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Improving the Glued Joint Strength by Modifying the Beechwood (*Fagus sylvatica* L.) with Gamma Rays

Povećanje čvrstoće lijepljenog spoja modifikacijom bukovine (*Fagus sylvatica* L.) ozračivanjem gama zrakama

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SUMMARY • *The reserach was done on the effects of gama rays on the change of the critical surface energy* γ_c*, work of adhesion* W_a*, penetration* W_p*, spreading* W_s*, and the change of the glued joint strength in beechwood. The samples absorbed the irradiated dosages of 25,50 and 100 kGy, which was followed by measurements of the wetting angle. The surface parameters were calculated from the wetting angle and the surface tension of the liquid. The results show the increase of all observed parameters with an increased dose of radiation. The glued joint strength did not significantly change at low absorbed irradiated dosages of 25 and 50 kGy, but at 100 kGy, the increase in shear strength of the glued joint was considerable.*

Key words: *gamma rays irradiated beechwood, wetting angle, glued joint strength*

SAŽETAK • *U radu su provedena istraživanja utjecaja zračenja bukovine gama zrakama na promjenu kritične površinske energije* γ_c*, rada adhezije* W_a*, penetracije* W_p *i razlijevanja* W_s *te promjenu čvrstoće lijepljenog spoja. Uzorci su ozračivani apsoribranim dozama zračenja od 25, 50 i 100 kGy, nakon čega je mjeren kut kvašenja, te su iz kuta kvašenja i površinske napetosti tekućine izračunati ranije navedeni parametri. Dobiveni rezultati ukazuju da sa*

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povećanjem doze zračenja rastu i svi promatrani parametri. Čvrstoća lijepljenog spoja nije se značajno promjenila kod malih apsorbiranih doza zračenja od 25 i 50 kGy, ali kod doze od 100 kGy došlo je do značajnog povećanja smične čvrstoće lijepljenog spoja.

Ključne riječi: *zračenje bukovine gama zrakama, kut kvašenja, čvrstoća lijepljenja*

1. INTRODUCTION
1. Uvod

Continuing rise in timber prices and constant quality decline, connected with the growing interest for solid wood products, has all led to the research into the adhesion and the application of new practices of wood gluing. This is apparently the only way to achieve a more economic conversion of timber into final products.

With the objective of obtaining higher strength of the glued joints, we tried to modify beechwood by exposing it to gamma-rays in order to increase the free surface energy, and consequently, to achieve higher adhesion when using the PVAC (polyvinylacetate) glue.

Nagieb (1988) did a research on the exposure of the cotton stems and wood sawdust to gamma rays for higher sugar yield. The absorbed dosages were 100, 500, and 1000 kGy. A more intensive formation of carboxyl and carbonyl groups was followed by a simultaneous fall of the polymerization degree. Sugar yields from lignocellulose residues exposed to doses higher than 500 kGy grew abruptly.

Ueno (1990) studied exposures to gamma rays of different materials used in paper production (hardwood and softwood kraft pulp, both bleached and unbleached). The absorbed dosages were 10, 25, 50, and 100 kGy. The results were that the higher the dose was, the lower the "dry" tensile strength and tearing resistance, whereas the "wet" tensile strength displayed a certain rise with the growth of the irradiation dosage.

In the gluing of wood, it is important to see what changes cellulose would undergo after it is exposed to gamma rays. According to Dole (1972), the main effect of the cellulose irradiation is the splitting of the main chains into shorter ones, supported by the formation of hydrogen ions and acid fragments. The aromatic compunds bound to the cellulose protect the main chain from breaking. The removal of water from the wood increases the number of formed radicals. An example is, where the water contents is 0.5%, three times as many radicals are formed than with a water content of 7%. The radicals that do not disintegrate in contact with water are "long-living", since they have been captured in the crystalline area. The free radicals captured in the cellulose may initiate a polymerization reaction with a suitable monomer, so that a secondary polymer may be split into cellulose. Thus formed polymer has apparently different properties than the original cellulose. The improvements include a higher elasticity, toughness and resistance to decay and abrasion. The cellulose may be first irradiated, then treated, but it can also be gamma-rayed together with the monomer. Homo- polymerization is present in both cases.

