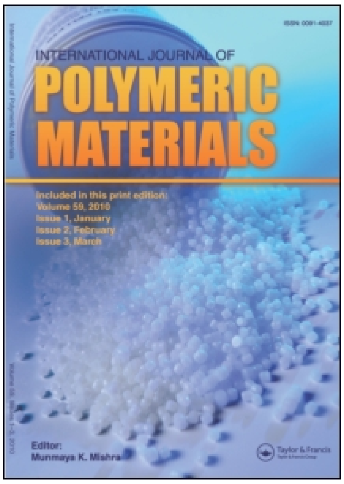

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H. Hargitai[a,b]; I. Rácz[a]
[a] Bay Zoltán Institute on Materials Science and Technology, Budapest, Hungary [b] Department of Polymer Engineering and Textile Technology, Technical University of Budapest, Hungary

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Influence of Water on Properties of Cellulosic Fibre Reinforced Polypropylene Composites

H. HARGITAI[a, b] and I. RÁCZ[a,*]

[a] *Bay Zoltán Institute on Materials Science and Technology, Fehérvári u 130, H-1116 Budapest, Hungary;* [b] *Technical University of Budapest, Department of Polymer Engineering and Textile Technology, Hungary*

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Flax fibers with different moisture content were used as reinforcement in polypropylene matrix with maleic anhydride grafted polypropylene coupling agent. Mechanical properties – three point bending and Izod impact strength – were investigated as a function of moisture content of the fiber, and amount of applied coupling agent.

By decreasing the moisture content of the fiber all the investigated properties can be improved. Using PPgMA as coupling agent the three point bending characteristics (flexural strength, flexural modulus, and ultimate bending stress) were better, while the impact strength decreased.

Keywords: Flax fibre reinforced polypropylene; moisture content; maleic anhydride grafted polypropylene; mechanical properties

INTRODUCTION

Cellulosic fibers – as renewable raw materials – have great importance in sustainable development. Their production is economical, with low requirements on equipment, and they can easily be recycled.

Compounding of cellulosic fibers with thermoplastics reduces the mass of the composite due to their low density, increases the stiffness of the composites but tends to reduce strength and toughness [1, 2].

*Corresponding author.

The poor strength properties results from an *insufficient adhesion* between the hydrophobic polymer and hydrophilic fiber. The efficiency of a fiber reinforced composite depends on the fiber-matrix interface and the ability to transfer stress from the matrix to the fiber. To improve the properties of the composites the reinforcing natural fibers can be modified by *physical treatments* (*e.g.*, thermotreatment [3], cold plasma treatment [4], corona treatment [5], *etc.*) or *chemical treatments* (using coupling agents like organosilanes [6, 7], acrylate reagents [8], triazine derivatives [9], isocyanates [6, 9, 10], vinyl-sulphone reagents [9], titanates [11], maleic anhydride grafted polypropylene (PPgMA) [12–17], *etc.*). A few surveys are given in scientific literature on the possibilities of physical and chemical modification [18–20].

PPgMA is one of the most widely studied coupling agent. Many papers deal with its preparation [21, 22] and its effect on mechanical properties [12–17]. According to Karnani *et al.* [15] the increase of PPgMA content increases both the tensile and flexural strength, while Raj and Kokta [23] found, that the tensile strength and modulus changed *via* maxima.

Maleic anhydride forms covalent bond with the hydroxyl groups of cellulose fiber (Fig. 1a) and assume compatibilization and adhesion with the matrix by wettability, cocrystallization, and/or entanglements [24, 25]. The esterification between succinic anhydride and cotton

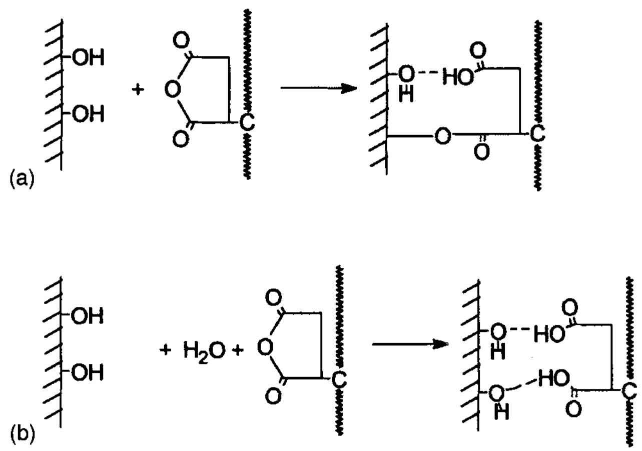

FIGURE 1 Reaction of maleic anhydride grafted polypropylene with cellulose (a) and with water (b).

cellulose occurred in the absence of catalyst at room temperature while the esterification of the second acid group requires higher temperature [26, 27]. Water on the fiber surface acts like separating agent on the fiber matrix interface [28], since PPgMA also can react with water (Fig. 1b) sorbed by the strongly hydrophobic fiber.

