Electrical Conductivity of Wood-polymer Composites

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This paper presents a study of the electrical conductivity of wood-polymer composites, determined by measurements done in DC and AC current. It is also noticed that the values of $\sigma_{dc}$, $\sigma'$ and $\sigma''$ are strongly influenced by the nature of matrix and filler content of the composite. Finally, it is shown that samples of PE and PP matrices can be used for applications in power frequency electric fields (0 ... 100 Hz) and at temperatures between 30 and 90 °C.

Keywords: compozite lemn-polimer, conductivitate electrica

The electrical conduction in an insulator, respectively the orderly displacement of charge carriers (electrons, holes, ions etc) in relation to the body depends on both the concentration and their respective charge, as well as the chemical nature and physical structure of the body, the characteristics of the environment (temperature, humidity etc.), the electric field strength and frequency etc. [1]. The electrical conductivity $\sigma$ of an insulator can be determined with the relation

$$\sigma = \sum N_k q_k \mu_k,$$  \hspace{1cm} (1)

where:

- $N_k$ is the charge carriers concentration;
- $q_k$ – the charge;
- $\mu_k$ – the mobility of charge carriers of species $k$ (ions, molions, electrons etc).

Although the conduction mechanism for metals is relatively simple (the displacement of free electrons within permitted bands), in the case of insulating bodies (amorphous or partially amorphous), the electrical conduction mechanisms are more complex, considering hopping conduction, polaron movement, impurities effect etc. The problem is much more complex for composites [2] and, especially, for wood-polymer composites, which are inhomogeneous and anisotropic materials with low electrical conductivity. The high water absorption characteristic of wood together with space charge accumulation on wood-polymer interfaces result in a far different current variation curves than those found in the case of pure polymer samples [3]. In time variable electric fields, these currents are also strongly influenced by the polarization mechanisms characteristic to inhomogeneous dielectrics analyzed in [4].

Direct current conductivity

When a step voltage $U_0$ is applied to a capacitor (with electrodes of surface $A$ and the dielectric of thickness $g$), the absorption current that appears $i_1(t)$ has four components (fig. 1):

$$i_1(t) = i_{ch}(t) + i_p(t) + i_{sc}(t) + i_c(t),$$  \hspace{1cm} (2)

where:

$\ i_{ch}(t)$ is the charging current of the capacitor with vacuum dielectric;
$\ i_p(t)$ is the polarization current;
$\ i_{sc}(t)$ - the space charge current;
$\ i_c(t)$ - the conduction current.

The current $i_{ch}(t) = \varepsilon_0 \frac{dE}{dt}$ is due to the charging of a capacitor with vacuum dielectric (with permittivity $\varepsilon_0$) and quickly drops to zero. As a consequence, in the case of usual measurements, the first part of curve $i_1(t)$ (fig. 1) is not recorded [5].

![Fig. 1. Time variation of the current $i$ in a capacitor dielectric for a step applied voltage ($i_{ch}(t)$) and after the supply disconnection and electrode short-circuit ($i_{sc}(t)$).](image)

The component $i_p(t)$ is given by the dielectric polarization phenomena (electronic, ionic, orientation, interfacial) that consist in very small motions of a large number of attached charges [4]. When the voltage is switched off, these charges return to their initial positions [6-7].

The current $i_{sc}(t)$ corresponds to the movement of the existing space charge in the dielectric volume that can be generated by the technological processes (molecules' fractures etc.), degradation process during the work service (thermal, electric, mechanic and radiation etc.) and charge injection at the surface of the electrodes with sharp edges and/or protuberances [8]. In a certain time
depending on dielectric properties, the currents \( i_{\text{d}}(t) \) and \( i_{\text{r}}(t) \) become zero.

The conduction current \( i(t) = I_c = A \times \sigma \times U_0 g \) (fig. 1) is given by the convection of electrons, ions and molions. The concentration of each charge type depends on the chemical nature and physical structure of dielectric and gives an electronic or ionic preponderant conduction [1]. This component of the current \( i(t) \) is unchanged in time (from the voltage application to its switch-off) and allows the determination of the electrical conductivity of dielectric. The charge carriers come from electrode injections, band-band tunneling, impurities ionization etc. On the other hand, the charge carrier concentration also depends on the humidity etc., and is correlated with the dielectric ageing [1].

If at moment \( t \), the voltage is switched off (\( U_0 = 0 \)) and the capacitor electrodes are connected in short-circuit, the dielectric is passed by a transitory current (resorption current) \( i_{\text{r}}(t) \):

\[
i_{\text{r}}(t) = i_{\text{d}}(t) + i_{\text{d}'}(t) + i_{\text{r}'}(t),
\]

where:

- \( i_{\text{d}}(t) \) is the discharge current of the vacuum dielectric capacitor;
- \( i_{\text{d}'}(t) \) - depolarization current;
- \( i_{\text{r}'}(t) \) - the current that corresponds to the space charge.

