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Simultaneous synthesis and densification of NiAl and Ni_3Al by pressure-assisted combustion

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Simultaneous synthesis and densification of NiAl and Ni_3Al from elemental powders of Ni and Al was investigated. Combustion synthesis was carried out under the uniaxial compressive pressure and preheating of die. It was shown that both the compressive pressure and preheating of die strongly affect the density, the grain size and the hardness of NiAl and Ni_3Al product. A high relative density of NiAl and Ni_3Al products as much as 99.8% was achieved under compressive pressure of 275 MPa and preheating temperature of 600°C.

1. Introduction

The intermetallic compounds of NiAl and Ni_3Al have received considerable attention recently as a material for high-temperature application. Its properties provide a desirable combination of a high melting temperature, excellent oxidation resistance, increasing flow stress with increasing temperature, and a relatively lower density than commercial nickel-based superalloy [1–3].

Many processing methods have been used to fabricate NiAl and Ni_3Al compounds, including powder processing, casting, and self-propagating high temperature synthesis [4–6]. SHS of the methods offers several advantages, including the use of elemental powders, short processing times, and energy saving. On the other hand, the problem such as porosity has prohibited the wide spread use of combustion method. In order to reduce the porosity of Ni_3Al, optimizing reaction sintering parameters, including particle size, heating rate, atmosphere and pressure was investigated [7–9].

In this work, to produce dense material of NiAl and Ni_3Al during combustion synthesis, the use of uniaxial pressure and preheating of die is demonstrated. And mechanical properties of NiAl and Ni_3Al products are also presented and discussed.

2. Experimental procedure

The material used in this work were 99% pure aluminium powder and 99% pure nickel powder with a sieve classification of −325 (∼44 μm) obtained from Chang-Sung co. (South Korea) and NiAl powder with a particle size of 10 μm as diluent obtained by crushing combustion-synthesized NiAl product using ball mill. The powder were mixed to give the two different stoichiometric composition, (Ni : Al = 1 : 1) and (Ni : Al = 3 : 1). The powder mixtures were then poured into a cylindrical steel die after being coated with a thin layer of boron nitride along areas of contact with the powder. The die was preheated at the temperature ranging from 200°C to 600°C using preheating furnace under 1 atm pressure of Ar gas. As soon as the powder mixture was ignited by tungsten ignition coil, the hydraulic rams were applied to the desired pressure ranging from 0 MPa to 275 MPa at set preheating temperature.

The relative density and porosity of the synthesized sample was measured using the Archimedes method and image analyzer. The product samples were polished and etched using a solution of HNO_3(33.3 vol%), CH_3COOH(33.3 vol%) and HCl(33.3 vol%). The grain size was determined from the micrographs using the linear-intercept technique [10]. Analysis of the products was made through x-ray diffraction, and microstructural analysis was made by optical microscopy. Vickers microhardness (under 400 g force) was made on the synthesized NiAl and Ni_3Al samples.

3. Results and discussion

3.1. Effect of preheating and pressure on densification

NiAl and Ni_3Al compounds were synthesized from elemental powder of Ni/Al and 3Ni/Al stoichiometric ratio respectively by pressure-assisted combustion. These phase was confirmed by analysis of XRD pattern. Optical micrographs of the NiAl with compressive pressure during combustion without preheating of die are shown in Fig. 1. The sample Fig. 1a without a compressive stress shows structure with large pore and porosity of about 50%. Fig. 1b and c with compressive stress show much less porosity and smaller pore as compared with the sample without compression Fig. 1a. The variation of porosity of NiAl and Ni_3Al

