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Contact angle and surface free energy of electron-beam irradiated polymer composites

Summary — The effects of the electron radiation dose and of compatibilizers on the contact angle and surface free energy (*SFE*) of the composites made of low-density polyethylene (PE-LD), high-density polyethylene (PE-HD), polypropylene (PP), polystyrene (PS), and polyethylene terephthalate (PET) were studied. Use of the high-energy electron radiation with doses up to 300 kGy and of compatibilizers was done to reach better mechanical and adhesion properties of the composites studied and, at the same time, to investigate the possibility of applying of this technique in the processes of polymeric materials recycling. The compatibilizers were the styrene-ethylene/butylene-styrene elastomer grafted with maleic anhydride (SEBS-*g*-MA), added at the amounts of 5, 10 or 15 wt. %, and trimethylol propane trimethylacrylate (TMPTA), added at the amounts of 1, 2 or 3 wt. %. The effects, discussed in the present article, are: enhancement of wettability and increase in *SFE* of the composites studied. It was found that the contact angle steadily decreased and *SFE* of the composites increased with the rising dose of the electron radiation and that TMPTA intensified these tendencies.
Key words: composites, surface modification, contact angle, surface free energy, recycling.

KĄT ZWILŻANIA I SWOBODNA ENERGIA POWIERZCHNIOWA KOMPOZYTÓW PODDANYCH DZIAŁANIU PROMIENIOWANIA ELEKTRONÓW O DUŻEJ ENERGII
Streszczenie — Zbadano wpływ promieniowania elektronowego i kompatybilizatorów na kąt zwilżania i swobodną energię powierzchniową (*SFE*) kompozytów składających się z polietylenu małej gęstości (PE-LD), polietylenu dużej gęstości (PE-LD), polipropylenu (PP), polistyrenu (PS) i poli(tereftalanu etylenu) (PET). Poddanie tych kompozytów działaniu promieniowania elektronów o dużej energii, w zakresie dawek do 300 kGy oraz wprowadzenie do nich kompatybilizatorów poprawiających ich właściwości mechaniczne miało na celu sprawdzenie możliwości stosowania tej technologii w procesach recyklingu odpadów tworzywowych. Jako kompatybilizatory zastosowano elastomer styren-(etylen/butylen)-styren, szczepiony bezwodnikiem kwasu maleinowego (SEBS-*g*-MA), dodawany w ilościach 5, 10 lub 15 % mas. oraz triakrylan trimetylolopropanu (TMPTA) dodawany w ilościach 1, 2 lub 3 % mas. Zmiany zachodzące w warstwie wierzchniej tak modyfikowanych kompozytów powodowały zmniejszenie wartości kąta zwilżania i wzrost wartości *SFE*. Efekty te rosły wraz ze wzrostem dawki promieniowania i były dodatkowo wzmacniane po dodaniu TMPTA jako kompatybilizatora.
Słowa kluczowe: kompozyty, modyfikacja warstwy wierzchniej, kąt zwilżania, swobodna energia powierzchniowa, recykling.

Wettability and proper surface free energy (*SFE*) of polymers are necessary conditions for good adhesion of these materials. Enhanced adhesion is especially important for industrial processes of gluing, printing, and laminating of polymers obtained in the recycling of polymeric materials. *SFE* is also very important parameter governing many industrial processes, especially technologies of polymers and polymeric materials [1—4]. It is more and more significant in economy, mainly because of an increasing contribution of recycled plastics in the production of new articles. Intensive research and engineering works in the area of recycling plastics were already initiated over thirty years ago. In spite of a significant progress in this field and development of numerous recycling techniques, more effective and versatile ways of plastics waste management, including mostly reuse of them as raw materials for industrial production, are still in demand. One of potential directions of expansion of this field is connected with a wider application of radiation techniques [5, 6].

Thermodynamic immiscibility of most polymers is a serious barrier in their processing. It causes too low adhesion between individual molecules of the materials

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formed from the blends of polymers. In many cases, too low mechanical strength of these materials is an unfavourable result. These effects occur during formation of composites from original polymers as well as from plastic wastes. In order to enhance the material strength, some chemical substances called compatibilizers are added to the polymer blends. They increase the interfacial adhesion of individual components of the complex materials. Irradiation of the composites (with or without use of the compatibilizers) by means of ionising radiation, mainly high-energy electron radiation generated in accelerators, is another way to increase the material strength [7, 8]. During the irradiation in an air, the oxidation of surface layer of the polymeric material occurs, which causes increase in wettability and *SFE* and, also at the same time, the improvement in its adhesion properties [9, 10].

