Studying the Influence of Temperatures, Used for Processing ABS, PA 6.6 and POM, on Certain Electrical Properties, when Injection Moulding Items for the Automotive Industry, by Methods Employed for Determining Relative Permittivity and the Dielectric Dissipation Factor

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This paper analyses the influence of processing temperatures of acrylonitrile butadiene styrene (ABS), polyamide 6.6 (PA 6.6) and polyoxymethylene (POM) on some electrical properties when injecting items used in the automotive industry. The ABS specimens were made at the following injection temperatures: 220, 230, 240 and 250°C. The PA 6.6 specimens were made at the following injection temperatures: 280, 290, 300, 310 and 320°C. The POM specimens were made at the following injection temperatures: 180, 190, 200, and 210°C. The determination of electrical properties were carried out using the methods for determining the relative permittivity and the dielectric dissipation factor using zero method with Schering bridge. It was found in the three polymers analysed that the temperature rise processing by injection leads to insignificant changes in relative permittivity ($\varepsilon_r$) and dielectric dissipation factor (tgδ).

Keywords: acrylonitrile butadiene styrene (ABS), polyamide 6.6 (PA 6.6), polyoxymethylene (POM), relative permittivity, dielectric dissipation factor

Automotive industry is one of the most important consumers of plastics. The manufacturing process of different car parts relies on the use of the following technopolymers: polyamides, thermoplastic polyurethanes, polyoxymethylene, polypropylene, methyl polymethacrylate, ethyl cellulose, plasticized polyvinyl chloride, acrylonitrile butadiene styrene, etc., and on the injection process, as the most commonly employed processing technology.

Polyamides are characterized by good dimensional stability; they are highly rigid, especially when reinforced with glass fibres, and resistant to compression, abrasion, shock and vibration; they are hard materials, which remain stiff and tough when subjected to heat, without noticeable changes at temperatures of up to 80-90°C [1, 2]. By adding glass fibres to polyamides, their tensile strength, flexural strength, elasticity and hardness are improved. They prove resistant in salt water, and are stable to oil, hydrocarbons, varnish, weak bases, esters, ethers, alcohols and automotive fuels. They are good electrical insulators. All these properties recommend them as appropriate for the manufacture of various parts in the automotive industry. Thus they are used in the production of: water reservoirs (being characterised by good thermal resistance, resistance to glycol, rigidity and low fuselage); the cooling system (they are resistant at fatigue and glycol, they have good thermal resistance, help reduce the number of materials used, are rigid, and behave well at vibration); pipelines; thermo-regulators (they are resistant to heat and glycol); petrol tank; hollow body for the carburettor; the air circulation system; lid for the cylinder cover (they are heat resistant, rigid, behave well at fuselage, demonstrate good chemical resistance to oil); skeleton covering the motor; fans; seat structures; supporting structures for front ventilation grilles; the structural door module; pedal box and pedals (they are rigid and behave very well at fatigue and impact); reservoir for brake fluid (heat resistant, rigid, chemically resistant); brake hand lever; support gear knob; door handle; front wing; door mirror; defrosting grill; ventilation grill; fuel movement systems; fastening systems for cables.

Polyoxymethylenes are opaque polymers, due to their high degree of crystallinity [3]; they are characterized by good dimensional stability in a wide range of temperature. The high degree of crystallinity gives polyoxymethylene better general mechanical properties (stiffness in particular) as compared to other thermoplastics within the temperature range of 50-120°C. These polymers are resistant to shocks, fatigue, friction and wear. They have good resistance to many organic chemical agents (aldehydes, esters, ethers) and are good electrical insulators. In the automotive industry they are used for the production of: gear wheels, housings, guides, active component parts for oil or diesel oil pumps, flotation gears, valves, windscreen cleaners, etc. [4].

The most important mechanical properties of acrylonitrile butadiene styrene (ABS) are resistance to impacts and toughness. ABS is rigid, wear-resistant and tear-resistant; it has good dimensional stability in a wide range of temperature, unlimited colouring options, easily forms into shape [5] and is a good electrical insulator. It is resistant to acids and weak bases and unstable to esters, ketones, ethers and gasoline. Applications in the
Experimental part
In order to obtain the specimens, the following materials were used: ABS MAGNUM 3453, the PA 6.6 \text{T\textsc{ECHNYL AR218V30 Blak polyamide type}, and the POM \text{EUROTAL C9 NAT Polyoxymethylene}, and \text{MAGNUM 3453ABS}, which are used, in this particular case, for producing specific items for the automotive industry.