In this work experiments were carried out to investigate the effect of moisture content of reinforcing fiber on the dinamic mechanical properties of flax fiber reinforced polypropylene composites.

EXPERIMENTS

Materials

Polypropylene (H681F) homopolymer matrix was supplied by Tiszai Vegyi Kombinát Co., Ltd. (Hungary) (Melt Flow Index 1.7 g/10 min, density 0.9 g/cm^3).

Maleic anhydride grafted polypropylene (Polybond 3002) from Uniroyal Chemical Ltd. was used as coupling agent (Melt Flow Index 7 g/10 min). Sclavons *et al.* [29] found 30 and 11 meq/g maleic anhydride in raw and in washed product, respectively, by titration.

Combed flax fibers with diameter range of 30–300 μm (from Hungarolen, Komárom, Hungary) were used as reinforcement.

Preparation of Composites

The fibers were stored above different saturated saline solution (Tab. I) at room temperature (25 ± 3°C) for at least 60 hours. The PP, PPgMA (0-1-3-5-7 weight%) and the fiber (20 weight%) were compounded at maximum temperature of 198°C in a Brabender PL2100 twin screw extruder. The fiber spent 2–2.5 min in normal atmosphere

TABLE I Relative humidity above different saturated saline solution

Saline	*Relative humidity at 25°C* [%]	*Moisture content of fiber* [%]
LiCl	11.3	3.99 ± 0.14
K_2CO_3	43.2	7.86 ± 0.06
NaCl	75.3	11.50 ± 0.08
K_2SO_4	97.3	26.73 ± 0.83

between the climatized box and extruder. The extrudate was air cooled, ground and extruded again to get granulable strang. All the composites were injection moulded in a 5 ton ENGEL ES 200H/80V/50HL-2K two component injection moulder. The test specimens were allowed to post-crystallize at least for ten days after injection moulding and were stored in a dessicator above phosphorous pentoxyde for 3 days before mechanical testing.

Three-point Bending

The specimens were tested by ZWICK 1464 instrument at 23 ± 2°C. The size of testspecimens, the applied support span, and the rate of crosshead motion were $80 \times 10 \times 4$ mm, 60 mm, and 5 mm/min, respectively, according to MSZ KGST 892-78.

Izod Type Impact Strength

The impact strength was tested by ZWICK equipment on standard laboratory temperature, using 2.75 J hammer with 124.4° release angle (striking nose speed 3.46 m/s at the moment of impact). The size of test specimen was $80 \times 10 \times 4$ mm with 2 mm V notch (45°) according to MSZ IZO 180.

RESULTS AND DISCUSSION

Three-point Bending Test

The *ultimate bending stress* (at 10% of deflection) can be seen on Figure 2. The stress changed *via* maxima as a function of moisture content of fiber. The best results was measures at 7.86% moisture content (43.2% RH). The samples prepared from fibers with the highest moisture contents gave the worst results, as it was expected.

Using PPgMA coupling agent the ultimate bending stress was improved. Application of 7% additive resulted about the same improvement in case of all investigated moisture content.

Similar effect can be observed in case of *flexural strength* (at maximal force) (Fig. 3). The best results – in case of both properties mentioned above – were obtained at maximal (7%) PPgMA content.

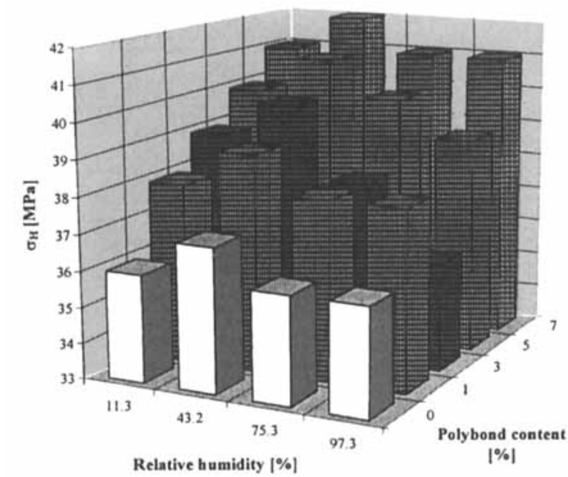

FIGURE 2 Ultimate bending stress as a function of relative humidity (moisture content of fiber) and amount Polybond 3002.

