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Magnetron deposition of yttria-stabilised zirconia electrolyte for solid oxide fuel cells

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The aim of the article is to review the latest achievements in the field of magnetron deposition of thin-film yttria-stabilised zirconia (YSZ) electrolyte for solid oxide fuel cells (SOFC). The main attention is paid to the use of magnetron sputtering for formation of YSZ electrolyte up to 10 μm thick on the anode substrates of intermediate-temperature SOFCs operating at a temperature of $(600-800)^{\circ}$C. The influence of the types of power sources and such deposition parameters as substrate temperature, substrate bias voltage, post-annealing treatment, etc., as well as the morphology of the anode substrate surface on the microstructure and properties of the deposited electrolyte is analyzed. It is shown that the magnetron sputtering method, despite its relatively high cost and complexity, is applicable to large area SOFC cells and is competitive compared to traditional methods of electrolyte formation, such as tape casting, tape calendering, electrophoretic deposition and screen printing.

Keywords: SOFC, nanotubes, thin-film electrolyte, magnetron sputtering.

Introduction

Among the tasks being solved in the field of hydrogen energy, the key task is creation of highly efficient and cheap solid oxide fuel cells (SOFCs). The main structural components of a single SOFC-cell are porous electrodes (an anode and a cathode) and a solid gas-tight electrolyte located between them.

SOFC stack, operating at the high temperature, is composed of unit cells, sealing materials, and interconnectors. Depending on the operating temperature,

SOFCs are divided into three types: high-temperature T>800 ^oC, intermediate-temperature 600 ^oC <T<800 ^oC (IT-SOFC) and low-temperature T<600 ^oC (LT-SOFC). High operating temperatures of SOFC entail the need for expensive construction materials. In addition, at high temperatures, a mutual diffusion of the electrode and electrolyte materials arises causing formation of non-conducting compounds, and mechanical stresses appear due to the difference in the coefficients of thermal expansion (CTE) of the individual layers [1]. Therefore, it is very important to reduce the operating SOFC temperature to (600-750) ^oC maintaining a high specific power.

Currently, two approaches are used to solve this problem:

•reduction of the thickness of the electrolyte by forming it into a thin film on a porous electrode (an anode or a cathode) [2];

•application of new ceramic materials with high ionic conductivity at moderate temperatures [3].

The electrolyte in SOFC is a conductor of oxygen ions from the cathode side to the anode, and therefore it must have a high ionic conductivity. At the same time, the electrolyte must provide reliable separation of the anode and cathode gas space, and hence the electrolyte must be gas-tight. Yttrium stabilized zirconia (YSZ) is the most commonly used electrolyte material for SOFC while Ni/YSZ and $La_{0.8}Sr_{0.2}MnO_3$ (LSM) are used as anodes and cathodes, respectively [4].

It was clearly shown that a reduction in the thickness of the electrolyte from hundreds of micrometers to several micrometers allows reducing the operating temperature of SOFC from (900-1000) ^oC to (650-800) ^oC [5]. In order to reduce the operating temperatures to (400-650) ^oC, formation of electrolyte with the thickness of several hundred nanometers is needed [6]. Traditional well-studied methods of electrolyte formation are various powder technologies, such as slip casting [7], screen printing [8], electrophoretic deposition [9], plasma spraying [10] and others. Despite the relative simplicity and low cost of these methods, they are designed for creation of thick coatings (tens of micrometers and higher) and are not suitable for formation of thin films.

To form a thin-film electrolyte various methods are used, which can be divided into three types. The first type includes the methods of physical vapor deposition (magnetron sputtering, electron beam and pulsed laser deposition) [11-13]. The second type includes the methods of chemical vapor deposition (spray pyrolysis, atomic layer deposition, metal-organic chemical vapor deposition) [14-16]. The third type includes such colloidal methods as sol-gel process, spin-coating, dip-coating [17-19].

The first review of vacuum deposition techniques for thin-film YSZ electrolyte, focused on such methods as magnetron sputtering, laser ablation, and electrochemical vapor deposition (EVD), was made by Pederson et al. in 2006 [20]. At that time, the authors of the review noted that the high cost of the equipment and relatively low deposition rates presented challenges to the implementation of the vacuum deposition methods in large-scale SOFC manufacturing. These limitations of the vacuum methods have not been overcome yet. Although it should be noted that substantial progress has recently been made in scaling of

the magnetron deposition of thin YSZ electrolyte films onto large-area substrates. The advantages of this method are the possibility of independent regulation of the main parameters of the sputtering process, formation of uniform coatings with required characteristics, and the possibility of processing of large-area surfaces. Regulating the process parameters such as particle energy, degree of ionization, substrate temperature, deposition rate, gas pressure, discharge power and operating mode of the magnetron sputtering system makes it possible to produce dense or nanoporous coatings.

