Journal of Ultrafine Grained and Nanostructured Materials
https://jufgnsm.ut.ac.ir
Vol. 55, No.1, June 2022, pp. 70-77
Print ISSN: 2423-6845 Online ISSN: 2423-6837
DOI: 10.22059/jufgnsm.2022.01.11

The effect of Al-BN Powder Preparation Method on Tensile Properties of Extruded bulk Nanocomposite

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Recieved: 15 November 2021; Accepted: 3 January 2022

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ABSTRACT

In this study, the Al-BN nanocomposite powders were prepared using two different methods of i) ultrasonic mixing, and ii) planetary ball milling. The Al-BN bulk nanocomposite samples were fabricated by hot extrusion. Morphology of powders through the preparation process and fracture surface were characterized using scanning electron microscopy and the microstructure of optimized bulk nanocomposite sample was investigated by a transmission electron microscopy and electron back scattered diffraction technique (EBSD). The mechanical properties of composite samples were examined by uniaxial tension test. With the use of planetary high energy ball milling and hot extrusion, the ultimate tensile strength of fabricated unmilled and milled pure Al samples reached to 104 MPa and 212 MPa, respectively. The ultimate tensile strength of extruded milled nanocomposite increased to 297, 330, 333MPa by adding 1, 2 and 4 wt.% BN, respectively. In the process of composite powders mixing and hot extrusion, the mechanical properties of samples were significantly decreased. By increasing the BN content within range of 0-6 wt.%, the tensile strength of mixing and extruded composite samples was not changed. On the other hand, the ductility is reduced from 24 % for pure Al to 5 % for Al-6 wt.% BN.

***Keywords**: Al-BN nanocomposite, Hot extrusion, Planetary ball milling, Tensile Property, Ultrasonic mixing.*

1. Introduction

In the last decade, the fabrication of bulk Al-BN nanocomposites has been significantly considered due to their high strength at room and elevated temperatures [1-3], light weight [4, 5], superior wear resistance [6], high damping capacity, and dimensional stability [7]. These properties are created in the mentioned nanocomposite due to unique feature of h-BN particles such as, its lower density compare with Al and high thermal conductivity, dissolution ability in Al matrix through the high energy milling [8-10], and in-situ phases formation capability during the subsequent heat processing [2, 4]. The weak Van Der Waals bonding between h-BN layers and low wettability in Al melting below 900 °C limit the nanocomposite fabrication method to powder metallurgy [9, 11].

Mechanical alloying is a solid-state composite powder preparation technique including repeated cold welding, fracturing, and rewelding of powder particles in a high-energy ball mill. In this process, the mixtures of composite powders contain reinforcement and control agent (PCA) are loaded into a vial with stainless-steel balls as a grinding media. It can also provide the chemical reaction between components to produce the in-situ nanocomposite [12]. It well founded that the in-situ AlN and AlB_2 phases are generated during mechanical alloying of Al-BN system with high energy ball milling and subsequent heating process as following [11]:

$$3Al + 2BN \rightarrow 2AlN + AlB_2 \quad (1)$$

Although, the mechanical properties of manufactured Al-BN nanocomposite are considerably increased by using high energy ball milling of composite powders, the self-lubricating feature of BN particles is removed by dissolution of this particle in Al matrix. In liquid process with using an ultrasonic method, the acoustic transient cavitation is produced by high-intensity ultrasonic waves which lead to produce an implosive impact by which the cluster of nanoparticle fragments and uniformly disperse in liquid [13].

Firestein et al. [1] prepared Al-BN nanocomposite powders using homogeneous mixing and planetary ball milling. Then, the bulk samples were fabricated by spark plasma sintering (SPS) at 600 ˚C. The tensile strength of Al-4.5 wt.% BN improved about 50 % and 130 % compare with pure Al as reference sample in the homogenous mixing and planetary milling of powders, respectively.

The several advantages including Achieving full density without the need for sintering, easy manufacturing process, low-cost production, and dynamic recrystallization occurrence and grain refinement lead to utilize the hot extrusion as the routine process in Al composite powder consolidation [14-16].

