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INFLUENCE OF LOW FREQUENCY ULTRASONIC FIELD ON THE CHANGES OF ANATOMICAL WOOD STRUCTURE

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Inwestigations were carried out on the defibration of the pine wood in a low frequency ultrasonic field. The highest defibration dynamic was observed during the two first hours of the sonication. The predominant effects of ultrasound treatment are: fibrilation of the external wood parts and deterioration at the middle lamella region.

INTRODUCTION

Limited base of the raw material and necessity of the environmental protection are principal reason conducted to improve chemical wood technology. There are investigating the possibilitys of use the new physical and chemical parameters to stimulate wood decomposition. Among these, there are the proof of use the wave energy and not only the high energy electromagnetic wave but also acoustic wave — ultrasounds. So called active use of the ultrasounds depend on the action of the wave on the matter and causing physical and chemical changes. These effects necessite concentration of the energy that can be obtained by the wave amplitude increase or frequency changes. At liquid, the strongest effect are observed at low frequency ultrasonic field. The possibilitys of use ultrasound as an active agent in wood processing was faund out in the middle of thirthieth when B u c k i n g h a m [14] confirmed that sonication of the cellulosic fibres at water caused effects similar to these during pulp milling. The others authors observed above all influence of the irradiation on the hydration and fibrilation [1, 3, 6, 7, 14]. The result of these study allowed to work out and introduce to industry methods of defibration with ultrasounds formed on the mechanical way (high frequency pulp treatment) [13].

In spite of investigation on ultrasonic delignification of wood, this method didn't lead to industrial practice [2, 4, 5, 8, 15]. The estimate of the effect of wood irradiation were more difficult than at the pulp. The reason is complicated anatomical and chemical structure of wood. The aim of this paper is archievement of the qualitative and quantitative evaluation of the effect ultrasonic wood irradiation at the medium favourable to delignification.

EXPERIMENTAL

The debarked pinewood (*Pinus silvestris* L.) used were preliminary grinding at disk mill and than at conical mill. After sieving, from the received sawdust, to following study were chosen main fraction 0,5 - 1,2 mm.

The sawdust were sonicated using the ultrasonic generator (Model UDM-10 from IPPT PAN „Techpan" Warszawa, Poland). Generator was connected to magnetostriction transducer having a nominal frequency 21 kHz. Apparatus make possible measurements of the wave amplitude at the range from 5 up to 50 μm and electrical power up to 2 kW. Visual inspection revealed that this combination of frequency and intensity, produced cavitation in the liquid. The diameter of the tip of the transformer was 1,8 cm. At 21 kHz, the ratio of diameter to wavelength of sound in water was 0,25. For such a value, the shape of the ultrasonic field should be approximately hemispherical, which would provide a relatively uniform distribution of energy over the cross-section of the treatment vessel (volume — 75 cm^3). The sonications were carried out at the thermostating (20 ± 1°C) glass vessel (diameter — 3,5 cm, height — 9 cm).

DEFIBRATION ANALYSIS

Weighed portion, near 2 g of the sawdust (5,2% humidity) was put at the vessel and flooded 75 cm^3 aqueous 8% NaOH or water (reference). After 15 min soaking, the sawdust were sonicated at the established parameters of ultrasonic treatment. The longest experiments were interrupted after an hour (5 min) in order to assure stability of work of the transducer and of the tip. The experiments were also carried out for the unsonicated samples.

After insonation at the vessel were three fraction of the wood: on the bottom sawdust, above fibrous fraction that in the following parts of paper is called „defibrous fraction" and at last, floating at the solution colloidal fraction. Solution with defibrous fraction were decanted and filtered G-4 drain). Filtrate analysis (centrifuging, decantation liquid phase and evaporation to total solids) showed that colloidal fraction contents is less than 0,5% abs. dry mass of weighed wood portion and that is contained at the assumed range of the methodic error for the defibrous fraction determinations.

Table 1

Variable factores of the experience
Parametry zmienne doświadczenia

Amplitude (μm) Amplituda	Time (min) Czas
0, 8, 16, 24, 32, 40, 48	15, 30, 45, 60, 120, 180, 240, 360, 480

In case of the experience performed at alkaline solutions, drains with wood were washed with hot water, 10% acetic acid and again hot water up to neutral reaction. Drains were drying to constant weight at 378 K.

Received after sonication amounts of the defibrous fraction were calculated as a % of the abs. dry mass of weighed wood portion. For any variant of the treatment were performed 15 sonications. Mean results are inserted at tab. 2 - 5.

