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Mechanical properties and consolidation of a nanocrystalline Al_2O_3 -reinforced Ni composite from mechanically synthesized powders by rapid sintering

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Nano-powders of Ni and Al_2O_3 were synthesized from NiO and Al powders by high energy ball milling. The nanocrystalline Al_2O_3 - reinfored composite was consolidated by a pulsed current activated sintering(PCAS)) method within 2 minutes from mechanically synthesized powders of Al_2O_3 and 3Ni. The average hardness and fracture toughness values obtained were 690 kg/mm^2 and 6.1 MPa·m$^{1/2}$, respectively.

Key words: Rapid sintering, Composite, Nanophase, Mechanical properties, Ni-Al_2O_3.

Introduction

It is increasingly being recognized that new applications for materials require functions and properties that are not achievable with monolithic materials. Metal matrix composites combine metallic properties (ductility and toughness) with ceramic characteristics (high strength and modulus), leading to greater strength in shear and compression and to higher service temperature capabilities. The attractive physical and mechanical properties that can be obtained with metal matrix composites, such as high specific modulus, strength-to-weight ratio, fatigue strengh, temperature stability and wear resistance, have been documented extensively [1-6]. Therefore, metal matrix composites are recognized as candidates for aerospace, automotive, bio-material and defense applications.

Traditionally, discontinuously reinforced metal matrix composites have been produced by several processing routes such as powder metallurgy, spray deposition mechanical alloying, various casting techniques and SHS(self-propagating high temperature synthesis). All these techniques are based on the addition of ceramic reinforcements to the matrix materials which may be in molten or powder form. One of these techniques, SHS developed by Merzhanov and coworker [7, 8] in the late 1960s, refers to a process in which materials with a sufficiently high heat of formation are synthesized in a combustion wave, which after ignition, spontaneously propagates throughout the reactants and converts them into the product. SHS is extremely attractive, producing a high- purity product due to the volatilization of low boiling point impurities at elevated temperature, and high productivity due to very high reaction rates.

Nanocrystalline materials have received much attention as advanced engineering materials with improved physical and mechanical properties [9, 10]. Since nanomaterials possess high strength, high hardness, excellent ductility and toughness, much attention has been paid to their application [11, 12]. Recently, nanocrystalline powders have been made by a thermochemical and thermomechanical process called as the spray conversion process (SCP), involving co-precipitation and high energy milling [13-15]. However, the grain size in sintered materials becomes much larger than that in the pre-sintered powders due to the rapid grain growth during a conventional sintering process. Therefore, even though the initial particle size is less than 100 nm, the grain size increases rapidly up to 500 nm or larger during conventional sintering [16]. So, controlling grain growth during sintering is one of the keys to the commercial success of nanostructured materials. In this regard, the pulsed current activated sintering method which can make dense materials within 2 minutes, has been shown to be effective in achieving this goal [17, 18].

The purpose of this study is to produce dense nanocrystalline Ni-Al_2O_3 composite within 2 minutes from mechanically synthesized powders using this pulsed current activated combustion method and to evaluate the mechanical properties of the resulting composite (hardness and fracture toughness).

Experimental Procedure

Powders of 99% NiO (–325 mesh, Alfa) and 99% pure Al (–325 mesh, Cerac, Inc) were used as a starting materials.

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The NiO and Al powder mixtures were first milled in a for High-energy ball mill, Pulverisette-5 planetary mill at 250 rpm 10 h. Tungsten carbide balls (8 mm in diameter) were used in a sealed cylindrical stainless steel vial with an argon atmosphere. The weight ratio of ball-to-powder was 30 : 1. Milling resulted in a significant reduction of the grain size.

The grain sizes of Ni and Al_2O_3 were calculated by Suryanarayana and Grant Norton's formula [19] :

$$B_r\ (B_{crystalline} + B_{strain})\ \cos\theta = k\lambda / L + \eta\sin\theta \qquad (1)$$

where B_r is the full width at half-maximum (FWHM) of a diffraction peak after instrumental correction; $B_{crystalline}$ and B_{strain} are the FWHM caused by a small grain size and the internal stress, respectively; k a is constant (with a value of 0.9); λ is the wavelength of the X-ray radiation; L and η are the grain size and internal strain, respectively; and θ is the Bragg angle of the diffraction peak. The parameters B and B_r follow a Cauchy form with the relationship: $B = B_r + B_s$, where B and B_s are the FWHM of the broadened Bragg peak and a Bragg peak from a standard sample, respectively.

