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Brazing of metallic conductors onto ceramic plates in solid oxide fuel cells

Part II *Attaching conducting wires*

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Commercialization of the solid oxide fuel cell (SOFC) will be facilitated by use of conventional materials and fabrication processes. In this paper, we discuss the results of brazing metallic wires, made of conventional heat-resisting alloys, onto metallic $CrFe5Y_2O_31$ conductor plates in SOFC. Such wires would be used for the current transport between individual SOFC stacks and to the power-consuming device. Aluminum-alloyed ferritic stainless steels (iron, 20 to 25 wt% chromium, 4.5 to 6 wt% aluminum) and Inconel 617 were found to be suitable materials for the wires. They can be brazed onto $CrFe5Y_2O_31$ using L-Ni 5 as filler for Inconel 617 and the ferritic steels, and Cr50Ni or L-Ni 5 for the ferritic steels. The effectiveness of the brazed conductor/$CrFe5Y_2O_31$ joints was verified by monitoring their resistance at 1000°C in air for 1000 h. © *2001 Kluwer Academic Publishers*

1. Introduction

The solid oxide fuel cell (SOFC) is a very promising candidate for stationary cogeneration and primary power for a wide range of applications due to its specific advantages, such as high efficiency, low emissions, tolerance against fuel impurities, and the flexibility of fuels that can be used. Use of conventional materials and fabrication processes would accelerate commercialization since they would reduce the cost for SOFC technology. The goal of the present work was to find conventional materials and processes that would allow the low-resistance current transfer between individual SOFC stacks and to the power consuming unit [1]. The development was done for a planar SOFC system that is operated at high temperatures, 1000°C and above [2]. This SOFC system possesses problems related to the bonding of metallic conductors to the ceramic end plates of the SOFC stack. Lanthanum chromite ($LaCrO_3$) is the state-of-the-art ceramic end plate for SOFC units operating at temperatures of 1000°C or above [2, 3]. It offers a good electrical conductivity as well as compatibility and good match of thermal expansion with other SOFC components.

Heat-resistant nickel alloys (e.g., Inconel 617) and ferritic stainless steels (e.g., iron alloyed with 22 wt% chromium and 5.5 wt% aluminum) are promising conventional conductor materials. However, the joining of these alloys to ceramic $LaCrO_3$ plates is problematic due to the large mismatch of thermal expansion. Between 20°C and 1000°C, $LaCrO_3$ has a thermal expansion coefficient of 10.4×10^{-6}/°C [1], while the heat-resisting alloys have a much higher thermal expansion coefficient, 14×10^{-6}/°C or more (Table I). The mismatch causes bending or cracking of the thin (<1 mm), fragile ceramic plate due to thermal stresses. Such stresses occur during the cooling after brazing and during the heating-up or cooling-down of the SOFC unit.

We developed a two-step process to connect such conventional conductors to ceramic end plates by vacuum furnace brazing. In a first step, a metallic current collector plate is attached to the ceramic end plate. The metallic plate compensates for the lower electronic conductivity of $LaCrO_3$. As reported in part 1, the alloy $CrFe5Y_2O_31$ (chromium alloyed with 5 wt% iron and 1 wt% yttria) is a suitable current collector material since it matches the thermal expansion of $LaCrO_3$. Such $LaCrO_3$/$CrFe5Y_2O_31$ joints can be brazed with SCP6 (copper with 18 wt% palladium) as filler alloy. The long-term stability of this joint under SOFC operating conditions has been proven.

In this paper, we present results concerning the second brazing step, in which wire conductors are brazed onto the current collector plate. For several reasons, the metallic current collector plate is highly advantageous for attaching the wire conductors. Due to the high in-plane conductivity of the metallic plate, the current collector needs to be contacted with only a few

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TABLE I Characteristics of the metallic conductors

