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Preparation of $La_{0.8}Sr_{0.2}CoO_{3-\delta}$ sputtering targets using a deformable compaction die

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ABSTRACT

Fabrication of sputtering targets deviates from the customary practices in ceramic processing as their production volumes are often quite low. The use of hot-press, in this context, greatly facilities the fabrication of sputter targets since both the density and the dimensions of the target are controlled during the pressing. In the absence of hot press, however, the fabrication requires extensive preliminary work, but difficult to justify due to limited volume production. In this study, in place of customary rigid die, we propose the use of a deformable die which greatly simplifies the fabrication procedure. In this approach, polytetrafluoroethylene (PTFE) rings are used as compaction die filled with powders, tapped to uniform density. The die is then deformed between parallel platens whereby compacting the powders. The method relies on the fact that the pressing leads to almost no change in the internal diameter of the ring. This approach was illustrated with the fabrication of 2 in. $La_{0.8}Sr_{0.2}CoO_{3-\delta}$ (LSC-113) target where the deformable die was dimensioned by preliminary experiments on PTFE rings of small diameter. Sputter targets of sintered density greater than 0.95 and dimensions within the tolerances of the sputter gun were successfully fabricated. It is proposed that the approach may also be applicable to flat products of irregular shape, as high friction in tapped particulate media makes the lateral flow difficult, confining the compaction mainly to axial direction.

1. Introduction

For voluminous processing of ceramics, it is customary to carry out an extensive preliminary work to determine the fabrication parameters. Once these are determined, a die of suitable dimension is prepared and the powders are compacted and sintered under carefully controlled conditions. In the case of sputter targets, the use of hot-press greatly simplifies this process as it ensures controlled densification with proper dimensional control [1]. Where the hot-press is not available, it is necessary to revert to the conventional practice of compaction and sintering which require, as pointed above, a lengthy preliminary work. This is quite demanding especially for a low volume production.

Sputter targets, especially for research purposes, of necessity, is of low volume, but with stringent requirements [2,3]. First, they should have high relative densities (better than 0.90) as otherwise there would be compositional deviations between the sputtered film and the target [4]. Also, they are expected to have a uniform distribution of grain size so as to suppress the nodule formation which could lead to problems in the deposited films [5].

The current work is a part of an effort to develop an efficient cathode material for solid oxide fuel cell via sputter deposition. The aim is to produce sputter targets of high density with uniform grain size suitable for the deposition of cathode thin films $La_{0.8}Sr_{0.2}CoO_{3-\delta}$ (LSC-113) as well as $La_{0.8}Sr_{0.2}CoO_{3-\delta}$ /$(La_{0.5}Sr_{0.5})_2CoO_4$ (LSC-113 / LSC-214) mixture. For this purpose, we utilize a method that relies on the use of deformable die for compaction.

2. Experimental procedure

LSC-113 as well as LSC-214 powders were synthesized using Pechini method. Although both powders were synthesized, the details reported below refer to LSC-113 only. In this method, the solution of respective nitrates is mixed with ethylene glycol and citric acid by stirring at 100 °C. After drying at 250 °C, calcination was carried out at 750 °C for 5 h. Powders obtained were crystalline without any additional phases. Particles were in the form of agglomerates approximately 5 μm in size made up of submicron oxides.

Compaction was carried out with an 800 kN hydraulic press. Dies used were in the form of PTFE (trade name Teflon) rings 5 mm in height with 5 mm wall thickness, Fig. 1. Sintering was carried out in a

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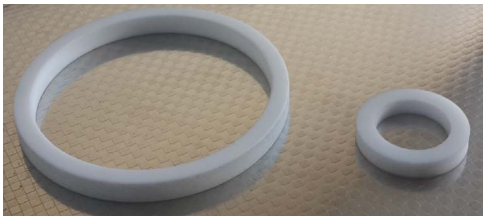

Fig. 1. PTFE rings used as a deformable die. Smaller diameter ring on the right was used for preliminary experiments. While the one on the left was used to fabricate 2 in. diameter $La_{0.8}Sr_{0.2}CoO_{3-\delta}$ (LSC-113) sputter target.

muffle furnace at the selected temperature, with heating /cooling rates of 2 °C/min with a hold during heating at 750 °C for 1 h.

Density of both compacted and sintered discs were measured with Archimedes method. For this purpose, the samples were made water impermeable by application of lacquer. Results were expressed as the relative density which is the ratio of measured density over the full density of the oxides. Microstructural characterization, with regard to grain structure of the deposited films, was carried out using a scanning electron microscope (SEM) with a FEG gun.

Sputter deposition was carried out in a special system designed for combinatorial thin film work [6], incorporating both R.F. and D.C. sputter guns. Target holders were 2 in. in diameter with a copper base. Targets were mounted onto this base with the use of a ring, 49 mm in inner diameter that could be fastened onto the base. This allows the use of targets of 50 mm in diameter ± 0.7 mm in tolerances.

