EFFECTS OF FILLER GEOMETRY ON PHYSICAL PROPERTIES OF POLY(BUTYLEN THEREPHTHALATE)/CLAY NANOCOMPOSITES

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Introduction

During the last decade, polymer-clay nanocomposites have attracted intense interest in material science because of their superior engineering properties compared to neat polymers [1]. In particular, improvements have been extensively reported in terms of stiffness and strength [2], thermal properties and heat distortion temperature [3], resistance to flammability [4] and barrier performances [5] with effects of magnitude usually related to the states of dispersion of the filler in the polymer matrices and to its geometrical features.

In a previous research [6], authors focused their attention on poly(butylene terephthalate) (PBT) based materials, loaded with 6 wt% of a lamellar clay (Somasif™ MEE: organically treated, synthetic fluoromica, density: 2.60 g/cc, specific surface: 9 m^2/g) and opportunely toughened with 20 wt% of Pibiflex, a thermoplastic with high elastomeric properties, to prevent that the inclusion of the rigid phase would sacrifice the impact properties of the hosting matrix. Results demonstrated that the dynamic-mechanical behaviour of the PBT matrix is really influenced by both additives.

Now, to evaluate any influence related to the filler geometry on viscoelastic parameters of the matrix, systems having the same composition, prepared under the same condition but loaded with a tubular clay (halloysite) have been analysed. This additive, only recently commercialised but already widely used for the production of nano-structured polymeric materials [9,10], consists of natural clays mainly available in the form of nanotubes with a fairly high L/D ratio.

Experimental

Materials
In this work an injection moulding grade poly buthylen terephtalate (PBT) POCAN B 1505 supplied by Bayer wad used as the matrix.

The PBT was reinforced by including 6 wt% of halloysite nanoclays purchased by Aldrich (density 2.53 g/cc, surface area 64 m^2/g and mean nanotube sizes 30/0.25-4 μm (D/L)) and toughened with the same approach in ref. 6.

All materials, opportunely dried before processing, were melt compounded by using a Polylab Haake corotating twin-screw extruder (L/D=40/2.5), applying a screw speed of 50 rpm and the temperature profile 210–230–230–228 °C from the hopper to the die.

Apparatus and Procedures
The extrudates were pelletized at the die exit, dried and then injection moulded into standard rectangular bars (30 mm length, 5 mm width and 2 mm thickness) for further mechanical characterisations.

Dynamic Mechanical Analysis of all investigated materials was performed with a TRITON instrument (mod Tritec 2000) in the single cantilever mode.

The temperature dependence of the dynamic storage modulus (G') and the damping factor (tan δ) were measured at a frequency of 1 Hz, with a strain amplitude of 0.02%, over a temperature range from 30 °C to 180 °C and heating rate of 5 °C/min.

Results and Discussion

The dynamic mechanical properties of all the composites were compared with those of the neat matrix, processed under the same conditions. The variation of the storage modulus (G') as a function of the temperature for halloysite based compounds can be seen in Fig. 1. It is evident that addition of the elastomeric Pibiflex yields to a relevant reduction of this dynamic parameter. This adverse effect, uniform all over the examined temperature range and explainable in terms of increased flexibility of the compounds, is almost totally recovered in particulate filled composites.

In Fig. 2 the temperature dependence of the damping factor (tan δ) for the investigated system is reported. Clearly, a single signal is always observed with a peak centred at 70 °C. In this case, addition of both additives does not seem to influence this signal significantly. In particular, the inclusion of Pibiflex leads to a slight upword shift and broadening of the signal as a sign of an increased structure flexibility. Again, the simultaneous presence of the rigid nanoclay resets the height of the matrix damping signal that remains slightly broadened.
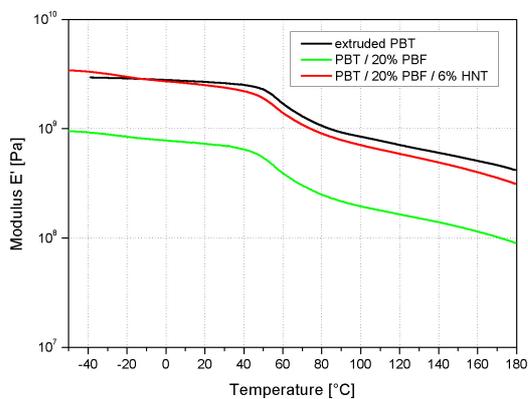

