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COMPOSITE MATERIALS ON THE BASIS OF WOOD AND SILICON ACRYLATES

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The main purpose of impregnating wood with plastics is not to enhance its mechanical properties in general, but first of all to stabilize it against atmospherical and biological attack, depending on moisture absorption. Hydrofobic silicon derivatives can be bound to cellulosic materials by radiation curing. Methacrylester of bimethoxy-propyloxy-silane can be impregnated and polymerized by gamma radiation in different wood species as beech, ash, oak and fir. Water absorption of the silicon-grafted beech- and ash-wood is reduced significantly, but the silicon-wood composite material is far from to be water-repellent.

INTRODUCTION

The organic silicon compounds are well known for their excellent water-repellent and "release" properties. To impart hydrofobic features to wood, impregnation with silicon oils has been proposed several years ago [5]. Impregnating wood with oligomer siloxanes, e.g. 20% alkyl-trialkoxy-silane dissolved in solvent naphta mixed with ethanol, results in products with improved dimensional stability after drying [4]. Wood-silicon polymer composites have been patented by Wacker by using chemical initiators and higher temperature for bonding silicones in wood [2, 3]. Dimethacrylate-methyl-dimethyl-siloxane oligomers were polymerized by peroxi initiation in wooden museum-objects. Alkylsiloxane coupling agents were used in wood-polymer composites [1, 6].

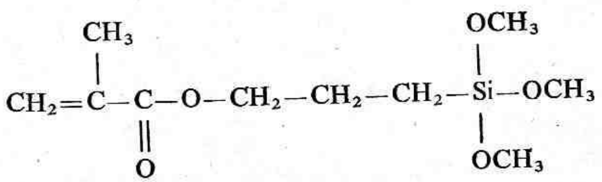
Silicon derivatives can easily be bound to cellulosic materials by radiation-curing. Recent publications are indicating that highly reactive silicon acrylate coating materials are applied on high-speed electron-beam-curing (EBC) lines producing siliconized "release-papers".

Our aim was to impregnate wood with silicone acrylates polymerized afterwords by radiation initiation to achieve stabilized wood.

MATERIALS

Air-dried samples of the following European woods have been prepared: fir (*Picea abies*), oak (*Quercus cerris*), ash (*Fraxinus excelsior*) and beech (*Fagus silvatica*). Dimension of the samples was: 30 mm (L) × 10 mm (R) × 10 mm (T). The average water content of the fir, oak, ash and beech samples before the treatment was 8.3; 8.6; 5.8 and 7.6% respectively, as related to the mass of dry wood.

As impregnating material methacrylester of trimethoxypropyloxy-silane has been used (SILAN GF 31 of Wacker Chemie GmbH):



This is a typical silicon acrylate which can be used as sizing material, a coupling agent for treating glassfibers or other reinforcements and fillers applied in composite plastics. Its double bond can be polymerized by radical initiation. On the other hand, through its methoxy groups involved in hydrolytic condensation, a 3-dimensional polysiloxane chain structure can also be formed.

METHODS

The air-dried wood samples were evacuated during 40 mins. The impregnation with the low-viscosity silicon-acrylate liquid (of boiling point 125°C at 15 mbar) was performed at room temperature under atmospheric pressure of pure nitrogen. After an overnight impregnation period the excess monomer has been drained off. The radiation polymerization was made in a Co-60 gamma source, applying a dose rate of 400 Gy/h and curing dose of 28 kGy at room temperature, in nitrogen atmosphere.

The water-absorption of the silicon-treated and untreated wood samples has been determined by immersion in water at 20±2°C, under 50 mm of water-level.

The following symbols have been used in the calculations:

w_u – mass of the untreated, air-dry wood sample;

w_t – mass of the silicon-treated wood sample in air-dry state;

w_0 – mass of the absolute dry wood material;

w_{wt} and w_{wu} – mass of the treated and untreated samples after immersion in water.

