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NOTICE

THE ACTION OF IONIZING RADIATION ON WOOD
AND ITS COMPONENTS

(Performed 3 November 1966 at the Institute of Wood Chemistry, Academy of Sciences, Latvian S.S.R.)

/Resume/

The effect of low doses of ionizing radiation on the properties of elementary spruce wood fibers, viz., cellulose, holocellulose, and cellolignin, was studied. Gamma doses were 10, 30, 50 and 70 Mrad at a radiation power of 600 rad/sec. Variation in properties after irradiating fibers was determined by their degree of swelling in a sulfuric acid-glucose solution.

A 10 Mrad dose destroyed hemicellulose and, particularly, cellulose in wood. Under these radiation conditions, fibers not containing lignin, cellulose and hemicellulose underwent considerably deeper destruction. Consequently, lignin exhibits protective action against radiation.

In turn, lignin under the effect of gamma rays varies more sharply in cellolignin than in wood.

At 50- and 70-Mrad doses, the primary tertiary walls of cellolignin fiber undergo marked destruction.

The action of radiation on high-polymer substances of natural origin, like wood, is of great interest from many points of view, which would include an effective method of investigating the chemical and morphological structure of lignin, cellulose and hemicellulose, and their interrelationships.

Literary data on the effect of radiation on wood, cellulose and other polysaccharides treats mainly of their chemical destruction at comparatively low doses. Little is known of the effect of low radiation doses on the properties and microstructure of individual wood components.

Korchemkin, Malinsk and Sukhov studied the effect of low radiation doses (1.5 and 10-Mrad) on variations in the microstructure of cellulose fibers (Ref. 1). They showed that 5 and 10-Mrad radiation doses so weakened the structure of the outer layer of the secondary wall that swelling of the fiber occurred without bead formation; while at 1 Mrad, the fibers, and also the control fibers, swell with the formation of numerous beads. This indicates that the onset of macromolecular transformation in cellulose is considerably earlier than established by Freydin (Ref. 2), who observed that at 50 Mrad hemicellulose undergoes destruction first.

Cellulose, according to his data, is more radiation resistant; lignin is still more resistant. He proposed that lignin plays a protective role in the cell wall by absorbing and scattering radiation energy.

The latter hypothesis was confirmed by research (Ref. 3) where, on the basis of chemical analyses of irradiated wood, it was shown that the disintegration of cellulose in wood in the presence of lignin is slower than that of free cellulose.

That cellulose in irradiated wood is protected to a considerable degree by lignin is also indicated in Ref. 4.

According to the data of Japanese researchers (Ref. 5), the 1 to 10 Mrad doses of ionizing radiation considerably affect lignin-carbohydrate bonds causing their partial destruction due to the advancing destruction of hemicellulose and cellulose, while at this time lignin remains practically unchanged. They found that cellulose undergoes destruction faster than hemicellulose, and at 10 Mrad, cellulose disappears completely. These data contradict those of Freydin, cited above.

Confirmation of the effect of low doses is also found in studies of the physical properties of wood. Freydin established (Ref. 6) that moisture-absorption of wood degrades even at 1 Mrad dose and reaches a minimum value at 10 Mrad. Increasing the dose to 100 Mrad does not change the picture. Haermans and Weidinger (Ref. 7) pointed out that water absorption by cellulose materials is proportional to their content of less-ordered polysaccharides. Consequently, the drop in moisture absorption by irradiated wood may perhaps be explained by the destruction of hemicellulose.

From the foregoing, it is obvious that the problem of why ionizing radiation causes the onset of destruction, why some wood components undergo destruction sooner, and why some are more radiation resistant is still inadequately explained and debatable.

As to the effect of radiation on variations in the microstructure of wood and its components, besides the studies on the swelling of irradiated cellulose fibers already cited above, we may point out the work of Polčin and others (Ref. 8). By conducting an electron microscope study of the morphological structure of the cell wall of spruce, beech, and poplar wood subjected to a 50 Mrad dose of gamma radiation, they established that ionizing radiation acts directly upon polysaccharide macromolecules to cause their destruction; and only further mechanical treatment causes destruction of other structural components of the wood.

In the work of Freydin (Ref. 2) there is also a short section which treats the microstructure of irradiated wood. However, it gives a weak presentation of variations in the cell wall which arise from radiation effect.

