

Acryl Resin Distribution In Lime Tree Wood Determined By ^{241}Am Analyser Of Density Profiles

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ABSTRACT: The aim of this study was to determine retention and distribution of the polybutylmethacrylate resin Solakryl BT-55 in conserved lime tree wood (*Tilia cordata* Mill.) using conventional method and gamma ray method working with radioisotope ^{241}Am . Conservation of sound lime tree samples 60 x 60 x 60 mm with known densities and density profiles in all three anatomical directions was performed with a 27.5 % toluene acrylic solution (Solakryl BT-55 diluted with toluene in a ratio of 1:1) in autoclave at 20 °C and 0.8 MPa for 5, 30 or 180 minutes.

Total retentions of the acryl resin increased nonlinearly with prolongation of the impregnation process when densities in the oven dry state of conserved samples increased about 20.5 % (5 min), 25.5 % (30 min) or 30 % (180 min). The gamma ray distribution analyses of the acryl resin showed that lime tree wood had the best impregnability in the longitudinal direction. Differential retentions of the solid acrylic resin into samples varied from 0.15 to 0.20 g/cm³ at distance of 5 mm from the axial surfaces, from 0.05 to 0.13 g/cm³ at distance of 20 mm from the axial surfaces, and only from 0.02 to 0.11 g/cm³ in the centre of samples (at distance of 30 mm). Penetrations of the acryl resin in the radial and tangential directions were negligible, unlike the longitudinal direction.

Keywords: acryl resin, distribution, lime tree, radioisotope ^{241}Am , retention

I. INTRODUCTION

Generally, for improving the esthetical, physical and mechanical properties of wood and wooden artefacts are used conservation agents. Efficiency of various conservation processes with natural and synthetic agents depends on more factors related to (1) wood structure, e.g. species, permeability, moisture, geometry, range and degree of damaging by bacteria, fungi, insect, etc., (2) macro- and micro-distribution of the conservation agents in wood which depends on their properties and also on technological parameters of the conservation process, e.g. pressure and time, (3) physical and chemical properties of conservation agents, e.g. viscosity, surface tension, polarity, molecular weight, possibility for reactions with -OH groups of cellulose, etc. [1,2].

Distribution of acryl and other conservation agents in wood can be determined by destructive methods, e.g. on small drilled samples, cross-sections and micro-slides by a scanning electron microscope – SEM [3], and also by various non-destructive methods, e.g. by acoustic, thermographic, nuclear magnetic resonance or radiographic [4, 5, 6].

At analysis the penetration, retention and distribution of conservation agents in wooden objects, similar to estimation of moisture, adhesive, mechanical

behavior, growth rings, knots, or defects caused by fungi and insect, The following stationary or mobile radiographic methods are usually applied [4, 7, 8, 9, 10, 11, 12, 13, 14, 15]:

- Conventional X-ray radiography – one line scanning,
- Gamma rays densitometry – one line scanning,
- X-ray computer tomography scanning (2D and 3D, e.g. SRXTM),
- γ -ray computer tomography scanning (2D and 3D),
- scanning neutron radiography.

The aim of this work was to evaluate the acryl resin “Solakryl BT-55” retention and distribution in lime tree wood in the longitudinal, radial and tangential directions after its pressure impregnation, using one line scanning gamma rays densitometry – the analyzer of density profiles. The density profiles of lime tree samples were monitored before and after conservation and from these profiles the acryl resin amount in defined distances from the external surfaces of conserved samples were computed. Used analyzer of density profiles is based on the measurement of the absorption coefficient attenuate of the gamma-radiation from the isotope ^{241}Am passing through tested material. More other researches preferred this isotope at testing the density and quality of wood and wooden composites [16, 17, 18, etc.].

II. MATERIAL AND METHODS

2.1 Lime tree wood

Samples, 60 mm x 60 mm x 60 mm (longitudinal x radial x tangential), were prepared from one lime tree (*Tilia cordata* Mill.) sound board 1400 x 400 x 100 mm which was previously kiln dried and conditioned on approximately 12 % moisture content. Natural samples without knots or other inhomogeneities were then selected for the experiment. In the oven dry state were determined their total densities “ ρ_N (g/cm³)” and density profiles in three anatomical directions “ $\rho_{N\text{-profile}}$ (g/cm³)”.

