U.P.B. Sci. Bull., Series 69, Vol. 69, No. 3, 2007 ISSN 1454-2331

STUDY ON THE IMPROVEMENT OF PHYSICAL-MECHANICAL CHARACTERISTICS IN THE EPDM-HDPE BLENDS

Maria Daniela STELESCU [1], Elena MANAILA[2], Ileana-Gabriela NICULESCU-ARON[3]

Lucrarea prezintă un studiu privind îmbunătățirea caracteristicilor fizico-mecanice ale amestecurilor pe bază de elastomer etilenă propilenă terpolimer (EPDM) şi polietilenă de înaltă densitate (PEID), prin introducerea unor agenți de compatibilizare sau iradiere cu electroni accelerați. Caracteristicile amestecurilor polimerice obținute sunt aditive şi depind de cantitatea de EPDM şi PEID introduse în amestec. Utilizarea agenților de compatibilizare nu a condus la o îmbunatățire semnificativă a caracteristicilor amestecurilor. Prin iradierea amestecurilor cu electroni accelerați la 5, 10, 15 si 20 Mrad, la creşterea dozei de iradiere, creşte modulul de elasticitate 100% şi alungirea la rupere, iar rezistența la sfâşiere prezintă un maxim urmat de o usoară scădere.

The work presents a study on the improvement of physical-mechanical characteristics of blends containing EPDM and HDPE by introducing some compatibilizing agents or irradiating them with accelerated electrons. The characteristics of the obtained polymer blends are additive and dependent on the amounts of EPDM and HDPE involved. The use of compatibilizing agents has not led to a significant improvement of the charasteristics of the blends. Irradiating the blends with accelerated electrons at 5, 10, 15 and 20 Mrad, when the irradiation dose was increased there were an increase in the elastic modulus of 100 % and elongation at break and tear strength showed a maximum followed by a light decrease.

Keywork: EPDM-HDPE blends, compatibilizing agent , accelerated electrons.

1. Introduction

Blends containing EPDM and polyolefins known as olefin thermoplastic elastomers (TPO) have a large range of applications, a significant one being in car manufacture [1-6]. Because of the importance of these types of thermoplastic

[1] Researcher, Eng., National Research and Development Institute for Textile and Leather – Leather and Footwear Research Institute, Bucharest, Romania
[2] Researcher, PhD, National Research and Development Institute for Laser Plasma and Radiation Physics, Romania
[3] Senior lecturer, PhD, Statistics and Economic Forecast Department, Academy of Economic Studies, Bucharest, RomaniaThe Economic Studies Academy

elastomers, many studies on the improvement of physical-mechanical and processing characteristics as well as on the enlargement of the application field were conducted.

The studies performed [7-11] have revealed that improved compatibility between the two phases (elastomer and polyolefin phases) enables the blend characteristics to be increased. The methods for improving the compatibility between the two phases are of two types, as follows: (1) unreactive compatibilization by involving a co-polymer or grafted homopolymer being miscible with the two phase and (2) reactive compatibilization by creating chemical linkages between the two co(polymers) at the interface [12-14].

In this work the improvement in the physical-mechanical characteristics of the blends containing EPDM and PEID was studied by two methods: (1) adding some co-polymers or grafted homopolymers and (2) irradiating with accelerated electrons. In the first method a non-reactive compatibility between two phases is created, and in the second method a new reactive compatibility. Process chemistry is based on building the super-radicals made up of the (co)polymer chains being recombined [15-17]

2. Experimentals

The following raw materials were used: (1) *EPDM rubber* Keltan 8340 A (54,7% ethylene, 5,41% ENB); (2) *HDPE* Hostalen GC 7260 (0,962 g/cm^3); (3) c*ompatibilization agents:* chlorinated polyethylene CPE TX10 (35% chlorine), polyethylene chemically modified by grafting with maleic anhydride Polybond 3009 (1% maleic anhydride, 0,95 g/cm^3), polypropylene chemically modified by grafting with maleic anhydride Polybond 3002 (0,2% maleic anhydride, 0,90 g/cm^3) and maleinized ethylene propylene terpolymer rubber Royaltuf 498 (1% maleic anhydride,0,90 g/cm^3).