A possibility to apply the electron radiation in the processes of recycling of packaging plastic wastes creates the need of a better understanding of the influence of this radiation on wettability and *SFE* of the composites made of the blends of polymers commonly used to produce the packaging. Earlier [11] we found that the electron radiation significantly affected the wettability of polypropylene (PP) film and we determined the quantitative changes in *SFE*, occurring upon irradiation. Now, we present the results of investigation of the influence of the high-energy electron radiation on the wettability and *SFE* of the composites made of various polymers commonly used in the packaging industry. The studied materials differed in the composition of polymers and content of two compatibilizers. The procedure of samples preparation including an additional operation of extrusion of granulated polymers, determines the basic mechanical and heat loads, occurring during reprocessing of plastic wastes. Therefore, the results of investigations of such samples may be used to estimate the properties of similar composite materials, produced from these wastes. Full identification of the components of the samples prepared according the described procedure is undoubtedly beneficial while such identification is generally impossible for composites formed from various plastic wastes. This advantage is especially important in case of investigation of the surface layer properties of these composites because these properties depend on all the components of a material studied.

The presented results constitute a part of a broader research work in which the electron radiation and various compatibilizers are applied in order to improve the properties of the composites produced from recycled plastics.

EXPERIMENTAL

Materials

The studied composites were made of the following twice-extruded granulated polymers:

— Low-density polyethylene (PE-LD) — „Malen-E FABS 23-D0022" (Basell Orlen Polyolefins, Płock, Poland), manufactured by the high-pressure polymerization. Its density was $\rho = 0.919—0.923$ g/cm^3 (23 ^{o}C) and melt flow rate, *MFR* = 1.6—2.5 g/10 min (2.16 kg, 190 ± 0.5 ^{o}C). The polymer contained a small amount of antioxidant.

— High-density polyethylene (PE-HD) — „Hostalen ACP 5831 D" (Basell Orlen Polyolefins, Płock, Poland), manufactured by the low-pressure polymerization over Ziegler-Natta catalysts. Its density was $\rho = 0.959$ g/cm^3 (23 ^{o}C) and *MFR* = 1.2 g/10 min (5.00 kg, 190 ± 0.5 ^{o}C). The polymer contained a small amount of antioxidant.

— Isotactic polypropylene (PP) — „Malen P F 401" (Basell Orlen Polyolefins, Płock, Poland), manufactured by the suspension polymerization. Its density was $\rho = 0.905—0.910$ g/cm^3 (23 ^{o}C) and *MFR* = 2.4—3.2 g/10 min (2.16 kg, 230 ± 0.5 ^{o}C). The polymer contained small amounts of antioxidant as well as of antistatic and antiblocking additives.

— Polystyrene (PS) — „Owispol 945 E" (Dwory S.A., Oświęcim, Poland), manufactured by the continuous bulk polymerization. Its density was $\rho = 1.03$ g/cm^3 (23 ^{o}C) and *MFR* = 4—5 g/10 min (5.00 kg, 200 ± 0.5 ^{o}C). The polymer was modified with rubber.

— Amorphous poly(ethylene terephthalate) (PET) — „Elpet-A" (Boryszew SA, Elana Branch, Toruń, Poland), manufactured by polycondensation of terephthalic acid and ethylene glycol. Its density was $\rho = 1.4$ g/cm^3 (23 ^{o}C) and intrinsic viscosity, $\eta = 0.615 \pm 0.010$ dL/g. The polymer contained no more than 170 ppm of acetic aldehyde.