The mechanical properties and microstructure evolution of Al-BN nanocomposite is fabricated by planetary ball milling for 300 min and hot extrusion at 580 ˚C were investigated. Decomposition BN and solid solution formation during milling process, and then subsequent AlN in-situ phase creation in Al matrix were reported as the main factor of mechanical properties improvement [4, 5]. Penchal Reddy et. al. [7] produced Al-BN nanocomposite by 60 min ball milling, microwave sintering at 550 ˚C for 12 min and hot extrusion at 350 ˚C. They found that the tensile strength of Al-1.5 % Vol. BN increased about 36 % compare with pure Al.

According to previous reports, the tensile properties of Al-BN nanocomposite with manufacture process of ultrasonic mixing of composite powders and hot extrusion have not been studied. In the present study, the effect of two composite powder preparation methods of homogeneous mixing and planetary ball milling on the tensile properties of bulk hot extruded Al-BN nanocomposite samples is investigated.

2. Experimental Procedure

2.1 Starting Materials

Commercial Al powders with an average size of 45 μm and hexagonal boron nitride (h-BN) powders with mean particle size of 70 nm were used as matrix and reinforcement materials, respectively.

2.2 Composite Fabrication

In the first composite powders method, the Al and BN powders separately were sonicated for 20 min at ethanol. Then, the Al powders were added to BN powders and the mixture of composite powders was ultrasound for 20 min. 1 g/lit sodium dodecyl sulfate (SDS) was used as the surfactant. After ultrasonic process, the mixture of composite powders in ethanol was placed on magnetic stirrer at 50 ˚C up to ethanol vapored and composite powders completely dried. Mixture of composite powders containing Al powders with 2, 4 and 6 wt.% BN were prepared. After composite powders consolidation, the hot extrusion was carried out at 450 ˚C.

In the second method, the Al-BN composite powders including 1, 2, and 4 wt.% were prepared using planetary ball milling under following technical parameters of 300 min milling in argon atmosphere, powder-to-ball weight ratio of 1:20, rotational speed of 430 RPM, and 1 wt.% stearic acid as process control agent. The weight percentage of BN reinforcement particles in Al matrix was selected to show how tensile properties of manufactured bulk composite change.

The composite powder was annealed at 200 ˚C and then was pressed at room temperature. The compacted billet was extruded at 580 ˚C. In order to fabricate control sample, the same experimental procedure was performed for pure Al. The extrusion temperature to consolidation of ultrasonic mixing powders is selected lower than planetary ball milled composite. This choice is considered to prevent melting of unmilled Al powders and avoid the chemical reaction between BN and Al at high temperature to remain BN particles in composite matrix.

2.3. Tensile Properties Evaluation

The mechanical properties of extruded samples were examined by tensile test. The cylindrical tensile specimens were machined along extrusion direction with diameter of 6 mm and 30 mm length according to ASTM E8. The tension test was conducted at room temperature and constant cross-head speed of 0.5 mm/min.

2.4. Microstructure Characterization

The microstructure observations and morphological evaluation of powders were investigated using a scanning electron microscope (Philips XL 30). The fracture surface of tensile test sample was observed using Cambridge S360 model scanning electron microscope (SEM). a transmission electron microscope (JEOL JEM-2100F) with an operating voltage of 200 kV, and Electron back-scattering diffraction (JEOL 7001 FE-SEM) with a field emission gun operating at 20 kV were used to investigate of in-situ phase formation and grain size. After grounding and final mechanical polishing, the electro-polishing of the EBSD sample and twin jet-polishing (Struers Co. Ltd.) of the TEM sample were performed in electrolyte solution of 20 vol. % $HClO_4$ acid and 80 vol. % of ethanol with the voltage of 35 V at -20 ˚C. Finally, the preparing of TEM specimen was carried out by ion beam using Gatan 691 precision ion polishing system (PIPS) for 1 min.