SCANNING ELECTRON MICROSCOPY

The sonicated sawdust and their defibrous fraction were examinated. The samples after cut-out (cross section analysis) were shadowed with a thick layer of carbon (fig. 1 - 2) and silver (fig. 3 - 9), spread uniformly from all sides. The defibrous fraction didn't cut-out. The observations and photos were performed using SEM: JEOL-JSM-50A (fig. 1 - 2) and TESLA (fig. 3 - 9) with resolution 15 nm and 12 nm.

RESULTS AND DISCUSSION

During the sonication in aqueous solution acoustic waves degrade wood causing above all defibration. Take into consideration that defibration is an agent making easier chemical wood processing, the maximalisation of this phenomenon were admited as an wood decomposition criterion.

The aim of the first experience (tab. 2) was determination of the dynamics wood decomposition dependent on time of the treatment. The sonication was performed at water using 40 μm wave amplitude. Amplitude was chosen following the early published results of the study, showed that intensification of the wood decomposition in ultrasonic fields occured from 32 μm up to 50 μm [11, 12]. The choice of the time of experience was caused by deficiency of the data concerned longterm ultrasonic treatment of wood and fibrous mass. The most of the experiences were performed using short time of the treatment for reason of lack of the thermostate and quick medium heating [10, 15].

Table 2

The effect of sonication time on the changes of the defibrous fraction amount
Wpływ czasu nadźwiękawiania na zmiany ilości frakcji rozwłóknionej

Time (min)	15	30	45	60	120	240	360	480	Czas (min)
Defibrous fraction in % dry mass of wood	3.73	4.39	5.56	7.50	6.57	7.85	7.60	18.05	Ilość frakcji rozwłóknionej w % masy b. s. drewna

The result (tab. 2) showed that the strongest wood decomposition occured during first hour of the treatment. The prolongation of the experiences didn't cause the intensification of the degradation. That can be result of the weakness and decay of the cavitation, that is particulary intensive during first twenty min. of sonication [9]. After 8 hours were observed increase of the amount defibrous fraction up to 18,05%, but even so long period of the sonication didn't cause total defibration of the wood.

Take into consideration observations performed after long term sawdust sonication in aqueous solution, the study on the influence of ultrasonic wave were carried out at the constant time of the experience — 1 hour. The aim of the experience was optimization of the defibration dependent on used wave amplitude.

Table 3

The effect of wave amplitude and chemical medium on wood defibration
Wpływ amplitudy fali ultradźwiękowej i środowiska obróbki na rozwłóknianie

	Defibrous fraction in % dry mass of wood Ilość frakcji rozwłóknionej w % b.s. drewna						
Medium Środowisko / Amplitude Amplituda (μm)	0	8	16	24	32	40	48
Neutral Obojętne	0.79	1.03	1.20	1.35	5.60	7.73	6.34
Alkaline Zasadowe	1.08	1.31	1.69	1.84	2.86	11.62	15.81

Result from tab. 3 show that wood decomposition at water begin at the amplitude — 32 μm. Determined amount of the defibrous fraction is above 4 time larger that one at 24 μm and 7 time larger in comparison to the reference. The wood decomposition intensify after increasing of the amplitude up to 40 μm. Mean quantity defibrous fraction attain than the biggest value for experience at aqueous solution — 7,33%.

At alkaline medium the wood decomposition intensify alike at water during treatment at the 32 μm. Determined amount of the defibrous fraction (2.86%) were yet less than at water (5,60%). This result can be explain by relative weakness of the influence of ultrasonic waves at relative dense 8% aqueous NaOH. The sawdust assembled on the bottom of the vessel didn't subject intensive mixing. In spite of the considerable swelling that was reason of the limitation of the sonication effects. Confirmed this, the results of the additional experience performed at 1% and 4% alkaline solution (tab. 4). In the most diluted 1% NaOH, after sonication at 32 μm were observed intensive mixing of the sawdust and determined amounts of the defibrous fraction (9,63%) were above three time bigger than that in 8% alkaline solution. After sonication at the same amplitude but in more concentrated solution (4%) were determined only 1,16% defibrous fraction. Sonication at the 40 μm caused circulation of the sawdust at the vessel, as well in 1-, 4- as in 8% alkaline solution. That caused diminution of the difference between results received at this solutions. It is worth to notice that the biggest amount of the defibrous fraction was received after sonication in 8% aqueous NaOH at 48 μm (15,81%).