Deposition at relatively low temperatures (<500 ^oC), which are typical for magnetron sputtering, leads to the lower level of unwanted interfacial reactions between the electrolyte and electrodes than the level observed during high-temperatures intering (T>1000 ^oC). Therefore, the purpose of this paper is to review the applicability of magnetron sputtering to large-scale SOFC technology, mainly focusing on yttria-stabilized zirconia (YSZ) electrolyte.

Magnetron sputtering of YSZ electrolyte. Effect of deposition parameters on electrolyte characteristics

Magnetron sputtering is a method of physical vapor deposition that is widely used to deposit a large variety of thin films, including industrial applications. The latest review of achievements in magnetron sputtering technology, in particular, unbalanced magnetron sputtering, closed field unbalanced magnetron sputtering, and pulsed magnetron sputtering was published by Kelly and his co-workers in 2000 [21]. Over the past years, the greatest changes have occurred in the magnetron power supplies. Previously, the most frequently used were DC, unipolar pulsed, bipolar pulsed [22] and dual magnetron sputtering systems. Now, high power impulse magnetron sputtering (HiPIMS) [23] and deep oscillation magnetron sputtering (DOMS) [24] are becoming increasingly popular. The latter methods enable scientists to make the reactive magnetron sputtering process more stable and repeatable, as well as to obtain films with a higher density.

A large number of investigations have been devoted to the study of the influence of various deposition parameters on the characteristics of YSZ electrolyte films. Parameters that have a significant effect on the structure of the electrolyte include oxygen partial pressure, substrate temperature, substrate bias voltage, post-annealing, and others.

YSZ films are usually deposited by sputtering of Zr-Y alloyed targets (84 at. % Zr and 16 at. % Y) in an argon and oxygen atmosphere. In this case, the substrate is preheated to a temperature (400-500) ^oC in order to increase the mobility of atoms during deposition of the film and to form a denser film. This also reduces the probability of electrolyte cracking during heating/cooling of the fuel cell due to the difference in thermal expansion coefficients of the anode and electrolyte.

One of the specific features of the magnetron sputtering method is formation of the columnar structure of the YSZ electrolyte. For the SOFC electrolyte, the columnar structure is undesirable, because the columnar grains provide an easy, low-energy path for crack propagation [25]. Therefore, the columnar structure of

the thin-film must be altered.

Kek et al. [26] showed that the YSZ films prepared by reactive DC-sputtering have different microstructures (from columnar growth to polycrystalline structure) depending on the partial pressure of oxygen in the atmosphere.YSZ films with a thickness of (4-8.5) mm were prepared on porous NiO-YSZ substrates. Denser and more compact YSZ films were fabricated by post-oxidation of sputtered films in air. No influence of the grain boundaries on the electrical conductivity of deposited films was observed.

Hobein et al. [5] deposited YSZ electrolytes with a thickness of (1.2-8.5) μm on the anode substrates by DC reactive sputtering. In this work, the temperature of the substrate was regulated in the range (500-700) ^oC, and the partial pressure of oxygen varied from 0.65×10^{-4} to 6.0×10^{-4} mbar. In all cases, along with the cubic phase, a tetragonal phase was formed in the film. Moreover, the percentage of the latter increased with a decrease in the partial pressure of oxygen. However, after annealing in air at $1100\,^oC$ for 3 hours the tetragonal phase completely transferred into the cubic phase. In this case, annealing caused a (2-10)-fold increase in the gas density of electrolytes.

In [27], a YSZ electrolyte with a thickness of 10 μm was deposited on the $(3.2 \times 3.2)\,\text{cm}^2$ anodes. During the deposition process, the substrates were heated to a temperature less than $200\,^oC$. The film had a fine fibrous structure in the cross-sectional view (the size of the fibrous structures is a few hundred nanometers in the lateral sizes), see Figure 1a. To suppress the columnar microstructure in the deposited films, the authors used high-temperature annealing in air in the temperature range (1150-1350) ^oC. The best result was achieved by annealing at $1250\,^oC$ for 4 hours (Figure 1b). It was found that the surface morphology of the anode substrate had a very important effect on the quality of the sputtered films. Application of the anode functional layer makes the anode surface smooth, which leads to the formation of a denser electrolyte film. The maximum specific SOFC power with annealed YSZ electrolyte (10-μm-thick) and LSM/YSZ cathode at $750\,^oC$ was 700 mW/cm^2.

