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PENETRATION OF UREA-FORMALDEHYDE ADHESIVES IN WOOD TISSUE
PART I: RADIAL PENETRATION OF UF ADHESIVES INTO BEECH

Abstract: Adhesive penetration plays an important role in wood adhesion, since wood is a porous material. The degree of penetration mostly depends on the wood factors, resin type and formulation and processing parameters. Tangentially cut 5 *mm* thick beech wood (*Fagus moesiaca*) plies, 100 *mm* long (parallel to grain) and 30 *mm* wide, were prepared for this study. The urea-formaldehyde (UF) adhesive was applied to the surface of one ply. Two plies were assembled into sample so that the grains of two plies were parallel. Samples were pressed in a hydraulic press at 120°*C* and 0,7 *MPa* for 15 *min*. Microtome test-specimens were cut of each sample. 20 μ*m* thick microtomes were cut by sliding microtome apparatus, exposing a bondline with a cross-sectional surface. The lack of more exhausting research on the penetration of urea-formaldehyde adhesives in wood is evident. Since urea-formaldehyde (UF) glue resins were the most important type of adhesives in the wood industry in the last 60 years (Dunky, 2000), the objective of this research was microscopic detection of UF adhesive penetration in wood tissue. Four types of UF resins with different levels of polycondensation were used in this research. Safranin was added in resins, since epi-fluorescence microscope was used in this research for measuring the adhesive penetration.

Key words: penetration, beech (*Fagus moesiaca*), degree of polycondensation, urea-formaldehyde (UF) adhesive, fluorescence microscopy

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ПЕНЕТРАЦИЈА УРЕА-ФОРМАЛДЕХИДНОГ АДХЕЗИВА У ТКИВО ДРВЕТА

ДЕО I: РАДИЈАЛНА ПЕНЕТРАЦИЈА УФ АДХЕЗИВА У ДРВО БУКВЕ

Извод: Пенетрација адхезива има значајну улогу у адхезији дрвета, пошто је дрво порозни материјал. Степен пенетрације највише зависи од фактора дрвета, врсте и рецептуре адхезива и параметара пресовања. За ово истраживање припремљене су тангенцијално резане дашчице (бочнице) буковог дрвета (*Fagus moesiaca*) дебљине 5 *mm*, дужине 100 *mm* (паралелно са влакнима) и ширине 30 *mm*. Уреа-формалдехидни адхезив је нанешен на површину једне плочице. Две дашчице су слепљене у узорак тако да су им паралелна дрвна влакна. Узорци су пресовани у хидрауличној преси 15 *min* на температури од 120°*C* и притиску од 0,7 *MPa*. Попречни микротомски препарати, дебљине 20 µ*m* исечени су на клизећем микротому. Евидентан је недостатак исцрпнијих истраживања пенетрације уреаформалдехидних (УФ) адхезива у дрво. Пошто су УФ-смоле најважнија врста адхезива у дрвној индустрији у последњих 60 година, циљ овог истраживања је микроскопско одређивање УФ-адхезивне пенетрације у ткиво дрвета. У сврху овог истраживања коришћене су четири УФ-смоле различитог степена поликондензације. У везива је додат сафранин пошто је за мерење дубине пенетрације адхезива коришћен епи-флуоресцентни микроскоп.

Кључне речи: пенетрација, букова дрво, притисак, степен поликондензације, УФ-адхезив, флуоресцентна микроскопија

1. INTRODUCTION

The use of polymeric resins to bond wood components has played a vital role in the development and growth of the forest products industry. Adhesives are indispensable for the process of manufacturing wood products. However, on an equivalent weight basis, adhesives are very expensive compared to wood, and therefore their use must be judiciously controlled (Kamke, Lee, 2007).

Since wood is a porous material, adhesive penetration plays an important role in wood adhesion. To facilitate adequate mechanical interlocking, the adhesives must have penetrated into the wood before it is cured (Wang, Yan, 2005). The degree of penetration depends on the wood factors (e.g. species, cutting direction: tangential, radial and longitudinal, earlywood and latewood, heartwood and sapwood, surface energy), resin type and formulation (e.g. molecular weight distribution, solid content, addition of fillers and other additives, surface tension of the liquid phase of the adhesive), processing parameters (e.g. open assembly time, pressing time, temperature and consolidation pressure).

