Dielectric characteristics of syntactic plastic foams with polyorganosiloxane binder in the ultrahigh frequency region

V. Yu. Chukhlanov and V. V. Kireev
Vladimir State University and the D. I. Mendeleev Russian Chemico-Technological University

Selected from International Polymer Science and Technology, 30, No. 7, 2003, reference PM 03/04/25; transl. serial no. 15013

At present, in connection with the increasing use in most sectors of industry of various devices and instruments whose operating principle is based on the use of ultrahigh frequency (UHF) electromagnetic energy (>300 MHz), interest has arisen in dielectric materials with minimum dielectric losses and accordingly greatest radioparency in this region of radio frequencies. For these purposes, syntactic plastic foams (SPFs) comprising a hollow spherical filler held together by the polymer matrix may be of particular interest owing to their low density and high physicomechanical characteristics. Naturally, both the filler and the binder should have fairly high dielectric characteristics. Furthermore, if SPF is additionally used as heat insulation material, both components should be characterised by fairly high heat stability.

The authors assumed that these requirements are satisfied by hollow glass microspheres (HGMs) and organosilicon polymer binder materials which, besides good dielectric properties, have high heat stability.

Since there are no published data on the dielectric characteristics of foam materials investigated in the UHF region, the aim of the present work was to study the dielectric characteristics of SPFs and also to reveal the influence of the nature and ratio of the binder and filler on these properties.

Standard-production sodium borosilicate HGMs of grade MSO A9 gr.A2 were used as the filler. The given HGMs have the following characteristics:

- Density of particles: 300 kg/m³
- Average particle size: 20 mm
- Coefficient of filling of volume: 60%

In the present work, low-modulus dimethylsiloxane rubber SKTN-1 with end hydroxyl groups was used as the organosilicon binder. The SKTN-1 was cured with catalyst K-18, which is a mixture of tetraethoxysilane and tin diethylcaprylate. The process of curing was accompanied with interaction between the end hydroxyl groups and reactive ethoxy groups of tetraethoxysilane and with the formation of ethyl alcohol. As a result of the reaction, polydimethylsiloxane (PDMS) was formed, which has increased heat stability and acceptable dielectric characteristics.

The process of producing SPFs consisted in mixing hollow microspheres with the prescribed amount of SKTN-1, into which 3% catalyst K-18 had been added beforehand. The specimens were formed in an aluminium mould under a small excess pressure of 0.4 MPa. After holding for 72 h at a temperature of 298 K, the specimens were removed from the mould and, if necessary, subjected to additional mechanical treatment. The physicomechanical and thermophysical characteristics of the SPFs produced in this way have been described in detail in studies published earlier [1, 2].

The dielectric characteristics of the specimens were determined in a volume resonator at a frequency of 9.8 GHz in accordance with GOST 8544.

Taking into account that SPFs consist of three phases (glass, PDMS, and air), and also that the dielectric losses in air are extremely low, it can be concluded that the determining influence on the dielectric characteristics of foam materials will be rendered by binder PDMS and the glass shell of the filler.
The conducted investigations showed that, with increase in the volume fraction of the binder, along with increase in radioparency losses, increase in the tangent of the angle of dielectric losses also occurs (Figure 1).

![Figure 1](image1.png)

**Figure 1. Dependence of dielectric permittivity and tangent of angle of dielectric losses on PDMS content in SPF**

For heterogeneous systems there is an analytical ratio (the Lichteneker formula) connecting the dielectric permittivity of the composite $\varepsilon$ with the dielectric permittivities of the components $[3]$

$$\varepsilon = \theta_1 \ln \varepsilon_1 + \theta_2 \ln \varepsilon_2$$

where $\varepsilon_1$ and $\varepsilon_2$ are the dielectric permittivities of the first and second components respectively, and $\theta_1$ and $\theta_2$ are the volume fractions of the first and second components respectively.

Comparing the calculated and experimental values of dielectric permittivity in Table 1, we see that the calculated values of dielectric permittivity are lower than the experimental values with a binder content of less than 40 vol.%. This may be attributed to the presence of open pores in the SPFs with a low volume content of PDMS owing to incomplete filling of the intersphere space and accordingly the appearance of sorbed moisture in the material.

