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A study of densification and microstructure evolution during hot pressing of NiAl/Al_2O_3 composite

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Evolution of the density and the microstructure during hot pressing of NiAl/Al_2O_3 composite has been investigated in the present paper. In particular, the effect of the process parameters, viz. compacting pressure, sintering temperature and sintering time, on the evolution of the density of the intermetallic–ceramic composite has been studied. Evolution of the density has been related to microstructure changing. Porosity, pore structures and grains rearrangement have been analysed in microscopic observations.

Keywords: hot pressing; sintering; intermetallic–ceramic composite; density evolution; microstructure

1. Introduction

Intermetallic-based composites reinforced with ceramic particles belong to novel materials with advantageous properties, which make these materials attractive for a wide range of industrial applications. NiAl matrix composite reinforced with Al_2O_3 particles is characterized by excellent mechanical and thermal properties, as well as high frictional wear resistance.[1–3] It can be used instead of steel or metal alloys in aerospace and automobile industries for elements working in conditions of intense friction (brake discs, clutches, cranes and valves) and for parts subjected to rapidly changing temperatures (nozzles, chambers combustion, engine guards and exhaust systems).

Hot pressing is widely used to produce intermetallic NiAl composites with ceramic reinforcements.[1,4–9] Densification is one of the main macroscopic effects in the manufacturing process caused by shrinkage and pore elimination in the sintered material. Densification during sintering is accompanied by complex microstructural changes. Evolution of the relative density is a good indicator of sintering kinetics and a useful parameter to predict the mechanical properties of the sintered component.[10–12]

Manufacturing and properties of intermetallic NiAl composites with ceramic reinforcements have been investigated in several papers, cf.[1,2,4–9,13,14] Properties of the composites can be tailored according to application requirements by taking suitable phase composition and designing specific microstructure.[15,16] Control of the

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manufacturing process and the understanding of the microstructure evolution during this process is essential in the optimization of the intermetallic–ceramic composites. Investigation of the intermetallic–ceramic composite properties and manufacture process, with its complexity, has become a major task of material science.

The main objective of the present work has been to investigate densification and microstructure evolution during hot pressing of $NiAl/Al_2O_3$ composite performed under different process parameters. Composite specimens have been manufactured at different process parameters like pressure, sintering temperature and sintering time. The density measurements for specimens with different levels of sintering have given the data-set of the density evolution. The microstructure observations complementing our studies are helpful to understand sintering mechanisms.

The outline of the paper is as follows. Section 2 presents the details of the specimen manufacturing process. Results of density measurements are given and discussed in Section 3. Analysis of microstructure evolution is done in Section 4.

2. Specimen manufacturing process

Intermetallic–ceramic composite specimens were manufactured from a mixture of nickel aluminide (NiAl) powder delivered by the Goodfellow Company and aluminium oxide (Al_2O_3) powder from NewMet Koch. Morphology of these powders is shown in Figure 1.

The grain size of the starting powders was analysed with the Clemex image analysing system. The average Feret diameter $d^{avg}_{NiAl} = 9.71\ \mu m$ was estimated for the NiAl powder and $d^{avg}_{Al_2O_3} = 2.28\ \mu m$ for the Al_2O_3 powder. The powder mixture prepared for technological experiments contained 20% volume fraction of Al_2O_3 and 80% of NiAl. This composition was taken after theoretical studies of mechanical properties of components of composite materials, taking into considerations the requirements of possible application in internal combustion engine as valve seat. In order to achieve uniform grain distribution in the whole volume, the powders were mixed in a Pulverisette 6 planetary mill in an air atmosphere with the following mixing process parameters: rotational speed $\omega = 100$ rpm, BPR coefficient 5:1 and time 1 h. The mixing conditions were chosen after earlier experiments of authors.[7,17]

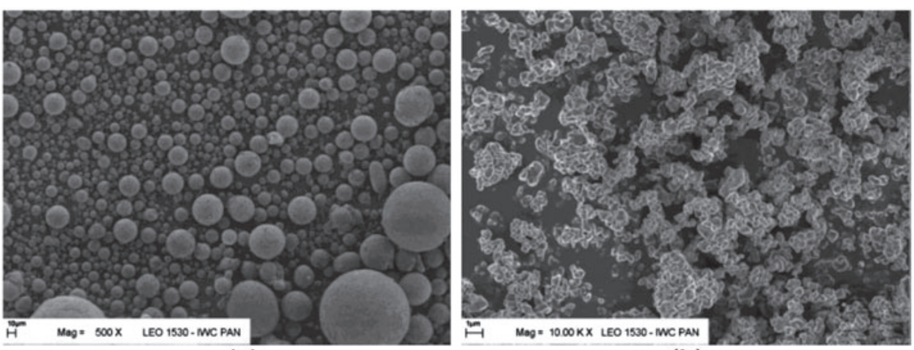

Figure 1. SEM images of the powders: (a) NiAl and (b) Al_2O_3.