3. Results and Discussion

3.1. Powder Morphology

Figure 1 shows the morphology of Al- 2 wt.% BN composite powders after two different preparation methods. As can be seen in Fig. 1(a, b), the BN particles are distributed in Al powders uniformly in the ultrasonic mixing method. As expected, the morphology and particle size of composite powders in this method has not been changed. On the other hand, the morphology of composite powders in planetary ball milling method is changed from irregularly initial morphology to mostly equiaxed (Fig. 1(c, d)). In planetary ball milling process, the composite powders mechanically alloyed in sequence of cold welding, fracture and rewelding the particles [12]. The particle size of Al-2 wt.% BN composite powders was increased from 45 µm to 100 µm through 300 min planetary ball milling. Therefore, the cold welding is dominant mechanism after 300 min milling process in this condition.

3.2. Mechanical Properties and Microstructure Observations

Figure 2 shows the tensile properties of the extruded samples including milled and unmilled pure Al powders, ultrasonic mixing and planetary milled composite powders at different contents of BN particles. As can be seen, the mechanical properties of Al-2, 4 and 6 wt.% are prepared using the ultrasonic mixing composite powders and extrusion, are significantly deteriorated. The ultimate tensile strength and elongation of control

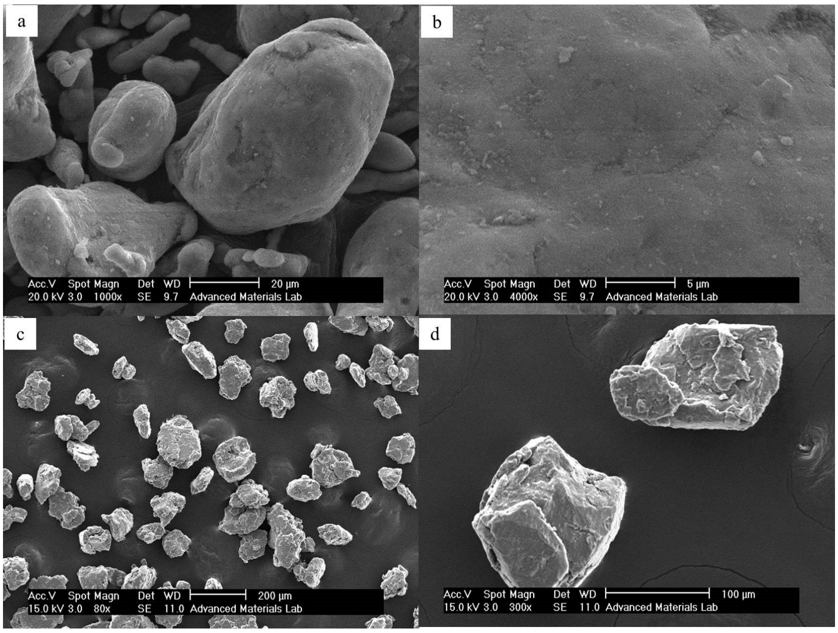

Fig. 1- Morphology of Al-2 wt. % BN composite powders after two different preparation methods a, b) Ultrasonic Mixing method with high magnification view on Al particle with uniform distribution of BN particles c, d) 300 min planetary ball milling process with high magnification view.

pure Al with manufacture method of ultrasonic mixing and hot extrusion at 450 °C are recorded 98 MPa and 24 %. On the other hand, the tensile strength of the Al-2, 4 and 6 wt.% increased about 2 % compare with pure Al. However, their ductility of the Al-2, 4 and 6 wt.% are decreased about 11 %, 7 %, and 5 %, respectively.

After hot extrusion of unmilled pure Al at 580 °C, the ultimate tensile strength and elongation of unmilled pure Al is reported 104 MPa and 26 %, respectively. After planetary milling and extrusion of pure Al, the ultimate tensile strength increased to 212 MPa and the elongation reduced to 15. On the other hand, the ultimate tensile strengths improvement of milled composite samples of Al-1, 2, and 4 wt.% are about 40, 70 and 90 % compare with milled pure Al. Among the all-extruded samples, the Al-1 wt.% BN have high ultimate tensile strength of 297 MPa with acceptable deceasing in elongation.

