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Combustion Synthesis Reactions in the Nb-Ni-Al System

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Abstract. Combustion synthesis of homogeneous Nb-Ni-Al alloys powder mixtures with nominal compositions Nb10Ni70Al, Nb20Ni65Al and Nb30Ni60Al have been investigated, with emphasis on the reaction mechanisms. Experiments were carried out by thermal explosion mode ("simultaneous combustion") conducted on cylindrical compacts along with differential scanning calorimetry (DSC) analysis of small fragments obtained by crushing the compacts. Microstructural characterization consisted of SEM, EDS and X-ray diffraction analysis. It was found that synthesis occurs as a two-stage reaction. Ni_2Al_3 and/or $NiAl_3$ are formed in the first stage, with relative amounts depending on the initial compact composition. The first stage reaction can trigger the second stage reaction related to the formation of $NbAl_3$.

INTRODUCTION

Combustion Synthesis in condensed system is a powerful process route, which involves strong exothermic reactions between constituent elemental powders to form a desired product. This method has increasingly becoming an attractive alternative route to process intermetallic materials [1,2]. There are two different modes of combustion. They have been referred to as SHS (acronym for Self-Propagating High Temperature Synthesis) and thermal explosion or simultaneous combustion. In the former, a powder mixture is ignited locally and the reaction propagates as a combustion front through the mixture. In the latter, a powder compact is heated in a furnace until the onset of the bulk reaction. Niobium and nickel aluminides are among the intermetallic compounds that can be synthesized by combustion reactions. Also $NbAl_3$-NiAl alloys can be obtained by this way. Many aspects concerning the synthesis and densification of niobium trialuminide and Nb-Ni-Al alloys have been investigated by the present authors [3-6]. In this work the main objective is to get extended knowledge on the reaction mechanisms involved in the combustion of Nb-Ni-Al powder mixtures by conducting experiments on thermal explosion mode along with differential scanning calorimetry (DSC) analysis.

EXPERIMENTAL

Simultaneous combustion synthesis was carried out on cylindrical pellets of 14mm diameter and 6 mm height. They were obtained by pressing uniaxially at 300MPa mixtures of Nb (less than 33 µm), Al (less than 33µm) and Ni (less than 10µm) powders according to the following nominal compositions: Nb10Ni70Al, Nb20Ni65Al and Nb30Ni60Al (at. %). Experiments consisted of heating the pellets at 15°C/min., under vacuum, to the ignition temperature as described previously [3]. After the reaction, pellets were cooled down to room temperature.

Differential scanning calorimetry analysis (DSC) under an argon flux (100ml/min.) were performed on small samples (100mg) consisting of fragments of compacted pellets with similar compositions. A 15°C/min. heating rate was used in these experiments. Also, fragments with similar compositions were submitted to heat treatment at 650°C for a short period of time followed by quick cooling.

The microstructures of the as reacted pellets and of the fragments submitted to the 650°C heat treatment were characterized by scanning electron microscopy (SEM). Phase identification was performed using mainly energy dispersive spectroscopy (EDS). X-ray diffraction was used to confirm phase identification for the as reacted pellets.

RESULTS AND DISCUSSION

Simultaneous Combustion

Table 1 summarizes the results of ignition temperature, some visual aspects and phase identification of the as reacted pellets according to nominal composition. Ignition temperature varies depending on the pellet compositions: 500°C for the 20 and 30 at.% Ni, and 850°C for the 10 at.% Ni. Shape variation of the pellet during the reaction was mainly observed for the 20 and 30 at.% Ni pellets.

$NbAl_3$ are present in all investigated pellet compositions as can be visualized in the micrographs of Fig. 1 (light gray areas). A Laves phase with composition Nb16Ni51Al was observed in 30 at.% Ni pellet and it corresponds to the fragmented white areas in the micrograph of Fig. 1a. For other compositions the white areas were identified as being unreacted Nb. The dark gray areas in the micrographs were identified as being NiAl containing 50 to 60 at.% Ni for the Nb30Ni60Al pellet (Fig. 1a), and Ni_2Al_3 for the other two pellet compositions (Figs. 1c and 1d). Fig. 1 also shows that the amount of each phase present in the microstructure depends on the pellet composition. The decrease in the nickel content leads to a decrease in NiAl and Ni_2Al_3 and an increase in the $NbAl_3$ contents. Low magnification views (not included) show that the volume fraction of unreacted Nb increases when the amount of Ni in the pellet is increased.

