# Influence of powders on ionic conductivity of polycrystalline zirconias

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## Introduction

Reduction of the operating temperature of solid oxide fuel cells (SOFCs) from 1000 $^{o}$C to 600-800°C simplifies materials compatibility issues, and allows the consideration of metallic components which should reduce system cost. Yttria stabilized zirconia is the electrolyte of choice for SOFC systems. However, as the operating temperature is lowered, grain boundary resistivity can dominate the total resistivity of the YSZ film. In this work, the influence of powders on the conductivity of 8 mol % yttria-stabilized zirconia (8YSZ) and a higher conductive material of 9 mol % scandia stabilized-zirconia (9SSZ) polycrystals is investigated using powders synthesized by different methods and commercial ones.

## Experimental

8YSZ powders were synthesized using Pechini [1], co-precipitation, and glycine-nitrate (GN) [2] methods. In the GN method, glycine was added to the solution in glycine / nitrate (G/N) ratios of 0.5 and 1.0. The powders synthesized were calcined at 1000°C or 1100°C for 4 h and then attoritor milled using YSZ balls at the rotation speed of 550 rpm for 1 h (one powder was for 320 m). The calcined powders were used to analyze crystalline phases from XRD spectra. The milled powders and two commercial 8YSZ powders (PSZ-13.5Y-HW from Stanford Materials Co. and TZ-8Y from Tosoh Corp.) were uniaxially pressed and sintered at 1500°C (some were at 1600°C) for 4h to make pellets. The sintered pellets were applied gold blocking electrodes on both sides for the AC conductivity measurements. 9ScSZ powders and pellets were also synthesized by the Pechini method.

## Results and Discussion

XRD analyses indicate that the calcined 8YSZ GN powders have a small amount of the monoclinic phase depending on the G/N ratio. The conductivity of the 8YSZ polycrystals by AC impedance spectroscopy has a factor of two differences between the lowest and highest in Table 1. Cole-Cole plots in Fig. 1 show that 8YSZ polycrystals prepared from synthesized powders have comparable total grain boundary resistivity (represented by the semi-circles in low frequencies) to the grain interior resistivity (the semi-circles in high frequencies). The total grain boundary contribution to the resistivity is more than 10 % for some powders at the temperatures 600-800°C, as shown in Fig. 2. The total grain boundary resistivity is drastically reduced by increasing the sintering temperature or reducing the powder particle size. As a result, the conductivity is improved by c.a. 50 % (See the Pechini specimens in Table 1). Similar results are obtained for the 9SSZ polycrystals prepared from the Pechini powders. Consequently, it is suggested that the total grain boundary contribution to the conductivity becomes significant if the sintering condition or the powder particle size are not appropriate, for intermediate temperature SOFCs.

## Acknowledgment

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## References

1. Pechini, U.S. Pat. No. 3330697, July 11 (1967).
2. L.A. Chick, L.R. Pederson, G.D. Maupin, J.L. Bates, L.E. Thomas, and G.J. Exarhos, *Mater. Lett.*, **10**, (1-2), 6-12 (1990).

Table 1 Ionic conductivity of 8YSZ and 9SSZ polycrystals measured by a.c. impedance in air (S cm$^{-1}$).

| Temperature (°C) | 600 | 650 | 700 | 750 | 800 | 850 | d (g/cm3) |
|---|---|---|---|---|---|---|---|
| 8YSZ Pechini | 0.0020 | 0.0042 | 0.0081 | 0.0145 | 0.0242 | 0.0383 | 5.44 |
| 8YSZ co-precipitate | 0.0024 | 0.0048 | 0.0089 | 0.0152 | 0.0242 | 0.0364 | 5.02 |
| 8YSZ glycine-nitrate | 0.0035 | 0.0068 | 0.0134 | 0.0234 | 0.0385 | 0.0584 | 5.50 |
| 8YSZ Pechini sintered at 1600°C | 0.0030 | 0.0057 | 0.0124 | 0.0224 | 0.0365 | 0.0550 | 5.49 |
| 8YSZ Pechini doping 2 mol% $Al_2O_3$ | 0.0026 | 0.0053 | 0.0098 | 0.0171 | 0.0271 | 0.0407 | |
| 8YSZ Pechini milled for 320 m | 0.0029 | 0.0082 | 0.0149 | 0.0250 | 0.0394 | 0.0581 | 5.66 |
| 8YSZ Pechini milled for 320 m and sintered at 1600°C | 0.0031 | 0.0065 | 0.0126 | 0.0219 | 0.0349 | 0.0521 | 5.63 |
| PSZ-13.5Y-HW | 0.0061 | 0.0114 | 0.0205 | 0.0338 | 0.0505 | 0.0734 | 5.83 |
| TZ-8Y | 0.0038 | 0.0075 | 0.0141 | 0.0242 | 0.0380 | 0.0565 | 5.50 |
| | | | | | | | |
| 9ScSZ Pechini | 0.0064 | 0.0136 | 0.0258 | 0.0449 | 0.0723 | 0.1084 | 5.28 |
| 9ScSZ Pechini sintered at 1600°C | 0.0099 | 0.0204 | 0.0372 | 0.0620 | 0.0937 | 0.1336 | 5.10 |

The specimens were sintered at 1500°C for 4h, and the starting powders synthesized were milled for 1h unless notified. The commercial powders were not milled.

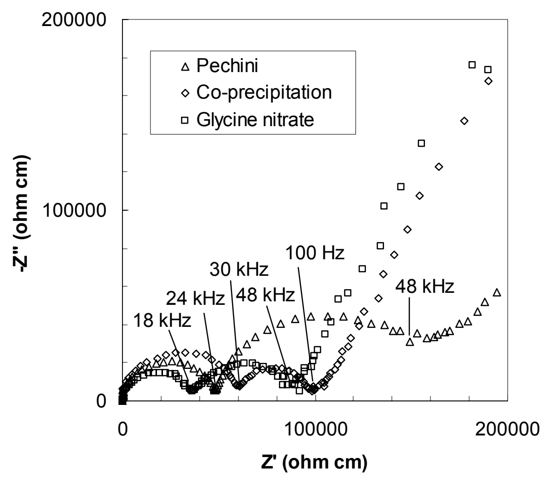


Fig. 1 Cole-Cole plots for 8YSZ polycrystals at 350°C in air.

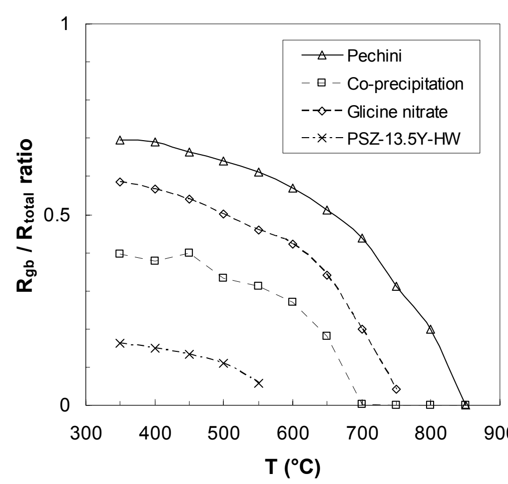


Fig. 2 Temperature dependence of the ratios of grain boundary / total resistivity for 8YSZ polycrystals.