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Taguchi design of experiments for optimization of ionic conductivity in nanocrystalline Gadolinium doped Ceria

Shrikant Kulkarni[a], Siddhartha Duttagupta[b], Girish Phatak[a,*]

[a]Centre for Materials for Electronics Technology (C-MET), Off Pashan road, Panchawati, Pune 411008, India
[b]Department of Electrical Engineering, Indian Institute of Technology (IITB), Powai, Mumbai 400076, India

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Abstract

Taguchi experiments are designed and carried out for glycine nitrate precursor (GNP) combustion method to study four preparation parameters, *viz.* Fuel to oxidizer molar ratio, oven temperature, calcination temperature and calcination dwell time for their influence on physical properties and ionic conductivity of sintered pellets of Gadolinium doped Ceria (GDC) powders. These four parameters with their three levels form L9 orthogonal array. Ionic conductivity measurements were done using Nyquist and Bode plots obtained using frequency perturbed impedance analysis in the frequency range of 1 Hz to 1 MHz at temperatures 500–700 °C. The optimum conditions was found out on the basis of ionic conductivity by using 'larger the better' analysis. Both, the Analysis of Mean (ANOM) and Analysis of Variance (ANOVA) indicate that Fuel to oxidizer ratio is the only influential process parameter. The ionic conductivity of GDC powder prepared using optimized conditions by Taguchi method was found to be 0.023 S cm^{-1} at 600 °C, which is 1.5 times higher than earlier measurements. Significantly, this is highest reported conductivity for GDC at 600 °C. The significant improvement in results following Taguchi optimization underlines importance of this method for materials synthesis.
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1. Introduction

Solid Oxide Fuel Cells (SOFCs) are the most promising non-conventional electrochemical power sources that convert chemical energy into electrical energy with high fuel efficiency [1]. SOFCs are capable of generating high power in the range of 10–1000 kW with energy density up to 1 W cm^{-2}. They are fuel flexible and can be operate using variety of fuels, such as, pure hydrogen, hydrocarbons and alcohols. However, one of the serious limitations of SOFC is the requirement of high operating temperature (~1000 °C). Such a high operating temperature results in difficulties of sealing, interconnects and causes limitations in its commercial use for mobile or hand-held device applications [2]. Lowering operating temperature to 300–600 °C range can resolve these issues and help in reducing its fabrication and operational cost [3]. However, it is a difficult task. The operating temperature of SOFC is dictated by the ionic conductivity of the electrolyte, which must have a reasonable value for a practical power source. Presently, Yttrium Stabilized Zirconia (YSZ) is a commercially used electrolyte in SOFC. YSZ offers oxygen ion conductivity of 0.1 S cm^{-1} at 1000 °C, which is a benchmark for developing alternative, low operating temperature electrolytes for SOFC applications [4]. In the past few years, Gadolinium doped Ceria (GDC) is considered to be one of the promising oxygen ion conducting electrolyte for intermediate temperature solid oxide fuel cells (ITSOFC) [5]. This electrolyte exhibits higher ionic conductivity than YSZ in the operating temperature range of 300–700 °C [1]. These properties of GDC have prompted investigations into various preparation methods and their optimization for ionic conduction. It is seen from these reports that combustion synthesis is a relatively less explored but promising method for the synthesis of GDC [6].

*Corresponding author.
E-mail address: gjp@cmet.gov.in (G. Phatak).

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Combustion synthesis is a simple wet chemical method mostly used to synthesize nanocrystalline oxide materials. The combustion synthesis process is greatly influenced by the type of fuel and the fuel-to-oxidizer ratio (F/O ratio). Depending upon the fuel used and the type of metal ion involved, the nature of combustion differs from flaming (gas phase) to non-flaming (smoldering and heterogeneous) type. The fuel specificity appears to be dictated by the metal–ligand complex formation, the thermodynamics of the reaction as well as thermal stability of the desired oxide [7]. Glycine nitrate precursor (GNP) method is one of the combustion synthesis methods that use Glycine as fuel. Several parameters influence the combustion reaction, such as, atmospheric conditions of temperature and humidity, fuel to oxidizer ratio, fuel content, ignition temperature, water content in precursor and pH of the solution *etc* [8]. Such multi-parameter influence on the reaction product demands enormous number of experiments to arrive at optimum preparation conditions. Using statistical method for experimental design, such as, Taguchi method, would be a better alternative.