Material	Vendor	Composition (main elements), wt%	α, $10^{-6}/^\circ$C	Specific Resistance at 20°C	Melting Range, °C	Form (∅ = diameter)
Inconel 600	BIBUS Inco Alloys GmbH, Duesseldorf, Germany	Ni bal., Cr 14-17, Fe 6-10, Mn < 1, Cu < 0.5	16.7 (20-982°C)	103 $\mu\Omega$·cm	1354-1413	wire, ∅ 1.8 mm
Inconel 617	BIBUS Inco Alloys GmbH, Duesseldorf, Germany	Ni bal., Cr 20-24, Co 10-15, Mo 8-10, Al 0.8-1.5, Fe < 3, Mn < 1, Ti < 0.6, Cu < 0.5	16.6 (20-1093°C)	122 $\mu\Omega$·cm	1330-1380	wire, ∅ 2.4 mm
Haynes 214	Haynes International Inc., USA, Kokomo, IN, USA	Ni bal., Cr 16, Al 4.5, Fe 3, Y < 0.01, Zr < 0.1, B < 0.01,	18.6 (25-1000°C)	136 $\mu\Omega$·cm	1355-1400	wire, ∅ 2 mm
Aluchrom I	Krupp VDM GmbH, Werdohl, Germany	Fe bal., Cr 19-22, Al 4.5-5.5	14.8 (20-1000°C)	137 $\mu\Omega$·cm	≈1500	wires, ∅ 2.5 and 3.5 mm
Aluchrom O	Krupp VDM GmbH, Werdohl, Germany	Fe bal., Cr 22-25, Al 4.5-6	15 (20-1000°C)	144 $\mu\Omega$·cm	≈1500	wires, ∅ 2 and 3.5 mm
Aluchrom Y	Krupp VDM GmbH, Werdohl, Germany	Fe bal., Cr 22, Al 5, Y 0.05-0.15, Ti 0.01-0.1, Zr 0.01-0.1	14.8 (20-1000°C)	139 $\mu\Omega$·cm	≈1500	wire, ∅ 2.5 mm
Hoskins 145	Berghuetten GmbH, Dietzenbach, Germany	Fe 71.7, Cr 21.1, Al 6.2[1]	≈15 (20-1000°C)	145 $\mu\Omega$·cm	≈1500	wire, ∅2 mm
Hoskins 145	Berghuetten GmbH, Dietzenbach, Germany	Fe 71.4, Cr 20.7, Al 5.4[1]	≈15 (20-1000°C)	145 $\mu\Omega$·cm	≈1500	strip, 0.6 × 6 mm

[1] Measured by inductively coupled plasma-atomic emission spectroscopy. All other data specified by vendors [4-9].

wire conductors. This joint area is therefore significantly smaller than the one between the end plate and the current collector. Furthermore, the metallic current collector plate can be much thicker than the ceramic end plate because of its superior conductivity. Bending or cracking of the metallic plate due to thermal stresses between plate and conductor is unlikely to occur. For these reasons, conventional heat-resistant nickel alloys and ferritic stainless steels can be used as conductors despite their much higher thermal expansion coefficients.

2. Experimental procedure

2.1. Materials

Oxide dispersion strengthened (ODS) alloys, as well as alloys that require a special heat treatment to guarantee the required heat resistance, were not employed as conductor materials. Such materials are expensive and difficult to process. The conductor materials tested and their physical characteristics are presented in Table I.

Three nickel alloys were examined. Inconel 600 and Inconel 617 are common oxidation resistant alloys. The third employed nickel alloy, Haynes 214, offers the best oxidation resistance because of its high aluminum content. This is attributable to the formation of a tightly adherent Al_2O_3-type oxide scale, which forms in preference to chromium oxide scales at temperatures of 955°C and above [8].

Additionally, various ferritic stainless steels were tested (Aluchrom I, Aluchrom O, Aluchrom Y, and Hoskins 145). Their high aluminum content of at least 4.5 wt% enables the formation of a well adhering, ductile Al_2O_3 layer, which provides excellent resistance to further oxidation. These materials are used as elements in resistance heating furnaces or heaters [5, 9]. Their oxidation resistance and their cost increase with increasing addition of aluminum. Aluchrom Y, which is also used as support for automotive catalysts, is alloyed with small amounts of zirconium, yttrium, and titanium to enhance the heat resistance [9]. Aluchrom I and Aluchrom O were used as wires with different diameters to evaluate the influence of the wire diameter on the oxidation behavior and the resulting resistance of the brazed joints. The material Hoskins 145 was used as wire and as strip with a cross-sectional area of 0.6 mm × 6 mm to determine if such a relatively thin strip can be used as conductor. Unlike the wires with a diameter greater 1.8 mm, formation of a 300 μm thick oxidation layer would cause complete oxidation in the strip, leading to the loss of its electrical conductivity.

