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Author(s)	Ohmi, Tatsuya; Kirihara, Kazuhiko; Kudoh, Masayuki
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Precision Casting of Ni–Al Alloy and Simultaneous Joining to Dissimilar Metals by Modified Centrifugal Combustion Synthesis [*1]

Tatsuya Ohmi, Kazuhiko Kirihara [*2] and Masayuki Kudoh

Division of Materials Science and Engineering, Graduate School of Engineering, Hokkaido University, Sapporo 060-8628, Japan

A modified centrifugal combustion synthesis process has been developed that enables precisely casting synthesized materials and simultaneously joining them to a dissimilar metal. The material to be synthesized was a Ni–25 mol%Al alloy; that to be bonded with the synthesized material was a stainless steel, an ultra-low carbon steel, pure nickel or a Ni–25 mol%Al alloy. The base material to be bonded; a graphite mold; and a green compact of reactants consisting of Al, Ni and NiO were set in a centrifugal caster. When the combustion synthesis reaction was induced in the centrifugal force field, synthesized molten Ni–Al alloy flew into the mold and collided with the base material. This process was successfully applied in joining the synthesized Ni–Al alloy and various base materials. Centrifugal force was also confirmed to assist the molten Ni–Al alloy fill the mold cavity and adhere to the surface of the base materials.

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1. Introduction

Intermetallic compounds such as transition-metal aluminides or transition-metal silicides have advantageous properties for high-temperature use. However, these intermetallic compounds are often difficult to work by ordinary metal-forming processes because of their poor machinability or weldability.[1] For this reason, current processes require many operations steps and much processing time to shape them into the intended configuration and size, and to join them to dissimilar materials.

Recently, we investigated a centrifugal combustion synthesis process for casting an intermetallic compound and simultaneously joining it to a dissimilar metal.[2] In this process, a combustion synthesis reaction[3–7] is induced to produce a molten alloy with a composition of the intermetallic compound, and then the molten alloy fills the mold cavity and concurrently contacts the dissimilar metal in a centrifugal force field.

Some centrifugally assisted combustion synthesis processes have been developed to produce metal-ceramic double-layered articles[8,9] or functionally graded metal-ceramic composite materials.[10] However, fabrication of a metal-intermetallic compound junction by centrifugal combustion synthesis has never been reported except our study.[2]

In our previous study,[2] we used a cylindrical block of stainless steel as a base material that was embedded in the basal part of the green compact of reactants. The green compact was set in a cylindrical alumina mold in a centrifugal caster, and the combustion synthesis reaction was induced in the field of centrifugal force. This process was successfully applied in joining the Ni–Al alloy and the stainless steel. Based on comparative experiments, we also found that joining is not successful in the absence of centrifugal force or close contact between the base material and the green compact.

This process, however, has some limitations. The most serious difficulty is that the base material and the green compact must be in close contact with each other before ignition. Therefore, intricately configured articles are difficult to produce by this process.

The purposes of the present study are to develop a modified centrifugal combustion synthesis process to overcome this difficulty and to examine the ability of the new process.

In the new process, the base material and the green compact are separated, and a mold with an open cavity is located between them. When the combustion synthesis reaction is induced in the centrifugal force field, synthesized molten alloy flows into the mold and collides with the base material. The molten alloy that adheres to the surface of the base material solidifies in the mold cavity, producing a joint of the precision-cast intermetallic compound and the base material. In the present study, we fabricated the joints of a Ni–25 mol%Al alloy and four kinds of metals, and investigated the soundness of the joining interface.

2. Experimental Procedure

2.1 Thermite-type combustion synthesis

We adopted a thermite-type combustion synthesis reaction,[11,12] eq. (3), which is a combination of a ordinary combustion synthesis reaction, eq. (1), and a modified thermite reaction, eq. (2), in order to produce a molten intermetallic compound.

$$3\mathrm{Ni} + \mathrm{Al} = \mathrm{Ni_3Al} \tag{1}$$

$$3\mathrm{NiO} + 3\mathrm{Al} = \mathrm{Ni_3Al} + \mathrm{Al_2O_3} \tag{2}$$

$$3a\mathrm{NiO} + 3(1-a)\mathrm{Ni} + (1+2a)\mathrm{Al} = \mathrm{Ni_3Al} + a\mathrm{Al_2O_3} \tag{3}$$

where a is the ratio of the modified thermite reaction to the thermite-type combustion synthesis reaction. The amount of heat evolved per unit mol reactant in eq. (2) is 3.6 times that

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[*2] Graduate Student, Hokkaido University.

in eq. (1). Molten Ni_3Al can thus be produced when the thermite-type combustion synthesis is used with an adequate value of a.

