A NEW LOW COST MASS PRODUCTION ROUTE FOR METALLIC SOFC-INTERCONNECTORS

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ABSTRACT

Due to their unique material characteristics chromium based alloys (e.g. Cr-5Fe-1Y2O3) have successfully demonstrated their excellent suitability for the use as metallic interconnector material for planar high-temperature solid oxide fuel cells (SOFC). Beside the technical performance requirements for Cr-based interconnectors integrated in the Sulzer HEXIS system the achievement of the cost target is gaining importance, as these systems are approaching commercial application. For this reason, Plansee has investigated a new near-net shape (NNS) powder metallurgical (PM) production route providing a cost reduction potential for mass production of interconnectors by a factor of 10 compared to the current state-of-the-art process. The main advantage of the NNS-route is the elimination of extensive machining required for parts made from rolled sheet material or other semifinished prematerials. The PM-manufacture additionally offers flexibility in alloy and interconnector design. For the PM-manufacture a variety of Cr-based alloys were specifically modified and adapted to prealloying, NNS-pressing and sintering. After the investigation of the corrosion behaviour NNS-current collectors were manufactured with selected alloys and integrated in Sulzer HEXIS stack tests. The performance of NNS-current collectors was equivalent to those made of conventional (rolled) Cr-5Fe-1Y2O3-material.

INTRODUCTION

The development of Cr-based alloys (Ducrolloy, e.g. Cr-5Fe-1Y2O3) with excellent SOFC relevant material characteristics has contributed to a significant progress in leading planar high-temperature SOFC concepts during the last five years. Especially, the exceptional corrosion resistance against hot gases and the thermal expansion compatibility with the stabilized zirconia electrolyte offer a great potential for Cr-based alloys to be utilized as metallic SOFC interconnectors (1-3).

Due to the intrinsic difficulties of Cr-melt metallurgy (e.g. high melting point, high reactivity and high evaporation rate of molten chromium) powder metallurgical (PM) processing is utilized at Plansee for the production of Cr-based SOFC interconnectors (4).
The conventional PM-route basically consists of prealloying the raw materials (powders), consolidating the powder mixtures by pressing and sintering, and final machining of the component. The consolidation of the powder is conducted by conventional pressing and sintering, optionally followed by hot isostatic pressing (HIP) or by hot rolling. Before rolling or HIP the compacts have to be canned. Finally, mechanical and/or electrochemical machining lead to finished parts. A more detailed discription of the processing as well as the cost share of the manufacture steps is given in reference (4).

In this work main emphasis was laid on the reduction of the significant final machining cost share for the given Sulzer HEXIS current collector design by developing a new near-net shape (NNS) PM production route. Beside the material savings resulting from NNS-manufacture the cost share of final machining can be lowered drastically leading to a projected cost reduction potential of at least an order of magnitude in industrial scale production compared to the conventional state-of-the-art PM manufacturing process. Apart from the cost-effectiveness of the NNS-PM route and its capability of meeting the cost target for commercialization of Cr-based interconnectors, the flexibility in alloy and interconnector design as well as the highly isotropic component properties are additional benefits. With the new NNS PM manufacturing route the medium-term (year 2005) commercial cost target for Cr-based metallic interconnectors (MICs) in large quantities, which is in the range of 10 US$ / MIC, can be considered to be realized.

The present paper summarizes the pilot near-net shape manufacture of interconnectors made of modified Cr-based alloys and their performance evaluation in stack tests compared to conventional Cr-5Fe-1Y2O3 interconnectors.

EXPERIMENTAL

Material Selection Program

An extensive material selection program was conducted for the evaluation of suitable alloy powders for the manufacture of NNS-interconnectors. This program included the investigation of three different powder production routes resulting in three powder qualities, in the following designated as grades (I), (II) and (III). The main difference between these three grades is the powder prealloying and preparation process.