In this study we have traced the effect of small doses of ionizing radiation on the properties of individual components of the wood cell wall and on the structure of its individual layers. For this purpose we have dissected spruce wood fibers and obtained from them cellulose, holocellulose, and cellolignin fibers.

Irradiation was performed at the RK-L Irradiation Center on the atomic reactor of the Physics Institute of the Academy of Sciences, Latvian S.S.R. (Ref. 18). The fiber was placed in glass capsules and subjected to 10, 30, 50 and 70 Mrad gamma radiation at a power of 600 rad/sec. Variability of irradiated fibers was based on their swellability in a solution of H_2SO_4 with glucose (1 mol glucose per 9 mol 64.5% H_2SO_4).

THE EFFECT OF RADIATION ON CELLULOSE AND HOLOCELLULOSE SPRUCE FIBERS

Cellulose fibers subjected to 10 Mrad radiation dose externally did not differ from control samples, but upon swelling, the general width of the fibers and the thickness of their walls increased to a great degree (Table 1). It may be assumed that the destruction of cellulose begins at radiation doses less than 10 Mrad. Lateral damages in the swollen fibers were very small.

Cellulose fibers subjected to 30 Mrad react otherwise upon swelling: Swelling becomes very extensive (Table 1), beginning after approximately ten minutes. Upon swelling, multiple ruptures are formed as a result of the destruction in the unoriented part of cellulose macromolecules. A high degree of swellability indicates impairment of the oriented, highly-ordered portions of the cellulose also. Severe swelling of the secondary layer (S_2) leads to ruptures of the primary wall and layer S_1.*

*) The outer layer of the secondary wall.

A 50 Mrad radiation dose makes the cellulose fibers brittle. Upon swelling in this case, as compared with the preceding radiation dose (Table 1), the fiber configurations became amorphous, and in almost half of the fibers studied, the swelling process transformed into a dissolution process. In fibers which maintained their shape, fibrillation of the cell wall was evident.

After 70 Mrad radiation dose, the fibers upon swelling became amorphous and transparent instantaneously, which indicates the transformation of cellulose into an easily-hydrolyzed state. This confirms the data of Sharkov and his co-workers (Ref. 9), who, upon studying the effect of gamma rays on cellulose, established that 100 Mrad radiation dose causes the transformation of difficultly soluble cellulose into an easily hydrolyzed state as a result of the depolymerization of cellulose macromolecules.

Holocellulose fibers subjected to 10 Mrad radiation dose reached maximum swelling after 5-7 minutes; and also the controlled fibers, but the dimensions of the swollen fibers was almost twice greater than that of the control fibers (Table 1) and clearly expressed fibrillation was exhibited.

30 Mrad radiation dose accelerates the swelling of fibers and increases the dimensions of swelling (Table 1). Maximum swelling is reached after 3-5 minutes. At this time, there is no fibrillation of the secondary wall, the fibers are severely swollen, and the majority are wound in thin spirals perpendicular to the axis of the fiber, similar to the spirals of layer S_1.

Radiation of fibers at a common dose of 50 Mrad causes rapid and severe swelling, which transforms into dissolution in most fibers. Upon swelling, the fibers rupture in the transverse direction in different places. The spirals which wind themselves around the fiber are not evident, which indicates loss of configuration by the fibrillous fascicules of layer S_1 due to the destruction of cellulose.

Fibers irradiated at 70 Mrad swell after 1-2 minutes, after which their configuration disappears and does not reappear after shrinking. In some fibers, which could still be measured, the dimensions were at a maximum (Table 1). In fibers which maintained their shape, lateral degradation was evident.

Differences in the reaction of some parts of the fiber upon swelling may be attributed to the unequal degree of aging of the fiber because the separation of fibers from the late wood of the yearly layer partially removes fibers from the early wood also. The latter fibers contain a characteristically high amount of hemicellulose, which is destroyed to a great extent even at 10 Mrad (Table 1).

THE EFFECT OF RADIATION ON SPRUCE WOOD FIBERS

Wood fibers irradiated at 10 and 30 Mrad react upon swelling just as the control fibers (Table 1). In external appearance, the swollen fibers also resemble the control fibers: the color is yellowish-green and the primary sheathing is ruptured lengthwise into individual threads which do not surpress swelling of the secondary wall. In the fibers there is a transverse striation of the S_1 layer which, apparently, does suppress swelling.

