical properties. It is difficult to obtain a fully dense ceramic with nanocrystalline grain size. The transition paths and temperatures vary depending on the particle size, chemical homogeneity, heating rate, and water vapor pressure [8, 9].

However attempts by applying high sintering pressure proxed useful in achieving higher densities with nanocrystalline microstructure [10]. Transition alumina powders normally have the highest surface area and smallest crystalline size, e. g. 15 ~ 70 nm [11].

The objective of this study was to investigate the effect of transition alumina powders on the properties of the high pressure sintered Al_2O_3-CNT nanocomposites.

Experimental Procedure

Two different alumina (γ-Al_2O_3 and AlOOH) powder were used as starting materials in this study. The first starting powder consists of spherical γ-Al_2O_3 (JSPDS Card No: 00-046-1215) phase with an average particle size of 20 nm (Plasma & Ceramic Technologies Ltd.-Latvia) and specific surface area of 50 m^2/g in the granulated state. The chemical composition of the starting γ -Al_2O_3 powder was given in Table 1. The second material used as a starting material for Al2O3-CNT composites is AlOOH obtained by hydrothermal processing.

Preperation of CNT reinforced γ-Al_2O_3 powder. A suspension of γ-Al_2O_3 powders were prepared. Ethanol is used as a solvent and the weigth percentage of alumina in the suspension was 5 %. A suspension of CNT (20 nm in diameter and 10-25 µm in length), (Thomas Swan Co. Ltd., UK), was also prepared in ethanol in the weigth percent of 1 %. Both suspensions were ball milled and ultrasonicated for 2 hours. The CNT suspension was added to the alumina suspension dropwisely. Then the composite powders were obtained after the drying process.

Preperation of CNT reinforced AlOOH (boehmite) powder. Aluminum acetate powders ($2Al(OH)(C_2H_3O_2)_2$) and CNT were mixed. The mixture was stirred indistilled water. 17 ml 25 % ammonia solution (NH_3) was then added to the mixture to adjust the

Chemical impurities [ppm]	Fe	Si	Na
According to supplier's analytical certificate	<1000	<200	< 1000

Table 1. Chemical impurities of the starting γ-Al_2O_3 powder

pH above 9 in order to obtain fine boehmite particles. The reason for the addition of such quantity of CNT in the mixture was the aim of obtaining 1 % reinforced final structure which was going to be in the form of CNT reinforced alumina (Al_2O_3) after the heat treatment. The resultant mixture was transferred into a teflon lined autoclave approximately filling 80 % of the 500 ml total volume and the hydrothermal synthesis was performed at 200 °C for 2 hours in a high pressure autoclave. The pressure of the autoclave was approximately 1 MPa. The autoclave was then cooled to room temperature. The resultant solution was vacuum filtered for four times to remove water soluble impurities. Then, the filtered solution was dried in an autoclave at 110 °C. Finally, CNT reinforced boehmite powder was obtained.

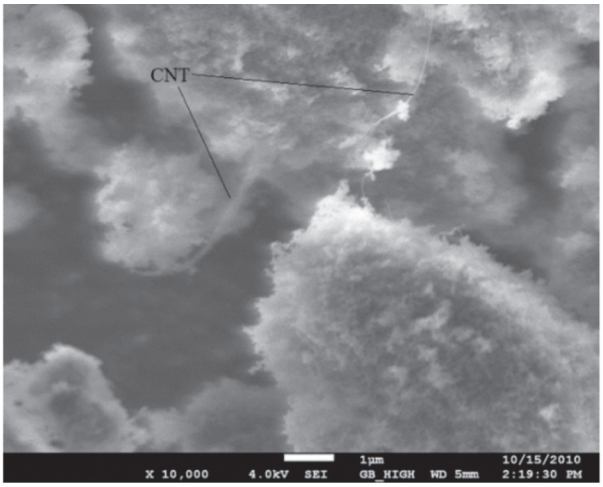
Sintering Conditions of Al_2O_3–CNT Nanocomposites. γ-Al_2O_3 or AlOOH-CNT powder containing no additives was uniaxially cold pressed at 20 MPa into cylinders 20 mm in diameter and 10mm in height. All the green compacts were pellets of 4 g. Green compact was encapsulated in a cube die made of pyrophyllite. The CNT-Al_2O_3 bodies were fabricated in cubic anvil high pressure (5 GPa) and at a temperature of 800 °C for 10 minutes. Phase analysis of the sintered samples was carried out by X-ray diffraction (XRD). Grain sizes were estimated from high-resolution scanning electron micrographs taken from fracture surfaces. Microhardness was determined on the polished surfaces under the load of 500 g.