As long as the voltage is applied, a part of the charge carriers which generate the space charge current \( i_{\text{r}'}(t) \) may become fixed in other potential traps, or, if they reach the plates of the capacitor, are neutralized. Therefore, \( i_{\text{r}'}(t) \) is lower than \( i_{\text{r}'}(t) \). On the other hand, because the space charge and the separable charge at interfaces between the homogeneous regions are reduced and, so, the interfacial polarization and its corresponding current, it results \( i_{\text{d}} < i_{\text{d}'} \).

From equations (1) and (2), the time dependent direct current conductivity \( \sigma_{dc}(t) \) can be defined as:

\[
\sigma_{dc}(t) = \frac{\varepsilon_0}{C_0 U_0} [i_{\text{d}}(t) + i_{\text{z}}(t)],
\]

where:

- \( C_0 = \frac{\varepsilon_0 A}{g} \) is the capacitance of the vacuum capacitor.

Because the charging \( i_{\text{d}}(t) \) and discharging \( i_{\text{d}}(t) \) currents drop to zero very quickly, it results:

\[
\sigma_{dc}(t) = -\frac{\varepsilon_0}{C_0 U_0} [i_{\text{d}}(t) + i_{\text{z}}(t) + I_c + i_{\text{d}'}(t) + i_{\text{r}'}(t)].
\]

From equation (5) results that \( \sigma_{dc}(t) \) has two components: a time dependant one \( \sigma_{dc}(t) \), corresponding to the polarization and space charge currents and a constant one \( \delta \), corresponding to the permanent conduction current \( I_c \) (called direct current constant conductivity):

\[
\sigma_{dc}(t) = \sigma_{dc}(t) + \delta
\]

\[
\sigma_{dc}(t) = -\frac{\varepsilon_0}{C_0 U_0} [i_{\text{d}}(t) + i_{\text{z}}(t) + i_{\text{d}'}(t) + i_{\text{r}'}(t)]
\]

\[
\delta = \frac{\varepsilon_0}{C_0 U_0} I_c
\]

Evidently, if the duration \( t \) of the voltage application is long enough, so that \( i_{\text{d}} = 0 \) and \( i_{\text{r}'} = 0 \), \( \sigma_{dc} \) can be determined from the following equation:

\[
\sigma_{dc}(t) = \frac{\varepsilon_0}{C_0 U_0} [i_{\text{d}}(t) + i_{\text{z}}(t)].
\]

Alternative current conductivity

Starting with the year 1970, variations of electric conductivity with temperature and frequency were presented in a series of papers (especially for different types of glass and flawed crystals) and a series of conduction models in alternative current were proposed (such as the hopping conduction) [9…12]. Modern frequency analyzers allow alternative current (ac) conductivity measurements in amorphous [13] and organic [14] semiconductors, polycrystals [15], conductive polymers [16-17], wood [18], bamboo [19], carbon black styrene-butadiene conductor composites [20] etc.

The experimental results are usually given by the real and imaginary part of the complex conductivity \( \sigma^*(\omega) = \sigma^r(\omega) + j \sigma^i(\omega) \), defined in relation to the complex permittivity \( \varepsilon^*(\omega) = \varepsilon^r(\omega) + j \varepsilon^i(\omega) \) by the relation:

\[
\sigma^*(\omega) = j \omega \varepsilon_0 \varepsilon^r(\omega),
\]

where:

- \( j = \sqrt{-1} \), \( \varepsilon^r(\omega) = \varepsilon_0 \varepsilon^r(\omega) = \varepsilon_0 [\varepsilon^i(\omega) - j \varepsilon^i(\omega)] \), \( \varepsilon^r(\omega) \) is the real part of the complex permittivity; \( \varepsilon^i(\omega) \) is the imaginary part of the complex relative permittivity; \( \omega = 2\pi f \) - the pulsation and \( f \) – the frequency of the applied electric field [21-22].

From equation (10) it results:

\[
\sigma(\omega) = \omega \varepsilon_0 \varepsilon^i(\omega)
\]

\[
\sigma^i(\omega) = \omega \varepsilon_0 \varepsilon^r(\omega),
\]

where:

- \( \sigma^r \) is the real part of the complex conductivity \( \sigma^r \);
- \( \sigma^i \) - the imaginary part of the complex conductivity;
- \( \varepsilon^r / \varepsilon^i \) - the dielectric loss factor.