After 50 Mrad dose fiber reactions upon swelling are the same as after 10 and 30 Mrad doses, but the degree of swelling is considerably greater. Swelling takes place outwardly because the primary wall is also ruptured, and, obviously, layer S_1 is weakened, which permits further swelling of the secondary wall. Transverse fissures in the secondary wall were not observed, and the ends of the swollen fiber were intact.

A 70 Mrad radiation dose increases the degree of swelling of wood fiber (Table 1). Swelling proceeds outwardly due to expansion of the lumen since cell wall thickness is almost invariant. This shows that, although layer S_1 is further weakened with increases in radiation doseage, the presence only inhibits and limits the destruction process of cellulose in the secondary layer.

Lateral fissures are observed in the secondary layer of the swollen fibers, which often leads to disintegration of the fiber into different pieces due to the destruction of cellulose in the weakly oriented parts.

The chlorine-zinc-iodine color reaction for all irradiated wood fibers was yellowish-green; but the dark or light-blue color, characteristic of cellulose, was not observed.

INVESTIGATION OF IRRADIATED CELLOLIGNIN FIBERS

Cellolignin fibers were obtained from spruce wood fibers. In order to trace mainly the reaction of lignin, hemicellulose was removed under more strict conditions than usual, viz., by treatment of the wood fibers with 55% sulfuric acid 45 minutes at 60°, after which the fibers were washed with distilled water and placed on glass slides to dry. Cellolignin fiber, obtained according to the stated method, showed high swellability.

The dried fibers were placed in glass capsules and irradiated as stated above.

Upon swelling of the non-irradiated fibers, numerous lateral fissures appeared in their second layer, which sometimes leads to the disintegration of the fiber into separate pieces, but the major part of the fissure extends only to the tubercle of the tertiary layer. The tertiary layer is distinctly separated upon swelling of the fiber (Fig. 1).

Cellolignin fiber, subjected to 30 Mrad, swells somewhat slower than the control fiber. Maximum swelling is reached after 8-10 minutes. Lateral fissures appear suddenly and deepen in proportion to the penetration of the solution inside the secondary wall, and extend to the tertiary sheathing. The tubercle of the tertiary layer remains intact. At the onset of swelling, the whole fiber is yellowish-brown, and after several hours appears bluish-brown: but the primary wall remains yellow, distinctly showing reticulation or interlacing - the natural structure of the primary wall. In all studies of the structure of the cell wall, it has been noted that this reticulation consists of cellulose microfibrils and its cells are filled with hemicelluloses, pectins and lignin. The unusual resistance to various chemicals and the weak swellability of this layer attest to its special properties.

According to our observations, this reticulation is more like lignin in nature (Fig. 2). In the photograph (at the right) it is apparent that in places its threads seem to be separated into individual structural parts, as we observed in the disintegration of lignin threads of the secondary wall (Ref. 10). In the photograph (at the left) changes in the reticulate structure of the primary wall are observed: it becomes disformed and homogenous. This is the result of the action of radiation because the control fibers do not exhibit these phenomena.

The swelling of cellolignin fibers subjected to 50 Mrad drops sharply (Table 1), which, apparently, is explained by the humification of sugars. Many lateral fissures are observed in the secondary layer which extend to the tertiary layer; which, as before, inhibit the disintegration of the fiber into pieces. The fiber upon swelling has a yellowish color from the chlorine-zinc-iodine in the solution, but gradually becomes brownish-blue.

The primary sheathing and the tubercle of the tertiary layer maintain their bright yellow color.

The primary sheathing almost never has a reticulate structure and most often its transformation into a formless mass is observed. Upon being colored with the chlorine-zinc-iodine mixture, it acquires a bright yellow color and after being colored with Sudan III it exhibits the bright orange color of the dye. This color is characteristic of fats, wax, and resin. This indicates that, due to the effect of radiation, variation in the chemical nature and morphological structure of the primary wall takes place.

A 70 Mrad radiation dose does not bring about variations in the degree of swelling of the fiber (Table 1), but the process of variation in the nature of the primary and tertiary sheathing is here more clearly expressed. The primary sheathing in the fibers is found in the form of cohesive tangled threads which, in places, run together, forming drops (Fig. 3).

The chlorine-zinc-iodine reaction gives a yellow color and Sudan III, a red-orange color, just as after 50 Mrad radiation doses. Interesting changes were found in the tertiary layer (Fig. 4). The photograph shows sections which retain the tubercles of the tertiary layer. It is totally covered with numerous, tiny bubbles. In some samples, these tubercles deform into large, swollen projections which, in places, lead to complete dissolution of the tubercles of the tertiary layer and to the formation of drops outside the fiber.