The poor mechanical properties of mixing and extruded samples attributed to weak strengthening of BN particles in matrix. The Van der Waals bonding between BN layers could not increase the dislocation in matrix with Orowan mechanism [11]. In addition, the presence the micro voids in microstructure is shown in Fig. 3 led to initiate cracks and reduce the ductility of nanocomposite. It seems that the presence of BN in microstructure prevents the maximum consolidation achievement in the bulk composite in the mixing and extrusion condition. However, solid solution formation due to BN particle decomposition in Al matrix, in-situ phase generation, and grain refinement are

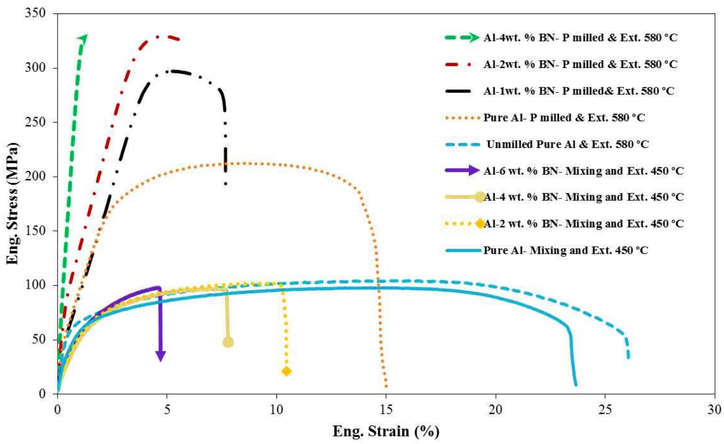

Fig. 2- Engineering Stress-strain curves for hot extruded samples including two powders preparation methods of ultrasonic mixing and planetary ball milling.

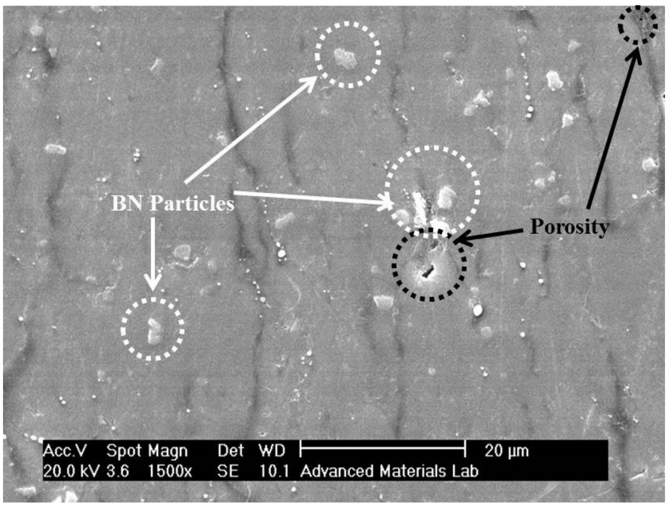

Fig. 3- Display the micro voids and agglomerated BN particles in SEM Microstructure of Al-4 wt. % BN is fabricated using mixing and hot extrusion.

strengthening mechanisms in the planetary ball milling and extruded samples.

Figure 4 displays the TEM micrograph and corresponding selected area pattern of Al-1 wt.% BN is prepared the using planetary milling and extrusion. As can be seen, the BN particles disappeared in the Al matrix after planetary ball milling process. Contrary to hard reinforcement such as B_4C, Al_2O_3, and SiC, the BN particles easily decomposed to boron and nitrogen elements due to weak Van Der Waals bonding between h-BN layers and then dissolved in the Al matrix in short time of high energy milling. This process led to work hardening, solid solution and grain refinement of Al matrix in planetary milling process [17].