2.2 Acryl resin

For the lime tree samples conservation was used commercial product Solakryl BT-55, which contains 55 % of the polybutylmethacrylate (PBMA) and it is manufactured in the Lučební závody, Draslovka a.s. Kolín, Czech Republic. In the experiment was used a 27.5 % solution of the acryl resin. It means the Solakryl BT-55 was diluted with toluene in a weight ratio 1:1 (w/w). This resin is convenient for restoration works, e.g. for consolidation of wooden artefacts. Its basic chemical-physical properties by the technical sheet PND 47-701-93 are as follows: $M_w = 40.103$ g/mol; $\rho = 900$ kg/m³; $T_g = 45$ °C.

2.3 Conservation process

Conservation of the lime tree samples with Solakryl BT-55 was performed in the Dreyer-Holand-

Merten KG device by the pressure-vacuum Lowry technique at a temperature of 20 °C, using pressure of 0.8 MPa, lasting for 5, 30, or 180 minutes. Conserved samples were conditioned 4 weeks at a relative humidity of 50 % and a temperature of 20 °C, then dried in a kiln at 103 ± 2 °C into the oven dry state, and finally re-analyzed their total densities " ρ_C (g/cm^3)" and density profiles in three anatomical directions " $\rho_{C\text{-profile}}$ (g/cm^3)". Volumes of samples remained the same during and after conservation, so the acryl resin was only in lumina and was not able to penetrate into cell walls of wood. It was confirmed by microscopic analyses, as well.

2.4 Total retention of acryl resin

Total retentions " R_S (g/cm^3)", i.e. total solid mass retentions of the acryl resin Solakryl BT-55, were calculated as difference between densities of the lime tree samples in their conserved " ρ_C (g/cm^3)" and natural " ρ_N (g/cm^3)" state by equation 1:

$$R_S = \rho_C - \rho_N \quad (1)$$

2.5 Assessment of acryl resin distribution in wood by the analyzer of density profiles

Density profiles of the natural and conserved lime tree samples were determined again in their oven dry state. Apparatus constructed in laboratories of the Faculty of Wood Sciences and Technology, Technical University in Zvolen was used for these analyses (Fig. 1). Source of the gamma radiation was a low-energy radiant AMG 50. It works with the radioisotope ^{241}Am having the energy of 59.5 keV, and its output power is 2.0 GBe.

During measurements, the tested samples were imbedded in initial position on the moving electrical car, then the cover of the emitter was removed and the beam of gamma-rays distributed from the radiant AMG 50 was transmitted through the samples gradually in their all three anatomical directions – longitudinal, radial and tangential. The shift of the samples was always after each 0.2 mm. The analyzer scanned the density profiles of respective samples in one plane (Fig. 1). The intensity of the gamma-radiation after crossing the slots is evaluated by the NaJ (TI) detector which is attached to the single-channel spectrum analyzer IH 10 made by the firm STADOS Prague, Czech Republic. This analyzer is furnished with IMS2 interface that makes it possible to control measure with personal computer [19]. Measurements of the density profiles were carried out in the same position of samples, i.e. in the natural " $\rho_{N\text{-profile}}$ (g/cm^3)" and then in the conserved " $\rho_{C\text{-profile}}$ (g/cm^3)" state, what was secured by small cuts on their edges (Fig. 1).

The differential retentions of the acryl resin Solakryl-BT55 into lime tree samples – test pieces " $R_{S\text{-profile}}$ (g/cm^3)" were calculated separately for their three anatomical directions (as difference between density profile of the conserved and natural sample) in the same plane for defined distances from their outer surface by equation 2:

$$R_{S\text{-profile}} = \rho_{C\text{-profile}} - \rho_{N\text{-profile}} \quad (2)$$