If there is oxygen near the place of the chemical change caused by irradiation, there is a higher probability of forming the hydrogen peroxide ($O_2 + H^o + H^o \rightarrow H_2O_2$) or the peroxide radical ($R^o + O_2 \rightarrow RO_2$), which are oxidants and therefore can chemically react with wood, i.e. they can thus modify it.

The purpose of the research was to find out the possibilities for improving the glued joint strength by modification of wood surface by ionization.

2. MATERIALS AND METHODS
2. Materijal i metode

All samples were made of beechwood (*Fagus sylvatica*), with an average ring width 5.8 mm, density 730.79 kg/m^3. All measurements were performed on an area with radial surface machined by fine planing (R_a = 5.5 μm). The samples were conditioned to the moisture content of 10%. Sample dimensions were 180x140x10mm. In the first three groups, each seven samples were gamma-rayed with absorbed dosages of 25 kGy, 50 kGy, and 100 kGy, while the fourth group consisted of seven unexposed samples to serve for comparison. One sample from each group was used for measuring the wetting angle, the remaining six from each group were glued together and processed into samples for testing the shear strength as shown in Figure 1.

Measuring the wetting angle
Mjerenje kuta kvašenja

At each step, the angles of wetting θ' were measured for 9 liquids with different, though known, surface tension $\gamma_{L,G}$, whose values ranged between 55.153 and 72.400 mN/m. For each liquid, the angle was measured

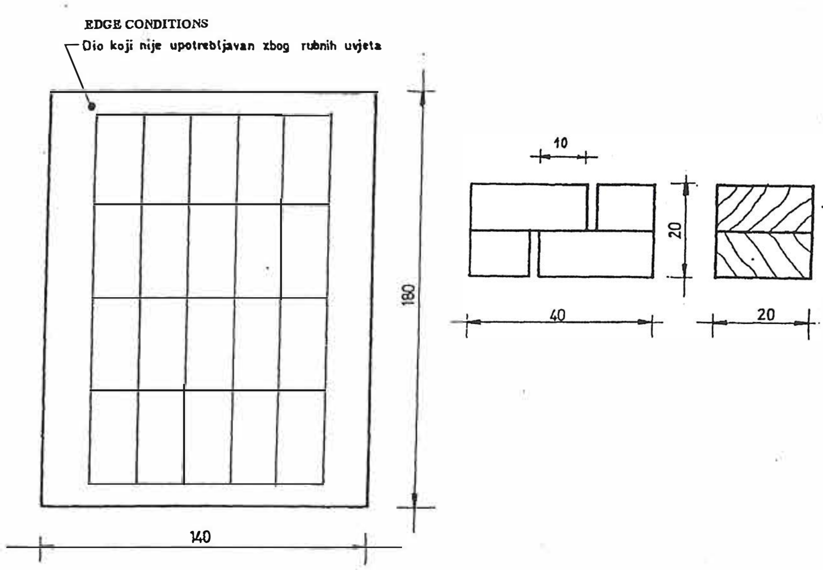

Figure 1.
Samples, their shape and size, for testing the shear strength • Shema izrade proba, oblik i dimenzije probe za ispitivanje smične čvrstoće

three times at three different places on the sample, and the mean value calculated. Using the wetting angles and the known surface liquid tensions, we determined the critical surface energy γ_c, work of adhesion W_a, work of penetration W_p, and work of spreading W_s. A detailed description of the method is given in Bogner's two previous studies (1993, 1995).

The work required for separation of the liquid from the solid phase is called the work of adhesion Wa. We calculated it using the equation (1) for each measured wetting angle.

$$W_a = \gamma_{L,G}\,(1+\cos\theta') \quad (1)$$

The work done by the liquid while entering the wood capillary is called the work of penetration W_p, and was calculated with equation (2) for each measured wetting angle:

$$W_p = \gamma_{L,G}(\cos\theta') \quad (2)$$

The work needed for the liquid to spread over the wood surface is called the work of spreading W_s, calculated with the equation (3) for each measured angle:

$$W_s = \gamma_{L,G}(\cos\theta'-1) \quad (3)$$

Values W_a, W_p, and W_s change with variations in the surface tension of the liquid wetting the wood surface by the parabola rule, so that the optimal values are at the top of the parabola.