The alternative current conductivity \( \sigma_{ac} \) corresponds to the dissipated energy during dielectric relaxation processes and is not related to the direct current constant conductivity \( \sigma_0 \) (which corresponds to electric charge displacement). It can be written:

\[
\sigma_{ac}(\omega) = \sigma^r(\omega) - \sigma_0.
\]

In this paper, the time dependent conductivity values \( \sigma_{ac}(t) \) as well as the direct current constant conductivity \( \sigma_0 \) were determined by measuring absorption and resorption currents for a duration \( t = 7200 \) s. The values of the complex conductivity components \( \sigma^r \) and \( \sigma^i \) were determined based on the complex relative permittivity \( (\varepsilon^r) \) components experimentally determined [4, 22].

Experimental part

Materials. Samples

The experiments were performed on compound wood-polymer composites made up of wood particles (wood flour) enclosed in a thermoplastic polymer matrix (polyethylene, polypropylene).
The wood flour (used as reinforcement) was obtained by the grinding of the sawdust resulted from hardwood processing, with a medium particle size of 0.4 mm (table 1) [4].

High-density polyethylene (HDPE) and polypropylene (PP) powders were used for the matrix (table 2). Vinyl acetate was added (as a compatibility agent) with 0.1% nonanoil peroxide, 10% of the wood flour mass.

The compounding process used to obtain the wood-polymer composites involved the following operations [4, 23]:

- pre-treating the wood flour and mixing it with vinyl acetate in a mixer, at 170–180 °C for 3 min;
- compounding process: adding the polymer into the mixer and continuing the process for 5 minutes for PE and 10 min for PP;
- removal of the mixture from the mixer and pressing it: 3 mm thick flat samples are obtained;
- grinding of the cooled plates in a BELMOS-PEEBLES equipment and obtaining pellets of sizes under 0.5 mm;
- hot pressing the pellets (using a SIEMPFLKAMP press with 600 x 600 mm² turntables, with cooling) and obtaining plates with the thickness g = 2..5 mm. First, a hot pressing is done (T = 185 - 200 °C, p = 75 atm, t = 2 min), followed by a cold pressing one (T = 80 °C, p = 2 - 3 atm, t = 4 - 5 min).

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charge \( i_1(t) \) currents are still not zero ([5], [7], [25]) and that the absorption current has not reached its steady state value. It can be also seen that the values of both \( i_1(t) \) and \( i_2(t) \) increase with wood flour insertion and these increments are more pronounced for the polyethylene matrix (fig. 2 a and b, curves 1).

Variation of \( \sigma_{dc} \) with time (computed from relation (4)) for three types of samples is shown in figure 3. It can be seen that similar to other composites ([15], [26]) \( \sigma_{dc} \) decreases in time and even after \( t = 2 \) h, the variable components of \( \sigma_{dc} \) do not become completely zero, respectively that in order to obtain \( \sigma_{dc} \) value, the voltage needs to be maintained for a period greater than 2 h. Therefore the use of the value of electrical conductivity (or resistivity) measured at 1 min after the application of DC voltage (according to the standards SR HD 429 S1:2002) is correct to compare materials between them, but it is not relevant for assessing the behavior of materials in long-term operation. For that it would be necessary to know the variation curve of resistivity (or electrical conductivity, fig. 3) in time.

On the other hand, an increase of the electrical conductivity value with wood flour content can also be observed. This is due to the greater polarization of wood flour (which means that \( i_p(t) \) has higher values), higher conductivity (higher values for \( \sigma_c \)) and space charge separation at wood-polymer interfaces (higher values for \( i_s(t) \)).

Figures 4 and 5 present the variations of the real and imaginary components of the complex conductivity (\( \sigma' \) and \( \sigma'' \)) for PE1 samples obtained for electric field...
frequency range from $10^4$ to $10^6$ Hz. A decrease of the values of $\sigma$ and $\sigma'$ with frequency decrease can be seen ($\sigma$ decreases by seven orders of magnitude and $\sigma'$ decreases by nine orders of magnitude when the frequency drops from 10 MHz to about 0.5 mHz) [27]. This decrease results, probably, from the slowing of dielectric relaxation processes at lower frequencies, respectively to the reduction of the alternative current component $\sigma_{ac}$. On the other hand, the values of $\sigma$ and $\sigma'$ increase with temperature increase: more in the case of $\sigma$ and less in the case of $\sigma'$. These increments are more significant at lower frequencies. For example, for PE2 samples, for $f = 2.8$ mHz, $\sigma$ increases from 6.13 pSm$^{-1}$ to 682 pSm$^{-1}$ (111 times) when the temperature increases from 30 °C to 90 °C.