In addition, the microstructure consists nano/ultrafine crystalline grains. Hence, the SAD pattern is indicated as a ring. The needle in-situ phases were generated in microstructure. The high-resolution TEM of in-situ phase is shown in Fig. 5. The results of EDS the in-situ phase indicated that Al_4C_3 as an unexpected phase was formed in matrix. The Al_4C_3 phase was generated by the reaction of Al and carbon (is created from dissolution of stearic acid in high energy milling)[18].

The EBSD observation of Al-1 wt.% BN is fabricated using the ball milling and hot extrusion was shown in Fig. 6. The microstructure consists recrystallized ultrafine/nano grains with the average size of 850 nm. According to high true strain of 2.3 in hot extrusion, the dynamic recrystallization is dominant mechanism in grain refinement. In addition, this nanocomposite matrix resists against grain growth and microstructure remains

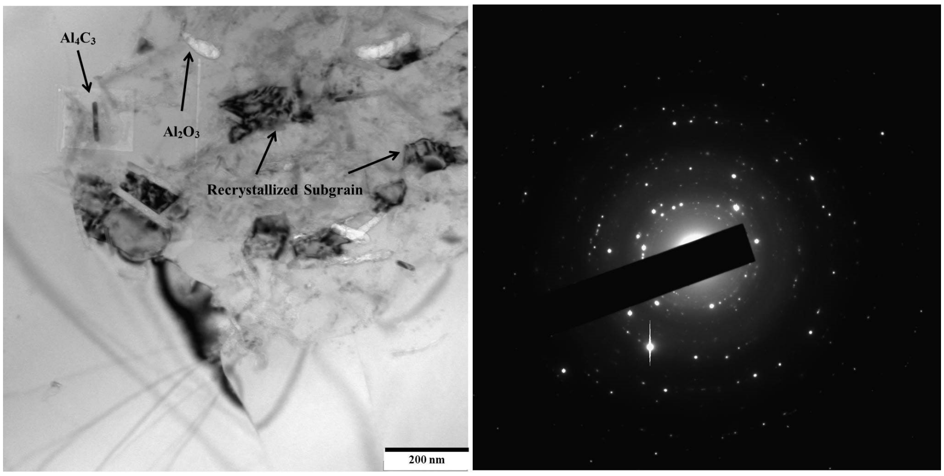

Fig. 4- TEM micrograph corresponding SAD pattern of Al-1 wt. % BN papered using planetary ball milling and hot extrusion.

Elements	Atomic Percent (Point 1)	Atomic Percent (Point 2)
Al	91.7	85.44
C	-	6.45
B	-	-
N	-	-
Cl	0.05	0.09
Cu	3.59	3.58
O	4.66	4.44

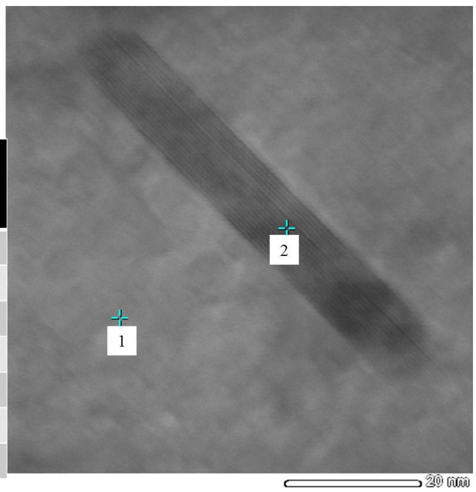

Fig. 5- Bright field corresponding of EDS results from in-situ Al_4C_3 in Al-1 wt. % BN nanocomposite matrix.

in nano/ultrafine grains in the hot extrusion. The grain boundary mapping shows that the high fraction of boundary (87.75 %) is high angle. The presence of low angle grain boundary indicated that the dynamic recovery as a possible softening mechanism in Al matrix with high stacking fault energy occurred in the initial stage of hot deformation. The recrystallization grains appeared with continuous dynamic recrystallization (CDRX) with increasing dislocations, and then the rotation and coalescence of low angle grain boundaries in matrix [19].

