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THE EFFECT OF POLYMETHYL METHACRYLATE IMPREGNATION ON MECHANICAL PROPERTIES OF PINE WOOD DEGRADED BY FUNGI

This paper presents results of investigations into possibility of using polymethyl methacrylate to reinforce wood degraded by Phellinus pini (Thore) Fr. fungus. It was found that impregnation of wood with the resin causes an increase in mechanical properties. Nevertheless this increase is relatively greater for wood degraded by biological agents than for undamaged wood. The behaviour of wood-resin system after artificial ageing in laboratory conditions was observed. The strength of wood treated with the resin was lower after artificial ageing than the strength of wood without ageing. Similar tendencies of changes were observed both in the case of wood degraded by biological agents and in the case of undamaged wood.

Keywords: synthetic resin, degraded wood, reinforced wood, wood impregnation with synthetic resin

Introduction

In practice of monument conservation nearly always there is a necessity to choose the most proper (optimal) method for treatment of damaged elements bearing significant historic values. The accepted conservation method should ensure a lasting effect. One has to keep in mind that monuments/antique objects, despite permanent care of the conservation service, are continuously exposed to damaging factors both biotic and abiotic and that leads to gradual deterioration of technical condition of these objects [Czajnik 1970]. A number of museums are faced with the necessity to increase their effectiveness in the field of wood protection, including also repairs of elements damaged by biotic agents. Too often the desire to restore the original technical efficiency of wooden construc-

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tion results in complete change of the original damaged elements and replacement of them by new ones. In many cases such attempt is contradictory to the need to keep the monuments in their original untouched shape. Hence there is a need for continuous search for new methods of saving the original elements in wooden monuments. Although one should not forget the verification of already existing, proven solutions capability to cope with specific problems under different environmental conditions, with special attention given to lasting character of the effects obtained.

Analysing project materials and carrying out field observations Krajewski [1995] was curious about the general construction problems resulting from mistakes made in design and executive stages. The mistakes result in establishment of conditions favourable to the expression of fungi which in turn may lead to establishment of an environment favouring corrosion of wooden elements in time. The weakened construction elements may then need mechanical support. According to Jankowski [2004], except biological corrosion, another frequent reason for wood damage are natural physical processes leading to cracks and deformations of wooden elements, thus to decreased strength of the whole construction. Moreover, any alteration of the way and range of the object use, such as reconstruction or development, also may result in an increased burden of the internal tensions in construction elements, thus lead to deflections or even damage of the elements.

The strengthening of characteristics of wood in weakened structures is a difficult and disputable task, particularly so when it concerns monument objects. One method of wooden monument conservation is to impregnate the wood elements subject to deterioration with synthetic resins. To optimise the effect of conservation it is necessary to use developed conservation agents that successfully passed lots of tests before they were recommended for practical (commercial) use in objects of great cultural and/or historic value. In the last few decades many authors presented results of their studies on the implementation of synthetic resins to wood strengthening. Czajnik [1968] investigated the effect of wood impregnation with impregnates containing resins, i.e. polyacetatevinyl, acrylic, epoxide and carbamide resins. He took into consideration the influence of resin on selected physical parameters of wood and on wood resistance to the infection by wood destroying fungi Coniophora cerebella (Pers.) Pers. and Polysticus versicolor (L. ex Fr.) Fr. After impregnation with resins wood attributes changed in the following way: bending strength and wood strength in the radial and tangential direction increased relatively the most significantly, whilst wood hygroscopicity, absorptiveness, and swelling decreased. Another effect observed in the reported study was the improved resistance to fungi. These results fitted well the earlier study results of other researchers [Domasłowski 1960, 1961] and the general conclusions were in line with the results of studies carried out with the use of different resins. The positive results of the above

reported studies were the well-grounded basis to give an impulse to implement epoxide and carbamide resins, and also, but to a smaller degree, polyvinyl carbamide and polymethyl methacrylate for the protection of wood against wood destroying fungi. The above listed resins were appreciated as applicable in conservation of wood damaged by fungi for wood impregnated with them was characterised by parameters similar to those of healthy wood.

