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Stainless Steel Foil with Excellent High-Temperature Strength and Oxidation Resistance "JFE20-5HS"

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Abstract:

Al-containing stainless steel foil "JFE20-5HS", which has 1.5 times higher high-temperature strength than that of conventional steel and excellent oxidation resistance, has been developed. There were two problems in increasing the high-temperature strength of the conventional steel: 1) to prevent deterioration of oxidation resistance, and 2) to ensure sufficient toughness of hot-rolled sheet for stable cold rolling. In order to solve these problems, the effect of the solid solution strengthening element on the oxidation resistance of the high-Al steel was investigated, and Mo was found as the element with little deterioration of the oxidation resistance. Furthermore, by clarifying the relationship between the amount of Mo addition and the toughness of the hot rolled sheet, the optimum amount of Mo addition was determined. By applying this product to metal honeycomb of automobiles, it is expected to improve heat resistance property of catalytic converters.

1. Introduction

Automobiles are equipped with a catalytic converter to detoxify harmful components contained in exhaust gas. Catalytic converters have a structure in which a catalyst carrier with a honeycomb structure is inserted inside a shell made from a stainless steel sheet. The materials used as catalyst carriers are ceramic carriers or stainless steel foil. **Figure 1**[1)] and **Figure 2** show the appearance of a catalyst carrier (metal honeycomb) made from stainless steel foil and a schematic diagram of its structure. Stainless steel foil having a thickness of approximately 30 to 100 μm is used in the metal honeycomb, which is formed by stacking corrugated foil produced by corrugation processing and unprocessed flat foil, and the winding the stacked foils into a honeycomb structure. The harmful components in exhaust gas are purified by passing the gas through the interior of this catalyst carrier, which carries the precious metal catalyst on its surfaces. Ceramic carriers are generally the main stream type used in catalyst carriers for four-wheeled vehicles, although metal honeycomb carriers are used in high performance automobiles that require high output and low pressure loss. At the same time,

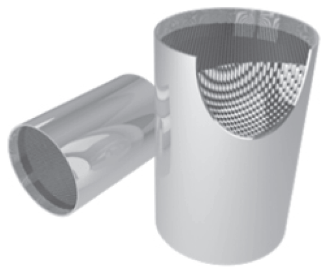

Fig. 1 Appearance of the metal honeycomb

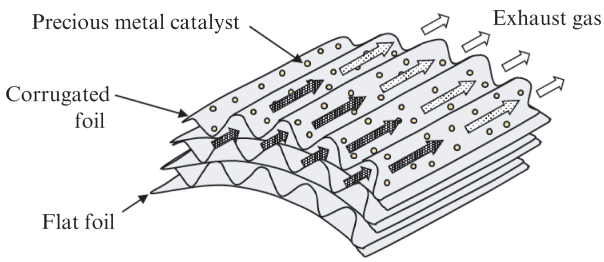

Fig. 2 Schematic of metal honeycomb system

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metal honeycombs, which possess higher vibration resistance than ceramics, are the main type used in motorcycles, as the catalytic converter is more easily subjected to impact and vibration than in four-wheeled automobiles for structural reasons[2–5].

In some cases, the interior of a catalytic converter can reach a high temperature exceeding 1 000°C due to the heat generated by the reaction between the catalyst and the exhaust gas in addition to the temperature of the exhaust gas itself, and for this reason, extremely high oxidation resistance is required in stainless steel foils used in metal honeycombs. In addition, because sudden temperature changes also occur accompanying changes in engine output, it is also necessary to prevent any large changes in the shape of the stainless steel foil or spalling of the surface oxidation film when the converter is subjected to cyclical heating and cooling. To satisfy these stringent requirements, SUH21 (18Cr-3Al steel) or 20Cr-5Al steel, in which 3% or more of Al is added to the stainless steel, is used in the metal honeycomb[5]. When ordinary stainless steel is oxidized at high temperature, a Cr oxide scale consisting mainly of Cr_2O_3 forms on the surface, but in severe high-temperature environments such as the interior of a catalytic converter, the oxidation resistance of the Cr_2O_3 is inadequate[6]. However, if 3% or more of Al is added to stainless steel, the Al will oxidize preferentially, forming a dense Al oxide scale consisting of Al_2O_3 as its main component[7]. Since this Al_2O_3 scale has higher stability at high temperatures than the Cr_2O_3scale, and its growth rate is also slow, if an Al_2O_3 scale is formed on the surface of stainless steel, it demonstrates an extremely high protective property[8]. As metal honeycombs materials, JFE Steel produces and sells the Al-containing stainless steel foil JFE20-5USR (20Cr-5Al-0.1La-0.03Zr steel) and JFE18-3USR (18Cr-3Al-0.1La-0.03Zr steel), in which oxidation resistance is dramatically improved by addition of La and Zr. These materials have an extensive record of use to date[9–10].