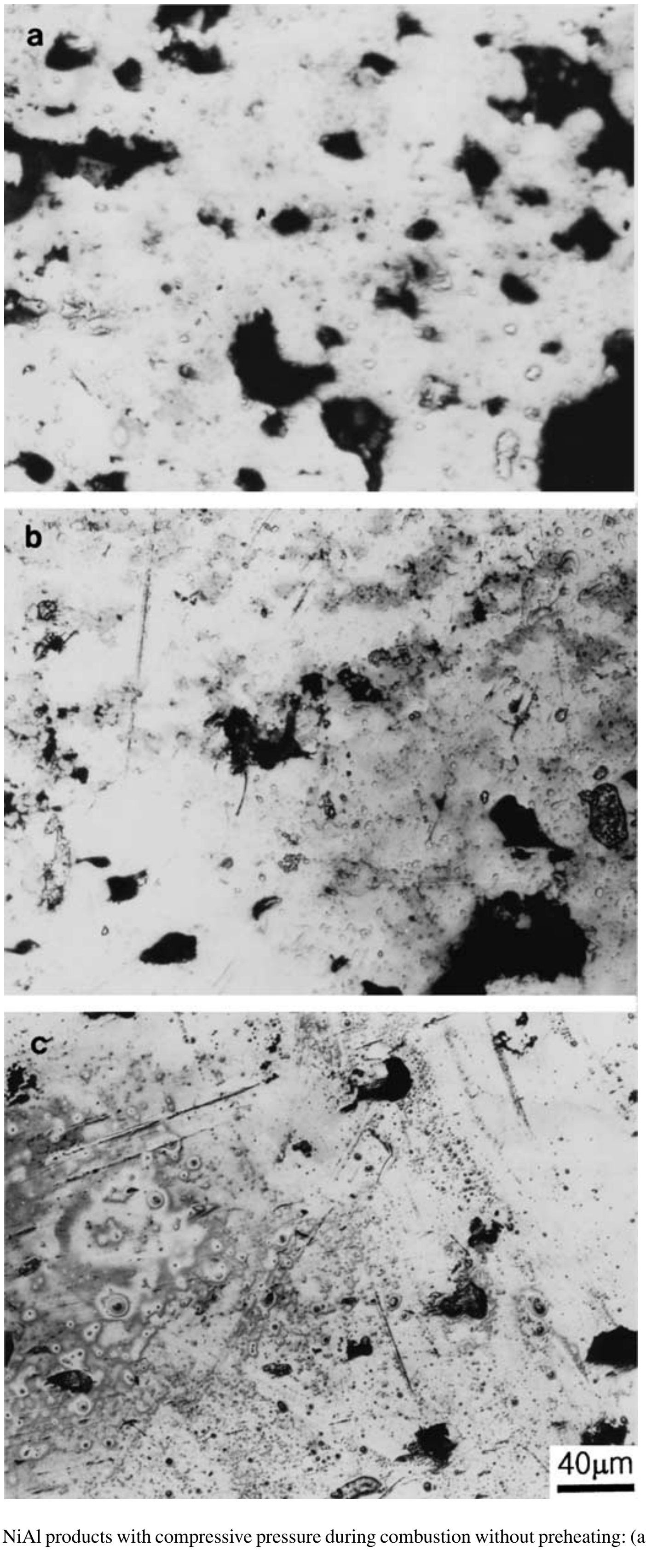

Figure 1 Optical micrographs of NiAl products with compressive pressure during combustion without preheating: (a) Without Pressure; (b) 135 MPa; (c) 275 MPa.

products with compressive pressure without preheating of die is shown in Fig. 2. The porosity of NiAl and Ni_3Al decreases with the imposition of compressive stress. And the porosity of NiAl is lower than that of Ni_3Al at the same compressive pressure. The nickel aluminides compound formation generates heat, which further accelerates the reaction, and lead to extensive liquid formation. The liquid quickly spreads throughout the structure, and compound formation continues until the reactants are consumed [11]. It is considered that the aluminum involved in the combustion reaction leaved considerable porosity unless the applied pressure is high enough to plastically deform the structure. Hence, higher liquid contents achieved with greater aluminum concentration of NiAl system show rather higher porosity at low applied pressure as shown in Fig. 2.

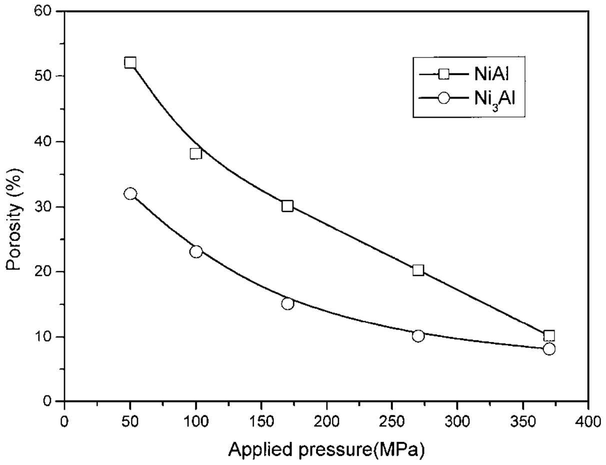

Figure 2 The variation of porosity of NiAl and Ni_3Al products with applied pressure without preheating.

