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Spark Plasma Sintering of Aluminum-Based Composites Reinforced by Nanocrystalline Carbon-Coated Intermetallic Particles

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***Abstract*—**Aluminum Matrix Composites reinforced with nanocrystalline Ni_3Al carbon-coated intermetallic particles, were synthesized by powder metallurgy. Powder mixture of aluminum with 0.5-volume fraction of reinforcement particles was compacted by spark plasma sintering (SPS) technique and the compared with conventional sintering process. The better results for SPS technique were obtained in 520ºC-5kN-3min.The hardness (70.5±8 HV) and the elastic modulus (95 GPa) were evaluated in function of sintering conditions for SPS technique; it was found that the incorporation of these kind of reinforcement particles in aluminum matrix improve its mechanical properties. The densities were about 94% and 97% of the theoretical density. The carbon coating avoided the interfacial reaction between matrix-particle at high temperature (520°C) without show composition change either intermetallic dissolution.

***Keywords*—**Aluminum matrix composites, Intermetallics Spark plasma sintering.

I. Introduction

ALUMINUM Matrix Composites (AMC) are new advanced engineering materials that offer the possibility to obtain unusual properties to meet specific requirements to develop lightweight high-performance materials with low density, high toughness and corrosion resistance in comparison to conventional unreinforced ones [1].

The most commonly used methods to produce AMCs are casting and powder metallurgy (P/M) techniques [2]. The major advantage of P/M is the microstructural control and homogeneous distribution of the reinforcement in the matrix. Also, it is necessary to avoid the limitations and defects of casting and producing materials with superior properties obtained from the powder characteristics [1]-[5].

Nickel aluminides has been recently investigated to use them as a reinforcing phase [6]-[9].

Particularly Ni_3Alphase shows unique mechanical properties, which make it potentially useful for structural and high temperature applications [10]-[12].

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Enayati et al. [4] studied the synthesis of Ni_3Al intermetallic by means of high-energy ball milling. Their results showed the formation of a solid solution Ni (Al) and later the formation of the Ni_3Al intermetallic compound with nanometric structure after 72 ks of milling. Lieblich et al. [13] studied the thermal stability of Al-5vol. % Ni_3Al composite processed by hot extrusion of spherical Ni_3Al powder particles at temperatures up to 773K. Their results showed that Ni_3Al is stable at temperatures up to 573K. After that the dissolution of particles occurs with formation of Al_3Ni and Al_3Ni_2 phases. In order to retain the nanocrystalline structure of the powder, the sintering process has to be fast in timescale with low heat input.

On the other hand, SPS has been employed for consolidation of metastable phases, nanostructures and synthesis of new compounds. The application of SPS produces a high pressure of microsecond duration and temperatures moderate enough to achieve bonding between powders, resulting in compacted samples with high density and retained novel structure [5].

Therefore, the purpose of this work is to synthesize and characterize the aluminum matrix composites reinforced with Ni_3Al obtained by mechanical alloying and sintering by SPS on the microstructural features, density and hardness behavior.

II. Experimental Procedure

A. Raw Materials

Elemental Al (99%, <100μm particle size) and Ni (99.98%, <30μm particle size) powders were used as raw materials to produce the stoichiometric composition Ni75-Al25 (atomic percent). The powders morphologies of aluminum and nickel were rounded and spongy, respectively.

B. Mechanical Alloying and Blending Processes

Mechanical alloying (MA) of the raw materials Al and Ni powders was carried out using a horizontal ball mill to produce Ni_3Al with nanocrystalline features. The ball-powder ratio was 36:1 and the rotational speed was 110 rpm under argon atmosphere to avoid oxidation. Methanol was used as milling control agent. X-ray diffraction and scanning electron microscopy analyzed morphological and structural characteristics. After MA process, a short blending process was carried out to obtain a homogeneous mixture of aluminum with 5 volume percent of Ni_3Al reinforcement.

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C. Composites Consolidation Processes

The pre-consolidation was done in a uniaxial press in order to obtain cylindrical specimens about 11mm in diameter and 3mm thickness of the previous mixture. The mixed was pre-compacted at 200 MPa of pressure. Then, it was inserted into graphite die and placed inside the SPS chamber.