In recent years, higher exhaust gas temperatures are increasingly adopted to meet demand for improved automotive fuel economy in response to stricter environmental regulations. Because the strength of the stainless steel foils used in metal honeycombs decreases at higher exhaust gas temperatures, this trend has heightened concerns about distortion or breakage of foil materials due to vibration and thermal stress during travel. Although JFE20-5USR and JFE18-3USR have extremely high oxidation resistance, the temperature of the catalytic converter environment is also expected to increase in future, and it is possible that the high-temperature strength of these materials may be inadequate for some high-temperature environments. Thus, increased high-temperature strength is required in high Al-containing ferritic stainless steel foils for use in metal honeycombs. JFE Steel succeeded in developing a new high Al-containing ferritic stainless steel foil, “JFE20-5HS,” with 1.5 times higher high-temperature strength than the conventional steel, JFE20-5USR, and the same excellent oxidation resistance as the conventional steel. This report describes the composition design policy and material properties of JFE20-5HS.

2. Composition Design of JFE20-5HS

In this development, composition design was carried out based on the composition of the conventional steel, JFE20-5USR, with the aim of increasing high-temperature strength. The development target was to increase high-temperature strength as much as possible without reducing the excellent oxidation resistance of the conventional steel within the range where stable production of foil materials is possible.

First, the chemical composition of the conventional steel will be explained. Al exceeding 3% is added to the stainless steel foils used in metal honeycombs in order to impart excellent oxidation resistance. Here, “excellent oxidation resistance” means the growth of the Al_2O_3 scale is slow, and as a result, the life of the foil until the Al in the steel is exhausted is long, deformation of the foil does not occur under the expansion and contraction caused by repeated heating and cooling, and the Al_2O_3 scale does not peel off due to repeated expansion and contraction. Although Al_2O_3 scales generally have higher stability at high temperatures than Cr_2O_3 scales, the adhesion of Al_2O_3 scale with the substrate is inferior, resulting in easy spalling of the scale[11]. To overcome this issue, reactive elements (REs) such as Y, Zr, La, and Hf or Ti, etc. are added to high Al-containing stainless steel foils for metal honeycombs in order to improve the spalling resistance of the Al_2O_3 scale[12]. Among these REs, JFE Steel produces and sells the Al-containing stainless steel foil JFE20-5USR (20Cr-5Al-0.1La-0.03Zr steel), which provides dramatically enhanced oxidation resistance as a result of compound addition of La and Zr[13].

Next, the method of improving the high-temperature strength of the conventional steel will be described. Among the strengthening mechanisms of steel, that is, solid solution strengthening, dislocation strengthening, dispersion strengthening and grain refinement strengthening[14], solid solution strengthening is also considered to be effective in the high-temperature region exceeding 1 000°C. As solid solution strengthening elements that can increase the high-temperature strength of stainless steel, for example, Mo, W, Nb and Ta may be mentioned. On the other hand, there are many unknowns regarding the effects of these solid solution strengthening elements on the oxidation resistance of high Al-containing ferritic stainless steel, and reduction of the toughness of the steel by the addition of these solid solution strengthening elements is also a concern. Since the toughness of high Al-containing ferritic stainless steels is inherently greatly inferior to that of general ferritic stainless steels[15], production of stainless steel foils may itself become impossible if the toughness of this type of steel is further reduced by the addition of solid solution strengthening elements. Thus, when attempting to increase the high-temperature strength of the conventional steel, a detailed composition design that also considers oxidation resistance and manufacturability is required.