Measruing the shear strength
Mjerenje smične čvrstoće

As the shear strength with this sample form was achieved by compression, the samples had slightly different dimensions (greater thickness and smaller length) than those prescribed by the Croatian Standard HRN H.K8.024, while the overlap dimensions are as usual (10 x 20 mm). We chose this sample form, as the overlaps cover a considerably bigger sample surface than it would be in case of using the standard form. For each group 60 samples were prepared.

The gluing was done with the PVAC glue of the Austrian firm WEGIN. The glue was applied at a spreading rate of 200 g/m^2, the assembly time being between 1 and 3 minutes.

Pressing parameters:
- gluing procedure was cold;
- all samples from each group were pressed at the same time
- the specific pressure was 0.3 MPa
- duration of pressing was 24 hours
- time of sample conditioning before preparation of the samples was one week.

The measuring of the shear strength was done on the WOLPERT tensile-testing machine at a constant speed of 6 mm/min.

The calculation of the shear strength was done by using the equation (4)

$$\tau = F/A \quad (4)$$

where τ = shear strength of the glued joint (Pa), F = breaking force (N), A = overlapping surface (m^2).

Using the shear strength of each individual sample, we calculated the mean strength $\bar{\tau}$ and evaluated the standard deviation S for the individual sample groups.

The standard strength was calculated with the equation (5)

$$\tau n = \bar{\tau} - 2S \quad (5)$$

and the strength equalization factor U by using equation (6)

$$U = \tau n / \bar{\tau} \quad (6)$$

Equation (6) shows that the joint strength increases with lower variability, the result being a higher equalization factor U.

3. RESEARCH RESULTS
3. Rezultati istraživanja

The rsaearch results of the critical surface energy γ_c are shown graphically on Fig. 2.

Figures 3 to 5 show the results of the research on the relation between the surface liquid tension and the work of adhesion, penetration and spreading, and the calculation of the maximum adhesion work $W_{a,max}$ and the related optimal surface liquid tension $\gamma_{L,G,opt}$. presented in Table 1.