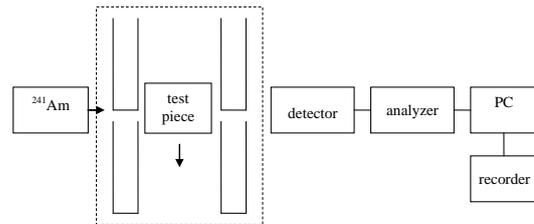


Fig. 1: Working flow diagram of the analyzer of density profiles - densities evaluated by equation 3

$$I = I_0 \cdot e^{-C \cdot \rho \cdot d} \quad (3)$$

where: " I_0 " is the intensity of the incident gamma beam, " I " is the intensity of the gamma beam transmitted through test piece (lime tree sample), " C " is the gamma-beam attenuation coefficient of the test piece, " ρ " is the density of the test piece and " d " is the thickness of the test piece in the direction of gamma beam.

III. RESULTS AND DISCUSSION

Total retentions of the acryl resin Solakryl BT-55 (R_S) into lime tree samples are listed in the Table 1.

Tab. 1: Total retentions of the solid part of acryl resin "Solakryl BT-55" (R_S) into lime tree samples

Time of 0.8 MPA in autoclave	5 min	30 min	180 min
Density of natural samples - ρ_N ($\text{g}\cdot\text{cm}^{-3}$)	0.470 (0.017)	0.468 (0.012)	0.467 (0.013)
Density of conserved samples - ρ_C ($\text{g}\cdot\text{cm}^{-3}$)	0.566 (0.024)	0.587 (0.022)	0.607 (0.021)
Total retention of acryl - R_S ($\text{g}\cdot\text{cm}^{-3}$)	0.096 (0.030)	0.119 (0.023)	0.140 (0.019)

Notes:

- Mean values are always from 6 replicates (5 min → No: A11-A16; 30 min → No: B21-26; 180 min → No: C31-C36)
- Values in the parentheses are the standard deviations

Differential retentions related to a solid part of the acryl resin Solakryl BT-55 ($R_{S\text{-profile}}$) into lime tree samples are presented in the Figures 2 and 3. For illustration are shown results only from one replicate selected from six replicates tested at a given impregnation time, i.e. replicates with numbers A-12, B-22 and C-31.

From the total retentions of the acryl resin into lime tree samples established after 5, 30 or 180 minutes of the pressure impregnation process (Table 1), and also from the differential retentions of the acryl resin rated in three anatomical directions of wood (Figs. 2 and 3), it is evident that the penetration kinetic of the conservation agent in wood structure had a non-linear character. Relatively the largest retentions of the acryl resin into lime tree wood were achieved during the first 5 minutes of impregnation while then retentions grew only moderately (Figs. 2 and 3). This result complies with knowledge of [20] and more other researches [e.g. 21] impregnated beech, maple, ash, pine and spruce samples of different dimensions from 15 to 150 minutes) under which the dependence between retention and impregnation time can be expressed by exponential, logarithmic or other non-linear equations.

The distribution analyses confirmed that the best impregnability of lime tree wood with the acryl resin was achieved in its longitudinal direction. This result complies also with a generic penetration model of adhesives for hardwoods proposed by [22] in which the vessel network is dominated. In our experiment, i.e. after pressure impregnation lasting from 5 to 180 minutes, the differential retention of the acryl macromolecules in the longitudinal direction at distance of 5 mm from the axial surfaces of samples varied usually from 0.15 to 0.20 g/cm³, at distance of 20 mm from 0.05 to 0.13 g/cm³, or in their centre – at distance of 30 mm only from 0.02 to 0.11 g/cm³ (Fig. 3).