Several reports present use of Taguchi method to optimize materials synthesis parameters. TiO_2 synthesized by the hydrolysis of tetraisopropyltitanate (TIPT) in the presence of de-ionized water and ethanol under high-intensity ultrasonic irradiation process was optimized by Taguchi method. In this study, L9 (3^4) orthogonal array was used to study four parameters, *viz.* water content, water-to-TIPT ratio, water-to-ethanol ratio and sonication time with three 'levels' for each parameter [9]. Synthesis parameters of fast hydrothermal method and Arc Spray method were also optimized using Taguchi method to synthesize TiO_2 nanoparticles using L9 and L16 orthogonal array respectively [10–11]. The Mechano-chemical synthesis parameters of CdO were optimized using Taguchi method to achieve Cauliflower-like nanostructured CdO [12]. Taguchi method was also used to optimize sol gel synthesis parameters for CuO–ZrO_2 nanoparticles [13]. Specifically, this method was used by Ghosh et al. for obtaining optimized synthesis parameters of Hydroxyapatite (HAp) by solution combustion method [14]. This paper attempts to evaluate the influence of process parameters on the flame temperature as well as physical characteristics of HAp powder using Taguchi method. A L16 orthogonal array with four parameters *viz.* batch size, diluents, fuel to oxidizer ratio and initial furnace temperature were studied to find optimal conditions. It is seen from these reports that Taguchi method has been used successfully for optimizing preparation conditions for various compounds. However, it is also seen that no such method is reported to optimize preparation conditions of GDC by GNP combustion method.

The key objective of this work is to optimize GNP combustion synthesis parameters for GDC to obtain optimum ionic conductivity in intermediate range of operating temperature (500–700 °C). In order to achieve this, we have studied the GNP synthesis parameters comprehensively because of their crucial role in phase purity, stoichiometry, crystallite size, grain growth and density of sintered pellets. Four important synthesis parameters *viz.* fuel to oxidizer ratio, oven temperature, calcination temperature and calcination dwell time have been focused upon in this study. Influence of these parameters on ionic conductivity of GDC has been studied with an aim of obtaining optimal experimental conditions for maximum conductivity of GDC at 600 °C.

2. Experimental procedure

2.1. Materials and process

The GNP combustion method was used to synthesis nanocrystalline GDC powders. The source of metal cations was Cerium nitrate ($Ce(NO_3)_3 \cdot 6H_2O$) and Gadolinium nitrate ($Gd(NO_3)_3 \cdot H_2O$) (Aldrich make) while Glycine (SISCO make) was used as fuel. A precursor solution was obtained by dissolving desired stoichiometric molar ratio of metal nitrates in DI water (Millipore ELIX 10). Solid state fuel (Glycine) was added as complexing agent in appropriate predetermined proportions with respect to metal nitrate. The precursor was kept stirring for 12 h. Water was removed by heating at 70 °C which results into gelation of solution [15]. The gel was kept in an oven at controlled temperature until spontaneous ignition took place. After combustion, the ash was collected and calcined at different temperatures and dwell times to obtain GDC powder.

2.2. Design of experiments

Three levels of four synthesis parameters, *viz.* Fuel to oxidant ratio, Oven temperature for combustion, calcination temperature and calcination dwell time were chosen for this study. The L9 orthogonal array of these parameters was prepared as given in Table 1. These permutations of the synthesis parameters are named as experimental conditions T1 to T9. The powders prepared under these conditions are also identified by this nomenclature. Each sample was prepared three times to ensure repeatability of the results.

2.3. Materials characterization

The calcined powders were characterized by X-ray Diffraction (XRD; PANanalytical PW3040/60 X'pert PRO diffractometer

Table 1
Taguchi L9 table prepared using four parameters with 3 steps of each.

Experimental condition	F/O molar ratio	Combustion temperature (°C)	Sintering temperature (°C)	Sintering time (h)
T1	1.00	200	600	2
T2	1.00	250	700	4
T3	1.00	300	800	8
T4	1.70	200	700	8
T5	1.70	250	800	2
T6	1.70	300	600	4
T7	2.50	200	800	4
T8	2.50	250	600	8
T9	2.50	300	700	2

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