In eq. (3), alumina is produced as a residual product. Centrifugal force can be expected to assist the separation of alumina from the products due to the density difference between the intermetallic compound and alumina.

In our experiments, the starting materials were aluminum powder of 99.5 mass% in purity and 100 µm in particle diameter, nickel powder of 99.8 mass% and 5 µm, and nickel oxide powder of 97.0 mass% and 5 µm. The powders were mixed after they were moistened with a small amount of ethanol. The ratio of each powder was set based on eq. (3) with the condition, $a = 0.25$, that enables the synthesized intermetallic compound to melt. The powder mixture was cold-pressed into a cylindrical green compact in a metal mold using a unidirectional pressure of 470 MPa. The green compact had a diameter of 20 mm and mass of 30 g.

2.2 Modified centrifugal combustion synthesis process

In the present paper, we refer to "the modified centrifugal combustion synthesis process" as "the centrifugal combustion synthesis process" or abbreviate it as "the CCS process" for simplicity. Figure 1(a) shows a schematic view of the main part of the vertical centrifugal caster used in the experiments. The mold part, which is located on the outer side of the green compact, consists of a base material and a graphite mold. We examined three mold configurations. Figure 1(b) shows the mold part used in the experiments. In Fig. 1(b), the type-A mold is used for joining a cylindrical base material (20-mm diameter and 10-mm length) and a cylindrical Ni–Al alloy cast with a different diameter (10 mm or 5 mm); the type-B mold, for joining a tubular base material (14-mm outer diameter, 10-mm inner diameter and 15-mm length) and a tubular Ni–Al alloy cast of the same size; and the type-C mold, for joining a cylindrical base material (10-mm diameter and 15-mm length) and a cylindrical Ni–Al alloy cast of the same size.

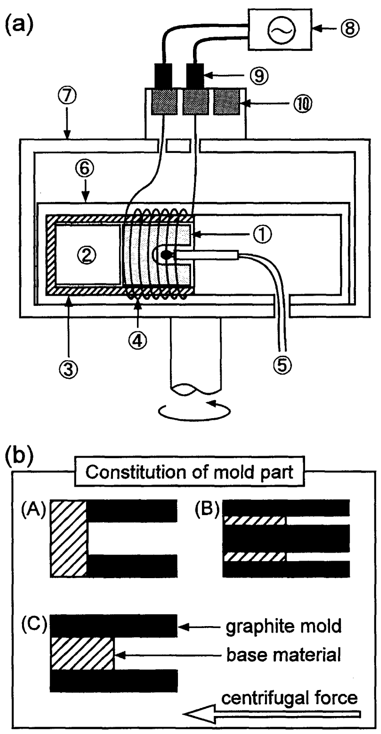

Fig. 1 Schematic illustration of experimental apparatus. ① green compact, ② mold part, ③ alumina container, ④ heating coil, ⑤ thermocouple, ⑥ inner steel container, ⑦ outer steel container, ⑧ electric source, ⑨ graphite brush, ⑩ slip ring.

The experimental procedure for the CCS process was as follows. The green compact in the rotating container was heated and then self-ignited at a temperature near the melting point of aluminum. The container rotation was continued until the synthesized Ni–Al alloy was completely solidified in the mold. The container rotation speed was 19 revolutions per second, which produced a centrifugal acceleration of 70 to 85 G at the joining interface of the specimen. All experiments were carried out in an air atmosphere.

Four kinds of metals were used for the base material: a stainless steel (SUS304L), an ultra-low carbon steel (carbon content: 0.08 mass%), pure nickel and a Ni–25 mol%Al alloy. The Ni–Al alloy base was produced by combining the thermite-type combustion synthesis process and the centrifugal casting process.

We performed comparative experiments without centrifugal force in order to investigate the role of centrifugal force in the CCS process. In these experiments, the green compact was located on the mold part so that the synthesized Ni–Al alloy melt could flow into the mold under the acceleration of gravity. In these comparative experiments, the case of $a = 0.4$, in which the adiabatic combustion temperature exceeds the case of $a = 0.25$, was also examined.

After processing, the joint specimens were sectioned longitudinally for metallographical observation. Marble's reagent, which is a mixture of a hydrochloric acid and a saturated aqueous solution of $CuSO_4$ in the volume ratio of 2 : 1, was used to reveal the structure of the specimens.

