Deposition of dense nanocrystalline Sm0.2Ce0.8O1.9 thin films by pulsed laser deposition

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Deposition of Dense Nanocrystalline $Sm_{0.2}Ce_{0.8}O_{1.9}$ Thin Films by Pulsed Laser Deposition

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Samarium doped ceria (SDC) of composition $Sm_{0.20}Ce_{0.80}O_{1.90}$ is a promising electrolyte material for intermediate temperature solid oxide fuel cells (SOFCs) [1,2]. Compared to yttria stabilized zirconia (YSZ) electrolyte, it has high ionic conductivity at temperatures below $T = 700$ °C and may even be operated as low as $T = 400$°C if hydrogen is used as the fuel [3]. Moreover, it has been reported that SDC electrolytes can successfully be used in single chamber fuel cell applications using air/hydrocarbon feed mixtures [4,5]. One of the limitations of the SDC electrolyte is its chemical stability under reducing environments, where electronic charge carriers may be formed from the partial reduction of Ce^{4+} to Ce^{3+} [1,2]. However, at lower operation temperatures the reduction of doped cerias becomes less of a problem [6,7], and it has been reported that above 500°C this only becomes significant when the SOFC is operated under open circuit conditions or at low current densities [8].

One approach to further decreasing the operation temperatures while maintaining the power densities of SOFCs is to reduce the ohmic loss by employing a thinner electrolyte layer [9,10]. Very thin electrolyte layers ($t \leq 10\mu m$) may be used if either the anode, cathode or the interconnect materials is employed as the structural support. It is for this reason that thin film deposition methods including pulsed laser deposition (PLD) are especially promising [9]. Recently, PLD has been used to deposit thin dense SDC layers onto cathode substrates to fabricate bilayer oxide membranes [11,12].

In this paper we examine the role of deposition temperature on film structure and morphology for depositing thin SDC films of composition $Sm_{0.2}Ce_{0.8}O_{1.9}$ onto model single crystal sapphire substrates. The goal is to establish the deposition conditions for which dense high quality SDC films of controlled nanostructure may be grown by PLD onto sapphire and then extend this to porous anode substrates for SOFC applications.

The films were deposited at deposition temperature between $200 \leq T \leq 700$°C under a reactive oxygen atmosphere of $p[O_2] = 100$ mTorr and varied in thickness from $500 \leq t \leq 1000$ nm. X-ray diffraction showed that the SDC films were all single phase and could be indexed to the cubic CeO_2 structure. The deposition temperature was shown to play a key role in determining the average crystallite size, preferential orientation and morphology of the as-deposited films.

The films deposited at lower temperatures ($200 \leq T \leq 300$°C) exhibited small average crystallite sizes ($20 \leq d \leq 40$ nm) but densified upon heat treatment at 500 °C forming microcracks. Films deposited at higher temperature, $400 \leq T \leq 700$°C showed much greater average crystallite size, $100 \leq d \leq 300$ nm and were often highly textured. SEM and AFM measurements of the film also confirmed the critical role of deposition temperature on film morphology. Electrical testing of the films between $200 \leq T \leq 480$°C showed that the films exhibited DC conductivities of $0.004 \leq \sigma \leq 0.01$ S/cm at T = 480°C with activation energies of conduction between $0.7 \leq Ea \leq 0.8$ eV.

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Figures

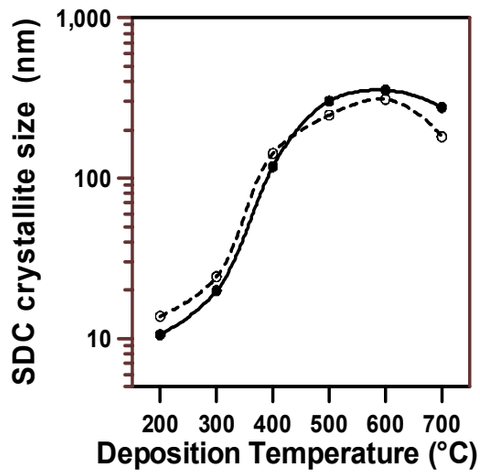

Figure 1: Plot showing the influence of deposition temperature (*Tdep*) on the average crystallite size for SDC films grown by PLD. Solid line: as-deposited films; dashed line: after testing at 480 °C.

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