Buksalewicz et al. [1987] defined the range of requirements that proper resin solutions need to fulfil. These requirements also presented the state-of-the-art in the field of research on penetration and spatial distribution of resin inside wood emphasizing the advantages of wood strengthening acrylic impregnate as modified with the use of pentachlorophenol (Petrifo). Paciorek [1993] reviewed in detail the literature in the area of resin applicability for wood strengthening using the example of an old sculpture destroyed by insects. He used the technique of computer tomography to see the resin distribution in the impregnated object. The author confirmed the effectiveness of implementation of the vacuum method to impregnation of lime wood, as well as the necessity of using proper mixture of solvents to avoid consequences of the reverse migration of resins. The following substances were among tested thermoplastic materials soluble in toluene: Winacet R-50, Osolan KL, and Paraloid B-72. Paraloid B-72 resin proved to have been definitely more resistant to damaging activity of microorganisms than any other tested product, and at the same time it was 100% resistant to colour change of impregnated wood.

Wieczorek [1992] listed and described all commercial products available on the Polish market till late 1980s and aimed at strengthening wood structure in monuments together with practical recipes and conditions of product use.

Schneider [1994] reviewed industrial methods of obtaining wood composites with addition of polymers (WPC). He also described methods for conservation of destroyed objects with the use of phenyl-formaldehyde resins and vinyl resins

The papers of Żaboklicki [1988] and Wiśniewski [1997] are also interesting items in the field of wooden construction element strengthening. Both authors described methods of structural wood strengthening and amended the wide literature review and characteristics of the applied resins: phenol-formaldehyde and epoxide. A synthetic resin solution applied on the surface of wood creates protective coat or decorative coat, whilst the same medium in the form of low percent solution (called an impregnate) applied to deeper regions of wood and then hardened actively changes a number of wood properties, first of all increasing wood resistance to mechanical factors.

There is a wide spectrum of materials useful for hardening of wood in antique objects [Soldenhoff 2004]. To achieve this synthetic resins are used. These resins, according to the unanimous opinion of many researchers, are all

characterised with advantageous features [Stamm, Seborg 1936; Nakhla 1986; Buksalewicz et al., 1987; Wieczorek 1992]:

- they have an increased ability to penetrate deep inside the wood,
- they increase mechanical strength of wood at minimum amounts of the resin used,
- they are resistant to both biotic and abiotic damaging factors,
- they do not cause any discoloration of impregnated wood, an attribute particularly important in the case of conservation of antique objects,
- they have no destructive effect on treated wood,
- water does not wash them off the wood which is an especially advantageous attribute if the objects are to be directly exposed to outdoor conditions,
- technologies of impregnation with resins and hardening them create no significant extra cost and fulfil current strict requirements of the environment protection.

One of very important decision-making factors concerning the choice of proper resin to be used for wood conservation concerns factors which the conserved piece of wood is to be exposed to. Those resins which become fragile or are not flexible enough are to a higher degree exposed to and threatened by destruction caused by external factors like e.g. changing humidity (alternate moistening and drying off), changing ambient temperature, alternately occurring changes in the state of water aggregation (freezing and thawing), as well as the activity of solar radiation in u-band and ultraviolet. The use of a resin characterised with little elasticity results in occurrence of inner tensions in treated wood which directly leads to occurrence of cracks along the wood-resin borderlines, as well as weakening of the composite resistance [Soldenhoff 1976].

Material and methods

The wood used for the present study was extracted from a Scots pine log whose heartwood region was heavily damaged by *Phellinus pini* (Thore) Fr. fungus under natural conditions. This fungus causes so-called white heterogeneous decay of coniferous wood. The wood samples subject to further testing were taken from the heartwood part of the log containing wood with decay. The decay had no signs of loss of the woody tissue (fig. 1).