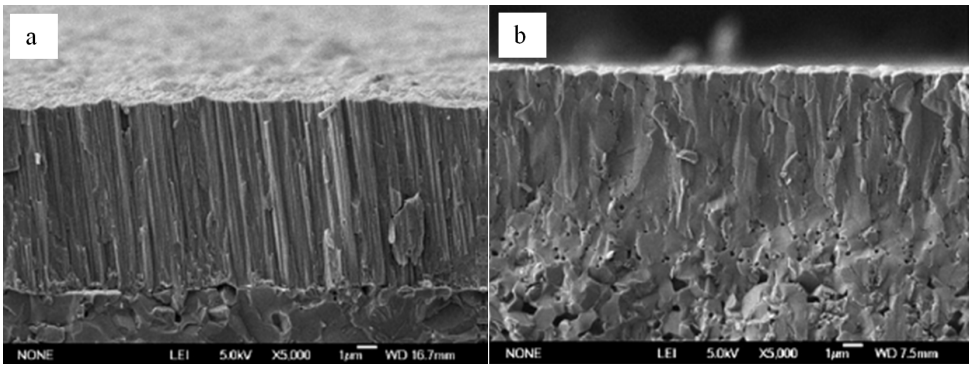

Figure 1. Microstructure of a YSZ electrolyte film deposited by magnetron sputtering (a) and filmannealed for 4 h at 1250°C (b) [27].

To suppress the columnar structure of the electrolyte film during its deposition, it is necessary to provide additional energy impact on the substrate. It can be provided by applying a negative bias voltage to the substrate. Positively charged ions of the inert gas (Ar) are accelerated towards the substrate and bombard

its surface. By varying the amplitude of the bias voltage of the substrate, it is possible to regulate the energy of ions.

Ion bombardment causes a number of effects, such as increase in atom mobility, densification of the film with a reduction in the microporosity, change in crystal size and shape, desorption of gases [28]. The reactive gas (oxygen) is activated and the reactivity of the gas molecule/deposited atom system is enhanced. In this case, at low ion energies, the bombardment effect will be hardly noticeable. On the other hand, too high ion energy can lead to radiation defects in the film and its re-sputtering, which will reduce the quality of the film and the rate of its growth.

The effect of applying the bias voltage to the substrate is demonstrated in [29], where 1-μm-thick YSZ electrolyte was deposited onto (5×5) cm^2 anodes by the reactive DC magnetron sputtering process. Before this procedure, a 15-μm-thick functional layer was formed on the anodes by vacuum slip casting. Due to this layer, it was possible to form a gas-tight electrolyte with a thickness of only 1 μm. During deposition, the substrate temperature was maintained at $800\,^oC$ and RF-bias with power varied from 0 to 0.5 W/cm^2 was applied to the substrate. Without a bias voltage, even at a substrate temperature of $800\,^oC$, the film had a columnar microstructure (Figure 2, a). By optimizing the specific power of the bias voltage, the authors managed to form a dense electrolyte without a columnar structure (Figure 2b). For SOFC with a 1-μm-thick YSZ electrolyte, a $Ce_{0.8}Gd_{0.2}O_{1.90}$ (GDC) barrier layer and a $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$ (LSCF) cathode, a power density of ≈ 1200 mW/cm^2 was obtained at a voltage of 0.7 V and a temperature of $755\,^oC$. This effect was achieved due to reduction of the area-specific resistance of the electrolyte layer.

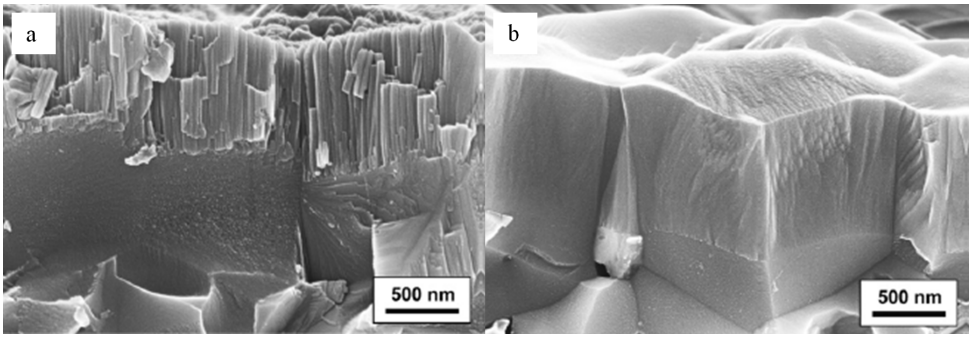

Figure 2. Microstructure of a YSZ electrolyte film deposited by magnetron sputtering without the substrate bias (a) and with the RF-bias of the substrate by the power of 0.3 W/cm^2 (b) [29].

Coddet et al. showed that the combination of DC-pulsed magnetron sputtering at low oxygen flow rates with high-frequency bias on the substrate makes it possible to obtain high-quality dense YSZ films of about 6.2 μm in thickness [30]. The deposition rate of 2.6 $\mu m/h$ was obtained by sputtering of two (Zr-Y) cathodes. In this case, the oxygen flow rate was selected so that there was no need in subsequent annealing of the YSZ film. The power density of SOFC with lanthanum cobaltite based perovskite (LSC) cathode produced in this work was 560 mW/cm^2 at a voltage of 0.7 V and a temperature of $800\,^oC$. The authors note that their results are close to the characteristics of the commercial cell ASC-800 (SOFC Power), tested under similar conditions. In addition, the paper

compares the characteristics of SOFC cells with the YSZ electrolyte, manufactured by various methods. Some of these data are given in Table 1.