To address these issues, in this development, we began by examining the effects of the solid solution strengthening elements on the high-temperature strength and high-temperature oxidation resistance of 20Cr-5Al steel. As a result, Mo was found to be a favorable solid solution strengthening element for this development. We then investigated the effect of Mo addition on the toughness of the hot-rolled sheet of the 20Cr-5Al steel in order to determine the optimum amount of Mo addition for increasing high-temperature strength within the range where stable manufacture of foil materials is possible. The following presents the results obtained through the process of these studies.

2.1 Effects of Solid Solution Strengthening Elements on High-Temperature Strength and Oxidation Resistance

2.1.1 Experimental method

Steel ingots of the compositions shown in **Table 1** were melted by vacuum melting, and cold-rolled sheets with a thickness of 1 mm were obtained through the hot rolling and cold rolling processes. Next, cold-rolled and annealed sheets were produced by performing heat treatment, in which these cold-rolled sheets were held at 1 000°C for 1 min, and the cold-rolled and annealed sheets were cold rolled to obtain foil materials with a thickness of 50 μm. High-temperature strength and oxidation resistance were evaluated using these cold-rolled and annealed sheets and foil materials.

High-temperature strength was evaluated by a high-temperature tensile test of the cold-rolled and annealed sheets using tensile strength (TS) at 900°C as an index of high-temperature strength. The high-temperature tensile test was conducted at 900°C in accordance with the method specified in JIS G 0567 using tensile test pieces with a sheet thickness of 1 mm, parallel part width of 10 mm and gauge length of 50 mm. The crosshead speed was 5 mm/min.

Oxidation resistance was evaluated by preparing small-sized test pieces simulating the metal honeycomb and measuring the amount of weight change and length change (evaluated here as length change) when the test pieces were held at high temperature. **Figure 3** shows the test piece preparation method for evaluation of oxidation resistance, and **Fig. 4** shows the length change measurement method. First, two samples with a width of 75 mm × length of 300 mm were taken from

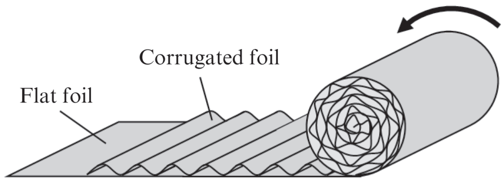


Fig. 3 Preparation method of test piece for oxidation test

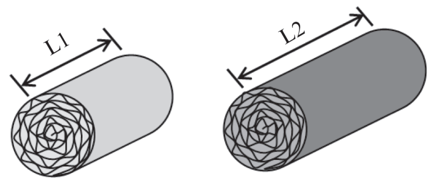


Fig. 4 Evaluation method of length change after oxidation test

Table 1 Chemical composition of test materials for evaluation of high temperature strength and oxidation resistance

(mass%)

Steel	C	Si	Mn	Al	Cr	Mo	Nb	Ta	W	La	Zr	Remarks
Base	0.005	0.17	0.15	5.3	19.9	—	—	—	—	0.12	0.03	Conventional steel
3Mo	0.005	0.15	0.15	5.3	20.1	3.1	—	—	—	0.14	0.03	
5Mo	0.005	0.18	0.16	5.8	20.3	4.9	—	—	—	0.10	0.03	
0.2Nb	0.008	0.16	0.13	5.5	20.0	—	0.2	—	—	0.05	0.03	
0.3Nb	0.009	0.16	0.13	5.4	19.9	—	0.35	—	—	0.03	0.02	
0.5Nb	0.009	0.16	0.14	5.5	19.8	—	0.5	—	—	0.06	0.03	
2Ta	0.007	0.14	0.15	5.7	20.0	—	—	2.1	—	0.10	0.03	
3Ta	0.008	0.13	0.15	5.6	20.0	—	—	3.3	—	0.08	0.03	
5Ta	0.008	0.11	0.15	5.7	20.0	—	—	5.5	—	0.13	0.03	
2W	0.006	0.16	0.15	5.6	20.2	—	—	—	2.1	0.07	0.03	
5W	0.006	0.14	0.15	5.7	20.2	—	—	—	5.2	0.06	0.03	