The selection of empirical wood material for the study was conditioned by observed in practice cases of using wood damaged by *Phellinus pini* for building construction purposes. As observed in preliminary tests, wood samples with white heterogeneous decay absorbed resin rather at a satisfactory rate which was the necessary precondition for the studies. Preparation of wood samples was in accordance with the general requirements of PN-77/D-04103 standard in the field of bending strength determination as well as with the requirements of

PN-79/D-04102 standard as regards testing for compression strength parallel to grain. The preliminarily selected samples were subjected to selection to eliminate all these samples with perceptible defects worsening homogeneity of the empirical material. All the samples of irregular shape, bearing sings of cracks or with knots were eliminated. Despite the selection, the remaining samples' density varied greatly from 356 to 526 kg/m³ at the absolute humidity level equal to about 12%.

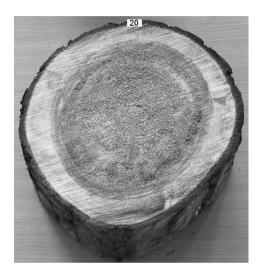


Fig. 1. Scots pine stem section with heartwood degraded by *Phellinus pini* (Thore) Fr.

Rys. 1. Wyrzynek pnia z rozkładem drewna twardzieli spowodowanym przez hubę sosnową

4 groups were established from the general collection of samples, each group containing 6 wooden pieces so that the wood parameters be possibly equal in every individual group. The following groups were selected in relation to their purpose:

- samples to be impregnated with resin, but not expected to be subjected to artificial ageing,
- samples to be impregnated with resin, expected to be subjected to artificial ageing,
- samples not to be impregnated with resin and not expected to be subjected to artificial ageing,
- samples not to be impregnated with resin, but expected to be subjected to artificial ageing.

A short description of tested wood samples' density is presented in table 1.

Table 1. Characteristic of density of tested wood samples
Tabela 1. Charakterystyka gęstości próbek drewna poddanych badaniu

		Wood density Gęstość drewna			Variation coefficient	Average variation	
Sample description Opis próbek			Min. Min.	Aver- age Średnia	Max. Maks.	Wsp. zmienności	coefficient Średni wsp. zmienności
			X_{min}	\overline{X}	X_{max}	υ	$\overline{\delta}$
				[kg/m ³]		[%	6]
	resin treated		356	429	493	14	
Degraded wood Drewno zdegradowane	nasycane żywicą	aged starzone	388	464	517	9	13
	untreated nienasycane żywicą	not aged niestarzone	366	459	526	14	13
		aged starzone	363	437	515	12	
	resin treated nasycane	not aged niestarzone	547	581	624	5	
Sound wood Drewno "zdrowe"	żywicą	aged starzone	512	556	603	5	5
	untreated	not aged niestarzone	523	555	596	4	3
	nienasycane żywicą aged starzone		508	566	625	6	

The wood was impregnated with a synthetic resin that was a combination of two products of commercial names Paraloid B-44 and Metaplex, each of them being a derivative of poly methacrylate of methyl. These two products (in crystalline form) were mixed together in a mass proportion of 1:4. A solution of so obtained mixture was used to impregnate the wood. The mixture itself was a 20% solution in toluene. The wood was impregnated in vacuum conditions during a 30-minute period at about 20 kPa pressure. Impregnated samples were seasoned for 30 days under conditions of limited solvent evaporation to minimise the effect of so-called backward/reverse migration of resin. Then samples were seasoned again this time for 60 years and in conditions of good ventilation to fully evaporate the solvent and harden the resin in the wood tissues.

The control wood material were sapwood samples extracted from undamaged wood of Scots pine. The size of control sample sets was comparable to that of the experimental ones (i.e. damaged by fungi) both in respect of number of groups and repeatability and the treatment methods applied.

