Influence of ionizing beta radiation on the adhesion properties and strength of bonded joint of linear low density polyethylene

Martin Bednarik, David Manas, Miroslav Manas, Michal Stanek, Jan Navratil, and Ales Mizera

***Abstract*—**In this study there was found that ionizing beta radiation increased the strength of bonded joints and improved the adhesion properties of linear low-density polyethylene (LLDPE). Generally, for the formation of quality bonded joint it is important to wet the adhesive bonding surface well. Wettability is characterized by the contact angle of wetting. The liquid has to have a lower surface tension than the solid in order to be able to wet the solid substance. The measurement results indicated that ionizing beta radiation was a very effective tool for the improvement of adhesive properties and increased the strength of bonded joints of linear low-density polyethylene. Bonded surfaces with ionizing beta radiation doses of 0, 33, 66, 99, 132, 165, and 198 kGy were irradiated. The best results were achieved by irradiation at dose of 132 kGy by which the highest surface energy and the highest strength of bonded joints of LLDPE were achieved. The strength of bonded joints after irradiation was increased up to 260 % compared to untreated material. A similar trend was observed even for contact angle of wetting and surface energy.

***Keywords*—**Surface energy, linear low-density polyethylene, adhesion, bonded joints.

I. Introduction

Bonding has experienced tremendous expansion in the field of joining materials in the last years. Because of that expansion bonding is classed as new techniques even when it is in fact very old.

In comparison with conventional joining methods (riveting, welding and screwing) bonding provides a new combination of options and it allows obtaining special shapes and properties which cannot be created by conventional methods of coupling. Joining materials using adhesive joints offers several benefits, but also limiting factors if compared with using mechanical joints. To decide about the type of coupling it is necessary to consider the advantages and disadvantages of bonding in comparison with traditional joining technique. [2, 4, 5, 9]

The advantage of bonded joints is that we can connect the same and dissimilar materials and do not take into account the thickness. Application of adhesives does not undermine the coherence of the connect parts and watertight and gastight joints may be prepared. It does not disturb profile or aesthetic appearance of bonded file and does not increase its weight. Joints can be transparent or colored and it is possible to achieve their high strength.

Adhesive bonding has also several technological disadvantages, in comparison with mechanical joints. The disadvantages of bonded joints are the requirements for planeness and cleanness of surface to be bonded and sensitivity to peel stress. Special surface treatments are required for bonded materials with poor adhesion properties and maximum bond strength is reached after the certain time. [2, 12, 14]

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II. EXPERIMENTAL

The aim of the experiment was to examine the effect of beta irradiation on the strength of bonded joints for selected type of material.

A. Material

For this experiment linear low-density polypropylene (LLDPE) were used. Linear low-density polyethylene is the youngest of all types of polyethylene. Its structure is similar to the structure of HDPE but has a low crystallinity given to a large number of short chains. Linear low-density has penetrated almost all traditional markets for polyethylene. It is used to produce plastic bags and sheets, plastic wrap, stretch wrap, pouches and toys. [15, 16, 19]

For bonding LLDPE it is necessary to modify surfaces which will be bonded. Surface modification improves its wettability in aqueous media and also improves adhesive properties. [10, 13]

The samples were made using the injection molding technology on the injection molding machine Arburg Allrounder 420C.

The samples had the shape and dimensions according to the CSN EN ISO 527 – 2 (Fig. 1, Table 1). Before bonding

surfaces of samples ionization beta radiation were irradiated with doses of 0, 33, 66, 99, 132, 165, and 198 kGy at Beta – Gamma Service GmbH & Co. KG, Germany. [3, 6, 17]