The uptaken silicon content of samples:

$$SC\,(\%) = \frac{w_t - w_u}{w_0}\,100$$

The water-absorption has been determined as follows:

$$WA_t(\%) = \frac{w_{wt} - w_t}{w_0} 100 \text{ for treated wood},$$

and

$$WA_u(\%) = \frac{w_{wu} - w_u}{w_0} 100 \text{ for untreated wood}.$$

The swelling (SW%) was determined from the difference of dimensions of water--immersed and air-dry samples measured in the tangential (T) and radial (R) directions. (In the longitudinal direction the swelling was undetectable during the silicone-impregnation as well as during water-immersion). The anti-swelling-efficiency (ASE%) was measured after 48 hours of immersion in water, as follows:

$$ASE(\%) = \frac{\text{original swelling} - \text{swelling of treated wood}}{\text{original swelling}} 100.$$

RESULTS AND DISCUSSION

The silicon-acrylate polymerized in glass vial without wood under the indicated radiation-parameters forms a glass-clear, rubbery block. This block shows after the opening of glass tube a further hardening on the surface, indicating hydrolytic polycondensation of the silicon-connected methoxy groups.

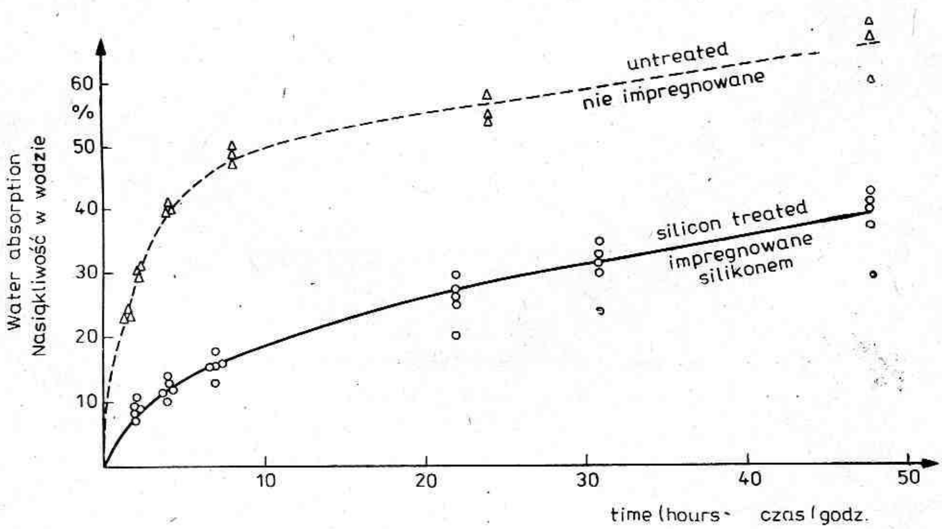


Fig. 1. Water absorption of silicon treated beech-wood

Rys. 1. Nasiąkliwość w wodzie drewna buka impregnowanego silikonem

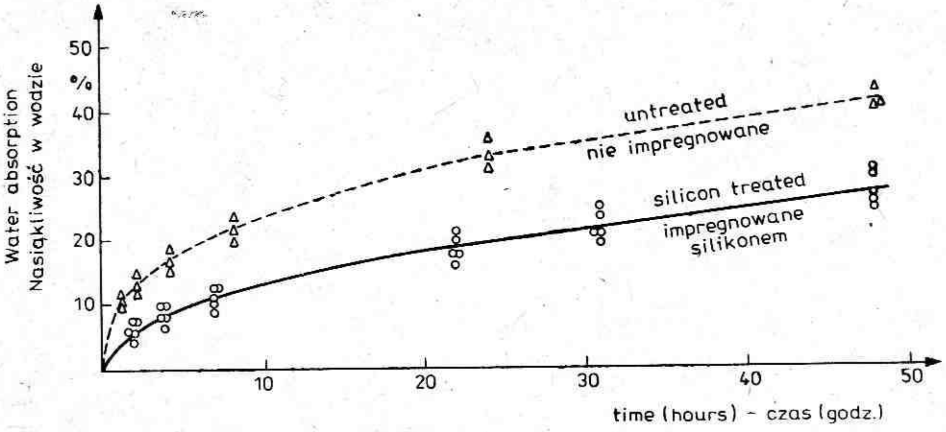


Fig. 2. Water absorption of silicon treated and untreated ash-wood