Adhesive penetration into wood may be categorized into gross penetration (micrometer level penetration) and cell-wall penetration (nanometer level penetration). The former results from the flow of liquid resin into the porous structure of wood, mostly filling cell lumens. The latter occurs when resin diffuses into the cell wall or flows into micro fissures. Gross penetration has been defined as a motion of adhesive from the external surface into the capillary structure of wood and encapsulating fractures and surface debris caused by processing (Marra, 1992). Gross penetration is due mainly to hydrodynamic flow and capillary action. Hydrodynamic flow is initiated by an external compression force, usually as a result of a clamp or a press employed to mate the wood surface to be bonded. Flow then proceeds into the interconnected network of lumens and pits, with flow moving primarily in the path of least resistance.

There are many techniques that have been successfully employed to study adhesive penetration in wood, such as transmitted end reflected microscopy, fluorescence microscopy, scanning electron microscopy (SEM) and transmission electron microscopy (TEM). Fluorescence microscopy was found superior to other optical techniques for applications where there is poor color contrast in the interphase region (Furuno, Saiki, 1988, Brady, Kamke, 1988). An epi-fluorescence microscope is equipped with a three-component optical filter set. A high intensity, broadbend light source produces a wide spectrum of wavelength (Kamke, Lee, 2007).

Based on literature review, the lack of more exhausting research in the penetration of urea-formaldehyde adhesives in wood tissue is evident. It is particularly true having in mind many variable factors mentioned above. Since urea-formaldehyde (UF) glue resins were the most important type of adhesives in the wood industry in the last 60 years, especially for the production of wood based panels, gluing of furniture elements and veneering (Dunky, 2000), the objective of this research was microscopic detection of urea-formaldehyde adhesive penetration in wood tissue.

2. EXPERIMENTAL

2.1. Materials

a) Samples and preparation of microtome sections

Tangentially cut 5 *mm* thick beech wood (*Fagus moesiaca*) plies, 100 *mm* long (parallel to grain) and 30 *mm* wide, were prepared for radial penetration. Before bonding, the wood plies were conditioned in standard climate conditions (t=20±2°*C* and φ=65±5%) and their moisture contents (MC) were 10%. The urea-formaldehyde (UF) adhesive was applied to the surface of one ply. Two plies (with and without adhesive) were assembled into sample so that the grains of two plies were parallel. Samples were pressed in a hydraulic press at 120°*C* and 0,7 *MPa* for 15 *min*.

After hot pressing, bonded samples were conditioned again in standard climate conditions. Three microtome test-specimens from different locations of each sample were prepared. Several 20 µ*m* thick microtome sections were cut of each test-specimen, by sliding microtome apparatus, exposing a bondline with a cross-sectional surface.

b) Urea-formaldehyde (UF) resins

Four types of UF resins with different levels of polycondensation produced in Dukol - Ostrava, Czech Republic, were used. Levels of polycondensation were in progress from resin UF I (the lowest) to UF III (the highest). The resin UF IV had an equal degree of polycondensation as resin UF III, but urea was added in the end of production UF IV in order to obtain commercial adhesive in E1 class. The resins UF I, UF II and UF III had a molar ratio of formaldehyde (Fd) vs. urea (U) 2.0, while the resin UF IV had this Fd vs U ratio of 1.45.

These UF resins were prepared by addition of 10% wheat flour and 0,05% safranin. The addition of ammonium sulphate catalyser for UF I and UF IV was 0,5% and for UF II and UF III was 0,3%. All ingredients were added on dry weight base.

Table 1. The characteristics of resins and prepared adhesives UF I, UF II, UF III and UF IV
Табела 1. Својства куповних и припремљених везива UF I, UF II, UF III и UF IV

<table>
<tr><td colspan="3" rowspan="3"></td><td colspan="3">Property
Својство</td></tr>
<tr><td>Dry matter
Садржај суве супстанце</td><td>Brookfield viscosity
Вискозитет по Брукфилду</td><td>Gel time
Време желирања</td></tr>
<tr><td>%</td><td>mPa</td><td>s</td></tr>
<tr><td colspan="3">Serbian standard
SRPS</td><td>JUS H.K8.023</td><td>JUS H.K8.022</td><td>JUS H.K8.025</td></tr>
<tr><td rowspan="8">Value
Вредност</td><td rowspan="2">UF I</td><td>Resin
Куповно везиво</td><td>53.66</td><td>218</td><td>58</td></tr>
<tr><td>Prepared adhesives
Припремљено везиво</td><td>54.43</td><td>545</td><td>59</td></tr>
<tr><td rowspan="2">UF II</td><td>Resin
Куповно везиво</td><td>53.65</td><td>281</td><td>59</td></tr>
<tr><td>Prepared adhesives
Припремљено везиво</td><td>54.32</td><td>745</td><td>60</td></tr>
<tr><td rowspan="2">UF III</td><td>Resin
Куповно везиво</td><td>53.83</td><td>555</td><td>58</td></tr>
<tr><td>Prepared adhesives
Припремљено везиво</td><td>54.56</td><td>1644</td><td>59</td></tr>
<tr><td rowspan="2">UF IV</td><td>Resin
Куповно везиво</td><td>65.65</td><td>2052</td><td>59</td></tr>
<tr><td>Prepared adhesives
Припремљено везиво</td><td>54.79</td><td>460</td><td>61</td></tr>
</table>

