

Aluminizing Process of Ni-base Superalloy by Spark Plasma Sintering Method

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Aluminizing was performed to form nickel aluminide on a nickel-based superalloy using the spark plasma sintering (SPS) method. Inconel 601, a nickel-base superalloy, was used as the substrate, and a powder mixture of Al, Al₂O₃, and NH₄Cl was used as the aluminizing source. The SPS aluminizing temperatures were 1073, 1173, and 1273 K, and the holding times were 1.8, 3.6, and 10.8 ks. The X-ray diffraction pattern of the sample subjected to SPS aluminizing revealed the formation of NiAl₃, Ni₂Al₃, and NiAl. The activation energy of the nickel aluminides formed by this method was estimated to be 310 kJmol⁻¹, which is less than that estimated by conventional pack cementation method. The oxidation resistance of the SPS-aluminized sample under high-temperature air was improved compared to that of untreated nickel-alloy substrate.

Keywords: aluminizing, diffusion coating, surface engineering, nickel-based superalloy, spark plasma sintering

1. Introduction

As the system requirements of high temperature material become ever more severe, material that has both excellent oxidation resistance and corrosion resistance is demanded. In this regard, intermetallic compounds, which show novel physical properties, have in recent years raised expectations. The Ni-Al system intermetallic compounds are excellent in oxidation resistance as an alternative material. Nickel-aluminide diffusion coatings have been applied to hot gas turbine components in order to improve their service life at high temperature. The β-NiAl phase plays an important role due to its excellent oxidation resistance, which is attributed to aluminum oxide formation over a wide range of temperatures and oxygen pressures^{1,2}.

Recently, works on the application of spark plasma sintering (SPS) or pulsed electric-current sintering (PECS) to the joining or surface modification process have been reported³⁻⁵, though much attention has been paid to this process as a fabrication method for new functional graded materials and composite materials. This process enables one to sinter these materials at lower temperatures and in shorter times than conventional sintering techniques^{6,7}.

In this investigation, pack aluminizing was performed to form nickel aluminide on nickel-base superalloy using the SPS method. And, the resulting interfacial microstructure and oxidation properties of the obtained aluminizing samples were investigated.

2. Experimental procedure

The sample material used in this study was a nickel-base superalloy Inconel 601. The sample disk was 10 mm in diameter and 10 mm in thickness. Al powder was used as an agent of pack powder, Al₂O₃ as a filler, and NH₄Cl as a halide activator salt. These powders were mixed at a weight ratio of 30% Al, 67% Al₂O₃, and 3% NH₄Cl. The aluminizing was carried out with a spark plasma sintering (SPS) apparatus, SPS-1020, produced by Sumitomo Coal Mining Co., Ltd (at present: Fuji Electronic Industrial Co., Ltd). Inconel 601 substrate and a powder mixture of Al, Al₂O₃, and NH₄Cl were loaded into a graphite die. SPS

aluminizing was carried out at 1073–1273 K for 1.8–10.8 ks with pressureless in a vacuum of 20 Pa.

The phase structures on the aluminized surface were determined by X-ray diffraction (XRD) studies. The aluminized microstructure and compositional analyses were using a scanning electron microscope-energy dispersive X-ray spectroscopy (SEM-EDX). In addition, the hardness of cross section of the aluminized sample was measured using a Vickers microhardness tester under a 0.25-N load. The samples were also subjected to cyclic oxidation tests in air, subjected to 5 cycles of oxidation at 1273 K, in 72 ks long for a total oxidation time of 360 ks. After every heating cycle, the samples were air-quenched to room temperature, then weighed using a microbalance.

3. Results and discussion

XRD results of the samples for various SPS aluminizing treatments revealed that NiAl₃, Ni₂Al₃, and NiAl were identified in all the aluminized samples. This result indicates that nickel aluminides intermetallic compounds were formed on the substrate by SPS aluminizing.

Figure 1 shows the effect of the aluminizing time and temperature on the microstructures of the cross section. It can be seen that a compound layer was on the top of the sample surfaces. The diffusion layer was also formed between the compound layer and the core material. Both the compound layer and diffusion layer that formed at the aluminizing interface were developed with increase in aluminizing time and temperature. Assuming that aluminum diffusion in the nickel aluminide layer is the primary factor affecting the aluminizing layer thickness and aluminized layer growth occurs perpendicular to the substrate, then on the basis of the classical kinetic theory, the squared thickness of the aluminized layer (d^2) as a function of treatment time (t) is described by Eq. (1)⁸.

$$d^2 = Dt \quad (1)$$

where D is the diffusion coefficient depending on the process temperature. It is clear that the layer thickness, d , changes linearly with the square root of aluminizing time, $t^{1/2}$, and consequently the diffusion coefficient, D , can be calculated from the slope of d versus $t^{1/2}$. The relationship