The powder mixture was sintered under pressure in an ASTRO HP50-7010 press using hot-pressing procedures in an argon protective atmosphere. The temperature and pressure profiles during sintering process are presented in Figure 2. The temperature was increased up to the sintering temperature T_s. Specimens were kept at T_s during the interval (sintering time) t_s and cooled to room temperature. The following sintering process parameters were employed in the present study:

- sintering temperature T_s – 1300, 1350 and 1400 °C,
- sintering time t_s – 0, 10 and 30 min,
- pressure p – 5 and 30 MPa.

$NiAl/Al_2O_3$ specimens were manufactured at different combinations of the above parameter values.

The sintered cylindrical specimens are shown in Figure 3. Bulk densities of the sintered composite material were measured using the hydrostatic method. The relative density, ρ_{rel}, was calculated as the ratio of the measured bulk density ρ and the theoretical density ρ_{theo} for the fully dense material:

$$\rho_{rel} = \frac{\rho}{\rho_{theo}} \tag{1}$$

3. Evolution of density

Results of density measurements are given in Table 1. Density evolution curves for different combinations of temperature and pressure are plotted in Figure 4 together with corresponding images of the composite microstructure. It can be noticed from these tables and figures that all the three technological parameters (temperature, time and pressure) have a strong influence on the degree of material densification.

The results confirm quite an obvious effect of sintering time on the densification. As it can be expected the density grows with the time of the process. Density growth is relatively flat, although it can be seen that it is slower when the bulk density approaches full density.

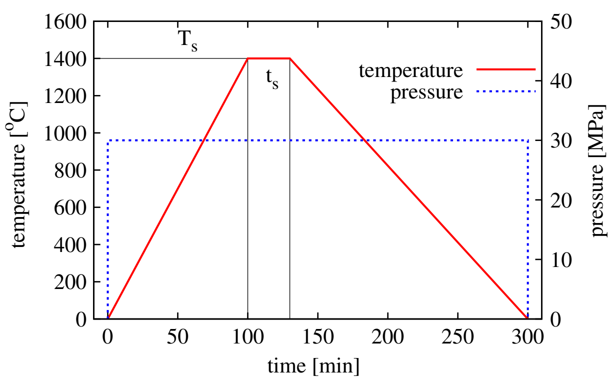

Figure 2. Temperature and pressure profiles for the sintering process of T_s = 1400 °C, t_s = 30 min and p = 30 MPa.

Figure 3. Sintered $NiAl/Al_2O_3$ specimens.

Table 1. Evolution of the bulk and relative density of the hot-pressed $NiAl/Al_2O_3$ composite manufactured under pressures of 5 and 30 MPa (NiAl/20% Al_2O_3 theoretical density – 5.52 g/cm^3).

Sintering temperature	t_s = 0 min		t_s = 10 min		t_s = 30 min	
T_s [°C]	ρ [g/cm^3]	ρ_{rel}	ρ [g/cm^3]	ρ_{rel}	ρ [g/cm^3]	ρ_{rel}
External pressure – p = 5 MPa						
1300	3.86	0.70	4.01	0.73	4.10	0.74
1350	4.02	0.73	4.16	0.75	4.42	0.80
1400	4.11	0.74	4.28	0.78	4.92	0.89
External pressure – p = 30 MPa						
1300	4.99	0.90	5.03	0.91	5.44	0.98
1350	5.24	0.95	5.35	0.97	5.50	0.99
1400	5.27	0.95	5.37	0.97	5.51	0.99

The results also show an increase of density with an increase of compaction pressure. Greater pressure corresponds to a higher density of the sintered material. The composites manufactured under a pressure of 5 MPa reached 89% relative density for the highest temperature and longest sintering time used in experiments. Increase of pressure to 30 MPa resulted in much higher degree of densification even at lower temperatures. Effect of compacting pressure on densification can be explained by easier grain regrouping process at the early stage, intensification of diffusion processes under pressure, activation of diffusion flows and, finally, easier pore elimination at the final stage of sintering.