Relative permittivity and the dielectric dissipation factor were measured for all the samples produced by injection. All tests were performed at room temperature, in the laboratories of Institut Supérieur d'Électronique et du Numérique (ISEN), of the Catholic University of Lille, France, and in the new laboratories of ISEN - Urban Wood, Lille, France.

Employing the zero method by Schering Bridge to determine relative permittivity and the dielectric dissipation factor.

To determine the relative permittivity and the dielectric dissipation factor the zero method was used. This method can be employed at measuring frequencies of up to 100 kHz. A Schering bridge balanced by adjusting one of the arms, both with and without samples, was used for the determination of the dielectric dissipation factor, at frequency values of: 200 Hz, 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz and 20000 Hz.

The Schering Bridge shown in figure 2 is a four-arm bridge, where two arms are capacitive: $C_n$ (without losses) and $C_r$, and two are resistive: $R_1$ and $R_2$. The variable $R_1$ resistance is opposed to the unknown capacity $C_x$ (the capacity of the specimen), this resistance being shunted by $C_n$, the standard bridge capacity.

To determine relative permittivity and the dielectric dissipation factor we used a device with two terminals between which the specimen was placed (fig. 3).

The relative permittivity ($\varepsilon_r$) of a dielectric environment is calculated using the formula:

$$\varepsilon_r = \frac{\varepsilon_r}{\varepsilon_0} \tag{1}$$

where:

- $\varepsilon_r$ is the absolute permittivity of that environment,
- $\varepsilon_0$ is the permittivity of the vacuum.

The relative permittivity of an electro-insulating material is determined experimentally by the relation:

$$\varepsilon_r = \frac{C_x}{C_0} \tag{2}$$

where:

- $C_x$ is the capacity of a capacitor in which the space between and around the electrodes is wholly and exclusively filled with insulating material,
- $C_0$ is the capacity of the same arrangement of electrodes in a vacuum.

The value of relative permittivity, for free-CO$_2$ dry air, at normal atmospheric pressure, is $1.00053$. In the experimental measurements of this paper we used the $C_{par}$ capacity of the electrodes-in-air device, instead of the $C_0$ capacity in vacuum, in order to determine the $\varepsilon_r$ relative permittivity with sufficient precision.

We determined the value of the $C_{par}$ parasitic capacitance of the device with the relation:

$$C_{par} = \varepsilon_0 \cdot \frac{S}{d} \tag{3}$$

where:

- $\varepsilon_0$ is the permittivity of the vacuum $\varepsilon_0 = 1/ 4 \pi \times 9 \times 10^9$ = 0.08854, [pF/cm]
- $S$ is the area of the device terminal, $S = 37.25$ cm$^2$
- $d$ is the distance between the terminals of the device, $d$ = 0.4 cm

The value obtained for $C_{par}$ was 8.25 pF.

The value of $C_x$ is calculated using the following equation:

$$C_x = \varepsilon_{max} \cdot \frac{S}{d} \tag{4}$$
where:

- $\varepsilon_{\text{max}}$ is the permittivity of the specimen,
- $S$ is the area of the device terminal, $S = 37.25 \text{ cm}^2$,
- $d$ is the distance between the terminals of the device, with the specimen between terminals, $d = 0.4 \text{ cm}$,
- $\varepsilon_r = \frac{\varepsilon_{\text{max}}}{\varepsilon_0} = \frac{(C_d \cdot d / S)}{(C_{\text{par}} \cdot d / S)} = \frac{C_d}{C_{\text{par}}}$ (5)

The $\delta$ dielectric loss angle of an electro-insulating material is the angle complementary to the ($\phi$) phase difference angle, between the applied voltage and the respective current, when the dielectric of the capacitor is made exclusively of this material (fig. 4).

Figure 5 presents the equivalent diagram in parallel for a capacitor with losses.

The $\text{tg} \, \delta$ tangent of the dielectric loss angle of an electro-insulating material is the size that indicates the result of measuring the $\delta$ angle of dielectric loss for this material.

