Fractal Analysis of Surface Topography of Solid Oxide Fuel Cell Materials

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Abstract. *In this work, we investigate the hierarchical surface topography of solid oxide fuel cell electrolytes consisting of zirconia stabilized with 10% Sc_2O_3 and 1% CeO_2 (1Ce10ScSZ) synthesized at 1300-1400 °C and anodes of 60 wt% 1Ce10ScSZ and 40 wt% NiO synthesized at 1250 to 1550 °C. The fractal dimension of AFM images of the films was determined by the cube counting method, the triangulation method and by means of the PSD slope at large wavenumbers. RMS roughness and correlation lengths were derived from the fit to the model PSD.*

Key words: Atomic force microscopy, solid oxide fuel cell materials, fractal analysis, power spectrum density.

I. Introduction

By taking advantage of the scaling property of many physical systems reducing the amount of data required for structural characterization [1], hierarchical structures might be adequately characterized by atomic force microscopy (AFM) with resolution from about 10 nm to the micrometer range. The investigation of surface topography of porous materials by means of atomic force microscopy (providing already digitized images which are ready for computer analysis) provides thus a tool for the prediction of surface-sensitive properties such as the catalytic reactivity. Characteristic parameters like surface roughness, local fractal dimension, grain size and superstructures in the morphology are most preferably estimated by the Fourier power spectral density (PSD) [2].

In this work, we evaluate the hierarchical surface topography of solid oxide fuel cell electrolytes consisting of zirconia stabilized with 10% Sc_2O_3 and 1% CeO_2 (1Ce10ScSZ) synthesized at 1300-1400°C and anodes of 60 wt% 1Ce10ScSZ and 40 wt% NiO synthesized at 1250 to 1550 °C.

II. Theory

Surface topography is usually described in terms of surface roughness. Surface roughness is solely a function of height, that is, information about lateral topography is lost.

Power spectral density (PSD) is a technique that calculates power (roughness amplitude squared) as a function of spatial wavelengths k of the features that are contributing to the surface image. It is suitable for surfaces that this can be well approximated by a series of sine waves. Thin film surfaces are known to be self-affine, e.g., they are well modelled using anisotropic fractals and their topography is well described using the k-correlation model (also called ABC model) yielding in the one-dimensional case (assuming perfectly isotropic surfaces) [3]

$$PSD = \frac{A}{(1 + B^2 k^2)^{C/2}}. \quad (1)$$

The PSD identifies wavelengths of features that contribute to the surface structure. It describes how the power of the Fourier transform of the surface height is distributed with wavenumber of the spatial frequency k in reciprocal space. The parameter A describes the low-frequency limit of the spectrum. The parameter B sets the point of the transition between the smooth low-frequency and fractal high-frequency behaviour of the PSD. It defines a correlation length beyond which the surface height fluctuations are not correlated thus representing a mean grain size. The parameter C allows analysing the growth mechanism [1]. The integration of the one-dimensional PSD yields the RMS roughness σ of the surface

$$\sigma_{1-D}{}^2 = \int_{f_{\min}}^{f_{\max}} PSD_{1-D}(k) \cdot dk. \quad (2)$$

III. Experimental

Microstructure of the SOFC materials was investigated by AFM using an NTEGRA Therma scanning probe microscope (NT-MDT, Zelenograd, Russia). Tapping mode imaging was performed in air using silicon cantilevers (Type NSG01, NDT-MDT, Zelenograd, Russia). The two-dimensional AFM image was averaged over the profiles traced in fast scan direction using the Open Source Software Gwyddion 2.21 using Daniell (equal weight) window [4]. This allows reducing noise due to instrument drift appearing more pronounced in the slow scan direction. Moreover, the averaging over 256 scans is approximately equivalent to the integration of the two-dimensional PSD over the slow scan direction [5]. In order to increase data reliability, PSDs were recorded using scans of 20x20 μm^2 and 40x40μm^2. The resulting PSD was calculated as the geometrical average over all PSDs [6].

The fractal dimension of AFM images of the films was determined by the cube counting method, the triangulation method and by means of the PSD slope at large wavenumbers [7]. RMS roughness, correlation lengths and local fractal dimension were derived from the fit to the model PSD.

IV. Results and Discussion

Fig. 1 illustrates the PSDs of the 60 wt% 1Ce10ScSZ -

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40 wt% NiO samples in dependence on annealing temperature. According to eq. (1), the PSD will run (with some fluctuations) at small wavenumbers parallel to the *k* axis. However, some of the PSD curves display a maximum in this region. This feature should be attributed to the fact that the low-frequency PSD components are not very reproducible because not enough data points are available in the averaging procedure to obtain adequate statistics [8]. This maximum is less pronounced when data of different scan lengths is geometrically averaged as proposed in [6].

The related fractal dimensions and PSD parameters are compiled in tables 1 and 2.