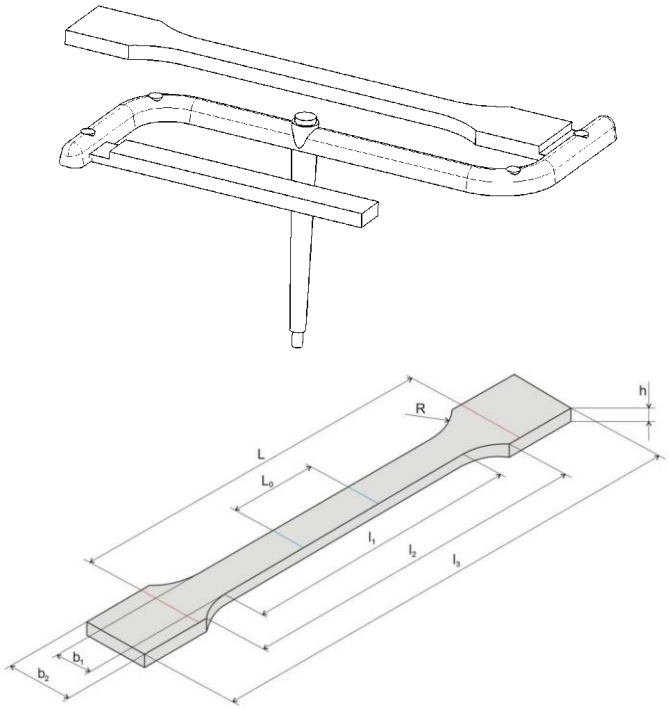

Fig. 1 Testing Specimen

Table 1 Specimen dimensions

Test specimen parameters	
b_1 - Width of Gage Length	10 ± 0.2 [mm]
b_2 - Width of Gripping End	20 ± 0.2 [mm]
l_1 - Length of gage Length	80 ± 2 [mm]
l_2 - Distance Between Gripping Ends	104-113 [mm]
l_3 - Specimen Length	≥ 150 [mm]
L_0 - Distance of Extensometers	30 ± 0.5 [mm]
L - Distance of Grips	115 ± 1 [mm]
h - Specimen Thickness	4 ± 0.2 [mm]
R - Radius	20 - 25 [mm]

B. Surface treatment by beta radiation

Ionizing beta radiation gives inexpensive commodity plastics and technical plastics the mechanical, thermal, and chemical properties of high-performance plastics. This upgrading of the plastics enables them to be used in conditions which they would not be able to with stand otherwise. The energy-rich beta rays trigger chemical reactions in the plastics which results in networking of molecules (comparable to the vulcanization of rubbers which has been in industrial use for so long). [1, 3, 7]

The energy from the rays is absorbed by the material and cleavage of chemical bonds takes place. This releases free radicals which in next phase from desired molecular bonds. [3, 8, 20]

The irradiation cross-linking of thermoplastic materials via electron beam or cobalt 60 (gamma rays) is performed separately, after processing. Generally, ionizing radiation includes accelerated electrons, gamma rays and X-rays. Radiation processing with an electron beam offers several distinct advantages when compared with other radiation sources, particularly γ-rays and x-rays.

The process is very fast, clean and can be controlled with much precision. There is no permanent radioactivity since the machine can be switched off. In contrast to γ-rays and x-rays, the electron beam can be steered relatively easily, thus allowing irradiation of a variety of physical shapes.

The electron beam radiation process is practically free of waste products and therefore is no serious environmental hazard. These are not only capable of converting monomeric and oligomeric liquids, but also can produce, due to cross-linking, major changes in the properties of solid polymers. The cross-linking level can be adjusted by the irradiation dosage. The absorbed dosage means the value of energy of ionizing radiation absorbed by a unit of mass of the processed material. The unit of absorbed dose is 1 Gray (1 Gy = 1J/kg). [1, 3-8]

The main difference between beta and gamma rays is in their different abilities to penetrate the irradiated material. Gamma rays have a high penetration capacity.

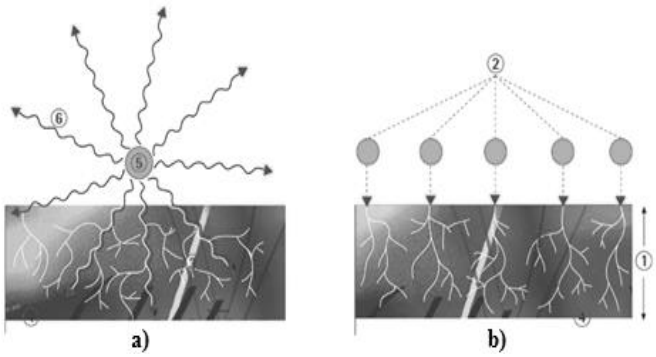

Fig. 2 Design of Gamma Rays (a) and Electron Rays (b) [3]

a) 3 – Secondary electrons, 4 – Irradiated material,
5 – Encapsulated Co – 60 Radiation source, 6 – Gamma rays
b) 1 – Penetration depth of an electron, 2 – Primary electron,
3 –Secondary electron, 4 – Irradiated material

