THERMALLY CONDUCTIVE BUT ELECTRICALLY INSULATING PLASTICS FOR THERMAL MANAGEMENT APPLICATIONS

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Abstract

Boron nitride (BN) is a synthetic ceramic material that exhibits both excellent thermal conductivity and dielectric properties. Loading BN into thermoplastic resins therefore enables unique composite materials that are thermally conductive but electrically insulating. However, compounding BN powders into plastics presents various challenges and the composites exhibit anisotropic behavior due to the platelet structure of BN crystals. Optimal processing methods and ways to maximize thermal conductivity of BN-plastic composites in the desired directions are reported in this paper. Predictions from the Lewis Nielsen model with one fit parameter (“A”) are compared to the thermal conductivity data.

Introduction

The ever-increasing use of electronic components and light emitting diodes (LEDs) in vehicles has elevated the need for thermal management therein. In addition to electronics and LEDs, thermal management problems are also widely prevalent in the power & battery systems of hybrid vehicles. In each case, inadequate or inefficient thermal management has resulted in higher operating temperatures that potentially lead to sub-optimal performance and sharply reduced lifetimes of the various components. Thermal management has therefore become an integral design consideration in the automotive industry today.

An important component of the thermal management solution is the heat sink. The main function of a heat sink is to move the dissipated heat away from the source and spread it out over a larger area to enable transfer to the ambient. There are several material options for the heat sink, but aluminum is largely the first choice for thermal management engineers due to its relatively light weight and high thermal conductivity. However, thermally conductive plastics have recently gained attention as a viable alternative to aluminum. Plastics offer various advantages over aluminum, most notably weight savings, design flexibility and parts integration. Design flexibility is especially important in the case of heat sinks, since the complex fin structures that are sometimes required may not be easily achievable with aluminum without costly secondary machining and processing. Detractors from thermally conductive plastics may point to their limited thermal conductivity, nominally 2 – 40 W/mK, which is significantly lower than aluminum at 200 W/mK. However, it has been recently demonstrated that moderate
thermal conductivities may be sufficient for thermal management in convection-limited environments [1,2]. It is important to note that even at 2 W/mK, the thermal conductivity is nearly ten times that of the neat resin thermal conductivity, and that may be sufficient in some applications.

**Background**

Thermally conductive plastics are made by incorporating high thermal conductivity fillers into the thermoplastic matrix. Graphite, expanded graphite and carbon fibers are commonly used additives, but these result in black and electrically conductive composites. In contrast, boron nitride based composites are white and electrically insulating. Boron nitride is a synthetic ceramic that is both an excellent conductor of heat and a dielectric material. A few other ceramic materials such as alumina, aluminum nitride, silica etc. exhibit similar characteristics, but the hexagonal allotrope of boron nitride ("BN") has the highest thermal conductivity (albeit anisotropic) and it is also much softer than virtually all of these other materials. Table 1 compares the material properties of BN to other similar, competing fillers.

*Table 1: Material properties of BN compared to competing fillers*

<table>
<thead>
<tr>
<th></th>
<th>BN</th>
<th>Al₂O₃</th>
<th>AlN</th>
<th>SiO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal conductivity (W/mK)</td>
<td>300</td>
<td>30</td>
<td>260</td>
<td>1.3</td>
</tr>
<tr>
<td>Specific heat capacity (J/gK at 25 °C)</td>
<td>0.81</td>
<td>0.79</td>
<td>0.73</td>
<td>0.69</td>
</tr>
<tr>
<td>Theoretical density (g/cc)</td>
<td>2.28</td>
<td>3.98</td>
<td>3.26</td>
<td>2.20</td>
</tr>
<tr>
<td>Dielectric constant</td>
<td>3.9</td>
<td>9.7</td>
<td>8.8</td>
<td>3.8</td>
</tr>
<tr>
<td>Volume resistivity (Ω cm)</td>
<td>10¹⁵</td>
<td>10¹⁴</td>
<td>10¹⁴</td>
<td>10¹⁴</td>
</tr>
<tr>
<td>Knoop Hardness (kg/mm²)</td>
<td>11</td>
<td>1500</td>
<td>1200</td>
<td>500</td>
</tr>
<tr>
<td>Mohs hardness</td>
<td>&lt; 2</td>
<td>&gt; 9</td>
<td>~ 7</td>
<td>~ 6.5</td>
</tr>
</tbody>
</table>

As can be seen in Table 1, BN has several desirable properties compared to other similar fillers. The softness of BN potentially enables applications in plastics, since BN can be compounded into plastics and such composites can generally be injection molded with minimal wear and damage to processing equipment.

