FORMATION OF INTERMETALLIC COMPOUND LAYER FORMED BY DIFFUSION TREATMENT OF THERMAL SPRAY COATINGS, AND ITS APPLICATION.

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ABSTRACT

In order to develop high temperature oxidation resistive coatings, multi-layer thermal spray coatings containing Ni-Cr-Al and Al layers were applied to austenitic stainless steel substrate. The Ni-Cr-Al layers were coated with Ni-20%Cr-5%Al pre-alloyed powder or mixtures of Ni-20%Cr powder with 5% of pure Al powder. Then, pure Al was sprayed on the Ni-Cr-Al coated surface. Prior to high temperature oxidation test, conducted at 1273 K in air, sprayed specimens were heat treated at 1123 K for 3.6 ks and 1423 K for 3.6 ks in air. The oxidation resistivity of coating sprayed with pre-mixed powder is superior to that made with pre-alloyed powder. This result is attributed to the difference in the distributions of the Al rich alloy phases formed by the diffusion between each layer.

INTRODUCTION

Thermal spraying is one of the practical method of surface coating to improve wear resistance, heat resistance and corrosion resistance of metals and alloys. Despite of the widespread use of the thermal spraying, the cohesive properties have not been researched from a point of view of the basic cohesion mechanism. The cohesivity expected to be enhanced through the mutual diffusion between the sprayed coating and the substrate. The authors have been aiming to improve the cohesive strength by appropriate diffusion treatment after spraying, and to develop the novel method of the surface modification by the use of the resulting intermetallic compound layers[1,2,3].

In this paper, the attempt to develop the high temperature anti-oxidation coating formed by the diffusion treatment of spray coating is described. This method was applied to the Ni-Cr-Al and Al multi-layer coatings on the austenitic stainless steel substrates, and the formation of the intermetallic compounds phases by diffusion treatment and the high temperature oxidation properties of these coatings were investigated.

It is well known that the Al-rich alloy layers formed on the surface of base-metals improve their oxidation properties by the resulting Al_2O_3 protective films[4]. However, Al coatings sprayed on stainless steel substrates did not form stable compound layer at high temperature[3].

The Ni-Cr-Al alloys were developed for high temperature protective coatings, and also have applied to the under-coat for ceramics coatings[5]. But the Ni-Cr-Al alloy spray coatings are available only by the Low Pressure Plasma Spraying method[6]. Hence, they have a limited field of use.

In view of these facts, we developed anti-oxidation coating available under oxidizing environment at about 1273 K. The prerequisites of the development are to form Al-rich intermetallic compound layers which have high cohesive strength with substrate and is stable at high temperature. In addition, it is an important requisite that the thermal spraying and the heat treatment can be conducted in air.

EXPERIMENTAL PROCEDURE

The Ni-Cr-Al alloys and pure Al multi-layers, as shown in Table 1, were coated on the commercial austenitic stainless steel substrates containing 18%Cr and 8%Ni (JIS SUS 304) by plasma spraying method in air. Plasma spraying was carried out using Bay State PG-100 torch. The stainless steel substrates were glit blasted prior to thermal spraying. The Ni-Cr-Al alloy layers were coated with pre-alloyed powders or powder mixtures of Ni-Cr pre-alloyed powders and pure Al powder. Then pure Al was sprayed on the Ni-Cr-Al of Ni-Cr sprayed surfaces. In table and figures, - and + show the pre-alloyed and the pre-mixed powders, respectively. The properties of material powders are listed in Table 2. The thickness of the Ni-Cr-Al layers and Al layers were 200 ～ 250 μm and 50 ～ 100 μm, respectively.

Table 1. Constitution of multi-layer coatings

Designation	1st layer	2nd layer
NC10A/A	Ni - 20%Cr + 10%Al	Al
NC5A/A	Ni - 20%Cr + 5%Al	Al
NCA/A	Ni - 20%Cr - 5%Al	Al
NCA	Ni - 20%Cr - 5%Al	-
NC	Ni - 20%Cr	-

Table 2. Properties of material powders

Powder	Chemical composition / mass%						Particle size / μm
	Al	Ni	Cr	C	Si	Fe	
Ni-Cr-Al	4.6	Bal.	18.12	1.1	-	-	53 - 10
Ni-Cr	-	Bal.	19.16	0.014	1.26	-	45 - 10
Al	Bal.	-	-	-	0.06	0.11	106 - 32

The sprayed specimens were heat treated at 1123 K for 3.6 ks and 1423 K for 3.6 ks in air.

High temperature oxidation properties were estimated by their mass gains during heat cycle tests conducted at 1273 K in air. The oxidation products were also determined by X-ray diffractometry (SHIMADZU XD-5A). Monochromated Cu K α radiation was used as the X-ray source.