After milling, the mixed powders were placed in a graphite die (outside diameter, 45 mm; inside diameter, 20 mm; height, 40 mm) and then introduced into the pulsed current activated sintering system made by Eltek in South Korea, shown schematically in Fig. 1. The four major stages in the synthesis are as follows. Stage 1-Evacuation of the system. Stage 2- Application of uniaxial pressure. Stage 3-Heating of the sample by an pulsed current. Stage 4-Cooling of the sample. The process was carried out under a vacuum of 40 mtorr(5.33 Pa).

The relative densities of the synthesized samples measured by the Archimedes method were over 95% of the theoretical value. Microstructural information was obtained from product samples which were polished at room temperature. Compositional and micro structural analyses of the products were made through X-ray diffraction (XRD) and scanning electron microscopy (SEM) with energy dispersive X-ray analysis (EDAX). Vickers hardness was measured by performing indentations at a load of 5 kg and a dwell time of 15 s on the sintered samples.

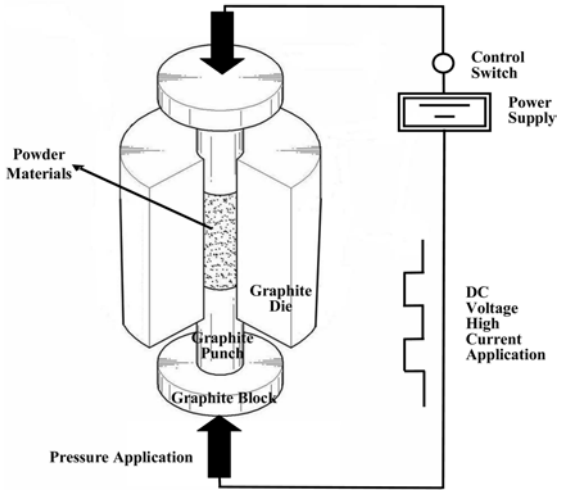

Fig. 1. Schematic diagram of the apparatus for pulsed current activated sintering.

Results and Discussion

The interaction between 3NiO and 2 Al, i.e. :

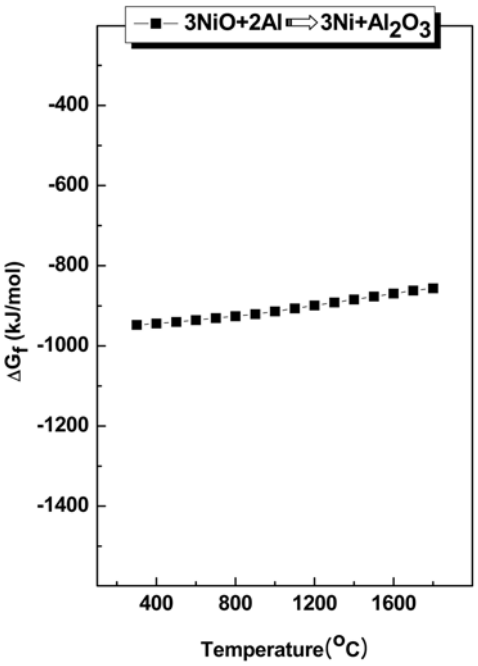

Fig. 2. Temperature dependence of Gibbs free energy variation by interaction of nickel oxide and aluminium.