Sintering is a thermally activated process; so the temperature plays an important role in it.[12,15] Sintering temperature is dependent on the physico-chemical properties of sintered powders, grain size and shape. Naturally, it is assumed that the sintering temperature of a unary system is 0.6–0.8 of the material melting point. In case of multiphase materials, the estimation of sintering temperature is more complicated. It is related to volume fractions of components, their solubility and wettabillity, and the surface energy connected with the global geometry of grains and specific surface area. The mass flow is strictly correlated to sintering temperature. Depending on the sintering temperature different mass transport mechanisms (surface diffusion, evaporation/condensation, grain boundary diffusion, viscous flow and volume diffusion) are dominant;

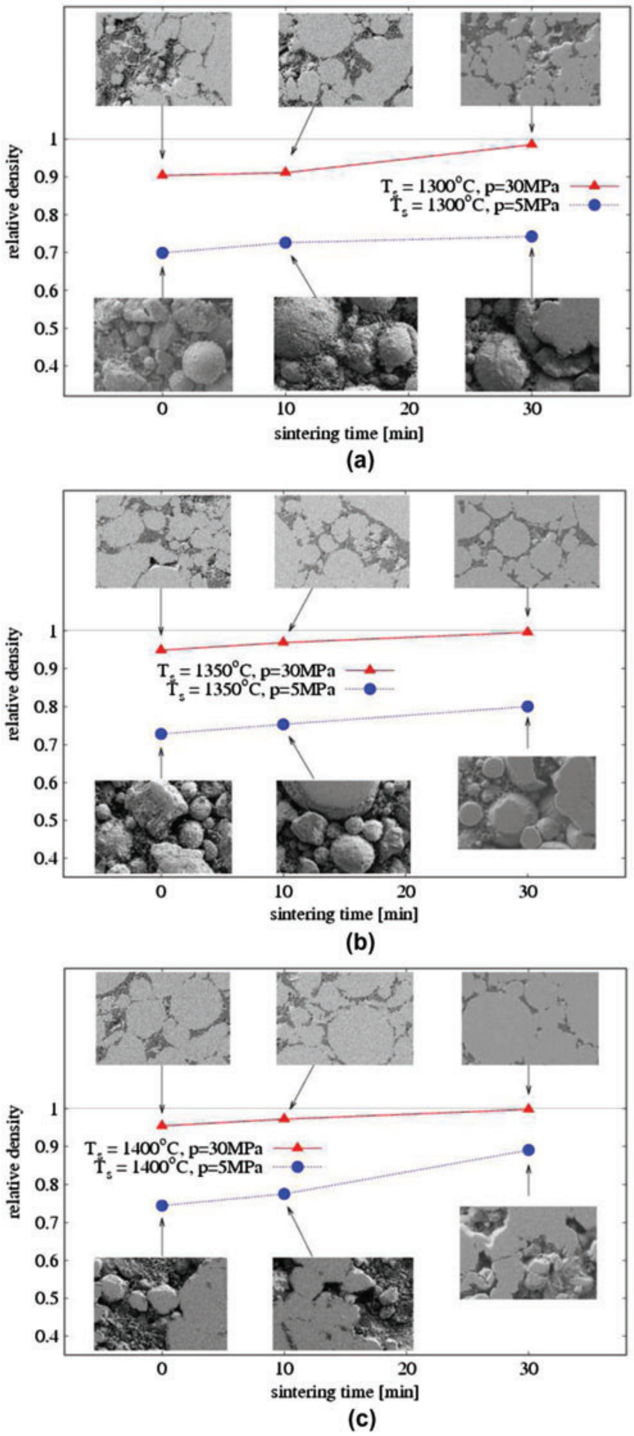

Figure 4. Density evolution – experimental results of $NiAl/Al_2O_3$ composite sintering with pressures 5 and 30 MPa in sintering temperature of: (a) $T_s = 1300$ °C, (b) $T_s = 1350$ °C and (c) $T_s = 1400$ °C.

e.g. surface diffusion dominates low-temperature sintering. Mass transport mechanism is an important factor in the densification of the sintered material.

It can be seen from Table 1 that for a constant sintering pressure, when the sintering temperature increases from 1300 to 1350 °C and 1400 °C, the sintered density remarkably increases. When the sintering process was performed below 1300 °C the specimen material had no integrity, indicating that sintering does not occur. Improvement of the densification with the increase of sintering temperature can also be observed in the plots in Figure 4.

4. Evolution of microstructure

A closer insight into microstructure development is provided by the SEM images in Figures 5–9 showing the microstructure details from the initial- to final-stage sintering. In the densification process during sintering we can distinguish the following stages (by different authors [18,19]): adhesion contacts, grain rearrangement and repacking, necks' formation at the initial stage of sintering, intermediate stage with the necks' growth and pore rounding, and the final stage with the pores' elimination. Duration and intensity of each stage can differ for different materials and various technological conditions of sintering process. Before starting the sintering process, the particles have to be in contact. It was estimated that the density of the green samples was 68%. The movement of particles in the direction of their dense packing occurs due to the applied external load. It was confirmed by the indications of punch displacement sensor. Beginning of sintering process results in enlargement of grains contact surfaces and decrease of pores. The forming of first necks between intermetallic particles is shown in Figure 5. At the beginning necks are small.