The following compounds were used as compatibilizers for the polymers mentioned above:

— The styrene-ethylene/butylene-styrene elastomer [SEBS, formula (I)] grafted with maleic anhydride [MA,

(I)

(II)

(III)
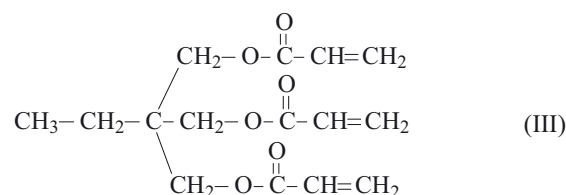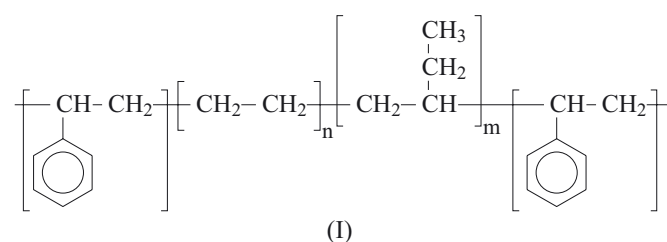
formula (II)], including 1.7 % of maleic anhydride and 30 % of styrene ("Kraton FG 1901X", Shell Chemicals, Houston, USA).

— Trimethylol propane trimethylacrylate [TMPTA, formula (III)] (Sigma-Aldrich, Germany).

Sample preparation

The samples prepared from two basic polymer blends containing one of the compatibilizers, SEBS-*g*--MA or TMPTA, were studied. Specification concerning the sample composition and irradiation dose has been included in the four-membered symbols of the samples, containing the following:

— As the first sign, the digit „1" or „2". „1" corresponds to the blend composed of 24 wt. % of PE-LD, 23 wt. % of PE-HD, 21 wt. % of PP, 15 wt. % of PS, and 17 wt. % of PET, whereas „2", to that of 33.4 wt. % of PE-LD, 33.3 % of PE-HD, and 33.3 wt. % of PP.

— As the second sign, the letter „C", „K" or „T". „C" corresponds to a blend without compatibilizer, „K" to that containing SEBS-*g*-MA, and „T" to that containing TMPTA.

— As the third sign, the digit „1", „2", or „3". A digit combined with „K" specifies the percentage (5, 10, or 15 wt. %, respectively) of SEBS-*g*-MA in a blend and combined with „T", the percentage (1, 2, or 3 wt. %, respectively) of TMPTA in a blend. For the blends without compatibilizer, zero (0) is used as the third element. The percentages of compatibilizers are specified in relation to the total mass of all polymers in a blend.

— As the fourth sign, the digit „0", „1", „2", „3", or „4". These digits determine irradiation doses equal 0, 25, 50, 100 or 300 kGy, respectively.

Apart from the above full symbols, some simplified symbols are also used to define the groups of samples, containing a specified number of polymers with or without a compatibilizer:

— 1C0; five polymers without compatibilizer;
— 2C0; three polymers without compatibilizer;
— 1K; five polymers with SEBS-*g*-MA;
— 2K; three polymers with SEBS-*g*-MA;
— 1T; five polymers with TMPTA;
— 2T; three polymers with TMPTA.

Individual polymers were weighed and carefully dried in an air at 120 $^{\circ}$C for ca. 6 h. Then, they were placed in a laboratory drum mixer and mixed for 20 min. After that, a compatibilizer was added and the blending was performed for the next 20 min. Each blend was granulated using a single-screw extruder of W25-30D type (IPTSz „Metalchem", Toruń, Poland), equipped with a segmented screw. To intensify the blending of the components, a replaceable mixing segment of the extruder screw was used. It was 8D long and mounted at the end of the screw. The obtained granulate was injected into a mold to yield a plate of the 5×5 cm size and of macroscopically smooth surface. The laboratory injec-

tion moulding press of Battenfeld Plus 35/75 type (Battenfeld GmbH, Germany), equipped with a two-cavity mold was used. The plates were used to measure the contact angle.

Irradiation of the samples was carried out at the Institute of Nuclear Chemistry and Technology in Warsaw (Poland) using an UELW-101-10 accelerator (NPO TO-RYJ, Russia). The samples with dosimetric indicators were placed in an aluminum container (42×50 cm). The container was put on a conveyor able to move with a precisely controlled speed. The speed determined the radiation dose absorbed by the samples. During a single crossing of the radiation zone, the samples absorbed a dose of 25 kGy, which corresponded to the conveyor speed of 0.58 m/min. The successive crossing of the samples through the radiation zone caused an increase in the absorbed dose by the next 25 kGy. This way, the doses of 25, 50, 100 or 300 kGy were applied to the samples. The doses were controlled by calorimetric method. During the irradiation, the maximum energy of electrons generated by the accelerator was 10 MeV, electron current intensity 480 mA, electron-beam power 10 kW, and sweeping frequency 12.5 Hz. The irradiation procedure was performed under ambient conditions of temperature and atmosphere.