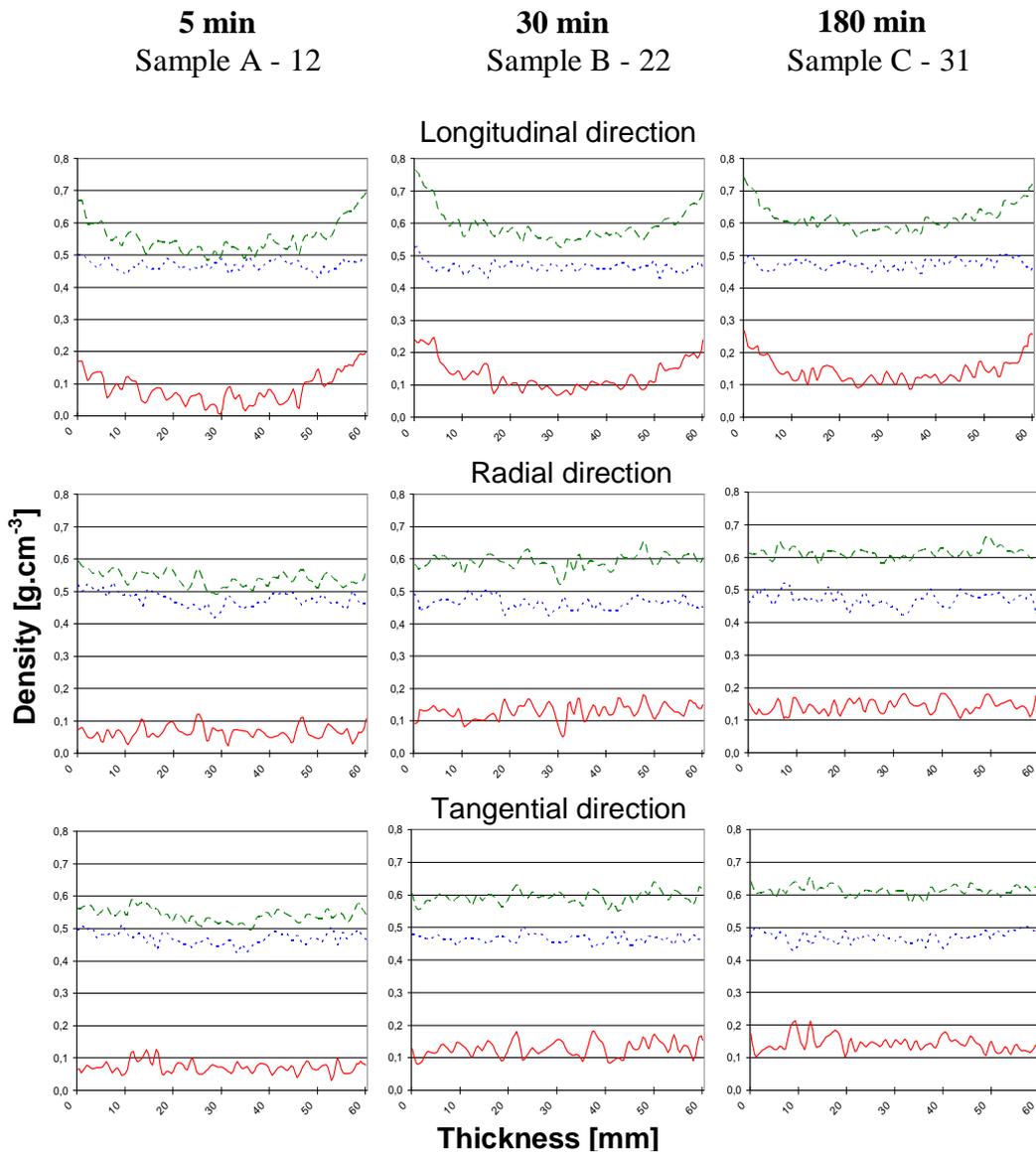


Fig. 2: Density profiles of the natural ($\rho_{N-profile}$ ) and conserved ($\rho_{C-profile}$ - - -) lime tree samples 60x60x60mm, and differential retentions of the acryl resin Solakryl BT 55 ($R_{S-profile}$ —)

Differential retentions of the acryl resin in the radial and tangential directions were negligible. It was indirectly found by almost the same differential retentions which were assessed in the radial and tangential directions of samples from various distances of their outer surfaces (Fig. 3). So, toluene solutions of the acryl resin penetrated into lime tree samples preferentially (or only) in the longitudinal direction (Fig. 3).