D. Samples Characterization

The phases evolution and crystal size were evaluated by X-ray diffraction using a D8 FOCUS BRUKER diffractometer with Cu-kα radiation. MA process was followed by scanning electron microscopy (SEM) using a JEOL 6300 microscope equipped with energy dispersive spectrometer (EDS). Also transmission electron microscopy (TEM) was used to analyze the formation of nanometric crystal size of Ni_3Al particles using a JEOL 2000 FXII microscope with EDS. The density of the samples after consolidation process was evaluated by the pycnometer method. Also samples were tested for Vickers micro-hardness with a load of 100g for 12 s, HV0.1/12s.

III. Results and Discussion

A. Mechanical Alloying

Fig. 1 shows the XRD patterns of raw powders and after 350h of milling. It is possible to see that aluminum atoms were dissolved in the Ni lattice and the Ni characteristic diffraction peaks displaced slightly to lower angles as Enayati et al. [4] observed. After 350 h of milling, the formation of nanocrystalline Ni_3Al intermetallic compound with disordered structure was observed [4], [14]. The grain size of particles was about 5 nm and it was estimated from broadening of XRD peaks using Scherrer method.

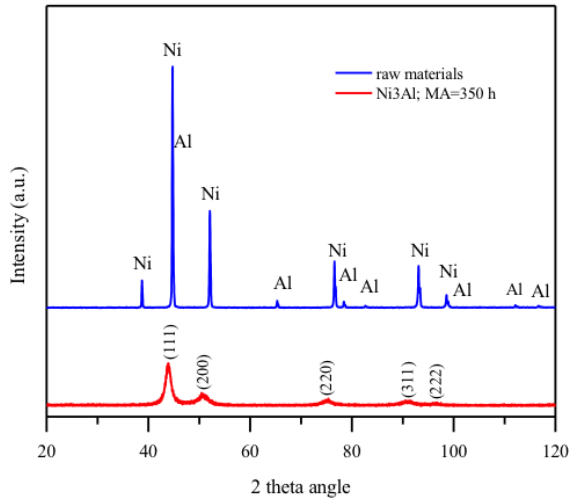


Fig. 1 XRD patterns of raw powders and after 350h of milling

Fig. 2 shows the TEM micrographs of the Ni_3Al powders. The indexing of the selected area diffraction pattern and also the bright field image confirm the presence of the Ni_3Al phase with nanometric features. Several particles were analyzed and only the presence of the Ni_3Al phase was detected.

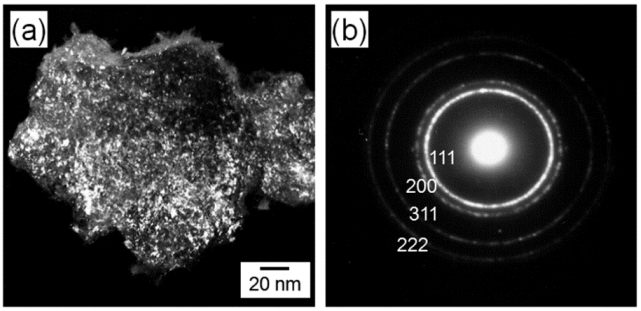


Fig. 2 Nanocrystalline Ni_3Al particle MA during 350 h (a) dark field image and (b) selected area diffraction pattern

Fig. 3, shows SEM images of composites Al-5vol.%(Ni_3Al-C). The sintering conditions were 12 tons of load at 350°C during 24, 100 y 250 h. Results showed the spatial distribution of reinforcement particles into the aluminum matrix with 48±7µmmain particle size. In 3000X after 250 h, it is possible to observe diffusion layers that suggest the interfacial reaction between matrix and reinforcement.

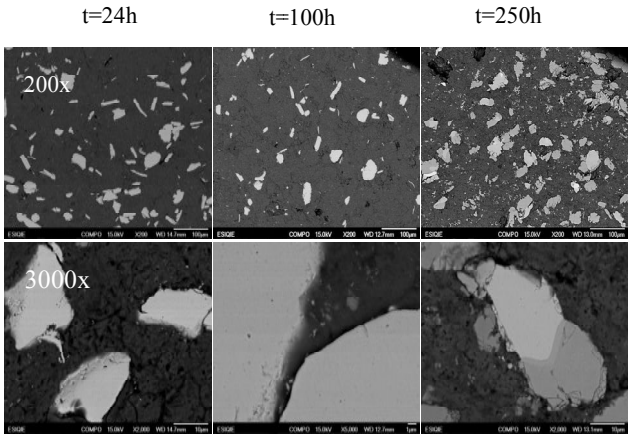


Fig. 3 SEM images of conventionally sintered composites at 350°C.