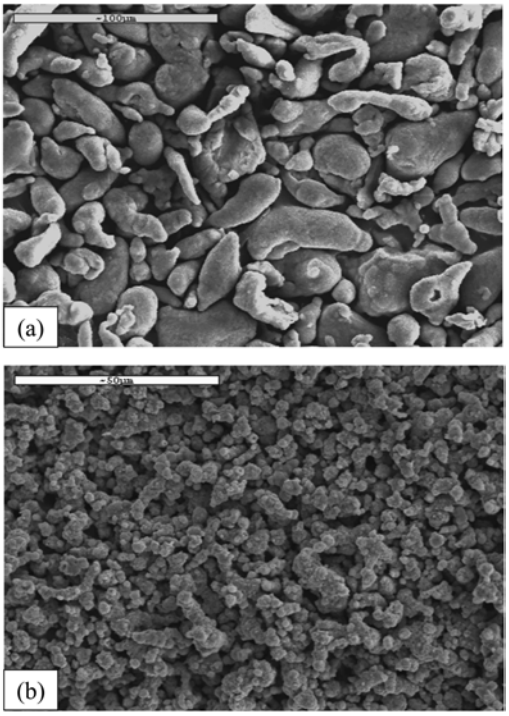

Fig. 3. Scanning electron microscope images of the raw materials : (a) aluminum powder, (b) nickel oxide powder.

$$3NiO + 2Al \rightarrow 3Ni + Al_2O_3 \quad (2)$$

is thermodynamically feasible as shown in Fig. 2.

Fig. 3 shows the SEM images of the raw materials used. The powders have a spherical shape. A X-ray diffraction patten result of the high energy ball milled powders is shown in Fig. 4(c). The reactant powders of NiO and Al were not detected but the products, Ni and Al_2O_3, were detected. From the above result the mechanical synthesis goes to completion during the milling. The average grain sizes of the Ni and Al_2O_3 measured by Suryanarayana and Grant Norton's formula are about 46 nm and 42 nm, respectively. A SEM image and X-ray mapping of the milled powders are shown in Fig. 5. The powders are very fine and the products (Ni, Al_2O_3) are uniformly distributed from the X-ray mapping results [Fig. 5 (b), (c) and (d)]. The variations in shrinkage displacement and temperature of the surface of the graphite die with heating time during the processing of the Ni and Al_2O_3 system are shown Fig. 6. As the pulsed current was applied the specimen showed some thermal expansion and the shrinkage displacement increased with temperature up to about 950 °C, but then abruptly increased at about 1000 °C. A X-ray diffraction pattern of a sample heated to 1200 °C is shown in Fig. 7. Ni and Al_2O_3, were detected. The structural parameters, i.e. the average grain sizes of Ni and Al_2O_3

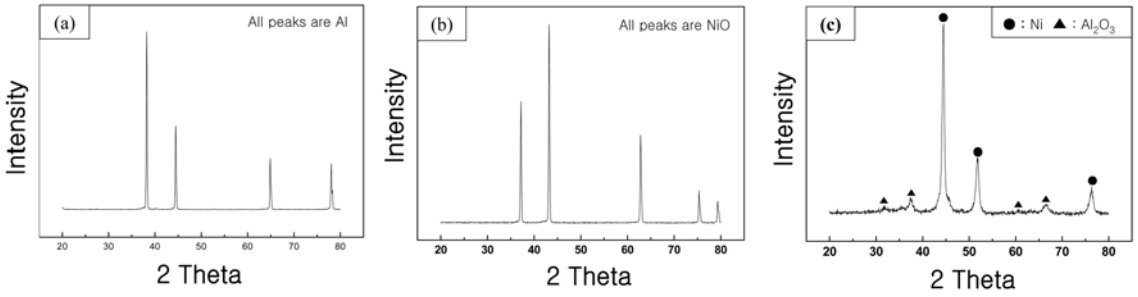

Fig. 4. XRD patterns of raw materials and mechanically syntheized powder : (a) Al, (b) NiO and (c) mechanically synthesized powder.