Table I: Ignition temperature, visual aspect and phases identified in the as reacted pellets.

Nominal Composition (at.%)			$T_{ignition}$ (°C)	Visual Aspects	Identified Phases
Nb	Ni	Al			
10	30	60	500	Partial fusion, large voids	NiAl (major), $NbAl_3$, NbNiAl (minor)
15	20	65	500	Small shape distortion	$NbAl_3$, Ni_2Al_3, Nb (minor)
20	10	70	850	Negligible distortion	$NbAl_3$(major),Ni_2Al_3, Nb (minor)

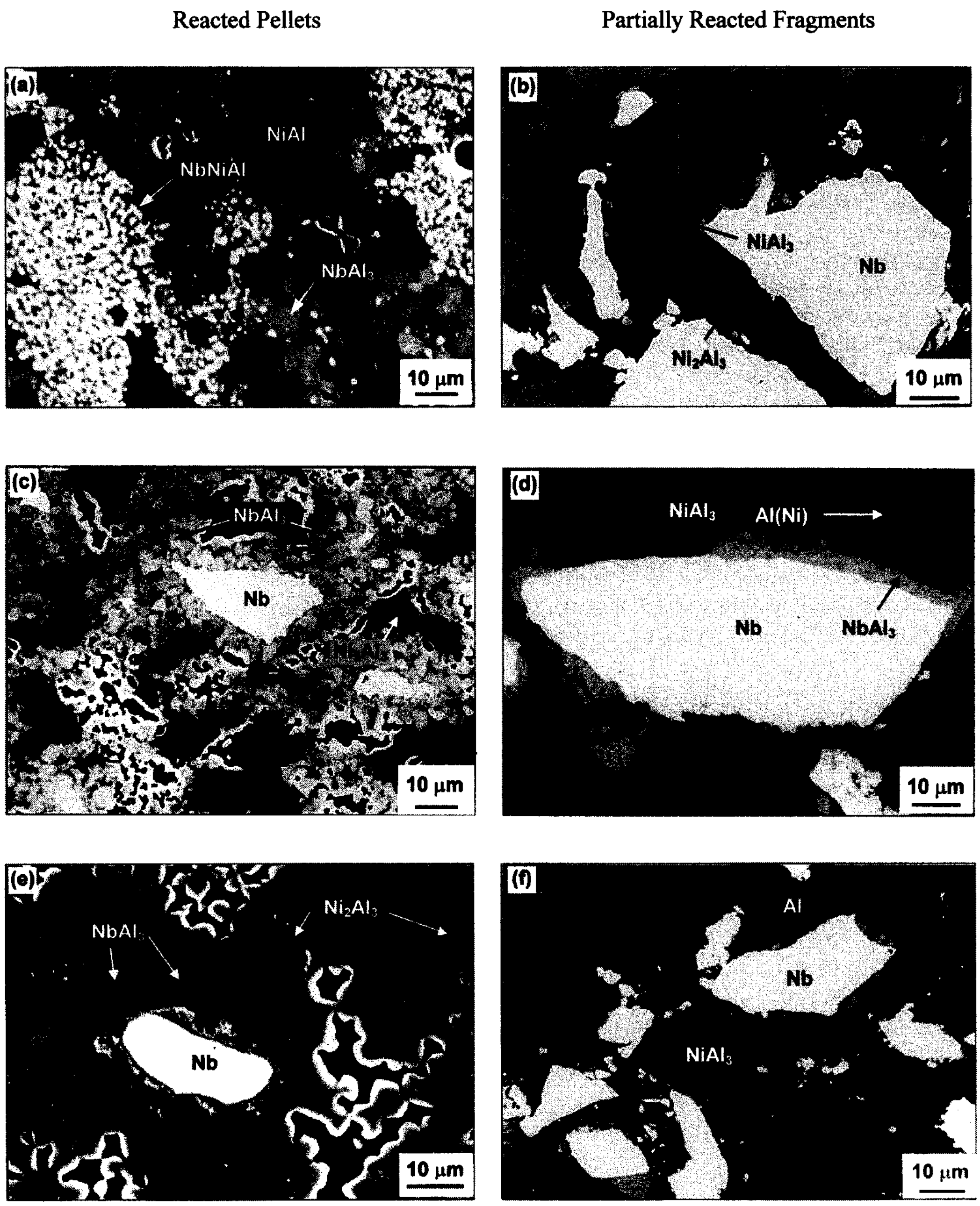

Figure 1: SEM micrographs of as reacted pellets and partially reacted fragments: (a) and (b) Nb30Ni60Al; (c) and (d) Nb20Ni65Al; (e) and (f) Nb10Ni70Al.

Differential Scanning Calorimetry

The results of DSC experiments are shown in Fig. 2 for the three investigated compositions. Results obtained for the $NbAl_3$ composition (Nb75Al) are also included in the figure for comparison. Two groups of peaks are generally observed for the tested compositions, the first around 600 °C and a second one around 900°C. For the Nb75Al composition the first peak around 660 °C is endothermic, and it corresponds to the melting of aluminum. The second peak around 900 °C is exothermic and it is associated with the $NbAl_3$ synthesis reaction as verified in previous investigations [3, 5]. Contrarily to what occurs with the $NbAl_3$ situation, in the Ni-containing compositions the first peak is exothermic and the second event can be decomposed on exothermic and endothermic subsequent events