Fig. 4 shows the SEM image of a single intermetallic particle of 24h sintered composite. The EDS and microanalysis showed that nickel (81.56w%) and aluminum (13.83w%) elements were present as stoichiometric composition of Ni_3Al. On the other hand, the quantified carbon (4.61w%) were majority found at particle surround avoiding the matrix-particle interaction.

After 250 h of sintering process the reinforcement particles showed compositional layers into them as it is possible to observe in Fig. 5. The punctual microanalysis revealed that these chemical compositions corresponds with the others intermetallics in diagram equilibria phases of Ni-Al system. Accordingly, it supposes the matrix-particle interfacial reaction by a diffusive process during the sintering processes as Lieblich predicted [13].

The microhardness (HV) results for each sintered composite were: 24h= 62.3±3; 100h= 56.08±5 and 250h= 42.85±3. The density was 92-94% of theoretical density.

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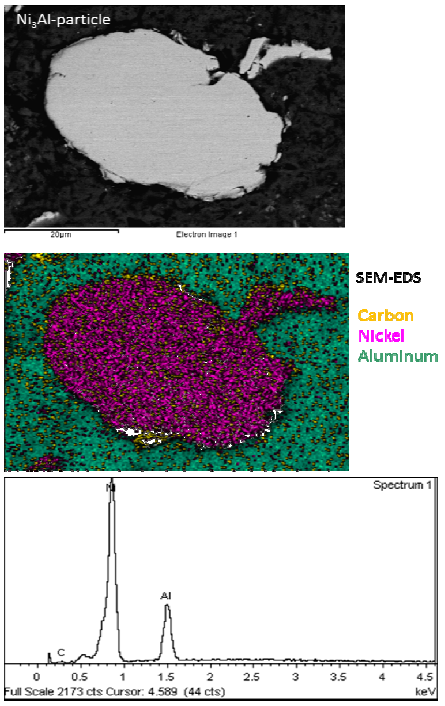


Fig. 4 EDS mapping of a single intermetallic particle of 24h sintered composite

On the other hand, Fig. 6 shows the SEM images of SPS sintered composites for different sintering conditions of temperature, pressure and time. In general, it is possible to observe a homogeneous particle distribution on matrix and a few irregular agglomerates in both temperature limits (450ºC and 520ºC). At 450ºC some particles exhibited interfacial diffusion in comparison with the particles at 520ºC that showed many interfacial reactions with the matrix even a total particle dissolution was observed at 520ºC-15kN-10min (M8 sample).

Resume of SPS sintering conditions and its relationship with microhardness and density measurements is showed on Table I. The best densification and hardness was obtained in M7 (520ºC-15kN-3min) and M2 (450ºC-15kN-3min) samples.

Fig. 7 shows the concentric rings in a single particle of M8 sample sintered by SPS. The linear microanalysis revealed that these extreme conditions (520ºC-15kN-10min) increase the intermetallic reactivity and promoted the interface reaction resulting in almost total dissolution and formation of Al_3Ni and NiAlintermetallics.

TABLE I
RESUME OF SPS SINTERING CONDITIONS AND MICROHARDNESS AND DENSITY RESULTS

Sample	T (°C)	P (kN)	t (min)	HV[a]	Density %
M1	450	5	3	29.3±5	91
M2		15	3	62.3±7	95
M3		5	10	32.8±4	87
M4		15	10	56.1±4	92
M5	520	5	10	55.3±5	87
M6		5	3	52.7±4	84
M7		15	3	70.5±8	97
M8		15	10	57.1±9	93