Table 4

The effect of alkaline solution concentration on ultrasonic wood defibration

Wpływ stężenia roztworu zasadowego na ilość rozwłóknionej frakcji drewna

Solution / Roztwór Amplitude / Amplituda (μm)	Defibrous fraction in % dry mass of wood / Ilość frakcji rozwłóknionej w % b.s. drewna	
	1% NaOH	4% NaOH
24	0.77	0.96
32	9.63	1.16
40	10.74	12.61

The results of the longterm sonications at water proved that wood decomposition is more intensive during first hour of treatment. Observed real influence of alkalies on the defibration at differents amplitudes indicates the possibility of the intensify that proces with prolongation of the treatment. Take that into consideration were carried out investigations on influence of time irradiation on the wood decomposition at alkalies. The aim of the study was optimization of the time defibration with ultrasound. The sonication time ranged from 1 up to 4 hours and was close to alkaline pulping time. The treatment were carried out at 48 µm — at that amplitude the amount of early determined (tab. 3) defibrous fraction was biggest.

Table 5

The effect of exposure time and chemical medium on the defibrous fraction amount

Wpływ czasu ekspozycji i środowiska obróbki na rozwłóknianie drewna

	Defibrous fraction in % dry mass of wood Ilość frakcji rozwłóknionej w % b.s. drewna							
Time Czas (min)	60		120		180		240	
Medium Środowisko	A	B	A	B	A	B	A	B
Neutral Obojętne	0.79	7.50	1.20	6.75	1.41	6.74	1.32	7.85
Alkaline Zasadowe	1.08	15.81	1.53	20.05	1.62	17.73	1.62	16.66

A – reference
– odniesienie
B – sonicated
– próby nadźwiękawiane

The results from tab. 5 prove that wood decomposition attained maximum after 2 hours of the sonication — 20,06% defibrous fraction. Prolongation of the treatment didn't cause increase of the defibration, on the contrary for 3- and 4 hours were determined: 17,73% and 16,66% defibrous fraction. Apparent lowering of the results with prolongation of the treatment can be explained by the repeated desintegration early defibred wood fraction. Formed during this process dusty fraction weren't stoped at the drains and therefore weren't seized in the results.

Presented above results of the study allowed to appreciate quantitative size of the ultrasonic wood defibration. Scaning electron microscopy allowed to appreciate, produced during insonation morphological changes of the wood. Because the determinants of the effectivity of ultrasonic action on wood are these parameters of the treatment that caused the biggest defibration, for investigations were chosen sawdust samples sonicated during 2 hours at 48 µm.

Fig. 1 and 2 illustrates defibration of the external wood parts. In fig. 1 partial defibration of the cell wall is clearly visible. Intensive insonation caused detaching of the fibril beans, formed essentialy from cellulose. The cell wall adjacent to defibrous part of the wood is separated in the middle lamella zone that means delignification. Fig. 2 presents the part of defibrous fraction that was subjected the quantitative analisys. This fraction is composed essentialy from the group of the tracheids or single tracheids with considerably destructed cell wall surfaces. Cross

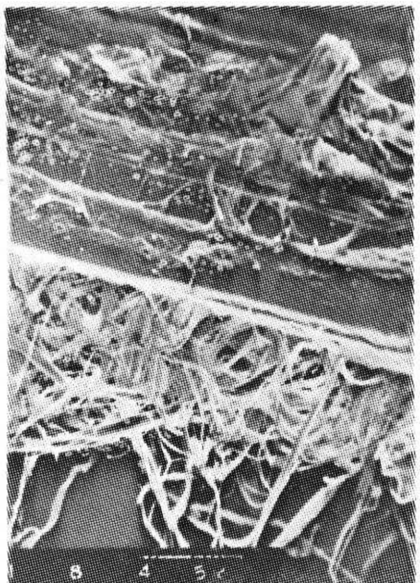

Fig. 1. Defibrous part of the cell wall: (×400)

Rys. 1. Rozwłókniony fragment ściany komórkowej (×400)

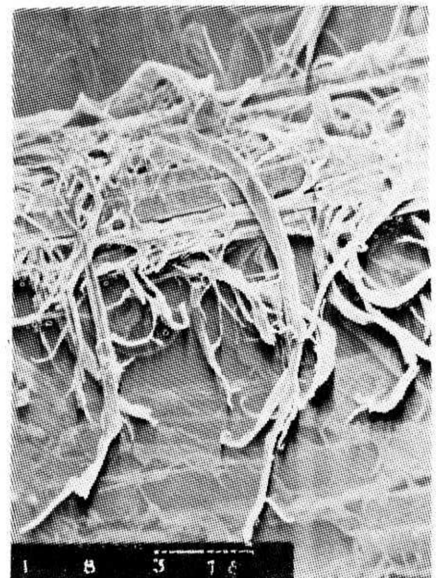

Fig. 2. Part of the defibrous fraction (×300)