Table 1.
Comparison of the characteristics of SOFC cells with the YSZ electrolyte, manufactured by various methods [30]. The data were obtained at a temperature of $800\,^oC$ and a voltage of 0.7 V.

Manufacturing process of the electrolyte	Electrolyte thickness (μm)	Power density (mW/cm^2)	Reference
Colloidal coating	~ 10	$\sim$1000	[1]
Tape casting	8$\pm$2	680	ASC-800 SOFC Power
Electron beam PVD	8	675	[31]
Magnetron sputtering	6.2$\pm$0.3	560	[30]
Thermal spraying	30	410	[32]
Electrophoretic deposition	17$\pm$1	282	[33]
Co-sintering	15	260	[34]
Electron beam PVD	12	115	[35]

The possibility of formation of 2-μm-thick YSZ electrolytes on NiO/YSZ anodes with an area of (13×13) cm^2 was first shown in [36]. The substrate temperature during the sputtering process was maintained at $500\,^oC$. The effect of a bias voltage of 350 kHz on the microstructure of electrolytes was studied. It was noted that it is necessary to avoid point electrical contacts between the anode substrate and the substrate holder through which strong currents can flow during the sputtering process, which in combination with a high temperature can lead to electrochemical blackening of YSZ. However, the influence of the latter on the YSZ characteristics was not studied in the work. Unfortunately, the electrochemical characteristics of SOFC with a thin-film electrolyte have not been studied either.

Most often to deposit YSZ electrolyte, sputtering in DC-pulsed mode is used. However, in the works of Sonderby et al. [37, 38], for depositing the YSZ electrolyte on NiO/YSZ anodes, high power impulse magnetron sputtering was used. HiPIMS utilizes extremely high power densities of the order of kW/cm^2 in short pulses of tens of microseconds at low duty cycle (on/off time ratio) of <10%. The main HIPIMS features are a high degree of ionization of the sputtered metal, high plasma concentration (up to 10^{13} cm^{-3}) [39] and a high rate of molecular gas dissociation which provide high density of deposited films. The use of HiPIMS for the deposition of metal and oxide coatings for various applications, as a rule, improves their characteristics.

Sonderby et al. [37] compared the microstructure of YSZ electrolytes deposited on (5×5) cm^2 anode substrates by the DC-pulsed magnetron sputtering and HiPIMS methods at a bias voltage in the range of 0 to -75 V. It was concluded that in the case of DC-pulsed magnetron sputtering, an increase in the bias voltage

does not lead to complete suppression of the columnar structure of the electrolyte, whereas at high power impulse magnetron sputtering with a bias voltage of -50 V the dense electrolyte films without a columnar structure are deposited (Figure 3). The electrochemical characteristics of SOFC have not been studied in this work.

In their next paper, Sonderby and co-authors [38] optimized the parameters of high power reactive magnetron sputtering and the magnitude of the bias voltage. As a result, YSZ electrolytes up to 3 μm thick are deposited on the anodes with an area of (13×13) cm^2. Heating of the substrates to 300 ^oC before sputtering is used to degasify porous anodes. With a specific impulse power of 0.6 kW/cm^2 and a bias voltage of -180 V, the deposition rate of the YSZ electrolyte is 0.23 $\mu m/h$. Electrochemical characteristics of SOFC were not studied.

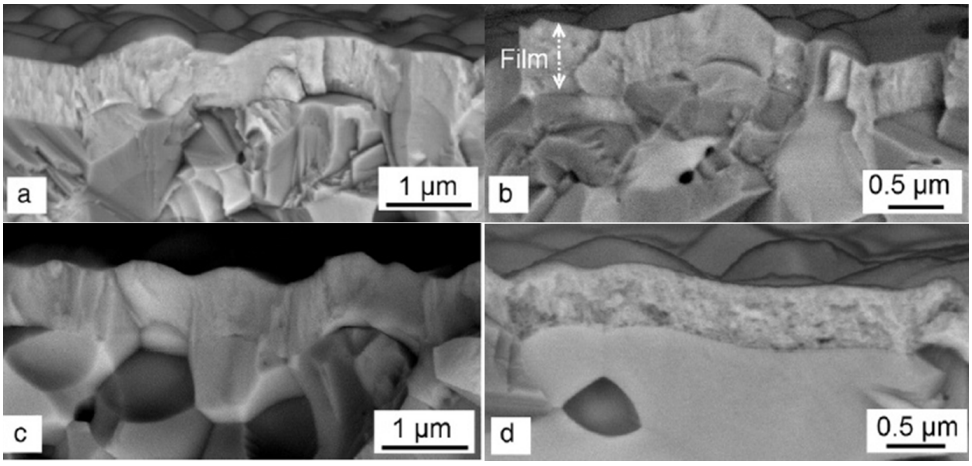

Figure 3. Cross section SEM images of YSZ films deposited by HiPIMS at different substrate biases: (a) floating potential, (b) -25 V, (c) -50 V, (d) -75 V [37].

