MECHANICAL AND PHYSICAL PROPERTIES OF SOME FLAX FIBER REINFORCED EPDM COMPOSITES

Anton Airinei[1], Maria Daniela Stelescu[2], Nicusor Fifere[1], Cristian Varganici[1], Elena Manaila[3]

[1]Petru Poni Institute of Macromolecular Chemistry, Aleea Grigore Ghica Voda 41A, 700487 Iasi, Romania
[2]National Research and Development Institute for Textile and Leather, Leather and Footwear Research Institute, 93 Ion Minulescu Street, 031215 Bucharest, Romania
[3]National Institute for Lasers, Plasma and Radiation Physics, 409 Atomistilor Street, 077125 Magurele, Romania

Keywords: EPDM, flax fibers, composites, mechanical properties, thermal stability

Abstract
Natural fiber reinforced thermoplastic composites are now potential materials for various engineering applications. Polymer composites based on EPDM and flax fibers added in different amounts have been obtained by melt blending technique. The structural characteristics of some EPDM based composites reinforced with flax fibers were evaluated by mechanical and water absorption properties, thermal stability, rubber-filler interactions. Based on the obtained results, the influence of the flax fiber content and electron beam irradiation dose on the mechanical properties and thermal behavior was very important. For compositions of 15 phr flax fiber the best performances were obtained.

1. Introduction

Fiber reinforced thermoplastic composites have been received more attention due to their various applications in automotive, electrical, construction and packaging field [1-3]. In these systems, both synthetic and natural fibers can be used as reinforcing agents. As an alternative reinforcement in polymer composites, the natural fibers have gained an important interest because of their advantages over conventional glass and carbon fibers [3]. The natural fibers have low density, low cost, good thermal and acoustic insulation characteristics, less health risks and they are nonabrasive, with reduced energy consumption. Also, the natural fibers are biodegradable, less irritating to the skin and they give environmental and technical added value and can be processed at relatively low temperatures [3-6]. The natural fibers used in thermoplastic compositions include flax, hemp, jute, kenaf, sisal, ramie and many others. In the case of natural fiber utilization, the environmental aspects play an important role since these fibers are made from renewable resources and their obtaining does not require a large amount of energy.

The major drawback of natural fibers in reinforcement of the polymer composites is the poor compatibility between the hydrophobic polymer matrix and the hydrophilic nature of the fibers. Also, the natural fibers possess low environmental and dimensional stability. The interfacial adhesion between the polymer matrix and natural fibers can be improved by the modification of fiber surface to make them more compatible with the polymer (corona or plasma treatments) or by chemical methods utilizing coupling agents, such as silanes, isocyanates and/or by grafting with reactive units (maleic anhydride, acrylic acid, etc.) [6-10].

Ethylene-propylene-diene terpolymers (EPDM) exhibit remarkable properties due to their nonpolar structure and saturated backbone such as ozone and heat resistance, flexibility to low temperatures, good resistance in polar media and excellent electrical characteristics. EPDM can be processed with large

Excerpt from ISBN 978-3-00-053387-7

amounts of fillers or plasticizers [2, 11, 12]. EPDM is prepared by polymerization of ethylene and propylene with low amounts of a nonconjugated diene, which gives higher resistance to aging, weathering and chemical resistance [13].

The use of natural fibers as reinforcing agents for some polymeric materials can be considered as, an interesting aspect of the valorization of the natural resources in order to develop advanced functional materials. Among the natural fibers, flax fibers play an important role due to their good mechanical properties and ease access to chemical and mechanical modifications. By irradiation with accelerated electron beams at different doses it is possible to enhance the water resistance and stability of the EPDM/natural fibers composites.

In this work the structural characteristics of some EPDM based composites reinforced with flax fibers were discussed in terms of mechanical properties, thermal stability, rubber-reinforcing agent interactions. The influence of flax fiber loading and electron beam irradiation dose on the composite properties was analyzed.

2. Experimental

EPDM rubber composites were obtained by melt blending procedure on laboratory electrically heated roller/mill equipped with a cooling system. The working parameters were chosen as follows: temperature 60-80^{0}C, friction 1:1.1, blending time 8-15 min. The mixing sequence started firstly with the introduction of EPDM rubber (1-2 min), then antioxidant (Irganox 1010) and polyethylene glycol (PEG 4000) were incorporated (2 min). When an homogeneous mixture was formed 5, 10, 15 and 20 phr ground flax fibers were added. Test specimens were mould at 160 ^{o}C and a pressure of 150 MPa using an electrical press. The plates were cooled at room temperature under pressure.

