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Low-cost, high-performance thermally conductive plastics

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Surface treatments and boron nitride-based mixed filler formulations are used to address property and cost challenges.

Boron nitride (BN) is a synthetic ceramic material with unique properties, which has attracted growing scientific and commercial interest. It enables resin formulations with high thermal conductivity (TC) that are also electrically insulating. Several thermally conductive plastics (TCPs) containing BN have been studied previously, but their high cost has limited their large-scale commercial adoption.[1–4]

Typical applications of BN-based TCPs include heat sinks for LEDs and other heat sources, battery housings, electronic device housings, temperature sensors, heat exchangers, and cooling systems. As devices become smaller and gain more functionality, demand for these applications is growing. Replacing aluminum with TCPs would have the advantages of light weight as well as design and color freedom. As costs come down, TCPs will also become more viable.

We have explored using BN powders with other thermally conductive and electrically insulating additives to enable more cost-effective formulations with improved physical properties and rheologies.[5] We prepared two types of thermoplastic composites filled with BN: one with polyamide (PA6) resin from Teknor Apex and one with polycarbonate (PC) resin from Sabic. We used and compared a number of high-purity BN powder grades: HCPL, PolarTherm PT100, and CoolFlow CF600 (all Momentive). For our laboratory-scale evaluations, we compounded the BN powders and other additives into the various plastic resins using a Brabender Plasti-Corder Intelli-Torque batch mixer. The PA6 and PC formulations were compounded at 265°C and 280°C, respectively. We mixed the formulations at 75rpm for five minutes under a nitrogen purge. After compounding, the samples were compression molded to a thickness of about 0.3–0.4mm using a Carver Monarch compression molder.

We investigated the effect on the resin formulation properties when we included a variety (alone, or in combination) of different surface treatments. These surface treatments included chopped glass fiber (GF) strands from Johns Manville, zinc oxide (ZnO) from US Zinc, and one of three silanes (organic compounds of silicon with reactive alkoxy groups). The silanes we studied were methacryloxy silane (MS), thiocarboxylate silane (TCS)—both from Momentive—and a halogen-containing silane (HS) from Sigma Aldrich. A list of the formulations that provided the optimal balance between properties and cost are given in Table 1. These were scaled up for twin screw extrusion and injection molding. The silane, where used, was loaded at 3wt% of the filler formulation. We chose ZnO and glass fibers to contribute to TC without affecting electrical insulation. The silane surface treatment improved dispersion, which in turn had a positive effect on both the thermal and mechanical properties of the compound. We also measured the tensile strength, extension at break, and tensile modulus of the resins using an Instron 4465 instrument with an extension rate of 0.2inch/min. We cut 2.5-inch bars from the middle impact bars and notched them on a Testing Machines, Inc. (TMI) notching cutter. In addition, we used a TMI impact tester—with a 1ft-lb pendulum—to measure the Izod impact strength, and we observed complete breaks in all cases.

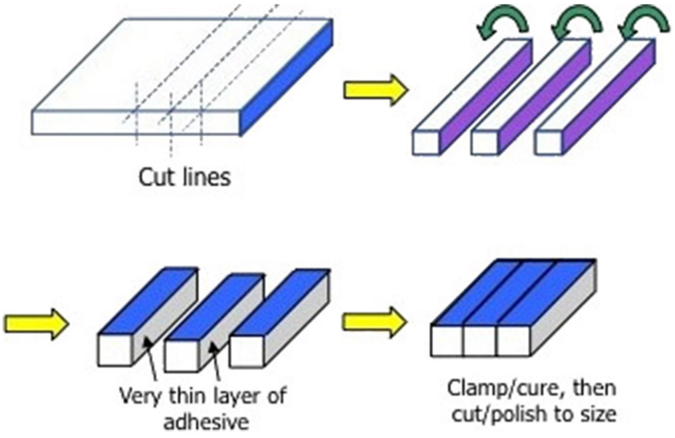

Figure 1. *Method to make laminate for in-plane thermal conductivity (TC) measurements.*

We used the laser flash method to conduct TC measurements. For the laboratory-scale samples, we used a modified sample holder and mask (from Netzsch) to measure the in-place TC. For the through-plane TC measurements of the injection-molded samples, we drilled half-inch