Methods of testing

Measurements of the contact angle were performed using a DSA 100 instrument (Krüss GmbH, Germany), equipped with a camera for recording a drop shape at a rate of 25 pictures per second. The instrument was also provided with a DSA-3 software that enabled making computations by the axisymmetric drop shape analysis [12].

The water contact angle (further called simply the contact angle) was measured by the method of a dynamic measurement of the advancing contact angle. Redistilled water (Aqua purificata, Maggie Co., Poland) of SFE = 72.8 mJ/m^2 was used for the measurements. During a measurement, the water drop volume was continuously increased from 5 to 10 µL at the rate of 6 µL/min, which enabled retaining the velocity of movement of the drop front less than 1 mm/min. The contact angle measured this way may be considered as equal to the advancing contact angle [13].

The measurements were carried out using two opposite points of the measuring drop. The mean arithmetic value of these two measurements was assumed as the contact angle value. One series of the measurements comprised 50 such values. The first 10 were neglected because the measuring drop was not fully stabilised. The next 40 values were used to calculate the mean arithmetic value and standard deviation which was in the range of 0.8—1.5 degree. This mean arithmetic value was accepted as a final value of the contact angle and used to determine SFE.

SFE was derived using so-called equation of state [14] that enabled determination of *SFE* from the advancing contact angle (Θ) of a single measuring liquid. The following form of the equation was applied:

$$\cos \Theta = -1 + 2(\gamma_S/\gamma_L)^{0.5} \exp\{-\beta(\gamma_L - \gamma_S)^2\} \qquad (1)$$

where: γ_S — *SFE of solid material*, γ_L — *SFE of measuring liquid*, $\beta = 0.0001247 \ (mJ/m^2)^{-2}$.

The values of γ_S were computed by a numerical method because eq. (1) is of the form of an implicit function.

The *SFE* of polymeric material is generally calculated from the values of the contact angle for two or three measuring liquids of different polarity (used in the methods of Owens-Wendt or van Oss-Chaudhury--Good). Such calculations being performed with use of the equation of state are less common, in spite of the advantage of this method, in which only one measuring liquid is applied. Keeping this advantage in mind, we have used the equation of state to calculate the *SFE* values of the studied composites. However, when the results obtained by these three methods are being compared, one has to realize the differences existing between these techniques [15, 16].

RESULTS AND DISCUSSION

Contact angle measurements

The mean values of the contact angle for the individual samples are listed in Table 1. The standard deviations (σ) of the mean values are less than 1 deg for 73 % of the studied samples and no more than 2 deg for the remaining ones. The difference in the values of σ is caused by the surface layer heterogeneity, resulting from the variety of polymers and compatibilizers, which constitute this layer.

T a b l e 1. Contact angle values (in deg) for the samples irradiated with different doses

Sample group	Dose, kGy				
	0	25	50	100	300
1C0	104.0	101.3	94.2	92.1	88.8
2C0	106.6	102.4	97.3	90.8	87.3
1K1	107.4	102.6	98.1	93.0	92.1
1K2	104.6	97.9	95.9	93.9	94.5
1K3	104.7	104.0	99.6	96.5	94.1
1T1	108.5	94.1	92.2	87.6	82.5
1T2	105.3	91.5	85.8	86.5	83.4
1T3	104.5	96.0	89.1	84.4	80.2
2K1	107.1	100.6	99.6	95.9	86.7
2K2	101.6	98.7	95.5	92.0	90.0
2K3	100.3	99.4	93.4	87.2	82.7
2T1	105.7	95.2	85.1	80.9	78.1
2T2	104.2	86.8	84.0	80.8	77.4
2T3	101.2	84.0	82.2	71.1	66.9

It was found that, the contact angle decreased steadily with the rising radiation dose for the studied samples, except for 1K24 (300 kGy) and 1T23 (100 kGy). The contact angle values for them are slightly larger than those for the preceding samples 1K23 and 1T22. The differences are 0.7 and 0.6 deg, respectively, whereas halves of the confidence interval for σ = 1 deg and σ = 2 deg are *ca*. 0.3 and 0.6 deg, respectively. These results were obtained while assuming a normal distribution of the contact angle values and accepting u_a = 1.96 (the value of u_a variable was taken from the table for a standardized normal distribution), α = 0.05 (the significance level), and n = 40 (the number of contact angle measurements used to calculate the mean arithmetic value).