The mechanical properties were performed with a Schopper strength tester using dumb-bell shaped specimens according to ISO 37/2012. The hardness of composites was measured using a hardner tester according to ISO 7691-1/2011. The elasticity was determined with a Schob test machine according to ISO 4662/2009. The thermogravimetric analysis was carried out on a STA 449F1 Jupiter instrument (Netzsch). The heating rate was 10 K/min and the nitrogen flow rate was 50 ml/min. The morphological characteristics of the EPDM/flax fiber composites were examined with a Quanta 200 scanning electron microscope operating at 20 kV in low vacuum mode using a secondary electron detector LFD. Irradiation of composites was performed by an electron beam accelerator ILU-6M in the dose range of 75-600 kGy on samples packed in a polyethylene film.

The raw materials EPDM rubber Nordel 4760 (Dow Chemical Co.), polyethylene glycol (PEG 4000 – Advance Petrochemical Ltd), Irganox 1010 (BASF Schwartz) and ground flax wastes were used as received.

3. Results and discussion

The mechanical properties of EPDM/flax composites were discussed taking into account the irradiation dose and the content of flax fiber. The results achieved based on tensile tests, hardness elasticity and elongation at break are listed in Table 1. The tensile strength increases both with the increase of fiber loading and absorbed dose. However, at higher doses (600 kGy), a significant decrease of tensile strength was observed. The hardness increases also with the increase of flax level and irradiation dose, excepting the sample having 20 phr flax fiber and dose of 600 kGy, where a slightly decrease was found out. The

Excerpt from ISBN 978-3-00-053387-7

increase of hardness of composites suggests that the incorporation of short flax fibers in polymer matrix leads to the reinforcement of rubber. The variation of hardness was marked for sample with higher content of fiber (PIn20). The elasticity decreases from 60% (PIn5) to 48% for the sample with 20 phr flax content.

Table 1. Physical-mechanical properties of some EPDM/flax fiber composites.

Sample	Dose (kGy)	Tensile strength (N/mm^2)	Hardness (Shore A)	Elasticity (%)	Elongation at break (%)
PIn 5	75	2.4	65	60	520
	150	2.3	69	58	340
	300	2.2	67	58	253
	600	2.4	66	56	493
PIn10	75	2.4	70	54	490
	150	2.2	73	52	480
	300	2.3	75	52	467
	600	1.8	75	52	180
PIn 20	75	2.9	78	50	220
	150	2.8	78	50	80
	300	3.0	82	48	87
	600	2.1	77	46	100

The elongation at break decreases with increasing fiber loading because higher content of flax fiber determine a reduced mobility of the polymer chains and the stiffness of the material [14].

The weight loss curves (TG) of EPDM/flax fibers composites are shown in Fig. 1. As can be seen from Fig. 1, the thermal decomposition of EPDM/flax fibers composites takes place by two steps. In the first degradation step a weight loss of about 10% was observed. The second step corresponds to the degradation of polymer chains with the maximum of decomposition temperature of 473 °C. The thermal stability of the composites decreases for higher fiber levels and irradiation doses (Fig. 1).

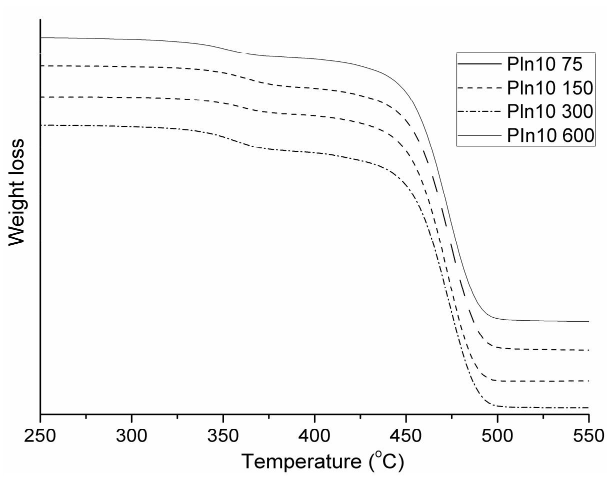
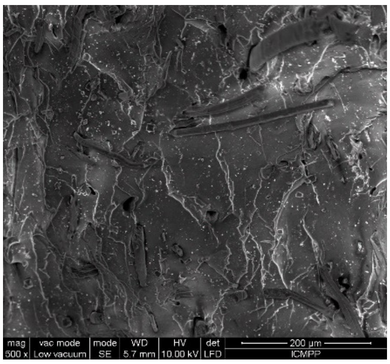


Figure 1. Thermogravimetrical curves of EPDM/flax fiber composites.

Excerpt from ISBN 978-3-00-053387-7

The amount of absorbed water in EPDM/flax fibers composites depends on the fiber content and accelerated electron dose. It was found out that the highest fiber loading (20 phr) provides the highest water uptake regardless the irradiation dose. Therefore, the electron beam treatment contributes to the improvement of the adhesion with polymer matrix arising thus a better interface and superior composite characteristics.