Effect of substrate on electrolyte characteristics

As already mentioned, the microstructure and properties of the deposited YSZ electrolyte are affected by the quality of the substrate surface. In anode-supported SOFC, a thin gas-tight electrolyte layer is formed on a porous substrate. Therefore, the minimum thickness of the electrolyte must be at least 3 times greater than the maximum pore size on the surface of the substrate. Thus, the pore size on the surface of the substrate should be in the range from hundreds of nanometers to (1-2) micrometers. For this, on the surface of the Ni/YSZ anode substrates with a thickness of hundreds of micrometers, anode functional layers with a fine-pore structure are formed [29, 40].

For example, in [29] it was possible to form a dense YSZ layer of electrolyte with a thickness of about 1 μm only due to the preliminary deposition of the fine-pored functional layer on the anode surface (Figure 4).

The microstructure of the cross-section of the fuel cell shows that the pore size of the functional layer does not exceed 200 nm, which is almost five times smaller than the size of the pores of the main large-pored anode.

Wanzenberg et al. proposed the following method for modifying a porous anode substrate [40]. A fine-pored functional layer was deposited on the anode

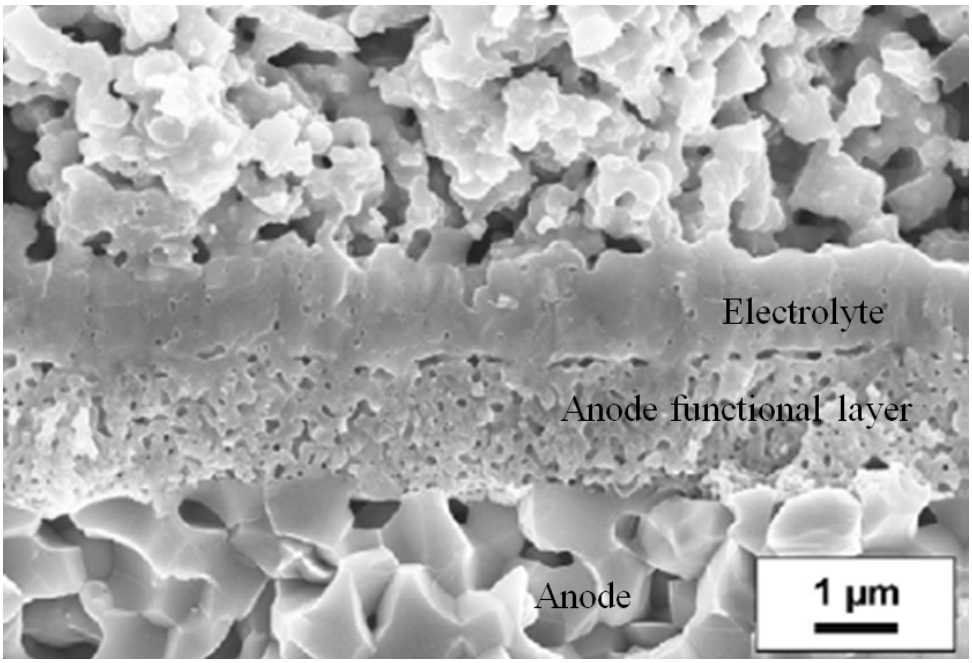

Figure 4. SEM polished cross-section of the $\approx 1\ \mu m$ thick electrolyte and the diffusion barrier layer (on top) [29].

substrate with a porosity of 45% (before NiO reduction) using a modified slip casting process. This layer was made of the same material but with a finer microstructure. The thickness of this layer was about 5 μm and the porosity was 15% to 20% after calcinations at $1000\,^{o}C$. The authors studied the influence of the sintering temperature (from 1300 to $1400\,^{o}C$) of the functional layer on changes in its microstructure, and the effect of this layer on the gas-tightness of the deposited YSZ films. Electrolyte films of thickness (5-8) μm were subjected to annealing after deposition to saturate them with oxygen and increase their density. It was shown that the porosity of the functional layer of the anode substrate decreases with increasing sintering temperature (Figure 5). A dense functional layer was obtained at a sintering temperature of $1400\,^{o}C$ (porosity was about 2%).