To calculate the $\text{tg} \, \delta$ dielectric loss tangent, we used the relation for parallel representation:

$$
\text{tg} \, \delta = \frac{1}{\omega C_p R_p} = \frac{C_p}{\omega} \cdot \frac{G_p}{2\pi f_{\text{max}} \cdot C_p} \quad (6)
$$

where:
- $f_{\text{max}}$ is the measurement frequency (200 Hz, 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz and 20000 Hz)
- $C_p$ is the capacity for parallel equivalent circuit, [F]
- $G_p$ is the conductance (reverse physical size of the electric resistance $G_p=1/R_p$) for the parallel equivalent circuit [S].

Both values, $C_p$ and $G_p$, are read using a Hewlett-Packard Model 4276A LCZ Meter (fig. 6). HP Agilent 4276A is a tool employed for measuring impedance, designed for the measurement of circuit components such as capacitors and inductors using frequency. The device feeds the Schering Bridge with a signal at a certain frequency, and on the two displays the device reads the values of both $C_p$ (display A) and of $G_p$ (display B). The values are obtained by the instantaneous reading of values indicated by the device (the reading of values from the machine was made no later than 1s from pushing the buttons).

Measuring the $C_p$ capacity was made by balancing the Schering Bridge while adjusting the values of the $R_1$ resistance and of the $C_1$ capacity. The measurement was made using a Tektronix oscilloscope MDO3024 model (fig. 7).

The Schering Bridge was fed (fig. 2) in the nodes A and B with a $U_{AB}$ sinusoidal signal, with an amplitude of 5V and the $f_{\text{max}}$ frequency, as it can be observed on the purple signal 3 on the display of the oscilloscope (fig. 7). The $R_1$ variable resistance values, as well as those of the $C_1$ capacity variable were adjusted, so that the current between node M and the node N will be zero, i.e. the two signals, $U_{\text{yellow}}$ (yellow signal 1) and $U_{\text{blue}}$ (blue signal 2), would have the same amplitude. Thus, the equivalent values of $C_1$ (Cp and Gp) capacity were determined, balancing the bridge by obtaining signals of the same amplitude, as shown in figure 7.

Measurements were made in accordance with the NFC 26-230: 1970 CEI IEC 60250 [11] European Standards on specimens with shapes and dimensions as shown in figure 8.

For the ABS MAGNUM 3453 material, a total of 24 measurements for determining relative permittivity and the dielectric dissipation factor were made. Thus, for the four samples produced at the following injection temperatures: 220, 230, 240, 250°C, six measurements were carried out, corresponding to the six selected frequency measures (200 Hz, 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz and 20000 Hz).

A total of 30 measurements were made for the material PA 6.6 TECHNYL AR218V30 Blak, in order to determine relative permittivity and the dielectric dissipation factor. Thus, for the five samples produced at the following injection temperatures: 280, 290, 300, 310 and 320°C, six measurements were carried out, corresponding to the six selected frequency measures (200 Hz, 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz and 20000 Hz).

A total of 24 measurements were made for the material POM EUROTAL C9 NAT, in order to determine relative permittivity and the dielectric dissipation factor. Thus, for the five samples produced at the following injection temperatures: 180, 190, of 200 and 210°C, six measurements were carried out, corresponding to the six selected frequency measures (200 Hz, 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz and 20000 Hz).
Results and discussions

After testing the ABS samples, the following values of relative permittivity were obtained (table 1).

It can be seen that, at the measurement frequencies of 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz and 20000 Hz, the increase of processing temperature for ABS, from 220 to 240°C, led to a slight decrease in relative permittivity values. A further increase of processing temperature, to the value of 250°C, led to a slight increase in the values of relative permittivity. At the measuring frequency of 200 Hz, the increase of processing temperatures for ABS, from 220°C to 250°C, determined the decrease of relative permittivity values from 2.03636 to 1.96363. The lowest value of relative permittivity (i.e. 1.92727) was recorded at the temperature of 240°C and at a frequency of 20000 Hz, while the highest value of relative permittivity (i.e. 2.07272) was recorded at 220 °C and at a frequency of 10000 Hz.

At testing PA 6.6 samples, the following results for relative permittivity were obtained (table 2).

Increasing temperature to 310°C led to a slight increase in relative permittivity values and finally raising the temperature to 320°C led to slight decrease in relative permittivity. The lowest value of relative permittivity, of 1.4424, was recorded at 280°C and at a frequency of 20000 Hz, while the highest value of relative permittivity, i.e. 1.8545, was recorded at 310°C and at a frequency of 500 Hz.