Additionally, the program included a comprehensive variation in alloy composition for all powder grades with the aim of optimizing the interaction between decisive criteria as pressability, the sintering activity (shrinkage behaviour) and the corrosion resistance. All alloys were Cr-5Fe based with modifications in the type and amount of dispersoid additions, in the Cr-powder quality (difference marked with „D“) and other alloying elements. Different Cr-powder qualities were considered due to the marked variations in costs, ranging from about 8 to 20 US$ / kg, and due to the influence of varying amounts of impurities which had to be evaluated. Ti was an additional alloying element in order to investigate its influence on the criteria mentioned above. The properties and advantages of oxide dispersion strengthened (ODS) Cr-based alloys are described in detail in references (1, 5, 6).
Table I summarizes the materials investigated. The Cr-5Fe alloy without any further additions was considered as a baseline.

Table I  Summary of investigated Cr-based alloys (compositions in wt.-%)

| Alloy composition          | Powder grades |
|----------------------------|---------------|
|                            |   (I)     |   (II)   |   (III)  |
| Cr-5Fe                     |   x       |   x      |     -    |
| Cr-5Fe-0.02Y2O3            |   x       |   x      |     -    |
| Cr-5Fe-0.5Y2O3             |   x       |   x      |     x    |
| Cr-5Fe-1Y2O3               |   x       |   x      |     x    |
| Cr-5Fe-0.3Ti-0.5Y2O3       |   x       |   x      |     x    |
| Cr-5Fe-0.3Ti-1Y2O3         |   x       |   x      |     x    |
| Cr-5Fe-0.5Y2O3 D           |   x       |   x      |     x    |
| Cr-5Fe-1Y2O3 D             |   x       |   x      |     x    |
| Cr-5Fe-0.3Ti-0.5Y2O3 D     |   x       |   x      |     x    |
| Cr-5Fe-0.3Ti-1Y2O3 D       |   x       |   x      |     x    |
| Cr-5Fe-0.5CeO2             |   x       |   x      |     x    |
| Cr-5Fe-1CeO2               |   x       |   x      |     x    |
| Cr-5Fe-0.3Ti-0.5CeO2       |   x       |   x      |     x    |
| Cr-5Fe-0.3Ti-1CeO2         |   x       |   x      |     x    |
| Cr-5Fe-0.5La2O3            |   x       |   x      |     x    |
| Cr-5Fe-1La2O3              |   x       |   x      |     x    |
| Cr-5Fe-0.3Ti-0.5La2O3      |   x       |   x      |     x    |
| Cr-5Fe-0.3Ti-1La2O3        |   x       |   x      |     x    |

...investigated  -...not investigated

**Sample Manufacture and Test conditions**

The evaluation of the different powder grades and compositions summarized in Table I was conducted with regard to powder metallurgical manufacturing criteria such as pressability, sintering behaviour, resulting density and the corrosion resistance as one main performance criterion in service.

For small scale tests plates with dimensions of Ø 30mm x 2.5mm were pressed on a 1000kN-laboratory press with a specific load of 5t/cm². The samples were subsequently sintered at temperatures > 0.6 Tm. The green densities as well as the resulting densities after sintering were determined geometrically. Corrosion tests were performed in laboratory air for 168h at 900°C. A selected number of samples was exposed to an additional corrosion test for 1008h at 1000°C. The corrosion behaviour was characterized by the mass change (weight gain) of the samples. The conventionally produced (pressed/sintered/ rolled) Cr-5Fe-1Y2O3 alloy was taken as a standard for comparison.
With a small number of promising alloys preliminary pressability and sintering tests were performed on Ø 48mm plates with a near-net shape nap-design on one side of the plates. Finally, the upscaling to the interconnector dimensions of Ø 120mm was conducted based on the experiences of the preliminary tests.

For Sulzer HEXIS stack tests about 100 NNS interconnectors, including three selected alloys, were produced on industrial facilities. The stack tests were evaluated after six weeks of stack operation and the performance of the NNS interconnectors was compared with conventional Cr-5Fe-1Y2O3 interconnectors.

RESULTS AND DISCUSSION

As a complete description of the results of all investigated alloys (see Table I) would go beyond the scope of this paper, emphasis will be laid in the following on the results of a selected number of alloys which are of enhanced interest for further development.

