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DIELETRIC ANALYSIS OF UNSATURATED POLYESTER COMPOSITE REINFORCED WITH RECYCLED COTTON TEXTILE RESIDUES

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ABSTRACT

Dielectric measurements were performed on unsaturated polyester matrix and its composite reinforced with non-woven cotton fibers. These latter were elaborated from recycled cotton residues generated throughout manufacturing textile processes. Two kinds of cotton wastes were used in the reinforcement: the white yarns and the indigo denim fabrics in the relative volume fraction 1:3. The composite material was elaborated using the classical contact mould method in the Solutions composites Company in Zaghouan. Dielectric spectra were measured in the frequency range from 0.1 Hz to 10^6 Hz and temperature interval 0°C to 150°C. Two dielectric relaxation processes were identified for the composite. Analysis of the interfacial relaxation by means of the Havriliak-Negami model allowed detecting the α mode relaxation. Comparing this relaxation to that appearing in the neat resin matrix evidenced the existence of the fibers/matrix interactions.

KEYWORDS: Biocomposite, recycled cotton, Unsaturated polyester, Dielectric relaxation.

1. INTRODUCTION

Bio-composites formed by bio-fibers and synthetic polymer matrix are partially biodegradable. Their growing areas of applications are in automotive parts, housing products and packaging. The challenge in replacing the conventional glass-reinforced plastics with bio-composites is to design materials that exhibit structural and functional stability during storage and use [Amar et al, 2005]. The potential of bio-composites consists in delivering a same performance as conventional composites with lower weight. Moreover, they exhibit non-brittle fracture on impact, which is another important requirement [Amar et al, 2005]. In the automotive industry, textile waste has been used for years to reinforce plastics used in cars [Sharma et al., 2017]. Cotton fibers have been used by early societies to produce textile [Amar et al, 2005]. Despite of their mechanical properties difference in comparison with other plant fibers, innumerable products are made from cotton, primarily textile and yarn goods, cordage and automobile tire cords [Amar et al, 2005]. Cotton fibers are the backbone of the textile trade of the world [Amar et al, 2005]. They are the most important textile vegetable fibers. A recent research project done on the composite panels reinforced with cotton fibers [Sharma et al., 2017, Koronis et al., 2017, Nassar et al., 2016, Nassar et al., 2017, Pickering et al., 2016, Robertson et al., 2013] revealed that the structural performance of cotton fiber composites is satisfactory for structural parts with low requirements, such as wall panels or doors [Raftoyiannis, 2012]. In this research study, it was evidenced that specimens with treated or untreated cotton fibers have similar property values with specimens with cotton textile [Raftoyiannis, 2012].

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The aim of the present study is to promote recycled cotton textile residues as reinforcement in plastic composites. These residues were provided by Society of Textile Industries (SITEX). Two kinds of textile wastes were used in the conception of the non-woven cotton fibers: the white yarns and the indigo denim fabrics. Unsaturated polyester (UP) resin was chosen as a matrix for the composite materiel because of its relatively low price, ease of handling and its good balance of mechanical, electrical and chemical properties [Amar et al, 2005]. In this work frame, dielectric measurements were performed on the UP matrix and its composite so that to evidence the existence of fibers/matrix interactions which control the mechanical performance of the composite.

2. MATERIALS AND EXPERIMENTAL TECHNIQUE

2.1 Materials

The matrix material used in this study was based on a commercially available unsaturated polyester, whose Trade Name is 'ENYDYNE® H 68372 TA' supplied by Polynt/Composites. The matrix was mixed with initiator Methyl ethyl Ketone peroxide, MEKP, and Cobalt octanone at a concentration of 1.5 % w/w before introducing the reinforcement.

Textile wastes cotton made of white yarns and indigo denim fabrics in the relative volume fraction 1:3 and supplied by the SITEX were used as reinforcement for the composite manufacturing. Textile wastes were chopped into small pieces with a length between 5 cm and 10 cm before using a fraying machine to transform these wastes into fibers. A needle punching machine in the laboratory of SITEX Company was then applied to the obtained frayed cotton fibers twice in order to transform them into nonwoven structure.