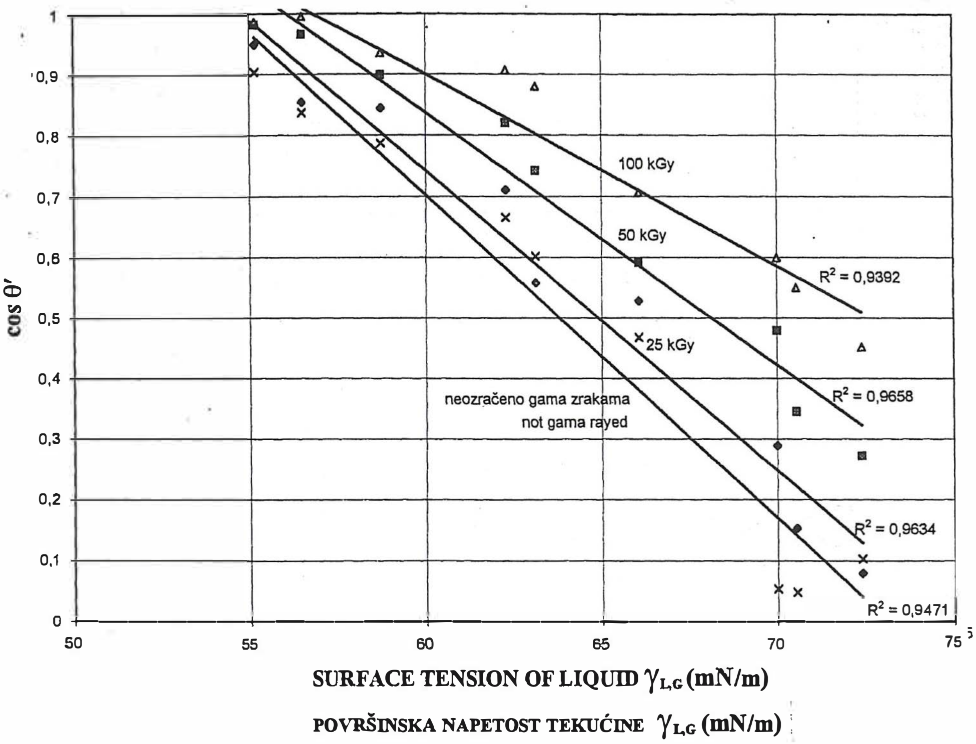

Fig. 2
The relation between $\gamma_{L,G}$ and COS θ' with the calculation of the critical surface energy γ_c. • Odnos $\gamma_{L,G}$ *i COS θ' uz proračun kritične površinske energije* γ_c

- unexposed to gamma rays - neozračeno gama zrakama γ_c =54,4 mN/m
- exposed to 25 kGy - ozračeno dozom od 25 kGy $\gamma_{L,G}$ =54,9 mN/m
- exposed to 50 kGy - ozračeno dozom od 50 kGy γ_c =56,1 mN/m
- exposed to 100 kGy - ozračeno dozom od 100 kGy γ_c =56,8 mN/m

γ_c=54,4 mN/m
γ_c=54,9 mN/m
γ_c=56,1 mN/m
γ_c=56,8 mN/m

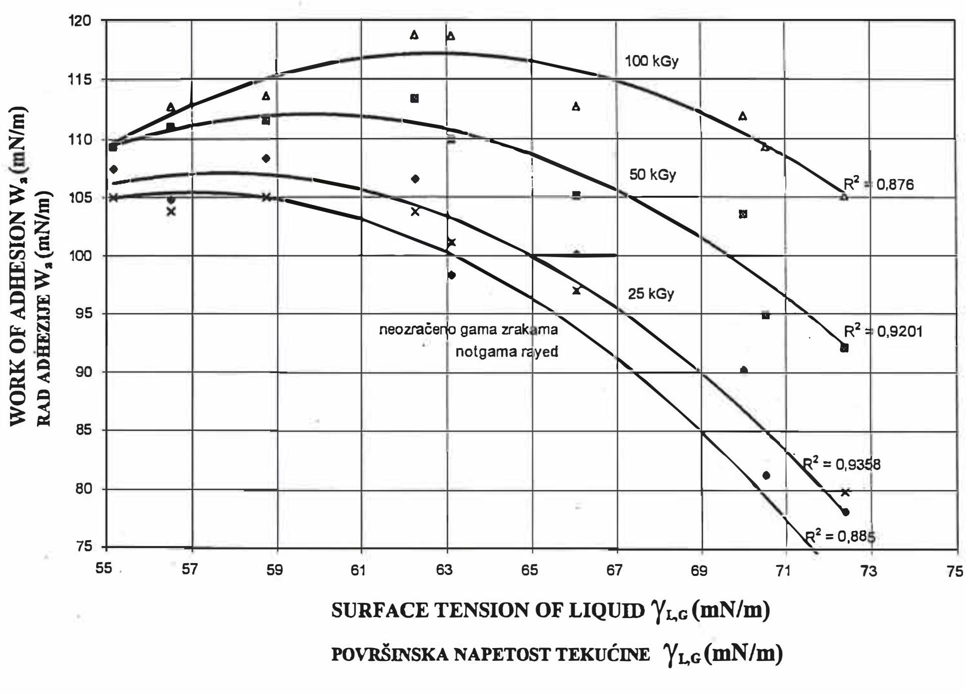

Figure 3.
The relation between the surface liquid tension $\gamma_{L,G}$ and adhesion work W_a. • *Odnos između površinske napetosti tekućine $\gamma_{L,G}$ i rada adhezije W_a.*

Figure 4.
The relation between the surface tension $\gamma_{L,G}$ and the work of penetration W_P. • Odnos između površinske napetosti tekućine $\gamma_{L,G}$ i rada penetracije W_P.