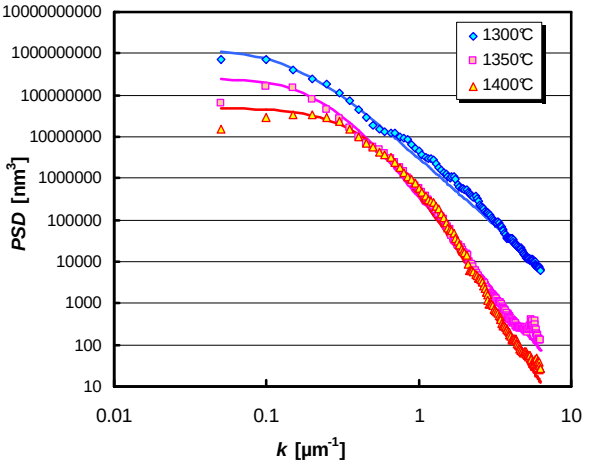

Fig. 1. Power spectral density of the surface profile calculated from AFM images of 60wt% 1Ce10ScSZ – 40 wt% NiO samples in dependence on annealing temperature. The fit to the ABC model is shown by solid lines.

Table 1. k-correlation approximation (ABC-model) of 60 wt% 1Ce10ScSZ-40 wt% NiO samples in dependence on annealing temperature.

Annealing temperature, °C	*A*, μm⁴	*B*, μm	*C*
1300	1.30	6.4	3.25
1350	0.28	4.0	4.7
1400	0.05	2.0	6.0

Table 2. Fractal dimensions and RMS roughness of 60wt% 1Ce10ScSZ-40wt% NiO samples in dependence on annealing temperature.

Annealing temperature, °C	Cube counting	Triangu-lation	PSD	RMS rough-ness, μm
1300	2.16	2.18	2.375	0.392
1350	2.16	2.22	(1.65)	0.180
1400	2.18	2.28	(1.0)	0.267

With regard to the finite numbers of pixels of an AFM image, the triangulation method provides a more reliable value of fractal dimension while cube counting underestimates the real value [7]. PSD does not allow deriving a reliable fractal dimension without applying before calculation of the PSD a more appropriate window function for the FFT algorithm in order to reduce finite pixel number effects and to minimize the arising from finite pixel numbers spectral leakage [9].

The fractal dimensions and ABC model parameters of the 1Ce10ScSZ samples are summarized in tables 3 and 4.

Table 3. k-correlation approximation (ABC-model) of 1Ce10ScSZ samples in dependence on annealing temperature.

Annealing temperature, °C	*A*, μm³	*B*, μm	*C*
1250	0.17	6.4	3.6
1300	0.80	7.2	3.5
1400	0.04	2.5	6
1450	0.11	4.8	3.6
1550	0.25	5.6	3.6

Table 4. Fractal dimensions and RMS roughness of 1Ce10ScSZ samples in dependence on annealing temperature.

Annealing temperature, °C	Cube counting	Triangu-lation	PSD	RMS rough-ness, μm
1250	2.23	2.3	2.20	0.318
1300	2.18	2.22	2.25	0.738
1400	2.23	2.32	(1.0)	0.253
1450	2.25	2.32	2.2	0.084
1550	2.2	2.25	2.2	0.433

V. Conclusions

In this work we have demonstrated that the local fractal dimension determined from AFM images is an appropriate and easy to use tool for the characterization of hierarchical surfaces of solid oxide fuel cell materials. With regard to the finite numbers of pixels of an AFM image, the triangulation method provides more reliable values of fractal dimension while the PSD allows analysis of structure evolution.

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References

[1] T. Viscek, *Fractal Growth Phenomena*, 2nd ed., World Scientific, Singapore, 1992.
[2] N.K. Sahoo, S. Thakur and R.B. Tokas, "Fractals and superstructures in gadolia thin film morphology: Influence of process variables on their characteristic parameters", *Thin Solid Films*, vol. 503, pp. 85-95, 2006.
[3] E.L. Church and P.Z. Takacz, "The optimal estimation of finish parameters", *Proc. SPIE*, vol. 1530, pp. 71-85, 1991.
[4] Gwyddion – data analysis software: http://gwyddion.net
[5] E. Marx, I.J. Malik, Y.E. Strausser, T. Bristow, N. Poduje and J.C. Stover, "Power spectral densities: A multiple technique study of different Si wafer substrates", *J. Vac. Sci. Technol. B*, vol. 20, no. 1, pp. 31-41, 2002.
[6] B.J. Ferré-Borrull, A. Duparré and E. Quesnel, "Roughness and Light Scattering of Ion-Beam-Sputtered Fluoride Coatings for 193 nm," *Applied Optics*, vol. 39, no. 31, pp. 5854-5864, 2000.
[7] C. Douketis, Z. Wang, T.L. Haslett and M. Moskovits, "Fractal character of cold-deposited silver films determined by low-temperature scanning tunneling microscopy", *Phys. Rev. B*, vol. 51, no. 16, pp. 11022-11031, 1995.
[8] S.J. Fang, S. Haplepete, W. Chen and C.R. Helms, "Analyzing atomic force microscopy images using spectral methods", *J. Appl. Phys*., vol. 82, no. 12, pp. 5891-5898, 1997.
[9] T. Itoh and N. Yamauchi, "Surface morphology characterization of pentacene thin film and its substrate with under-layers by power spectral density using fast Fourier transform algorithms", *Appl. Surf. Sci.*, vol. 253, pp. 6196–6202, 2007.