The composite was manufactured in the Solutions composites Company using the classical 'contact mould method' [Kaynak, Akgul, 2001]. Fibers were deposited on the mould and impregnated with the liquid resin mixed with suitable proportions of Methyl ethyl Ketone peroxide and Cobalt octanone as hardener and catalyst, respectively. The saturated material was then pressed by a roller to remove bubbles. After the hardness of the resin, the composite was withdrawn from the mould. The composite obtained had 3 % fiber weight fraction.

2.2 Experimental technique

Dielectric measurements were carried out with an Alpha Dielectric/impedance Analyser (Novocontrol) in the temperature range from 0°C to 150 °C and a frequency interval from $10^{-1}$to 10^{6} Hz. In this study, the sample was heated from the ambient up to 150°C. Then measurements were performed on the sample during the cooling process. In the dielectric characterization, the sample was placed between two gold parallel electrodes. A sinusoidal voltage was applied creating an alternating electric field. This produced polarization in the sample, which oscillated at the same frequency as the electric field, but had a phase angle shift δ. This phase angle shift was measured by comparing the applied voltage to the measured current, which was separated into capacitive and conductive components.

Measurements of capacitance and conductance were used to calculate:

• Real part of the permittivity (apparent permittivity) ε' which is proportional to the capacitance and measures the alignment of dipoles.

• Imaginary part of permittivity (loss factor) ε'' which is proportional to the conductance and represents the energy required to align dipoles and move ions.

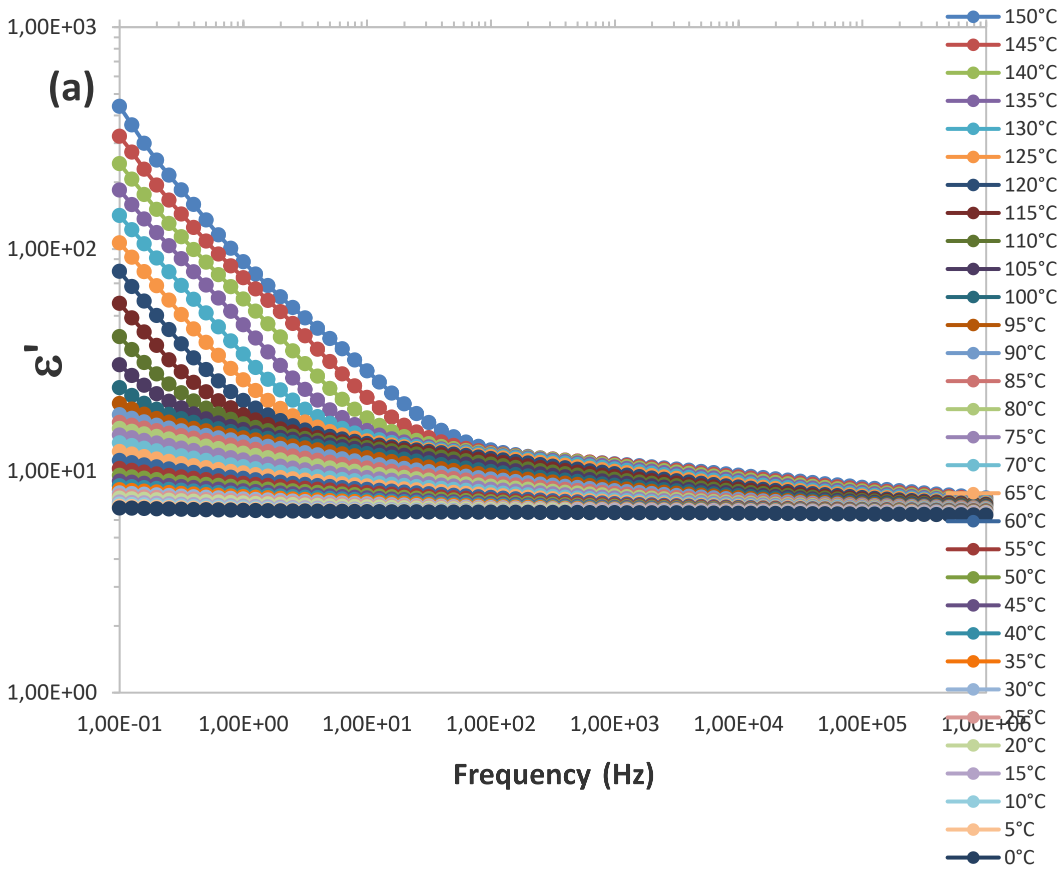
Accordingly, the complex permittivity was given by the following equation:

$$\varepsilon^* = \varepsilon' - j\,\varepsilon'' \qquad (1)$$

Dielectric experiments were conducted in isothermal runs with fixed temperatures and scanning frequencies from 10^{-1} to 10^{6} Hz.

