# Effect of the elongational flow on mechanical properties and thermal conductivity of polypropylene-boron nitride composite fibers

# EFFECT OF THE ELONGATIONAL FLOW ON MECHANICAL PROPERTIES AND THERMAL CONDUCTIVITY OF POLYPROPYLENE-BORON NITRIDE COMPOSITE FIBERS

R. Arrigo[1], A. Frache[1], G. Malucelli[1]
[1] Dipartimento di Scienza Applicata e Tecnologia, Politecnico di Torino, Viale Teresa Michel 5, 15121 Alessandria, Italy
e-mail: rossella.arrigo@polito.it

## Introduction

Polymers are broadly employed in advanced technological applications, ranging from organic electronic to wereable computing devices, owing to their unique properties such as low cost, lightweight and easy processing. Nowadays, however, due to the rapid development in integration and miniaturization of electronic devices, high performance thermally conductive materials are required for thermal management. Therefore pure polymers, due to their low thermal conductivity, cannot be exploited in several functional applications [1]. The formulation of polymer-based composites containing thermally conductive fillers such as boron nitride (BN) may offer a solution to this drawback; unfortunately, adequate values of thermal conductivity are usually obtained through the incorporation of high amounts (about 50 wt.%) of fillers, which causes a significant modification of the matrix rheological behavior, hence compromising its processability. Furthermore, high filler loadings usually induce particle agglomeration phenomena, thus worsening the polymer mechanical properties. Therefore, several strategies have been pursued for obtaining highly conductive polymer-based composites, while maintaining adequate loadings of fillers. Most of them concern the exploitation of filler orientation and alignment, as well as of tailored interfacial properties for the creation of thermally conductive patterns within the host polymer matrix which allow achieving an effective thermal transport through the material, while maintaining adequate processability and mechanical performance. Among the aforementioned methods, filler alignment is less exploited, due to technical difficulties in controlling and governing this phenomenon. Different approaches have been proposed to prepare thermally conductive polymer composites with aligned fillers, including the use of prealigned fillers (e.g., carbon nanotube arrays) or of magnetic-induced alignment methods. However, all these strategies present several issues, such as the use of solvents and/or the need of chemical functionalization of the fillers, causing complex process management and low production efficiency [2]. As widely documented in the literature, the application of the elongational flow during the processing of polymer-based composites is effective in inducing a preferential orientation of the embedded particles along the flow direction, especially when the stretching is applied in non-isothermal conditions [3].

In this work, we propose an easy and industrially viable method to obtain polypropylene (PP)-based composites containing high loadings of BN (up to 30 wt.%) with enhanced thermal conductivity and mechanical properties. In particular, PP+BN composites obtained through melt compounding in a twin-screw extruder (Process11, Thermofisher) were subjected to uniaxial elongational flow at the exit of the extruder through the utilization of a Reospin elongational rheometer. This way, composite fibers at different draw ratio (DR) were obtained and fully characterized through reological, mechanical and morphological analyses. Besides, the thermal conductivity of the fibers was assessed and compared to that of isotropic as-extruded materials.

## Results and discussion

Figure 1 plots the complex viscosity curves as a function of frequency for unfilled PP and BN-based composites. The introduction of increasing loadings of BN causes a progressive rise of the complex viscosity values, though composites containing up to 10 wt.% of BN show a rheological behavior quite similar to that of the unfilled PP. At variance, PP+20BN and, especially, PP+30BN systems exhibit a pronounced non-Newtonian behavior, involving the occurrence of an apparent yield stress at low frequencies; this phenomenon can be associated with the restriction of the dynamics of PP macromolecules resulting from the established filler/filler and polymer/filler interactions.

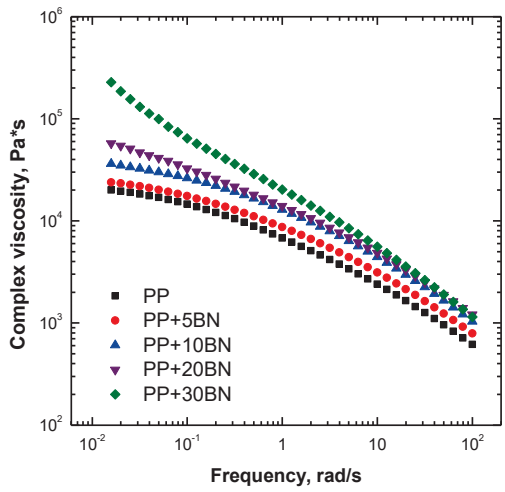


Figure 1: Complex viscosity curves for BN-containing composites and unfilled PP.