the foil material with the thickness of 50 μm, and corrugation processing of one of the sample sheets was performed by passing the sample between two gears. The corrugated foil and flat foil were then stacked and wound, after which the end part was welded. The test pieces for evaluation in the oxidation test were prepared by performing heat treatment by holding the samples in a vacuum at 1 150°C for 30 min to simulate the brazing process. The oxidation test was carried out at 1 100°C in atmospheric air. During the test, the test pieces were removed from the furnace at holding intervals of 5 to 50 h, and the weight change and the length change were measured.

2.1.2 Results and discussion

The effects of the various added elements on the high-temperature strength of the 20Cr-5Al steel are shown in **Fig. 5**. The high-temperature strength of the 20Cr-5Al steel increased when each of the solid solution strengthening elements was added. Comparing the increase in high-temperature strength per amount of addition, the highest unit increase was 74% with addition of 0.35% Nb. The next highest increase after Nb was obtained by addition of Mo, and the increase in high-temperature strength decreased with Ta and W, in that order. The increases obtained with addition of approximately 5% Mo, Ta and W were 82%, 69% and 48%.

Next, the oxidation resistance of the steels with these added elements was evaluated. **Figure 6** shows the effect of the added elements on the increase in oxidation amount (measured as weight change) and the length change of the 20Cr-5Al steel after holding at 1 100°C for 400 h in the air. In this oxidation test, spalling of the Al_2O_3 scale did not occur in any of the steels. Comparing the amount of oxidation, weight change increased with the addition of each of the solid solution strengthening elements in comparison with that of the base steel without addition. The weight change after oxidation differed depending on the element. There was little difference between the increase in oxidation of the Mo-added steel, W-added steels and base steel. However, in comparison with the base steel, the weight change was remarkably larger in the Nb-added and Ta-added steels. In particular, the influence of Nb addition was large, as brealaway oxidation occurred with the 0.5 Nb steel after holding at 1 100°C for 400 h. From these results, although the weight change increased with addition of all of the elements Mo, Nb, Ta and W, this test revealed that the increase in weight change was comparatively slight with Mo and W. Following this, the amount of length change after the oxidation test was compared. The effects of the solid solution strengthening elements on length change also differed greatly depending on the element. The change in length during the oxidation test was extremely large with the Nb-added and Ta-added steels, but in contrast, shape change was slight in the Mo-added and W-added steels. An analysis of the oxide that formed

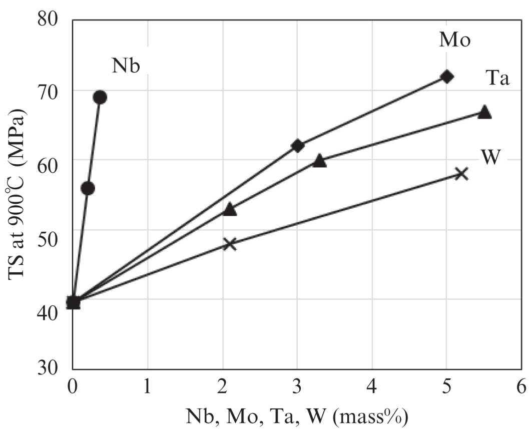
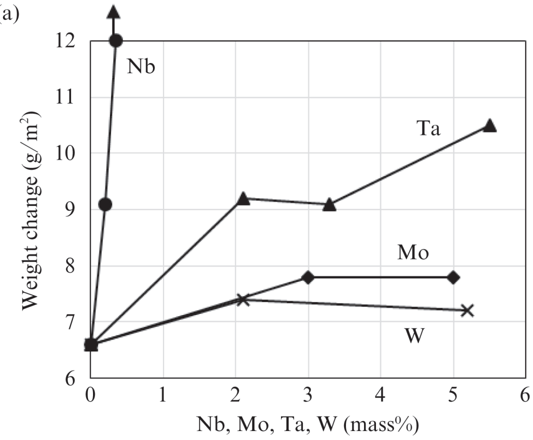


