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Interface Reaction between Nickel-Base Self-Fluxing Alloy Coating and Steel Substrate

F. Otsubo, H. Era, and K. Kishitake

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The interface reaction between a nickel-base, self-fluxing alloy coating and a steel substrate has been investigated to examine the formation of "pores," which are observed along the interface of used boiler tubes. It was found that lumpy precipitates form along the interface instead of pores after heating at high temperatures and that the precipitates are of Fe_2B boride. The adhesion strength of the coating is not decreased by the formation of Fe_2B precipitates along the interface because of the increase of the adhesion due to interdiffusion.

Keywords nickel-base self-fluxing alloy, plasma spraying, interface structure, adhesion strength

1. Introduction

Nickel-base self-fluxing alloy coatings are most widely used in the field of thermal spraying for boiler tubes, rolls, continuous casting molds, and machine parts in order to provide wear resistance.[1,2,3] In case of coating boiler tubes used in high-temperature atmospheres, it is predicted that interdiffusion occurs at the interface. Figure 1 shows the microstructure at the interface between a nickel-base coating and the substrate of a boiler tube used for 2 years. Many pores are observed along the interface on steel substrate side, and there is concern that these decrease the adhesion strength of the coatings.

Kamota *et al.* investigated the interface structure between a mild steel and a nickel-base coating during the joining of steels by using a self-fluxing alloy coating as a bond material.[4,5] However, they reported no interface reaction or pore formation. In this study, the interface reaction between a mild steel and a nickel-base coating during heating at a high temperature was investigated in detail by means of optical microscopy, electron microprobe analysis (EPMA), X-ray diffractometry (XRD), and transmission electron microscopy (TEM). The change in adhesion strength by heating the coating was also examined.

2. Experimental Procedure

Nickel-base powder (JIS-MSFNi4,[6] Table 1) was sprayed onto a mild steel substrate using a 40 kW plasma spraying apparatus under the conditions of 600 A, 26 V, and 100 mm spray distance. The coating thickness was about 500 μm. Subsequently, the coatings were fused at 1323 K for 300 s in air. The coatings were further heated at 1273 K up to 28.8 ks after fusing to investigate the change of the interface structure. Although the heating temperature, 1273 K, is higher than the operational temperature of conventional boiler tubes, the high temperature was adopted to accelerate the interface reaction between the coating and the steel substrate.

The specimen for TEM was prepared by means of the ion milling method under conditions of 5 kV at a tilt 10° angle for the argon ion beam. The phases revealed in the interface were also examined by electron beam diffraction. Furthermore, the adhesion strength of the coatings was measured by a shear test. The shear test pieces have a semicircular notch, as shown in Fig. 2, to reduce the stress concentration at the corner of the protruded coating.[7] The adhesion of six test piece, which was debonded at the interface of the coating and substrate, was evaluated.

3. Results and Discussion

Figure 3 shows a backscattered electron (BSE) image of the coating heated at 1273 K for 7.2 ks. Lumpy gray precipitates formed in the substrate along the interface, as shown by the arrow. It is also seen that the surroundings of the precipitates are concave, as shown by triangles. The formation of the concave

Table 1 Chemical composition of powder (mass.%)

Ni	Cr	B	C	Si	Fe	Mo
Bal	16.6	3.9	0.6	4.1	2.4	4

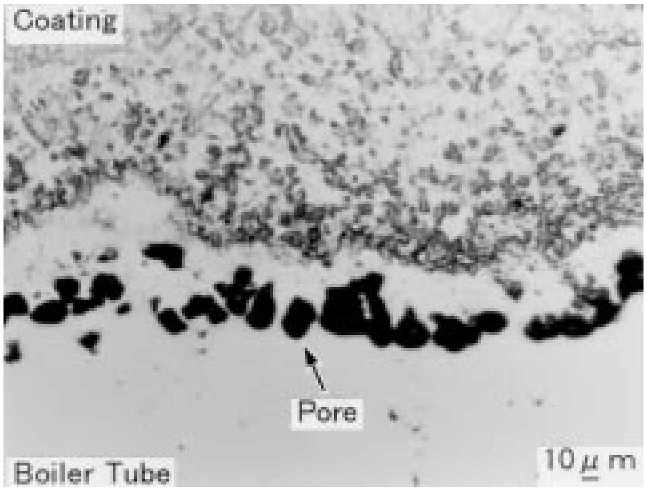

Fig. 1 Optical micrograph of interface between coating and boiler tube

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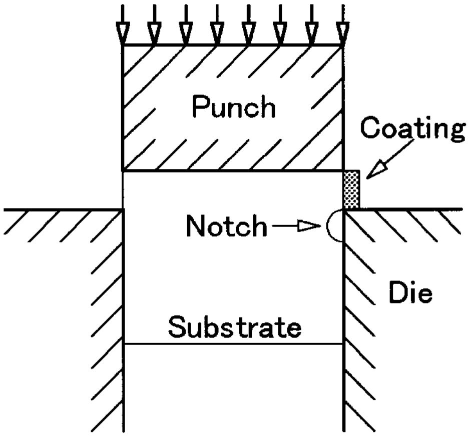