Continued on next page

Table 1. *Resin formulations scaled up for twin screw extrusion, as well as their in-plane TC values and measured physical properties. Form#: Formulation reference number. Res.: Resin. BN: Boron nitride. ZnO: Zinc oxide. GF: Chopped glass fiber strands. PA6: Polyamide 6. PC: Polycarbonate. CF600: Coolflow CF600 grade of BN. HCPL: BN grade of single-crystal hexagonal platelets refined for a low surface area. PT100: Polartherm PT100 grade of BN. TCS: Thiocarboxylate silane. MS: Methacryloxy silane.*

Form#	Res.	BN Grade	BN (wt%)	ZnO (wt%)	GF (wt%)	Silane	Tensile Strength (psi)	TC (W/mK)	Elongation at Break (%)	Notched Izod Impact Strength (J/m)	Viscosity ($\sim$10s^{-1}, Pa.s)
1	PA6	CF600	41%	–	–	–	8747	3.2	1.4	21	–
2	PA6	CF600	50%	–	–	–	6500	4.9	0.8	15	–
3	PA6	HCPL	40%	10%	7.5%	TCS	9713	4.8	1.1	32	–
4	PA6	CF600	20%	50%	–	TCS	7579	5.1	0.9	28	–
5	PC	PT100	39%	–	–	–	6857	3.3	–	–	529
6	PC	PT100	50%	–	–	–	5807	4.9	–	–	1120
7	PC	PT100	43%	–	–	MS	6736	4.9	–	–	552
8	PC	PT100	30%	–	20%	–	8550	3.5	–	–	–

discs from the tab section of the dog-bone tensile bars away from the mold gate. We made laminates (10mm × 10mm)—also from the tab portion of the dog-bone bars—to measure the in-plane TC (see Figure 1). Using this procedure, the in-plane TC was measured using a through-plane fixture (because the tensile bars were too thick for a direct in-plane measurement). Such a direct measurement would have required samples with 1 inch diameters and thicknesses less than 0.4mm, as described above. The laminates were made in the in-plane, ‘cross-flow’ direction. The through-plane TC values we report are the average of measurements on three discs for each sample. Our in-plane results are reported from the average of two laminates for each sample. For all TC measurements, the theoretical specific heat capacity was calculated for each formulation based on the composition.

The final physical properties we measured after injection molding are also shown in Table 1. Comparing the properties of the different formulations shows that silane can significantly increase the TC of the formulations and also reduce the viscosity. With the correct silane treatment, it was possible to achieve a given TC with a lower BN loading, which directly reduces the formulation cost. However, an incorrect choice of silane may have an adverse effect on the TC of the composite (data not shown). Excessive use of the silane, or insufficient silane treatment, both led to suboptimal TC performance. As indicated by the results in Table 1, it is possible—with intelligent formulation—to lower the amount of BN and yet achieve high TC. We also find it interesting that there were significant improvements in typical physical properties with these mixed filler formulations.

In summary, we have investigated the benefits of surface treatments on TC, rheology, and physical properties of BN-thermoplastic composites. Our results show that surface treatment with the correct silane and loading level can both increase the TC and lower the viscosity of BN-filled thermoplastic composites. We have also explored the use of ZnO and GF in combination with BN. The results show that mixed filler formulations may be a practical approach to achieving cost-effective formulations of TCPs, while also delivering significant improvements in typical physical properties. We are now concentrating on better understanding the applications for thermally conductive and electrically insulating plastics.

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Chandra Raman is leading a team of application development engineers while continuing to work closely with Momentive’s customers to support their development efforts. His research focuses on the use of BN powders in thermally conductive plastics, driven by growing demand for LED bulb housings and other applications.

Continued on next page

Bei Xiang is the application development engineer dedicated to assisting Momentive's customers with the use of BN powder in polymer-based thermal management solutions. He has extensive research experience in polymer synthesis and functionalization, as well as in solid phase chemistry. His current focus is on the preparation, modification, and application of BN powder as a filler in polymer-based thermal interface materials for applications in electronics and LED lighting.

Anand Murugaiah has a background in materials science and extensive experience in high-temperature materials synthesis, as well as in ceramics and composites processing and characterization. He has worked on BN materials, processes, and their applications for over 10 years. He is currently leading a project focusing on next-generation BN materials and their applications including in polymers.

Allison Howard is currently leading the global marketing efforts of Momentive's ceramics business unit. Her focus is on BN technology for thermoplastics applications to increase thermal conductivity, improve lubrication, and/or enhance nucleation in a wide range of resins.

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