[a]HV= microhardness Vickers

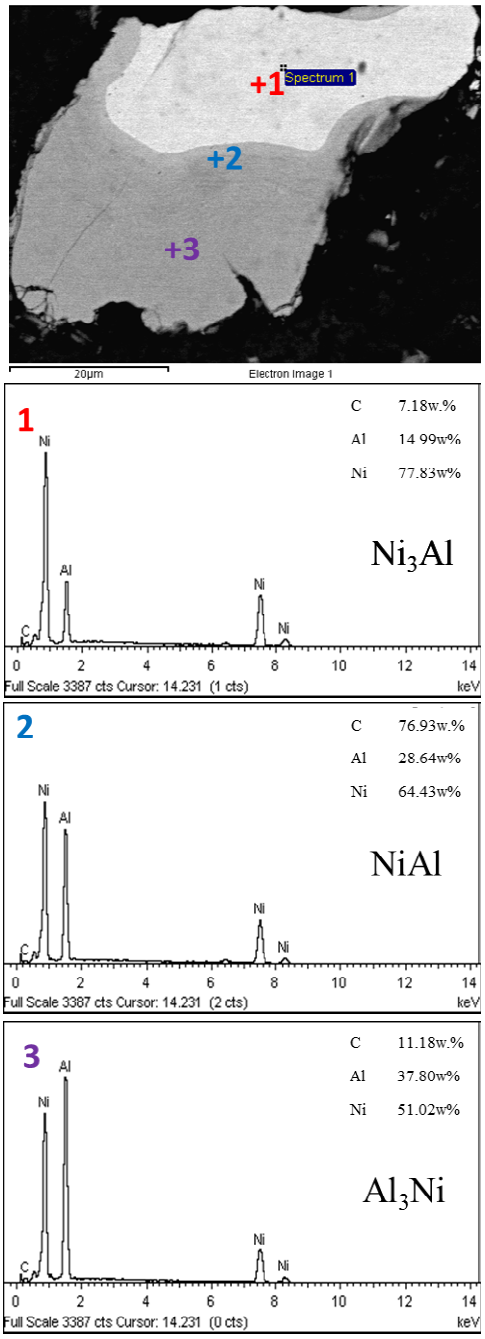


Fig. 5 Punctual microanalysis of 250h sintered sample

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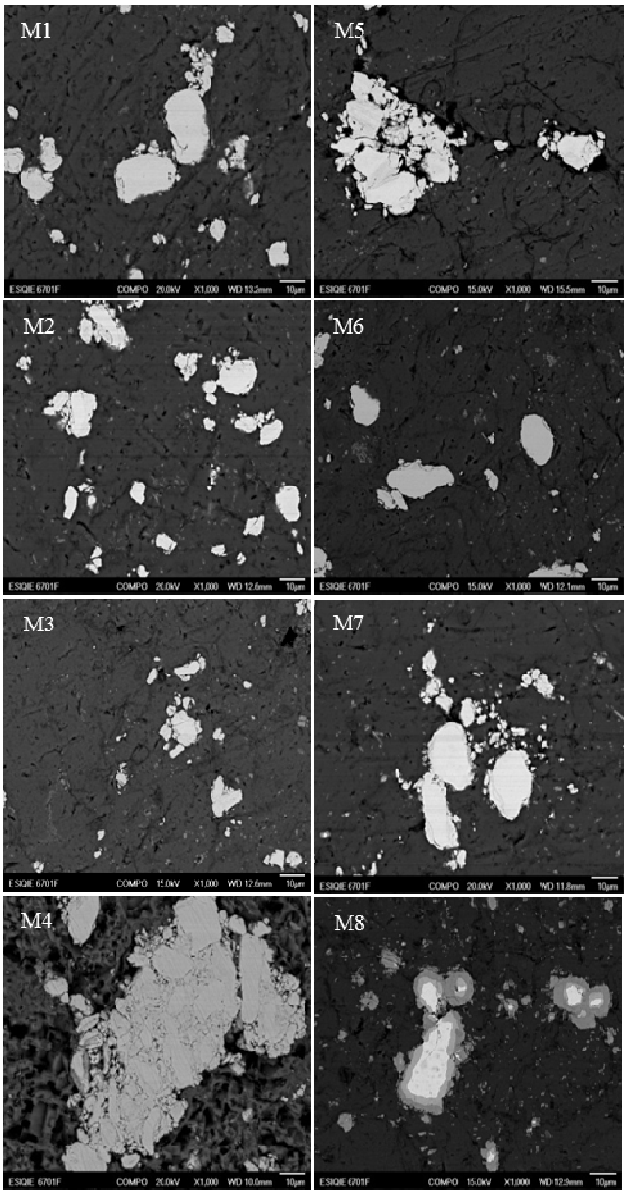


Fig. 6 SEM images of SPS sintered composites at different operating conditions

Operating conditions for SPS process: **M1**: T= 450°C; P=5kN; t= 3min. **M2**: T= 450°C; P=15kN; t= 3min. **M3**: T= 450°C; P=5kN; t= 10min. **M4**: T= 450°C; P=15kN; t= 10min. **M5**: T= 520°C; P=5kN; t= 10min. **M6**: T= 520°C; P=5kN; t= 3min. **M7**: T= 520°C; P=15kN; t= 3min. **M8**: T= 520°C; P=15kN; t= 10min

Fig. 8 shows a theoretical graphic about elastic modulus versus reinforcement volume fraction calculated by mixing rules. The upper limit was calculated by (1):

$$E_c = E_m V_m + E_r V_r \quad (1)$$

and the lower limit by (2)

$$E_c = \frac{E_m E_r}{E_m V_r + E_r V_m} \quad (2)$$

where Ec, Er and Em are the elastic modulus of the composite, reinforcement and matrix, respectively. Moreover Vm and Vr are the matrix and reinforcement volume respectively.