The penetration capacity of electron rays depends on the energy of the accelerated electrons. Due to electron accelerator, the required dosage can be applied within seconds, whereas several hours are required in the gamma radiation plant (Fig. 2). [3-5]

Beta and gamma rays can be used for the irradiation of polyolefines, polyesters, halogen polymers and polyamides from the thermoplastics group, elastomers and thermoplastic elastomers. Some of them need the addition of a cross-linking agent. Polymers can be classified into two groups according to their response to ionizing radiation. One group exhibits predominant cross-linking, the other predominant chain scission. [1, 3, 11, 18]

Irradiation cross-linking of thermoplastic materials deals with creation of a cross-link among the macromolecular strings. Intermolecular forces are replaced by a covalent bond. As a result, we can optimise properties of standard and engineering polymers and impart them the properties of high performance polymers (Fig. 3). [25-39]

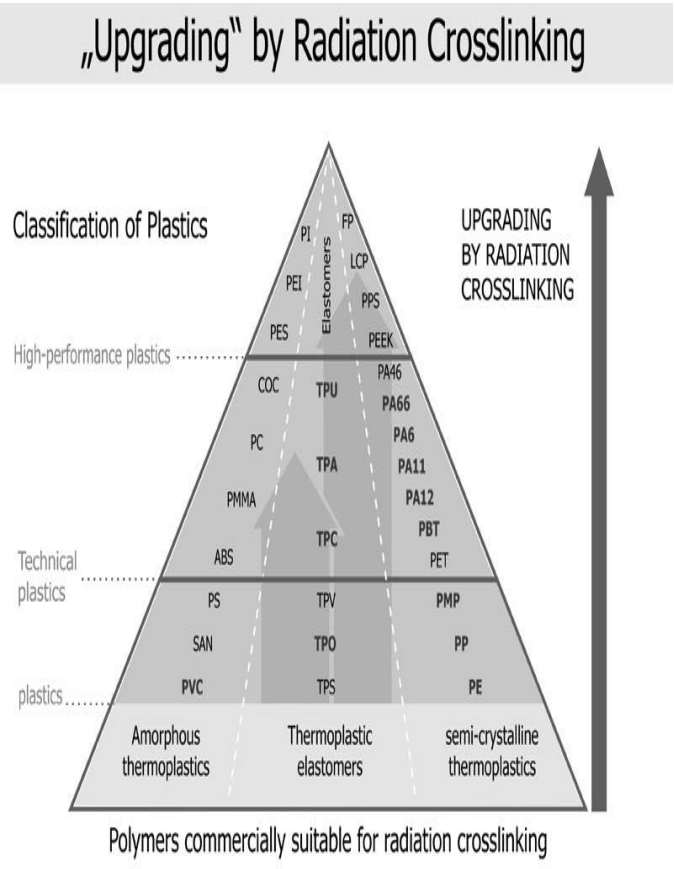

Fig. 3 Pyramid of Polymers [3]

Irradiation of polymers turned out to be interesting because of economic reasons, production costs and a life time of products. However, these benefits depend on the type of irradiated polymer and the radiation dosage. Behaviour of each material is different after irradiation.

We cannot expect the improvement in all areas (in mechanical, thermal and chemical). Most of polymers are not suitable for irradiation because of degradation and deterioration of their properties. [3-8, 49-62]

C. Wetting contact angle measurements

The angle of contact was measured by using the sessile drop (Fig. 4) and surface energy was determined by OWRK method (Owens – Wendt – Rabel – Kaeble method).