One can notice that the behaviour of ceramic particles is affected at this stage of sintering. Individual aluminium oxide grains are located on the surface of the intermetallic particles. The bonds are rather weak, the aluminium oxide grains are mechanically pressed into the NiAl and some deformation of NiAl surfaces is visible. The great amount of ceramic grains is placed at triangle contacts of NiAl, where they form agglomerates and their sintering starts.

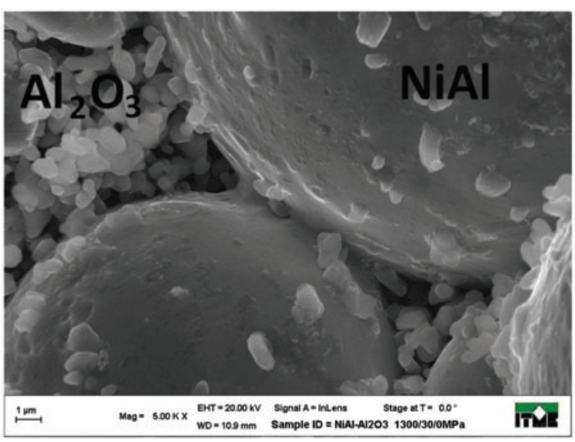

Figure 5. Formation of necks between NiAl grains ($T_s = 1400$ °C, $t_s = 30$ min and $p = 5$ MPa).

The intermediate stage is the most important stage for densifcation and determining the properties of sintered materials. It is characterized by simultaneous pore rounding, densification and grain growth.[20] This stage of sintering is controlled by diffusion processes. As it can be seen in the Figure 6(a), with the increase of temperature the necks are growing and the more compact structure is formed. There are visible bonded grains in systems consisting of more than two grains. The average distance between the adjacent grains decreases and the size of necks is enlarging. It is a typical behaviour of sintered materials described by other authors and it is consistent with our observations.[16,19–21] The ceramic grains are also connected between themselves. The rise of temperature to 1400 ºC results in sintering of Al_2O_3 particles (Figure 6(b)). At this stage we can observe bonding ceramic reinforcement with NiAl matrix too. In some cases they appear in the necks on the grain boundary of sintered NiAl grains (Figure 7).

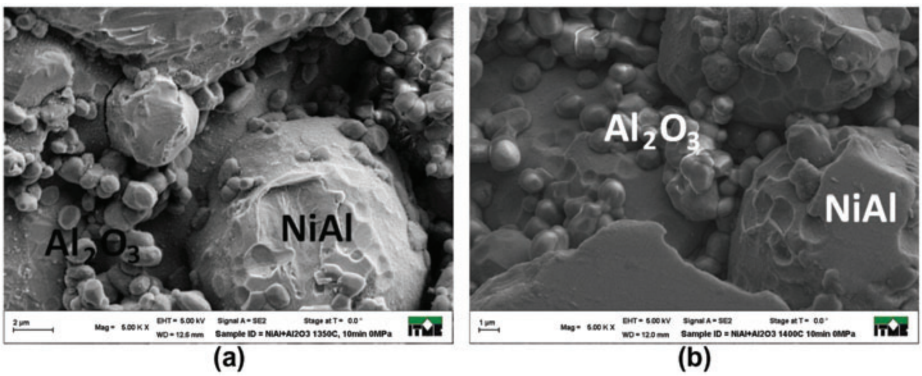

Figure 6. Intermediate stage of NiAl/Al_2O_3 composite material sintering: (a) $T_s = 1350$ °C, $t_s = 10$ min, $p = 5$ MPa and (b) $T_s = 1400$ °C, $t_s = 10$ min, $p = 5$ MPa.

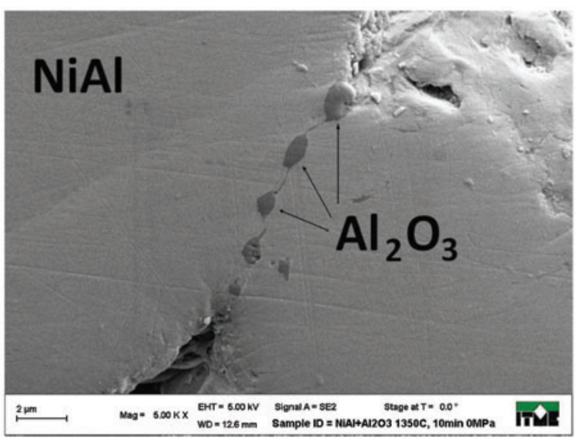

Figure 7. Al_2O_3 distribution on the grain boundary of intermetallic particles.

