



## Dielectric analysis of the interfacial polarization of alkali treated woven flax fibers reinforced epoxy composites



A. Triki <sup>a,\*</sup>, M. Karray <sup>b,d</sup>, C. Poilâne <sup>c</sup>, P. Picart <sup>d</sup>, M. Gargouri <sup>b</sup>

<sup>a</sup> Laboratoire des matériaux composites, polymères et céramiques, Faculté des Sciences de Sfax, Route Soukra, 3018, Tunisia

<sup>b</sup> Unité de l'état solide, Faculté des Sciences de Sfax, Route Soukra, 3018, Tunisia

<sup>c</sup> CIMAP, UMR6252 CNRS-CEA-ENSICAEN-Université de Caen Basse-Normandie, F-14032, Caen, France

<sup>d</sup> LAUM UMR CNRS 6613, École Nationale Supérieure d'Ingénieurs du Mans, Université du Maine, Av. O. Messiaen, F-72085, Le Mans, France

### ARTICLE INFO

#### Article history:

Received 26 February 2015

Accepted 11 May 2015

Available online 23 May 2015

#### Keywords:

Laminates

Flax fibers

Dielectric relaxation

Adhesion

### ABSTRACT

Dielectric measurements were performed on alkali treated flax fibers reinforced epoxy composites in the frequency range 0.1 Hz–1 MHz and the temperature range 40–170 °C. Two common dielectric relaxations were observed for all composites. The first one appearing at low temperatures was attributed to the water dipoles polarization due to the hydrophilic character of flax fibers. The second one observed at high temperatures was identified to the interfacial polarization. Analysis of this latter using the Havriliak–Negami model showed an improvement of the fibers/matrix adhesion by these alkali treatments. The best adhesion was obtained by the leaching treatment.

© 2015 Elsevier B.V. All rights reserved.

### Introduction

Natural fiber-reinforced composites which are environmental friendly materials have been studied extensively [1–4] for environmental and economic considerations. Many natural fibers have been considered as reinforcement in structural composites such as flax, jute, bamboo, sisal, hemp, ramie, abaca, kapok, etc. However, despite of the numerous advantages characterizing natural fibers [5–13] their incorporation in polymeric matrix isn't without challenges. Indeed, their hydrophilic character makes them incompatible with the hydrophobic polymeric matrix which results in weak interface in composite material. Such poor interfacial adhesion can be improved by pretreatments of natural fibers. These latter can clean the fiber surface, modify the surface chemically, stop the moisture absorption process, and increase the surface roughness [14]. The effect of these treatments on the fiber/matrix interfacial adhesion was evidenced by many techniques. The ones mostly used are dynamic mechanical analysis and scanning electron microscopy (SEM) observation.

The dielectric spectrometry is an additional technique that can be used to probe the composite interface and investigate the effect of fiber treatment on the evolution of composites' interfacial

properties [15–18]. Indeed, when an electrical field is applied across a parallel-plate capacitor containing a dielectric material, the various atomic and molecular charges present in the dielectric are displaced from their equilibrium positions and the material is said to be polarized. Different polarization mechanisms can occur, including dipole orientation, extrinsic free charges or intrinsic charge migration, electrode polarization and the interfacial or Maxwell–Wagner–Sillars (MWS) polarization in the case of heterogeneous or composite system [19,20]. The MWS polarization is attributable to the accumulation of charges at interfaces in composite materials. The analysis of this relaxation using the Havriliak–Negami model and calculation of its intensity allows probing the interfacial region.

The purpose of this work is to analyze the dielectric relaxation of woven flax fibers reinforced epoxy composites so as to study the effect of the reinforcement alkali treatment (e.g. leaching, bleaching and mercerization), used in textile industry to clean fabrics, in interfacial region of the composites.

### Experimental

#### Materials

Flax fibers fabric/epoxy resin prepregs have been supplied by the LINEO NV Company [21]. The fabric of flax fibers was made with yarns coming from the flax semiwet spinning. These yarns were

\* Corresponding author.

E-mail address: [trikilamacop@yahoo.fr](mailto:trikilamacop@yahoo.fr) (A. Triki).

woven in the form of 2/2 twill fabric [22]. The obtained fabric was a subject of pretreatments before impregnation by the epoxy resin. In this study three pre-treatments were used. The first one was the leaching treatment done with a detergent. This pre-treatment was an alkaline boiling off. The second one was the bleaching treatment done with a combination of hydrogen peroxide and sodium hypochlorite. The third one was the mercerization done with sodium hydroxide. These pretreatments are classical alkali treatments used in the textile industry to clean fabrics and thus facilitate the finishing process [23]. To optimize the impregnation of these fabrics with the epoxy resin, they undergo a compatibilization treatment patented by LINEO NV [24]. Finally, the fabrics are impregnated by an epoxy resin especially formulated for prepregs (Hunstman Araldite LY5150/Aradur 5021/Hardener XB 3471) [25]. The composite materials were elaborated by means of a hot platen press [26]. Laminates were produced with seven layers of prepreg fabrics for hot platen press. The obtained volume fractions are about 50%.