Table 1
THE RELATIVE PERMITTIVITY FOR THE ABS MAGNUM 3453 SAMPLES, PROCESSED BY INJECTION, DEPENDING ON PROCESSING TEMPERATURES AT DIFFERENT FREQUENCIES

<table>
<thead>
<tr>
<th>Processing Temperature[°C]</th>
<th>200 Hz</th>
<th>500 Hz</th>
<th>1000 Hz</th>
<th>5000 Hz</th>
<th>10000 Hz</th>
<th>20000 Hz</th>
</tr>
</thead>
<tbody>
<tr>
<td>220</td>
<td>2.03636</td>
<td>2.06060</td>
<td>2.06060</td>
<td>2.04848</td>
<td>2.07272</td>
<td>2.02424</td>
</tr>
<tr>
<td>230</td>
<td>2.02424</td>
<td>2.00000</td>
<td>2.01212</td>
<td>2.00000</td>
<td>2.02424</td>
<td>1.97575</td>
</tr>
<tr>
<td>240</td>
<td>1.97575</td>
<td>1.95151</td>
<td>1.95151</td>
<td>1.93959</td>
<td>1.97575</td>
<td>1.92727</td>
</tr>
<tr>
<td>250</td>
<td>1.96363</td>
<td>1.98787</td>
<td>2.00000</td>
<td>1.98787</td>
<td>2.01212</td>
<td>1.97575</td>
</tr>
</tbody>
</table>

Table 2
THE RELATIVE PERMITTIVITY OF PA 6.6 TECHNYL AR218V30 Blak SAMPLES PROCESSED BY INJECTION, DEPENDING ON PROCESSING TEMPERATURES AT DIFFERENT FREQUENCIES

<table>
<thead>
<tr>
<th>Processing temperature[°C]</th>
<th>200 Hz</th>
<th>500 Hz</th>
<th>1000 Hz</th>
<th>5000 Hz</th>
<th>10000 Hz</th>
<th>20000 Hz</th>
</tr>
</thead>
<tbody>
<tr>
<td>280</td>
<td>1.4545</td>
<td>1.5030</td>
<td>1.4909</td>
<td>1.4666</td>
<td>1.4666</td>
<td>1.4424</td>
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<tr>
<td>290</td>
<td>1.5272</td>
<td>1.6127</td>
<td>1.6363</td>
<td>1.6000</td>
<td>1.6363</td>
<td>1.6121</td>
</tr>
<tr>
<td>300</td>
<td>1.6727</td>
<td>1.6969</td>
<td>1.6484</td>
<td>1.7090</td>
<td>1.6242</td>
<td>1.5757</td>
</tr>
<tr>
<td>310</td>
<td>1.7575</td>
<td>1.8545</td>
<td>1.8060</td>
<td>1.7454</td>
<td>1.7696</td>
<td>1.7212</td>
</tr>
<tr>
<td>320</td>
<td>1.5878</td>
<td>1.6363</td>
<td>1.6484</td>
<td>1.6484</td>
<td>1.6484</td>
<td>1.6000</td>
</tr>
</tbody>
</table>
After testing POM samples, the following results of relative permittivity were obtained (table 3).

It can be seen that, at all the six measurement frequencies (200 Hz, 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz, 20000 Hz), the increase of processing temperature for POM, from 180 to 200°C, led to a slight decrease in relative permittivity values. Further increasing the processing temperature to 210°C led to a slight increase in relative permittivity values. The lowest value of relative permittivity, of 2.09696, was recorded at 200°C and a frequency of 200 Hz, while the highest value of relative permittivity, of 2.46060, was recorded both at 180°C and a frequency of 200 Hz, and at 180°C and a frequency of 10000 Hz.

In the case of both ABS and POM, when processing temperatures are the lowest (ABS - 220°C and POM - 180°C) and when mechanical degradation occurs in the injection barrel, the highest values of relative permittivity were recorded. Further increasing processing temperatures led to a slight decrease in relative permittivity values, while at the highest processing temperatures (ABS - 250°C and POM - 210°C), when thermal degradation occurs in the injection barrel, the values of relative permittivity increased again, with the exception of the ABS sample injected at a temperature of 250°C and measured at 200 Hz.

After testing the ABS samples, the following results were obtained for the dielectric dissipation factor (table 4).