3. Results and Discussion

3.1 Structure of the joint specimen produced by the CCS process

Figure 2 shows the structures of a specimen produced by the CCS process with a type-A mold. The specimen is the joint of a 20-mm diameter cylindrical stainless steel base and a 10-mm diameter cast cylinder of a Ni–Al alloy. Figures 2(a) and (b) show the macrostructure and the microstructure near the joining interface of the specimen. As seen in Fig. 2(a), the surface of the base material at the joining interface was slightly melted because it closely contacted the high-temperature molten Ni–Al alloy. We also found, from microstructure observation, that there are no defects at the interface as shown in Fig. 2(b).

The characteristics of the solidification structure of the Ni–Al alloy produced by the CCS process are as follows.

(1) Primary NiAl crystals, which are observed as white dendrites in the macrostructure, are distributed in a Ni_3Al matrix. A similar structure was observed in Ni–25 mol%Al alloys produced by the thermite-type combustion synthesis process as previously reported.$^{2)}$

(2) Only a few alumina particles were observed in the cast Ni–Al alloy part. The alumina particles are spherical and

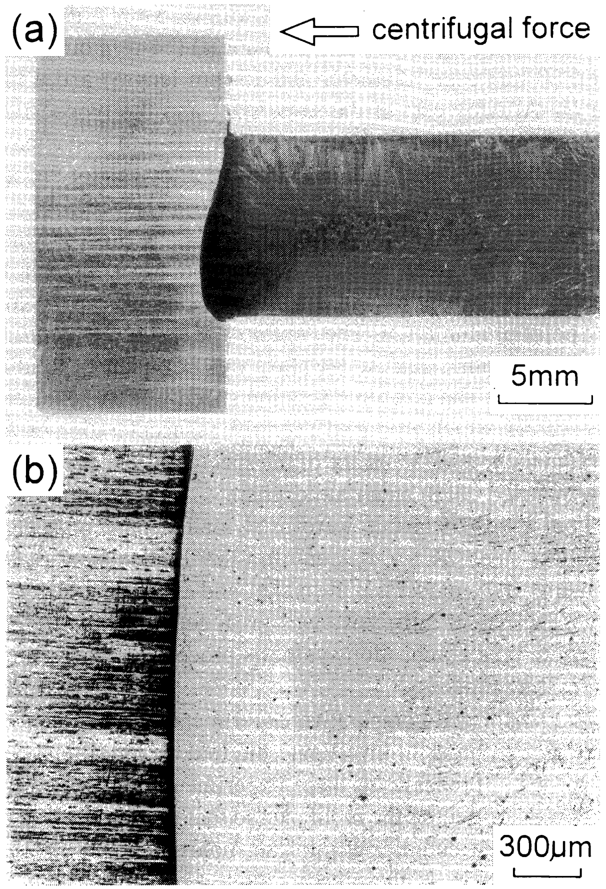

Fig. 2 Structure of a Ni–Al alloy-stainless steel jointing produced by the CCS process. (a) Macrostructure. (b) Microstructure near the joining interface.

their maximum diameter is about 10 μm as shown in Fig. 3. Most of the alumina produced in the reaction remained where the green compact was located.

In the present study, we did not use any means to prevent the alumina from flowing into the mold cavity. However, we expect that alumina will be prevented from contaminating the product by using a filter to capture the alumina. From a different point of view, we also expect that a joint with a cast composite material containing alumina particles will be produced when alumina particles with controlled size and distribution are intentionally dispersed. Development of these techniques is a subject for future study.

3.2 Effect of centrifugal force on the soundness of the joining interface

Figure 4 shows the result of a comparison experiment in which the combustion-synthesized Ni–Al alloy was cast, without centrifugal force, into a mold similar to that used in Fig. 2. Figures 4(a) and (b) show the macrostructure and the microstructure near the joining interface of the specimen. In this experiment, *a* was set at 0.4 in order to achieve a higher adiabatic combustion temperature than in Fig. 2. However, little melting of the base material surface is seen. In addition, cracks and coarse alumina inclusions are observed near the joining interface as seen in Fig. 4(b).

Figure 5 shows the results of the experiments carried out using a type-A mold with a slender cavity (diameter: 5 mm).

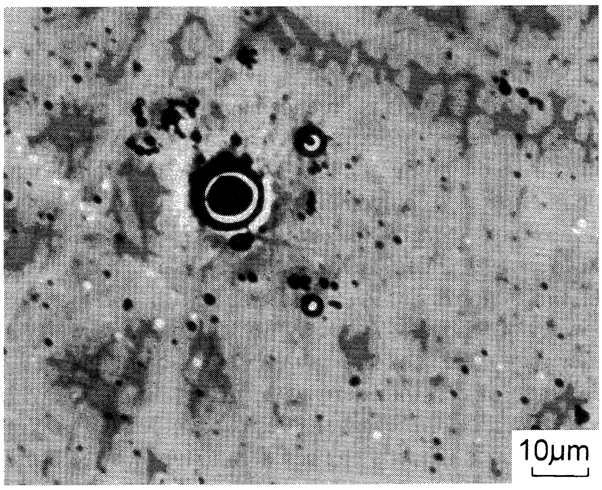

Fig. 3 Fine alumina particles observed in the specimen shown in Fig. 2.