Also, the deviation of the porosity between NiAl and Ni_3Al decreased as the applied pressure increased. This means that the liquid more actively spreads into the pores with deformation of structure and provides an interconnected liquid structure. However, it is hard to obtain near full density without preheating of die. It is considered that this is ascribed to the fact that contact with steel die under pressure apparently suppresses the self-sustaining reaction due to an enhanced heat transfer from sample to steel die by heat conduction. To reduce heat loss through steel die, the die is preheated at the temperature ranging from 200°C to 600°C. Optical micrographs and variation of porosity of the NiAl product with preheating temperature and compressive pressure during combustion are shown in Figs 3 and 4 respectively. Porosity of NiAl decreases with increasing of compressive stress and preheating temperature.

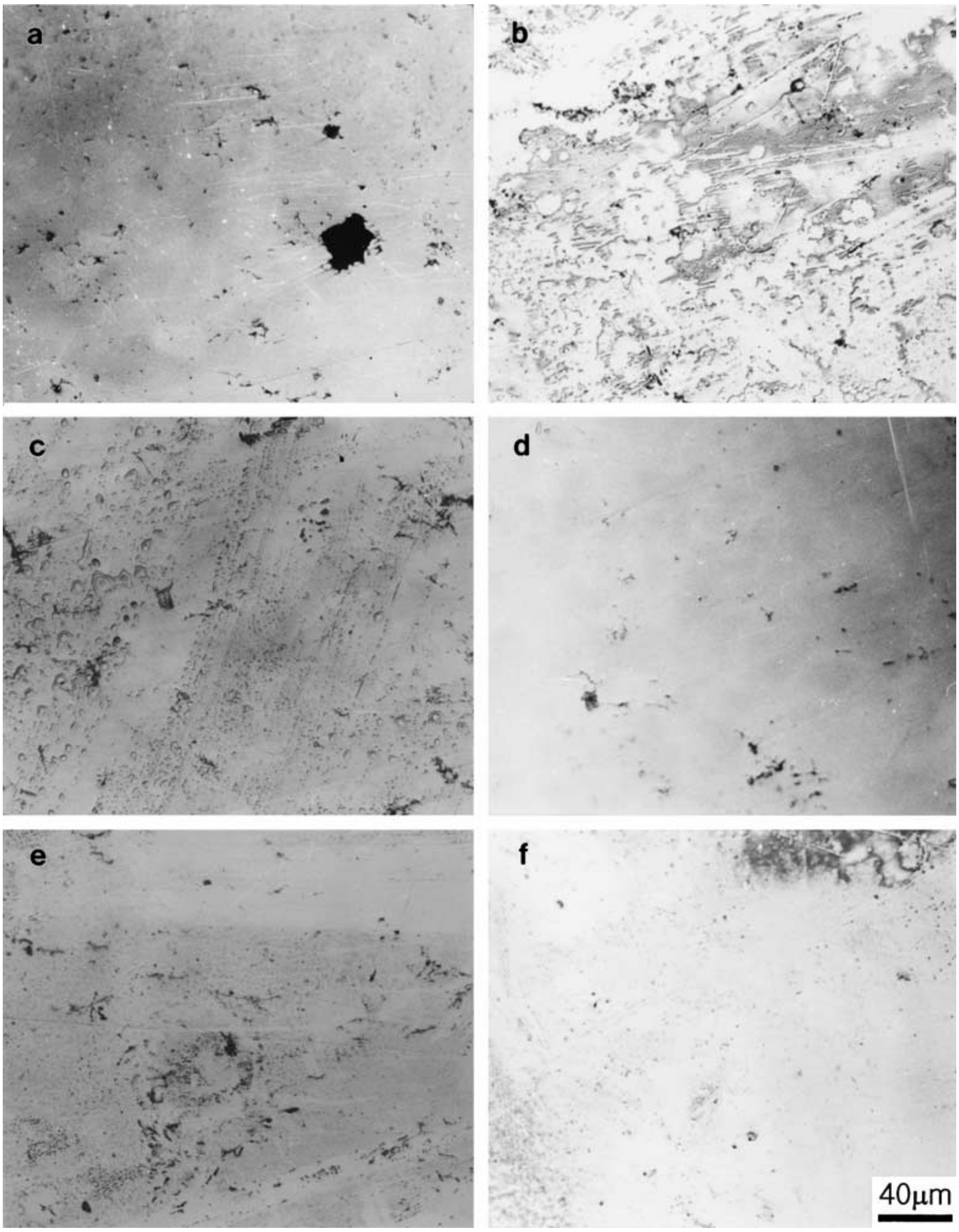

Figure 3 Optical micrographs of NiAl products with preheating temperature and compressive pressure during combustion: (a) 200°C, 135 MPa; (b) 200°C, 275 MPa; (c) 400°C, 135 MPa; (b) 400°C, 275 MPa; (e) 600°C, 135 MPs; (f) 600°C, 275 MPa.