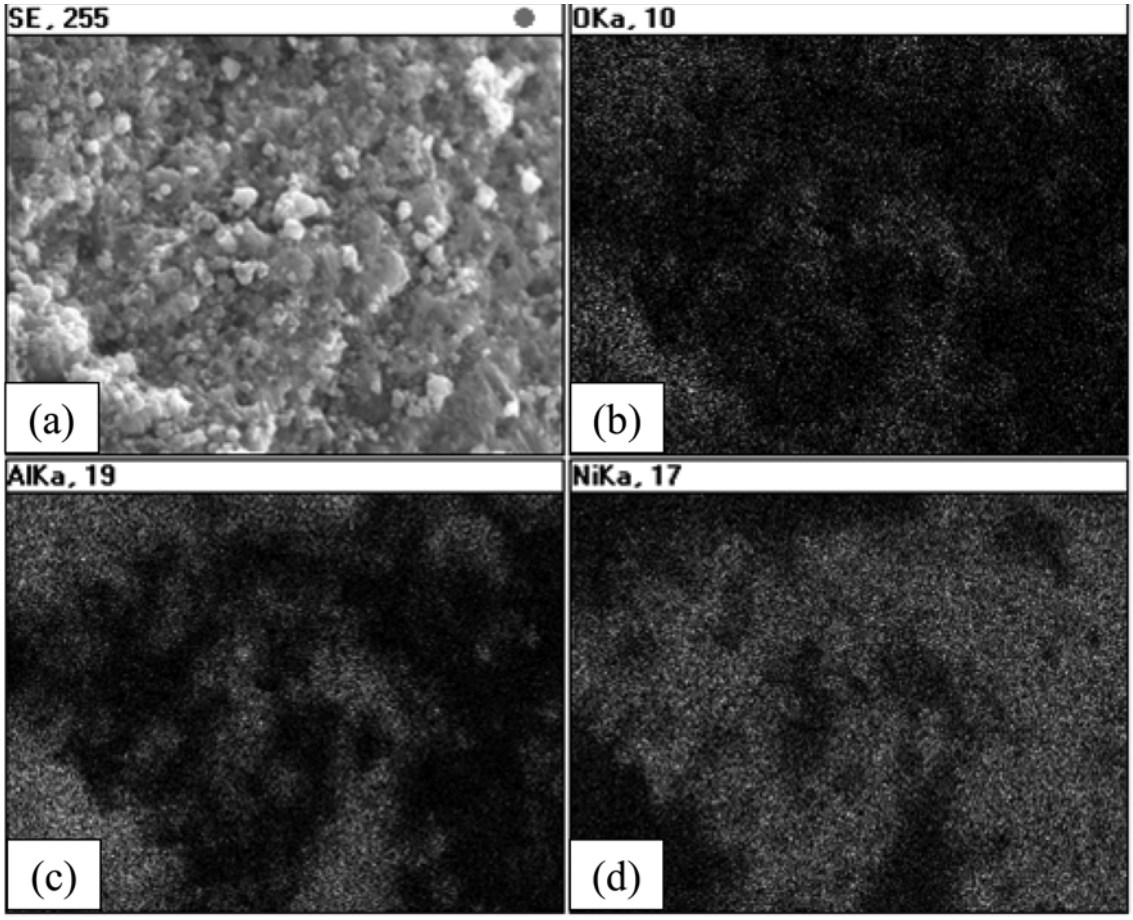

Fig. 5. SEM image and X-ray maps of Ni and Al_2O_3 powders : (a) SEM image, (b) oxygen mapping, (c) aluminum map, (d) nickel map.

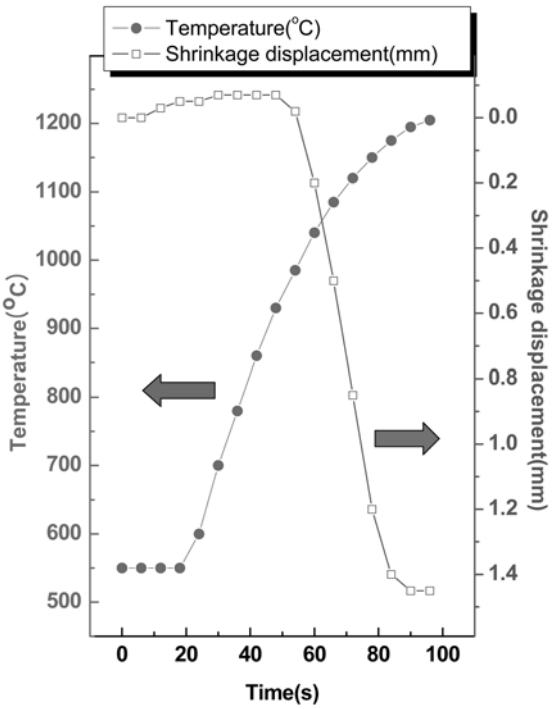

Fig. 6. Variation of temperature and shrinkage displacement with heating time during pulsed current activated sintering of 3Ni + Al_2O_3 (80 MPa, 2800A).

obtained from the X-ray data using Suryanarayana and Grant Norton's formula, were 690 nm and 46 nm, respectively. Fig. 8 shows the SEM image of sample heated to 1200 °C and X-ray maps. The dark color and grey color regions are Al_2O_3 and Ni, respectively. The Al_2O_3 particles were well distributed in the matrix, as ascertained by the SEM image and X-ray maps shown in Fig. 8.