Fig. 2 Schematic illustration of shear adhesion test

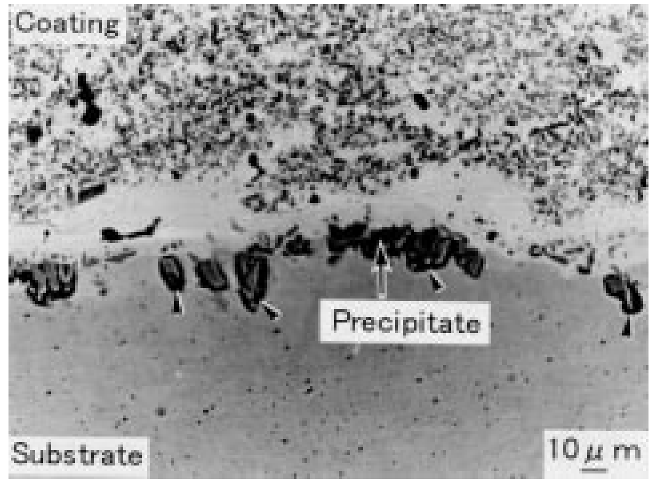

Fig. 3 BSE image of coating held at 1273 K for 7.2 ks.

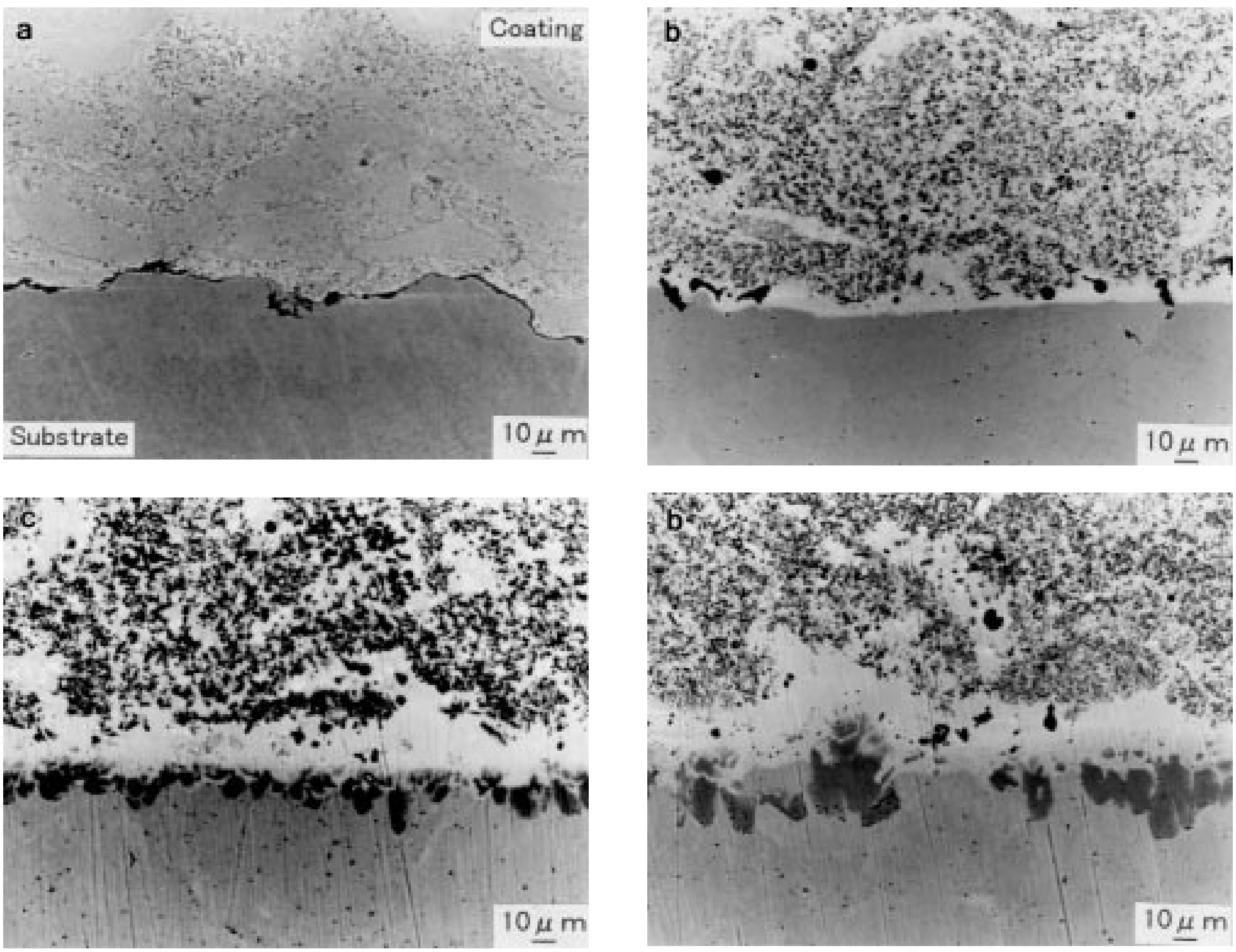

Fig. 4 BSE images of interface between coating and substrate: (**a**) as-sprayed, (**b**) after fusing, (**c**) and (**d**) heated at 1273 K for (**c**) 7.2 ks and (**d**) 14.4 ks

may be due to corrosion through lapping of the specimen. The lumpy precipitates are not observed when the polishing time is extended, but, instead, a line of pores appears, shown in Fig. 1. Therefore, it is considered that the pores shown in Fig. 1 are formed by pull out of the precipitates through lapping. Figure 4 indicates changes in the interface structure between the coating and substrate by heating at 1273 K. The lamellae boundaries in vanish and fine precipitates in the coating become coarse after fusing. The coating heated at 1273 K after fusing was polished with abrasive paper up to #1500 (grit size of 10 μm) to prevent the preferential polishing or pull out of the lumpy precipitates and, therefore, the specimen exhibits some scratches. Precipitates less than 10 μm in diameter are seen at the interface in the substrate side after heating at 1273 K for 7.2 ks (Fig. 4c). These precipitates grow on heating at 1273 K for 14.4 ks (Fig. 4d). The interface of the coating heated at 1273 K for 14.4 ks is diffuse compared with that of the coating after fusing.