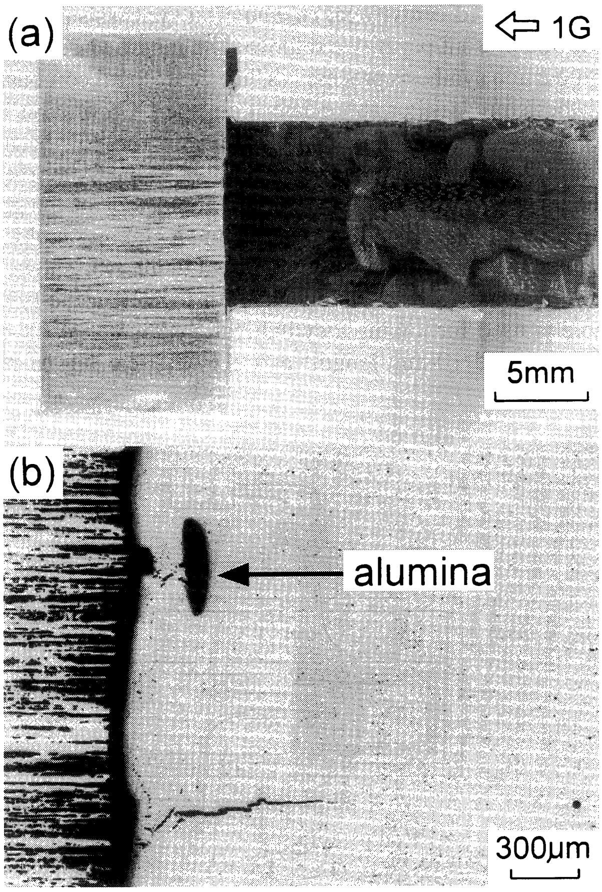

Fig. 4 Structure of a specimen produced under 1 G gravitational acceleration. The base material is a stainless steel. (a) Macrostructure. (b) Microstructure near the joining interface.

Figures 5(a) and (b) show the specimens produced by the CCS process and by the comparison experiment without centrifugal force. In the latter experiment, *a* was set at 0.4. As seen in Fig. 5(a), the CCS process produced a good joint specimen. In contrast, the synthesized Ni–Al alloy in the comparison experiment did not flow into the mold cavity as seen in Fig. 5(b).

The above results indicate that the centrifugal force in the CCS process plays an important role in casting a synthesized

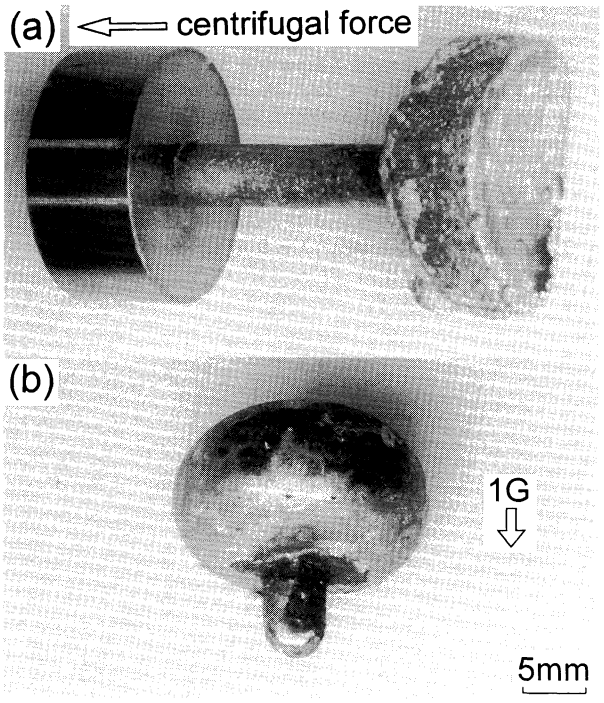

Fig. 5 Specimens produced by using a mold with a slender cavity (diameter: 5 mm). (a) Specimen produced by the CCS process. (b) Specimen produced under 1 G (Ni–Al alloy that did not flow into the mold cavity).