All the samples (control and experimental) intended to have been used for artificial ageing were exposed to physical factors in accordance with PN-71/C-89038 standard. They all were subjected to artificial ageing repeated eight times. The stages of ageing are presented in table 2.

Table 2. The stages of one cycle of accelerated ageing Tabela 2. Etapy jednego cyklu procesu przyspieszonego starzenia

No. Lp.	Procedure Zabieg	Temperature Temperatura [°C]	Time Czas [h]
1	Soaking in water Moczenie w wodzie	+ 20	48
2	Freezing Zamrażanie	20	12
3	Drying Suszenie	+ 70	12
4	Exposure to ultraviolet Naświetlanie promieniami UV	+ 20	4

The study reported in this paper was carried out in the laboratories of the Department of Wood Science and Wood Protection at the Faculty of Wood Technology of the Warsaw University of Life Sciences (SGGW), Poland.

Results

The present study has made it possible to obtain new information concerning the pattern of the resin penetration into wood degraded by *Phellinius pini* (Thore) Fr. fungus.

Table 3. The effect of wood impregnation with synthetic resin on selected wood properties

Tabela 3. Wpływ nasycania drewna żywicą syntetyczną na wybrane cechy drewna

	Wood d Gęstość	,	Amount of absorbed resin Ilość wchłoniętej żywicy		
Sample description Opis próbek	Average value Wartość średnia Wartość średnia Coefficient of variation Współczynnik zmienności		Average value Wartość średnia	Variation coefficient Współczynnik zmienności	
	\overline{X}	υ	\overline{X}	v	
	$[kg/m^3]$	[%]	[kg/m ³]	[%]	
Degraded wood Drewno zdegradowane	447	13	144	13	
Sound wood Drewno,,zdrowe"	568	6	145	16	

The results of wood impregnation with the resin are collected in table 3 and their graphical visualisation is shown in fig. 2.

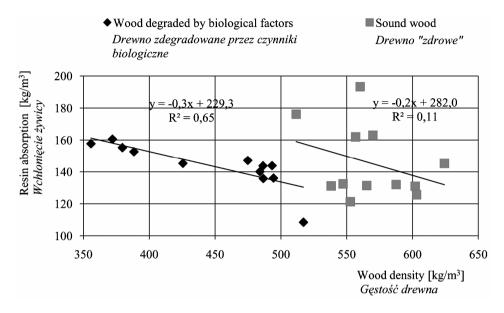


Fig. 2. The effect of wood density on absorption of synthetic resin Rys. 2. Wplyw gestości drewna na wchłoniecie żywicy syntetycznej

The resin penetration efficiencies into wood samples damaged by fungi and into sound samples were similar and in both case they equalled to about 145 kg/m³ on average.

Both degraded wood and sound wood showed similar patterns of the resin absorption which allows a general conclusion that the amount of absorbed resin becomes reduced with growing density of wood. The relation between resin absorption and wood density is of linear character and in the case of fungus degraded wood is characterised by rather high value of determination coefficient ($R^2 = 0.65$).

Tables 4, 5, and 6 contain collective results of the artificial ageing impact on selected parameters of wood impregnated with the resin, both in the case of fungi degraded wood samples and samples extracted from sound wood. The comparison of wood bending strength, as well as of the values of modulus of elasticity at bending and compression strength parallel to grain was done in two variants: for artificially aged wood and untreated wood.

The impregnation of wood with the resin resulted in increased bending strength of wood. In the case of degraded wood the increase was about 32.7% (from the initial 49 to 65 MPa), whilst in the case of sound wood the increase

was about 10.2% (from 128 to 141 MPa). The strength increase of degraded wood was greater than that of sound wood. Despite the visible increase of degraded wood strength after it was treated with the resin, the absolute value of strength was about two times lower than that of sound wood not treated with the resin.