Result and Discussion

The SEM micrograph and the XRD spectrum of the AlOOH-CNT composite powder obtained by hydrothermal processing are presented in Figure 1. As can be seen in the figure, AlOOH and CNT are obtained.

The SEM micrograph and the XRD spectrum of Al_2O_3-CNT nanocomposites (γ-Al_2O_3 used as starting material) sintered at 800 °C under 5 GPa for 10 minutes are presented in Figure 2. γ-Al_2O_3 transformed to α-Al_2O_3 in this sintering conditions (Figure 2b). After sintering, average grain size of α-Al_2O_3-CNT is 150 nm and agglomeration of CNT in the microstructure can be seen in Figure 2a. An alumina hydrate, AlOOH, was found in the samples sintered at 5 GPa and 800 °C as can be seen in Figure 2b. This phase is caused by trapped water or surface OH groups which forms the hydrate phase during low temperature sintering.

The SEM micrograph and the XRD spectrum of Al_2O_3-CNT nanocomposites (AlOOH used as starting material) sintered at 800 °C under 5 GPa for 10 minutes are presented in Figure 3. The average grain size of AlOOH-CNT composite is 3 µm. Accord-



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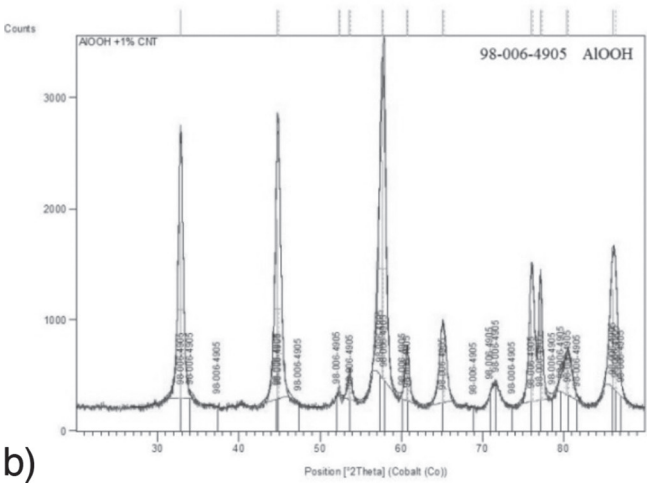
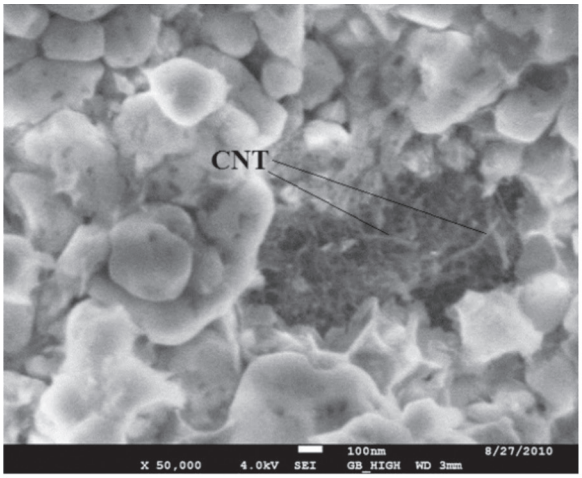


b)

Figure 1. AlOOH-CNT composite powder obtained by hydrothermal processing, a) SEM micrograph, b) XRD spectrum