The blends were prepared on a Brabender plastificator at a temperature of 160 – 170^{o}C and 60 rpm, 7 min blending time. For homogenization was used an electrically heated laboratory roll. The plates 150 mm x 150 mm x 2 mm intended to be used for the physical mechanical tests were made on a laboratory hydraulic press at a temperature of 160^{o}C, pressing time 6 min.

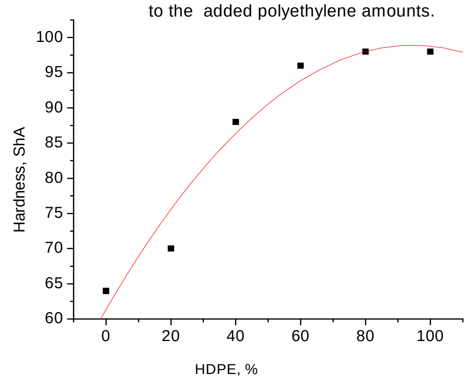
EPDM/HDPE blends containing 100 %, 80 %, 60 %, 40 %, 20% and 0% (by weight) elastomer in the blend were prepared. For the blend containing 60% EPDM (by weight) two methods of compatibilizing have been used: (1) the addition of compatibilizing agent 5% (percent by polymer weight) and (2) blend was subjected to the accelerated electron irradiation, using a particle accelerator ILU–6M with a medium energy of 2 MeV and maximum power of the electron beam of 20 kW. The samples were protected from the oxygene in irradiation chamber by wrapping them in polyethilene foil

Physico-mechanical tests were performed according to the following standards: ISO 7619:2001 (hardness), ISO 37:1997 (modulus, tensile strength, elongation at break) and ISO 34-1:2000 (tear strength).

3. Results and Discussions

Some EPDM/HDPE blends containing 100, 80, 60, 40, 20 and 0% (by weight) elastomer were prepared. Figs. 1-4 reveal that as the HDPE level is increased, the hardness, tearing strenght and the tensile strength are higher, elongation at break diminish. The resulted physical-mechanical characteristics have revealed that the blend properties are *additive* according to Paul R. [18] (with no minimum or maximum) and dependent on every constituent; thus, with a higher (80%) level of elastomer, the elastomer characteristics are predominant, and with 80% of HDPE, the plastic characteristics are predominant.

Figure 1 - Changes in the hardness according to the added polyethylene amounts.

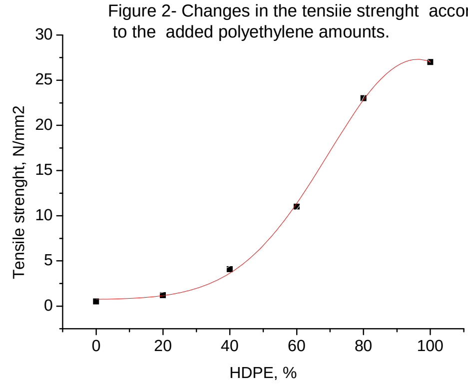


$$Y = 61.35714 + 0.79393\,X - 0.0042\,X^2$$

Parameter	Value	Error
A	61.35714	3.53019
B1	0.79393	0.16603
B2	-0.0042	0.00159

R-Square(COD)	SD	N	P
0.95977	3.89505	6	0.00807

Figure 2- Changes in the tensiie strenght according to the added polyethylene amounts.

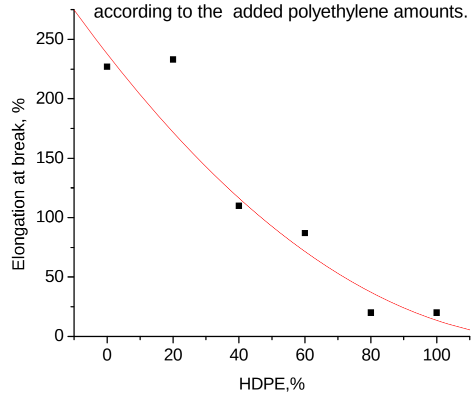


Data: Data2_B
Model: Gauss

Chi^2 = 0.20908
R^2 = 0.99936

y0	0.707	±0.35999
xc	96.29641	±1.41371
w	53.684	±2.75573
A	1789.30719	±106.48203

Figure 3 - Changes in the elongation at break according to the added polyethylene amounts.