### Measurement

Dielectric measurements were conducted with an Alpha dielectric-impedance analyzer (Novocontrol), with the measurements of the studied samples taken over the temperature range from 40 to 170 °C and in a frequency interval from  $10^{-1}$ – $10^6$  Hz. A circular gold electrode (2 cm in diameter) was sputtered on both surfaces of the sample to ensure good electrical contact with the gold-plated measuring electrodes. A sinusoidal voltage was applied to create an alternating electric field that produced polarization in the sample, which oscillated at the same frequency as the electric field but had a phase angle shift ( $\delta$ ). This phase angle shift was measured by the comparison of the applied voltage with the measured current, which was divided into capacitive and conductive components. With the following equations [27], the dielectric parameters were calculated:

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

$$\varepsilon' = \frac{C_p(\text{sample}) d}{A \varepsilon_0} \quad (2)$$

$$\varepsilon'' = \frac{G(\text{sample}) d}{\omega A \varepsilon_0} \quad (3)$$

where ( $j^2$ ) = -1;  $\varepsilon'$  and  $\varepsilon''$  are the real and imaginary parts of the complex permittivity ( $\varepsilon^*$ ); A and d are the area and thickness, respectively, of the sample;  $C_p$  is the capacitance; G is the conductance; and  $\varepsilon_0$  is the permittivity of the free space and is equal to  $8.854 \times 10^{-12}$  F/m. Dielectric experiments were carried out with fixed temperatures and scanning frequencies from  $10^{-1}$ – $10^6$  Hz.

### Results and discussions

Comparative plots of the frequency dependence of the dielectric permittivity ( $\varepsilon'$ ) in the untreated (UWFFRE), leached (LWFFRE), bleached (BWFFRE) and mercerized (MWFFRE) woven flax fibers reinforced epoxy composites for different temperatures from 40 to 170 °C in increments of 20 °C are shown in Fig. 1(a–d). An overall increase in  $\varepsilon'$  with temperature at low frequencies and a decrease of the behavior with increasing frequency were observed. For low temperatures, the enhancement of the dielectric permittivity  $\varepsilon'$  at low frequencies is attributed to the water dipoles polarization. This enhancement remained the same after the bleaching treatment whereas it decreased after leaching and mercerization treatments.

As temperature increases,  $\varepsilon'$  increases due to the enhanced conductivity of the composites at higher temperatures. Fig. 2(a–d) illustrate the isothermal variation of the loss factor ( $\varepsilon''$ ) for the UWFFRE, LWFFRE, BWFFRE and MWFFRE composites, respectively. At low frequencies and low temperatures the loss factor  $\varepsilon''$  shows a decreasing tendency with increasing the frequency for the UWFFRE composite. A similar behavior was observed for the isothermal curves of the dielectric permittivity  $\varepsilon'$  at the same temperatures and frequencies ranges. This phenomenon presents a situation of low frequencies dispersion which is a characteristic of the systems dominated by the carriers as mentioned in Refs. [28,29]. This dispersion mechanism was explained according to the model of cluster [30]. Indeed, the existing cluster in heterogeneous material can be regarded as a site containing charge carriers. This dispersion mechanism was enhanced by the bleaching treatment but lowered by the leaching and the mercerization treatments. For all composites, -1 slope of the loss factor  $\varepsilon''$  in the low frequencies and high temperatures region is typical for dc conductivity.

Electric modulus formalism ( $M^*$ ) was introduced in order to minimize the effect of this dc conductivity. This  $M^*$  has recently been adapted for the investigation of dielectric processes occurring in composite polymeric systems and those proposed for the description of systems with ionic conductivity<sup>15</sup>.  $M^*$  is defined by Eq. (4) [31]:

$$M^* = \frac{1}{\varepsilon^*} = \frac{1}{\varepsilon' - j\varepsilon''} = \frac{\varepsilon'}{\varepsilon'^2 + \varepsilon''^2} + \frac{\varepsilon''}{\varepsilon'^2 + \varepsilon''^2} \quad (4)$$

where  $M'$  and  $M''$  are the real and imaginary parts of the electric modulus, respectively. An advantage of using  $M^*$  to interpret bulk relaxation properties is that the variation in the large values of the real part of the permittivity and the loss factor at low frequencies are minimized. In this way, common difficulties of the electrode nature and contact, space-charge-injection phenomena, and absorbed impurity conduction effects, which appear to hide the relaxation in the permittivity representation, can be solved or even ignored [32]. Fig. 3(a–d) show the variation of  $M''$  as a function of frequency for the UWFFRE, LWFFRE, BWFFRE and MWFFRE composites when they were heated over the temperature range from 40 to 170 °C.

A series of two common distinct relaxations could be considered for all composites. The first one was related to the water dipoles relaxation which appeared at low temperatures. At high temperatures, the second relaxation was attributed to the interfacial polarization effect. An additional relaxation associated with the dc conductivity effect has been observed only for the UWFFRE and BWFFRE composites at high temperatures. Nevertheless, one can notice the absence of the  $\alpha$  relaxation in all composites which is hidden by the MWS polarization effect. To further support these assignments, the activation energy ( $E_a$ ) relative to the different relaxations was evaluated with the following Arrhenius relation:

$$f_{\max} = f_0 \exp\left(-\frac{E_a}{k_B T}\right) \quad (5)$$

where  $f_{\max}$ ,  $k_B$ ,  $T$ ,  $E_a$  and  $f_0$  are frequency associated with the maximum of  $M''$  for a fixed temperature, boltzmann constant, temperature, activation energy of the process and frequency at high temperature. Fig. 4 shows the evolution of  $\log f_{\max}$  versus  $1/T$  for each one of the different observed relaxations.  $E_a$  and  $f_0$  were extracted from the slopes and the intercepts of the plots of  $\log f_{\max}$  versus  $1/T$ . The relaxation time  $\tau_0 = \left(\frac{1}{2\pi f_0}\right)$  is then calculated for each dielectric relaxation. Analysis of the activation energy values related to the interfacial polarization effect (Table 1) has revealed their alterations after these alkali treatments. This indicated

Download English Version:

<https://daneshyari.com/en/article/724211>

Download Persian Version:

<https://daneshyari.com/article/724211>

[Daneshyari.com](https://daneshyari.com)