The same dry content and gel time of prepared adhesives, were essential for the purpose of clear understanding and comparison of their penetration into wood tissue.

Therefore, the gel time was adjusted by addition of different quantity of catalyser, while dry content was adjusted by addition of water (into UF IV only).

Prepared UF adhesives were applied on the surface of one ply at a loading level of 200 $g \cdot m^{-2}$.

Table 1 shows the characteristics of both UF resins and prepared adhesives.

2.2 Methods

The interphase region of the adhesive bond is defined as the volume containing both wood cells and adhesive (Brady, Kamke, 1988). The size of the interphase region is determined by the depth of penetration of the adhesive. The average penetration (AP) was calculated as the mean value of penetration depth measured on the 25 positions along 1400 μ*m* of bondline for each microtome slide section. The filled interphase region (FIR) was expressed as the percentage of the ratio of the area covered by adhesive vs. total interphase region (including the unfilled lumen area).

Epi-fluorescence microscope (LEICA DM LS) was used in this research for measuring the adhesive penetration. The optical filter set used consisted of a 450 *nm* excitation filter, 510 *nm* dichromatic mirror and a 515 *nm* emission filter. The image analysis system included a colour video camera (LEICA DC 300), a Pentium personal computer, image processing and analysis software (IM1000) and a LG high resolution image monitor.

2. RESULTS AND DISCUSSION

In this research dry matter and gel time of prepared adhesives, as well as loading level and press factors (press time, temperature, and pressure) were constant.

The average depth of radial penetration as a function of different degree of polycondensation of UF adhesives was presented in Figure 1.

Degree of polycondensation increases from UF I (the lowest) to UF III (the highest). Indirect measure for degree of polycondensation was shown as a Brookfield viscosity (*mPa*) in Table 1. Viscosity of prepared adhesives rises from 545 *mPa* UF I, to 745 *mPa* UF II, and successively to 1644 *mPa* UF III. The higher the degree of polycondensation, the larger the molecules of prepared polymer adhesive. Hence, the penetration depends on the ratio of an average anatomical vessel (lumen) diameters vs. the average size of polymer molecules. For the purpose of gross penetration measurement, depth of penetration might be correlated to the viscosity. It is evident from Fig. 1 that depth of penetration decreases with the raise of viscosity i.e. with the raise of degree of polycondensation and molecular size. However, prepared adhesive UF IV was diluted having the lowest viscosity of 460 *mPa* and consequently high penetration depth again. This adhesive thus does not follow the decreasing trend.