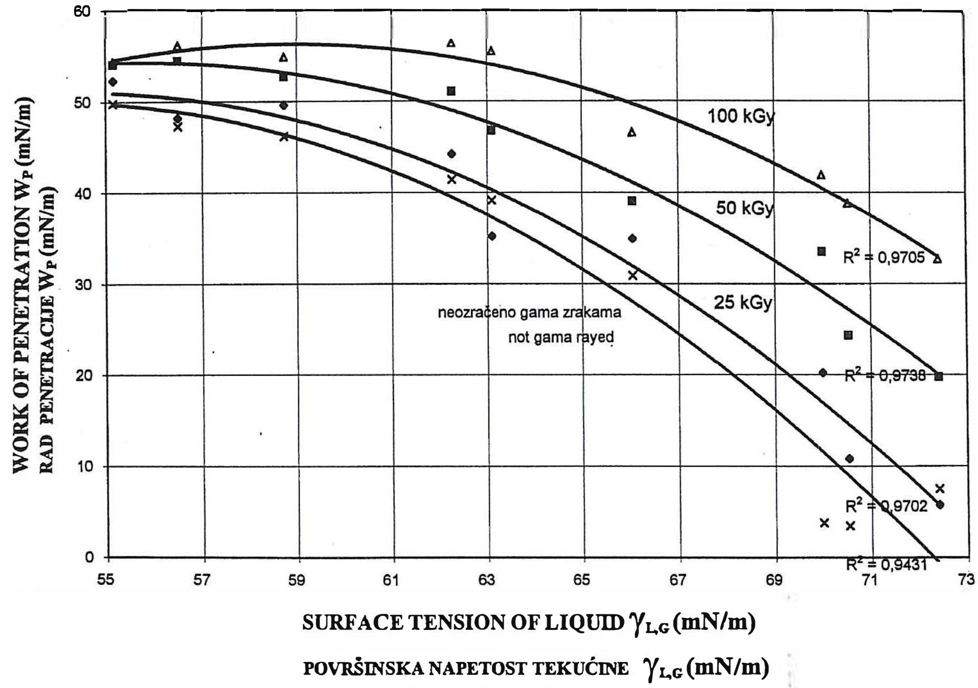

Figure 5.
The relation between the surface liquid tension $\gamma_{L,G}$ and the work of spreading W_S. • Odnos između površinske napetosti tekućine $\gamma_{L,G}$ i rada razlijevanja W_S.

