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TEMPERATURE DEPENDENCE DIELECTRIC BEHAVIOUR OF 20GDC ($CE_{0.8}GD_{0.2}O_{2-\delta}$) EFFECT FOR SOLID OXIDE FUEL CELL APPLICATION

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ABSTRACT

In the present investigation 20GDC ($Ce_{0.8}Gd_{0.2}O_{2-\delta}$), **nano powder** was synthesized from the sol-gel method. The nano powders were sintered at 1400 ^oC for 2hours by converting into pellets. The density of sintered pellets measured from the Archimedes method was found to be around 96%. XRD of 20GDC powder and sintered pellets shown were in cubic fluorite structure. Morphology of 20GDC powder was sintered pellets by using SEM. Grain size was measured from the linear intercept method. Dielectric properties (AC conductivity, Dielectric constant and loss tangent) were measured for 20 GDC samples using Solatron Impedance analyzer with temperatures ranging from 300^{o}C to 800^{o}C and with a variation of frequency from 1 KHz to 1MHz.

Keywords: GDC Nano Powder, Sol-gel Method, Pellet, SEM, Dielectric Constant.

INTRODUCTION

In present years, solid oxide fuel cells (SOFCs) have involved much attention due to a large amount of efficient and environmentally energy conversion devices[1-2]. Generally, yttrium-stabilized zirconia (YSZ) materials are used as electrolyte materials which require high operation temperature (Above 1000^{o}C), so these materials are not useful for engineering applications due to material degradability at high temperature and high cost in the economical point of view [3-4]. To reduce the operating temperature of solid oxide fuel cells (SOFCs) to below 800^{o}C is more important due to improving stack life span and cost reduction [5-6]. However, in case of SOFCs at Intermediate temperatures, there are two possibilities of the following:

1. Electrode reaction rates decrease
2. Minimize cell losses(Ohmic and polarization losses)at the electrode-electrolyte interface

Generally, Ceria-based materials are considered as great alternative electrolytes for lowering the operating temperature of SOFC than conventional YSZ. Ceria-based materials are one of the most exciting electrolyte materials which have been used at low temperature[6] due to the great formation as catalyst and oxygen sensors etc[18].

Ionic conductivity in oxide element plays an important role for SOFC applications, Brook and Kilner was observed maximum ionic conductivity in oxide fluorites with the addition of a type of dopant of its ionic radius, causes less elastic strain in the parent crystal lattice. By doping with oxides of metals with lower valences, oxide vacancies may arise e.g., by the dissolution of Gd_2O_3=$2Gd'_{Ce}+V^{oo}_o+3O_o$. In the present study, gadolinium doped ceria-nano powders are prepared using Sol-gel and sintered at 1400^{o}C for 2

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hours by converting into circular pellets. Dielectric properties of sintered 20 GDC sample is analyzed and presented.

EXPERIMENTAL

In the present investigation, nano-crystalline ceria powders 20 mol% of gadolinium (**$Ce_{0.8}Gd_{0.2}O_{2-\delta}$**) were synthesized by a modified sol-gel technique featuring a nitrate-fuel exothermic reaction[9]. GDC pellets were prepared from powders and sintered at 1400°C for 2 hours at a heating rate being 2°C/minute and cooling rate was 5°C/minute. The crystalline structure and phase behavior is analyzed using powder X-ray diffraction technique with model XPERTO-PRO Diffractometer along with Cu-Kα radiations at wavelength 1.54060 A^{o}. The crystalline grain size is calculated using Scherrer equation [8]. The scanning electron microscope of model CRL-ZESIS-EVO-MAI5 is used to study the microstructure behavior of sintered GDC samples and electrical properties such as AC conductivity, loss tangent and dielectric constant were measured at different temperatures ranging from 300^{o}C to 800^{o}C with a variation of frequency from 1 KHz to 1MHz.