The increase of wood flour content leads to an increase of both components ($\sigma$ and $\sigma'$) of the complex conductivity (curves 1 and 2, figs. 6 and 7). This is due to higher filler conductivity, as well as the increase in the total matrix-filling interface area and space charge separation.

For the same filler content, the complex conductivity components are influenced by the matrix nature. Therefore, in the case of a 40% filler content, the values of $\sigma$ and $\sigma'$ take higher values for a polyethylene matrix (curves 1, figs. 6 and 7) than for a polypropylene matrix (curves 3, figs. 6 and 7). This is due to the lower polypropylene conductivity, as well as the more compact physical structure of the polypropylene-based composite [23].

Analyzing the curves $\sigma(\omega)$ (fig. 6) and taking into account the relation (13), the following variation law was considered for $\sigma(f)$

\[
\sigma(f) = \sigma_0 + \sigma_{ac}(f) = \sigma_0 + a f^n, \quad (14)
\]

where:

- $\sigma_0$ is the frequency-independent component (direct current constant conductivity);
- $a$ and $n$ – two material constants (which depend on the electric field frequency $f$).

In order to determine $\sigma_0$, $n$ and $a$, groups of three values for $\sigma$ were used, each corresponding to frequencies of between $4.5 \cdot 10^4$ and 50 Hz or between 50 Hz and 1 MHz.

Their values, for PE2 and PP2 samples, are shown in table 4. Analyzing the values of $\sigma_0$, $\sigma_1$ and $\sigma_2$, a relative difference $\Delta \sigma_0 = (\sigma_1 - \sigma_0) / \sigma_0$ up to 85% is noticeable. This is probably due to the errors in measuring both the absorption and resorption currents, as well as the impedance $Z(f)$ in alternative fields [27] and/or not using adherent electrodes.

The direct current conductivity values $\sigma_{dc}$ and $\sigma_{ac}$ for all samples, with various matrices and wood flour content, at voltages lower than 1000 V are relatively low (below $10^{-10}$ S/m) which would allow these materials to be used in various low voltage equipment.

On the other hand, because the values of $\sigma$ and $\sigma'$ are low at power frequencies ($50 \ldots 100$ Hz), these materials can be used for regular alternative current applications (for example, $50 \ldots 60$ Hz) (figs. 4-7).

For high frequency electric fields ($10^6$ Hz), $\sigma$ and $\sigma'$ have important values (figs. 4-7). Therefore, wood-polymer composites are not recommended in equipment which operate at high frequencies.

### Conclusions

Polymeric composites made of polyethylene or polypropylene matrices and wood have the electrical conductivity strongly dependent on the voltage application time – for dc fields – and of frequency – for ac fields.

Continuous current electrical conductivity of wood-polymer composites presents important variations during voltage application. Therefore, the use of $\sigma_{dc}(f)$ conductivity to characterize the composites would be more useful than using conductivity values determined at 1 min from the voltage application.

The electrical conductivity values depend on the matrix nature, higher values for the conductivities $\sigma$ and $\sigma'$ are obtained for samples with a polyethylene matrix.

The values for $\sigma$ and $\sigma'$ increase with filler content, more for samples with a polyethylene matrix and less for those with a polypropylene matrix.

The wood-polymer composites analyzed in this paper have sufficient insulating properties (conductivity lower

### Table 4

VALUES OF THE QUANTITIES $\sigma_0$, $\sigma_1$ AND $\sigma_2$

<table>
<thead>
<tr>
<th>Samples</th>
<th>Frequency [Hz]</th>
<th>$\sigma_0$ [S/m]</th>
<th>$\Delta \sigma_0$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>PE2</td>
<td>4.6 $\cdot$ 10^4...50</td>
<td>2.5 $\cdot$ 10^{-11}</td>
<td>85</td>
</tr>
<tr>
<td></td>
<td>50...2.7 $\cdot$ 10^6</td>
<td>2.24 $\cdot$ 10^{-13}</td>
<td>85</td>
</tr>
<tr>
<td>PP2</td>
<td>4.6 $\cdot$ 10^4...50</td>
<td>2.46 $\cdot$ 10^{-9}</td>
<td>85</td>
</tr>
<tr>
<td></td>
<td>50...2.7 $\cdot$ 10^6</td>
<td>3.16 $\cdot$ 10^{-13}</td>
<td>85</td>
</tr>
</tbody>
</table>

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than $10^{-10}$ S/m) to be used in electro-technical applications, for low voltage equipment and power frequencies.

A study regarding the modification of the electrical properties of wood-polymer composites due to the electrical and/or thermal aging will be discussed in a future paper.

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