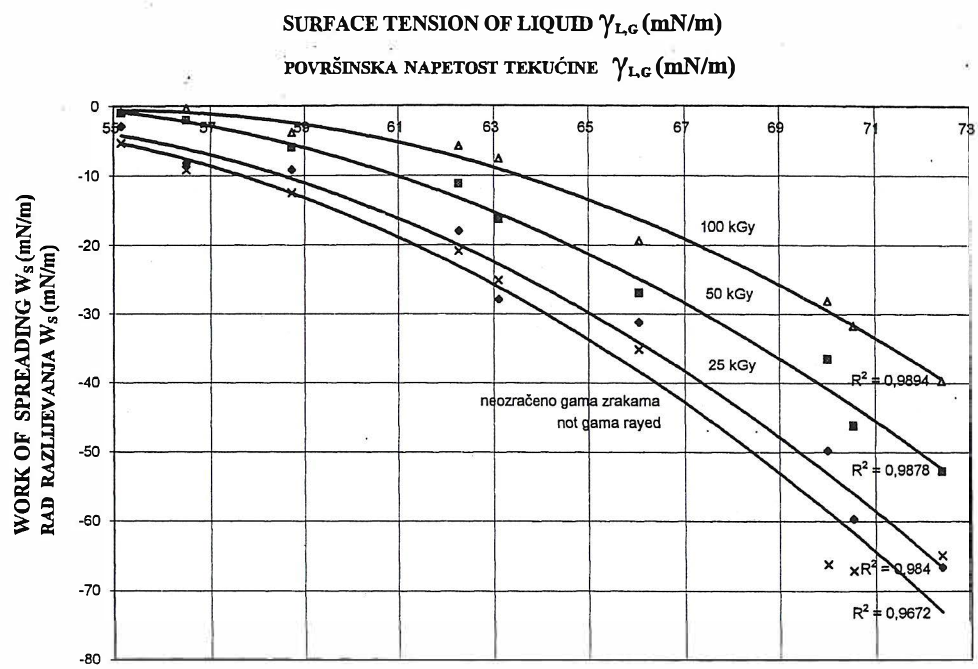

Table 1
Maximum adhesion work and optimal surface liquid tension • Maksimalni rad adhezije i optimalna površinska napetost tekućine

Treatment Obrada	$W_{a,max}$ mN/m)	$\gamma_{L,G,opt}$ (mN/m)
unexposed to gamma rays neozračeno gama zrakama	105.91	57.1
exposed to 25 kGy ozračeno sa 25 kGy	107.13	57.7
exposed to 50 kGy ozračeno sa 50 kGy	112.17	59.7
exposed to 100 kGy ozračeno sa 100 kGy	117.2	62.6

The results of the research on gluing strength are shown in Table 2.

Treatment Obrada	τ(MPa)	S (MPa)	τ_n (MPa)	U
unexposed to gamma rays neozračeno gama zrakama	14.88	1.78	11.32	0.761
exposed to 25 kGy ozračeno sa 25 kGy	14.33	1.68	10.97	0.765
exposed to 50 kGy ozračeno sa 50 kGy	14.85	1.52	11.81	0.795
exposed to 100 kGy ozračeno sa 100 kGy	16.98	2.26	12.46	0.734

Table 2
The results of the research on shear strength τ of the glued joints • Rezultati ispitivanja smične čvrstoće τ lijepljenih spojeva

4 DISCUSSION AND CONCLUSION
4. Diskusija i zaključak

The results show that the increase of the absorbed irradiation dosage causes the increase of all observed parameters. Thus increases the critical surface energy, which was in unexposed samples 54.4 mN/m, and in those exposed to 100 kGy, it amounted to 56.8 mN/m. Likewise, the values of adhesion, penetration and spreading were increased. It was the growth of the critical surface energy, which is a good approximation for the real wood surface energy, that influenced the growth of other observed parameters. Thus was the maximum adhesion of the unexposed samples 105.91 mN/m, and 117.22 mN/m in samples exposed to 100 kGy. To achieve maximum adhesion on wood surface with certain surface energy, it is necessary that the liquid has a certain surface tension, which was marked in the table 1. To achieve strength and durability of the joint, it is of greatest importance to obtain the best possible adhesion, since, one way or another, the penetration and spreading support each other by the pressure in almost all wood gluing processes. The glued joint strength has not changed singificantly with small absorbed dosages of 25 and 50 kGy, while with 100 kGy a considerable increase occured in the shear strength of the glued joint. The research did not include the changes of the physical and mechanical wood properties resulting from the exposure to gamma rays. They will be the subject of our future research.

5. REFERENCES
5. Literatura

1. Bogner, A. (1993): Kvašenje drva i adhezija. Drvna industrija 44 (4) 139-143
2. Bogner, A. (1995): Work of adhesion as a criterion for determination of optimum surface tension in adhesives. Drvna industrija 46 (4) 187-194
3. Dole, M. (1972): The Radiation Chemistry of Macromolecules. Vol. 1&2, Academic Press, N.Y.
4. Nagieb, Z.A., El-Meadaway, S.A. (1988): Study on the Effect of Gamma-Radiation on Different Egyptian Lignocelluloses. Holzforschung und Holzverwertung, 40 (6), 147-149
5. Ueno, T., Takahashi, M. (1990): Effect of Irradiation on the Properties of Paper and Paperboard I. Journal of the Japan Wood Research Society 36 (8), 665-671