Rys. 2. Fragment rozwłóknionej frakcji drewna (×300)

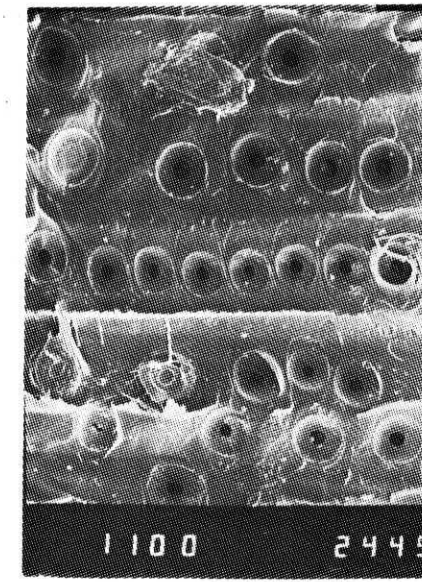

Fig. 3. Radial section of the unsonicated pinewood (×1100)

Rys. 3. Przekrój promieniowy drewna — próbka wzorcowa (×1100)

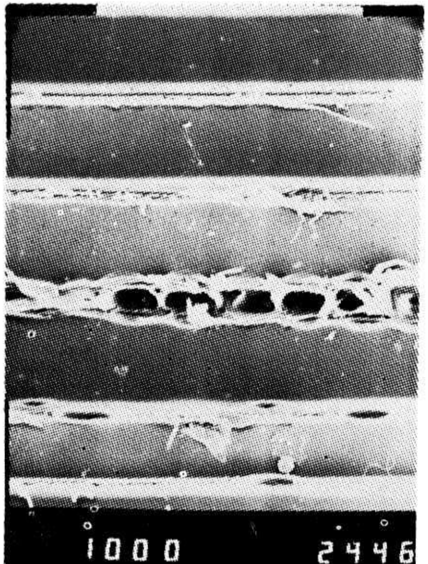

Fig. 4. Tangential section of the unsonicated pinewood (×1100)
Rys. 4. Przekrój styczny drewna — próbka wzorcowa (×1100)

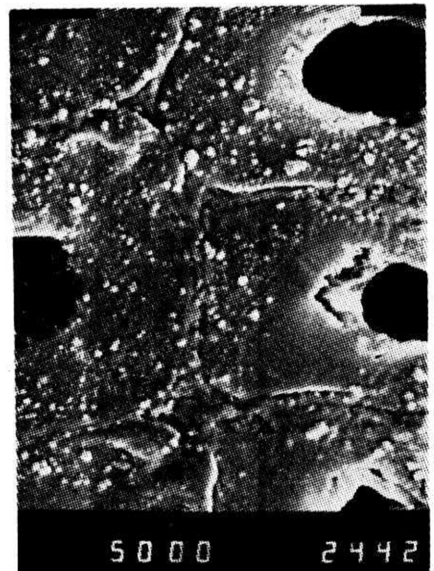

Fig. 5. Transvers section of the unsonicated pinewood (×5000)
Rys. 5. Przekrój poprzeczny drewna — próbka wzorcowa (×5000)

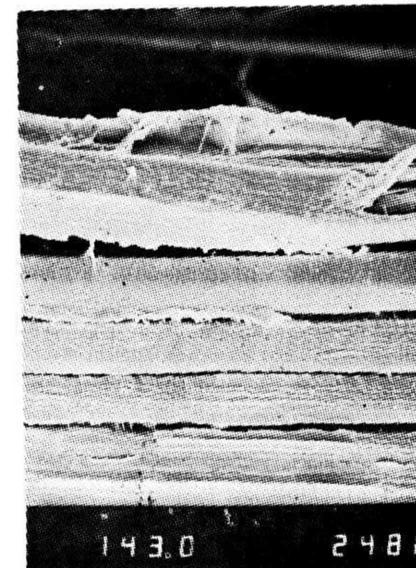

Fig. 6. Decomposition of the sonicated pinewood. Radial section (×1430)
Rys. 6. Zjawiska rozkładowe na przekraju promieniowym nadźwiękawianego drewna sosny (×1430)

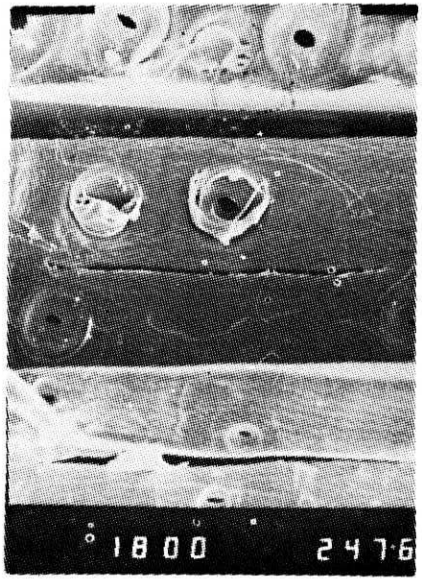

Fig. 7. Tracheids liberated from the group. Radial section of the sonicated pinewood (×1800)