Fig. 5 Effect of additive elements on high-temperature strength of 20Cr-5Al Steel

(a)



(b)

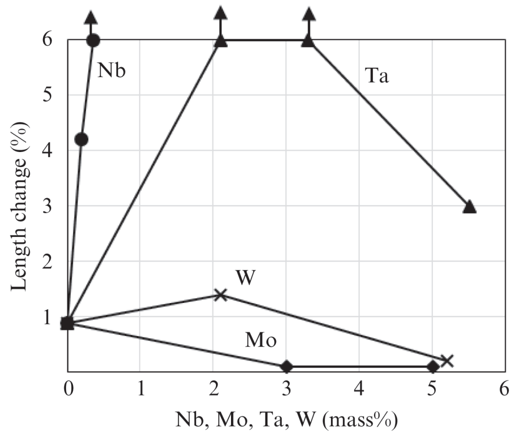


Fig. 6 Effect of additive elements on (a) weight change and (b) length change of 20Cr-5Al steel after 400 h oxidation at 1 100°C

on the surface revealed that only an oxide consisting mainly of fine, dense Al_2O_3 formed on the surface of the Mo-added and W-added steels, while a complex oxide consisting of $FeNbO_4$ or $FeTaO_4$ in addition to Al_2O_3 had formed on the surface of the Nb-added steel and Ta-added steel, respectively. Because these complex oxides are porous and large volumetric changes occur during their formation, it is thought that the shape changes observed in these test pieces were caused by the large stress generated by the growth of the oxide. Moreover, it can also be pointed out that the formation of these oxides may lead to an increase in the oxidation velocity of the Nb-added and Ta-added steels.

If breakaway oxidation and/or shape change occur in a metal honeycomb, spalling of the catalyst held on the carrier surface and blockage of the gas channel occur. Considering this point, we concluded that Nb and Ta are not suitable for use as solid solution strengthening elements added to materials for metal honeycombs, as these elements invite remarkable increases in the oxidation velocity and shape change of the foil. Comparing the amount of increase in high-temperature strength per unit addition of Mo and W, which did not cause a remarkable decrease in oxidation resistance, the unit strength increment of Mo was larger than that of W. Therefore, based on the investigation results described above, Mo was selected as the solid solution strengthening element in this development.

2.2 Effect of Mo on Toughness of Hot-Rolled Sheet

2.2.1 Experimental method

If Mo is added to 20Cr-5Al steel, high-temperature strength increases, but on the other hand, the toughness of the hot-rolled sheet decreases. For stable foil rolling, it is necessary to determine the amount of Mo addition by also considering the toughness of the hot-rolled sheet. Therefore, we investigated the effect of the amount of Mo addition on the toughness of hot-rolled sheets of 20Cr-5Al steel. Here, the toughness of hot-rolled sheets was investigated by the Charpy impact test. Among the steels shown in Table 1, ingots of steels with different Mo contents from 0 to 5% (Base, 3Mo, 5Mo) were processed into sheet bars with a thickness of 30 mm by hot rolling. These sheet bars were reheated to 1 200°C and hot rolled to obtain hot-rolled sheets with a thickness of 3 mm. Next, 2 mm V-notch Charpy impact test pieces with a thickness of 3 mm (conforming to the JIS No. 4 test piece except for the sheet thickness) were taken from these sheets and used in the Charpy impact test (testing machine capacity: 490.3 J). As the sampling direction of the test pieces, the longitudinal direction of the test pieces was the same as the rolling direction, and the notch was made so that the crack would grow in the vertical direction with respect to the rolling direction. The test was conducted with 3 samples of each material at 1 temperature level, and the transition curve was obtained by measuring the absorbed energy and brittle fracture ratio. The ductile-brittle transition temperature (DBTT) was defined as the temperature at which the brittle fracture ratio became 50%.