depending on the compact composition. In an attempt to understand the reactions occurring at temperatures up to 650 °C, small fragments of Nb-Ni-Al compacts with similar compositions were heated to this temperature, under vacuum and quickly cooled to room temperature. This temperature is sufficient to solely start the first exothermic reactions when small fragments are used. The results of a metallographic analysis performed on these fragments are shown in Fig. 1 (b, d, f). As it can be verified in the micrographs, in general the $NbAl_3$ synthesis reaction has been suppressed and the problem can be treated as a reaction between aluminum and nickel. The phases observed in the 10 at.% Ni are non-reacted aluminum and niobium, and $NiAl_3$. Besides unreacted Nb, in the 20 at.% Ni fragment the presence of $NiAl_3$ and a solid solution of nickel in aluminum containing between 1.1 to 2.9 at.% Ni are observed. In the 30 at.% Ni fragment, all the aluminum has been consumed and the phases $NiAl_3$ (minor), Ni_2Al_3 (major), and unreacted niobium are observed. The peaks shown in the DSC spectrum around 600°C can be well correlated with the reactions between Al and Ni for the formation of $NiAl_3$ and Ni_2Al_3 [7]:

$$Ni + 3Al \rightarrow NiAl_3 \quad (\Delta H^{\circ}_{298} = -37.66 \text{ kJ/mol}) \qquad (1)$$

and

$$NiAl_3 + Ni \rightarrow Ni_2Al_3 \quad (\Delta H^{\circ}_{298} = -56.48 \text{ kJ/mol}) \qquad (2)$$

