

THE IMPEDANCE MEASURE OF YTTRIUM ALUMINIUM GARNET DOPED WITH CADMIUM OXIDE.

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ABSTRACT

Yttrium Aluminium Garnet ($Y_3Al_5O_{12}$ or YAG) doped with Cadmium oxide (CdO) was fabricated and the electrical conductance investigated. The impedance technique was applied in the study of the electrical conductance whereas the famous sintering by powder mixing process was used in producing the pellets of the system. The system showed two distinct dumb-bell envelopes of impedance curves. This assisted in the discrimination and extraction of the conductance components as grain and grain boundary conductance. The analyses of the results showed a mixed ionic n-type electronic conductivity with the main contributor as defects clusters at the intergranular grain junctions where impurities appear to segregate. It has been suggested that Cd^{++} and other impurity ions were responsible for the electronic conductivity of the samples. An activation energy of about 1.62 eV was calculated for the best sample in terms of composition. Consequently, the characteristics of the system suggest that the materials is corrosion resistance and translucent and could be applied in most electrical and electronic industries.

KEY WORDS: Impedance, doping, Yttrium Aluminum, Conductance

INTRODUCTION

Yttrium materials and their related components have been generally regarded as good structural system with high degree of compressive strengths (Masanori and Hiroshi, 1991). Thus the choice of studying the present system by doping it with CdO arose from its unique mechanical strength, sintering characteristic properties, corrosion resistant ability, thermal resistance capability and the ease of processing the materials coupled with the electrical properties of CdO (Asokan, 1988). It is expected that CdO will impact positively on the system by diffusion process of its atoms into the microstructure of the system. The previous work of Kingery and Tuller (1980) on the sintering behaviour of ceramics and their properties emphasized more on the mechanical properties than the electrical behaviour of the system. Hence, the search for suitable materials with comparable electrical properties has continuously been a subject of challenging interest. This has, in part, prompted most researchers (Quadir, 1998 and Lackey, 1989) into new region of material development with regard to electrical characteristics.

In addition to the establishment and growing interest in the ionic conductivity phenomena in some ceramic, workshops and seminars were organized by professionals and interest groups with a view to encouraging and motivating researcher on the relevance of this area of study (Fisher, 1996). This generated very strong interest among specialists in development of electrical materials eventhough Idem, (1988) and Schuh and DeWith (1994) have previously made some initial strides in opening up this area. Besides, it was reported by Lackey (1989) and Kingery & Tuller (1980) that MgO is a mixed electrical conductor with a predominant electronic conductivity at both low and high oxygen pressures. Also, Yamaka (1984) and Fisher (1996) showed that negative thermoelectrical signals in vacuum and high temperature conditions are normally present in most mixed oxides. This implies that electronic transport mechanism may be responsible for the conductance, which are commonly noticed in difference mixed-oxides of ceramics.

In most of these studies, it has been shown that proper ceramic processing of the starting powder material plays a paramount role in assisting the quality of the final product

required. Consequently, the theoretical and experimental data are normally needed for comparison and derivation of impedance model curves that may help in detail investigation.

The conditions for processing and sintering usually influences the mechanism of most systems. Thus, several electronic band structure arising from this have been detected (Francis, 1985 and Hales, 1992). The procedures adopted for the present study include among other factors; doping by mixed oxide method, powder, processing and impedance spectroscopy. By characterizing the starting material (powder), it has been possible to determine any possible source of error since electrical material requires high degree of purity. Thus the activation energy of the system, transport number and Hall coefficient were determined from the impedance plots.

MATERIALS AND METHODS

Yttrium aluminum garnet (YAG) and Cadmium oxide powders (Reachim, USA) average particles sizes of approximately $3\ \mu\text{m}$ were wet-mixed and electrically vibrated for about 12 hours. The CdO was applied as an impurity dopant, which meant that the weight percentage used was carefully determined and these are shown in Table 1. The entire mixture was later oven-dried at about 100°C and calcined at 350°C for three hours.

The calcined powder product was then crushed, sieved, air-blown and sprayed-dried to form nice powder spheroid particles. Thereafter, 5% poly-vinyl alcohol was added as a binder and thoroughly mixed before pressing out the pallet samples. This increased the consolidation and ease of pressing the samples into pallets. Green samples were hydrostatically pressed into discs of about 16mm diameter and about 1-mm thickness.