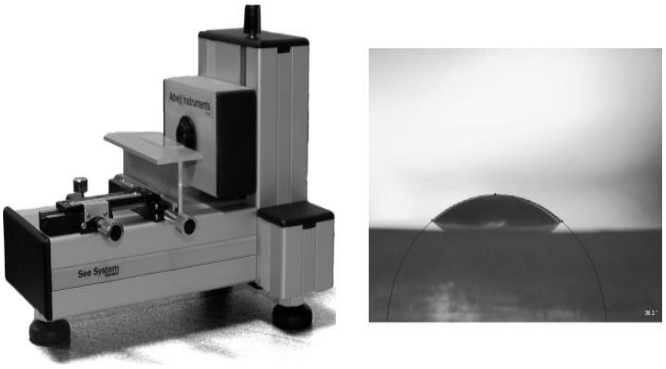

Fig. 4 Surface Energy Evaluation System (See System)

The liquids water, glycerol and ethylene glycol with known γ^p (polar component) and γ^d (dispersive component) were used for calculating the surface energy of LLDPE. [2, 11, 40-48]

Table 2 Surface energy of liquids

Liquid	γ_1 (mJ/m^2)	γ_1^d (mJ/m^2)	γ_1^p (mJ/m^2)
Distilled water	72.8	21.8	51.0
Glycerol	64.0	34.0	30.0
Ethylene glycol	48.0	29.0	19.0

D. Testing the strength of bonded joints

After sample preparation (production and irradiation) contact angles were measured and surface energy was calculated. Then the samples were bonded and their strength was measured. For testing the strength of bonded joints there was used a tensile test on the test machine Zwick 1456 (Fig. 5, Fig. 6). Test conditions were according to the CSN EN ISO 527-1 and CSN EN ISO 527-2. Speed was 10 mm/min and evaluation software was Test Expert Standard. [18, 20-24]

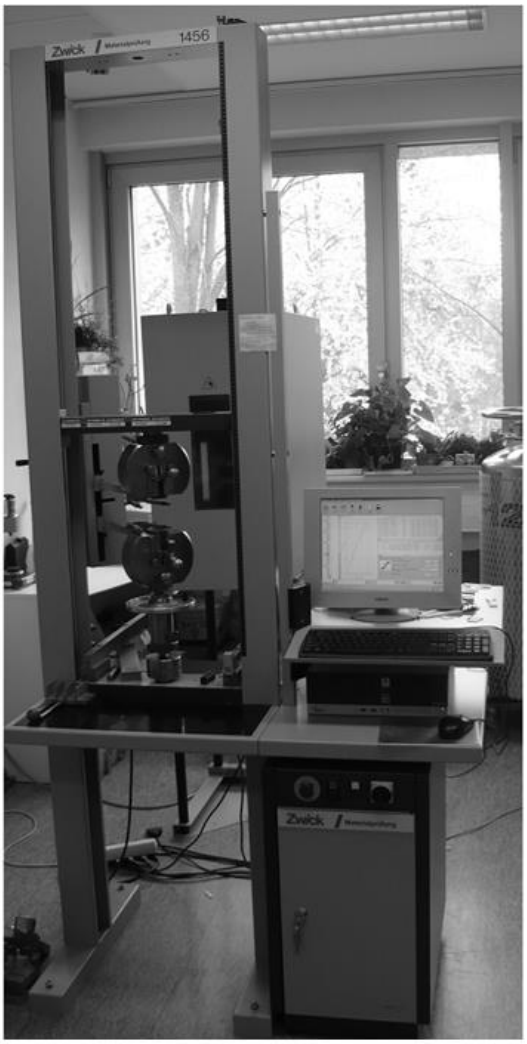

Fig. 5 Zwick 1456

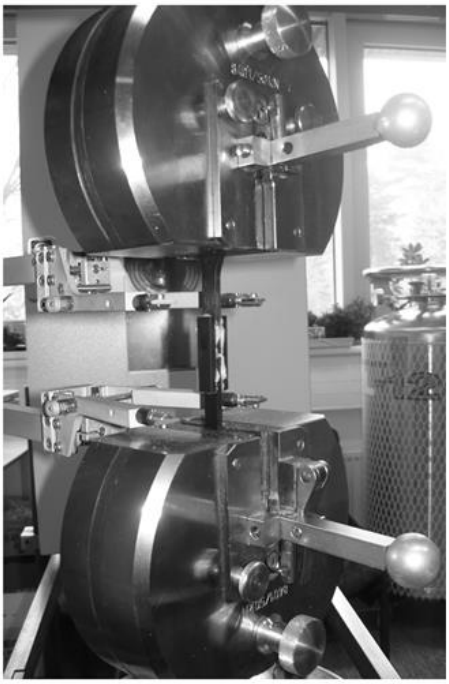

Fig. 6 Bonded joint

III. Results and Discussion

A. Contact angle and surface energy

The variation in contact angle of LLDPE for different doses of radiation and for different test liquids is shown in Fig. 7.