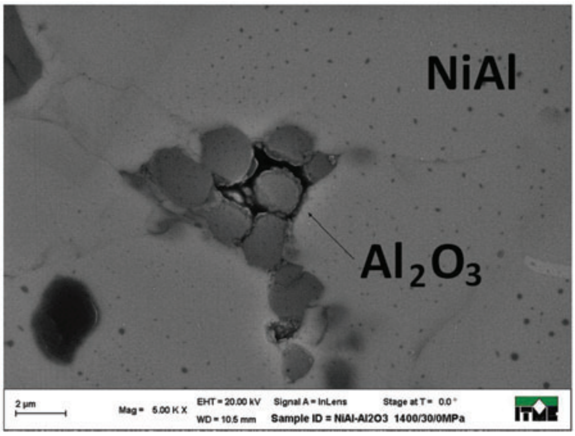

Figure 8. Channel pores in NiAl–Al_2O_3 composite structure ($T_s = 1400\,°C$, $t_s = 30$ min, $p = 5$ MPa).

At the beginning of this stage the pore volume amounts to over 20% of the total volume. Finally, it decreases to a few percent. Pores with irregular shapes become channel pores (Figure 8). This proves the earlier investigation on the theory of sintering provided by German [20]. They form a network which is limited by the surfaces of three adjacent grains and node, where the four channels meet. Enlargement of neck size leads to a progressive decrease in the number of pores.

The final sintering stage is characterized by pore elimination from the composite structure. Compared with the initial and intermediate stages, final-stage sintering is relatively a slow process. A complex interaction between particles, pores and grain boundaries is decisive for the final densification. According to the literature these interaction can take three forms. Pores retard grain growth to a point causing the grain boundary to bow and pull against a slow-moving pore. Pores can be dragged by the moving grain boundaries, or the grain boundaries can break away from the pores, leaving them isolated in the grain interior.[20] Some porosity exists between the ceramic particles

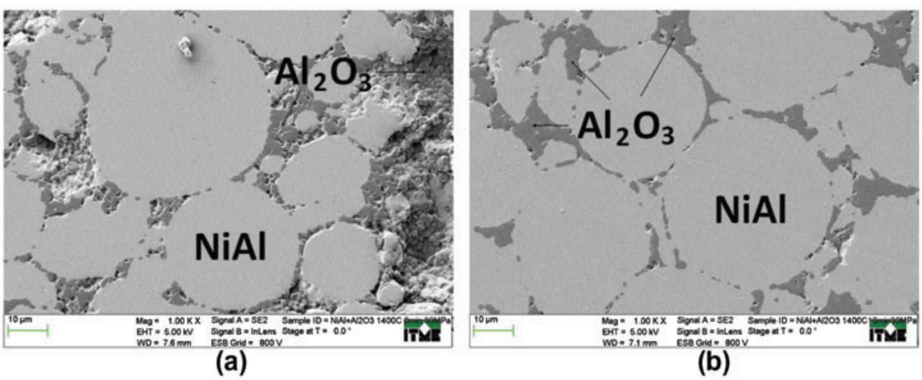

Figure 9. Final stage of sintering – pores elimination: (a) $T_s = 1400\,°C$, $t_s = 0$ min, $p = 30$ MPa and (b) $T_s = 1400\,°C$, $t_s = 10$ min, $p = 30$ MPa.

even when the process ends. In our case, we observed some structure discontinuities at the area of metal–ceramic interface, which can have a significant effect on the materials' properties. It should be underlined that finally we obtained almost fully dense materials with low amount of porosity (less than 1%).

5. Conclusions

A comprehensive study of densification and microstructure evolution during hot pressing of an intermetallic–ceramic NiAl/Al_2O_3 composite has been presented in this work. Increase of pressure and sintering temperature is favourable to the sintering kinetics in hot pressing of NiAl/Al_2O_3; however, the pressure increase seems to have a bigger impact than the increase of temperature. Increase of external pressure resulted in a significantly higher density even at lower temperatures. The effect of pressure can be explained by its favourable influence on crucial sintering mechanisms: grains regrouping, activation of diffusion flows and pores' elimination. By taking a pressure of 30 MPa, sintering temperature of 1400 °C and sintering time of 30 min, a nearly fully dense NiAl/Al_2O_3 composite is obtained.

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