<table>
<thead>
<tr>
<th>Content of PDMS in SPF, vol.%</th>
<th>Dielectric permittivity</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Calculated</td>
</tr>
<tr>
<td>10</td>
<td>1.26</td>
</tr>
<tr>
<td>30</td>
<td>1.82</td>
</tr>
<tr>
<td>50</td>
<td>2.39</td>
</tr>
<tr>
<td>70</td>
<td>2.72</td>
</tr>
<tr>
<td>90</td>
<td>2.97</td>
</tr>
<tr>
<td>100</td>
<td>3.15</td>
</tr>
</tbody>
</table>

Experimental data confirm that the introduction of waterproofing fluid 136-41 into the SPFs with an open-pore structure leads to a marked improvement in the dielectric characteristics (Figure 2). Thus, the tangent of the angle of dielectric losses decreases by 30% when the waterproofing fluid is introduced in a quantity

![Figure 2](image2.png)

**Figure 2. Dependence of tangent of angle of dielectric losses on content of waterproofing fluid 136-41 in SPF**

The action of moisture on the surface of the glass leads to a decrease in the surface electrical resistance of the material from $1 \times 10^{12} - 1 \times 10^{14}$ $\Omega$ to $0.5 \times 10^{7} - 3 \times 10^{9}$ $\Omega$. To reduce the moisture absorption of SPFs, experiments were carried out on the introduction into the foam material of waterproofing fluid 136-41. This fluid is by nature an organosiloxane containing, along with side organic radicals, hydrogen substituents. The application of water repellants should lead to retention of a very high surface electrical resistance of the HGMs, hardly dependent on the humidity of the ambient air. Increase in surface electrical resistance leads accordingly to a reduction in the tangent of the angle of dielectric losses. Thus, it can be assumed that the introduction of waterproofing fluid into the SPFs with a volume fraction of the binder of less than 40% will lead to an increase in the dielectric characteristics in the UHF range.

The conducted investigations showed that, with increase in the volume fraction of the binder, along with increase in radioparency losses, increase in the tangent of the angle of dielectric losses also occurs (Figure 1).
Further introduction of the waterproofing fluid is inexpedient since in this case the dielectric characteristics of the foam material hardly change. The dependence of the losses of radioparency of the material and moisture absorption on the content of waterproofing fluid 136-41 in the SPFs (Figure 3) also indicates the mutual dependence of these quantities.

Retention of the dielectric characteristics of the SPFs with increase in temperature depends on the temperature dependence of the dielectric characteristics of the binder and filler. Thus, the dielectric properties of the glass depend largely on its composition. For example, the dielectric characteristics of lithium-containing glass under conditions of high temperatures are considerably lower than those of potassium-containing glass, which is due to the small size of the lithium atom and, accordingly, its high mobility and thereby its reduced electrical resistance [3]. Accordingly, the dielectric characteristics of the sodium-containing glass as a function of temperature will occupy an intermediate position between the above glasses since the size of the sodium atom occupies an intermediate position between the sizes of the lithium and potassium atoms.

An important property of materials based on organosilicon binders is retention of high dielectric characteristics after exposure to a flame. However, in this case the application only of organosilicon binders that contain no phenyl groups as side substituents at the silicon atoms is allowed. The phenyl groups lead to the formation of a conductive coke layer which greatly degrades the dielectric properties of the materials [4].

As shown by investigations, the action of a flame on SPFs with binder PDMS does not lead to any significant changes in the dielectric properties of the foam material (Table 2).

Thus, the conducted investigations make it possible to draw the following conclusions:

- Increase in the content of binder in the SPFs above 15–20 vol.% leads to considerable loss of radioparency (>1 db per 10 mm thickness of the material);
- Reduction in the tangent of the angle of dielectric losses and increase in radioparency practically by a factor of 4 in the UHF range for SPFs with a binder content of less than 15 vol.% is achieved by introducing waterproofing organosilicon fluid in a quantity of 1.0–1.5 wt.% of the foam material;
• the temperature dependence of the dielectric characteristics in the range 273–423 K is insignificant, which makes it possible to use the SPF under conditions of increased temperatures;
• exposure to an open flame does not lead to any marked deterioration in the dielectric characteristics of highly filled SPF with binder PDMS.

REFERENCES


(No date given)