The fundamental crystal structure of hexagonal BN is shown in Figure 1. The a-b crystal planes are made of tight, covalently-bonded boron and nitrogen atoms. The a-b planes are repeated along the c-direction held together by weak van der Waal’s forces. This crystal structure leads to inherent anisotropy in the material and hence several physical properties, such as thermal conductivity, coefficient of expansion, refractive index, etc., are different in the a-b plane versus the c-direction. This anisotropy is most notable in thermal conductivity where the in-plane thermal conductivity (i.e. in the a-b plane) is estimated to be as high ~300 W/mK, while the through-plane (in the c-direction) thermal conductivity is less than 10 W/mK [3, 4].
Due to the crystal structure shown above, the natural particle shape of hexagonal BN particles is a platelet. Figures 2(a) and 2(b) show SEM images of grades PT110 and PT120 (Momentive Performance Materials) that are examples of platy BN crystals.

As seen in Figure 2, BN crystal platelets are not isometric and have a high aspect ratio. The aspect ratio, defined as ratio of crystal diameter to crystal thickness, is ~20 for these materials. When anisotropic behavior is acceptable, the high aspect ratio of these BN fillers typically can
be leveraged to make composites with high in-plane conductivity, as will be discussed in later sections. But if more isotropic thermal conductivity is desired, agglomerate grades of BN might also be considered. Agglomerate grades, as shown in Figure 2(d) (Grade PT350), consist of larger particles that are aggregates of smaller BN crystals. In the case of PT350, the average particle size is \(\approx 150 \mu m\), but the fundamental crystal size is only \(\approx 8 \mu m\). Within these larger aggregates, since the BN crystals are randomly oriented the agglomerate is significantly more isotropic than single crystal platelets. Such agglomerate grades have found application in thermal interface materials (TIMs) that are widely used for thermal management in the electronics industry. It is well known in these applications that maintaining agglomerate structure is critical to achieving high and isotropic thermal conductivity. Among the grades shown in Figure 2, CF600 (Figure 2(c)) is a mix of platelets and some small agglomerates. However, due to the fine particle size and lower tap density, it behaves much like the fine platelet grades such as PT120.

Platelet and agglomerate BN powders both present challenges at various processing steps. Platelet BN grades are extremely light, fluffy materials that are difficult to handle and to feed into compounding equipment. The higher surface areas typical of platelet BN powders and the inert surface chemistry of BN in general, make these materials difficult to wet out in extrusion. Improper wetting of the BN crystals by the resins leads to poor interfaces between the filler and matrix and results in inferior composite properties, including thermal conductivity. Agglomerate grades, on the other hand, are much easier to feed into the extruder due to their larger particle size and higher densities; the challenge in this latter case is to maintain the agglomerate structure through the various processing steps. If isotropic thermal conductivity is required, agglomerate grades may be the only recourse.

The Lewis Nielsen model has been used extensively to predict dynamic properties of composites [6]. Various authors have used this model to examine behavior of composites utilizing high-aspect ratio fillers and have successfully predicted the thermal conductivity in the direction of the aligned fillers [1, 7].

The equations of the Lewis Nielsen model are shown below:

\[ K_c = K_m \left( \frac{1 + AB \phi}{1 - B \psi \phi} \right) \]

Where

\[ B = \frac{\lambda - 1}{\lambda + A} \quad \lambda = \frac{K_f}{K_m} \quad A = 2 \frac{L}{D} \]

\[ \psi = 1 + \left( \frac{1 - \phi_m}{\phi_m^2} \right) \phi \]

In the above equations, \(K_f\) is the thermal conductivity of the filler, \(K_m\) is that of the matrix and \(K_c\) is final composite thermal conductivity. \(\phi\) is the volume fraction of the filler in the system and \(\phi_m\) is the maximum packing fraction for the filler system. For high aspect ratio fillers, \(A\) is related to the aspect ratio of the fillers as shown above, but is also dependent on the extent of orientation of the fillers. The Lewis-Nielsen model is especially useful and practical since it is an explicit equation for the composite thermal conductivity and predicts thermal conductivity over a wide range from \(\phi = 0\) to \(\phi \to \phi_m\).
It is important to note that this model assumes perfect interfaces between the filler and matrix and does not explicitly account for contact resistances between the matrix and fillers. Hence the model is independent of filler particle size and depends only on the volume fraction of the filler.