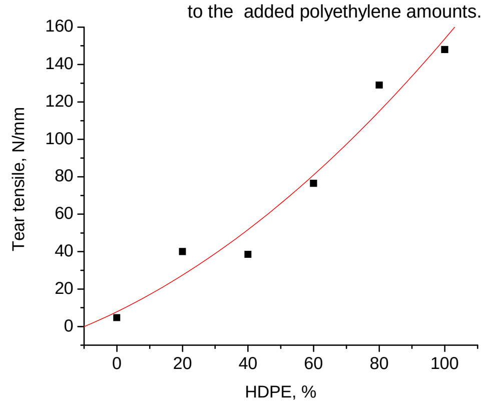


$$Y = 237.86121 - 3.56392\,X + 0.0132\,X^2$$

Parameter	Value	Error
A	237.86121	13.09098
B1	-3.56392	0.68486
B2	0.0132	0.00695

R-Square(COD)	SD	N	P
0.98021	17.29142	6	0.00278

Figure 4 - Changes in the tear tensile according to the added polyethylene amounts.



$$Y = 7.825 + 0.85884\,X + 0.006\,X^2$$

Parameter	Value	Error
A	7.825	12.76543
B1	0.85884	0.60038
B2	0.006	0.00576

R-Square(COD)	SD	N	P
0.96213	14.0848	6	0.00737

1. An amount of 5% compatibilizing agent such as maleinized polyethylene (PEm), chlorinated polyethylene (CPE), maleinized polypropylene (PPm) and maleinized EPDM (EPDMm) was added to the blend of 60% EPDM and 40% HDPE.

The results of physico-mechanical tests (Figs. 5-7) have revealed the following features:

- By adding *maleinized polyethylene* to the blend, the best values for tensile strength and tear strength and a slight decrease in elongation at break were revealed.
- Addition of *chlorinated polyethylene* has led to decreased elongation at break, tensile strength and a slight increased in tear strength.

- Compatibilization agent of *maleinized ethylene propylene* (Royaltuf 498) has led to a slight decrease in tear tensile and an increase in tensile strength and the best value for elongation at break.
- *Maleinized polypropylene* has led to increased elongation at break and tear strength and a slight decrease in tensile strength.