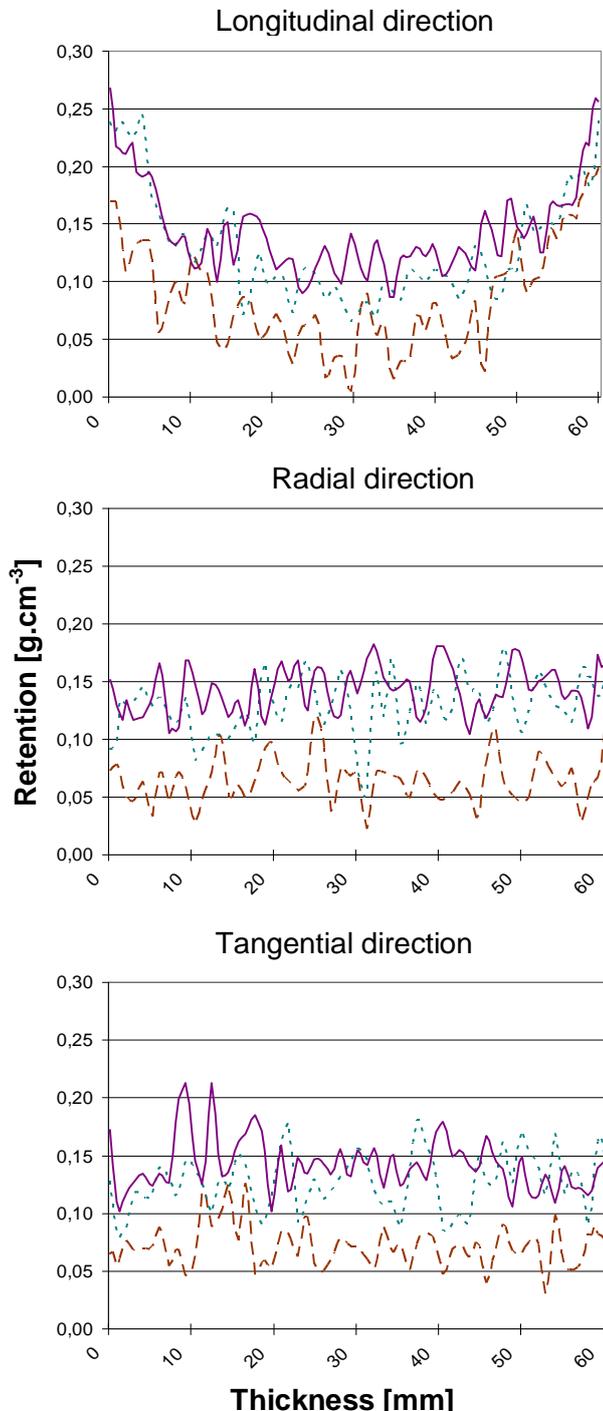


Fig. 3: Differential retentions of the acryl resin Solakryl BT-55 ($R_{S-profile}$) into lime tree samples 60x60x60 mm (see Fig. 2) after 5 minutes ---, 30 minutes or 180 — minutes of the pressure impregnation processes

IV. CONCLUSIONS

The achieved results give the opportunity to do the following conclusions:

- The gamma ray densitometry working with the isotope ^{241}Am is a suitable method for distribution analyses of solid conservation substances, e.g. the acryl resin Solakryl BT-55, in wood.
- Clearly the highest penetration of the acryl resin into lime tree wood occurred from its axial surfaces in the longitudinal direction.
- The most significant penetration and retention of the acryl resin into lime tree wood was measured at beginning of the pressure impregnation processes, and this is in compliance with exponential behavior of transport processes in wood.