**Materials and methods**

**Extruded & injection molded samples**

The grade names and typical properties of various BN powder grades are listed in Table 2. The BN powders were compounded into polycarbonate (“PC”, Lexan® HF1110 from SABIC Innovative Plastics), Nylon 6 (“PA6”, Chemlon® 212/212H from Teknor Apex) and Nylon 66 (“PA66”, Chemlon® 100 from Teknor Apex) on Steer Omega-20® (20 mm) and Steer Omega-40® (40 mm) twin screw extruders at Steer America’s Application Development Lab in Uniontown, OH.

<table>
<thead>
<tr>
<th>BN grade</th>
<th>Oxygen (wt%)</th>
<th>Surface Area (m²/g)</th>
<th>Tap density (g/cc)</th>
<th>Crystal size (μm)</th>
<th>Particle size (D50, μm)</th>
<th>Powder type</th>
</tr>
</thead>
<tbody>
<tr>
<td>PT110</td>
<td>&lt; 0.6</td>
<td>0.7</td>
<td>0.70</td>
<td>50</td>
<td>50</td>
<td>Platelet</td>
</tr>
<tr>
<td>PT120</td>
<td>&lt; 0.6</td>
<td>4.0</td>
<td>0.40</td>
<td>12</td>
<td>12</td>
<td>Platelet</td>
</tr>
<tr>
<td>CF600</td>
<td>&lt; 0.6</td>
<td>6.0</td>
<td>0.45</td>
<td>8</td>
<td>16</td>
<td>Mixed</td>
</tr>
<tr>
<td>PT350</td>
<td>&lt; 0.6</td>
<td>4.0</td>
<td>0.70</td>
<td>8</td>
<td>125</td>
<td>Agglomerate</td>
</tr>
</tbody>
</table>

Typical data are average values. The actual values may vary and should not be used as specifications.

The compounding was carried out in two configurations:

a. A general purpose mixing screw with multiple kneading block and all fillers were fed at the feed throat along with the polymer pellets, is referred to as “Baseline method”, and

b. An optimized configuration where the fillers were fed into the extruder using a side-feeder, downstream of the kneading blocks, is referred to as “Optimized method”.

Pellets of the various formulations were collected from the extruder and injection molded at the University of Akron on a Van Dorn 55 Ton Injection Molder at 1 inch/s to make tensile bars. Samples were obtained from tensile bars at the location shown in Figure 3 and thermal conductivity was measured by the laser flash method (Figure 4).

The through-plane thermal conductivity measurement was relatively straightforward utilizing the configuration shown in Figure 4(a). The in-plane thermal conductivity was measured in two ways – in the first method, pellets were compression molded to a thin plaque ~0.3 mm and measured using the in-plane sample holder on an LFA447 Nano Flash® from Netzsch Instruments, shown in Figure 4(b). The other method was to construct a special “laminate” sample as shown in Figure 4(d) and the in-plane conductivity was measured using a laminate sample holder (Figure 4(c)). Both methods were in reasonable agreement. The data in the rest of the paper show the in-plane conductivity as measured by the first method, i.e., compression molding and using a modified laser flash method.
The experimental results and the corresponding model predictions are discussed separately for platelet, agglomerate and mixed BN powder grades.

**Platelet BN grades: PT110 and PT120**

The in-plane and through-plane thermal conductivity of PT110 processed in PC (HF1110) with the Baseline method (Page 5) is shown in Figure 5.
The thermal conductivity is significantly higher in the in-plane direction. The platelet structure of the BN crystals typically enables significant alignment of BN particles during injection molding and the aligned structure is frozen as the part cools in the mold. This aligned structure, along with the inherent anisotropy of BN crystals, has been shown to lead to highly anisotropic thermal conductivity in the final composites.