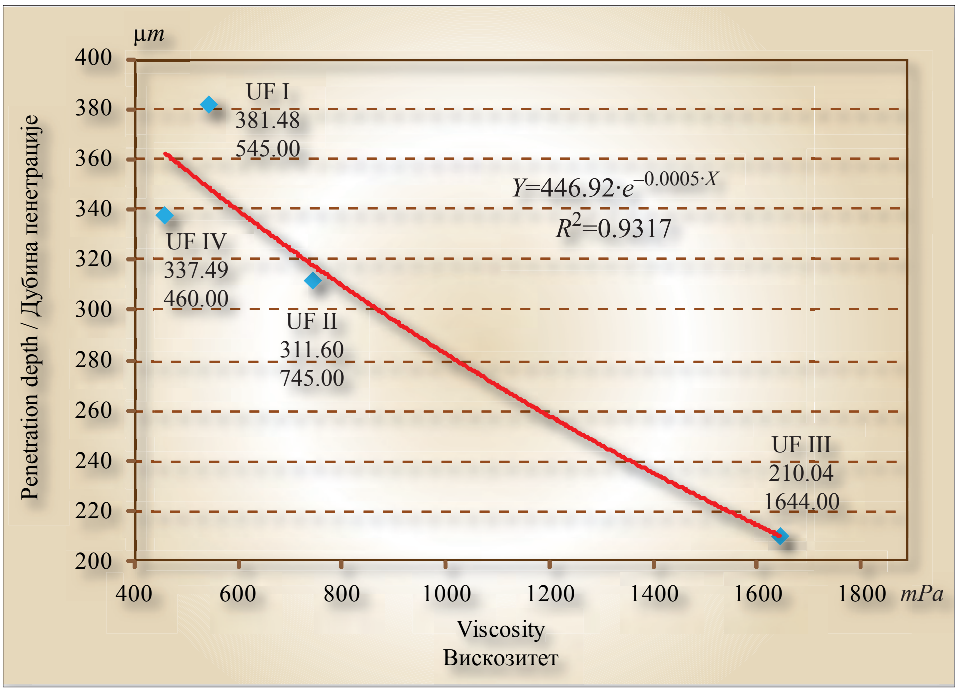

Figure 1. The average penetration depth as a function of viscosity (different degree of polycondensation) of UF adhesives

Слика 1. Просечна дубина пенетрације у зависности од вискозитета (различитог степена поликондензације) УФ-адхезива

The photomicrographs (Figure 2 and Figure 3) confirmed the mentioned statements. Thereafter, it might be observed that the adhesive fills the lumens of vessels. The percentage of vessels at the bondline, directly influences the depth of penetration. Since, beech is a diffuse porous species, the distribution of vessels was uniform within annual rings. It was noticed that penetration of adhesive into ray cells is omitted, presumably because of presented cells content in their lumens. The effect of adhesive penetration could be expressed by partly filled or fully filled anatomical vessels of wood tissues in interphase region.

Mechanical elements of anatomic structure are not convenient for the penetration of adhesives. This is probably because they have thick cell walls and small lumens. The narrow pits contribute to this effect.

The percentage filled interphase region as a function of different degree of polycondensation of UF adhesives was illustrated in Figure 4.

The filled interphase region, as well as the depth of penetration, correlated to the viscosity. It is evident from Fig. 4 that filled interphase region decreases with the raise of viscosity, i.e. with the raise of the degree of polycondensation and molecular size. On the

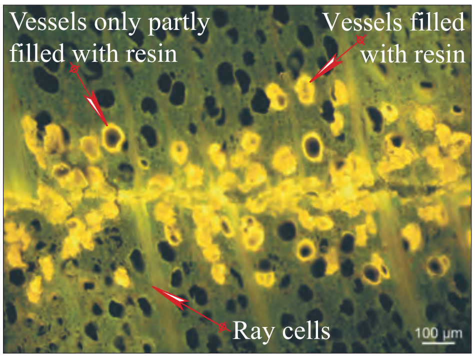

Figure 2. Photomicrograph of UF I with safranin bondline, using epi-fluorescence

Слика 2. Микрофотографија линије лепљења UF I са сафранином, помоћу епи-флуоресцентног микроскопа

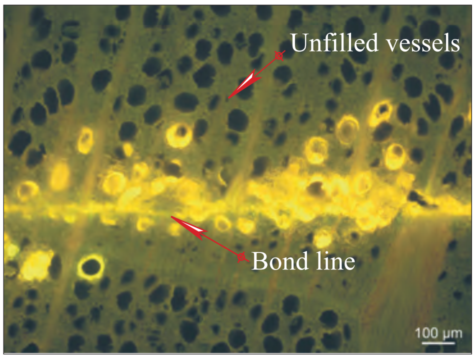

Figure 3. Photomicrograph of UF II with Safranin bondline, using epi-fluorescence

Слика 3. Микрофотографија линије лепљења UF II са сафранином, помоћу епи-флуоресцентног микроскопа