Morphologically, a significant difference between the initial and irradiated samples was observed (Fig. 2). In the electron beam irradiated fiber composites the crosslinking process between adjacent polymer chains occurred.


(1)

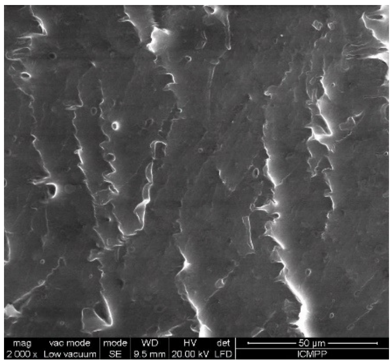

(2)

Figure 2. SEM micrographs of EPDM composites reinforced with flax fiber:
1-PIn10; 2-PIn10, irradiation dose 150 kGy.

4. Conclusions

The mechanical and thermal characteristics, water uptake of some EPDM composites reinforced with flax fibers have been evaluated. The mechanical properties are improved in terms of tensile strength, elasticity and hardness. The irradiation with accelerating electron beams in suitable doses favors the interfacial adhesion between polymer matrix and flax fibers leading to better mechanical properties.

References

[1] A. Kalapakdee and T. Amornsakchi. Mechanical properties of preferentially aligned short pineapple leaf fiber reinforced thermoplastic elastomer: Effect of fiber content and matrix orientation. *Polymer Testing*, 37:36-44, 2014.

[2] N.N. Rozik, S.L. Messieh, A.A. Yaseen and A.A. Shouk. Dielectric and mechanical properties of natural nanofibers – reinforced ethylene propylene diene rubber: Carrot foliage and corn gluten. *Polymer Engineering and Science*, 53:874-881, 2013.

[3] E. Danaila, M.D. Stelescu, L. Surdu, G. Craciun, C.L. Dinca and D. Gurau. Polymeric composites based on flax wastes and natural rubber. *Industria Textila*, 65:53-60, 2014.

[4] G.S. Sarkel and A. Choudhury. Dynamic mechanical and thermal properties of PE-EPDM based jute fiber composites. *Journal of Applied Polymer Science*, 108:3442-3453, 2008.

[5] K. Ramanaiah, A.V.R. Prasad and K.H.C. Reddy. Mechanical and thermophysical properties of fish tail palm tree natural fiber-reinforced polyester composites. *International Journal of Polymer Analysis and Characterization*, 18:126-136, 2013.

Excerpt from ISBN 978-3-00-053387-7

[6] F. Puch and C. Hopmann. Experimental investigation of the influence of the compounding process and the composite composition of the mechanical properties of a short flax fiber-reinforced polypropylene composite. *Polymer Composites*, 36:2282-2290, 2015.

[7] O. Faruk, A.K. Bledzki, H.P. Fink and M. Sain. Biocomposites reinforced with natural fibers: 2000-2010. *Progress in Polymer Science*, 37:1552-1596, 2012.

[8] M. Soleimani, L. Tabil, S. Panigrahi and A. Opoku, The effect of fiber treatment and compatibility on mechanical and physical properties of flax fiber-polypropylene composites, *Journal of Polymers and the Environment*, 16:74-83, 2008.

[9] K. Salasinska, M. Polka, M. Gloc and J. Ryszkowska. Natural fiber composites: the effect of the kind and content of filler on the dimensional and fire stability of polyolefin-based composites, *Polimery*, 61:255-265, 2016.

[10] L. Sobczak, O. Bruggemann and R.F. Putz. Polyolefin composites with natural fibers and wood-modification of the fiber/filler-matrix interaction. *Journal of Applied polymer Science*, 127: Article 36935, 2013.

[11] M. Homocianu, A. Airinei, D.M. Stelescu, D. Timpu, A. Ioanid. Morphological structure and surface properties of maleated ethylene propylene diene monomer/organoclay nanocomposites. *Polymer Composites*, 33:379-387, 2012.

[12] R. D. Allen. Fundamentals of compounding EPDM for cost/performance. *Journal of Elastomers and Plastics*, 15:19-32, 1983.

[13] F. Delor-Jestin, J. Lacoste, N. Barrois-Oudin, C. Cardinet and J. Lemaire. Photothermal and natural ageing of ethylene-propylene-diene monomer (EPDM) rubber used in automotive applications. *Polymer Degradation and Stability*, 67:469-477, 2000.

[14] M. Cristea, A. Airinei, D. Ionita and D.M. Stelescu. Relaxation behavior of flax-reinforced ethylene-propylene-diene rubber. *High Performance Polymers*, 27:676-682, 2015.

Excerpt from ISBN 978-3-00-053387-7