3. RESULTS AND DISCUSSION

Figure 1-(a, b) illustrates the isothermal runs of the dielectric permittivity, ε', for the matrix and its composite (C1), respectively. An overall increase with temperature at low frequencies and a decrease of the behavior with increasing frequency were observed.



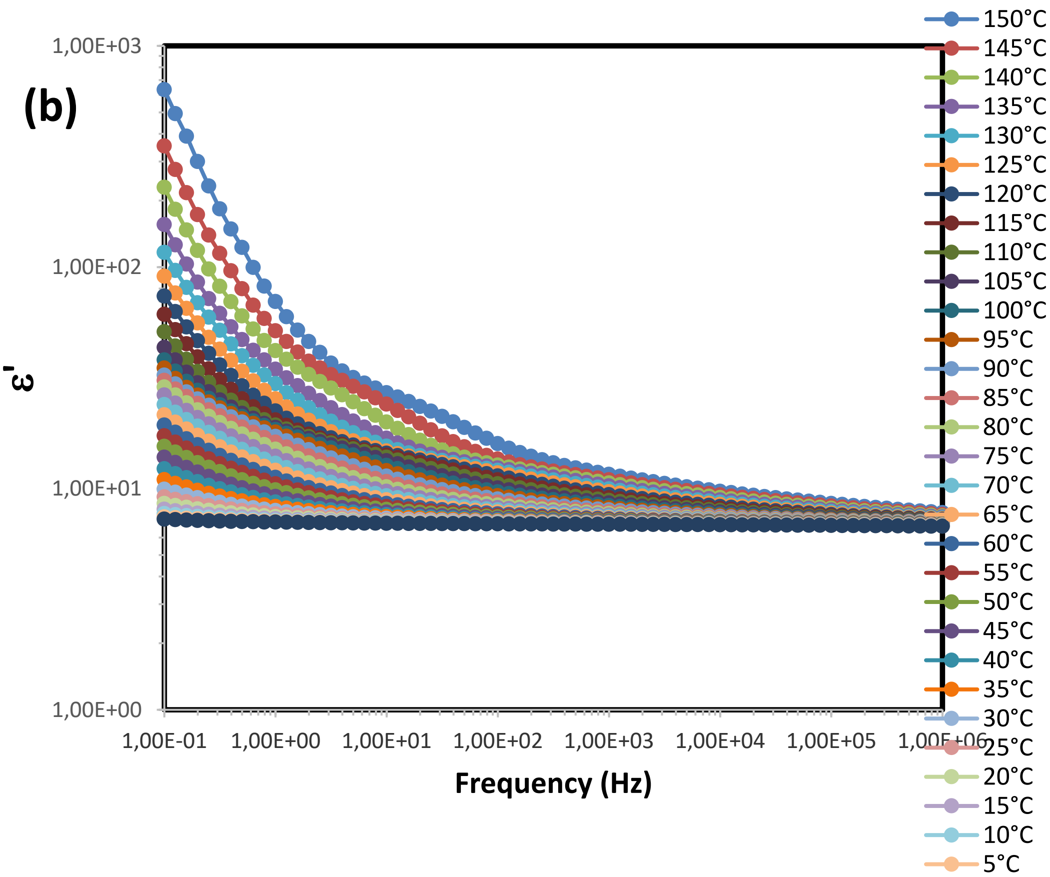


Figure 1: Isothermal runs of the dielectric permittivity for the matrix (a) and its composite (b).

Analysis of the isothermal runs of the loss factor, ε'', didn't reveal any dielectric relaxations either at low temperatures or at high ones as depicted in figure 2. Hence, water dipoles polarization was absent in the case of the composite C1. The heat treatment adopted before carrying out the dielectric characterization allowed removing the hydrophilic character of plant fibers [Arous et al., 2007]. Nevertheless, these isothermal runs showed a -1 slope at low frequencies and high temperatures which is a typical characteristic of dc-conductivity effect.