Table 4. Bending strength of wood

Tabela 4. Wytrzymałość drewna na zginanie statyczne

			Bending strength Wytrzymałość na zginanie statyczne	
	le description pis próbek	Average value Wartość średnia	Variation coefficient Współczynnik zmienności	
		\overline{X}	v	
			[MPa]	[%]
	resin treated	not aged niestarzone	65	13,8
Degraded wood Drewno	nasycane żywicą	aged starzone	57	22,8
Drewno zdegradowane	untreated nienasycane żywicą	not aged niestarzone	49	23,2
		aged starzone	37	35,1
	resin treated nasycane żywicą	not aged niestarzone	141	11,3
Sound wood Drewno "zdrowe"		aged starzone	134	11,2
	untreated nienasycane żywicą	not aged niestarzone	128	7,8
		aged starzone	114	11,4

Artificial ageing of impregnated wood resulted in diminished bending strength of wood samples. The reduction in the strength degree varied from 65 to 57 MPa (that is about 12.3%) in the case of degraded wood treated with the resin, whilst in the case of treated sound wood the reduction in strength was relatively small – only by about 5.0% (from 141 to 134 MPa).

After artificial ageing samples of degraded wood not treated with the resin demonstrated reduced bending strength. The bending strength diminished from the initial about 49 MPa to the level of 37 MPa (by about 24.5%), whilst bending strength of untreated samples of sound wood dropped by about 10.9% from the original level of about 128 MPa to the final level equal to about 114 MPa.

Table 5. Modulus of elasticity values Tabela 5. Wyniki oznaczania modułu sprężystości

			Modulus of elasticity at bending Moduł sprężystości przy zginaniu statycznym		
	ole description pis próbek	Average value Wartość średnia	Variation coefficient Współczynnik zmienności		
			\overline{X}	v	
		[GPa]	[%]		
	resin treated	not aged niestarzone	8.1	17.3	
Degraded wood Drewno zdegradowane	nasycane żywicą	aged starzone	7.9	21,0	
	untreated nienasycane żywicą	not aged niestarzone	6.4	13.3	
		aged starzone	5.8	19.5	
Sound wood Drewno "zdrowe"	resin treated nasycane żywicą	not aged niestarzone	12.9	10.7	
		aged starzone	12.1	17.0	
	untreated nienasycane żywicą	not aged niestarzone	11.2	19.5	
		aged starzone	10.1	15.6	

The analysis of the average values of the modulus of elasticity proved that impregnation of degraded wood with the resin caused the rise of the modulus value from 6.4 up to 8.1 GPa, i.e. by about 26.7%. In the case of the sound wood samples the introduction of the same volume of the resin has resulted in a significantly smaller strengthening effect, because observed increase in modulus of elasticity was merely about 15.1% (from 11.2 to 12.9 GPa). It can be concluded from the above that fungi degraded wood samples were characterised by a significantly higher increased in the value of modulus of elasticity than samples of sound wood. Despite the visible increase in the modulus of elasticity value in the case of degraded wood after its impregnation with the resin, the absolute value of modulus of elasticity was by about 3 GPa lower compared with respective value for sound wood.

The artificial weathering of wood samples led to a decrease in the value of modulus of elasticity of sampled wood. After the last (eighth) cycle of ageing a decrease of about 2.5% (from the initial 8.1 to the final 7.9 GPa) in the value of modulus of elasticity was observed in the wood degraded by fungi and then

impregnated with the resin, whilst in the case of sound wood impregnated with the resin the fall was merely of about 6.2% (from 12.9 to 12.1 GPa).

