Obtaining Composites Powders of Al₂O₃ / Ni and Al₂O₃ / Nb by Mechanical Alloying

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Mechanical alloying (MA) can be highlighted among the various methods and powder processing forms, which consists of producing homogeneous materials from elementary powders. This process is of paramount importance as it has the ability to reduce the particle size of the powders and can often promote bonding between elements which have low interaction and wettability. This method is usually used in the route of powder metallurgy, and generally improves the sinterability and mechanical properties of the materials. In this context, the objective of this work was to analyze the evolution of mixtures of Al_2O_3 -5wt%-Ni and Al_2O_3 -5wt%-Nb powders submitted to the mechanical alloying process in a planetary ball mill for up to 40h. The morphological evolution was observed through Scanning Electron Microscopy (Field Emission Gun, SEM-FEG), energy dispersive spectrometry (EDAX) phases and particle sizes by Particulometry. It was observed that a significant micro structural change occurred in both compositions, indicating the formation of composite powders. This behavior was more intensely observed with the increase in the grinding time, obtaining composite powders with a more homogeneous and uniform structure with the time of 40 hours of MA.

Keywords: Alumina, nickel, niobium, composite powders, Mechanical alloying.

1. Introduction

Alumina is a widely used material in engineering, especially in structural applications, and has very characteristic properties such as high mechanical strength, chemical inertia, refractoriness, hot hardness, and corrosion resistance, among others. However, its use is restricted in several other applications because it has low fracture toughness. In this context, a solution to improve this toughness could be, found by adding impurities, which can drastically affect the properties of alumina ceramics even in small quantities¹, as well as including metal in ceramics, which can serve as fronts for inhibiting crack propagation, resulting in an improvement in the toughness of the ceramic material^{2,} ^{3, 17}. In this context, nickel and niobium gain importance because they are metals with desirable characteristics and are present in abundance in Brazil. According to data from the Brazilian Mining Institute, Brazil is the seventh largest producer of nickel and holds about 98% of the world's niobium reserves. Powder Metallurgy is an industrially developed and established process for manufacturing ferrous and non-ferrous components. It is economically advantageous over other production techniques, since it minimizes the need for machining and surface finishing, and maintains a close dimensional tolerance 4, 5. One of the powder metallurgy stages is preparing and processing powders and mechanical alloying (MA) is a widely used technique, mainly because it enables producing homogeneous materials starting from the mixture of elementary powders. One of the frequently used kind of equipment in MA is the planetary ball mill. It received this name because of the planetary movement of its milling containers. The containers (crucibles) rotate counterclockwise, causing the balls to travel and collide against the opposing wall of the crucible^{6, 10}. At the beginning of the process, the fragile particles fragment and the ductile particles become flattened by a micro forging process. The flattened particles undergo welding, while the fragile particles are deposited on the surface of the ductile material.^{7,8}. MA has numerous advantages, for example: production of a single second dispersion phase (usually oxide), extending the solid solubility limits, enabling the possibility of reducing the particle size up to the nanometer range, synthesis of new crystalline phases, development of phases (vitreous), the possibility of binding immiscible elements and the induction of chemical reactions at low temperatures^{6, 7, 8}. In this context, the objective of this work is to produce composite powders from the mixtures of Al₂O₂-5wt%-Ni and Al₂O₂-5wt%-Nb using mechanical alloying and to study the evolution of this process considering its morphological and phase aspects.

2. Materials and Experimental Procedures

The alumina powder was obtained from the manufacturer Alumina do Norte do Brasil S/A with 98.99% purity, the nickel from the Aldrich Chemical Company (99.99% purity) and the niobium from Chemcraft Ltda, with 99.99% Purity. Two alumina compositions were prepared with nickel and niobium additions, both of 5% by weight, for a total of 10 grams for each composition.

The preparations were subjected to mechanical alloying in a Pulverissite 7 planetary ball mill using 5mm diameter beads and crucible, both of alumina, to avoid possible contamination. Ethyl alcohol was used as a process control agent (PCA) and a powder oxidation inhibitor. The ball-topowder mass ratio was 3:1, and the rotation speed was 400 RPM. The milling time was up to 40 hours, and samples were taken at 0 (manual mixing), 5, 10 and 20h. At each interval, an amount of 2 g of powder was withdrawn. Next, they were placed to dry in an oven at room temperature for 24 hours. Finally, particle size analyzes were performed using a Cilas 920 laser granulometer, Scanning Electron Microscopy and energy dispersive spectrometry (EDAX) through a ZEISS Model AURIGA 40 SEM-FEG.

Figure 1 (a-c) shows the micrographies of the elementary powders. The alumina (Figure 1-a) with characteristics a more irregular particle morphology, resembling the spherical shape. The powder of nickel, presenting a more rough morphology and the niobium powder, having a squamous morphology.

3. Results and Discussion

The Figure 2 shows the particle size analysis. It is observed that a slight increase of the particle size of the manual mix (0 hours) up to 5 hours of milling occurred in both compositions. However, from there until the time of 40 hours there was a gradual reduction in this mean diameter, and more significant in the first 20 hours. This behavior indicates that the cold welding process which occurred during the MA was predominant in the first 5 hours, causing the greatest agglomeration between particles. Then a reduction in the average particle size was observed from 5 hours onwards, indicating that the fracture process prevailed in these cases^{7,9}. However, the efficiency of mechanical alloying is gradually reduced with increasing grinding time due to the decrease in the angulation of both curves, with the tendency to become asymptotes to the x axis of the (milling time). In other words, from 5 hours, the MA is efficient in reducing particle size, yet this efficiency decreases with increasing time, especially after 20 hours. In comparing the behavior of the added metals to alumina, it is noticed that the nickel suffers a greater influence of the welding process to the cold occurred during the first 5 hours of MA than niobium.