It shows that the contact angle on the untreated surface is 84.9°, 76.6°, and 62.9° for distilled water, glycerol, and ethylene glycol, respectively. The contact angle values were considerably reduced after irradiation by a dose of 132 kGy to lower values of 60.1°, 59.3°, and 49.0° for distilled water, glycerol, and ethylene glycol, respectively (referring to: Fig. 7).

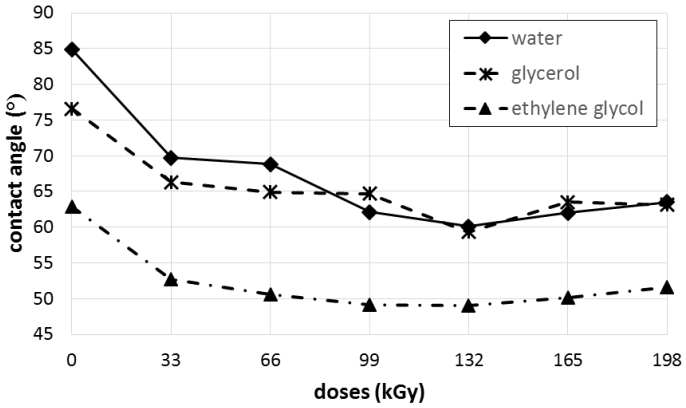

Fig. 7 Variation of contact angle with respect to radiation dose for material LLDPE

Fig. 8 shows a plot of surface energy γ_s from the measured contact angles on the surface of the LLDPE. It shows that surface energy of untreated surface is 27.1 mJ/m^2 and 26.7 mJ/m^2 for water + glycerol and water + ethylene glycol, respectively. Ionization beta radiation increases the surface energy. The surface energy values considerably increased after irradiation by a dose of 132 kGy to higher values of 42.1 mJ/m^2 for water + glycerol and 42.3 mJ/m^2 for water + ethylene glycol.

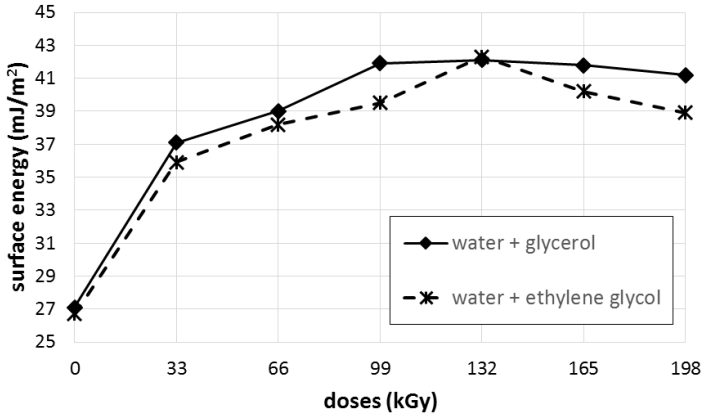

Fig. 8 Variation of surface energy with respect to radiation dose for material LLDPE

