FULLERENES AND OTHER CARBON FILLERS APPLICATION FOR MODIFICATION STRUCTURE AND PHYSICO-MECHANICAL PROPERTIES OF POLYIMIDE COMPOSITES

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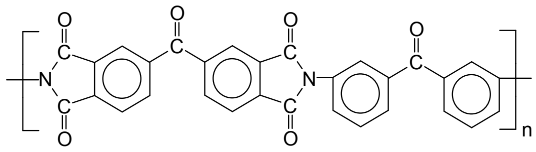
Introduction

Polyimides (PI) are among the most promising polymer for composite materials and moldings with high thermal stability (up to 500°C). Polyimides (especially crystallize) are widely used for tribological purposes as film coatings, moldings, and a matrices of composite materials [1].

Addition in polymer particles of different nature fillers increased strength and thermal stability, lead to optimal structure of matrices on intramolecular level [2]. Adding in polymer up to 10-15 wt.% of filler increase wear resistance in two orders without any notable change of friction coefficient. Degree of polymer's crystallinity does not change also and decreases only during friction processes. It was shown that friction surface became amorphous [3].

In this work crystallizable PI's were developed on the basis of a dianhydride of 3,3′,4,4′-benzophenonetetracarboxylic (BTDA) acid and meta-substituted diamine, in particular 3,3′-diaminobenzophenone (3,3′ DABP):



Structure, tribological and thermal properties of polymer molding based on (DABP-BTDA) and doped by fullerenes (C_{60}-C_{70}) and other carbon fillers (shungit) were investigated.

Result and discussion

BTDA-DABP PI powders obtained under the conditions of low temperature chemical imidization in solution in an amide solvent is semi crystalline and its melting temperature is unusually low for this class of polymers (T_{melt} =290°C). BTDA-DABP PI powders became amorphous after heating higher than 350°C and is characterized only be glass transition temperature T_g=245°C [4]. It was shown that chemically imidized PI could be processed into the matrix of a composite material and molding due to low melting temperature which is far from temperature of thermal destruction of PI. The mechanical properties of this polyimide molding parts are as follows at: flexural strength is 120 MPa, flexural modulus is 2.2 GPa, and elongation at break is 5.5 %.

To obtain fullerene-containing compositions of semi-crystalline BTDA-DABP PI, several methods of fullerenes introduction into the polymer were employed (table 1). A mixture of C_{60}-C_{70} fullerene in the weight ratio of 78:22% was used.

Table 1.

Sample
PI crystalline
PI amorphous
1M, fullerene added in diamine(0.015 %)
2M, fullerene added in diamine (0.1 %)
2M1 (sample.2M with phthalaldepyde (PhA)
3M, fullerene added in poly-amic acid(0.1 %)
3M1, fullerene added in poly-amic acid (1 %)
4M, after the introduction of the imidizing mixture (0.1 %)

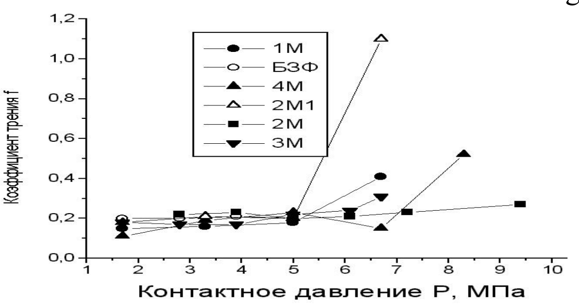
The DSC and WAWD data confirmed that degree of crystallinity in PI powders with fullerenes mixture was the same as in pure PI.

Melting temperature T_{m1} decreased to 280°C. Degree of crystallinity estimated by Sherrer's method was 20%.

Moldings, based on BTDA-DABT PI-fullerene compositions, were tested for a friction behavior. Fig.1 shows the coefficient of sliding friction (**f**) vs. pressure in tribocontact (**P**) for moldings based on polyimide-fullerene compositions and for a molding based on pure BTDA-DABT PI. Sliding speed was 0.18m/sec. It should be noted that up to a pressure of 5MPa friction coefficient for all compositions remains almost unchanged and is in the 0.15-0.2 range.

At contact pressures above 5 MPa, the coefficient of friction increases linearly only for the moldings, obtained by method 2M. Hence it was promising for application at contact pressure up to 8 MPa. Method 2M was the best method for introduction of fullerene mixture in PI. Moldings, obtained by other methods, are stable only up to 5 MPa.

Fig.1



(Contact pressure, MPa)

The molding sample obtained by method 2M was also optimal with respect to the second tribological characteristic-wear (wear decreased in three orders of magnitude).

Introduction of C_{60}-C_{70} fullerene mixture by mechanical blending did not get any advantageous comparison with chemical method neither wear resistance nor antifriction properties of obtained moldings. Increasing of wear resistance in twice times and decreasing of friction coefficient from 2 to 0.16 was observed only for moldings based on PI with 10 wt.% of shungit, prepared by mechanical blending.

Thermal properties of moldings were studied during friction under pressure at 5.5 MPa and sliding speed 0.18m/sec. The thermocouple was placed in the depth 1mm under friction surface.

The increasing of temperature during experiment (100sec.) was about 8-10^{o}C for materials with high wear resistance (optimal introduction fullerenes or shungit in PI). For materials with low wear resistance the temperature increased only 3-4^{o} C. This fact indicated that thermal conductivity in the first material was higher than in the second material. Consequently in the second case only thin layer combined heating energy and it leads to high wear. In friction surface in this case large wear particles appeared. Dimensions of wear particles for materials with low wear resistance were 30-60 microns, for materials with high wear resistance – 12-17 microns.

Conclusion

1. Several methods of fullerene introduction into PI BTDA-DABT were proposed. It was shown that in all cases fullerene containing polyimide had low melting temperature and the degree of crystallinity was about 20%. Hence, it can be processed into a molded product or a matrix of the composite
2. An optimal method of fullerene introduction into PI BTDA-DABT was found: fullerene in an o-dichlorobenzene solution is added to diamine solution. The molding obtained on the basis of this composition exhibited tribological properties (friction coefficient, mass wear) which exceeded those of both moldings based on pure PI BTDA-DABT and of other fullerene – containing compositions
3. It was shown the efficiency of mechanical blending PI with shungit for increasing wear resistance and antifriction properties of moldings.

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