By using the isostatic cold hydrostatic pressing technique. Green specimens of between 45 and 60 percent theoretical densities were produced before the sintering process.

The specimens were all sintered in a muffle furnace between 800°C and 1300°C for a comprehensive sintering time of six hours at the rate of $10^\circ\text{C}/\text{min}$. (initially) and later 20°C (during the hold period). A typical sintering cycle is shown in Fig. 1.

By simple weighing and measurement of dimensions, the densities of the green and final products were determined. Also the weight losses, shrinkage and porosity of the samples were estimated by standard methods which have been applied elsewhere (Owate and Otounefor, 2000).

The electrical conductivity of the specimen was measured as a function of both frequency and temperature using the frequency response analyzed and the impedance meter

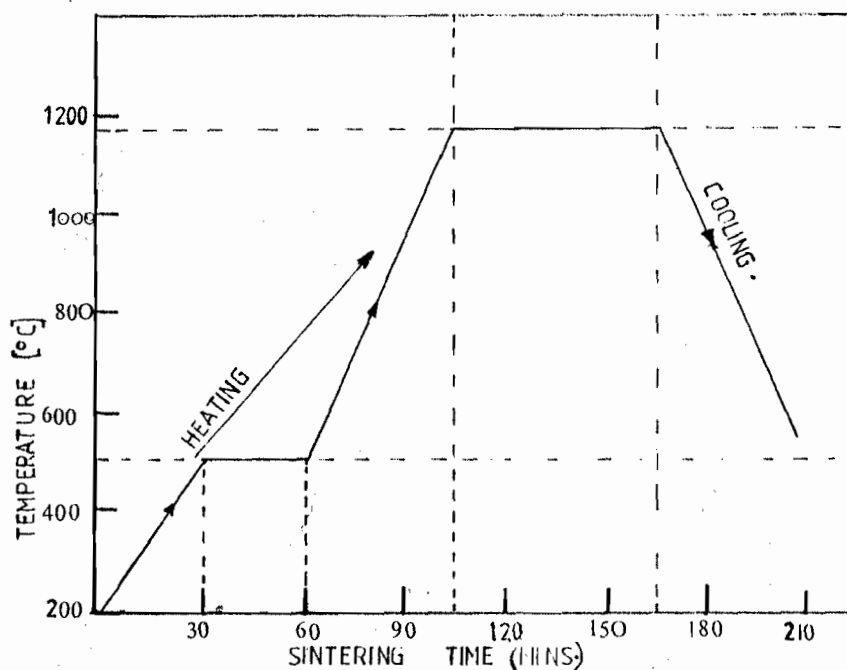


Fig.1 A typical sintering cycle.

Table 1: Compositions and Parameters of YAG

SAMPLES	CONCENTRATION OF COMPONENTS (Wt %)		PARAMETERS		
	Y ₃ Al ₅ O ₁₂	CdO	% Theoretical Density.	Activation Energy (eV)	Grain Size (μm)
A	99.80	0.20	98.85	1.94	3
B	99.50	0.50	99.16	1.69	4
C	99.00	1.00	98.42	3.39	6
D	98.50	1.50	97.03	3.54	7
F	98.00	2.00	95.11	3.98	7