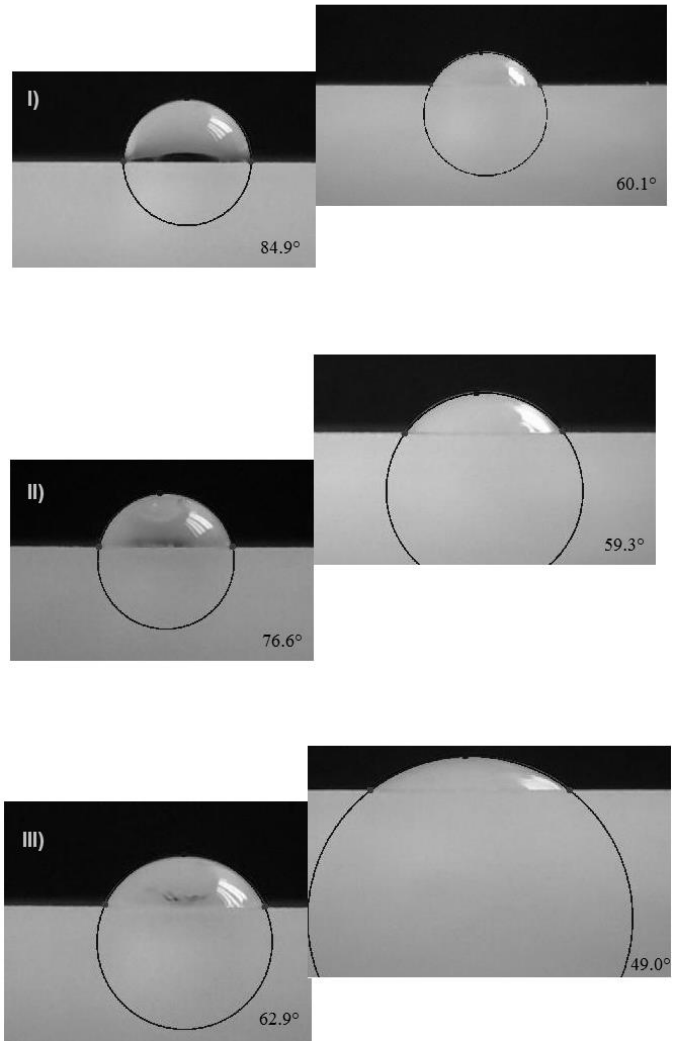

Fig. 9 Contact angle before and after irradiation with dose of 132 kGy (I – water; II – glycerol; ethylene glycol)

B. Strength of bonded joints (at 23 °C)

Strength of bonded joints is characterized by the maximum burdensome force which endured bonded sample. For bonding of LLDPE one type of two-component methacrylate adhesive (Cyberbond A806), one type of two-component epoxy adhesive (Cyberbond E705), and one type of cyanoacrylate adhesive (Cyberbond 2028) were used. Basic properties of the adhesives used are shown in Table 3, Table 4, and Table 5.

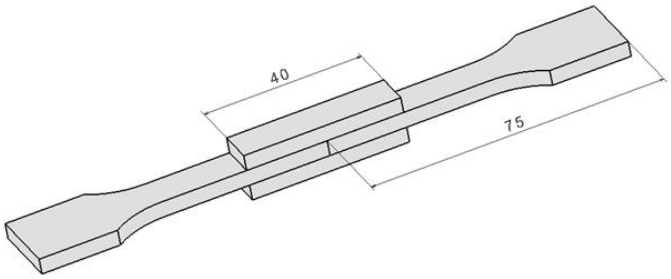

Fig. 10 Bonded joint

Table 3 Properties of adhesive Cyberbond A806 [63]

A+B Mixture (liquid)	
Basis Monomer	Methylmethacrylat + Hardener
Appearance	Beige
Viscosity Plate/Cone	50.000 mPa.s
Flashpoint (acc. Abel Pensky)	10 °C
Mixing Ratio	1:1
Gel Time at Room Temperature	15 – 18 min.
Fixture Time	30 min.
Shelf Life in Unopened Packaging	9 months
Polymer (solid)	
Shore D Hardness	65 – 70
Peeling Strength acc. to DIN 53283 (steel)	5 – 10 N/cm
Tensile Shear Strength acc. to DIN 53283 (steel)	20 – 30 N/mm^2
Elongation at Break	1.5 – 2.5 %
Temperature Range	-40 / +120 °C

Table 4 Properties of adhesive Cyberbond E705 [63]

Properties (uncured)	
Chemical Base	Epoxy
Appearance	Light amber
Mixed Viscosity	10.000 mPa.s
Spec. Gravity	1.2 g/cm^3
Shelf Life in Unopened Packaging	12 months
Curing Behavior…(fast)	
Potlife at Room Temperature	5 – 8 min.
Mixture Ratio	1:1
Functional Cure	15 min.
Final Cure after	3 – 6 hours
Polymer Properties	
Peel Strength	Not Recommended
Operating Temperature	-40 / +95 °C