Small deviations of the contact angle values for 1K24 and 1T23 samples from a commonly observed tendency may be due to differences in compositions of their surface layers, caused by incomplete homogenization of the blends used to form these samples.

Changes in the contact angle values upon the electron radiation for 1C0 blend are similar to those for 2C0. Within the entire studied range of doses (0—300 kGy), the contact angle decreases by 15 and 19 deg for 1C0 and 2C0, respectively. Also, differences in the contact angle values for these two composites irradiated with the same dose are small (1.1—3.1 deg).

The SEBS-*g*-MA elastomer introduced into the 1C0 blend usually increases the contact angle of the composites, *i.e.*, it worsens their wettability. The 1K21 sample (25 kGy), the contact angle of which is by 3.4 deg smaller than that for 1C0, is the only exception. In case of 2C0, the effect of addition of SEBS-*g*-MA is less explicit because a clear decrease in the contact angle in relation to that value for 2C0 occurs only for the samples with the largest contents of the compatibilizer (2K30—2K34). Nevertheless, the basic regularity observed is that the contact angle for 1K and 2K diminishes with the rising radiation dose. For 1K the values of contact angle decrease by 10.1—15.3 deg whereas for 2K, by 11.6—20.4 deg, dependently on the content of the compatibilizer and value of the radiation dose.

The influence of TMPTA on the contact angle of the studied materials is clearly larger and more defined than that of SEBS-*g*-MA. For 1T, the contact angle values increase with the radiation dose by 21.9—26.0 deg, dependently on TMPTA content. However, when comparing the 1T samples irradiated with the same dose, the effect of TMPTA addition is unclear. This effect for 2T is more pronounced. The contact angle values decrease with the radiation dose and, for the sample groups irradiated with the same dose, the contact angle increases with TMPTA content. For the sample groups with the same content of TMPTA (2T1, 2T2, and 2T3), the contact angle values decrease by 26.6—34.2 deg with the increasing dose. Such a clear effect of TMPTA may be explained as follows: due to interaction of high-energy electrons with TMPTA molecules, a great number of free

radicals are generated that lead to formation of polar functional groups in the surface layer [17]. The possibility of generation of these radicals results from the structure of TMPTA molecule that contains six oxygen atoms, including three ones bound to carbon atoms by double bonds [formula (III)]. The functional groups formed due to reactions initiated by these radicals are hydrophilic and cause a decrease in contact angle.

Surface free energy

The effect of the radiation dose on *SFE* of the individual samples is shown in Figs. 1—5. As can be seen, the changes in *SFE* upon the radiation are similar for all the composites studied. *SFE* increases steadily within the entire range of doses. The fastest increase occurs for the doses up to 50 kGy, slower one for 50—100 kGy, and the slowest for the doses above 100 kGy. The course of *SFE* changes reflects the variation in the contact angle, which was used to calculate *SFE*. The *SFE* changes upon the radiation for the 1C0 blend are similar to those for 2C0. It means that the effect of PET on *SFE* of 1C0 is negligible, although the content of this polymer in 1C0 composite is
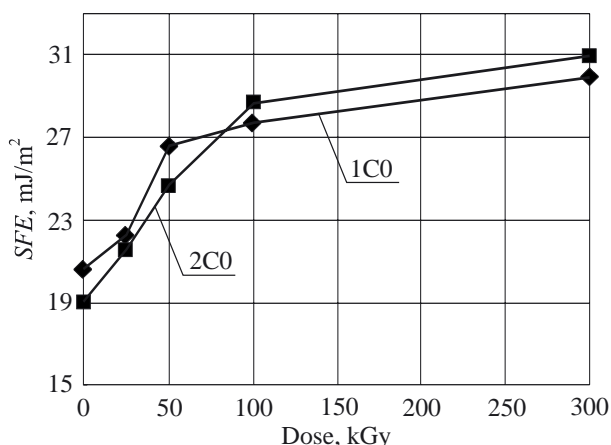