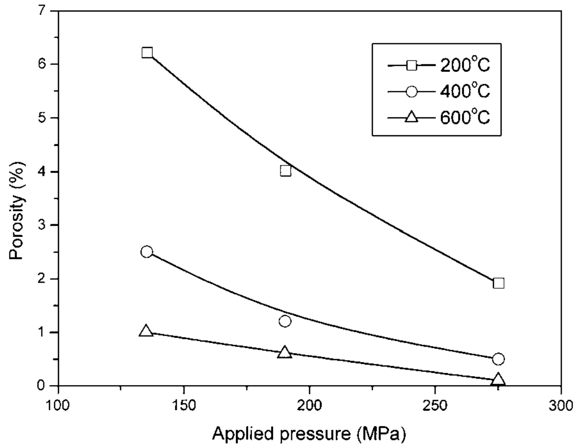

Figure 4 The variation of porosity of NiAl product with preheating temperature and compressive pressure.

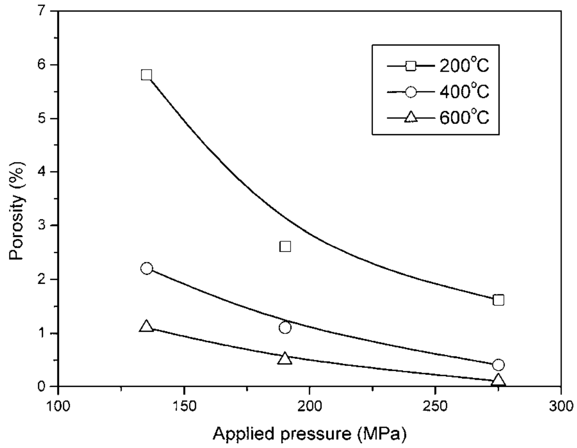

Figure 5 The variation of porosity of Ni_3Al product with preheating temperature and compressive pressure.

This result from preheating effect suggests that the formation of a high-temperature liquid associated with a self-sustaining reaction is a major factor for densification. The variation of porosity of Ni_3Al with preheating temperature and compressive pressure is shown in Fig. 5. The observed trend is similar to that of NiAl. A high relative density of NiAl and Ni_3Al product as much as 99.8% was achieved under compressive pressure of 275 MPa at preheating temperature of 600°C.

3.2. Effect of preheating on grain size

Optical microstructures of NiAl products with various preheating temperature are shown in Fig. 6. The grain size noticeably increases with preheating temperature. The variation of grain size of NiAl and Ni_3Al with preheating temperature is shown in Fig. 7. The grain size linearly increases from about 5 μm to about 25 μm with preheating temperature. The variation of grain size with preheating temperature can be explained as follow. Phase transformation consists of nucleation and grain growth which depend on supercooling. And supercooling also depends on preheating temperature. Therefore it is considered that variation of grain size is ascribed to degree of supercooling associated with variation of preheating temperature. The effect of compressive pressure on grain size is shown in Fig. 8. Compressive stress has little effect on grain size. The effect of NiAl diluent content on the grain size of NiAl product is shown in Fig. 9. The grain size decreases with an addition of NiAl diluent due to the fact that combustion temperature decreases with increasing of diluent.