 1 µm
 EHT = 5.00 k/
 Meg = 5.00 KX
 Signal A = InLens
 Date :14 Feb 2019

 WD = 5.0 mm
 Pixel Size = 22.92 nm
 Photo No. = 12334 Time :9:56:30

Figure 1. Scanning Electron Microscopy of the initial powders of Alumina (a), Nickel (b) and Niobium (c), at 5000X.

This fact can be explained by presence greater aglomerates this powder, indicating the predominance the welding phenomena of the nickel particles during this milling time.



Figure 2. Average particle diameter X milling time.

No significant changes were observed until the 5 hours of milling (Figure 3-a) (Figure 3-b), with squamous morphology mainly being maintained relative to niobium. Specifically, in the 10-hour milling time, the fracture process of the previously cold-welded particles can be clearly seen. The microstructure refinement is observed with more rounded and uniform particles after 20 hours^{8, 10}, constituting a possible indication of forming the Al₂O₃/Nb composite powder structure. The composite powder formation is confirmed in 40 hours of milling, when the distinction between the initial elements was not perceived, becoming a single structure with uniform tendency. Figures 4 show the energy dispersive spectrometry (EDAX) analyzes in 0 and 40 hours of grinding respectively, which confirm the formation of the composite powders.



Figure 3. Microstructural evolution of Al₂O₃-5wt-Nb powders after a)0 h, b)5h, c)10h, d)20h and e)40h MA.



Figure 4. EDAX analysis with mapping of Al₂O₃ -5wt%-Nb powders (a- 0 hours, b-40 hours MA).

In relation to the microstructural evolution of Al_2O_3 -5%wt-Ni powders described in Figure 5 (a-e), with the powders initially submitted to manual mixing without milling (fig.5-a), we observed the structure of each powder in its initial aspect with clear differentiation between them. The alumina is characterized by the spherical morphology of its particles, and the nickel has irregular and rough structure. The structures were practically undone in the 5-hour milling time (Fig. 5-b), forming considerable particle agglomerates with small rounded particles of alumina still being visible (Fig. 6). This occurred through the deformation and the micro-welding process of the nickel particles, mainly^{9,10,11,12}. The more fragile alumina particles primarily suffer from fracture, but tend to be incorporated by the more ductile



Figure 5. Microstructural evolution of Al₂O₃-5wt-Ni powders after a)0 h, b)5h, c)10h, d)20h and e)40h MA.



Figure 6. Al₂O₃/Ni powders (5% wt) after 5 hours MA (10000X).

particles, thereby forming a lamellar composition of the alloying elements^{13,14}.

The welding of the ductile particles occurs due to the localized increase of the temperature at the time of the collisions together with the high mechanical energy of the impact^{11, 12, 15}. In the range of 10 to 20 hours of milling (Figure 5c-d), it is noted that the nickel particles lose the ductile fracture aspect and incorporate the alumina particles. Then from 20 hours of milling, the presence of nano-sized composite particles was observed on the outer surface of larger remaining agglomerates (Fig. 7). Lastly, the powder structure refinement predominantly occurs in the 40-hour milling time (Fig. 5-e) for the fracture process during MA. Fracture of the fragile particles, deformation



Figure 7. Al₂O₃/Ni powders (5% wt) after 20 hours MA (10000X).

with subsequent embrittlement, and fracture of the ductile particles occurs due to the increase in the internal stress of the grains generated by the great pressure exerted on the dust particles from the collisions of the milling bodies¹⁶. Figure 8 shows the results of the EDAX analyzes with mapping for the mixture of alumina and nickel powders after 0 and 40 hours MA, respectively.

In Figure 8-a, as expected, the nickel particles are in their initial morphology, and are clearly identifiable in the analysis by the color density emitted in the mapping. In this case, we have two immiscible and low wettability powders that will be "forced" to interact through the MA process^{15, 16, 17}.

In Figure 8-b, after 40 hours of milling, a greater interaction and dispersion between the alumina and



Figure 8. EDAX analysis with mapping of Al₂O₃-5wt%-Nb powders (a- 0 hours, b-40 hours MA).

nickel particles was observed, in addition to the change in their structures becoming of irregular morphology¹⁷. Only a squamous phase of composite powders is observed even in a relatively large cluster of nickel particles, indicating that a considerable interaction between the elements occurred for this milling time.

4. Conclusions

- Mechanical alloy with a range of up to 40 hours was efficient as a processing and formation method of composite Al₂O₃-5wt%-Ni and Al₂O₃-5wt%-Nb powders. The use of this technique resulted in finer powders and homogeneous structure.
- A predominance of the hardening and cold welding process was observed until the 5 hours of milling time, mainly of Ni and Nb particles. Then the fracture, refinement and embrittlement processes of the powders prevail from 5 hours up to 40 MA hours.
- The formation process of composite powders through Mechanical Alloying was more efficient in the Al₂O₃/Nb system than in the Al₂O₃/Ni system. At the beginning, uniform refinement and dispersion was verified after 20h of milling, while some nickel agglomerates were still observed from 40h.

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6. References

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