



Impedance and modulus spectroscopy of $\text{ZrO}_2\text{--TiO}_2\text{--V}_2\text{O}_5$ nanocomposite

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Abstract

High-k $\text{ZrO}_2\text{--V}_2\text{O}_5\text{--TiO}_2$ nanocomposite prepared by coprecipitation-calcination method was investigated by impedance and modulus spectroscopy over a frequency range of 50 Hz–5 MHz, for different temperatures. Distinct peaks at characteristic frequencies were obtained. The shifting of the peaks towards high frequency with increase in temperature indicated a temperature dependent relaxation in the nanocomposite. Complex impedance formalism showed that the maximum value of the impedance decreases as temperature increases suggesting that the sample had negative temperature coefficient of resistance. The electric analysis confirms the findings of the impedance analysis. The f_{max} values for Z'' and M'' are different confirming non-Debye type of relaxation. The values of activation energies based on impedance and modulus relaxation were calculated and were equal to 1.02 eV and 0.51 eV respectively.

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1. Introduction

Polycrystalline dielectric layers in any electronic device are subjected to alternating fields. The grain boundary interface and the metal electrode interface play an important part in their electrical response. Impedance spectroscopy is an attractive method used to distinguish between the bulk and boundary effects. This technique enables us to study the individual electrical properties of component parts of a material, thereby enabling us to modify the material in a

favourable manner. It deals with the measurement of the real and imaginary parts of the impedance Z at various temperatures and frequencies. The impedance plots are in the form of semicircles that are associated with the grain boundary (R_{gb}) and bulk resistances (R_{b}). The effect of the intergrain and intragrain impedance plays a dominant role in deciding the material for a high-k dielectric layer. Zirconia and its composites have been studied extensively with respect to its applications in catalysis, SOFCs, dental and structural ceramics, dielectric layers on electronic devices etc [1–8]. The use of zirconia based materials as high-k dielectrics for gate dielectrics has been explored widely. Dielectric properties of binary oxides of Zirconium and titanium individually and with other

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dopants have been explored previously [9,10]. The addition of V_2O_5 to dielectric materials enhanced their dielectric properties [11,12]. A number of methods have been used to synthesize nanomaterials, but the simplest and effective method for synthesizing nanopowders is a wet chemical method. This method has been widely used in recent times to effectively synthesize nanomaterials [13,14]. In our previous study the morphology, optical and basic dielectric behaviour of the $ZrO_2-TiO_2-V_2O_5$ nanocomposite, synthesized by a coprecipitation method, were discussed [15]. A high dielectric permittivity of around 3000 was obtained. Here, the impedance spectroscopy and electric modulus analysis of the nanocomposite are studied and the results are discussed in detail.

2. Experimental details

A simple coprecipitation-calcination method was used to prepare the nanocrystalline $ZrO_2-V_2O_5-TiO_2$ composite [16–18]. Zirconium oxychloride, titanium tetrachloride and vanadium oxide (AR grade) were taken in the molar ratio 1:1:1 and dissolved in double distilled water. The water solution of $ZrOCl_2$, $TiCl_4$ and V_2O_5 was treated with aqueous ammonia at a pH of 9. This results in the formation of NH_4Cl . The hydrogel obtained was washed with distilled water to remove NH_4Cl and dried at $110\text{ }^\circ\text{C}$ for 5 h in a hot air oven to a constant mass. Further, the samples were calcined at $700\text{ }^\circ\text{C}$ in a furnace for 10 h. The synthesized samples were collected and powdered for characterization. A HIOKI 3532 LCR HITESTER instrument was used for obtaining the impedance parameters.

3. Results and discussion

3.1. Impedance analysis

The impedance of the material can be calculated using the formula [19].

$$Z(j\omega) = Z' - jZ'' = \frac{1}{1/R + j\omega C} = \frac{R}{1 + j\omega RC} \\ = \frac{R}{1 + (\omega RC)^2} - \frac{j\omega R^2 C}{1 + (\omega RC)^2} \quad (1)$$

where $\omega = 2\pi f$, is the angular frequency, R is the resistance and C is the capacitance.

The variation of real part of the impedance with frequency and temperature of the $ZrO_2-V_2O_5-TiO_2$

nanocomposite is shown in Fig. 1(a) and (b). The high value of Z' at low frequencies is due to space charge polarization. It is evident that the impedance curves merge at higher frequencies (Fig. 1(a)), denoting that the real part of impedance (Z') is independent of temperature at higher frequencies. This behaviour indicates the dominance of space charge polarization, which occurs at low frequencies and also suggests a reduction in the barrier. The decrease in impedance at higher frequencies may be attributed to the hopping of electrons between localized ions [12]. At peak temperature the impedance values increase with decreasing frequency (Fig. 1(b)). This may be due to the accumulation of charges at grain boundaries.

Fig. 2(a) and (b) show the imaginary part of the impedance (Z'') with respect to frequency. It can be seen that Z'' reaches a maximum at a particular frequency, f_{max} . The maximum frequency (f_{max}) increases with increase in temperature. The maximum value of the impedance decreases as temperature increases. Hence it is evident that the material has a negative temperature coefficient of resistance (NTCR). Here again the impedance curves merge at higher frequencies, irrespective of the temperature. Broadening of the impedance peaks with increasing temperature may be due to a temperature dependent relaxation mechanism.

The effect of temperature on the impedance can be clearly understood from the Nyquist plots shown in Fig. 3(a) and (b). The intercepts of the semicircles on low frequency end gives the bulk resistance (R_b) of the sample at different temperatures [13]. The frequency at the maximum of the semicircle is equal to

$$\omega = 1/RC \quad (2)$$

The relaxation time is given by

$$\tau = RC \quad (3)$$

Therefore

$$\omega\tau = 1 \quad (4)$$

The values of the bulk or grain resistance (R_b) and the corresponding bulk capacitance (C_b) for the sample at different temperatures are given in Table 1. The presence of a single semicircle denotes a deviation from an ideal Debye behaviour [13]. The impedance plots shown can be represented by a parallel R–C combination shown in Fig. 3(a), where R and C represent the bulk resistance and capacitance respectively.

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