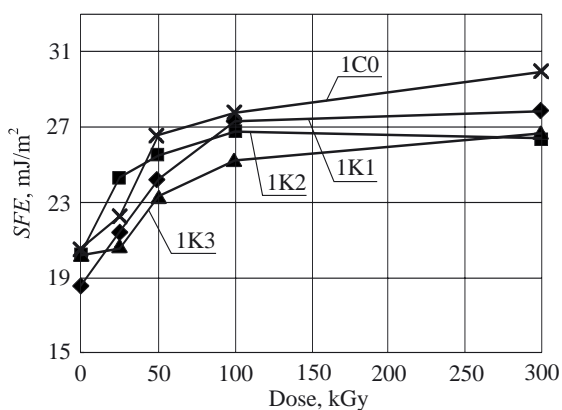Fig. 1. *Effect of the radiation dose on surface free energy (SFE) values of samples 1C0 and 2C0*

Fig. 2. *Effect of the radiation dose on surface free energy (SFE) values of samples 1K1, 1K2 and 1K3 (1C0 is a reference sample)*
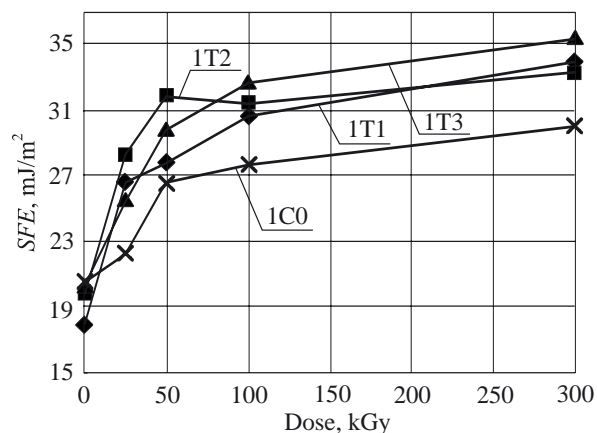

Fig. 3. *Effect of the radiation dose on surface free energy (SFE) values of samples 1T1, 1T2 and 1T3 (1C0 is a reference sample)*
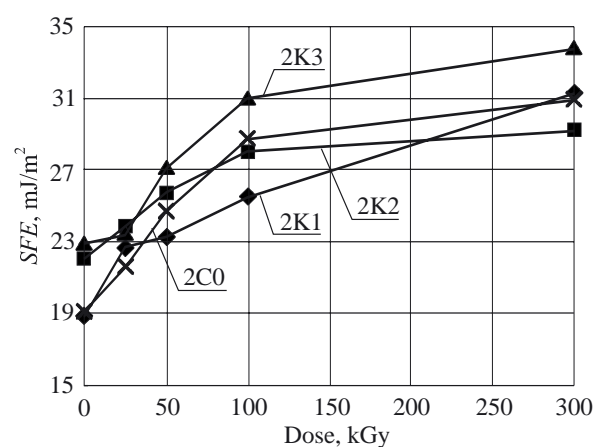

Fig. 4. *Effect of the radiation dose on surface free energy (SFE) values of samples 2K1, 2K2 and 2K3 (2C0 is a reference sample)*
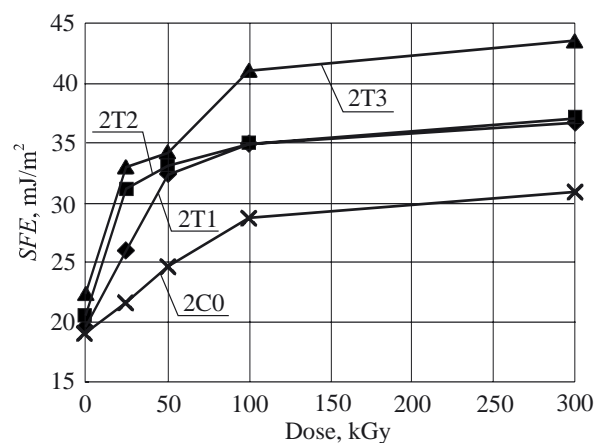

Fig. 5. *Effect of the radiation dose on surface free energy (SFE) values of samples 2T1, 2T2 and 2T3 (2C0 is a reference sample)*

17 % and its *SFE* is the highest among all the components of this blend.