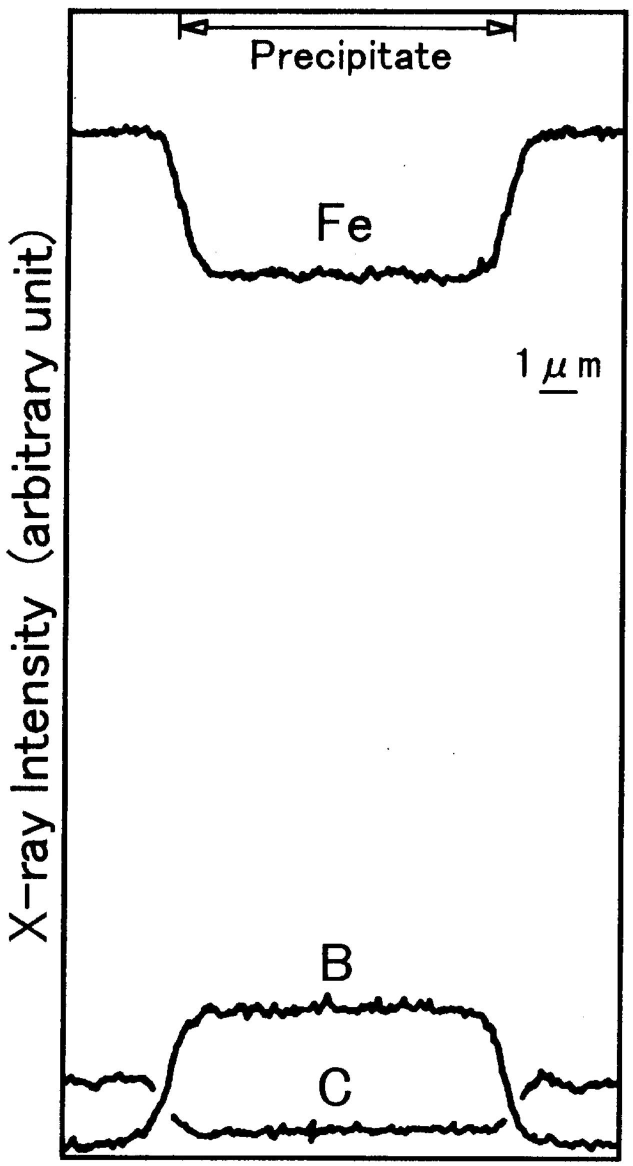

Fig. 5 X-ray intensity profiles across lumpy precipitate

Figure 5 shows the X-ray intensity profiles across the precipitate in the substrate. The precipitate contains iron and boron, while nickel and chromium are not detected in the precipitate. It is clear that the precipitate is iron-boride. X-ray diffraction analysis of the interface was also carried out to investigate the precipitates by grinding off the coating from the top surface to the interface. The XRD pattern reveals the peaks of Fe_2B boride in addition to those of nickel and ferrite of the substrate (Fig. 6, thereby concluding that the precipitate is Fe_2B boride).

The interface structure was further investigated by using TEM. Figure 7 shows the TEM images of the interface heated at 1273 K for 1.8 ks. A lumpy crystal is observed in the substrate near the interface (Fig. 7a). The electron beam diffraction pattern (Fig. 7b) obtained from the crystal reflects Fe_2B with a tetragonal structure of lattice parameters of a = 0.511 nm and b = 0.425 nm. Consequently, it is confirmed that the precipitate in the substrate at the interface is Fe_2B boride.

It was found that the borides in the nickel-base alloy (JIS-MSFNi4) are $(Cr,Mo)_3B_2$ and Ni_3B phases.[8] Although the standard free energies of formation of these borides are not known, it is considered that $(Cr,Mo)_3B_2$ and/or Ni_3B borides are reduced and boron diffuses to make Fe_2B at the interface between the coating and the steel substrate.

The adhesion strength of the self-fluxing coating exceeded values that could be evaluated by the standard tensile test method