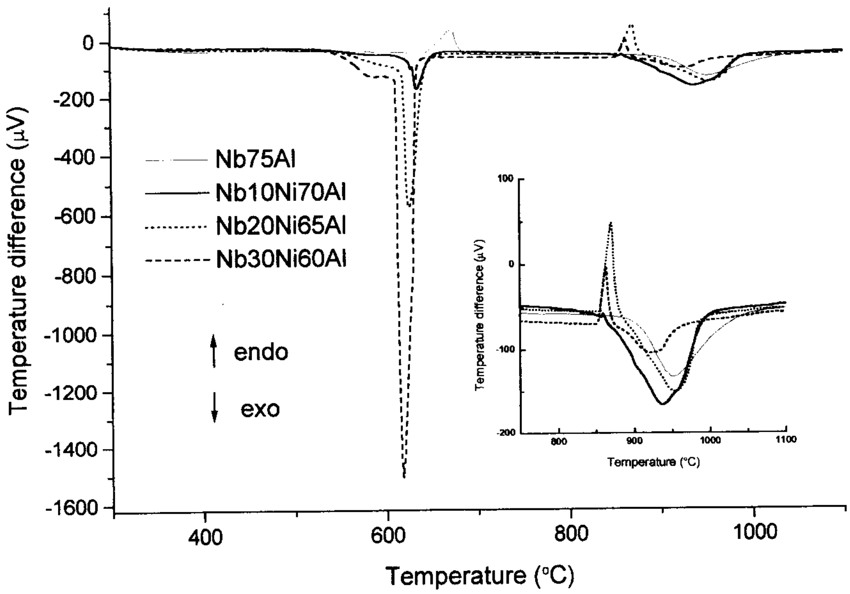

Figure 2: DSC curves for samples with different compositions investigated. Higher temperature peaks region is enlarged.

Reaction (1) has been observed to occur in all the three investigated fragments, being most intense in the 10 at.% Ni composition. Reaction (2) is only observed in the 20 at. % and 30 at.% Ni samples being most intense in the latter. The occurrence of similar reactions has also been reported during the first stages of Ni_3Al synthesis [8-10].

The peaks observed for these compositions in the DSC experiments around 800 °C can be associated with transformations occurring with the phases previously synthesized at lower temperature. Since Nb do not participate in the first reactions, the use of the Al-Ni phase diagram [11] is elucidative (Fig. 3). In fact, for the Nb10Ni70Al composition (Ni/Al = 0.14), one can verify that at 830°C all the $NiAl_3$ should dissolve in a liquid rich in aluminum. This dissolution takes place quasi-simultaneously with the $NbAl_3$ synthesis, which occurs through the reaction:

$$Nb + 3Al_{(liq.)} \rightarrow NbAl_3 \qquad (3)$$

For the composition Nb20Ni65Al (Ni/Al=0.31) the presence of the endothermic peak can be explained by the transformation that takes place at 854°C according to the reaction:

$$NiAl_3 \rightarrow Ni_2Al_3 + Al_{(liq.)} \qquad (4)$$

Above 854°C, Ni_2Al_3 coexists with a liquid containing around 15 at. % Ni. An exothermic reaction (3) then takes place above this temperature. For the Nb30Ni60Al composition (Ni/Al=0.50) it is observed that Ni_2Al_3 and $NiAl_3$ are stable up to 854°C. At this temperature the endothermic peak associated with the melting of $NiAl_3$ has lower intensity since the amount of this phase (27 wt.%) is now much small. The reaction (3) then takes place at higher temperatures.