Rys. 2. Nasiąkliwość w wodzie drewna jesionu impregnowanego silikonem

The uptaken silicon content (SC) of the wood as determined taking an average of five samples was the following: 63.2% for beech, 43.0% for ash, 35.0% for oak and 27.6% for fir.

The water-absorption (WA) of silicon-treated and untreated wood are seen on figures 1 - 4.

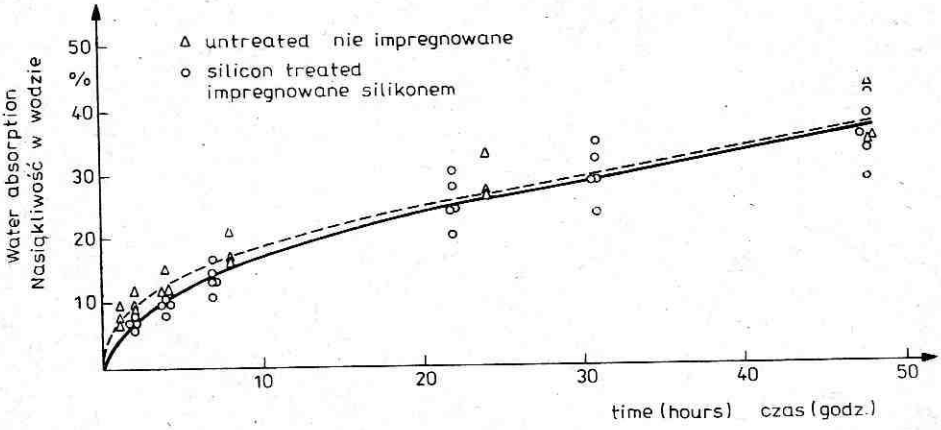


Fig. 3. Water absorption of silicon treated and untreated oak-wood

Rys. 3. Nasiąkliwość w wodzie drewna dębu impregnowanego silikonem

In case of beech- and ash-wood there is a significant difference in water-absorption between silicon-treated and untreated wood. The silicon-treatment diminishes the rate of water absorption especially at the first hours of immersion.

In case of oak-wood (fig. 3) there is practically no difference between treated and untreated wood. In the case of fir-wood (fig. 4) the situation is even worse: the silicon-

-treated wood absorbs seemingly more water than the untreated wood after about 8 hours of immersion. The water absorption at this critical time is about 20% which is close to the fiber-saturation-point (FSP) of the coniferous woods of high resin content. It seems that the silicon acrylate treatment retards water absorption only

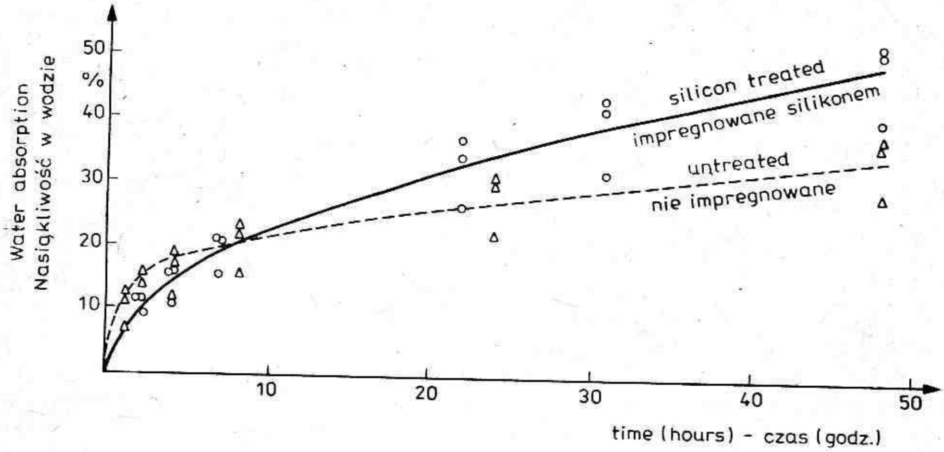


Fig. 4. Water absorption of silicon treated and untreated fir-wood
Rys. 4. Nasiąkliwość w wodzie drewna świerka impregnowanego silikonem