To minimize the effect of the dc conductivity, the formalism of ''electric modulus'' is introduced. The electric modulus M^* is defined by the following equation [Omri et al., 2016] :

$$M^* = \frac{1}{\varepsilon^*} = \frac{1}{\varepsilon' - j\varepsilon''} = \frac{\varepsilon'}{\varepsilon'^2 + \varepsilon''^2} + j\frac{\varepsilon''}{\varepsilon'^2 + \varepsilon''^2}$$
$$= M' + jM'' \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots(2)$$

Where M' and M'' are the real and imaginary parts of the electric modulus, respectively.

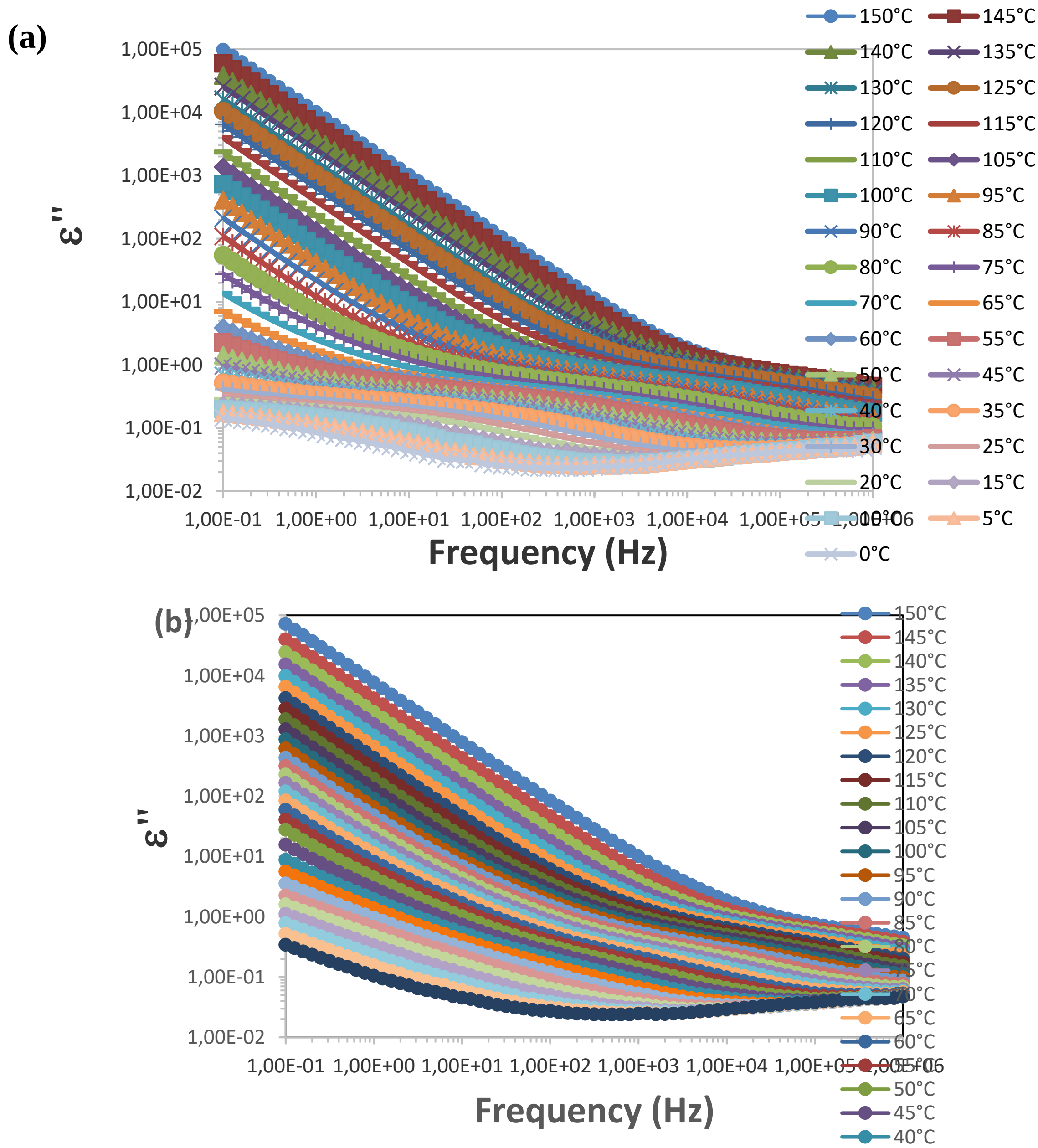
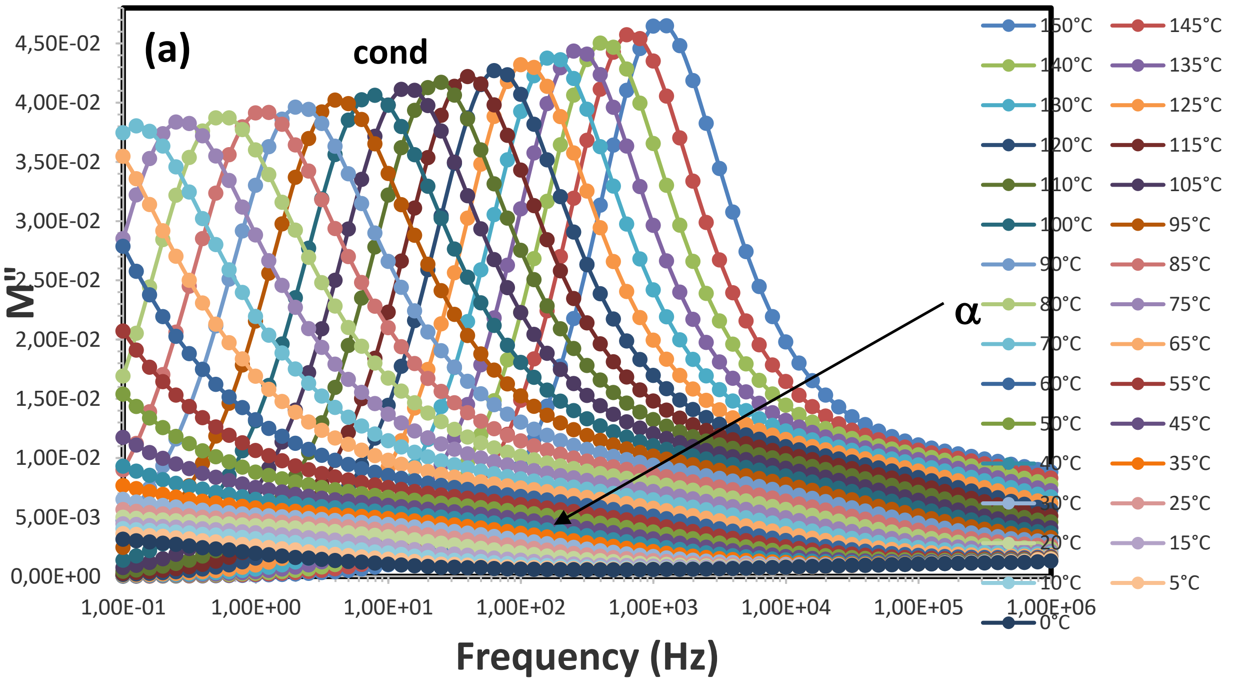


