

Effect of Fe₂O₃ on the electrical properties of Kaolin

Abdeltif Bouchehema¹, Mohamed Essaleh², Rachid Bouferra², Amal Bourezza², Soufiane Belhouideg¹

¹Research Laboratory of Physics and Engineers Sciences, Team of Applied Physics and New Technologies, Polydisciplinary Faculty (FP-BM), Sultan Moulay Slimane University, Béni Mellal, Morocco

²Laboratoire de Géosciences, Géoenvironnement et Génie Civil. Cadi-Ayyad University, Faculty of Sciences and Technology, B.P.549, 40000, Marrakech, Morocco

Abstract: Impedance spectroscopy is an useful non destructive experimental technique largely used in the electrical characterization of ceramic materials Kaolin is a clay material which is one of the potential ways to support sustainable development in both urban and rural areas. In this context, electrical impedance spectroscopy method with frequency varying between 20 Hz to 1 MHz is used in order to measure electrical complex impedance in a given temperature domain. It permits to estimate the activation and relaxation energies for the dominant electrical conduction mechanisms and also to give an electric picture of the electrical behaviour of the material.

Keywords: Clay materials, Kaolin, Impedance spectroscopy.

1 INTRODUCTION

Very few experimental data of electrical conductivity in the dynamic regime of clay materials are cited in the literature (De Lima O.A.(1992)). In the high temperature range up to 750°C, there is no real investigation on the characterization of the microstructure of clays by studying the temperature and frequency dependence of their electrical conductivity. Impedance spectroscopy is an useful non destructive experimental technique largely used in the electrical characterization of ceramic materials (Bona (2001)). It permits to measure the frequency dependence of the real part, Z' and the imaginary part, Z'' of complex impedance Z^* ($Z^* = Z' + jZ''$ where $j^2 = -1$) at a given temperature T . For an homogeneous material, the Z'' versus Z' diagram (Cole-Cole diagram) is a half circle that can be modeled by a resistance R mounted in parallel with a capacitance C . This capacitance is replaced by the constant phase element (CPE) if there is a distribution of relaxation time. In this paper, we report on the electrical characterization of an industrial kaolin clay mineral providing. Impedance spectroscopy measurements were done with an impedance meter HP 4284A connected to computer, in the frequency range from 20Hz to 1MHz. For this study, the temperature varies between 25 to 750°C. The electric modulus formalism is used in order to identify the predominant microscopic contributions of bulk, grain boundary and electrode polarization effects. To our knowledge, there are no similar experimental data reported for this material in the considered high temperature region. Hence, the aim of this work is to investigate the electrical characteristics of kaolin in continuation to our previous works on disordered semiconducting materials (Bouchehema (2021), Bouferra (2019), Amhil (2019), Essaleh (2017), Essaleh (2018), and Kirou (2019)).

2 EXPERIMENTAL DETAILS

The kaolin considered in this work was collected from different commercial societies. Samples in form of pellets of 13 mm in diameter and 2 mm in thickness using 0.6 g of Kaolin were prepared by uniaxial pressing of 5 MPa and covered on both sides with silver (Ag) electrodes. The various electrical and dielectric parameters (Complex impedance (Z^*), capacitance (C), dielectric losses ($D = \tan \delta$), electrical conductivity (σ), Modulus M^* , etc.) can be measured using an "HP 4284A" impedance meter operating in the frequency domain ranging from 20 Hz to 1 MHz and under an excitation level of the order of (500 mV). The ceramic is placed into a temperature-controlled programmable cylindrical furnace that allows temperatures ranging from 25 °C to 750 °C. A "HP-34401A" type multimeter is used to measure the voltage variation across a K type thermocouple placed close to the sample, in order to follow the evolution of its temperature. The temperature is programmed using a "Eurotherm 2416" type regulator, with a speed of around 5 °C / min. Finally, the whole (impedance-meter, furnace) is controlled automatically by a computer.