The X-ray diffraction analysis of the 20GDC synthesis powder and sample sintered at 1400^{o}C-2h GDC samples are shown in Fig.-1. XRD patterns have illustrates CeO_2 solid solution as the well-known cubic phase. However, with increasing temperature, the lattice parameter increases from 5.410 Å to 5.424 Å. Fig.1 shows that there is no sign of unreacted do-pants or secondary phase formation with doping of Gadolinium (Gd^{3+}) to ceria. The XRD pattern is identical with the cubic fluorite structure of ceria lattice [9].

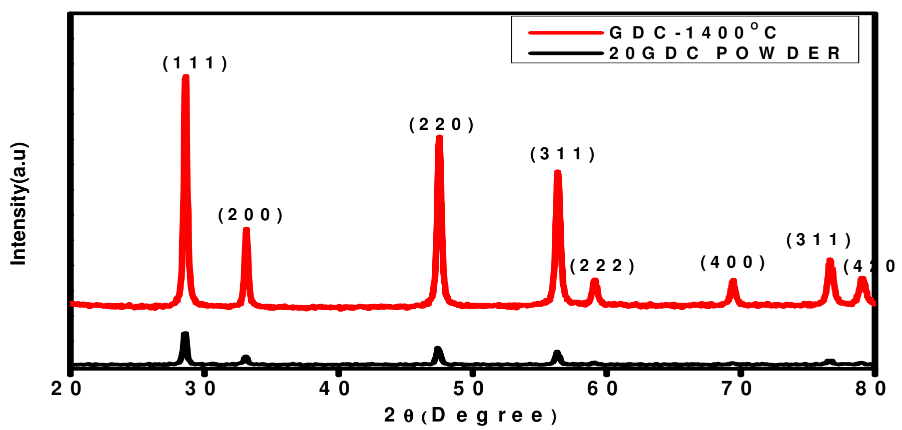

Fig.-1: X-ray Diffraction Pattern of 20 GDC ($Ce_{0.8}Gd_{0.2}O_{2-\delta}$)

The scanning electron micrographs of Pure and sintered 20 GDC at 1400 ^{o}C were presented in Fig.-2a and Fig.-2b. It shows that surface morphology of 20GDC with well-grained structure with size being calculated by the linear intercept method as in 1.72 µm. The density of sintered pellet calculated from Archimedes method was found to be around 96%. It was evident that no pores were observed in Fig.-2b in the sample due to high dense material[7-8]

Dielectric properties such as AC conductivity, loss tangent, dielectric constant, were measured for 20 GDC samples with temperatures 300^{o}C to 800^{o}C and with a variation of frequency from 1 KHz to 1MHz. In this analysis, dielectric constant (Fig.-3) was measured as function of frequency decreased in frequency and increased in temperature. From the plots, it is observed that 'universal' dielectric response and the Jonscher's law is applicable to such materials which do not show loss in peaks[10], and at lower frequencies, high values of dielectric constant were observed the sign of the electrode-electrolyte interface. The shift of peaks towards higher frequencies was evident from graphs ascribed to Maxwell-Wagnertype of interfacial polarization in agreement with Koop's phenomenological theory[18-19]. Koop's theory suggests that dielectric materials have large number of conducting grains that are divided by defined grain boundaries. If external electric field applied on material produces large polarization with high dielectric constant due to the acquisition of charge carriers takes place between grain and grain boundaries and also because of this process dielectric constant decreases with the increase of frequency. The value of dielectric constant arises due to all four types of polarization mechanisms identified at lower frequencies and low dielectric constant values at higher frequencies arises along with dipoles and highest relaxation times which stop to react with the applied frequency[19].