Figure 2: Isothermal runs of the loss factor for the matrix (a) and its composite (b).

Figure 3-(a, b) shows isothermal runs of the behavior of M'' as a function of frequency for the matrix UP and its composite C1, respectively, during the cooling process over the temperature range from 150 to 0°C. A slight shoulder can be noticed at intermediary frequency and temperature ranges for the matrix. This shoulder was identified to the α mode relaxation associated with the glass transition of the UP resin matrix. A second dielectric relaxation appearing at high temperatures was attributed to the dc conductivity which occurs as a result of the carriers' charges diffusion.



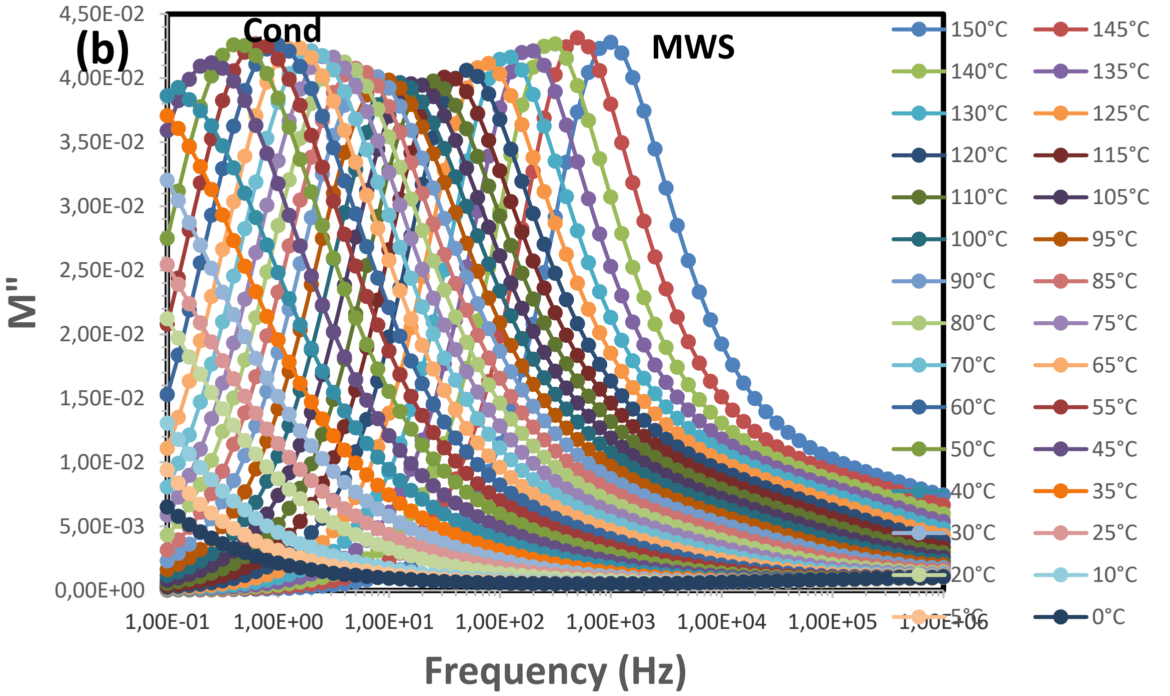


Figure 3-(a, b): The isothermal runs of the imaginary part M'' of the electric modulus versus frequency for the UP matrix (a) and its composite C1 (b).

The incorporation of the nonwoven sheet of cotton fibers into the matrix led to the appearance of another dielectric relaxation which was associated with the interfacial or Maxwell-Wagner-Sillars (MWS) relaxation that was attributable to the accumulation of charges at the cotton fibers/polyester resin interfaces. The α relaxation, which was already seen in the UP resin, was completely masked by the MWS relaxation in the case of the composite C1.

To further support the dielectric relaxation identifications, the α and MWS relaxations for the UP matrix and its composite C1, respectively, were analyzed according to the Havriliak-Negami model given by the following equation :

$$\varepsilon^* = j\left(\frac{\sigma_o}{\omega\,\varepsilon_o}\right)^N + \sum_{k=1}^{2}\left[\frac{\Delta\,\varepsilon_k}{(1+(j\,\omega\,\tau_k)^{\alpha_k})^{\beta_k}} + \varepsilon_{\infty k}\right] \tag{3}$$

Where, N is the positive exponential factor which is inferior or equal to 1, σ_o.is the dc-conductivity, $\omega = 2\,\pi$ frequency, $\Delta\,\varepsilon_k$ is the strength of the dielectric relaxation k whose maximum is located at relaxation time τ_k. α_k and β_k are fractional shape parameters describing the skewing and broadening, of the dielectric function, respectively.

Argand's plots in electric modulus of the UP matrix and its composite C1 at 95°C are depicted in figure 4-(a, b).