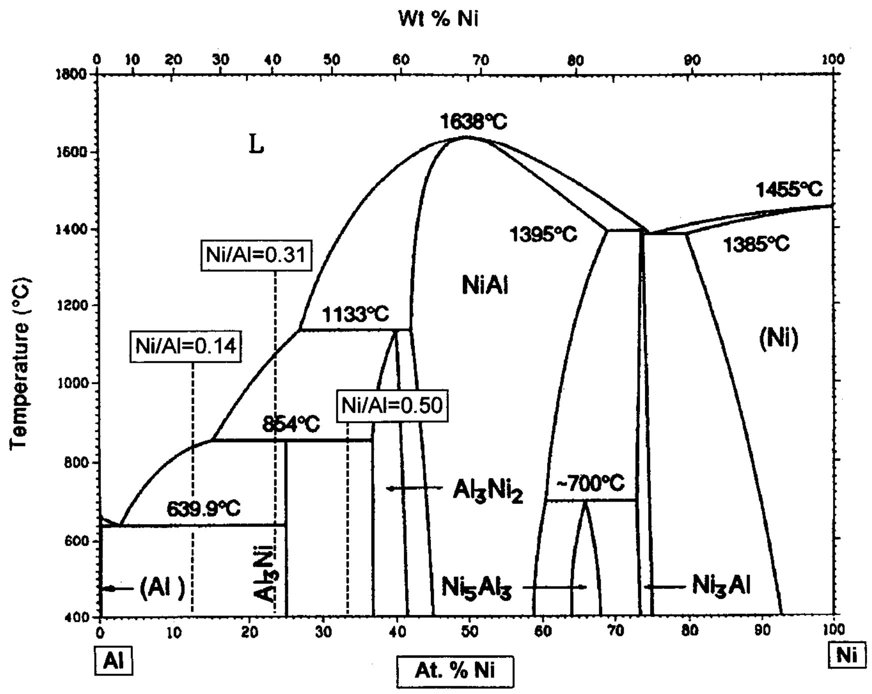

Figure 3: Phase diagram for the nickel-aluminum system [11].

NiAl present in Nb30Ni60Al samples can be formed from Ni_2Al_3 decomposition through the peritectic reaction at 1133 °C:

$$Ni_2Al_3 \rightarrow 2NiAl + Al_{(liq.)} \quad (5)$$

This reaction, not shown in Fig. 2, is detected in the DSC experiment.

The presence of NbNiAl phase in the Nb30Ni60Al reacted pellets could be explained by the high temperatures involved in this reaction. According to previous works [5,12] this Laves phase is obtained after long heat treatments at temperature of 1140 °C, indicating that it is an equilibrium phase contrarily to what is evidenced by the available section of the NbNiAl phase diagram [13].

CONCLUSIONS

The microstructural analysis performed on pellets (simultaneous combustion synthesized) and on partially reacted fragments of Nb-Ni-Al alloys evidenced that the synthesis reaction takes place in two stages. The first stage involves the reaction between Ni and Al, which starts at temperatures below the melting point of aluminum. Ni_2Al_3 and/or $NiAl_3$ can be formed as a consequence of this reaction, depending on the initial compact composition. Ni_2Al_3 can be formed either by the reaction of Ni_3Al with Ni or by the decomposition (peritectic reaction) of $NiAl_3$. The second stage involves the formation of $NbAl_3$ which takes place when local temperatures is around 850°C. The formation of $NbAl_3$ is initiated on the surface of the niobium particles. Depending on the relative amount of Ni and Al present in the compact, this first reaction can trigger the second reaction, leading to a drastic decrease of the compact ignition temperature.

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REFERENCES

[1] J. J. Moore and H. J. Feng, Prog. Mater. Sci. 39 (1995) p. 243-273.
[2] Z. A. Munir and U. Aselmi-Tamburini, Mater. Sci. Rep. 3 (1989) p. 277-365.
[3] P. I. Ferreira and R. M. Leal Neto, Int. J. Powder Metall., 30 (1994) p. 313-321.
[4]; J. G. B. Taveira, W. Inoue and P. I Ferreira, Proc. of the 11th Cong.Eng. Ci. Mat., Águas de São Pedro, SP, Brazil, Vol. 1., 1994. p. 107-110.
[5] R. M. Leal Neto and P. I. Ferreira, Mat. Sci. Eng. A, 192/193 (1995) p. 549-555.
[6] R. M. Leal Neto; S. Buganza and P. I. Ferreira, Proc.IV Semin. Met. do Pó, 1995, S. Paulo, S.P., Brazil, 1995. p. 313-323.
[7] R. Hultgren, Selected values of thermodynamic properties of metals and alloys. Metals Park, OH, ASM, 1973, p.192.
[8] K. A. Philpot, Z. A. Munir and J. B. Holt, J. Mater. Sci., 22 (1987) p. 159-169.
[9] C. Nishimura and C. T. Liu, Acta Metall. Mater. 41 (1993) p. 113-220.
[10] J-. P. Lebrat, A. Varma and P. J. Mcginn, J. Mater. Res., 9 (1994) p. 1184-1192.
[11] T. B. Massalski; J. L. Murray; L. H. Bennett and H. Baker, Binary alloy phase diagrams. Metals Park, OH, ASM, 1986, V.1, p.140.
[12] C. P. Reip; G. Sauthoff, Intermetallics, 1 (1993) p. 159-169.
[13] J. S. Benjamin; B. C Giessen; N. J. Grant, Trans. Metall. Soc. AIME, 236 (1966) p.224-226.

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