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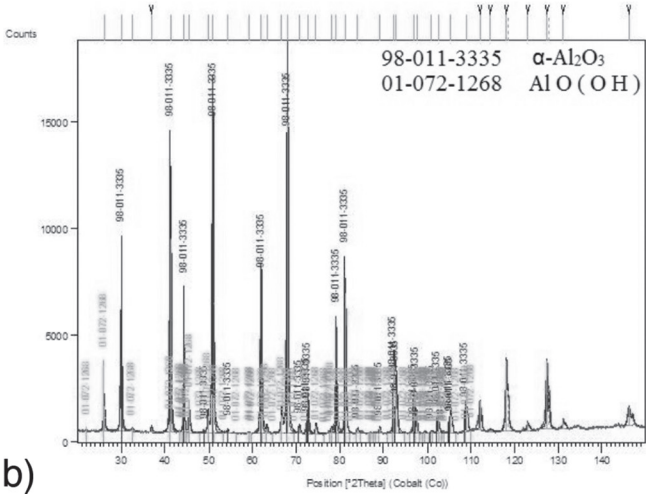
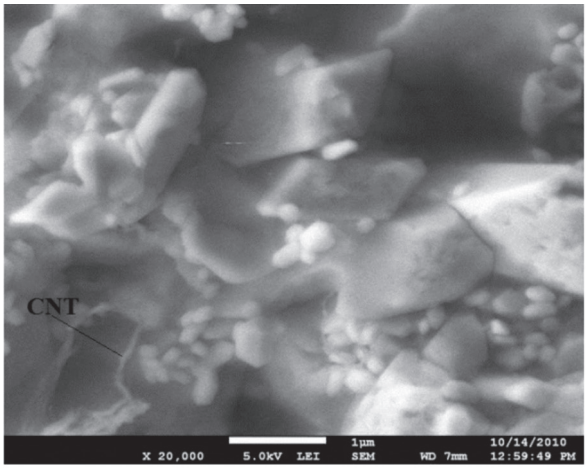


b)

Figure 2. Al_2O_3-CNT nanocomposite (γ-Al_2O_3 used as starting material) sintered at 800 °C under 5 GPa for 10 minutes, a) SEM micrograph, b) XRD spectrum



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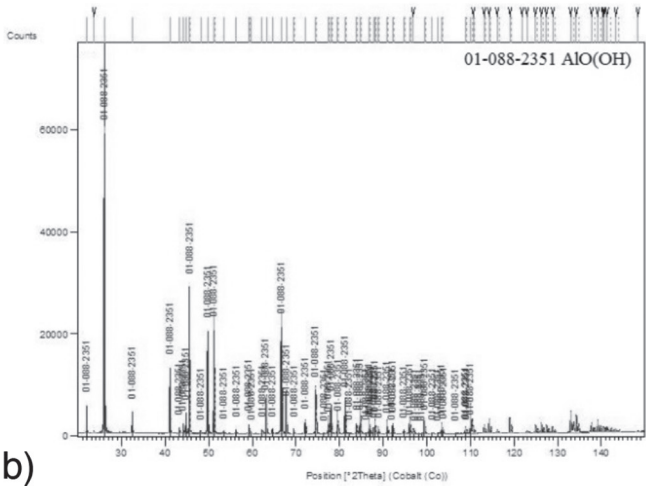
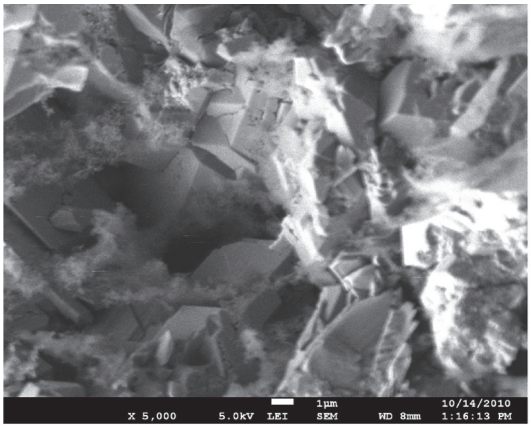


b)

Figure 3. Al_2O_3-CNT nanocomposite (AlOOH used as starting material) sintered at 800 °C under 5 GPa for 10 minutes, a) SEM micrograph, b) XRD spectrum