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REFERENCES

- [1] L. Reinprecht, Rekonštrukcia objektov z dreva "Reconstruction of wooden structures" (TU Zvolen – Slovakia, 1998).
- [2] C. A. S. Hill, Wood modification – chemical, thermal and other processes (John Wiley & Sons Ltd, Chichester – UK, 2006).
- [3] L. Reinprecht, and I. Makovíny, Hardening of aminoplasts in modified wood with catalytic and thermic dielectric heating, Modyfikacja drewna, 1987, 288-298.
- [4] A. Unger, A. P. Schniewind, and W. Unger, Conservation of wood artifacts (Springer-Verlag Berlin Heidelberg, 2001).
- [5] E. Lehmann, S. Hartmann, and P. Wyer, Neutron radiography as visualization and quantification method for conservation measures of wood firmness enhancement, Nuclear Instruments and Methods in Physics Research – Section A, 2005, 542: 87-94.
- [6] I. Kučerová, B. Schillinger, E. Calzada, and E. Lehmann, Monitoring transport of acrylate consolidants through wood by neutron radiography, Wood Science for Conservation of Cultural Heritage, 2009, 80-85.
- [7] V. Bucur, S. Garros, A. Navarrete, M.T. de Troya, and R. Guyonnet, Kinetics of wood degradation by fungi with X-ray microdensitometric technique, 10th International Symposium on Nondestructive Testing of Wood, Lausanne – Switzerland, 1996, 209-215.
- [8] I. Kučerová, and J. Lisý, Sledování pruniku impregnační látky do dřeva metodou počítačové tomografie "Study of the penetration ability of consolidating agent into wood by using the X-ray computer tomography" Rekonštrukcia a konzervácia historického dreva '99, TU Zvolen – Slovakia, 1999, 167-170.
- [9] L. Reinprecht, H. Novotná, and V. Štefka, Density profiles of spruce wood changed by brown-rot and white-rot fungi, Wood Research, 52(4), 2007, 17-28.

- [10] D. Mannes, L. Josic, E. Lehmann, and P. Niemz, Neutron attenuation coefficients for non-invasive quantification of wood properties, *Holzforschung*, 63, 2009, 472-478.
- [11] D. Mannes, F. Marone, E. Lehmann, M. Stampononi, and P. Niemz, Application areas of synchrotron radiation tomographic microscopy for wood research, *Wood Sci. Technol*, 44, 2010, 67-84.
- [12] J.Y. Buffiere, E. Maire, J. Adrien, J.P. Masse, and E. Boller, In situ experiments with X ray tool for experimental mechanics, *Experimental mechanics*, 50, 2010, 289-305.
- [13] F. Forsberg, M. Sjødahl, R. Mooser, E. Hack, and P. Wyss, Full three-dimensional measurements on wood exposed to three-point bending: Analysis by use of digital volume correlation applied to synchrotron radiation micro-computed tomography image data, *Strain*, 46, 2010, 47-60.
- [14] E.H. Lehmann, D. Mannes, P. Scherrer, K. Hunger, M. Wörle, S. Braovac, H. Kutzke, and M. Christensen, Wood investigations by means of radiation transmission techniques in the analysis of cultural heritage objects of different size scale, Joint final conference of COST Actions IE0601 and MP0601 - Wood science and conservation: on-going work and challenges for the future, Paris - France, 2011, 53-54.
- [15] P. Hass, F.K. Wittel, M. Mendoza, H.J. Herrmann, and P. Niemz, Adhesive penetration in beech wood: experiments, *Wood Sci. Technol.*, 46, 2012, 243-256.
- [16] F. Divós, S. Szegedi, and P. Raics, Local densitometry of wood by gamma batch-scattering, *Holz als Roh- und Werkstoff*, 54 (4), 1996, 279-281.
- [17] M. Tiitta, H. Olkkonen, T. Lappalainen, and T. Kanko, Density measurement of particleboard, veneer and wood specimens by narrow beam gamma absorption technique, *Nondestructive Testing of Wood*, Lausanne - Switzerland, 1996, 187-200.
- [18] W. Dźbeński, P. Mańkowski, and S. Krzosek, Theoretical and practical usefulness of radiation methods for wood density testing, *Folia Forestalia Polonica – Series B*, 39, 2008, 31-43.
- [19] V. Bahýl, Analyzátor hustotného profilu aglomerovaných materiálov “Analyzer of density profiles of composite materials”, *Drevo*, 47(2), 1992, 48-49.
- [20] J.F. Siau, *Transport processes in wood* (Springer Verlag, New York, 1984).
- [21] L. Reinprecht, and D. Horský, Impregnability of tree species as a function of their shape, Latest achievements in research of structure and physics, IUFRO Conference, TU Zvolen – Slovakia, 1990, 163-177.
- [22] M. Mendoza, P. Hass, F.K. Wittel, P. Niemz, and H.J. Herrmann, Adhesive penetration of hardwood: a generic penetration model, *Wood Sci. Technol.*, 46, 2012, 529-549.