In order to determine the intermetallic compound phases formed by the heat treatment and during the heat cycle test, some metallographical examinations, i.e. metallography, electron probe microanalysis (JEOL JXA-8600), and X-ray diffractometry were conducted.

RESULTS and DISCUSSION

The result of high temperature heat cycle test was shown in Fig. 1. As shown in this figure, the multi-layer coating made from Ni-Cr-Al pre-alloyed powder (NCA/A) was peeled in a few number of heat cycles. The multi-layer coating which did not contain Al in the first layer (NC/A) shows the same tendency. On the other hand, no peeling can be detected in the coatings made from Ni-Cr+Al pre-mixed powders (NC5A/A and NC10A/A). Mass gain of Ni-Cr single layer coating (NC) during the heat cycle test were less than that of Ni-Cr+Al pre-mixed multi-layer coatings. This fact is attributed to the high vapor pressure of Cr_2O_3 formed on the Ni-Cr coated specimen during high temperature heat cycle test. The same tendency was observed in the case of the Ni-Cr-Al single layer coating (NCA).

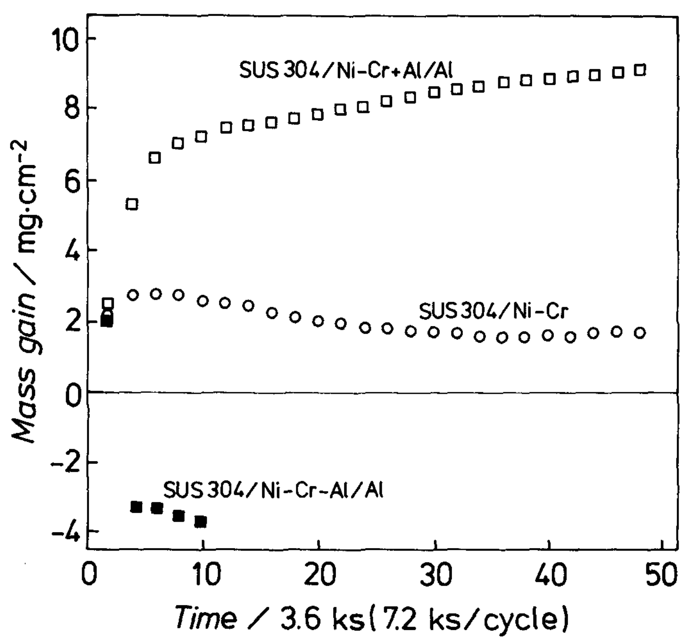

Figure 1. Effect of constitution of multi-layer coatings on high temperature oxidation properties at 1273 K.

The results mentioned above shows that the oxidation resistivity of the coating sprayed with pre-mixed powder is superior to that made with pre-alloyed powder, although their compositions are the same. Then, the microstructures of the specimens coated with pre-alloyed powder and pre-mixed powder were examined for revealing to the effect of the method of Al addition on the oxidation properties.

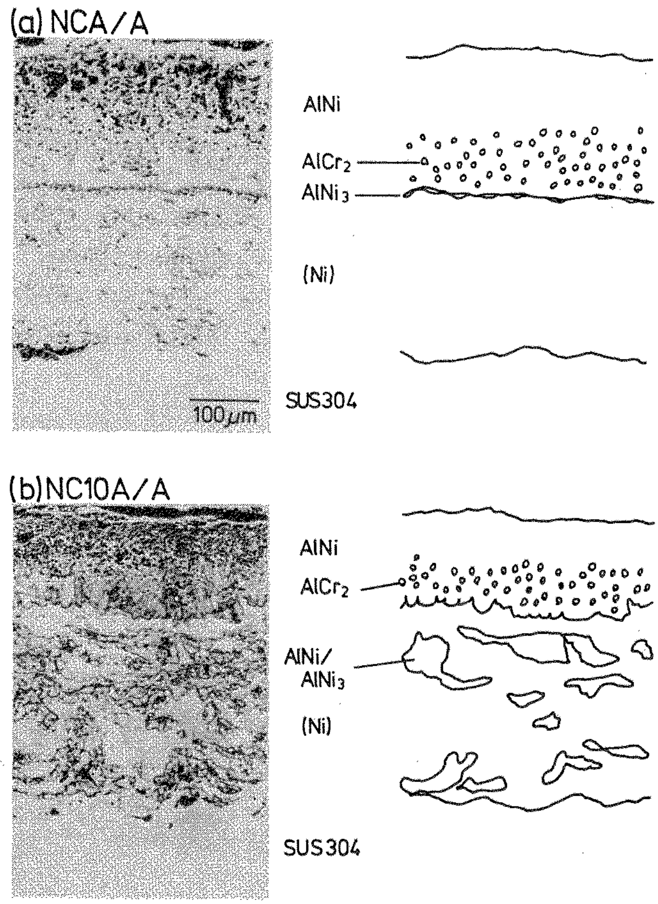

Figure 2. Microstructures and their schematic illustration of multi-layer coatings made from (a) pre-alloyed powder and (b) pre-mixed powder, after heat treatment.