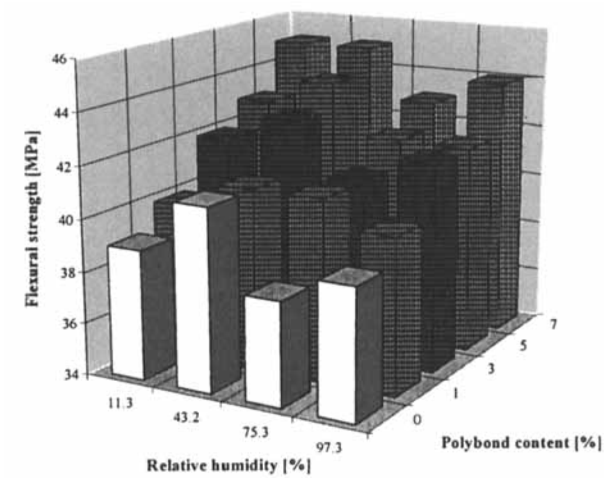

FIGURE 3 Flexural strength (at maximal force) as a function of relative humidity (moisture content of fiber) and amount Polybond 3002.

The highest *flexural modulus* values were found at the lowest moisture contents (Fig. 4). The flexural modulus changed *via* maxima as a function of PPgMA content. The maximum values were found at 1% PPgMA content.

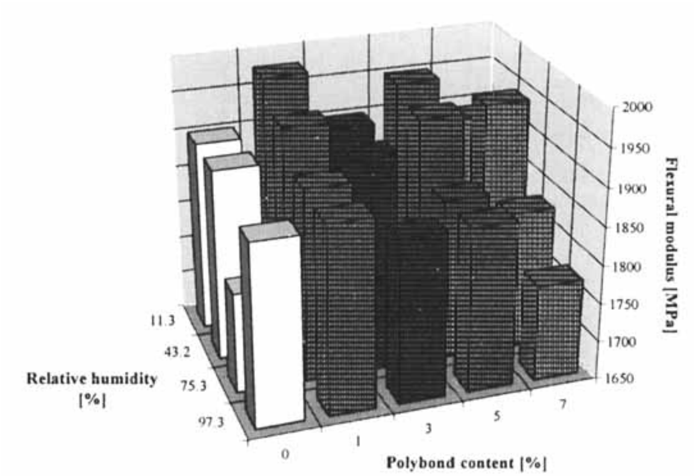

FIGURE 4 Flexural modulus as a function of relative humidity (moisture content of fiber) and amount Polybond 3002.

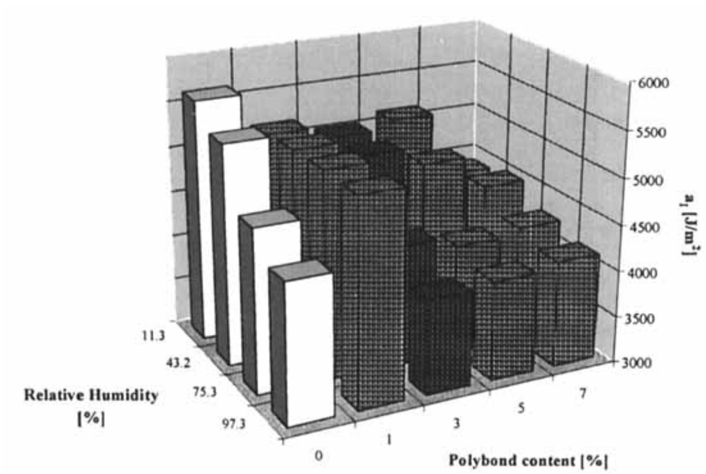

FIGURE 5 Impact strength as a function of relative humidity (moisture content of fiber) and amount Polybond 3002.

Izod Type Impact Test

Figure 5 shows *Izod type impact strength* as a function of relative humidity and Polybond content. The impact strength decreased by increasing moisture content. The highest drop in impact strength can be observed between samples prepared from flax fibres with 7.68% and 11.50% moisture content. The exceptions are the samples with 1% polybond content, in this case the moisture content had no effect.

Using 1% PPgMA seems to be optimal from the point of view of impact strength too. This effect is more definite in case of higher relative humidity.

Increasing the polybond content from 3% to 7% caused no changes above 75% relative humidity.

CONCLUSION

Flax fibers with different moisture content were used as reinforcement in polypropylene matrix with maleic anhydride grafted polypropylene as coupling agent.

Application of 1% Polybond seems to be optimal from the point of view of flexural modulus and Izod impact strength.

As it was expected the mechanical properties worsened with increasing moisture content. Presumably due to the chemical reaction between water and PPgMA. Further investigation is needed to prove this statement.

On the other hand the applied coupling agent in humid environment is very corrosive against the vital part of processing machines (screws, valves, *etc.*) as well as tooling.

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