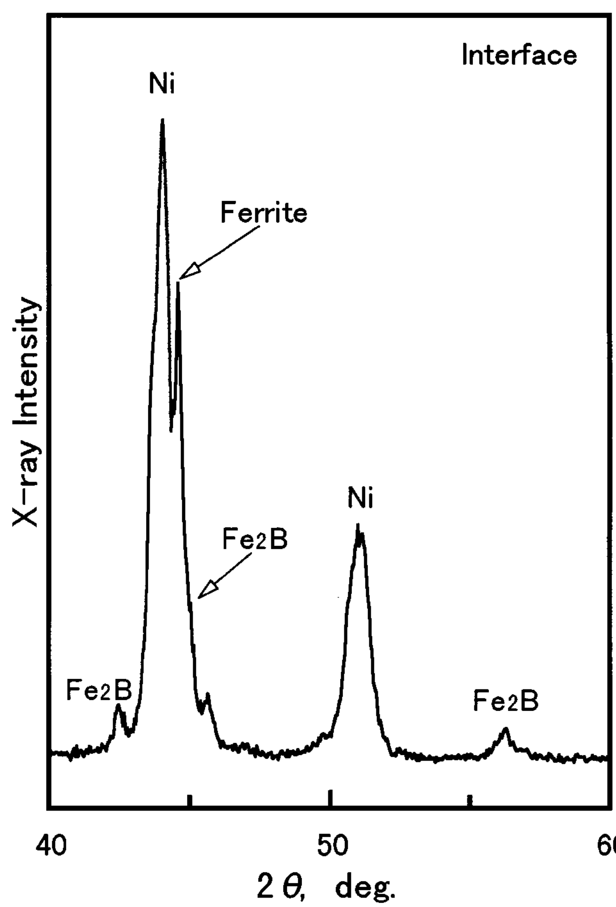

Fig. 6 XRD pattern of interface between coating and substrate

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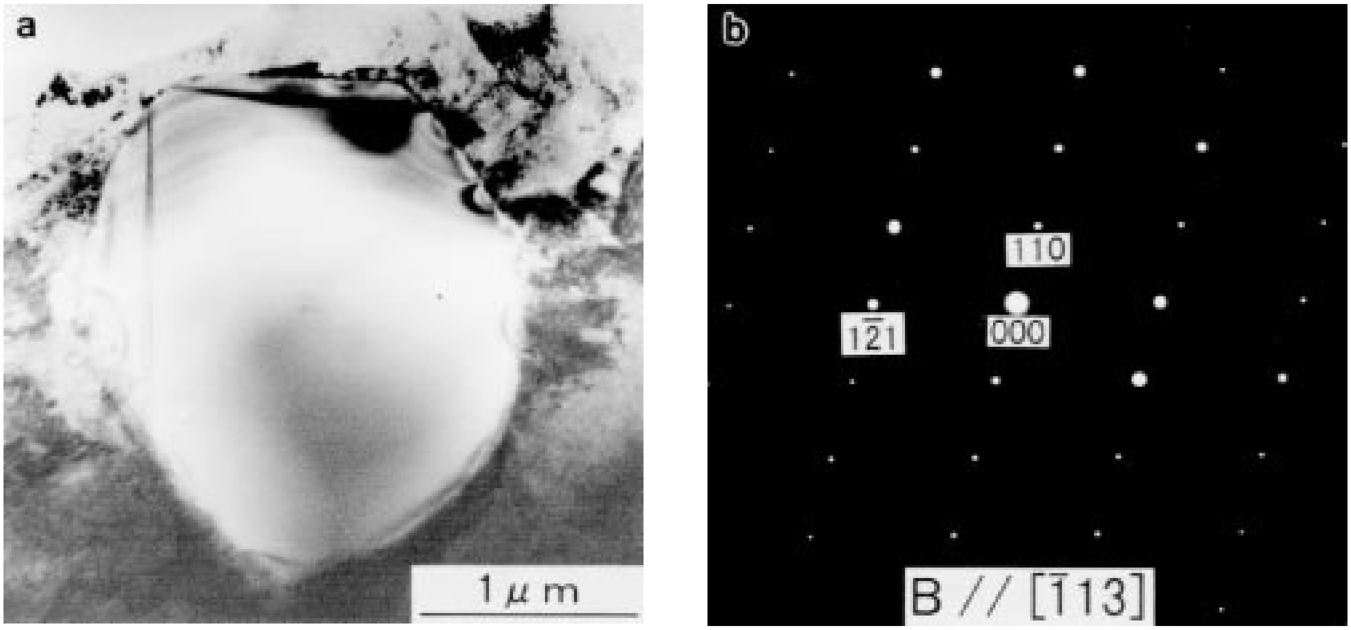

Fig. 7 TEM images of lumpy precipitate: (**a**) bright-field image and (**b**) diffraction pattern

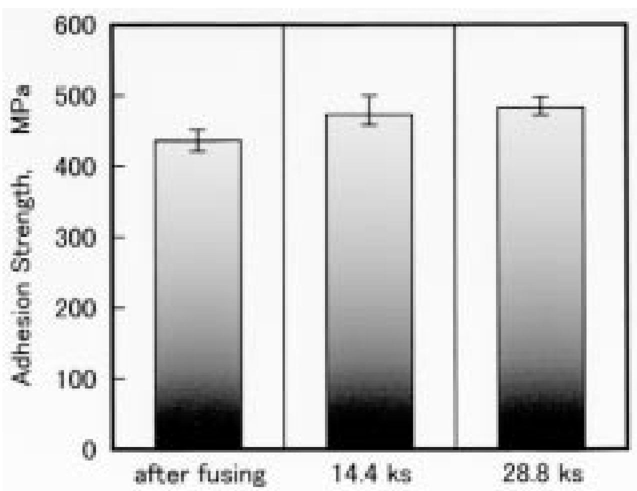

Fig. 8 Shear adhesion strength of coatings. Coatings were heated at 1273 K for 14.4 and 28.8 ks in air, except for as-sprayed condition. Longitudinal bars show maximum and minimum strength.

(JIS-H8666). Therefore, the adhesion was evaluated by a shear coating and the steel substrate.

The adhesion strength of the self-fluxing coating exceeded values that could be evaluated by the standard tensile test method (JIS-H8666). Therefore, the adhesion was evaluated by a shear test using notched specimens. Figure 8 shows the shear adhesion strength of the coating after fusing and heating at 1273 K for 14.4 and 28.8 ks, respectively. The adhesion strength increases slightly after heating at 1273 K. It is clear that adhesion does not decrease even if Fe_2B boride precipitates form at the interface. This may be because the interdiffusion at the interface compensates for the reduction of the interface strength by the Fe_2B formation.

4. Conclusions

The structure of the interface between nickel-base coating (JIS-MSFNi4) and mild steel substrate was investigated by using EPMA, XRD, and TEM. The shear adhesion strength of the coating was also measured.

The results are summarized as follows:

- Lumpy Fe_2B boride precipitates form in the substrate at the interface between the coating and the steel substrate after heating at 1273 K for 1.8 ks or more.
- The adhesion strength of the coating is not decreased by Fe_2B formation at the interface.

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