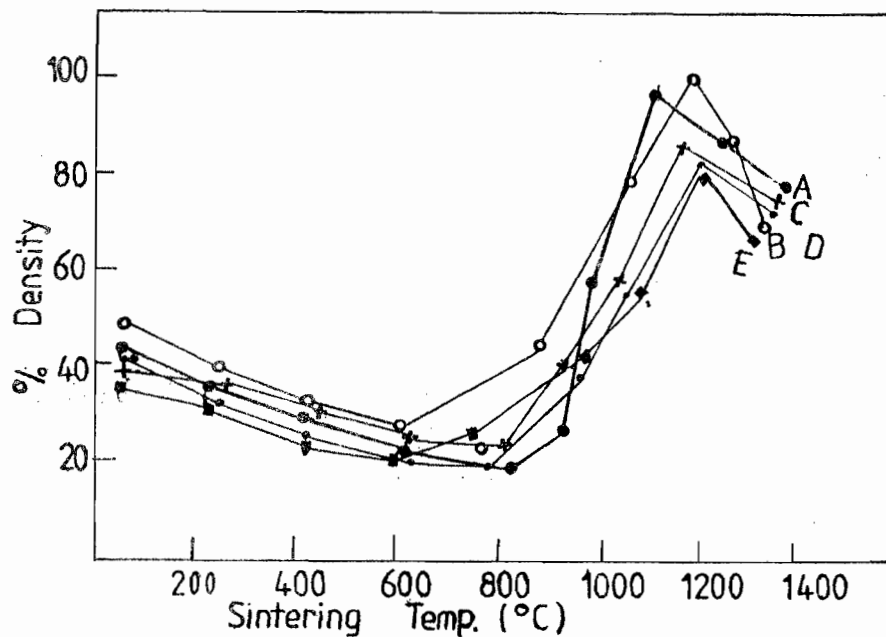


FIGURE 2

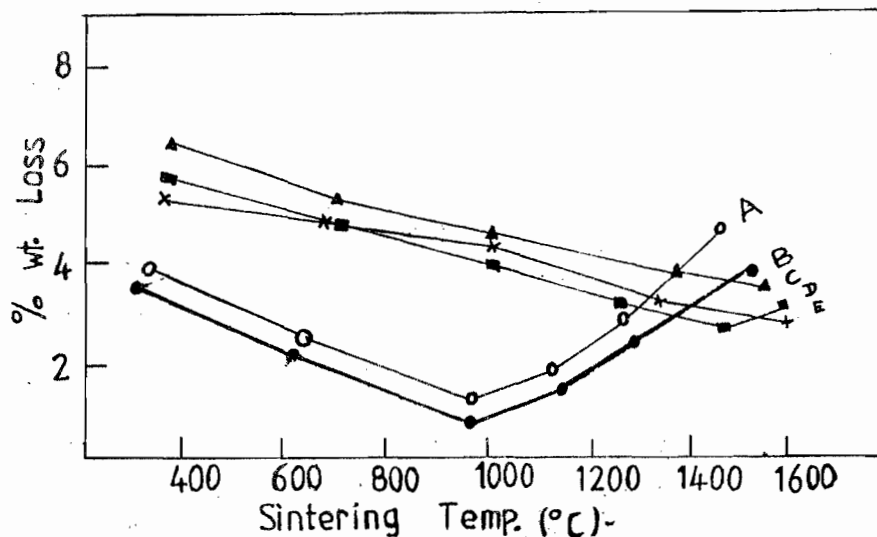


FIGURE 3

(SCLLUMBERERGER - Frequency Response, Model 18900 and Solariton impedance Analyzer Model 1176, 10 Hz-MHz). Each composition was singularly analysed and the activation energy for the conduction process determined at ambient atmosphere since the electrical conductivity experiment was conducted as a function of temperature. Consequently, the transport numbers for the systems were calculated whereas the Hall-coefficients were estimated.

RESULTS AND DISCUSSION

The functional dependence of density, shrinkage and weight losses sintering temperature are presented in Figures 2 - 9.

The densities for the sample general decreased slightly from the original green density and then increased proportionate with temperature up to 1150°C and 1150°C. Thus the maximum density was obtainable between 1050°C remarked that it has been possible to obtain higher % of theoretical density under controlled sintering conditions as observed in Table 1 for sample B, C and D. These values do not however contradict Fig. 1 due to the fact that factors such as rate of firing holding time and, the sintering cycle were skillfully manipulated to obtain the values in Table 1 as opposed to the trial experiment of Fig. 2. Fig. 2 shows the actual sintering conditions with greater level of the control of