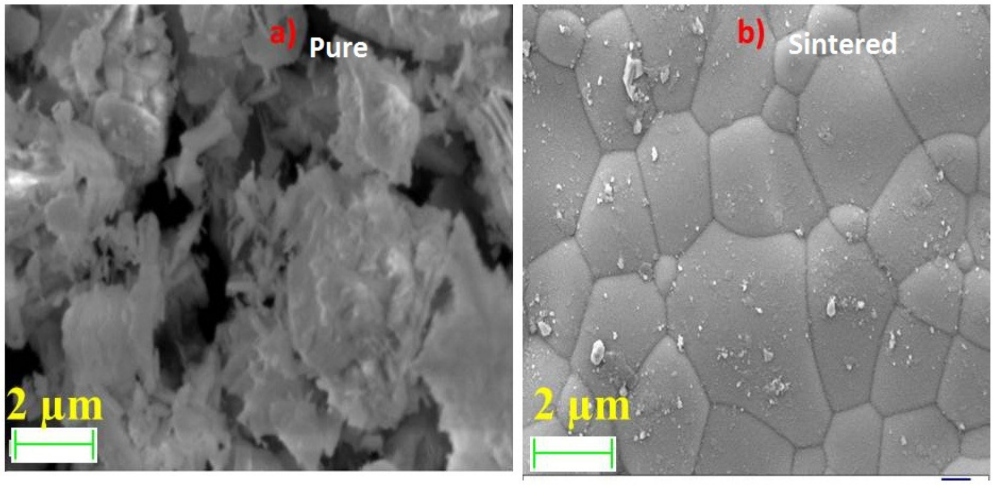

Fig.-2: Microstructure of 20GDC Powder and Sintered of 20 GDC at 1400°C

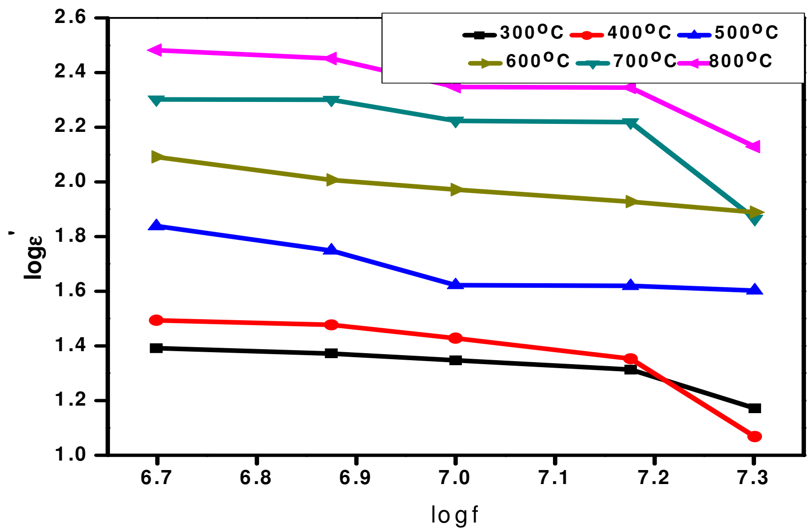

Fig.-3: Dielectric Constant as a Function of the Frequency of $Ce_{0.8}Gd_{0.2}O_{2-\delta}$ at Different Temperature

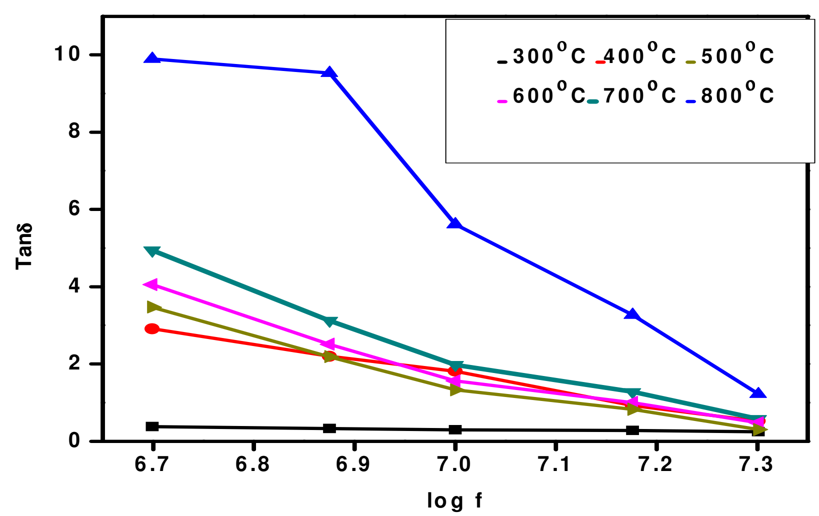

Fig.-4: Dielectric Loss Tangent (Tanδ) as a Function of the Frequency of $Ce_{0.8}$ $Gd_{0.2}O_{2-\delta}$at Different Temperatures.