One criterion for selecting suitable filler alloys was that their melting temperatures be higher than 1000°C, the present SOFC system operating temperature. [2]. This is required to avoid softening of the braze filler metal or metallurgical degradation due to excessive diffusion during SOFC operation. The filler alloys used for this study are described in Table II. Nickel-based

TABLE II Characteristics of the filler alloys

Filler	Manufacturer	Composition, wt%	Form	Melting range, °C	Brazing Program (in vacuum < 10^{-5} mbar)
L-Ni 5	Degussa AG, Hanau, Germany [10]	Ni 71, Cr 19, Si 10, C < 0.1	powder	1080-1135	Heating with 10°C/min Holding at 1200°C for 5 min Cooling with 20°C/min
Cr50Ni	Plansee, Reutte (Tirol), Austria	Cr 50, Ni 50	foil, 100 μm thick	≈1350	Heating with 10°C/min Holding at 1370°C for 5 min Cooling with 20°C/min

conductors were only brazed with L-Ni 5, since the brazing temperature of Cr50Ni (1370°C) is higher than the solidus temperature of the nickel-based conductors. L-Ni 5 was mainly used to braze the ferritic stainless steels. One material (Aluchrom I, 3.5 mm diameter) was brazed with both filler materials to determine the influence of the filler alloy on the electrical resistance of the joint. Cr50Ni was used to braze the Hoskins 145 strip, since this filler alloy came in the form of a foil, which is more suitable for producing an areal joint than a powder (L-Ni 5 came as a powder).

The material used for the current collector plates, $CrFe5Y_2O_31$, was developed by Plansee (Reutte, Austria) for SOFC parts such as metallic interconnects [11]. Major advantages of this alloy are its good resistance against corrosion and the good match of its thermal expansion coefficient (11.3×10^{-6}/°C between 20°C and 1000°C) to that of $LaCrO_3$ (10.4×10^{-6}/°C in the same temperature range) [1]. The $CrFe5Y_2O_31$ plates used for this study had an area of 25 × 25 mm and a thickness ranging from 3 to 3.5 mm.

2.2. Furnace brazing parameters

The surfaces of the $CrFe5Y_2O_31$ plates were prepared by grinding and etching, and finally cleaned before brazing. A notch 10 mm long and 2 mm deep was machined into the plates if wires had to be attached. The notch was as wide, or slightly (between 0.1 and 0.2 mm) wider, than the diameter of the wire intended to be used. Filler alloy powder or, if the filler alloy came as a foil, tiny pieces of the foil were placed into the notch. Afterwards, the wire was put into the notch and covered with some filler powder or foil fragments. The Hoskins 145 strip was brazed directly onto the surface of the $CrFe5Y_2O_31$ plate. A Cr50Ni foil, 6 × 10 mm, was used, and a small weight was applied on the assembly to retain the strip in position during brazing. The length of the strip/plate bond was 10 mm for all joints. All of them were brazed in a vacuum of 10^{-5} mbar, following the brazing programs given in Table II.

2.3. Characterization methods

The most important criterion for the brazed joints is the long-term behavior of their electrical resistance during SOFC operation. Therefore, the resistances of the joints were measured at room temperature by four-point dc technique after various times of annealing at 1000°C in air (Fig. 1).

Four copper electrodes were employed: the two outer ones as current-carrying electrodes and the two inner ones as voltage probes. The voltage drop was measured at two positions, shown in Fig. 1. Voltage drop U1 was used to calculate the specific resistance of the metallic conductor (units of $\mu\Omega \cdot$ cm) close to the conductor/$CrFe5Y_2O_31$ bond. Voltage drop U2 was used to determine the integral resistance of the brazed conductor/plate joint (units of mΩ). The latter resistance was not converted to a specific resistance, because it is determined for several materials, and the cross-sectional area changes within the measuring length. Direct current ranging from 0.5 A to 5 A was applied, and the resolution of the voltmeter amounted to ± 1 μV. The samples were not affected by the resistance measurement. Thus, the resistance of the same joint could be measured after various times of annealing. As a result, we could accurately determine resistance changes in the metallic conductors as well as in the conductor/plate joints.

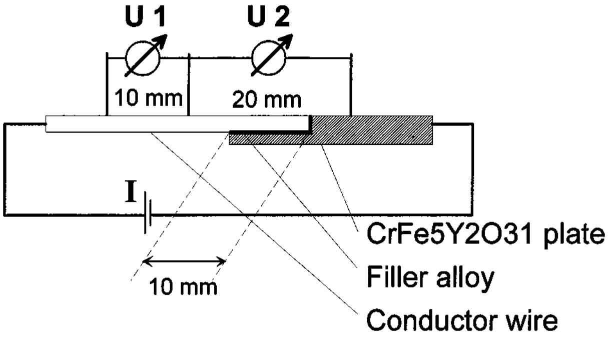

Figure 1 Schematic for resistance measurement of brazed joints by four-point de technique.