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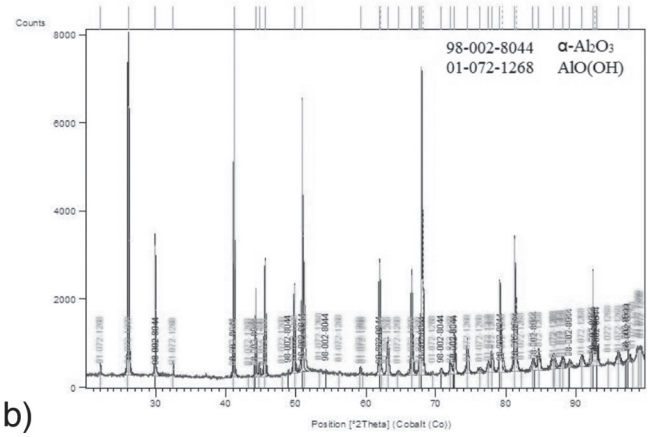

b)

Figure 4. Al_2O_3-CNT nanocomposite (AlOOH used as starting material) sintered at 1000 °C under 5 GPa for 10 minutes, a) SEM micrograph, b) XRD spectrum

ing to XRD spectrum, phase transformation did not occur.

The SEM micrograph and the XRD spectrum of Al_2O_3-CNT nanocomposites (AlOOH used as starting material) sintered at 1000 °C under 5 GPa for 10 minutes are presented in Figure 4. Average grain size of α-Al_2O_3 -CNT composite is 5 µm. According to XRD spectrum, phase transformation occured, the presence of the α-Al_2O_3 could be seen. In the presence of the hydrates, it is difficult to control grain growth according to S. C. Liao et al. [10].

Sintering γ-Al_2O_3-CNT at 5 GPa/800 °C for 10 minutes resulted in compacts with a theoretical density of 95.40 % and with a hardness of 882.62 kg × mm^{-2}.

Sintering AlOOH-CNT at 5 GPa/800 °C for 10 minutes resulted in compacts with a theoretical density of 84.34 % and with a hardness of 472.95 kg × mm^{-2}.

Conclusions

- Dense and crack free compacts were obtained by high pressure processing at 5 GPa/800-1000 °C for 10 minutes.
- CNT agglomarated in γ-Al_2O_3 powder prepared with ethanol, whereas CNT was dispersed homogenously in AlOOH prepared by hydrothermal processing.
- The theoretical density of γ-Al_2O_3 –CNT nanocomposite sintered at 5 GPa/800 °C for 10 minutes is 95.40 %, while the theoretical density of AlOOH-CNT composite sintered at the same conditions is 84.34 %.
- An avarage grain size of 150 nm is obtained in the γ-Al_2O_3–CNT nanocomposite sintered at 5 GPa/800 °C for 10 minutes. But in the AlOOH–CNT composite sintered in the same conditions, the average grain size is 3 µm.
- The microhardness of γ-Al_2O_3–CNT nanocomposite is almost the two times the value of microhardness of AlOOH–CNT composite. The reason for this is the difficulty of the control of grain growth due to the presence of the hydrates.

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Abstract

Hochdrucksintern von Al_2O_3-CNT Nanokompositen. In der diesem Beitrag zugrunde liegenden Studie wurden Al_2O_3-CNT Nanokomposite bei 800 bis 1000 °C unter 5 GPa für 10 Minuten unter Verwendung von zwei verschiedenen Pulvern γ-Al_2O_3 and AlOOH gesintert. Die Auswirkung der Übergangsaluminiumpulver auf die Eigenschaften der Hochdruck-gesinterten Al_2O_3-CNT Nanokomposite wurde untersucht. Al_2O_3 Keramiken werden breitflächig für struturelle, elektrische und optische Anwendungen eingesetzt. Carbon-Nanoröhrchen (Carbon Nanotubes (CNTs)) finden in der letzten Zeit als Verstärkungselemente in Keramikmatrixkompositen aufgrund ihrer einzigartigen mechanischen Eigenschaften Berücksichtigung. Während der Fertigung der CNT-verstärkten Keramikkomposite wurde das Reaktivdrucksintern bei niedrigen Temperaturen angewendet, um die Agglomeration der Carbon-Nanoröhrchen zu vermeiden. Die Härte und die Dichte wurden untersucht und mit der Mikrostruktur der dichten Materialien korreliert. Eine REM-Analyse wurde durchgeführt, um die CNT-Verteilung in der Matrix sowie die Morphologie und die Größe der Partikel zu untersuchen.

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