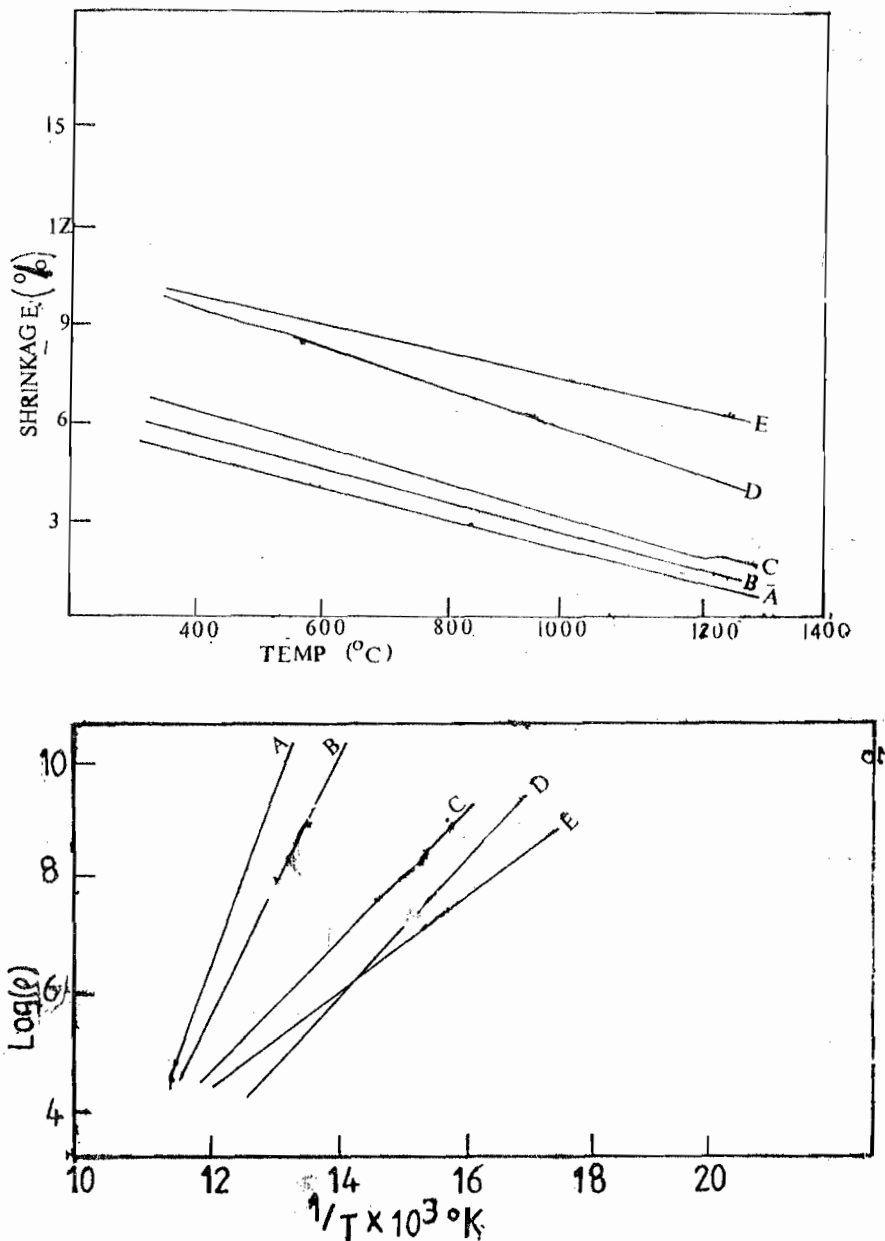


FIGURE 4

factors such as temperature and sintering time. The degree of weight loss and densities obtained justifies the influence of such factors. The weight loss decreased with increasing sintering temperature up to about 950°C for samples A and B and then later increased with increasing temperature. Samples C, D and E decreased in weight loss with increasing temperature. On the other hand Fig.4 shows that the percentage shrinkage decreased with increasing temperature eventhough samples A, B and C had lower percentage shrinkage than D and E samples.

The slight decrease in density at relatively low temperature observed in Fig.2 is attributable to high loss in the binder and some other volatile impurity materials with low vaporization temperature that were burnt off during the first stage of sintering. These weight losses coupled with considerable dimension shrinkage could have resulted to high consolidation process and the resultant increasing densities with temperature. The decreasing density with further increases in temperature outside the temperature range of 1050°C and 1150°C indicates that there is a suitable range of temperature for the attainment of maximum density. Outside this range of temperature, other processes that inhibit densification might begin to occur. This implies that at very high temperatures for the present material, microstructural defects such as pore formation and stress microcraks (Fig. 5, 6 & 7) were now inter-playing with the sintering mechanism. This might have been responsible for the low density recorded in the system. Thus, densification process for the system is achievable under controlled conditions between 1050°C. Above this range of temperature, grain growth and porosity that are the usual phenomena for solid state sintering might have started exerting undue influence on the system. In addition by increasing the dopant concentration beyond 1.50 wt % a negative impact was observed upon the system as presented in Table1. For example there was a reduction in density and increasing grain growth (Fig.7). the impedance plots for sample B and D are presented in Fig. 8 & 9 respectively. These curves were chosen because they illustrated the best and worse cases for the specimens based on the doping concentrations. Also, the Arrhenious plots for the samples are shown in Fig. 10 and the estimate ionic (-1.87 & + 4.89) and electronic transport (11) numbers in atmospheric oxygen