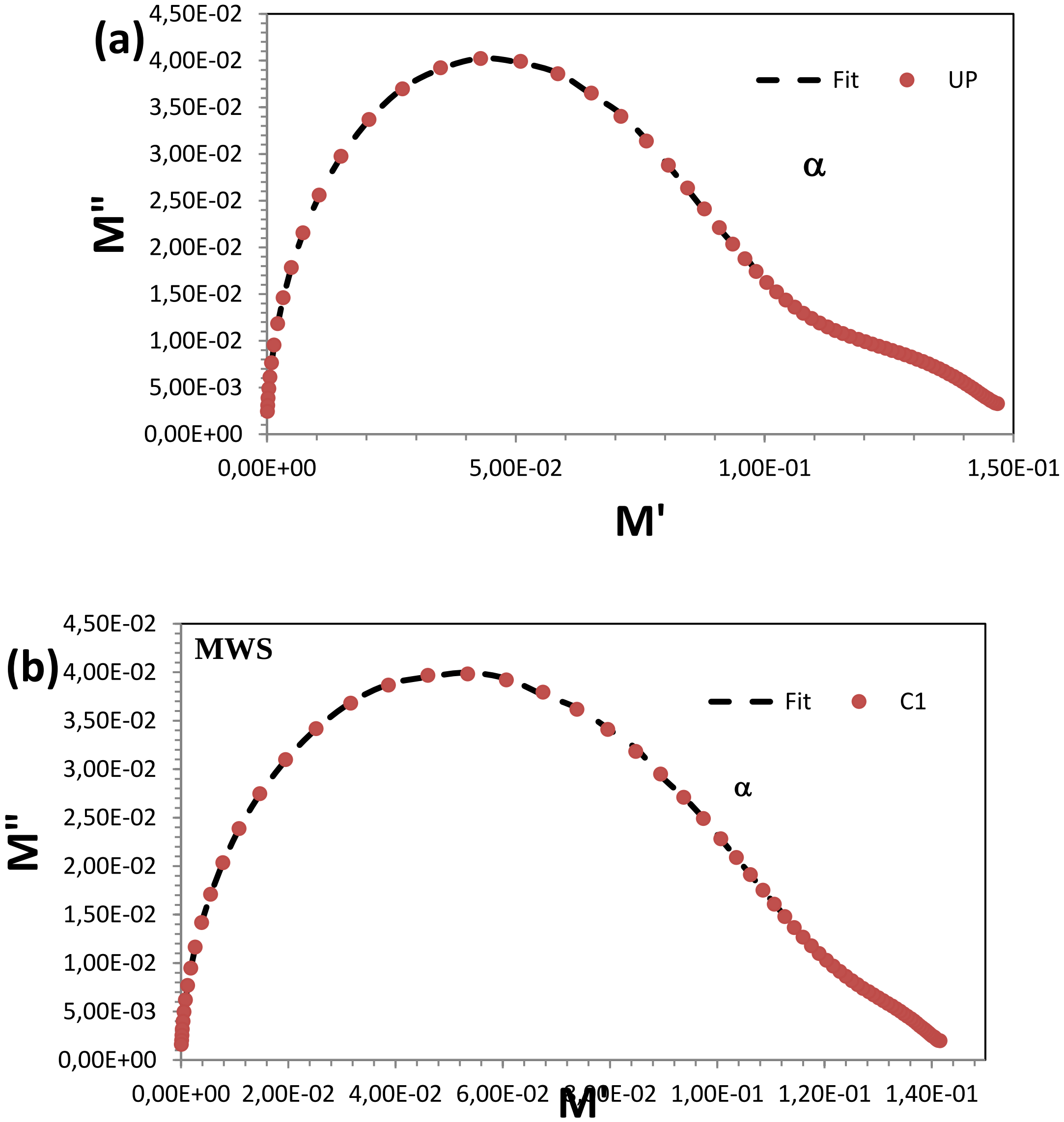


Figure 4 : Argand's plot of the electric modulus, M* for the UP matrix (a) and its composite C1 (b).

The Cole-Cole diagram of the UP resin corresponds to the α relaxation. Nevertheless, for the composite C1, we note that it was impossible to fit to the Havriliak-Negami model with all the experimental points taking into account the MWS relaxation. Hence, a second dielectric relaxation is detected originating from the α relaxation. Adjustment parameters obtained for UP matrix and the composite C1 at each temperature allowed determining the activation energy of the α relaxation for the matrix which decreases when the reinforcement was introduced in the resin. Such result reveals the existence of interactions at cotton fibers/matrix interfaces. Further, the α relaxation strength for the composite C1 is lower than that for the matrix UP. Such decrease can be explained by the effect of the interfacial relaxation on the α relaxation which confirms the adhesion of the non-woven cotton fibers in the UP matrix.

4. CONCLUSION

Dielectric analysis of the UP matrix and its composite reinforced with non-woven frayed cotton fibers revealed that a heating treatment allowed analyzing the α dielectric relaxation in both samples. The comparison of the obtained results evidenced a decrease of either its activation energy or its dielectric strength when introducing the reinforcement in the matrix. These tendencies were attributed to the existence of fibers/matrix interactions.

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