The addition of SEBS-*g*-MA in 1K blends causes a decrease in *SFE* in comparison with the samples irradiated with the same dose (Fig. 2) while this effect in case of 2K is unclear and requires further investigation (Fig. 4). The

introduction of TMPTA into both 1C0 and 2C0 leads to an increase in *SFE* upon sample irradiation. It is especially visible in case of 2T3, for which *SFE* reaches the largest values, *e.g.* above 43 mJ/m^2 for 2T34 (300 kGy, Fig. 5). Thus, one may conclude that TMPTA migrates to the surface layer and causes formation of increased number of functional groups, which leads to an increase in *SFE*, in spite of a relatively low contribution of this compatibilizer. However, confirmation of this hypothesis requires further investigation.

Variations in *SFE* may be described using a function called „susceptibility (*P*) of a composite to *SFE* changes occurring upon radiation". This function has been defined as the derivative of the function *SFE* = *f*(*D*) with respect to the radiation dose (*D*). The relation *SFE* = *f*(*D*) are graphically shown in Figs. 1—5. In order to determine *P*, the analytical form of the function *SFE* = *f*(*D*) is needed. However, the relations presented in Figs. 1—5 can hardly be approximated to the function *P* with sufficient accuracy. Thus, for the purpose of further considerations, mean values (*P'*) of *P* in three ranges of doses were assumed, the ranges being A (0—50 kGy), B (50—100 kGy), and C (100—300 kGy). The value of *P'* [in (mJ/m^2)/kGy] was defined as the ratio of *SFE* difference to the dose difference of the values at the ends of an individual range. It reflects an increase in SFE (in mJ/m^2) upon a unit dose (1 kGy) of the electron radiation.

T a b l e 2. Mean values of susceptibility *P'* [in (mJ/m^2)/kGy] of the studied samples to *SFE* changes occurring upon radiation

Sample group	Dose range[a]		
	A	B	C
1C0	0.120	0.026	0.010
2C0	0.112	0.080	0.011
1K1	0.112	0.063	0.003
1K2	0.106	0.025	-0.002
1K3	0.062	0.038	0.008
1T1	0.197	0.057	0.016
1T2	0.240	-0.009	0.010
1T3	0.190	0.059	0.013
2K1	0.090	0.045	0.029
2K2	0.075	0.044	0.006
2K3	0.085	0.077	0.014
2T1	0.254	0.052	0.009
2T2	0.250	0.038	0.011
2T3	0.236	0.138	0.013

[a] A: 0—50 kGy, B: 50—100 kGy, C: 100—300 kGy.

The calculated values of *P'* are listed in Table 2. Generally, these values clearly decrease with the rising radiation dose for the individual samples groups. This tendency can be expressed using the inequalities: *P'*(A) > *P'*(B) > *P'*(C), being valid for the samples groups specified in Table 2. The only exceptions to this regularity are observed for 1K24 (300 kGy) and 1T23 (100 kGy) and the reason for that was given above while discussing the changes in the contact angle. When neglecting these two samples, one may point out that the values of *P'* for 1C0

and 2C0 in the range A are 10—12 times higher than those in the range C, the values in the range B being intermediate. The largest changes in *P'* for 1C0 and 2C0 composites occur in consequence of TMPTA addition. The values of *P'* for the samples containing this compatibilizer are 1.6—2.3 times larger than those for 1C0 and 2C0. When considering the effect of a dose, it is seen that the values of *P'* in the range A are 12.3—28.5 times higher than those in the range C. Contrary to TMPTA, the addition of SEBS-*g*-MA to 1C0 and 2C0 blends leads to a decrease in *P'* values.

CONCLUSIONS

— The contact angle decreases with the increasing electron radiation dose for all the composites studied. The fastest diminishing occurs in the dose range of 0—50 kGy.

— The relation between the contact angle and a dose for the 1C0 blend is similar to that for 2C0. Differences in the contact angle values for these two composites irradiated with the same dose are small (1.1—3.1 deg).

— Addition of small amounts (1—3 wt. %) of TMPTA into a five- and three-component blends (1C0 and 2C0) enables a significantly higher increase in *SFE* of the composites due to irradiation.

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