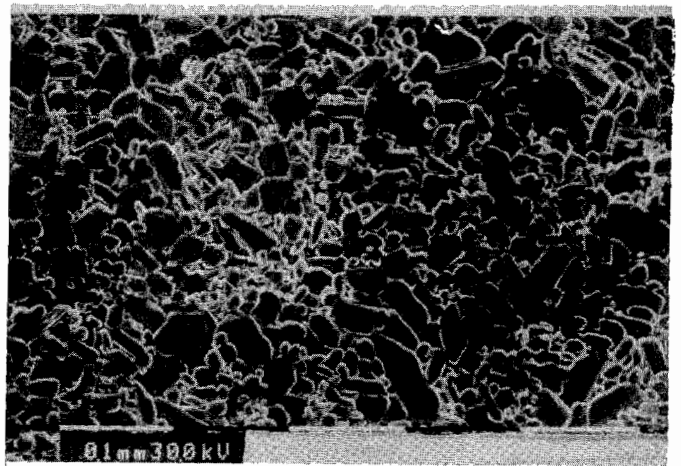


FIGURE 5

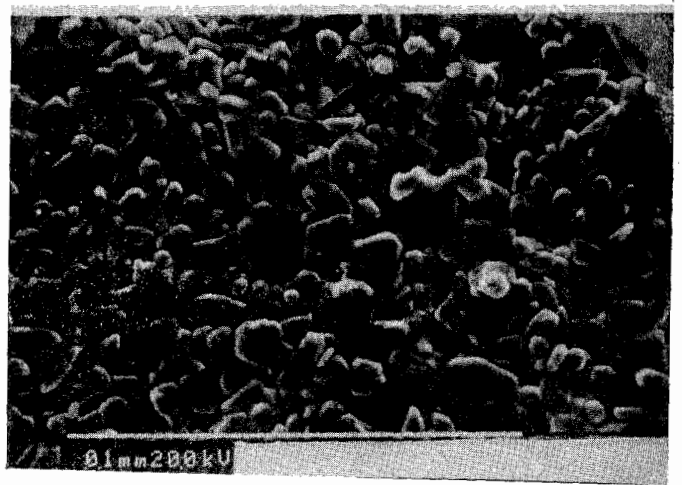


FIGURE 6

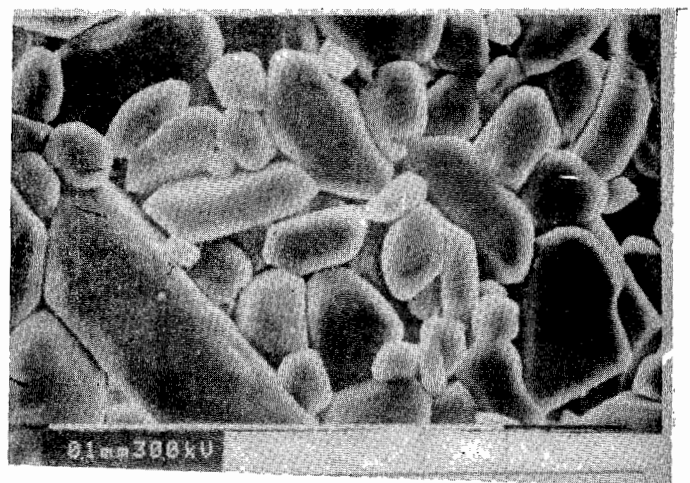


FIGURE 7

condition were computed. The dumb-bell shaped impedance spectroscopy curves were more distant and well pronounced for sample B (Fig. 8) than that of sample B (with minimal