After NiO reduction, porosity of this functional layer increased to 16%, which ensures sufficient fuel diffusion to the three-phase boundary. In this case, the most gas-tight YSZ coating was obtained on the anode with a functional layer sintered at $1320\,^{o}C$ (the film was annealed at a temperature of $1360\,^{o}C$).

It was also shown that the higher the annealing temperature of the electrolyte film compared to the sintering temperature of the anode functional layer, the greater the probability of defects appearing in the film in the form of bulges and film peeling from the substrate. The authors suppose that such defects are formed because of a stronger shrinkage of the anode substrate during the repeated heat treatment (i.e., during annealing of YSZ films).

Specialists of the Institute of High Current Electronics of the SB RAS (Russia) proposed to use pulsed electron beam treatment (EBT) as a method of modification of the porous structure in anode substrates [41]. This treatment can be carried out in a single vacuum cycle with deposition of an electrolyte film. Figure 6 shows changes in the microstructure of the anode substrate after an electron beam treatment at different electron beam energy densities (E_s). The initial anode substrate (Figure 6a) fabricated using the tape casting method had a

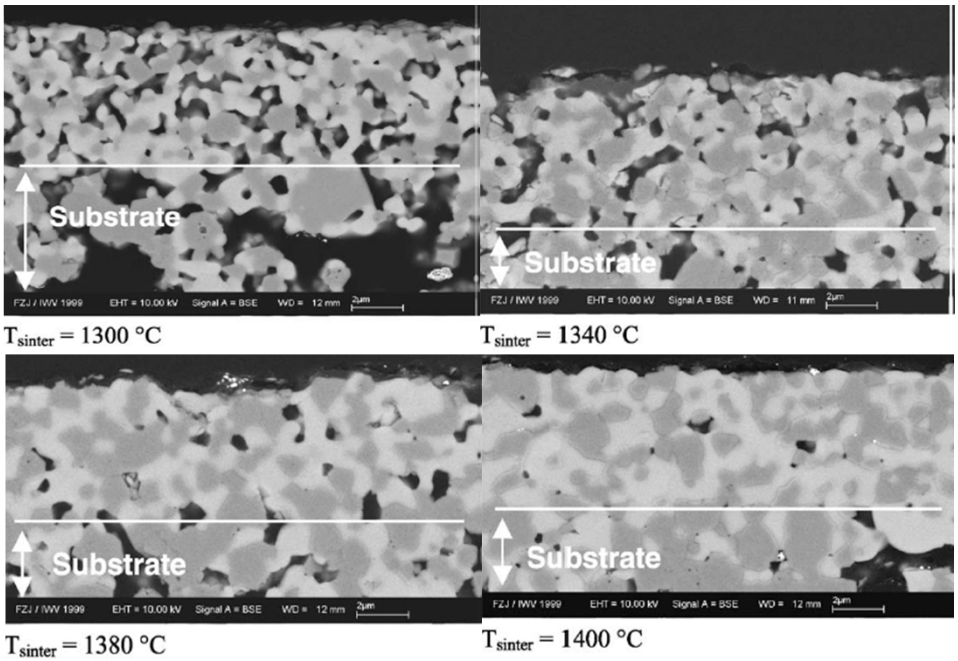

Figure 5. SEM pictures of the anode functional coating sintered at different temperatures (in backscattering mode; black: pores, dark grey: 8YSZ, light grey: NiO) [40].

porous structure with a coarse surface. The anode surface after the electron beam treatment is much denser and smoother than the original anode surface. Such treatment leads to melting of the substrate surface layer down to a depth of about (1-1.5) μm. In addition, it was shown that the electron beam treatment of the porous anode substrate with a thin YSZ sublayer (thickness of 1-2 μm) facilitates further deposition of a dense film of electrolyte without a columnar structure. SOFC with a 4-μm-thick YSZ electrolyte, deposited by magnetron sputtering in combination with electron beam treatment, and the LSM cathode demonstrated a maximum power density of 600 mW/cm^2 at a temperature 800 ^oC [42].