The microstructure of some joints was studied by scanning electron microscopy (SEM), and the element distribution was measured by energy dispersive X-ray spectroscopy (EDS).

3. Results and discussion

Fig. 2 shows the specific electrical resistances of the conducting wires brazed to $CrFe5Y_2O_31$ plates versus time of annealing (1000°C in air). If the same material was tested in different sizes, then the values given in Fig. 2 represent the smallest wire. The resistances of larger wires consisting of the same material are more stable during annealing, since the current-carrying cross-sectional area of thicker wires is less reduced by the formation of oxide layers.

Before annealing (0 h in Fig. 2), specific resistances similar to the literature values (see Table I) were measured for all but one conductor. This finding indicates a good accuracy of the four-point dc technique used. Only the specific resistance of the Hoskins 145 wire (156 $\mu\Omega \cdot$ cm) was higher than the literature value (145 $\mu\Omega \cdot$ cm). The higher aluminum amount of 6.2 wt% in this sample compared with the amount specified (e.g., the Hoskins 145 strip contained only 5.4 wt% aluminum) most likely caused the high resistance. Generally, the electrical resistance of an alloy increases with increasing amount of dissolved alloying elements, since the dissolved atoms distort the host lattice and thereby increase the resistance for electron transport.

The resistance of Haynes 214 and the ferritic stainless steels are significantly higher than those of the other nickel alloys, but remain constant during annealing. Both effects can mainly be attributed to the formation of an Al_2O_3 layer on the surface of these alloys. The Al_2O_3 layer protects them against further oxidation, but has itself a high electrical resistance.

In contrast to Haynes 214, the resistance of Inconel 600 and Inconel 617 increases by 5% or 6% during annealing. This degradation is most likely caused by

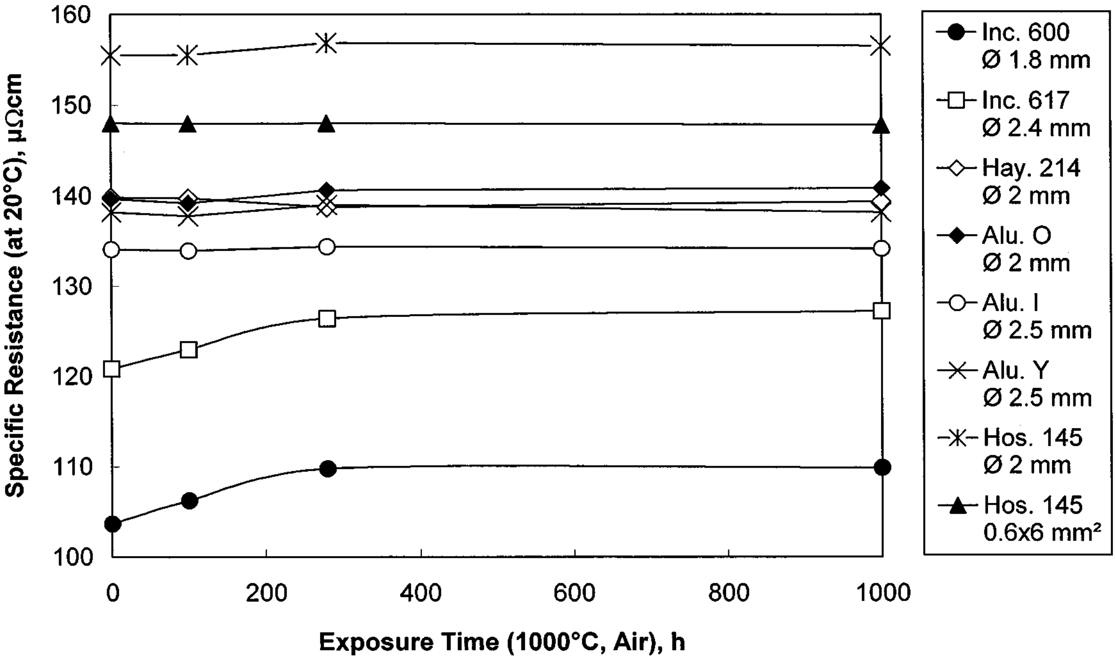

Figure 2 Electrical resistance of conductors, measured close to the conductor/CrFe5$Y_2O_3$1 bond. (Alu. = Aluchrom, Hay. = Haynes, Hos. = Hoskins, Inc. = Inconel).