2.2.2 Results and discussion

Figure 7 shows the results of the Charpy impact tests of the 20Cr-5.8Al hot-rolled sheets with different Mo contents. As the Mo content increased, the Charpy impact value and the transition curve of the brittle fracture ratio shifted to the high temperature side. However, there was almost no change in the upper shelf energy when the Mo content was changed. **Figure 8** shows the relationship between DBTT and the Mo content read from Fig. 7 (b). It can be understood that DBTT increased accompanying increases in the Mo content.

Next, from the viewpoint of securing hot-rolled

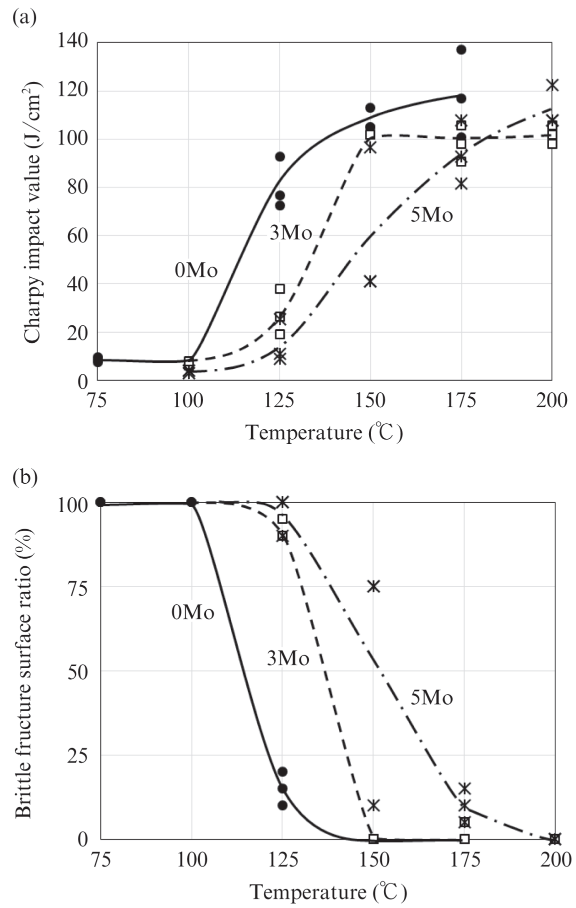


Fig. 7 Charpy impact test results of 20Cr-5Al-(0∼5) Mo steel
(a) Charpy impact value (b) Brittle fracture surface ratio

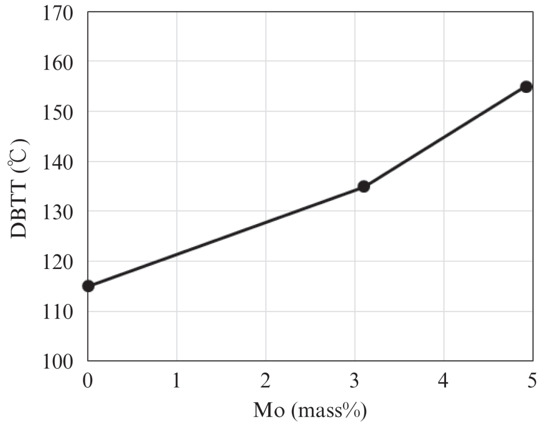


Fig. 8 Effect of Mo on ductile-brittle transition temperature of hot rolled 20Cr-5Al steel