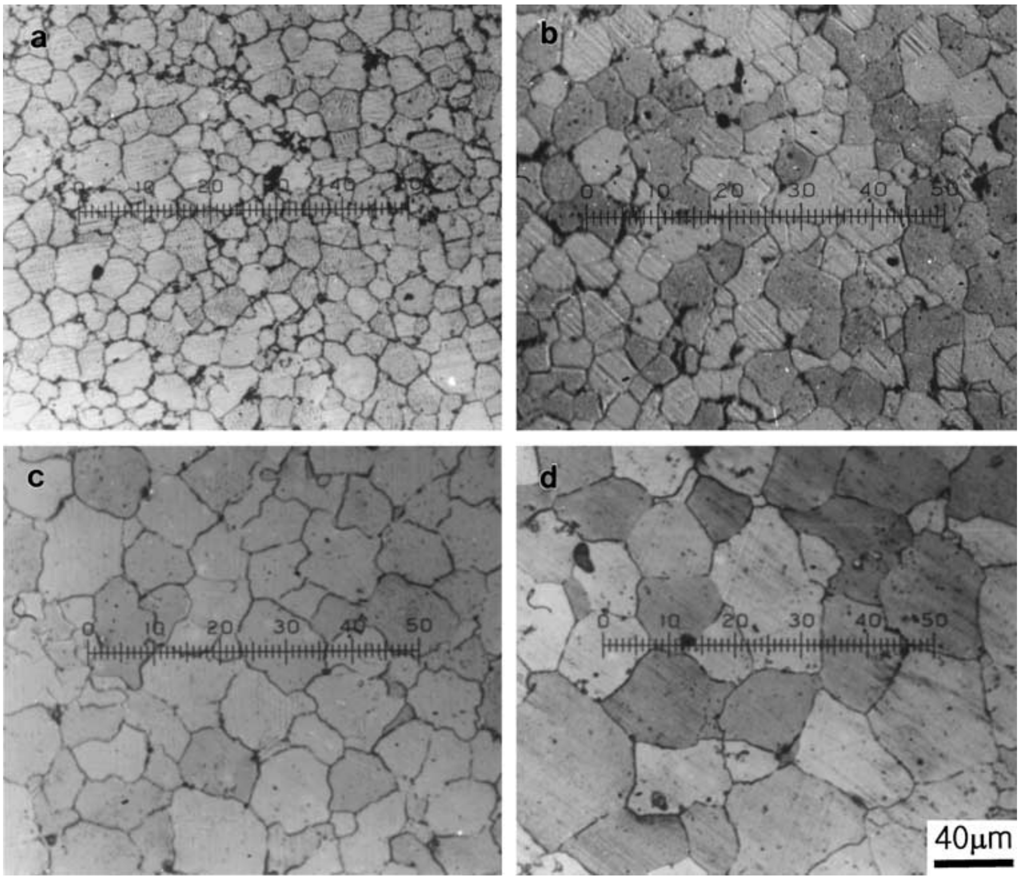

Figure 6 Optical microstructures of NiAl products with various preheating temperature at compressive pressure of 135 MPa: (a) Without preheating; (b) 200°C; (c) 400°C; (d) 600°C.

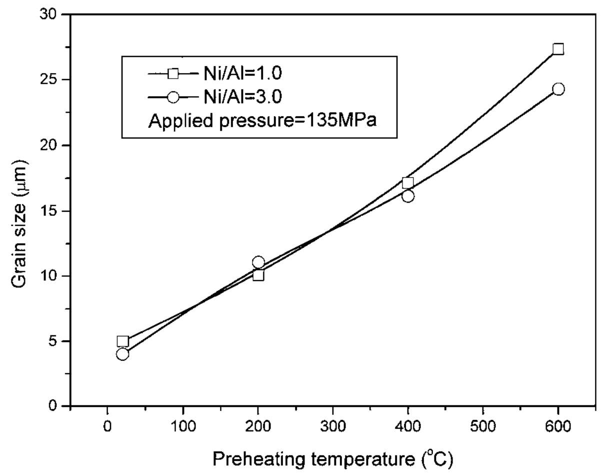

Figure 7 The effect of the preheating temperature on the grain size of NiAl and Ni_3Al products under compressive pressure of 135 MPa.

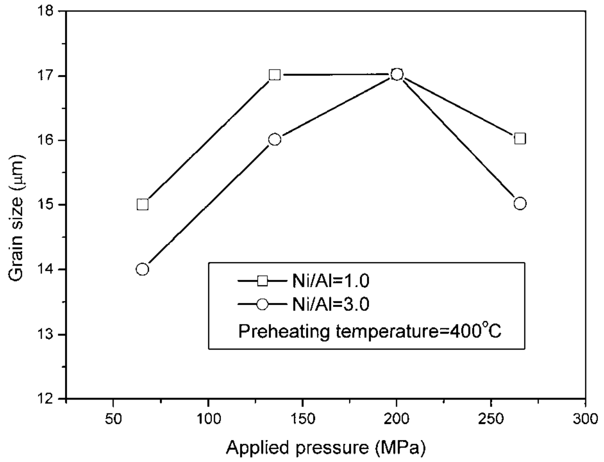

Figure 8 The effect of compressive pressure on the grain size of NiAl and Ni_3Al at preheating temperature of 400°C.

