A STUDY OF THERMAL CONDUCTIVITY OF BORON-NITRIDE EPOXY-MATRIX COMPOSITES.

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Abstract
The aim of the work is assessment of the possibility of increasing the thermal conductivity of the filled epoxy resins. The heat management in the electronic and power devices is a crucial issue. It is an important problem to be solved in order to prevent such devices from malfunction. The effective heat dissipation is thus one of the main challenging problems to be solved. The pure boron nitride micro-powders with different size distribution and surface modification, as well boron nitride mixtures with silica were used as the fillers for preparation of the epoxy resin composites. A representative set of filled epoxy samples has been prepared with the different content of the investigated fillers. The thermal conductivity measurements have been performed on these samples at the room temperature and SEM observations have been showed.

1 Introduction
Thermal conductivity of neat-epoxy resin is one of the lowest among all known solid materials. In order to increase the thermal conductivity the addition of filler particles with high thermal conductivity into epoxy resin is used as common method. The properties of obtained epoxy composite depend on properties of used filler, i.e. size, shape, surface area or dispersion in a matrix. The filled epoxy composites are broadly used in power and microelectronic industry. The heat management in the electronic and power devices is a crucial issue. It is an important problem to be solved in order to prevent such devices from malfunction. The effective heat dissipation is thus one of the main challenging problems to be solved [4]. Hexagonal boron-nitride has high in-plane thermal conductivity which goes up to 600W/mK and at the same time possess good electrical insulating properties[1,2]. From this reasons h-BN was used as a filler. In this work two types of hexagonal boron nitride has been used, spherical PTX 25 and plate-like PT 100. Thermal conductivity was measured by two complementary methods and morphology of fillers was investigated.

2 Materials and methods
For this study a typical industrial liquid epoxy derived from bisphenol A (DGEBA) CY 228-1 hardened with HY918 (Huntsman) was used. As a filler a commercial h-BN powders PT100 and PTX25 (Momentive) were used. The properties of fillers are shown in Table 1.
<table>
<thead>
<tr>
<th>Filler type</th>
<th>Surface Area [m²/g]</th>
<th>APS [µm]</th>
<th>Tap Density [g/cc]</th>
<th>Bulk Density [g/cc]</th>
</tr>
</thead>
<tbody>
<tr>
<td>BN PT100</td>
<td>5.5</td>
<td>13</td>
<td>0.3</td>
<td>2.25</td>
</tr>
<tr>
<td>BN PTX25</td>
<td>7</td>
<td>25</td>
<td>0.3</td>
<td>2.25</td>
</tr>
</tbody>
</table>

Table 1 Properties of used fillers BN PT 100 and BN PTX 25

Epoxy resin samples, filled with 5%, 10%, 15%, 20%, 25% and 30% by volume fraction with boron nitride powders were produced by conventional vacuum casting method with the use of typical curing profile found in producer’s technical data sheet. The cylindrical specimens, with the diameter of 6 mm and high from 3 mm to 6 mm, were prepared for thermal measurements. Thermal conductivity measurements were performed by means of PPMS (Physical Property Measurement System) manufactured by Quantum Design Inc using thermal transport option (TTO) (Figure 1). The sample was mounted in the two probe lead configuration, i.e. spliced to the copper leads by the epoxy adhesive layer. Heat was applied by the current running through the copper lead. The thermal conductivity measurements have been performed in the high vacuum conditions at room temperature.

![Figure 1](image1.png)

**Figure 1** Thermal transport option puck (left) used in thermal conductivity measurements, and sample prepared for measurement (right).

![Figure 2](image2.png)

**Figure 2** Hot Disc Thermal Constant Analyser TPS 500 based on Transient Plane Source technique.

Additional measurements with Hot Disc Thermal Constant Analyser TPS 500 (Figure 2) have been carried out in room temperature. The measurement system is based on Transient Plane Source technique [3]. The sensor is sandwiched between two pieces of sample (cylindrical shape with 35 mm diameter and 20 mm high). By marking upper and lower side of the samples it was possible to test 3 configuration of sample set-up. Such arrangement provide information about possible differences, as filler inhomogeneous concentration on upper and lower side.

3 Results
3.1 Morphology of fillers
Many studies indicated, that properties of filler such as shape, size and dispersion have an significant impact into effective thermal conductivity of obtained composites. In Lewis-
Nielsen model to predict effective conductivity, shape factor is taken into account [5]. The scanning electron microscopy (SEM) images shown in Figure 3 show morphology of investigated h-BN. For BN PTX25 agglomerate structure with tendency to create spherical agglomerates with diameter about 30 µm is observed. BN PT100 (left image in Figure 3) has plate-like irregular shape with average particle size of 13 µm.

![SEM images of BN PT100 and BN PTX25](image)

**Figure 3.** Results of SEM observation (found in producer’s technical data sheet): BN PT 100 flakes shape (left), BN PTX spherical agglomerates (right).

### 3.2 Thermal conductivity measurements

The thermal conductivity results of epoxy-BN composite measured with two methods at room temperature are shown in Figure 4 for BN PT100 and in Figure 5 for BN PTX25. Effective thermal conductivity increases with subsequent increasing of filler concentration according to prediction by theoretical models [5]. In both cases more significant increase is observed for 20% volume filler content. This tendency indicates consistency with percolation model, i.e. the efficient thermal conductivity paths of filler are created within epoxy matrix. Only for concentration of 30 vol.% in both cases thermal conductivity decreases. The reason of this behaviour is existence of porosity in samples with 30 vol.% concentration and very high viscosity.

![Thermal conductivity graph](image)

**Figure 4.** Results of thermal conductivity measurements obtained with Hot Disc analyzer and PPMS device in room temperature for composites filled with BN PT 100.

The results obtained for BN PTX 25 (Figure 5) indicate that spherical particles are optimal for creation of more effective paths for heat transport. Phonon-scattering across the interfaces between polymer and filler surface play a significant role. Spherical agglomerates contacted with polymer have smaller interface surface than dispersed flakes particles and consequently
smaller phonon scattering. However BN PT100 during preparation can also create agglomerates. Results obtained using two different methods give comparable results.

![Figure 5. Results of thermal conductivity measurements obtained with Hot Disc analyzer and PPMS device at room temperature for composites filled with BN PTX 25](image)

**4 Conclusions**
The aim of this study was to investigate the influence of filler content on the effective thermal conductivity of the obtained epoxy based composites. It was observed that thermal conductivity increases with increased filler content. Additionally particles shape influence on thermal properties was observed. Filler able to create spherical agglomerates, namely BN PTX25, showed slightly better thermal conductivity results than plate-like PT 100. Such behavior can be explained by the fact that flakes particles have larger interfacial areas than spherical agglomerates what leads to creation of the thermal resistance and causes increase of the phonon scattering. The obtained results shows that optimization of thermal conductivity of filled epoxy composites is crucial from application point of view.

**References**