Composite fibres at different DR values were obtained using the elongational rheometer and characterized through tensile tests; the trends of the dimensionless elastic modulus (obtained as the ratio between the value of the modulus at a specific DR and that of the isotropic sample) as a function of DR for unfilled PP and BN-containing composites are reported in Figure 2. For all investigated systems, the dimensionless elastic modulus shows an increasing trend as a function of DR, due to the progressive alignment of the polymer chains along the fiber direction, notwithstanding the greater growth rate for the fibers containing high amounts of BN particles (i.e., beyond 20 wt.%). This finding can be explained considering the well-documented ability of the elongational flow in ameliorating the morphology of polymer-based composites. In particular, the application of the elongational flow during processing (especially in non-isothermal conditions) promotes the distruption of the filler agglomerates possible present in the isotropic systems, as well as a preferential orientation of the embedded fillers along the flow direction [3].

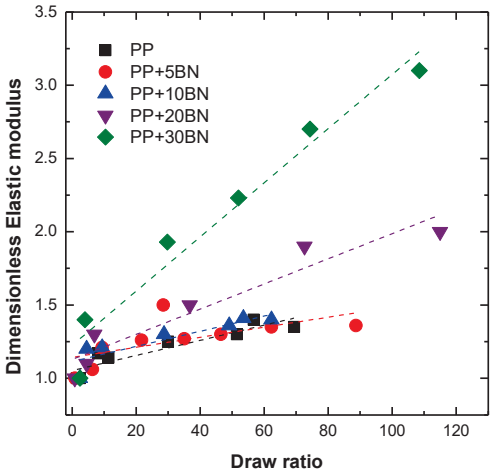


Figure 2: Elastic modulus as a function of draw ratio for BN-containing composite fibres and unfilled PP.

The beneficial effect of the non-isothermal elongational flow on the morphology of the formulated composite fibers was confirmed through SEM observations. The micrographs presented in Figure 3 clearly indicate that in the PP+30BN fibers the filler agglomerates present in the isotropic material were no longer observable; furthermore, a preferential orientation of the embedded BN particles can be detected. The elongational flow-induced morphology enhancement (further proven through XRD analyses performed on stretched samples), also promoted an enhancement of the thermal conductivity of the materials. In fact, PP+BN composites subjected to elongational flow exhibited enhanced thermal conductivity as compared to their as-extruded counterparts; in particular, PP+30BN fibers showed a thermal conductivity of 0.723 W/mK, with an increment of 173% with respect to unfilled PP sample. Finally, electrical meaurements confirmed that the introduction of BN does not modify the electrical insulation of the PP matrix, highlighting the effectiveness of the proposed strategy in the obtainment of polymer-based materials potentially suitable for applications requiring effective thermal management.

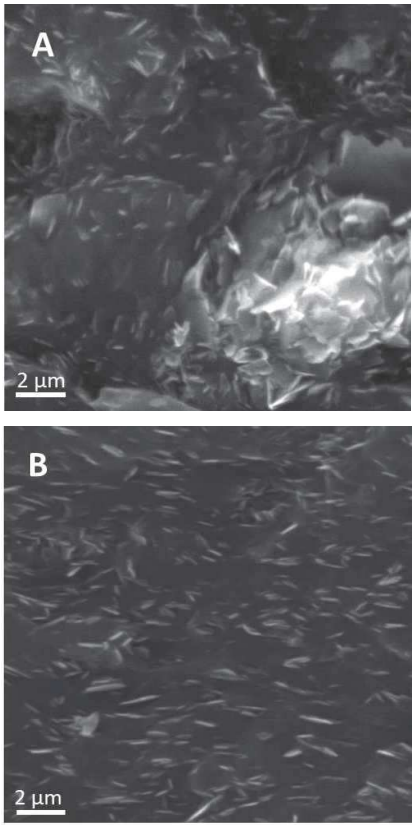


Figure 3: SEM micrographs for (A) as-.extruded and (B) drawn (DR=80) PP+30BN sample.

## Conclusions

This work clearly demonstrated the effectiveness of the application of non-isothermal elongation flow for the obtainment of PP-based composites containing high loadings of BN with enhanced thermal conductivity and mechanical properties. In particular, PP+BN fibers exhibited a progressive increase of the tensile properties as a function of the DR, as well as enhanced thermal conductivity as compared to their isotropic counterparts. These results were ascribed to the ability on the elongational flow in ameliorating the filler dispersion; in fact, the morphological characterizations pointed out the achievement of a more homogeneous morphology for the composites fibers, with the disappearance of BN agglomerates observed in the isotropic materials and some preferential orientation of the embedded fillers along the flow direction.

## References

[1] D. Zou, X. Huang, Y. Zhu, J. Chen, P. Jiang, *Comp. Sci. Technol.* 177, 88 (2019).
[2] T. Terao, C. Zhi, Y. Bando, M. Mitome, C. Tang, D. Golberg. *J. Phys. Chem. C* 114, 4340 (2010).
[3] R. Arrigo, G. Malucelli, F.P. La Mantia. *Polymers* 13, 3529 (2021).