It can be seen that, at measurement frequencies of 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz and 20000 Hz, raising the processing temperature of the ABS, from 220°C to 240°C resulted in a slight increase in the values of the dielectric dissipation factor. A further increase of processing temperature, to the value of 250°C, led to a slight decrease of dielectric dissipation factor values. At the measuring frequency of 200Hz, increasing ABS processing temperatures, from 220 to 250°C, determined the increase of the dielectric dissipation factor values, from 0.04736 to 0.04912. The lowest value of the dielectric dissipation factor, of 0.00476, was recorded at 220°C and at a frequency of 20000 Hz, while the highest value of the dielectric dissipation factor, of 0.04912, was registered at the temperature of 250°C and at a frequency of 200 Hz. Increasing the frequency, from 200 Hz to 20000 Hz, at any injection temperature value for the ABS samples, led to the decrease, by an order of magnitude, of the material’s dielectric dissipation factor.

After testing the PA 6.6 samples, the following results were obtained for the dielectric dissipation factor (table 5).

It can be seen that, at all the six frequency measurements (200 Hz, 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz and 20000 Hz), raising the processing temperature of the PA 6.6 sample from 280 to 320°C led to insignificant changes in the dielectric dissipation factor. Increasing the frequency from 200 Hz to 20000 Hz, at any of the temperatures of injection for the PA 6.6 samples, lowered the dielectric dissipation factor of the material. The lowest
value of the dielectric dissipation factor, of 0.00598, was recorded at 290°C and at a frequency of 20000 Hz, while the higher value for the dielectric dissipation factor, of 0.30373, was registered at the temperature of 320°C and at a frequency of 200 Hz.

As a result of testing POM samples, the following results were obtained for the dielectric dissipation factor (table 6).

It can be seen that, at all the six frequency measurements (200 Hz, 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz and 20000 Hz), increasing the processing temperature for POM, from 180°C to 200°C, led to a slight increase in the dielectric dissipation factor values. A further increase of temperature, up to 210°C, determined a slight decrease in dielectric dissipation factor values. Increasing the frequency from 200 Hz to 20000 Hz, at any of the injection temperatures for the POM samples, determined the decrease of the material’s dielectric dissipation factor. The lowest value of the dielectric dissipation factor, of 0.00784, was recorded at 180°C and at a frequency of 10000 Hz, while the highest value of the dielectric dissipation factor, of 0.04599, was registered at 200°C and at a frequency of 200 Hz.

The graphic representations of the relative permittivity variations, in relation to the measuring frequency, in the case of the three polymers studied are shown in figure 9.
Based on the charts shown in figure 9, we might conclude that, in the case of the three polymers analyzed (ABS, PA 6.6 and POM), the increase of measurement frequency from the value of 200 Hz to the value of 20000 Hz has an insignificant influence on the relative permittivity of materials.

The graphical representations of variations in the dielectric dissipation factor ($\tan \delta$), depending on measurement frequency, as regards the three polymers studied, are shown in figure 10.

Based on the graphic representations shown in figure 10, it can be concluded that, for the three polymers analyzed (ABS, PA 6.6 and POM), increasing the measuring frequency value from 200 Hz to the value of 20000 Hz leads to a significant reduction of the dielectric dissipation factor.

Conclusions

This paper has analysed changes in the electrical properties of acrylonitrile butadiene styrene of the ABS MAGNUM 3453 type, of the PA 6.6 TECHNYL AR218V30 Blak polyamide type and of POM Euroteam C9 NAT Polyoxymethylene, which are polymers used in the production of specific parts for the automotive industry, in relation to the injection processing temperature and the measuring frequency. The ABS specimens were produced at the following injection temperatures: 220, 230, 240 and 250°C. The PA 6.6 specimens were made at the following injection temperatures: 280, 290, 300, 310 and 320°C. The POM specimens were made at the following injection temperatures: 180, 190, 200, and 210°C. Samples were injection moulded on an ENGEL CC 100 Type ES 80/50 HL injection machine.

The determination of electrical properties was performed using methods for determining relative permittivity and the dielectric dissipation factor, namely the zero method by Schering Bridge. The tests were performed using as measuring device a Hewlett-Packard Model 4276 and a Model MDO3024 Tektronix oscilloscope. It has been found, in the three polymers analysed, that the increase of temperature during injection-processing leads to insignificant changes in relative permittivity ($\varepsilon_r$) and in the dielectric dissipation factor ($\tan \delta$). At the same time it has been found that increasing the frequency of measurement, from the value of 200 Hz to the value of 20000 Hz, has an insignificant influence on the relative permittivity of materials, though, at the same time, it leads to a significant reduction of the dielectric dissipation factor.

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