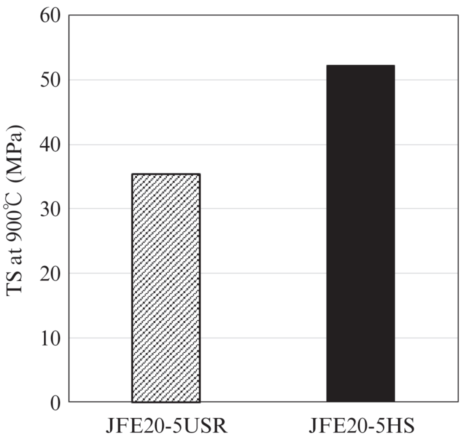


Fig. 9 High-temperature strength of conventional steel and developed steel

sheet toughness that enables stable foil rolling, the optimum amount of Mo addition was studied. A separate investigation of the relationship between the DBTT of the hot-rolled sheet and the strip breakage rate during cold rolling revealed that the breakage rate decreases greatly and stable cold rolling is possible if DBTT is 140°C or less. On the other hand, from the results shown in Fig. 8, it can be understood that DBTT decreases to less than 140°C when the Mo content is no more than 3%. Based on these study results, the Mo content of the developed steel was set at 3%.

3. Characteristics of JFE20-5HS

The composition of the developed steel was decided based on the composition design guidelines described up to this point. **Table 2** shows the chemical composition of the conventional steel, JFE20-5USR, and the developed steel, JFE20-5HS. In comparison with the conventional steel, the high-temperature strength of the developed steel was increased by 3% addition of Mo. In the following, the high-temperature strength and high-temperature oxidation resistance of the developed steel are shown by comparison with the conventional steel.

3.1 High-Temperature Strength

Hot-rolled sheets of the developed steel and the conventional steel produced at East Japan Works were sampled, and cold-rolled sheets with a thickness of 1 mm were prepared using a laboratory cold rolling mill. The cold-rolled sheets were heat-treated by holding at 1 000°C for 1 min, followed by air cooling, and the high-temperature strength of the obtained materials was investigated by performing a high-temperature tensile test under the same conditions as in section 2.1.1. The results are shown in **Fig. 9**. The tensile strength (TS) at 900°C of the conventional steel was 35 MPa, while that of the developed steel was 52 MPa, showing that approximately 1.5 times higher high-temperature strength was realized in the developed steel.

3.2 High-Temperature Oxidation Resistance

Foils (thickness: 40 μm) of the developed steel and the conventional steel produced at East Japan Works were sampled, and an oxidation test was carried out under the same conditions as in section 2.1.1. The materials investigated in this experiment had two types of final finish, as-rolled (HARD material) and bright annealed (BA material). These two types of final finish are used appropriately depending on the degree of processing and the part to which the material is applied. The results of the oxidation tests of the HARD material and BA material are shown in **Fig. 10** and **Fig. 11**, respectively. With both materials, no breakaway oxidation or spalling of the Al_2O_3 scale was observed after the oxidation test, and a sound external appearance was maintained. The increase in weight change after oxidation was slightly larger in the developed steel than in the conventional steel. However, since this anomaly was very slight, it can be judged that the developed steel has substantially the same oxidation life as the

Table 2 Chemical composition of JFE20-5USR and JFE20-5HS

(mass%)

Steel	C	Si	Mn	Al	Cr	Mo	La	Zr	Remarks
JFE20-5USR	0.06	0.15	0.15	5.5	20	—	0.1	0.03	Conventional steel
JFE20-5HS	0.06	0.15	0.15	5.5	20	**3**	0.1	0.03	Developed steel

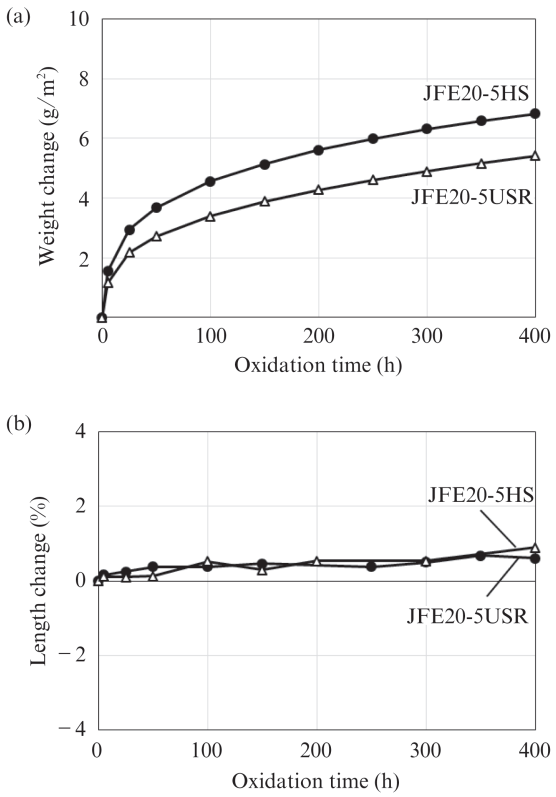


Fig. 10 Oxidation behavior of conventional steel and developed steel at 1 100˚C (40 μm thickness, HARD)
(a) Weight change (b) Length change