3.3. Fracture Surface of Extruded Milled Composite Samples

Fracture surfaces of milled pure Al and milled nanocomposite samples are shown in Fig. 7. As can be seen in Fig. 7 (a), the fracture image of hot extruded milled pure Al is a dimple like structure. On the other hand, the fracture surface dimples of Al-1 wt.% BN are shallow, which shows that not so much plastic deformation occurred up to ductile fracture in this nanocomposite (Fig. 7 (b)). Some cracks with micro voids (indicated by arrows in Fig. 7 (c)) which were observed in fracture surface of Al-2 wt.% BN and Al-4 wt.% BN nanocomposites, indicated that the fracture type of nanocomposite including more than 1 wt.% BN nanoparticles was different.

The dimple like structure in Fig.7 (a) which shows the ductile fracture of pure Al, is created due to nucleation, growth and coalescence of created voids in matrix, during the tensile test [20]. Contrary to pure Al, in which failure occurred due to dimple rupture, the initiation and growth of crack led to fracture in nanocomposite with BN content of 2 wt.% and higher (Fig.7 (c-d)). Decohesion in matrix and also dimple like structures with micro void size of 0.1-0.5 µm, which are known as characteristics of intergranular failure mode [21], are shown by arrows in Fig. 7 (c). The flat fracture surface view of Al-2 wt.% BN indicated low plastic

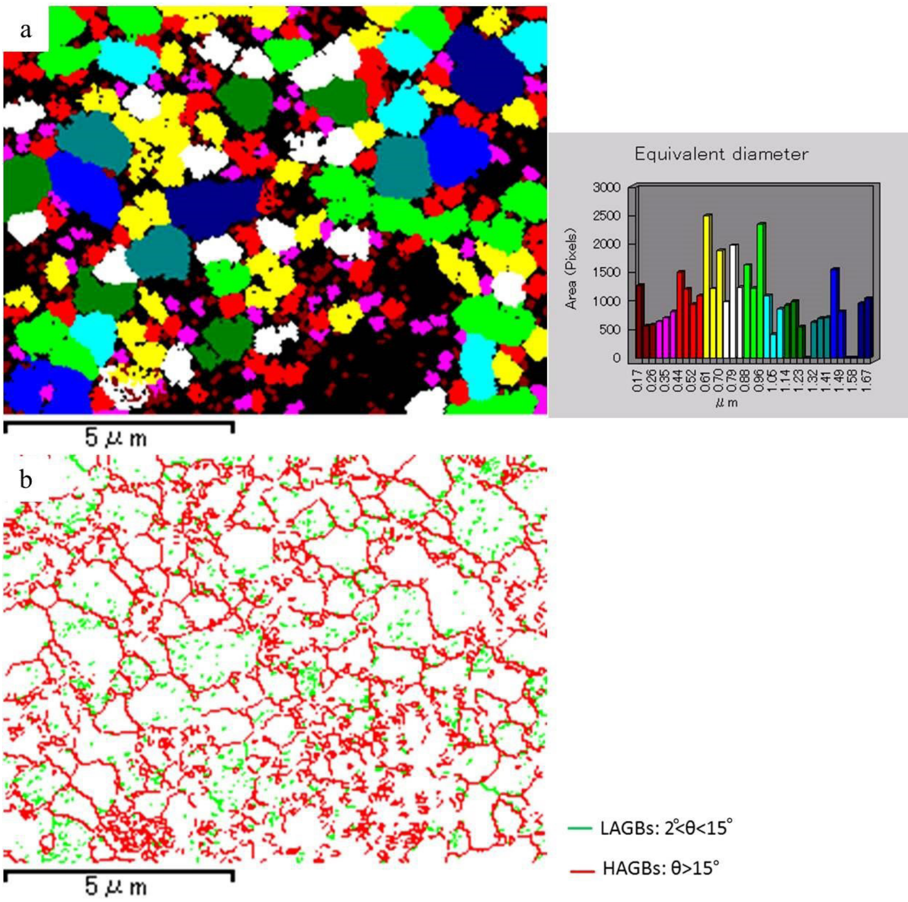

Fig. 6- a) EBSD map of Al-1 wt. % BN corresponding grain diameter distribution histogram b) gain boundary mapping.