Fig. 1 Comparison of dynamic storage modulus curves
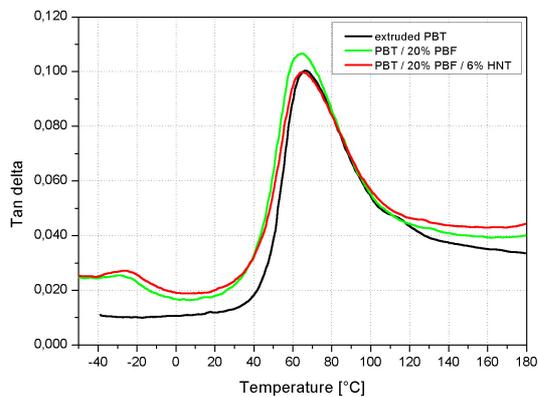

Fig. 2   Temperature dependence of the damping factor for investigated systems

Finally, with the aim to emphasize the effect of the filler geometry on dynamic-mechanical behaviour of polyester based compounds, Fig. 3 compares above cited results with data reported in ref. 6.

A greater effect on the viscoelastic properties of the matrix is verified in presence of the lamellar additive. This behaviour certainly linked, under the same process conditions, to a different level of achieved dispersion, could be attributed to a partial failure of the halloysite nanotubes: consideration to be verified by further structural and morphological investigations.

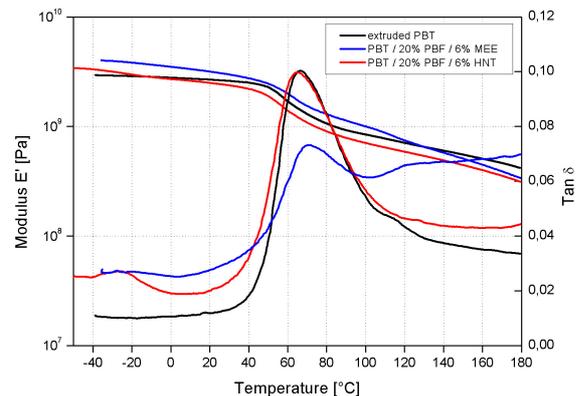Anyway evidences will be supported also by static mechanical tests.

Fig. 3   Storage modulus and tan δ curves of ternary compounds compared with the reference material ones

References
 1. Pinnavaia, T.J., Beall, G.W. "*Polymer-clay nanocomposites*", Editors Pinnavaia, T.J. and Beall, G.W., John Wiley & Sons, Inc., N.Y. (2000).
 2. Vaia, R.A., Price, G., Ruth, P.N., Nguyen, H.T., Lichtenhan, J., Polymer/layered silicate nanocomposites as high performance ablative materials. *Appl. Clay Sci.* **15** (1999) 67-92.
 3. Kawasumi, M., Kohzaki, M., Kojima, Y., Okada, A. and Kamigaito, O., *United States Patent No. 4810734*, Assigned to Toyota Motor Co., Japan (1989).
 4. Gilman, J.W., Flammability and thermal stability studies of polymer-layered silicate (clay) nanocomposites. *Appl. Clay Sci.* **15** (1999) 31-49.
 5. Messersmith, P.B., Giannelis, E.P., Synthesis and barrier properties of poly(ε-caprolactone)-layered silicate nanocomposites. *J. Polym. Sci. Part A: Polym. Chem.* **33** (1995) 1047-1057.
 6. Acierno, D., Amendola, E., Russo, P., Dynamic-mechanical investigation of melt compounded poly(butylenterephthalate)/organoclay nanocomposites. *Macromol. Symp.*, **247** (2007) 156-161.