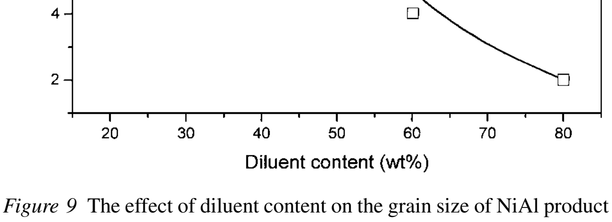

Figure 9 The effect of diluent content on the grain size of NiAl product under compressive pressure of 135 MPa at preheating temperature of 400°C.

3.3. Mechanical behavior

The variation of Vickers micro-hardness of NiAl and Ni_3Al products with preheating temperature and compressive pressure is shown in Fig. 10. Vickers hardness of NiAl and Ni_3Al increases with increasing of preheating and compressive pressure. This result is closely related to the fact that the porosity of NiAl and Ni_3Al decreases with increasing of preheating and compressive stress.

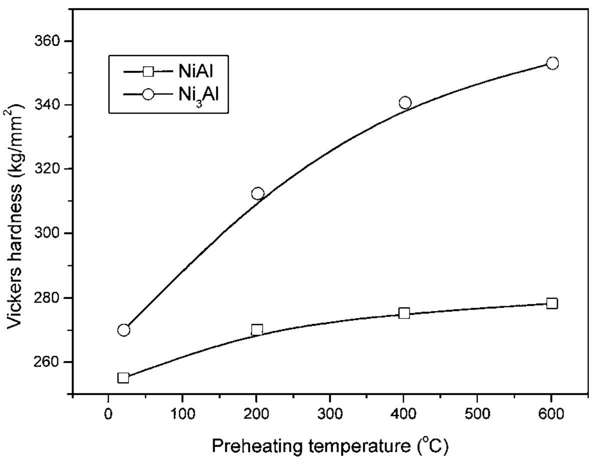

Figure 10 The variation of Vickers hardness with preheating temperature (applied pressure: 280 MPa, punch speed: 2.0 cm/sec).

4. Conclusion

Our results have clearly demonstrated that compressive pressure and preheating of die strongly affect the density, the grain size, and the hardness of NiAl and Ni_3Al products. A high relative density of NiAl and Ni_3Al products as much as 99.8% was achieved under compressive pressure of 275 MPa and at preheating temperature of 600°C. Vickers hardness of the NiAl and the Ni_3Al products was closely related to porosity of these products with preheating temperature and compressive pressure. The hardness of NiAl and Ni_3Al products increased with increasing of preheating temperature and compressive pressure.

References

1. S. MIURA, S. OCHIAI, Y. OYA, Y. MISHIMA and T. SUZUKI, in "High-Temperature Ordered Intermetallic Alloys," edited by C. T. Liu, A. I. Taub, N. S. Stoloff and C. C. Koch (MRS, Pittsburgh, 1988) p. 341.
2. W. A. KAYSSER, R. LAAG, J. C. MURRAY and G. E. PETZOW, *The International Journal of Powder Metallurgy* **27** (1991) 43.
3. J. A. NESBITT, E. J. VINARCIK, C. A. BARRETT and J. DOYCHAK, in "High Temperature Aluminides and Intermetallics," edited by S. H. Whang, D. P. Pope and C. T. Liu (Elsevier Science Publishers Ltd., England, 1992) p. 561.
4. A. S. MURTHY and E. GOO, *Acta Metall. Mater.* **41** (1993) 2135.
5. ZHOU YIZHANG, ZHANG TIANXIANG, TONG YINGJIE, ZHANG BINGDA, ZHU YAOXIAO and HU ZHUANGGI, in "High-temperature ordered Intermetallic Alloys," edited by C. T. Liu, A. I. Taub, N. S. Stoloff and C. C. Koch (MRS, Pittsburgh, 1988) p. 555.
6. D. E. ALMAN and N. S. STOLOFF, *The International Journal of Powder Metallurgy* **27** (1991) 29.
7. C. NISHIMURA and C. T. LIE, *Acta Metall. Mater.* **41** (1993) 113.
8. A. BOSE, B. H. RABIN and R. M. GERMAN, *Powder Metall. Int.* **20** (1988) 25.
9. D. M. SIMS, A. BOSE and R. M. GERMAN, *Prog. Powder Metall.* **43** (1987) 575.
10. M. I. MENDELSON, *J. Amer. Ceram. Soc.* **52** (1969) 443.
11. B. H. RABIN, A. BOSE and R. M. GERMAN, in "Combustion and Plasma Synthesis of High-Temperature Materials," edited by Z. A. Munir and J. B. Holt (VCH, New York, 1990) p. 114.

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