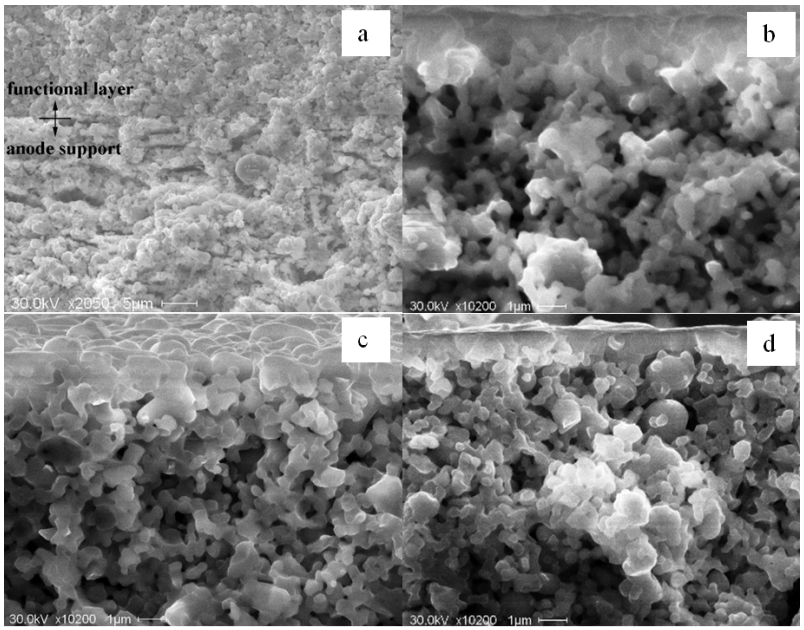

Figure 6. Cross-sectional SEM images of the anode substrate: (a) initial; (b), (c) and (d) the anode substrate modified by EBT at energy density E_s =0.8, 2.5 and 4.5 J/cm^2 (number of pulses N=3), respectively [41].

It was shown that the electron beam treatment allows us to abandon the stage of high-temperature annealing of the electrolyte after its deposition, which, as shown above, is often used to increase the density of films deposited by magnetron sputtering.

Scaling of the technology of magnetron deposition of YSZ electrolyte

Any technology of electrolyte deposition, which claims to be of industrial use, must provide formation of a uniform gas-tight electrolyte layer on large-area substrates. Practical size of anode-supported solid oxide fuel cells is equal to about (12×12) cm^2. For example, in commercial stacks E1000 (Elcogen AS, Estonia) of power of 1 kW and SOFCMAN-A-Stack-304 (Ningbo SOFCMAN Energy Technology Co., China) of power of 2 kW, (12×12) cm^2 fuel cells are used.

As mentioned above, S?nderby et al. [36] demonstrated the possibility of forming YSZ electrolytes with a thickness on NiO/YSZ anodes of (13×13) cm^2 area. However, a whole fuel cell was not produced and studied in this work.

Practical-size anode-supported solid oxide fuel cells with magnetron sputtered electrolyte were first studied in [43]. In this work a planar SOFC was fabricated using a commercial NiO/YSZ anode support, an YSZ/GDC thin-film electrolyte, and a composite cathode of $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$ /$Gd_{0.1}Ce_{0.9}O_{1.95}$ (LSCF/GDC). The cross-sectional microstructure of a single SOFC cell is shown in Figure 7a; the appearance of the cell is shown in Figure 7b.

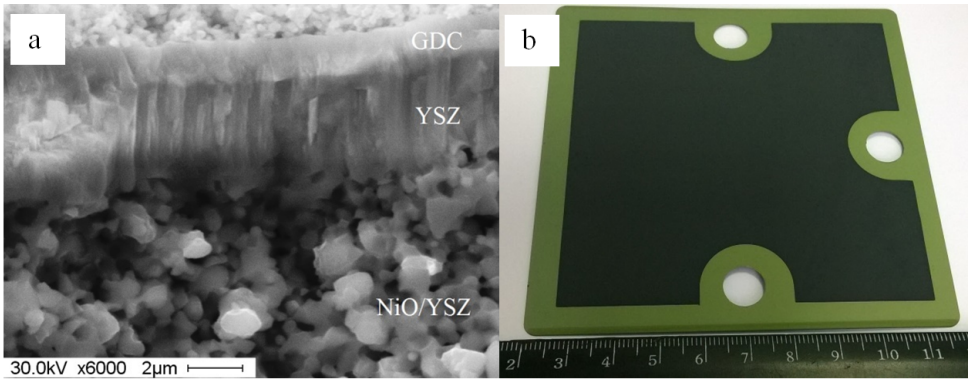

Figure 7. Cross-sectional SEM image (a) and photograph [43] (b) of the (10×10) cm^2 anode-supported single cell with magnetron sputtered electrolyte.

YSZ/GDC bilayer electrolyte was deposited by reactive medium-frequency dual magnetron sputtering technique. Eight substrates (10×10) cm^2 were simultaneously mounted on the rotating drum. TwoZr-Y (86:14 at. %) and two Ce-Gd (90:10 at. %) targets of size (8.3×26.9) cm^2 were sputtered in an Ar/O_2 mixture atmosphere. The current-voltage characteristic of a (10×10) cm^2 anode-supported single cell with magnetron sputtered electrolyte is shown in Figure 8. The OCV obtained for the single cell at $750\,^oC$ was 1.04 V and remained stable at this value during the testing process. This indicates that the YSZ/GDC electrolyte

prepared by magnetron sputtering was dense enough. The power and power density of the present cell are 12 W and 200 mW/cm^2 at 0.7 V, respectively. This result is comparable to the one that was obtained by Zhou et al. [44], in which the (10×10) cm^2 cell's electrolyte, 6.75 μm thick, was fabricated by aqueous-based tape casting, and the peak power density was 265.8 mW/cm^2 at $800\,^oC$.