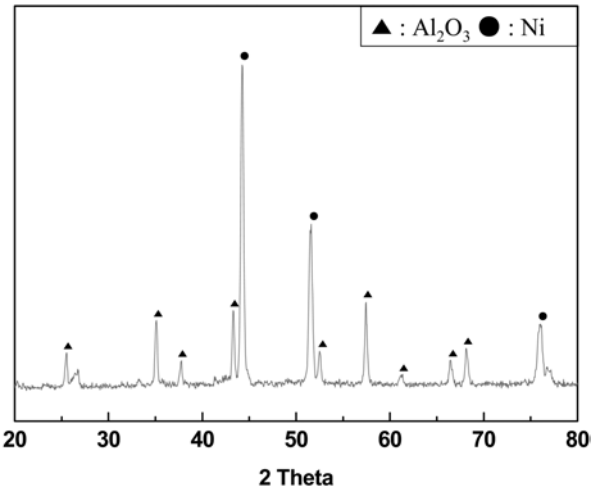

Fig. 7. XRD pattern of the Ni-Al_2O_3 composite heated to 1200 °C.

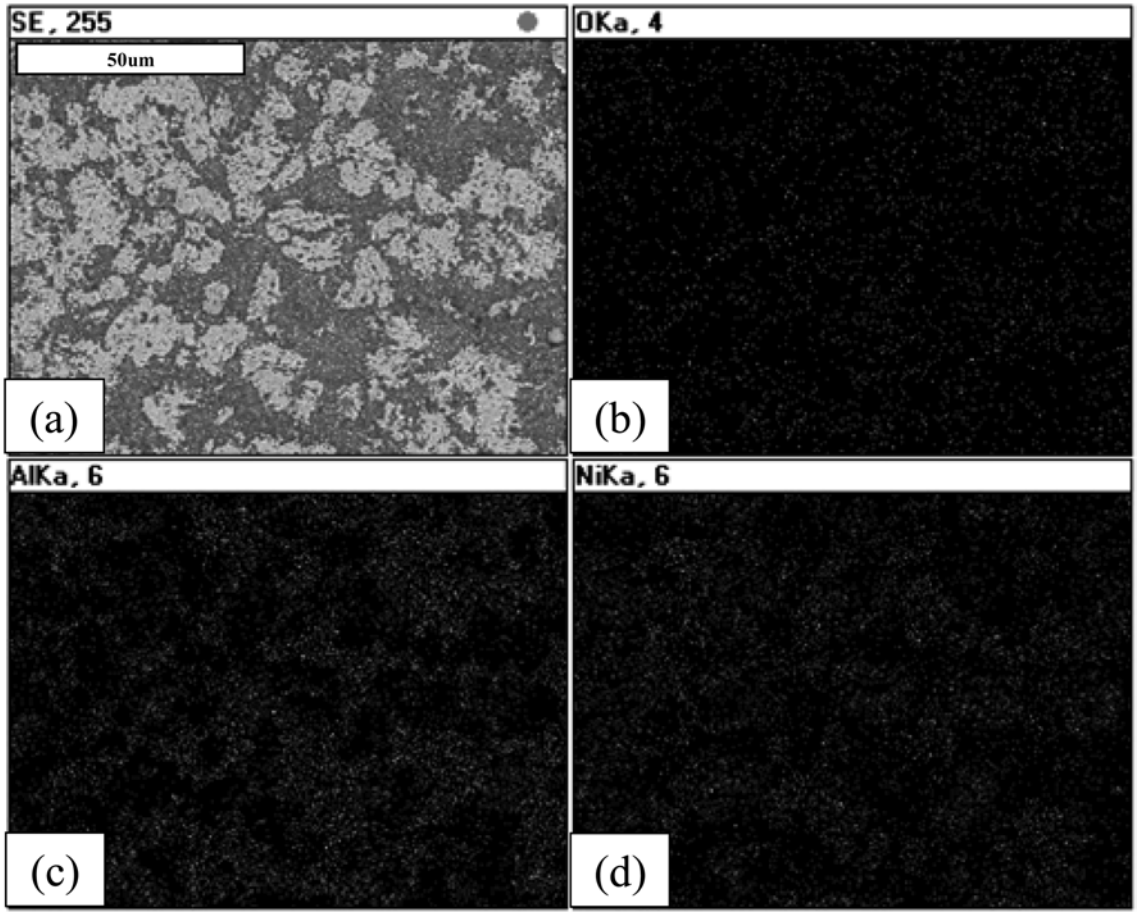

Fig. 8. Scanning electron microscope image and X-ray maps of a Ni-Al_2O_3 composite: (a) SEM image, (b) oxygen map, (c) aluminum map, (d) nickel map.

Vickers hardness measurements were made on polished sections of the Ni-Al_2O_3 composite using a 5 kg_f load and a 15 s dwell time. The calculated hardness value of 3Ni-Al_2O_3 composite was 690 Kg/mm^2. This value represents an average of five measurements. Indentations with large enough loads produced median cracks around the indent. The length of these cracks permits an estimation of the fracture toughness of the material by means of Anstis's expression [20]. :

$$K_{IC} = 0.016(E/H)^{1/2}P/C^{3/2} \quad (3)$$

where E is Young's modulus, H the indentation hardness, P the indentation load, and C the trace length of the crack measured from the center of the indentation. The modulus was estimated by the rule mixtures for the 0.5657 volume fraction of Al_2O_3 and the 0.4343 volume fraction of Ni using E(Al_2O_3) = 380 GPa [21] and E(Ni) = 200 GPa [22]. The calculated fracture toughness value of 3Ni-Al_2O_3 composite is about 6.1 MPa·m$^{1/2}$. As in the case of the hardness value, the toughness value is the average of measurements on five measurements. The