Similar data for PT120 processed using the Baseline method are shown in Figure 6. While the thermal conductivity performance is comparable at 30 wt% and 40 wt%, it is important to note that the highest loading of PT120 could not be processed with the Baseline configuration due to issues with feeding the material into the extruder. The low tap density of PT120 (<20% of BN’s theoretical density) indicates that large volumes of air will be displaced and released when the powder is wet out by the resin. Non-optimal screw elements at the feed throat and insufficient venting led to inconsistent feeding of PT120 into the extruder, leading to surging and blocked dies at 50wt% loading.

**Lewis Nielsen model for platelet BN grades:**

The in-plane thermal conductivity data presented above for PT110 and PT120 were fit with Lewis Nielsen model using the built-in Solver function in Microsoft Excel† 2010 to minimize the error of the prediction with $A$ as a fit parameter. The rest of the fixed parameters used for the Lewis Nielsen model and the densities of the component materials (to convert from weight fraction to volume fraction) are shown in Table 3, consistent with values suggested by other authors.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>$K_m$ (W/mK)</th>
<th>$K_f$ (W/mK)</th>
<th>$\rho_{BN}$ (g/cc)</th>
<th>$\rho_{PC}$ (g/cc)</th>
<th>$\phi_m$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Value</td>
<td>0.2</td>
<td>200</td>
<td>2.28</td>
<td>1.20</td>
<td>0.80</td>
</tr>
</tbody>
</table>
Figure 6: In-plane and through-plane conductivity of PC-PT120 composites processed using the Baseline method

Table 4: In-plane thermal conductivity predicted by Lewis Nielsen model versus measured values for PT110 and PT120

<table>
<thead>
<tr>
<th>BN grade</th>
<th>Weight fraction ($\omega_{BN}$)</th>
<th>Volume fraction ($\phi_{BN}$)</th>
<th>$\psi(\phi_{BN})$</th>
<th>$K_c$ predicted (W/mK)</th>
<th>$K_c$ measured (W/mK)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PT110</td>
<td>0.32</td>
<td>0.20</td>
<td>1.06</td>
<td>2.45</td>
<td>2.36</td>
</tr>
<tr>
<td>PT110</td>
<td>0.40</td>
<td>0.26</td>
<td>1.08</td>
<td>3.38</td>
<td>3.38</td>
</tr>
<tr>
<td>PT110</td>
<td>0.50</td>
<td>0.34</td>
<td>1.11</td>
<td>4.97</td>
<td>5.06</td>
</tr>
<tr>
<td>PT120</td>
<td>0.30</td>
<td>0.18</td>
<td>1.06</td>
<td>2.18</td>
<td>2.10</td>
</tr>
<tr>
<td>PT120</td>
<td>0.40</td>
<td>0.26</td>
<td>1.08</td>
<td>3.31</td>
<td>3.43</td>
</tr>
</tbody>
</table>

Note: Test data. Actual results may vary.

Table 4 shows the thermal conductivity predicted by the Lewis Nielsen model using the best fit values for A for PT110 and PT120. A was chosen to be 45.6 and 44.6 for PT110 and PT120 respectively (obtained as described earlier) and the model prediction agreed extremely well with the data with just one fit parameter. It is important to note that the best fit value of A of ~45 is very much in the ball-park of what one might estimate from the crystal dimensions as discussed earlier. The results for PT110 versus the predicted values are also shown graphically in Figure 7.
Predicting the through-plane conductivity of these composites is not covered in this paper since it would involve a more detailed analysis of platelet orientation in the through-plane direction as described by Heinle et al [7].

**Agglomerate BN grade: PT350**

Table 5 shows the in-plane and through-plane conductivities of PT350 composites with PC, PA6 and PA66 processed with the Baseline and Optimized screws at 45wt% PT350 loading.