While cellulose of the secondary layer is sufficiently well preserved and, as before, shows the characteristic color (blue) when acted upon by chlorine-zinc-iodine, it is possible that here the protective role of lignin is eminent, inasmuch as lignin is distributed in a uniform network in the secondary layer of coniferous trees, which network gives cellulose its color.

The observation of the tertiary wall is interesting because this part of the cell wall has only recently been the object of study; although in view of numerous technological processes which deal with materials like wood, the tertiary wall plays a no less significant role than the primary. It has been cited earlier in many investigations of the cell wall of wood (Nechesanyy /Ref. 11/,Wadrop /Ref. 12/et al) that the tertiary sheathing is highly ligneous; and this is also confirmed by the new studies of Clark (Ref. 13) where it is stated that lignin reaches the same density in the tertiary walls of wood cells as in the middle layer. However, the position of lignin in the tertiary sheathing has not been clarified in recent years.

In a number of wood species the tertiary layer on the lumen side has a special outgrowth, like a beard. This was first described by Kobashi and Utsumi, and then by Liese (1951). But the most detailed study of the development of the tufted layer, its optical properties, solubility, and color reactions was carried out by Wardrop in 1959 (Ref. 14). He established that the tufts--these are spherical bodies, 0.1-0.5 microns in diameter located on the internal surface of the tertiary wall--completely, or almost completely, foliate the cell and are covered with membranes; they coat the lumen and have a thickness of 0.06 microns. Investigation showed that ultraviolet was strongly absorbed by the tufts (while the membranes themselves did not absorb) which indicates that these formations may be ligneous in nature. Solubility tests of the tufted formations confirmed this supposition. Color reactions (phloroglucinol with hydrochloric acid) of the tufts gave a bright red color, while the membrane remained colorless.

Electron microscope studies by Liese (Ref. 15, 16) on arboreous and herbaceous plants showed that the age tracheids of the majority of conifers are covered on the inside by tufted layers.

In many deciduous species and herbaceous plants, this layer has granular structure. The tufts develop in the last stages of cell differentiation from the remains of the protoplast.

The special character of the tertiary sheathing was observed earlier by us upon studying the effect of different temperatures on the cell wall of wood (Ref. 17). It was shown that the tertiary layer at high temperatures becomes resistant to further heating and to the action of acidic hydrolyzing reagents, and may be separated from the fiber in the form of a separate tubercle. We attributed these properties of the tertiary sheathing to the special structure of cellulose in this layer.

Observation of cellolignin fibers before and after irradiation indicates the indisputable presence of ligneous substances in the tertiary layer, which determine the special properties of this layer under the effect of radiation.

TABLE I

A) B) Процессы, характеризующие набухание	C) Для контрольных волокон	D) Интегральные дозы облучения, Мрад			
		10	30	50	70
E) Увеличение поперечной ширины набухших волокон в % от ширины сухих, принятой за 100 %					
F) целлюлоза	132	215	406	408	413
холоцеллюлоза	189	290	419	398	425
древесина	218	220	227	267	[illegible]
целлолигнин	486	—	399	311	308
G) Увеличение толщины клеточных стенок в % от их толщины в сухом состоянии, принятой за 100%					
H) целлюлоза	221	339	710	689	669
холоцеллюлоза	264	500	589	662	737
древесина	288	285	296	328	339
целлолигнин	800	—	663	507	510
I) Увеличение ширины полостей набухших волокон в % от их ширины в сухих волокнах					
J) целлюлоза	66	103	133	214	224
холоцеллюлоза	52	130	185	187	165
древесина	182	137	165	187	275
целлолигнин	216	—	180	141	130

A) Variation in general width and thickness of cell walls and lumen width during the swelling of the irradiated fibers in an acid solution of glucose.

B) Processes characterizing swelling.

C) Control fibers.

D) Integral radiation dose, Mrad.

E) Increase in lateral width of swollen fibers in % of dry fiber width, taken as 100%.

F) (Downward) Cellulose, holocellulose, wood, cellolignin.

G) Increase in cell wall thickness in % of their thickness in the dry state, taken as 100%.

H) Same as F).

I) Increase in lumen width of swollen fibers in % of their width in dry fibers.

J) Same as F).