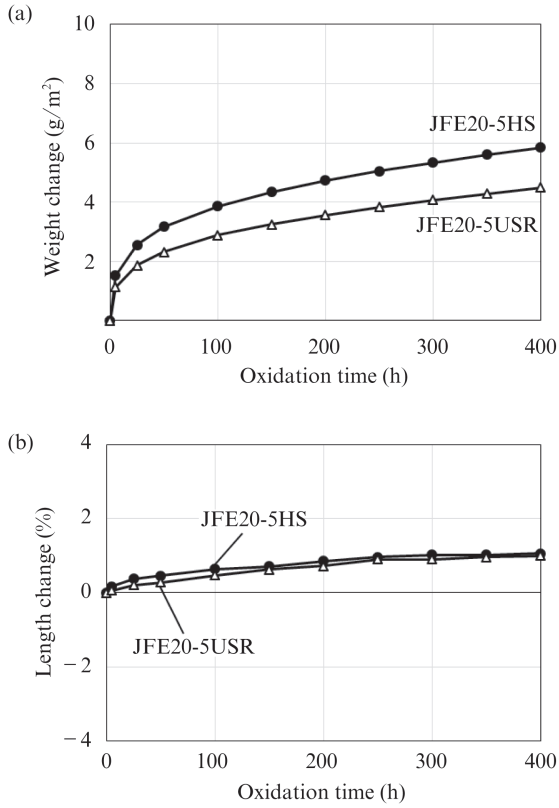


Fig. 11 Oxidation behavior of conventional steel and developed steel at 1 100˚C (40 μm thickness, BA)
(a) Weight change (b) Length change

conventional steel. The shape change (length change) of the developed steel after the oxidation test was also extremely small, at less the ±1%, which was substantially the same as that of the conventional steel. From these results, it can be understood that the developed steel has extremely high oxidation resistance, equivalent to that of the conventional steel.

Comparing the weight change after oxidation of the HARD and BA materials, the increase in oxidation was slightly smaller in the BA materials of both the conventional steel and the developed steel. Here, it is thought that the growth rate of the oxide decreased due to changes in the properties of the surface oxide formed by bright annealing and the oxide formed by subsequent oxidation in air[16].

4. Conclusion

This paper has described the composition design guidelines and material properties of the newly-developed high Al-containing ferritic stainless steel foil, “JFE20-5HS,” which has both excellent high-temperature strength and oxidation resistance. The key points are as follows.

- Mo was found as a solid solution strengthening element that effectively increases high-temperature strength while maintaining the excellent oxidation resistance of 20Cr-5Al steel.
- The amount of Mo addition was set at 3% in order to secure hot-rolled sheet toughness that enables stable foil rolling.
- The tensile strength at 900˚C of JFE20-5HS is 1.5 times higher than that of the conventional steel, JFE20-5USR.
- JFE20-5HS has excellent oxidation resistance, equivalent to that of JFE20-5USR, and no breakaway oxidation occurred even when foil with a thickness of 40 μm was held at 1 100˚C for 400 h. Shape stability at high temperature was also excellent, as the amount of shape change during high-temperature oxidation was extremely small.

Because the developed steel has higher strength in the high-temperature region than the conventional steel, it is possible to apply this product to automobiles or motorcycles with a higher exhaust gas temperature than the conventional type. Improved heat resistance and longer life can also be expected in metal honeycombs by application of this product. JFE Steel intends to contribute to further improvement of automotive fuel economy and solution of global environmental problems by expanding the application of this product.

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