Table 6. The index of compression strength parallel to grain
Tabela 6. Wyniki badania wytrzymałości na ściskanie wzdłuż włókien

			Compression strength parallel to grain Wytrzymałość na ściskanie wzdłuż włókien		
1	le description pis próbek		Average value Wartość średnia	Variation coefficient Współczynnik zmienności	
	•		\overline{X}	v	
			[MPa]	[%]	
resin treated		not aged niestarzone	49	16.3	
Degraded wood Drewno zdegradowane	nasycane żywicą	aged starzone	43	23.2	
	untreated nienasycane żywicą	not aged niestarzone	40	25.0	
		aged starzone	35	20.0	
Sound wood Drewno "zdrowe"	resin treated nasycane żywicą	not aged niestarzone	80	6.3	
		aged starzone	73	8.2	
	untreated	not aged niestarzone	71	5.6	
	nienasycane żywicą	aged starzone	62	14.5	

At the same time wood samples degraded by fungi and not subjected to impregnation with the resin demonstrated a fall in the value of modulus of elasticity by about 12.5% (from 6.4 to 5.8 GPa), whilst sound wood samples, not treated with the resin, reacted by a smaller fall by about 9.8% (from 11.2 to 10.1 GPa).

Impregnation of wood samples with the resin caused an increase in the value of the index of bending parallel to grain. In the case of degraded wood the index value rose from the initial 40 MPa to 49 MPa, (an increase of about 22.5%), and in the case of sound wood the respective rise was from 71 MPa to 80 MPa (i.e. by about 12.6%). Even though after impregnation with the resin wood degraded by fungi reacted by a more visible increase in the value of the index, its absolute strength was still lower by about 22 MPa compared with untreated sound wood. The process of artificial ageing caused a drop in the value of the index of bending parallel to grain. In the case of degraded wood treated with the

resin, ageing resulted in a drop in strength from 49 MPa to 43 MPa, i.e. by about 12.2%, whilst in the case of sound wood the respective drop was smaller – only of about 8.8% (from 80 MPa to 73 MPa).

After ageing samples of degraded wood not treated with the resin demonstrated a drop in bending strength from 40 MPa to 35 MPa (change by about 11.1%), whilst bending strength of sound wood samples not treated with the resin diminished from the initial 71 MPa to 62 MPa (by about 12.7%).

The analysis of results obtained in this research project allows formulation of the following sentence: Scots pine heartwood decay initiated by *Phellinus pini* (Thore) Fr. fungus makes the wood structure permeable which enables effective impregnation of wood with the synthetic resin in an amount similar to that absorbed by the undamaged sapwood of Scots pine. It is worth stressing that sound Scots pine heartwood is commonly believed to be impossible to impregnate.

Impregnation of wood with the resin results in an increased resistance of wood. This effect was observed for fungi damaged wood and for wood without any symptoms of damage. This pattern was also true for static bending of wood and test for compression strength parallel to grain. Always a more perceptible effect of wood strengthening was observed in the case of wood samples damaged by fungi, yet never the increase in strength fully set off the negative effect of the strength change caused by fungi causing damage of wood structure.

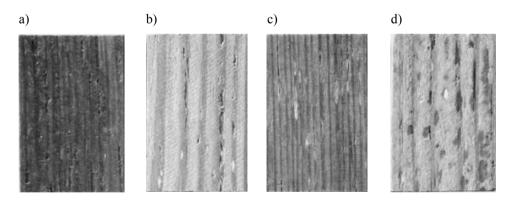


Fig. 3. The appearance of wood samples degraded by *Phellinus pini* (Thore) Fr.:

- a) wood treated with the resin, not aged,
- b) wood not treated with the resin, not aged,
- c) wood treated with the resin, aged,
- d) wood not treated with the resin, aged.
- Rys. 3. Wygląd próbek drewna zdegradowanego przez grzyba Phellinus pini (Thore) Fr.:
- a) drewno nasycone żywicą, nie poddane procesowi sztucznego starzenia,
- b) drewno nienasycone żywicą, nie poddane procesowi sztucznego starzenia,
- c) drewno nasycone żywicą, poddane sztucznemu starzeniu,
- d) drewno nienasycone żywicą, poddane sztucznemu starzeniu.