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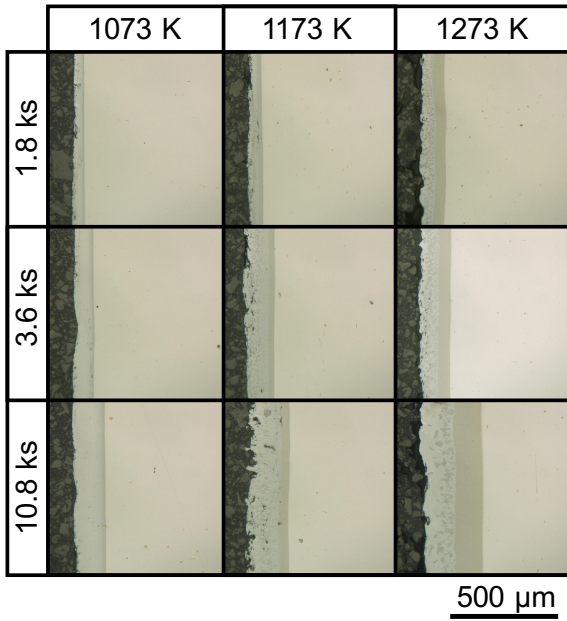


Fig. 1 Cross-sectional micrographs of SPS-aluminized nickel alloy.

between the diffusion coefficient, D , activation energy, Q , and the temperature in Kelvin, T , can be expressed as an Arrhenius-type equation, Eq. (2).

$$D = D_0 \exp(-Q/RT) \quad (2)$$

where D_0 is the pre-exponential constant and R is the gas constant. Q can be calculated from a plot of $\ln(D)$ versus $1/T$, shown in Fig. 2. The mean activation energy in the case of SPS aluminizing was calculated as 310 kJmol^{-1} , which is smaller than the activation energies for self-diffusion of aluminum in $\beta\text{-NiAl}$ phase for a conventional pack cementation process ($332\text{-}414 \text{ kJmol}^{-1}$)⁹. This result suggests that the diffusion of aluminum in this study is promoted by thermal and electromagnetic energies during SPS aluminizing. In addition, this result exhibited that SPS method was beneficial for diffusion coating of aluminum.

Figure 3 shows the relationship between oxidation time and mass increase for a high-temperature oxidation test conducted for 360 ks at 1273 K. Comparison of the

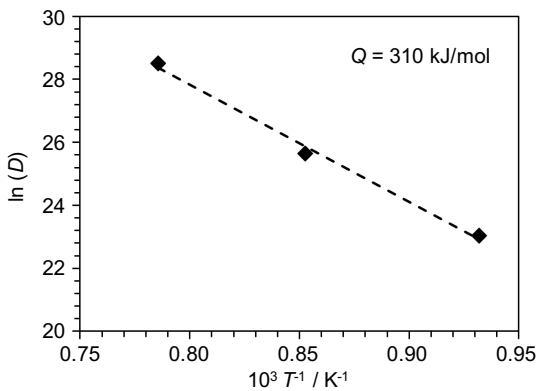


Fig. 2 A plot of $\ln(D)$ versus $1/T$ of aluminized nickel alloy by SPS.

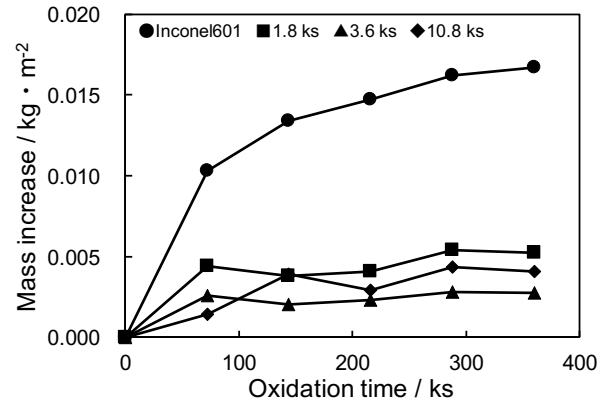


Fig. 3 Oxidation characteristics of aluminized nickel alloy for a high temperature oxidation test at 1273 K for 360 ks.

aluminized samples and the untreated nickel-base superalloy specimen reveals that the mass increase was lower and high-temperature oxidation resistance was higher for the aluminized specimens in all the cases. The mass increase of aluminized specimen was less than that of the untreated nickel alloy specimen. XRD results of the samples after the oxidation test revealed that Al_2O_3 phase was identified. This could be because a protective Al_2O_3 film was formed on the aluminized sample. This result indicates that the formation of Ni-Al intermetallic compound on the nickel-base superalloy markedly improved the oxidation resistance of the nickel alloy.

4. Conclusions

- (1) From the XRD result, the sample subjected to SPS aluminizing indicated the formation of NiAl_3 , Ni_2Al_3 , and NiAl .
- (2) The activation energy of the nickel aluminide formation in this method was estimated to be 310 kJmol^{-1} .
- (3) The oxidation resistance of the aluminized sample in high-temperature air was improved compared to that of untreated nickel alloy substrate.

References

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