higher oxidation rates in air for these materials due to the formation of oxides other than alumina. The higher oxidation rates lead to a measurable reduction of the current-carrying cross-sectional area. Compared with Inconel 600, the amount of degradation is slightly smaller for Inconel 617, because this alloy contains a small amount of aluminum, which leads to the formation of a better-protecting oxide layer. The resistance of both alloys increased mainly during the first hours of annealing, and leveled out after 280 h. Thus, these materials are still promising as conductors for the present purpose despite their inferior resistance against oxidation.

The comparatively thin Hoskins 145 strip exhibited a constant resistance during annealing. The oxidation occurs so slowly in this material that even the cross-sectional area of such a thin strip is not reduced significantly during annealing; this leads to a constant resistance.

The electrical resistance of the brazed conductor/CrFe5$Y_2O_3$1 joints, calculated from voltage drop U2, versus time of annealing (1000°C, air) is given in Fig. 3. This electrical resistance is influenced by the conductor, the CrFe5$Y_2O_3$1 plate, and the filler alloy. The influence of the conductor material and geometry is thereby dominating. This was concluded from the following two observations: (i) joints manufactured with a relatively thick wire ($\varnothing = 3.5$ mm) exhibit a much lower resistance than such manufactured with thinner wires ($\varnothing = 2$ to 2.5 mm) consisting of the same material, and (ii) the resistance of joints manufactured with nickel-based conductors is significantly lower than that of joints manufactured with steel conductors having a similar cross-sectional area. However, the resistance of some joints made with the nickel-based conductors (Inconel 600 and Haynes 145) increased by 12% and 25% during annealing, respectively. For both materials, the resistance of the joint increased more than the specific resistance of the wire itself. Therefore, we assume that the deterioration is not only caused by oxidation of the conducting wires, but also by changes in the conductor/plate bond or in the CrFe5$Y_2O_3$1 plate. Since other suitable conductor materials were found in this study, no further investigations to understand the deterioration were performed. The resistance of the joint made with the third nickel-based alloy (i.e., Inconel 617) remained constant during annealing and was significantly lower than that of joints made with steel wires having approximately the same diameter. Consequently, Inconel 617 is regarded as a very suitable conductor material for the present purpose.

The distribution of the main elements of a CrFe5$Y_2O_3$1/L-Ni 5/Inconel 617 joint in the bond area (Cr, Fe, Al, Ni, Co, Si) was determined by EDS (Fig. 4). The peak for molybdenum, another main constituent of Inconel 617, interfered with a peak from the gold coating (the samples have been coated for SEM and EDS analysis). Therefore, the molybdenum values have been left out of consideration. The joint was brazed with a 100 μm filler layer, and it was examined after annealing (1000°C in air for 1000 h). Little diffusion from the conductor and filler material, respectively, into the CrFe5$Y_2O_3$1 plate occurred. As a result, there is a sharp change in the element distribution at the CrFe5$Y_2O_3$1/L-Ni 5 interface. In contrast, no clear transition was observed between the filler layer and the Inconel 617 wire. The EDS spectra indicate that materials were homogeneously mixed, disregarding some peaks for Cr, Ni, Al, and Si very close to the CrFe5$Y_2O_3$1/L-Ni 5 interface. Dissolution of Inconel 617 in the liquid filler during brazing as well as diffusion during the long-term annealing caused the homogeneous mixing. Additionally, chromium was diffusing from the CrFe5$Y_2O_3$1 plate into the filler and wire material.