The frequency-dependent tanδ plots were shown in Fig.-4 for $Ce_{0.8}Gd_{0.2}O_{2-\delta}$. It was evident that dielectric loss tangent tanδ values changed from larger to lower values and decreased with the increase of frequency. The effect was reflexive because the ions were not able to respond and reorient themselves with applied frequency. It is clear from the cited plots that with increasing temperatures, the reorientation and relaxation peaks shift towards higher.

The variation of AC conductivity of sintered GDC at 1400°C with frequency at different temperatures ranging from 300°C to 800°C was shown in Fig.-5. It was deduced that with the increasing frequency, AC conductivity values were decreased. This behavior could be explained by jump relaxation model.[15-16]

AC conductivity was at lower frequencies due to the jumping of ion from one vacant state comprehensively where dispersion occurs at higher frequencies due to relaxation and hopping of ions. Dispersion region moves towards the high-frequency region with increasing temperature. This may confirm the ionic hopping mechanism in addition to Arrhenius relation by Jonscher's law.[15]

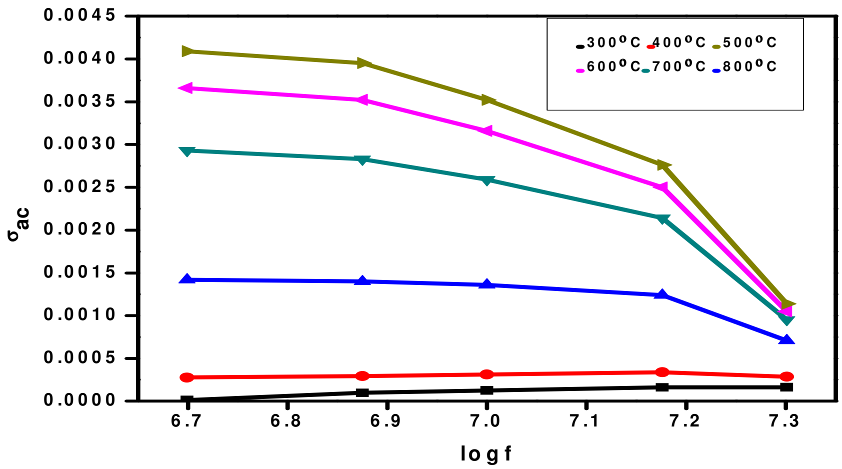

Fig.-5: AC Conductivity as a Function of the Frequency of $Ce_{0.8}Gd_{0.2}O_{2-\delta}$ at Different Temperatures

CONCLUSION

Nano-crystalline gadolinium doped ceria, $Ce_{0.8}Gd_{0.2}O_{2-\delta}$ were successfully synthesized with the sol-gel method. The X-ray diffraction confirms that the formation of single-phase and lattice constants were calculated and were observed in the recorded range of 5.40 Å to 5.42 Å due to the large ionic radius of Gd than Ceria. SEM was used to study the morphology of the materials and grain size was 1.72 µm at a sintered temperature of 1400°C. The dielectric properties such as dielectric constant, loss tangent, and Ac conductivity values decrease with increasing frequency in the range 1 KHz to 1MHz and variation with temperature ranging from 300°C to 800°C due to Maxwell-Wagner type of interfacial polarization in agreement with Koop's phenomenological theory. The variation of Ac conductivity was calculated in the frequency range 1 KHz to 1MHz with a variation of temperature 300°C to 800°C exhibited an increase in conductivities observed with the increase of temperature due to the universal dielectric response with Jonscher's law and jump relaxation model, which will affect the conduction phenomena for nano synthesized 20GDC material.

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