at the beginning of water-interaction. On the other hand the polymerized silicon acrylate holds the pits of the wood cells in open position making the whole wood structure more accesible to the water.

The table 1 shows the swelling properties of silicon-treated woods. As mentioned before the longitudinal swelling was insignificant, and the tangential swelling was

Table 1

Swelling of untreated and silicon-treated wood after 48 hours immersion
Pęcznienie naturalnego i impregnowanego silikonem drewna po 48 godz. moczenia w wodzie

Wood species Rodzaj drewna	Silicon content Zawartość silikonu	Swelling Pęcznienie				Anti-swelling efficiency Wskaźnik zmniejszenia pęcznienia	
		radial promieniowe		tangential styczne			
		untreated naturalne	treated impregnowane	untreated naturalne	treated impregnowane	radial promieniowego	tangential stycznego
	%						
Beech Buk	63.2	4.0	4.6	12.1	11.2	−15.0	7.4
Ash Jesion	43.0	4.1	3.3	8.0	7.1	19.5	11.3
Oak Dąb	35.0	4.6	3.1	6.8	5.4	32.6	20.6
Fir Świerk	27.6	4.9	3.5	7.0	6.4	32.7	−22.4

always higher than the radial one in untreated and in treated samples as well. Generally, the silicon-acrylate treatment reduces the dimensional swelling of the woods but not more than 20 - 30%.

In conclusion it can be stated that the silicon acrylate can be easily polymerized in wood by gamma irradiation. The composite material obtained can be considered as silicon-grafted wood, because of the radical initiation which is started – partially – by free radicals formed on ligno-cellulosic complex. The silicon grafting modifies somewhat the water absorption properties of the wood but the composite material obtained is far from to be watei-repellent.

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KOMPOZYTY MATERIAŁOWE NA BAZIE DREWNA I SILIKONÓW AKRYLOWYCH

Streszczenie

Głównym celem impregnacji drewna tworzywami syntetycznymi nie jest zmiana właściwości mechanicznych, lecz w pierwszym rzędzie uodpornienie drewna na działanie czynników atmosferycznych, a także biologicznych. Wiąże się to ściśle z absorpcją wody przez drewno. Hydrofobowe pochodne silikonów mogą łączyć się z materiałami celulozowymi z udziałem promieniowania radiacyjnego. Ester metakrylowy bimetoksypropyloksysilanu może być użyty do impregnacji różnych rodzajów drewna i spolimeryzowany za pomocą promieni gamma. Absorpcja wody przez drewno buka i jesionu z zaszczepionym silikonem uległa znacznemu obniżeniu, jednak kompozytu drewno-silikon nie można uznać za w pełni odporny na działanie wody.

КОМПОЗИТЫ НА ОСНОВЕ ДРЕВЕСИНЫ И АКРИЛОВЫХ СИЛИКОНОВ

Резюме

Главной целью пропитки древесины синтетическими соединениями является не изменение механических свойств, а, прежде всего, повышение устойчивости древесины к действию атмосферных и биологических факторов. Это тесно связано с абсорбцией древесиной

воды. Гидрофобные производные силиконов могут соединяться с целлюлозными материалами при радиационном облучении. Метакриловый эфир биметоксипропилоксисилана можно применять для пропитки разных древесных пород и подвергать полимеризации гамма--лучами. Абсорбция воды древесиной бука и ясеня с привитым силиконом значительно снизилась, однако нельзя считать композит древесина-силикон полностью устойчивым к действию воды.

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