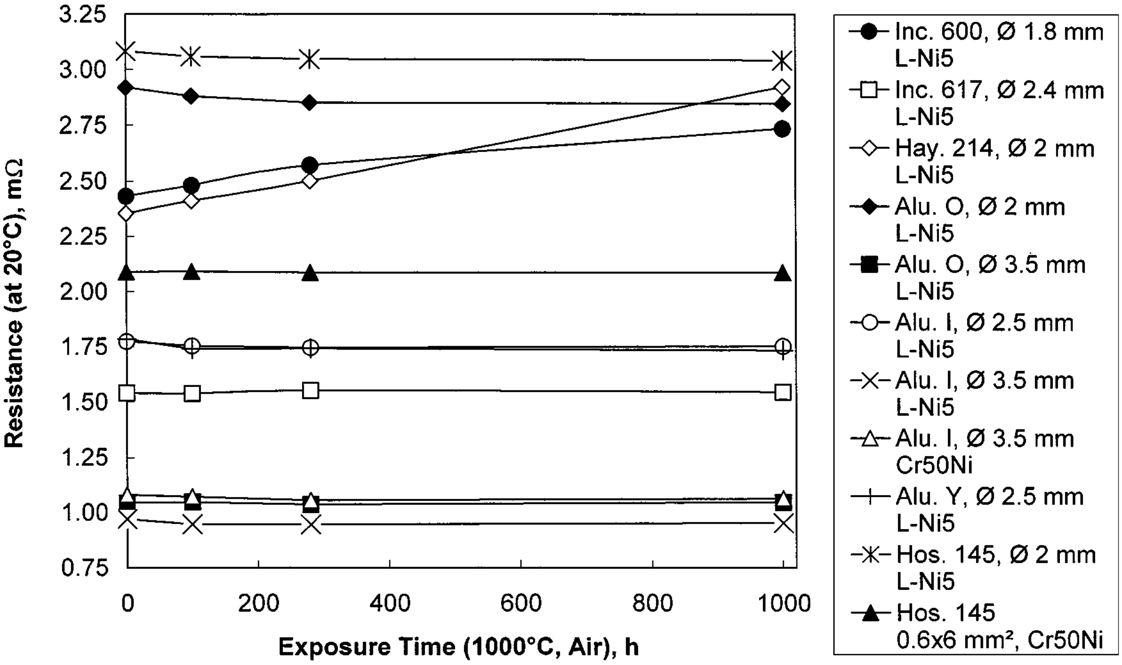

Figure 3 Electrical resistance of conductor/CrFe5$Y_2O_3$1 joints.

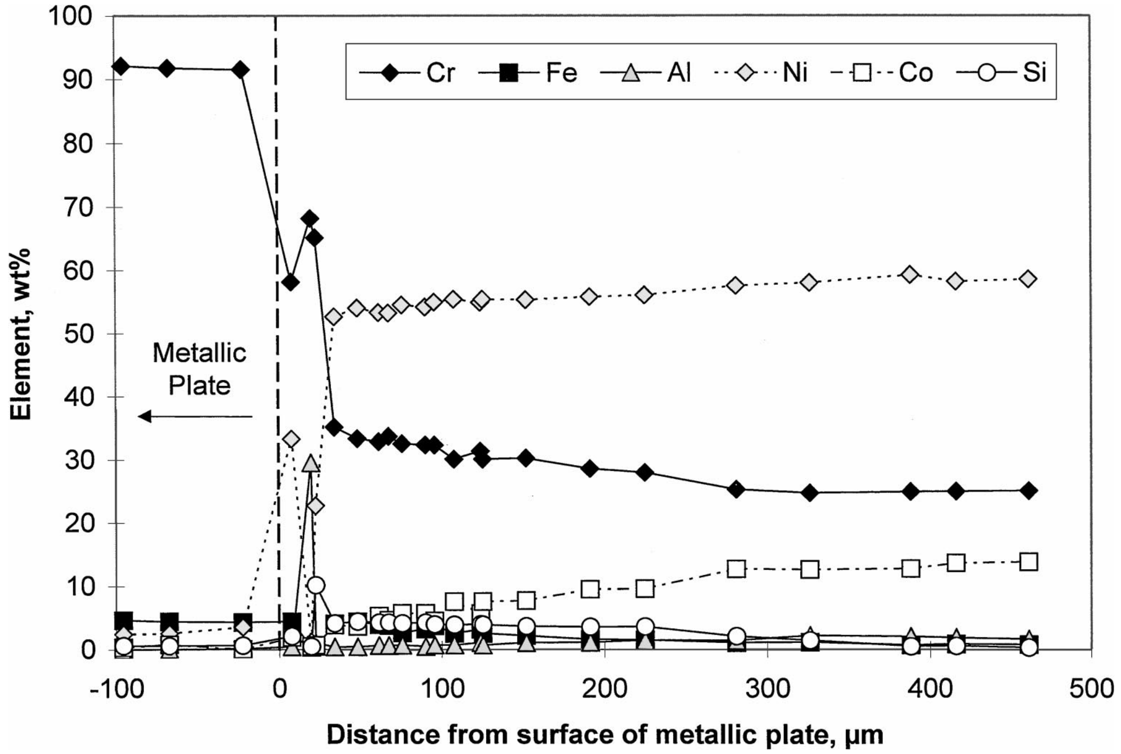

Figure 4 Element distribution for a CrFe5$Y_2O_3$1/Inconel 617 joint brazed with 100 µm L-Ni 5, after annealing (1000°C in air for 1000 h).