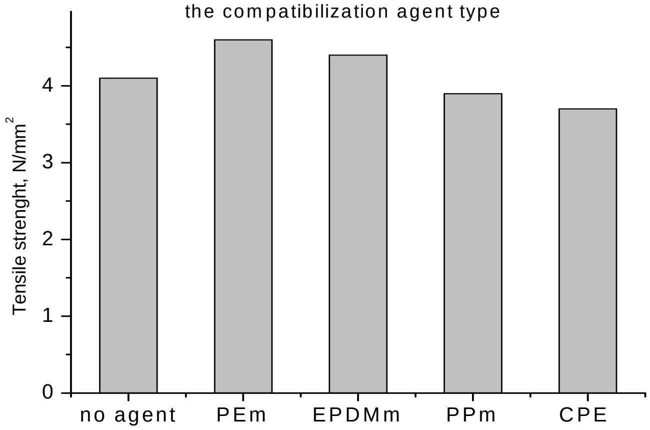


Figure 5 Tensile strenght versus the compatibilization agent type

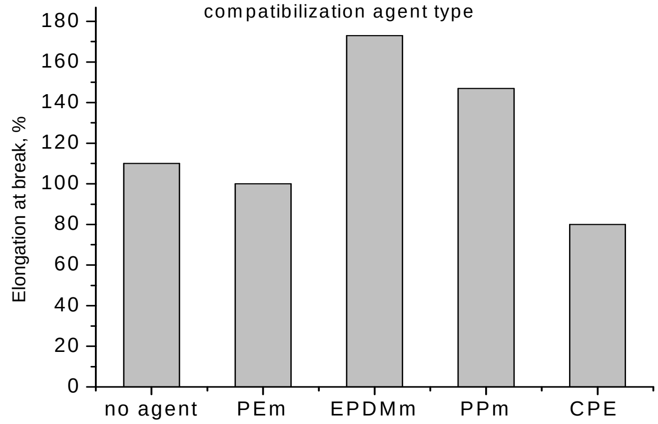


Figure 6 Elongation at break versus compatibilization agent type

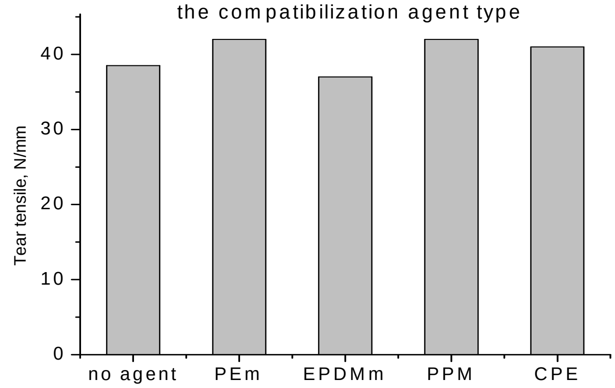


Figure 7 Tear tensile versus the compatibilization agent type

Concluding, using compatibilisation agents has not led to significant modification of the characteristics of the blends; the most efficient agent was PEm.

2. The blend of 60% EPDM and 40% HDPE was irradiated with 5, 10, 15 and 20 Mrad. The obtained results (Figs. 8–10) reveal an abrupt increase in tear strength and elongation at break following the irradiation of the EPDM/HDPE blend with 5 Mrad, this showing that the elastomers have turned from plastic to elastic condition. In this case crosslinking bridges both between the elastomer chains and between the elastomer and HDPE are made, the last ones leading to the formation of polymers at the interface which increase the compatibility between the two phases. When the irradiation dose was increased up to 10, 15 and 20 Mrad, the crosslinking extent in the blend and elastic modulus increased, elongation at break decreased and tear strength showed a maximum followed by a slight decrease.

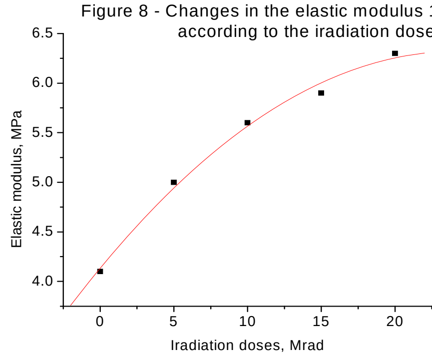


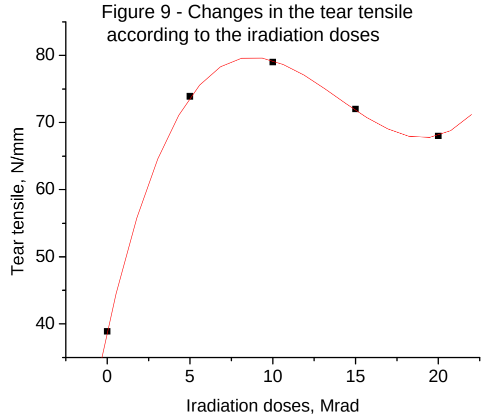
Figure 8 - Changes in the elastic modulus 100% according to the iradiation doses

$Y = 4.13429 + 0.18029\ X - 0.00371\ X^2$

Parameter	Value	Error
A	4.13429	0.08999
B1	0.18029	0.02132
B2	-0.00371	0.00102

R-Square(COD)	SD	N	P
0.9938	0.09562	5	0.0062

Figure 9 - Changes in the tear tensile according to the iradiation doses

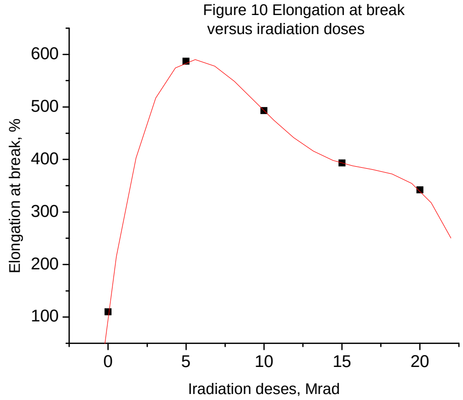


$Y = 38.93857 + 10.99024\,X - 0.91543\,X^2 + 0.02193\,X^3$

Parameter	Value	Error
A	38.93857	0.3204
B1	10.99024	0.16301
B2	-0.91543	0.0207
B3	0.02193	6.80336E-4