Table 5 Properties of adhesive Cyberbond 2028 [63]

Physical Properties – Monomer (Uncured)	
Base Compound	Ethyl
Appearance	Clear
Viscosity	200 +/- 40 cPs
Specific Gravity	1.06 g/cc
Flash Point	85 °C
Shelf Life	12 mo
Storage Condition	20 °C
RoHS – Compliant	yes
Physical Properties – Polymer (Cured)	
Full Cure Time	24 hours
Appearance	Clear
Service Temp Range	-55 to 95 °C
Setting Time	
Steel	20 to 50 seconds
ABS	2 to 4 seconds
EPDM	1 to 4 seconds
Performance of Cured Adhesive	

Substrate	**N/mm^2**	**PSI**
Steel	16.3 to 18.8	2360 to 2730
Rubber*	5.1 to 15.2	735 to 2200
AL	15.3 to 18.1	2225 to 2630
PC**	22.3 to 24.9	3240 to 3605
PVC**	18.3 to 26.7	2660 to 3875
ABS**	12.0 to 18.3	1740 to 2660

**Rubber figures given are typical. Your results may vary by specific rubber type.*
***Tested to ASTM 4501.*
****n/r = not recommended*

The highest strength of bonded joints samples of LLDPE (adhesives Cyberbond 2028) have those which were irradiated by a dose of 132 kGy. After the irradiation by a dose of 132 kGy strength is increased by 260 % (referring to: Fig. 11).

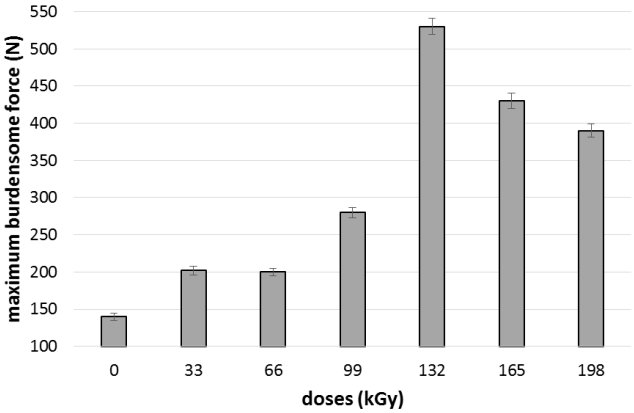

Fig. 11 Variation of strength of bonded joints with respect to radiation dose (Cyberbond 2028)

The highest strength of bonded joints samples of LLDPE (adhesives Cyberbond A806) have those which were irradiated by a dose of 132 kGy. After the irradiation by a dose of 132 kGy strength is increased by 200 % (referring to: Fig. 12).

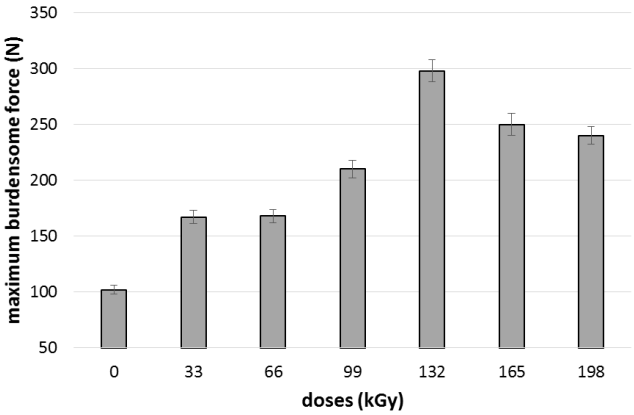

Fig. 12 Variation of strength of bonded joints with respect to radiation dose (Cyberbond A806)

The highest strength of bonded joints samples of LLDPE (adhesives Cyberbond E705) have those which were irradiated by a dose of 132 kGy. After the irradiation by a dose of 132 kGy strength is increased by 200 % (referring to: Fig. 13).