3. Results and discussion

Initial work was carried out with a conventional die. Thus a steel die, 50 mm in inner diameter, was used for compaction of LSC 113 powder. Compaction was carried out under a pressure ranging from 80 MPa to 120 MPa. Densities of the green compacts are reported in Table 1. It is seen that the density increases with increase in the compaction pressure. Compacts were then sintered at 1300 °C for 4 h. Representative microstructures after sintering are given in Fig. 2. Here, the sample compacted with 80 MPa was not considered satisfactory as there was a lack of fusion in the sintered oxide, Fig. 2(a). The relative density measured for this sample was 0.80. With the compaction pressure of 110 MPa, the powders were fused together with the presence of occasional small pores yielding a relative density of 0. 96. The microstructure in this sample was also quite satisfactory in terms of its uniformity. As a result, a relative density of 0.95 was taken as the minimum target value.

Based on the sintered density and the associated shrinkage, a rigid compaction die could be prepared for the fabrication of 2 in. LSC-113 target. It appears that a compaction die of 65 mm inner diameter, would be required for this purpose. The compact would then shrink to 50 mm after sintering. Since the relative density and shrinkage after sintering would change depending on choice of powder, it is necessary to repeat the above work for LSC-214 as well as LSC-113/LSC-214 mixtures.

Compaction dies are normally fabricated from tool steels machined

Table 1
Green density and the relative density of LSC113 compacted in rigid and deformable die at different compaction pressures.

Pressure	Rigid Die		Deformable Die	
	Density (g/cm^3)	Relative density	Density (g/cm^3)	Relative density
80 MPa	3.974	0.552	3.953	0.549
100 MPa	4.075	0.566	4.018	0.558
120 MPa	4.142	0.575	4.147	0.576

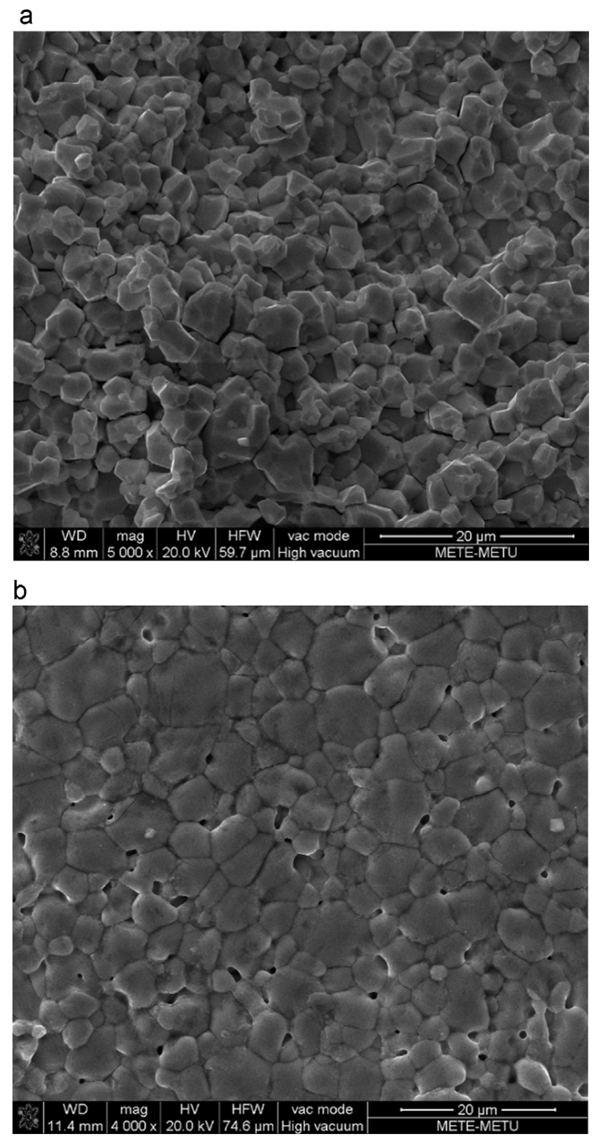

Fig. 2. SEM micrographs of LSC-113 after sintering at 1300 °C compacted with a conventional die with a pressure of a) 80 MPa, b) 110 MPa.

to close tolerances followed by quenching and tempering. The fabrication is, therefore, rather an involved process and a simpler alternative would be highly desirable. Here we propose the use of deformable die as an alternative.

Deformable dies used for the fabrication of LSC-113 target are in the form of rings of low aspect ratio, i.e. height is small in comparison to the diameter, see Fig. 1. For preliminary experiments, in order to save the powder, small diameter dies were used. Therefore, a large number of PTFE rings of 20 mm inner diameter were machined from PTFE rods of a suitable diameter.

The experiments, described for the conventional die for LSC-113 target, were repeated for 20 mm PTFE die where the compaction pressure varied from 50 to 135 MPa. It should be pointed out that the geometry of compaction, here, is different from the conventional practice. The PTFE ring is pressed between two parallel platens deforming the ring and compacting the powder. Thus the ring is used once and then discarded. This is in contrast to the conventional compaction where powders are compacted within a die – punch system, a process that can be repeated many times with the same toolset, Fig. 3.

In the experiments, PTFE rings were filled with powder, tapped to obtain uniform density. They were then pressed uniaxially to the