The microstructures of the multi-layer coatings made from pre-alloyed powder (NCA/A) and pre-mixed powder (NC10A/A) are shown in Fig. 2. They consisted of AlNi phase and Ni solid solution, and islands of $AlCr_2$ phases were dispersed in the AlNi layers. $AlNi_3$ phases were formed in the interface between AlNi and Ni layers of the coating made from pre-alloyed powder (Fig.

2 (a) NCA/A). On the other hand, the Ni solid solution layer which made from pre-mixed powder (Fig. 2 (b) NC10A/A) had dispersion of Al rich phases consisted of AlNi and $AlNi_3$.

Furthermore, the secondary images and characteristic X-ray images of Ni, Cr, Al and O were shown in Fig. 3. There were some differences in distributions of alloying elements between pre-alloyed and pre-mixed coatings. In the case of the coatings made from pre-alloyed powder (Fig. 3 (a) NCA/A), distributions of Al and O formed a continuous net-work. This fact could be attributed to the oxidation of the surface of the molten particles and the interface of the deposited particles during spraying, and/or heat treatment. On the other hand, in the case of the pre-mixed coating (Fig. 3 (b) NC10A/A), the pre-mixed Al particles were oxidized preferentially, and the cohesive strength between deposited particles were consequently higher. The same tendency has been observed in the coating sprayed from Ni-Cr+5%Al pre-mixed powder (NC5A/A), which has the same composition as the Ni-Cr-Al pre-alloyed powder.

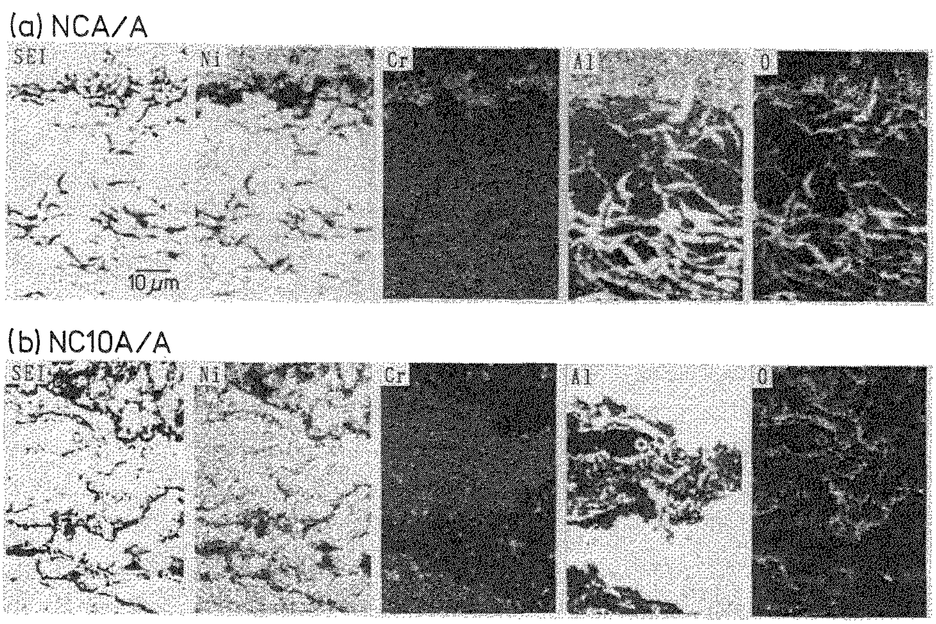

Figure 3. Secondary electron images and characteristic X-ray images of sprayed and heat treated multi-layer coatings made from (a) pre-alloyed powder and (b) pre-mixed powder.

Therefore, the difference of the oxidation properties between pre-alloyed and pre-mixed coatings could be ascribed to their distinct morphology of oxides in the spray coated layers. We can conclude that the pre-mixed Al addition is available in an attempt to develop the high temperature oxidation resistive coatings by means of thermal spraying method.

CONCLUSIONS

In order to develop high temperature oxidation resistive coating, Ni-Cr-Al and Al multi-layer thermal spray coatings were applied to austenitic stainless steel substrate. It has been proved that the oxidation resistivity of the coating sprayed with pre-mixed powder was superior to that made from pre-alloyed powder. This fact is attributed to their distinct morphology of oxides in the sprayed coatings.

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