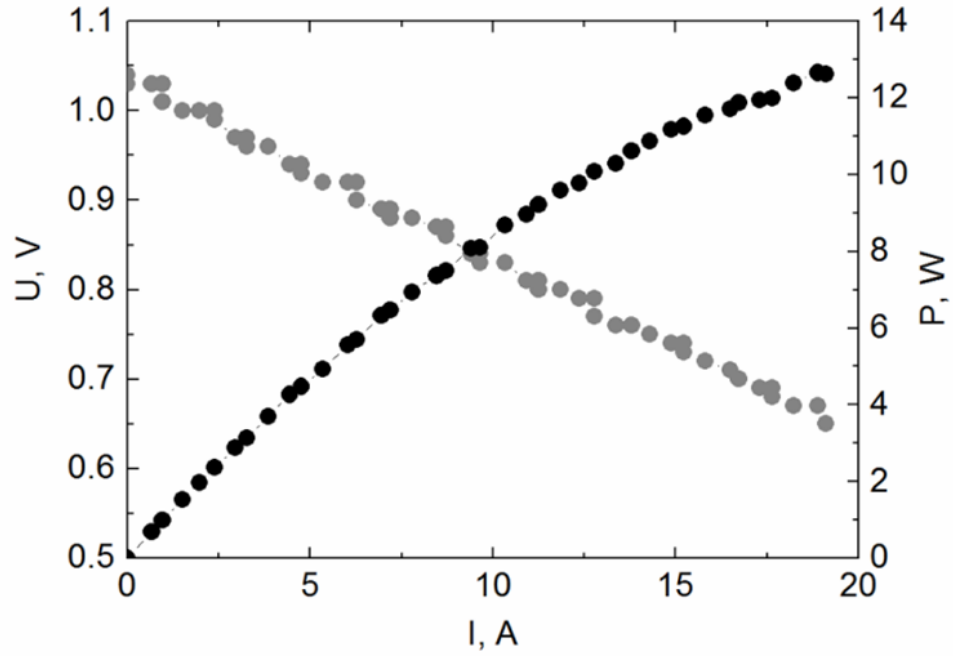

Figure 8. The current–voltage characteristic of a (10×10) cm^2 anode-supported single cell with magnetron sputtered electrolyte [43].

The obtained values of the cell power density (10×10) cm^2 are quite different from the power density of cells with a diameter of 20 mm (button cell) manufactured using the same technology (1000 mW/cm^2 at $750\,^oC$ under voltage of 0.7 V) [45]. This is caused by the fact that the testing setup for a planar cell with a large area is quite different from that for a button-sized cell. Stainless steel interconnectors (bipolar plates) used in a stack cause a significant contact resistance that leads to a decrease in the power density [46, 47]. Therefore, further studies should be aimed at reducing the contact resistance in SOFC stacks by optimizing the construction of bipolar plates and the composition of the cathode contact layer providing a minimum resistance between the bipolar plates and the SOFC cathode.

Estimations have shown that the cost of depositing a YSZ/GDC thin film electrolyte on a vacuum coater with a capacity of 70.000 (10×10) cm^2 fuel cells per year will be $\approx$3.5 US dollars per item. If we compare the cost of depositing of a thin-film electrolyte with the cost of commercial SOFCs presented on the well-known fuelcellmaterials.com website, it becomes obvious that the magnetron sputtering technology has a great potential in the SOFC market.

Conclusion

This review surveys capabilities of magnetron sputtering technique as a method used to prepare electrolyte for SOFCs and its scalability to a manufacturing scale. Although this method has disadvantages of vacuum film deposition methods, such as relatively high equipment cost and complexity, relatively low deposition rates ($\approx 5\ \mu m/h$) compared to traditional processing methods [20], the above results show competitiveness of the proposed technique. It allows us to produce dense, crack-free films with a wide range of compositions on dense and porous substrates at a relatively low substrate temperature.

This literature review showed that for the deposition of a thin gas-tight YSZ electrolyte on porous anode substrates by the magnetron sputtering method, one should take into account the following factors:

• It is necessary to preheat the substrates to a temperature of (300-500) ^oC to desorb gases from their porous structure;

• The columnar structure of the electrolyte must be suppressed in terms of its mechanical strength, as the columnar grains provide an easy, low-energy path for crack propagation;

• Surface morphology of the anode substrate has a very important effect on the quality of the sputtered YSZ films and it is necessary to form finely porous anode functional layers;

• Denser and more compact YSZ films are fabricated by post-oxidation of sputtered films in air;

• Use of substrate bias allows the change of the layer growth morphology from columnar to a denser structure.

Acknowledgments

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