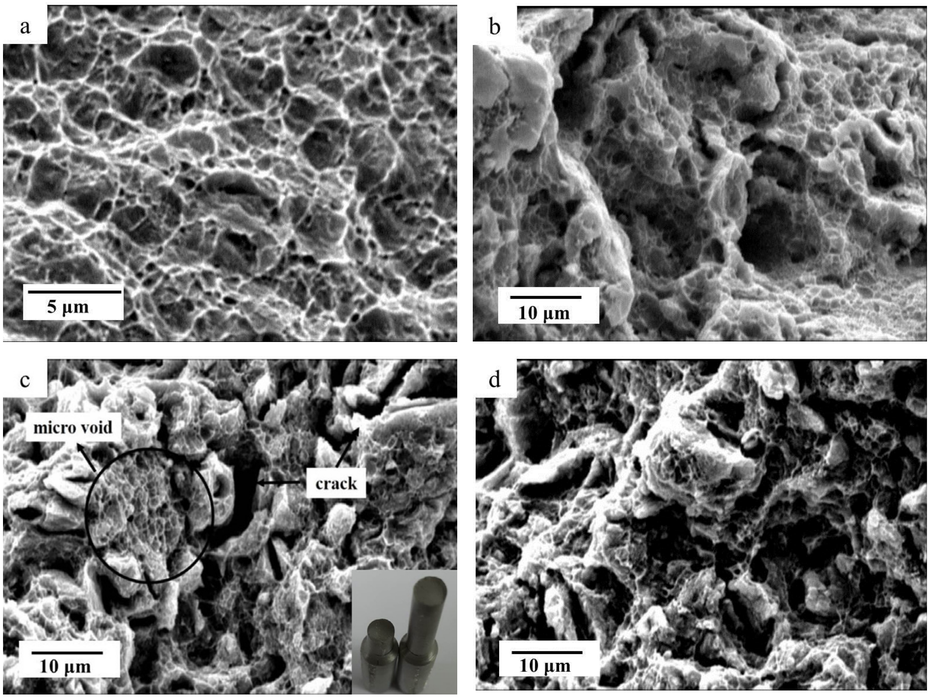

Fig. 7- Fracture surface of milled and extruded samples: a) pure Al b) Al-1 wt. % BN c) Al-2 wt. % BN d) Al-4 wt. % BN.

deformation without necking occurrence in tensile test (Fig. 7 (c)). Therefore, the fracture type of the fabricated nanocomposites including more than 1 wt.% BN nanoparticles is recognized as the brittle intergranular fracture.

4. Summary

In this study, the Al-BN bulk nanocomposite was fabricated using the hot extrusion of composite powders which has been prepared with two different methods of ultrasonic mixing and planetary ball milling. The results show that the ultrasonic mixing method not only has no effect on improving the tensile strength, but also decreases the ductility of the extruded Al-BN nanocomposite. On the other hand, the tensile properties of Al-1 wt.% BN nanocomposite is prepared using planetary ball milling process increased about 40 % and 185 % in comparison with milled and unmilled pure Al as control samples. The BN decomposition in Al matrix and solid solution formation occurred in milling process and then Al_4C_3 in-situ phase generation and grain refinement after hot extrusion led to increase the mechanical properties. Based on fracture surface observations, the growth of cracks and micro voids formation are rupture mechanisms at milled and extruded composite sample with higher than 2 wt.% BN. Therefore, the optimum concentration of BN in Al matrix composite is selected 1 wt.%.

Acknowledgements

The authors would like to acknowledge the financial support from the Shiraz University. This work was also supported by the research council office of Shiraz University through Grant No. 94-GR-ENG-15.

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