Hence, a good metallurgical bond between the filler and Inconel 617 was achieved by diffusion, while the bond between CrFe5$Y_2O_3$1 and the filler was significantly less strong. A few cracks parallel to this interface were observed by SEM. These cracks were induced either during brazing or during the preparation of the sample for SEM examination. They did not occur or grow substantially during the long-term annealing, since the resistance of the CrFe5$Y_2O_3$1/Inconel 617 joint was stable during annealing.

The resistances of all joints made with steel conductors were constant during annealing, even when a conductor in the form of thin strip (Hoskins 145) was used. Accordingly, these materials can be used as conductors for the present task. Given the costs of the materials tested, the alloy with the lowest aluminum amount should be used (Aluchrom I, corresponding to steel 1.4765 according to [5]). Furthermore, no additional alloying elements are required in order to enhance the oxidation resistance (such as Ti, Zr, and Y in the case of Aluchrom Y). Future work should focus on the question of whether steels having a lower aluminum content, which do not form a pure Al_2O_3 protecting layer, offer a high enough resistance against oxidation for the present purpose. Such materials (e.g., steel 1.4742 according to [5], or see [12] for examples) are more cost-effective and have a lower electrical resistance than the steels investigated in this study.

We conclude that L-Ni 5 is a more suitable filler alloy than Cr50Ni for manufacturing a low-resistance conductor/CrFe5$Y_2O_3$1 joint. This follows from the lower resistance of the CrFe5$Y_2O_3$1/Aluchrom I joint brazed with L-Ni 5(0.95 Ω) compared to the one manufactured with the same wire, but Cr50Ni as filler alloy (1.06 Ω). The higher amount of the better conducting γ-phase (Ni) compared to the α-phase (Cr) in L-Ni 5 can explain the effect. Furthermore, the brazing temperature required for L-Ni 5 (1200°C) is close to the one required for SCP6 (1140°C), which was found to be the most suitable filler alloy for brazing CrFe5$Y_2O_3$1 plates to $LaCrO_3$ plates [1]. Thus, both furnace brazing steps required to connect conducting wires to $LaCrO_3$ end plates in SOFC can be performed cost effectively in one furnace brazing step if these filler alloys are used.

4. Conclusions

The following conclusions were drawn from this study:

1. Conductors made of various aluminum-alloyed ferritic stainless steels can be brazed onto CrFe5$Y_2O_3$1 using L-Ni 5 or Cr50Ni as filler alloy.

2. The electrical resistances of such joints are constant during a long-term thermal exposure (1000°C in air for 1000 h). Additionally, the specific resistances of the steels themselves, measured close to the joint area, are stable. Hence, these materials are very promising as conductor materials.

3. Considering their specific electrical resistance, the alloy Aluchrom I should be used, since it has the lowest aluminum content and therefore the lowest specific resistance (137 $\mu\Omega \cdot$ cm at 20°C). Future work should reveal if ferritic stainless steels with even lower aluminum amounts (<4.5 wt%) can be used for the present task. Such alloys have a lower electrical resistance since they do not form a pure Al_2O_3 protecting layer, but this advantage goes along with the disadvantage of an inferior resistance against oxidation.

4. The nickel-based conducting wires (Inconel 600, Inconel 617, and Haynes 214) can be successfully brazed onto CrFe5$Y_2O_3$1 with L-Ni 5 as filler alloy.

5. The electrical resistance of the joint made with Inconel 617 wire was constant during the long-term thermal exposure, while joints made with Inconel 617 and Haynes 214 wires showed an increase in their resistance. Inconel 617 is, therefore, a suitable material for the conducting wire.

6. The specific resistance of Inconel 617 after annealing (127 $\mu\Omega \cdot$ cm at 20°C) is lower than that of the tested ferritic stainless steels. Thus, this alloy is considered to be the more suitable conductor material for the present task.

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