R-Square(COD)	SD	N	P
0.9999	0.32271	5	0.01296

Figure 10 Elongation at break versus iradiation doses



$Y = 110 + 215.66667\,X - 32.07\,X^2 + 1.77333\,X^3 - 0.034\,X^4$

Parameter	Value	Error
A	117.28571	60.51969
B1	154.95238	30.79118
B2	-15.43429	3.90992
B3	0.41333	0.12851

R-Square(COD)	SD	N	P
0.97142	60.95666	5	0.21422

4. Conclusions

From the study on the improvement of the physical - mechanical characteristics in the blends containing EPDM and HDPE have resulted the following:

1. The characteristics of the EPDM/HDPE blends are additive and dependent on the amounts of EPDM and HDPE added;
2. Adding compatibilisation agents has not led to significant improvement of the physical – mechanical of the EPDM/HDPE blends. Maleinized polyethylene is a good compatibilizing agent for the EPDM/HDPE blends;
3. By irradiating the blend of 60% EPDM and 40% HDPE with 5, 10, 15 and 20 Mrad, the elastic modulus has shown an increase, elongation at break and tear strength have shown a maximum after a slight decrease when the

irradiation dose was increased. This method leads to an improvement of the characteristics, the optimal dose being 5 Mrad.

4. This blends have shown elastomer specific characteristics and could be processed by methods used currently in processing thermoplastic materials (injection, jet moulding etc.).

REFERENCES

[1] *E. N. Kresge,* Thermoplastic Elastomers, G. Holden N. R. Legge, R. P. Quirk si H. E. Schroder eds., Hanser / Gardner (1996), Munich. Vienna, New York.
[2] *L. E. Mirci*, Elastomeri termoplastici, Editura Art Press si Editura Augusta, Timisoara (2005).
[3] *Nakamura Eiji, Masuda Haruhisa,* PCT Int. Appl. WO 14.nov.2002
[4] *T Augustin,* Rubber Chemistry and Technology, **Vol. 63**, nr. 1 (90)
[5] *Tasaka Michihisa, Ogawa Tomozo*, Jpn. Kodai Tokkyo Koho JRP (2002)
[6] *Jin Xuechih, Takahaschi Hideki,* Jpn. Kokai Tokkyo Koho, JP 2002
[7] *Josephine George,* Rubber Chemistry and Technology, **Vol**. **78**, p. 286-311 (2005)
[8] *A.Y.Coran, R.Patel,* Rubber Chemistry**, Vol. 54**, p. 91.nr. **1** (1998)
[9] *O.Chung, A.Y.Coran,* Rubber Chemistry and Technology, **Vol. 70,** nr. **5**. (1997).
[10] *D. Mangaraj,* Rubber Chemistry and Technology, **Vol. 78,** no.3 , (2005)
[11] *R.Asaletha, Sabu Thomans,* Rubber Chemistry and Technology, *Vol. 68*, no. 4, (1995).
[12] *T.Marinovic, Z.Susteric, I.Dimitrievski,* Kautschuk Gummi Kunststoffe, no. 3, (1998).
[13] *T.A. Guguva, A.A. Kanauzova,* International Polymer Science and Technology, **Vol. 25**, nr. 10, (1998).
[14] *M. D.Ellul,* Rubber Chemistry and Technology, **Vol. 68,** nr. 1, (1995).
[15] *S.K. Datta, N.K. Pradhan, T.K. Chaki*, Kautschuk Gummi Kunststoffe, no. 7-8/97, p.554-559.
[16] *Han Do Hung, Shin Seung Ho,* Radiation Physics and Chemistry, **Vol. 69,** no. 02/2004, p.239-244.
[17] *Rajesh Chowdhury, M S Banerji,* Journal of Applied Polymer Science, 2/97, p. 968 – 975.
[18] *D.R.Paul,* Polymer Blends, J.Macromol. Sci. – Rev. Macromol. Chem., **C18 (1),** p. 109 – 168 (1980).