MECHANISMS OF INTERACTION BETWEEN POLYPROPYLENE FOIL
AND LIQUID DIELECTRIC

1. INTRODUCTION

Interaction between the polypropylene foil and insulating liquid is
the complex of phenomena which must be taken into account in the construc-
tion and technology of all-film capacitors. Both in the technological
as in the ageing process in working conditions gas cavities cannot re-
main or be generated and the products of this interaction should not de-
crease the dielectric properties of the insulating system.

The interaction between the foil and liquid dielectric consists of so-
me phenomena such as adhesion of liquid to the surface of PP-film, disso-
vling of solid phase in the liquid one, PP-swelling, cavitation of gas bu-
bles under the influence of electrical stress at the interface.

2. Adhesion

Adhesion depends on the contact angle $\Theta$ between the liquid and solid
phase, which fulfils the Young's equation [1]:

$$W_a = \gamma_1 \cdot (1 + \cos \Theta)$$  \hspace{1cm} (1)

where:
- $\gamma_1$ - the surface tension of liquid,
- $W_a$ - the adhesion work between solid and liquid phase.

Bearing in mind, that $\gamma_1$ is proportional to the molar cohesive energy
$\gamma_1 \sim W_c$, it can be concluded that the liquids with relatively low value

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of \( \gamma_c \) should be applied. The contact angle \( \theta \) decreases in this case and it results in the better wettability of the solid phase by the liquid one. Surface tension \( \gamma_\ell \) of liquid hydrocarbons is of the value of 0.02 to 0.04 N/m [2].

3. Dissolving

The process of dissolving is governed by the free energy equation:

\[
\Delta F = \Delta H - T \cdot \Delta S
\]  

(2)

where:
- \( \Delta F \) - the change of energy,
- \( \Delta H \) - enthalpy (heat of mixing),
- \( T \) - absolute temperature,
- \( \Delta S \) - entropy of mixing.

Dissolving is characterized by such an increase of \( T \cdot \Delta S \) component in the equation (2) in comparison with the change of \( \Delta H \) that the free energy change \( \Delta F \) becomes negative.

The enthalpy \( \Delta H \) is proportional to the difference \( (\delta_1 - \delta_s)^2 \), where \( \delta_1 \) and \( \delta_s \) are the solubility parameters of mixing materials. They are equal the square root of the volume density of the molar-cohesive energy:

\[
\delta = \sqrt{\frac{\Delta E}{V}}
\]  

(3)

where:
- \( \Delta E \) - the energy of vaporization of the components 1 or s respectively,
- \( V \) - molar volume of the component 1 or s.

The \( \delta \) values of polymer solids are tabularized [3]. For liquid solvents the values of \( \Delta H_\ell \) can be calculated as follows:

\[
\Delta H_\ell = \Delta H_1 - R \cdot T
\]  

(4)

where:
- \( \Delta H_1 = 99.1 \cdot T_b + 0.06 \cdot T_b^2 - 12336 \) [J/mol]
- \( R = 8.314 \) gas constant,
- \( T_b \) - the boiling temperature [K].

To decrease the dissolving of solid phase in liquid one the insulating liquids with increased \( (\delta_1 - \delta_s) \) value should be applied. Comparing the conclusions from parts 2 and 3, it can be said that the require-
ment of good adhesion is in the contradiction to the requirement of small solubility. Thus a compromise should be taken into account.

4. Swelling

Swelling is characterized by such an increase of the enthalpy $\Delta H$ in comparison with the change of $T$. $\Delta S$ in equation (2) that the free energy change $\Delta F$ becomes positive. Swelling is determined by the coefficient of the relative mass increase:

$$\Delta m = \frac{m - m_0}{m_0}$$  

where:
- $m_0$ - mass of swelling polymer before this process,
- $m$ - mass of swelling polymer after this process.

5. Experimental results

Two synthetic insulating liquids were investigated:

Liquid No 1: the refined dodecylbenzene $C_{18}H_{30}$
- $d = 0.859 \text{ g/cm}^3$
- $T_b = 426 \text{ K}$

Liquid No 2: the blend of monobenzyltoluene $C_{14}H_{14}$ and dibenzylbenzene $C_{21}H_{20}$
- $d = 1.005 \text{ g/cm}^3$
- $T_b = 472 \text{ K}$

The polypropylene films 0.01 mm were immersed in these liquids and the dynamic of mass increase of solid dielectric was measured. The experiments were carried out at two temperatures: 353 and 293 K. In equilibrium state the following results of swelling were obtained (table 1).

<table>
<thead>
<tr>
<th>Liquid No</th>
<th>$\Delta m$ (g/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$T = 293 K$</td>
</tr>
<tr>
<td>1</td>
<td>0.08</td>
</tr>
<tr>
<td>2</td>
<td>0.04</td>
</tr>
</tbody>
</table>
6. Calculations

The solubility parameter for polypropylene was assumed: \( \delta_{pp} = 18.81 \) \( [J/cm^3]^{1/2} \).

Results of the calculations of quantities influencing the dissolving and swelling processes are given in the table 2.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Liquid No 1</th>
<th>Liquid No 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \Delta H ) [J/mol]</td>
<td>44398</td>
<td>53077</td>
</tr>
<tr>
<td>( \Delta H ) at 353 K [J/mol]</td>
<td>41453</td>
<td>50142</td>
</tr>
<tr>
<td>at 294 K [cm^3]</td>
<td>41961</td>
<td>50640</td>
</tr>
<tr>
<td>( V ) [cm^3]</td>
<td>283</td>
<td>190 - 270</td>
</tr>
<tr>
<td>( \delta ) at 353 K [J/cm^3]^{1/2}</td>
<td>12.25</td>
<td>16.7 - 13.6</td>
</tr>
<tr>
<td>at 293 K</td>
<td>12.17</td>
<td>16.32 - 13.69</td>
</tr>
<tr>
<td>( (\delta - \delta_{pp})^2 ) at 353 K [J/cm^3]</td>
<td>43.50</td>
<td>4.5 - 27.1</td>
</tr>
<tr>
<td>at 293 K</td>
<td>44.08</td>
<td>6.2 - 26.2</td>
</tr>
</tbody>
</table>

From the above results it is seen that the \( (\delta - \delta_{pp})^2 \) value is greater in the case of liquid No 1.

Thus it can be concluded that the swelling of polypropylene film in contact with this liquid should be greater in comparison with liquid No 2. The experimental result of coefficient \( \Delta m \) are in good accordance with this conclusion.

On the basis of relatively high values of \( (\delta - \delta_{pp}) \) in both liquids it can be concluded that the dissolving process is of the less meaning in comparison with the swelling one.

7. Observations

Samples of investigated PP-foils have been observed by the use of electron scanning microscopy technique. The examples of photos of the PP-
foil surface attacked by investigated liquids are presented in the Fig. 1.

Fig. 1. Examples of PP-surface image in ESEM
a - Liquid No 1, 3000 x, b - Liquid No 2, 3500 x.

The different results of the penetration of each of both liquids are seen. Further investigations correlated to the morphology of PP foil and to the chemical structure of liquids are being continued.

References