FIGURE A

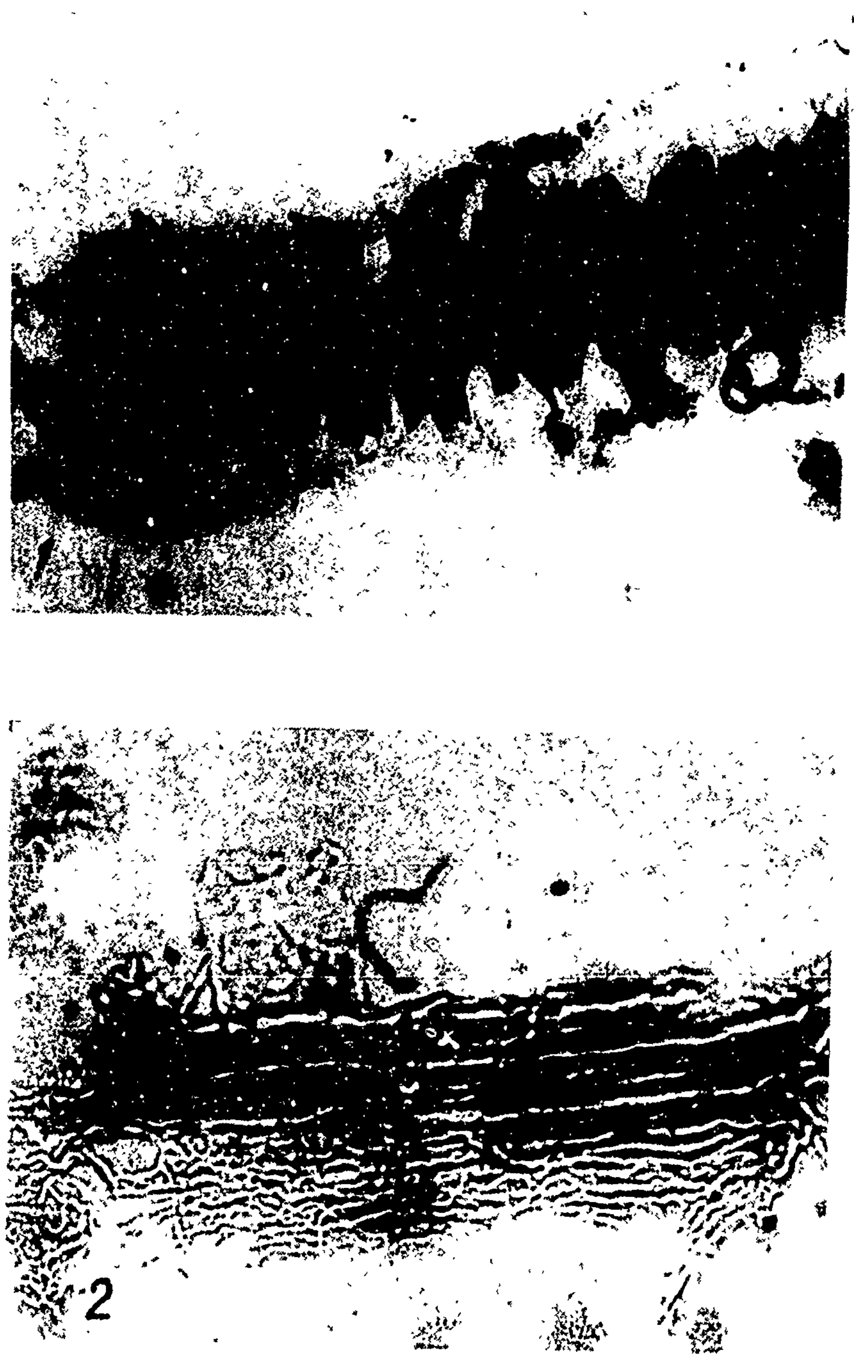

Fig. A: 1 - Control (non-irradiated) fiber of cellolignin, swolien in an acid solution of glucose. x680;
2 - Part of the primary sheathing of cellolignin fiber irradiated at 30 Mrad. Disintegration of the network into individual and particles. x780.

FIGURE B

Fig. B: 3 - Cellolignin fiber irradiated at 70 Mrad. Threads of the primary wall network form bundles, with places that take on the shape of individual drops. x680;

4 - Part of the tubercle of the tertiary layer of cellolignin fiber irradiated at 70 Mrad. The entire tubercle has become blistered. x780.

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13. ABSTRACT

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