hardness and fracture toughness of Al_2O_3 with a grain size of 4.5 μm are reported to be 1800 kg/mm^2 and 4 MPa·m$^{1/2}$, respectively [21]. The hardness of Ni-Al_2O_3 composite is lower than that of monolithic Al_2O_3 but the fracture toughness is higher than the value of Al_2O_3 due to the addition of ductile Ni.

A typical indentation pattern for Ni-Al_2O_3 composite is shown in Fig. 9. Typically, one to three cracks were observed to propagate from the indentation corner. This demonstrates that the crack propagates linearly.

Conclusions

Nanopowders of Ni and Al_2O_3 are synthesized from NiO and 2 Al powders by high energy ball milling. Using the pulsed current activated sintering method, the densification of nanocystalline an Al_2O_3-reinforced Ni composite was accomplished from the mechanically synthesized powders. Complete densification can be achieved within 2 minutes. The relative density of the composite was 95.6% for an applied pressure of 80 MPa with the pulsed current. The average grain sizes of the Ni and Al_2O_3 prepared by PCAS were about 690 nm and 46 nm, respectively. The average hardness and fracture toughness values obtained were 690 kg/mm^2 and 6.1 MPa·m$^{1/2}$, respectively.

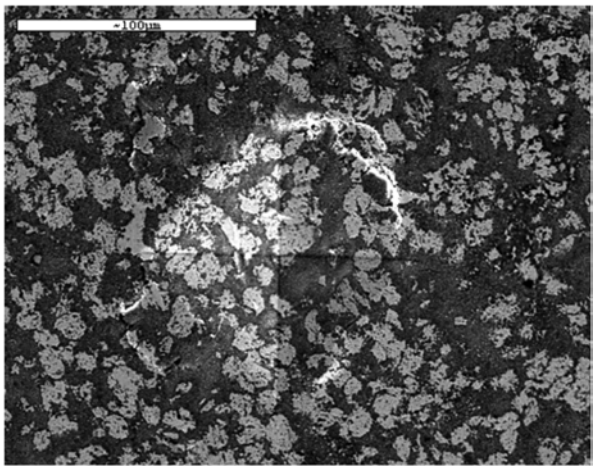

Fig. 9. Vickers hardness indentation on the Ni-Al_2O_3 composite.

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