3 RESULTS AND DISCUSSION

It is well known that the complex impedance (Z^*) and the electric modulus (M^*) formalisms are usually used to identify and distinguish the contribution of largest resistance from those of smallest capacitance. According to the classical theory of Debye (Barsoukov (2005)), Z^* is given by:

$$Z^* = Z' + jZ'', \quad \text{where } Z' = \frac{R}{1+(\omega\tau)^2} \text{ and } Z'' = -R \left(\frac{\omega\tau}{1+(\omega\tau)^2} \right) \quad (1)$$

and M^* , by:

$$M^* = j\omega C_0 Z^* = M' + jM'', \quad \text{where } M' = \frac{C_0}{C} \left(\frac{(\omega\tau)^2}{1+(\omega\tau)^2} \right) \text{ and } M'' = \frac{C_0}{C} \left(\frac{\omega\tau}{1+(\omega\tau)^2} \right) \quad (2)$$

In these expressions, $C_0 = \frac{\epsilon_0 S}{d}$ is the capacitance of the material in vacuum where S is the area of the electrode, d is the thickness of the sample and ϵ_0 is the permittivity of free space. R and C represent the resistance and the capacitance of the material. The relaxation time τ satisfies the condition $2\pi f_0 \tau = 1$ where f_0 is the frequency at which the curve Z'' versus frequency presents a minimum. This minimum must correspond to a maximum in M'' versus frequency.

The behaviour of the electrical circuits of Resistance-Capacitance ($R-C$) or Resistance-Constant Phase Element ($R-CPE$) type connected in parallel describes the experimental results in semiconducting materials. The complex impedance, defined for a series of values of the frequency, of the current and voltage, can be represented in different diagrams. The impedance spectrum can be represented in two different modes, Nyquist ($Z'' = f(Z')$) and Bode (Z'' and $Z' = f(\text{frequency})$). Bode's representation offers a complete view of the frequency domain, on this representation, the phase shift ϕ as well as the module of the impedance $|Z^*|$ are plotted as a function of frequency. On the other hand, the most used graphic representation concerns the imaginary part $Z'' = \text{Im}(Z^*)$ as a function of the real part $Z' = \text{Re}(Z^*)$ of the complex impedance Z^* . It is to be understood in three dimensions (frequency f , Z' and Z''). These two representations give different visualizations of the same result but they are complementary; each of them shows a particular aspect of the impedance diagram. The Nyquist representation gives detailed information about the studied system, such as relaxation frequency, diameters of semicircles, etc., but sometimes obscures high frequency results while Bode's representation offers a complete view of the domain of frequency, while being less meaningful in identifying certain characteristic phenomena. These two diagrams are illustrated in Fig.1 for a simple RC circuit.

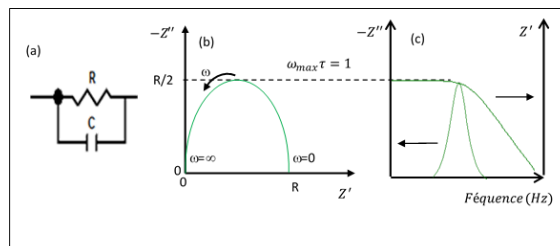


Fig. 1: Simple RC circuit Model (a), Nyquist (b) and Bode (c) diagrams.

Figure 2 show the variation of the imaginary part (Z'') of the complex impedance with frequency for various temperatures for the investigated Kaolin sample. Only one single peak is observed at certain frequency (f_0) suggesting the existence of a single contribution in all samples. These peaks shift towards high frequencies when the temperature increases, indicating the relaxation behaviour. In addition, a peak broadening is noticed with increasing temperature, which suggests the presence of a distribution of relaxation times in this material.

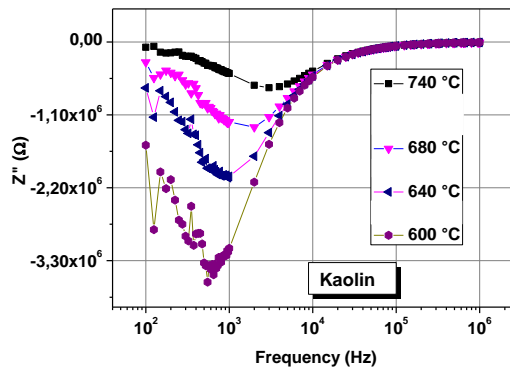


Fig. 2: Z'' as a function of frequency of Kaolin for some representative temperatures.

On the other hand, impedance spectroscopy measurements were done by mixing the pure kaolin with Fe_2O_3 in different percentage from 10% to 100%. Similar curves with Fig.3 were obtained (not shown here) for each percentage. The analysis of these data permits us to analyse the conduction and the relaxation behaviour of the synthesized composite and to give an electric picture of the system.

4 CONCLUSIONS

Some new experimental data of impedance spectroscopy are presented in this work for the Kaolin clay material. The data show a relaxation behaviour. The data indicates the predominance of the grain contribution to the electrical conduction and the distribution of the relaxation time in the system. More analysis is needed to identify the dominant conduction mechanism of polaron and carriers tunnelling in Fe_2O_3 doped kaolin.

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