And this means that wood degraded by fungi does not fully recover after it was impregnated with resin and it cannot regain 100% of the original strength.

It was observed that every time artificial ageing caused reduction in wood strength. The wood destroyed by fungi always demonstrated noticeably smaller change in strength compared with sound wood. Most likely this pattern could have been caused by the fact that the wood with damaged cellulose structure (following the enzymatic activity of fungi) is no more susceptible to influence of physical factors at a degree characteristic for sound wood.

It was found that wood treated with the resin and then subjected to artificial ageing was weakened to a smaller degree compared with untreated wood. Therefore it may be concluded that impregnation of wood with resins increases wood resistance to external physical factors.

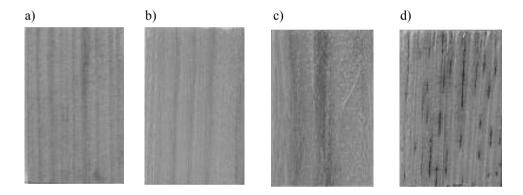


Fig. 4. The appearance of sound wood samples (wood free of decay):

- a) wood treated with the resin, not aged,
- b) wood not treated with the resin, not aged,
- c) wood treated with the resin, aged,
- d) wood not treated with the resin, age.
- Rys. 4. Wygląd próbek drewna "zdrowego":
- a) drewno nasycone żywica, nie poddane procesowi sztucznego starzenia,
- b) drewno nienasycone żywicą, nie poddane procesowi sztucznego starzenia,
- c) drewno nasycone żywicą, poddane sztucznemu starzeni,
- d) drewno nienasycone żywicą, poddane sztucznemu starzeniu.

Impregnation of wood with the resin causes changes in the shape and colour of treated wood samples regardless of whether the samples had been artificially aged or not (fig. 3). Moreover, it was found that changes of wood shape and colour occurring in wood not treated with the resin are less compared with treated wood. This holds true both for degraded wood and sound wood (fig. 4).

Conclusions

- 1. This study found that in both cases, i.e. in the case of wood damaged by *Phellinus pini* (Thore) Fr. and sound wood, impregnation of wood with the resin resulted in increased bending strength and wood stability in terms of compression strength parallel to grain.
- 2. The increased mechanical stability of wood impregnated with the resin is more evident in the case of damaged wood than in the case of sound wood.
- 3. Scots pine heartwood degraded by the fungus is absorbing the resin at a degree comparable to that of undamaged wood samples.
- 4. Resin impregnated wood is less susceptible to artificial ageing compared with untreated wood.

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WPŁYW NASYCANIA POLIMETAKRYLANEM METYLU NA WŁAŚCIWOŚCI MECHANICZNE DREWNA SOSNY ZDEGRADOWANEGO PRZEZ GRZYBY

Streszczenie

Przedstawiono wyniki badań nad możliwościami wykorzystania polimetakrylanu metylu do wzmacniania drewna zdegradowanego przez grzyba *Phellinus pini (Thore) Fr.* Zaobserwowano, że impregnacja drewna żywicą powoduje wzrost właściwości mechanicznych, niemniej jednak wzrost ten jest relatywnie wyższy w przypadku drewna zdegradowanego przez czynniki biologiczne, aniżeli w przypadku drewna nieuszkodzonego. Badano zachowanie się układu drewno – żywica po poddaniu go sztucznemu starzeniu w warunkach laboratoryjnych. Po procesie sztucznego starzenia wytrzymałość drewna nasyconego żywicą obniżyła się w porównaniu z wytrzymałością drewna niepoddanego starzeniu. Podobne tendencje zmian zaobserwowano zarówno dla drewna zdegradowanego przez czynniki biologiczne, jak i dla drewna nieuszkodzonego.

Słowa kluczowe: żywica syntetyczna, drewno zdegradowane, drewno wzmocnione, impregnacja drewna żywicą syntetyczną