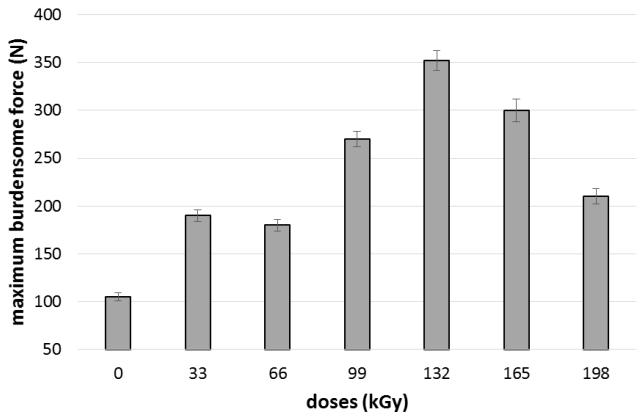

Fig. 13 Variation of strength of bonded joints with respect to radiation dose (Cyberbond E705)

C. Strength of bonded joints at elevated temperature (60 °C)

The highest strength of bonded joints (at elevated temperature) samples of LLDPE (adhesives Cyberbond 2028) have those which were irradiated by a dose of 165 kGy. After the irradiation by a dose of 165 kGy strength is increased by 230 % (referring to: Fig. 14).

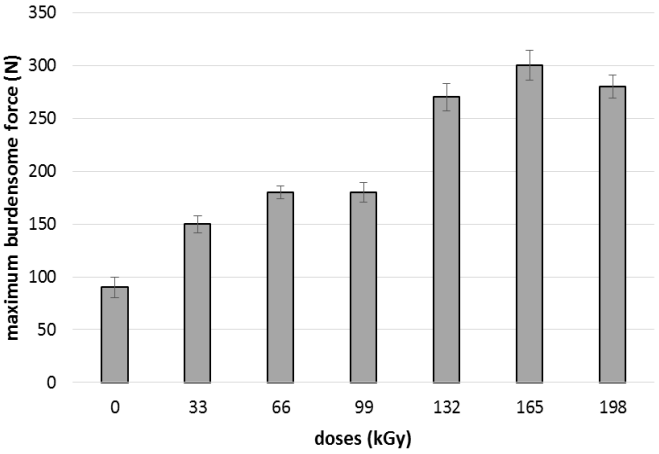

Fig. 14 Variation of strength of bonded joints (at elevated temperature) with respect to radiation dose (Cyberbond 2028)

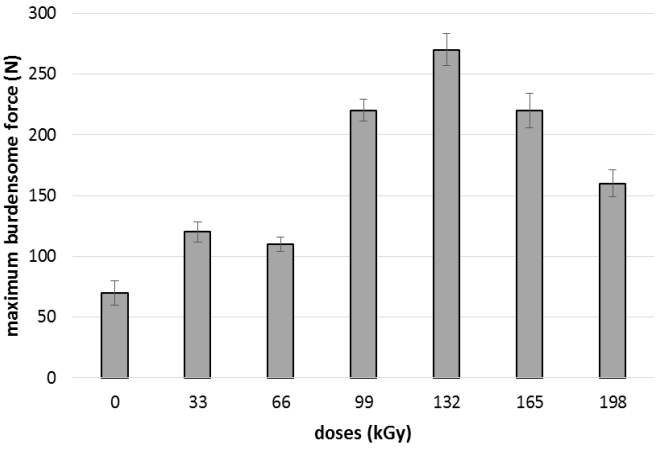

Fig. 15 Variation of strength of bonded joints (at elevated temperature) with respect to radiation dose (Cyberbond E705)

The highest strength of bonded joints (at elevated temperature) samples of LLDPE (adhesives Cyberbond E705) have those which were irradiated by a dose of 132 kGy. After the irradiation by a dose of 132 kGy strength is increased by 260 % (referring to: Fig. 15).

IV. Conclusion

This article describes the effect of beta radiation on the contact angle of wetting, on the surface energy and on the final strength of bonded joints of LLDPE. Beta radiation increases the strength of bonded joints of LLDPE and improves their adhesion properties. The best results were achieved by irradiation at doses of 132 kGy by which the highest surface energy and the highest strength of bonded joints of LLDPE were achieved.

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