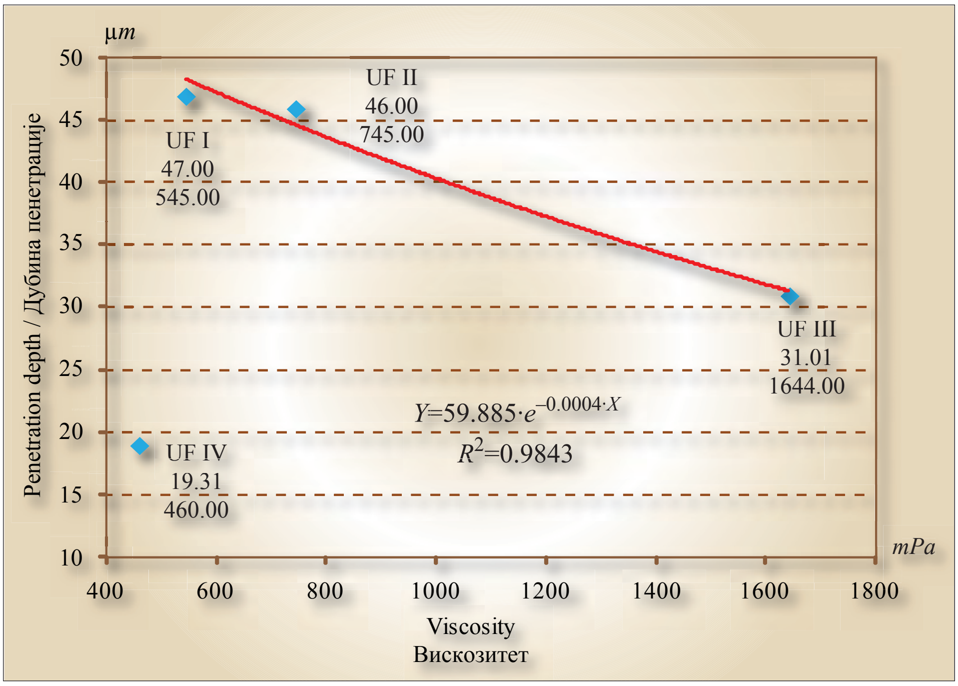

Figure 4. The percentage filled interphase region as a function of viscosity (different degree of polycondensation) of UF adhesives

Слика 4. Степен испуњености зоне лепљења у зависности од вискозитета (различитог степена поликондензације) УФ-адхезива contrary of dept of penetration, in the case of prepared adhesive UF IV, filled interphase region follows the decreasing trend. This is probably because there were filled vessels with adhesive far from bondline, which enlarged the interphase region. It was noticed during the measurement of penetration depth of prepared UF IV adhesive.

4. CONCLUSION

Depth of adhesive penetration is very variable in beech wood tissue. The effect of adhesive penetration could be expressed by partly filled or full filled anatomic vessels in interphase region. For instance, adhesive penetration into lumens of ray cells was inhibited by their cell content. Thus, filled interphase region gives an additional information on penetration character.

Depth of radial penetration of prepared urea-formaldehyde adhesives of different viscosity in the beech wood tissue, decreases with raising adhesive viscosity (degree of polycondensation). For each viscosity raise of 10 *mPa*, the depth of penetration decreases averagely for 1,54 μ*m*. Similarly, the percentage of filled interphase region mostly follows the depth of penetration. For each viscosity raise of 10 *mPa*, the percentage of filled interphase region decreases averagely by 0,156%.

The above mentioned relations, as well as the new relations, will be the objective of further research and evaluation.

Epi-fluorescence microscope supplied with appropriate computer program for calculation of depth of penetration and percentage of filled interphase region, presents a proper technique for the diagnostic of phenomenon of adhesive penetration in this research.

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ПЕНЕТРАЦИЈА УРЕАФОРМАЛДЕХИДНОГ АДХЕЗИВА У ТКИВО ДРВЕТА ДЕО I: РАДИЈАЛНА ПЕНЕТРАЦИЈА УФ-АДХЕЗИВА У ДРВО БУКВЕ

Резиме

Дубина радијалне пенетрације припремљених уреа-формалдехидних адхезива различитих вискозитета у ткиво буковог дрвета опада са порастом вискозитета (степена поликондензације). Пенетрација зависи од односа просечних пречника трахеја и просечне величине молекула полимера.

Ефекат пенетрације адхезива може се изразити делимично и потпуно испуњеним трахејама дрвног ткива у зони лепљења. Ћелијски садржај у луменима трака лигнума спречава пенетрацију адхезива у њих. Епи-флуоресцентна микроскопија је одговарајућа техника за дијагностику феномена пенетрације адхезива.

Дубина пенетрације адхезива је веома варијабилна код дрвног ткива букве, па испуњеност зоне лепљења даје додатне неопходне информације за оцену пенетрације. На основу резултата из овог рада, може се закључити да испуњеност зоне лепљења опада са порастом вискозитета адхезива.

Степен испуњености зоне лепљења углавном прати дубину пенетрације.