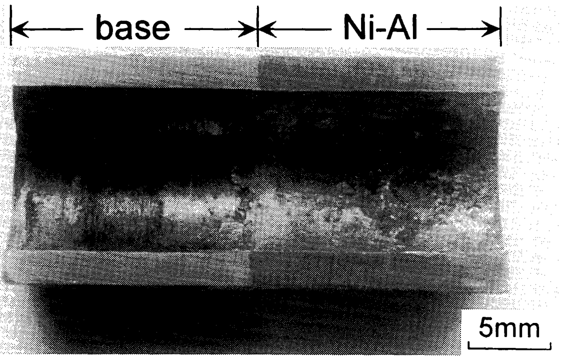

Fig. 6 Longitudinal cross section of a tubular specimen produced by the CCS process. The base material is a stainless steel.

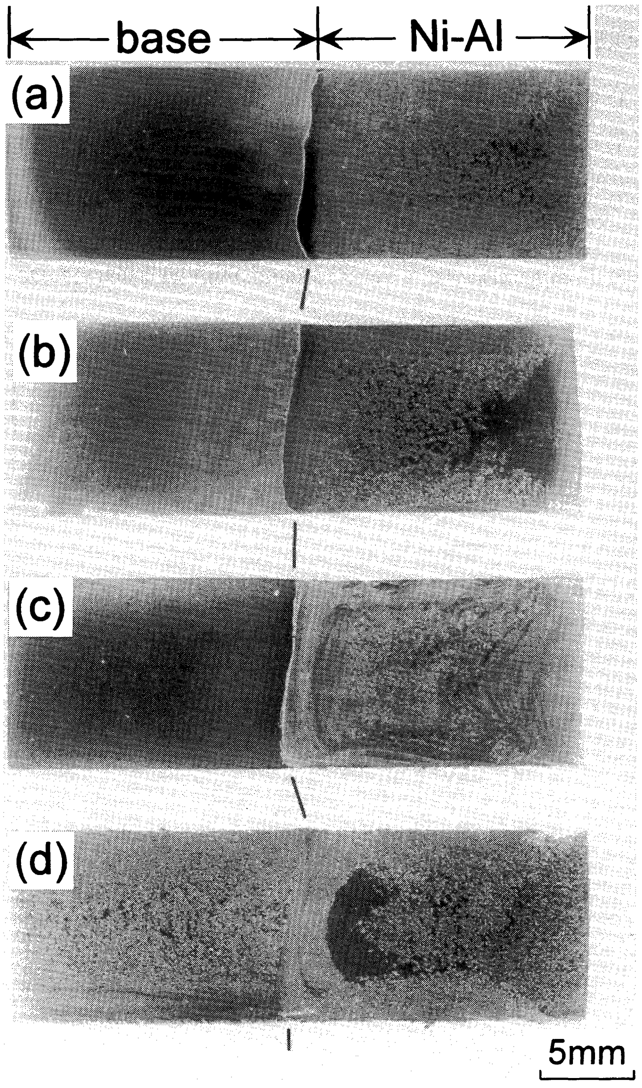

Fig. 7 Macrostructures of specimens produced by the CCS process with various base materials. (a) Stainless steel. (b) Ultra-low carbon steel. (c) Ni. (d) Ni–25 mol%Al alloy.

molten alloy and in producing a sound joining interface.

Figure 6 shows the longitudinal cross section of the tubular specimens produced by the CCS process with a type-B mold. The base material was a stainless steel. This result shows clearly that the CCS process enables precisely casting intermetallic compounds and simultaneously joining them to a dissimilar metal.

3.3 Joint specimens with various base materials

Figures 7(a) to (d) show the macrostructures of a longitudinal cross section of the joint specimens produced by the CCS process with various base materials; *i.e.* a stainless steel (a), an ultra-low carbon steel (b), pure nickel (c) and a Ni–25 mol%Al alloy (d). A type-C mold was used in these experiments. In the specimens shown in Figs. 7(a) to (c), the white-etched region along the joining interface is a transition region from the base material to the cast alloy. In all specimens shown in Fig. 7, the cast alloy adheres well to the base material, and no defects were observed at the joining interface. These results suggest that the CCS process can be applied to join various metals.

4. Conclusions

We attempted to fabricate a joint of a precision-cast intermetallic compound and a dissimilar metal by a modified centrifugal combustion synthesis process (the CCS process) in which the combustion synthesis process and the centrifugal casting process are merged. The results of our investigation are summarized as follows.

(1) The CCS process enables precisely casting a synthesized Ni–25 mol%Al alloy and simultaneously joining it to a dissimilar metal.

(2) In this process, centrifugal force assists the molten Ni–Al alloy to flow into the mold cavity and to adhere to the surface of the base materials.

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