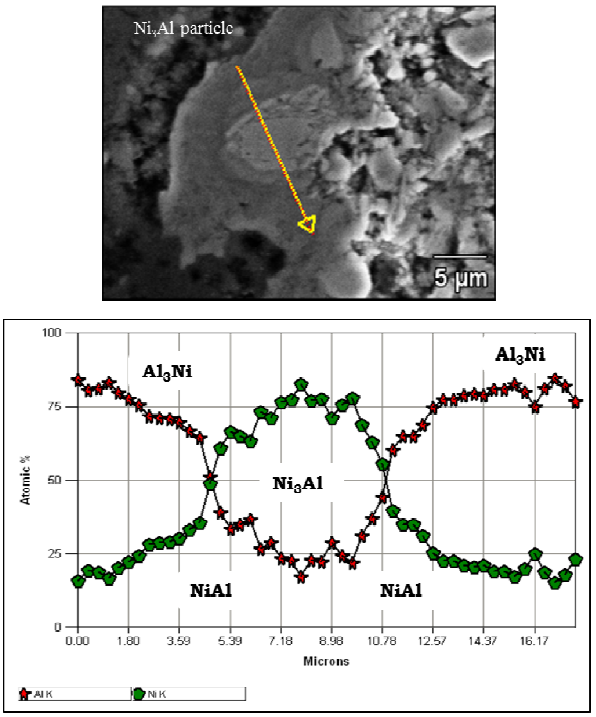


Fig. 7 Lineal microanalysis of a single particle of sample M8 of SPS sintered composite

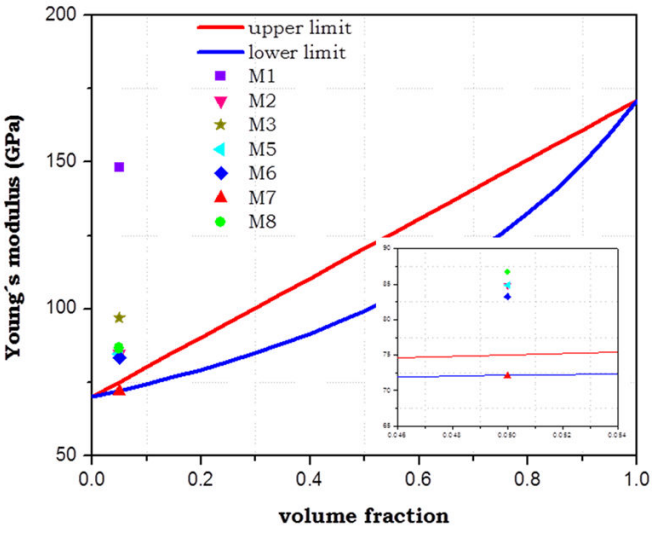


Fig. 8 Theoretical graphic about elastic modulus vs. reinforcement volume fraction calculated by mixing rules

From these results we assumed the following: for the M1 sample (450ºC-5 kN-3min) the elastic modulus and hardness values were the highest, which is related to the accumulated stress during MA process. The density of M1sample was smaller than M2 and M7 samples because of the presence of porosity. M2 and M7 samples showed the major densification compared with the other samples of the experimental matrix.

Even, these samples presented the best elastic modulus and hardness values due to the homogeneous distribution of intermetallic particles in the matrix as well as the temperature were the dominant factor on the densification result for these operating conditions.

IV. Conclusions

Aluminum Matrix Composites can be hardened with nanocrystalline-Ni_3Al intermetallic. Nearly dense Al-(Ni_3Al-C) composites could be produced by SPS showing good particle distribution and high density. The conventional sintered composites showed gradual intermetallic decomposition as Lieblich predicted in the study about Ni_3Al thermal stability at high temperature, however, the carbon presence delayed this process. The fact that there were not matrix-reinforcement interfacial reactions or intermetallic dissolution after 520°C-15 kN-3min (M7 sample) of SPS process indicates that this technique has significant potential applications if is used as consolidation process. It was obtained an excellent densification (97%), highest hardness (70.5±8 HV). The elastic modulus was significantly improved according with the reinforcement volume fraction on the matrix. Finally, a clear intermetallic particle dispersion strengthening was observed for all tests with the improvements of aluminum mechanical properties.

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