Rys. 7. Cewki uwolnione z zespołu. Przekrój promieniowy nadźwiękawianego drewna sosny (×1800)

Fig. 8. Transvers section of the sonicated pinewood (×400)

Rys. 8. Przekrój poprzeczny — próbka nadźwiękawiana (×400)

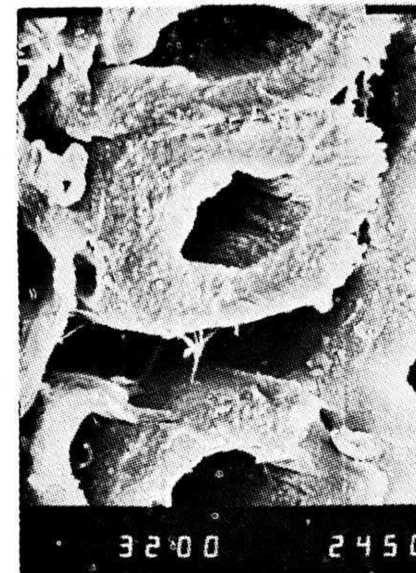

Fig. 9. Erosion at the middle lamella region. Transwers section of the sonicated pinewood (×3200)

Rys. 9. Erozja w warstwie blaszki środkowej. Przekrój poprzeczny nadźwiękawianego drewna sosnowego (×3200) sections of the pinewood are normally quite simple and homogeneous (fig. 3 - 5). In the fig. 6 is visible radial section of the sonicated wood. The micrograph shows decomposition of the anatomical elements. This effect might have been caused by the knife during cutting but even if this were the case, it may have been facilitated by the ultrasonic irradiation. Fig. 7 shows radial section of the sonicated wood. There is cleary visible a tracheid with oblong cracks liberated from the group. The cracks appears not only in the middle lamella region but also in the middle of cell wall. As shown in fig. 8 and 9 (transwers section) earlywood presents more homogeneus structure so were more resisted on the ultrasonic degradation. In the region of the latewood were observed lot of oblong cracks. The SEM analisys performed at bigger magnification (up to 5000) proved that separation of the cell walls and erosion in the middle lamella region occured as well in the earlywood as in the latewood.

CONCLUSION

1. During ultrasonic wood irradiation occures co-operation between the agents: time of sonication, wave amplitude and chemical medium:

a) the biggest intensity of the wood decomposition occures during first hour of the treatment; longterm sonication caused the repeated desintegration early defibred wood fraction and so lowering of the results,

b) the more dynamics wood decomposition were observed after sonication ranged from 40 up to 48 μm; in this range ultrasonic intensity is near to 60 W/cm² of the radiating surface of concentrator horn.

2. The chemical medium differences the effects of the sonication; during insonation at alkalies the essential phenomenons are:

a) fibrilation of the external layers of cell walls,

b) erosion in the middle lamella region.

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WPŁYW POLA ULTRADŹWIĘKOWEGO NISKIEJ CZĘSTOTLIWOŚCI NA ZMIANY STRUKTURY ANATOMICZNEJ DREWNA

Streszczenie

Celem badań było dokonanie oceny ilościowej i jakościowej zjawiska rozwłókniania i degradacji elementów budowy anatomicznej drewna poddanego obróbce ultradźwiękowej w środowisku zasadowym. Nadźwiękawianie trocin sosnowych prowadzono przy różnych parametrach czasu i amplitudy fali ultradźwiękowej. Analiza ilościowa powstającej w trakcie ekspozycji rozwłóknionej frakcji drewna pozwoliła ustalić optymalne z punktu widzenia maksymalizacji rozwłókniania parametry obróbki. Uzupełnieniem badań ilościowych są wyniki obserwacji mikroskopowych. Stwierdzono, że procesy degradacyjne z największą dynamiką występują

w ciągu pierwszej godziny nadźwiękawiania; przedłużenie czasu obróbki ponad dwie godziny powoduje występowanie wtórnego rozdrobnienia rozwłóknionej frakcji drewna. Zjawiskami dominującymi w trakcie nadźwiękawiania w środowisku zasadowym są: fibrylacja zewnętrznych, przyobwodowych partii trocin i erozja w warstwie blaszki środkowej.