*Table 5: In-plane, through-plane thermal conductivities and their ratio for various PT350 composites manufactured by Baseline and Optimized extrusion methods*

<table>
<thead>
<tr>
<th>Row</th>
<th>Resin</th>
<th>Screw Config</th>
<th>Thru’plane TC (W/mK)</th>
<th>In-plane TC (W/mK)</th>
<th>Ratio of IP:TP</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>PA66</td>
<td>Baseline</td>
<td>1.02</td>
<td>4.26</td>
<td>4.18</td>
</tr>
<tr>
<td>2</td>
<td>PA66</td>
<td>Optimized</td>
<td>1.33</td>
<td>4.29</td>
<td>3.23</td>
</tr>
<tr>
<td>3</td>
<td>PA6</td>
<td>Baseline</td>
<td>0.87</td>
<td>4.66</td>
<td>5.36</td>
</tr>
<tr>
<td>4</td>
<td>PA6</td>
<td>Optimized</td>
<td>1.17</td>
<td>4.06</td>
<td>3.47</td>
</tr>
<tr>
<td>5</td>
<td>PC</td>
<td>Baseline</td>
<td>0.88</td>
<td>3.88</td>
<td>4.41</td>
</tr>
<tr>
<td>6</td>
<td>PC</td>
<td>Optimized</td>
<td>1.08</td>
<td>3.49</td>
<td>3.23</td>
</tr>
</tbody>
</table>

*Note: Test data. Actual results may vary.*

The Optimized extrusion process showed significantly improved through-plane conductivity performance, since agglomerate integrity was retained better in this process set up. It is also interesting to note a corresponding drop in the in-plane thermal conductivity. Examining the ratio of in-plane to through-plane conductivity showed a clear difference between the Baseline and Optimized configurations where the Optimized configuration delivered more isotropic thermal conductivity composites.
**Lewis Nielsen model for agglomerates**

The thermal conductivity data for PT350-based composites were not analyzed due to subtleties in calculation of the volume fraction of the agglomerate powders. Since the agglomerate particles are porous, the volume fraction occupied by these agglomerates is larger than what one would estimate using the theoretical density of BN for a given weight loading. The “envelope density” of the agglomerates related to the porosity of the particles would be necessary in order to calculate the volume fraction of PT350. This exercise is beyond the scope of the current paper.

However a closer examination of this model provides some insight into the results shown in Table 5. If the Optimized extrusion method did indeed retain the agglomerate shape better than the Baseline method, it is reasonable to expect that the value of A might be effectively lower for the Optimized method, since A is ~2.0 for spherical fillers. The effective aspect ratio of PT350 particles in the composite might therefore be lower with the Optimized method than the Baseline method, thus might lead to more isotropic conductivity and lower in-plane thermal conductivity.

**Mixed BN grade: CF600**

The potential efficacy of the Optimized extrusion method is also indicated by the fact that it potentially enables significantly improved handling and processing of CF600. CF600 is a fluffy, higher surface area grade of BN with poor flow characteristics that are difficult to process using the Baseline extrusion process. The results of PC-CF600 composites compounded using the Optimized method are shown in Figure 8.

The data in Figure 8 are consistent with the previously described expectation that CF600 may be likely to behave more like a platelet grade than an agglomerate grade. We can also see this in the SEM images (Figure 2) and typical characteristics of CF600 listed Table 1. Fitting the Lewis Nielsen model to the in-plane data in Figure 8 yields a value of 52.3 for A for CF600.

![Figure 8: In-plane and through-plane conductivity of PC-CF600 composites](image)

*Note: Test data. Actual results may vary.*
Conclusions and future work

An optimized extrusion method to process BN powders has been developed. The optimized method potentially enabled both high through-plane conductivity with BN agglomerates and stable processing of difficult-to-process BN powders such as fine platelet grades. Predictions of the in-plane thermal conductivity from the Lewis Nielsen model matched well with experimental results with just one fit parameter $A$. The best fit value of $A$ is ~45 for PT120 and PT110, and ~52 for CF600. Future work would include predictions with the Lewis Nielsen model in other resin systems, through-plane conductivity of composites with platelet grades, and tailoring the model to accommodate agglomerate grades of BN.

Acknowledgements

Momentive would like to thank the State of Ohio and the Ohio Department of Development for their support. This work was funded in part by the Ohio Third Frontier Grant TECH 10-081. The authors would also like to thank Shayma Mouhammed and Yu Chen for their help with thermal conductivity measurements.

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References

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