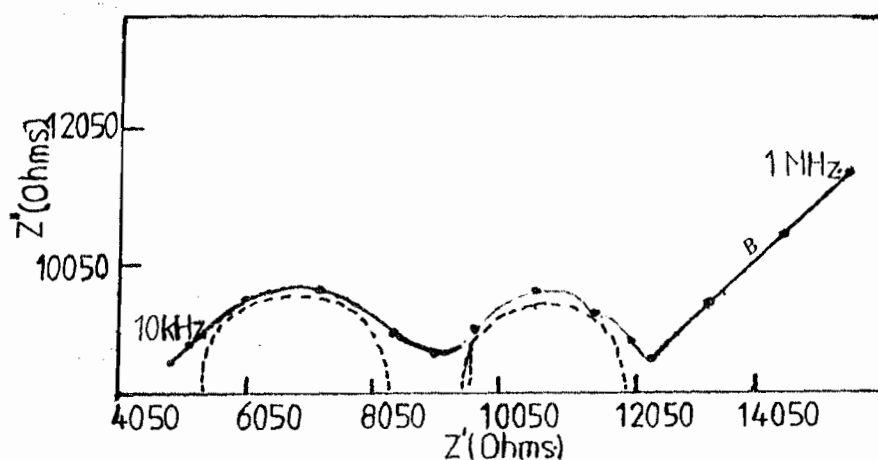


FIGURE 8

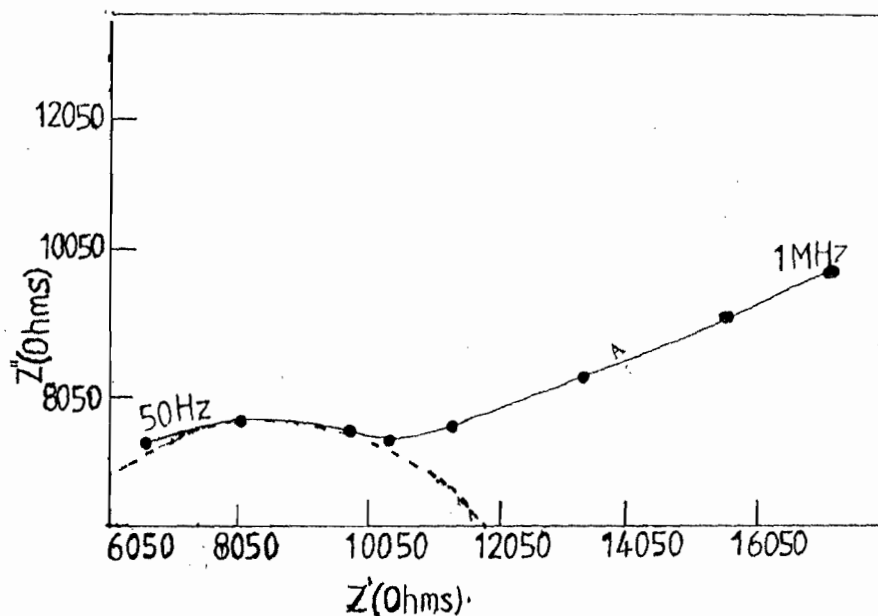


FIGURE 9

concentration of dopant) a fairly good approximation of impedance plot-shape was obtained. This indicates that low dopant concentration within the ambient oxygen atmosphere had influence on the bulk (grain) material and minimal effect on the grain boundary. However, increasing the concentration of the dopant reduces this influence of the impurity segregation into the grain boundaries as observed in both samples B and D. These observations are based upon the fact that the calculated conductivity shown in Fig.10 is that of a typical mixed ionic n-type electronic conductance. The major ionic defects that could be responsible for this might be impurities, gases within the pores and vacancy clusters. In addition, the

estimated Hall-coefficient indicated negative values, which suggest that an n-type electronic conductivity within the grain boundary interface was in operation for the system. Consequently protons are to be expected within the YAG garnet lattice, as this should balance the entire system as well as affect the grain interior as to balance the activities within the microstructure. Similar observations have been reported by Schuh et al, 1993 for Zirconium-doped YAG. Enhancing the above facts is the low activation energy of about 1.68eV computed for sample B. The value and that of sample A in Table 1 show that a small quantity of CdO reduces the relative activation energy for the Yttrium Aluminum garnet system.

CONCLUSIONS

It has been observed that in the Yttrium aluminium garnet (YAG) doped with 0.5-wt % cadmium oxides 99.16% of the theoretical density was achieved. The product exhibited an n-type electronic conductivity with average activation energy of 1.68 eV. The impedance spectroscopy has clearly demonstrated that sample B with a dopant concentration of about 0.5 wt % had distinct grain and grain boundary conductivities. Consequently, the impurities (dopants) were partly responsible for the conductance of the system.

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