Pressability and Sintering tests

Figure 1 illustrates the results of pressability and sintering tests for a number of alloys representing the three different powder grades. Concerning the pressability of the different powder grades at a given specific load a significant increase in green density can be observed from grade (I) to grade (III), the values for grade (II) powders being in between. The differences are mainly caused by the specific morphologies and grains size distributions for each powder grade. Within a certain powder grade the green densities are at a comparable level and do not depend on the alloy composition.

Figure 1. Green density and sintering density of a selected number of Cr-based alloys.
As far as the sintering behaviour is concerned specific characteristics for the three powder grades can be derived from Figure 1. The grade (III) powders hardly exhibited any sintering activity resulting in negligible shrinkage. In contrast, some of the grade (I) and grade (II) powders sintered to densities exceeding 90% of the theoretical value. Additionally, specific alloy compositions within a given powder grade had a distinct influence on the sintering activity and consequently on the time and temperature response of the shrinkage behaviour.

Corrosion Tests

Based on the corrosion tests at 900°C for 168 hours in air and the pressability and sintering tests samples of the favoured alloys of each powder grade were exposed to an additional corrosion test at 1000°C for 1008 hours in air. The mass change of the samples was determined after periods of 168 hours respectively. The standard Cr-5Fe-1Y2O3 alloy taken from PM sheet material served as reference material. Figure 2 summarizes the results of the 1000°C corrosion tests.

Generally, the grade (I) and (II) alloys exhibit significantly lower mass gains compared to grade (III) alloys. This result can directly be related to the lower sintering densities of the grade (III) alloys. Concerning the alloy composition effects the CeO2-strengthened alloys show slightly higher corrosion rates than the Y2O3-containing alloys what may be due to the size and distribution of the dispersoids. However, despite of the excellent corrosion resistance of the investigated Cr-based alloys it should be mentioned that the interconnectors are additionally protected with adequate coatings during service (3, 7).

![Figure 2](image_url). Corrosion behaviour of a selected number of Cr-based alloys investigated. Test conditions: 1000°C, 1008 hours, laboratory air (samples were preoxidized at 900°C for 168 hours in laboratory air).
NNS Manufacturing Results

The NNS-manufacture was built up in several upscaling steps with regard to the size of the pressed plates. Figure 3a shows results of upscaling tests which were performed with grade (II) powders. The scanning electron microscope (SEM) micrograph in Figure 3b illustrates a geometrical detail of the near-net shape surface after sintering. The surface exhibits excellent contour characteristics comparable to mechanically machined components.

![Image](https://via.placeholder.com/150)

**Figure 3.** (a) Results of preliminary near-net shape manufacturing trials. (b) SEM micrograph of a contour detail after NNS pressing and sintering.

Based on the small scale manufacturing trials interconnectors were produced on industrial scale utilizing the NNS-manufacturing route causing a significant overall reduction of machining time and costs. Figure 4 shows some finalized Cr-based interconnectors (diameter 120mm) made of Cr-based alloys via the NNS PM manufacturing route.

![Image](https://via.placeholder.com/150)

**Figure 4.** Metallic interconnectors (Ø 120mm) made of Cr-based alloys via the near-net shape powder metallurgical production route on industrial facilities.
Stack Performance Tests

Several SOFC stack tests with NNS PM interconnectors were conducted at Sulzer HEXIS AG / CH. Figure 5 shows the performance of a test stack equipped with NNS PM current collectors. A more detailed description of the stack performance is given in reference (8) in the present proceedings.

Figure 5. Performance of a SOFC test stack equipped with NNS PM current collectors made of grade (II) Cr-based alloys.

SUMMARY AND OUTLOOK

An extensive number of Cr-based alloys with different grades was investigated with regard to their suitability for a new near-net shape powder metallurgical production route of SOFC interconnectors for the Sulzer HEXIS system. This production route has successfully demonstrated its viability and high potential for a significant cost reduction compared to the state-of-the-art manufacturing process and for the realization of the projected cost target for large scale commercialisation, which is around 10 US$ per interconnector.

Further activities will be focused on the specific optimization of Cr-based powders for the near-net shape manufacture and the installation of a highly automatized production line for interconnectors.
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