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THE MORPHOLOGY, CHEMISTRY AND PULPING CHARACTERISTICS OF REACTION WOOD

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Abstract

In this paper, 'reaction wood' has been described and reference made to its wide occurrence in forest trees and the reasons for such occurrence. It has been emphasised that there may be all gradations from mild to severe reaction wood formation depending on the nature and intensity of the stimuli responsible. The macroscopic and microscopic features of the reaction wood of softwoods (compression wood) and that of hardwoods (tension wood) have been recorded. Particular attention has been paid to the variations in cell wall organisation found in the two types of reaction wood and comparisons have been made with that of normal wood. Ways in which the cell wall organisation might influence properties of both wood and pulp have been discussed. The chemical composition of reaction wood differs from that of comparable normal wood and these differences are particularly marked where the reaction wood is most severe. Compression wood is

higher in lignin content and lower in cellulose content than normal wood of the same tree; the reverse is the case with tension wood and, in addition, the pentosan content is much lower. By staining techniques and ultra-violet microscopy, it has been shown that the cell wall of compression wood fibres is highly lignified and that of tension wood fibres is virtually unlignified. The lower pentosan content of tension wood has been correlated with the very poor papermaking qualities of chemical pulps prepared from it. On the other hand, the higher lignin content of compression wood does not apparently interfere with either the preparation or the properties of the chemical pulps prepared from it. although such pulps contain considerable quantities of lignin (in the cell wall). The association of cell wall deformations with the development of reaction wood has been referred to and it has been pointed out that such deformations are a source of weakness in pulps prepared by acid pulping methods. It was observed that tension wood produces a mechanical pulp much superior to that obtained from normal wood: on the other hand, compression wood gives a very poor mechanical pulp. Numerous investigations carried out have established that, although the presence of certain amounts of reaction wood adversely affects pulp strength properties, the decrease in strength, from a practical aspect, at least in the case of alkaline pulps, is not too great for most purposes.

Finally, it has been stressed that, for the best integration of industries, those logs without reaction wood should be reserved for peeling and sawing, because in the converted timber the presence of reaction wood is likely to be extremely troublesome. This means, of course, that those logs in which reaction wood is present must go to the pulpmill, but here they can be converted into useful pulp by alkaline processes, although the strength of such pulp may be reduced somewhat, depending on the severity of the reaction wood present.

Introduction

The term *reaction wood* is not one that is commonly encountered in the pulp and paper industry. It is important, therefore, to set out clearly what is meant by it.

In recent years, wood technologists (see Dadswell and Wardrop⁽¹⁾) have accepted it as a general term applicable to the wood, formed by hardwoods and softwoods, which is anatomically different from normal wood, but which is developed typically in parts of leaning or crooked stems and in

branches undergoing movements of orientation. The specific term applied in the case of hardwoods is *tension wood* and that applied in the case of softwoods is *compression wood*. Both of these terms are probably better known than reaction wood. These tissues are formed under certain environmental, particularly gravitational, influences and partly as a result of intrinsic factors and are associated with the development of considerable stress gradients.

Reaction wood formation usually involves eccentric radial growth of the stem; in hardwoods, this is on the upper side of leaning stems and branches, whereas in softwoods it is on the lower side. It should be appreciated that reaction wood is of fairly wide occurrence; in any forest, either of hardwoods or softwoods, there is a considerable proportion of trees showing eccentric growth and external evidence of its presence in the form of crooked or leaning stems. In addition, since reaction wood is developed as part of an orientation mechanism (governing the relation between stem and branch under changing environmental conditions), it is commonly found in the early growth rings of stems and branches. Because of its wide occurrence, reaction wood is often found in sawn timber and in much of the material sent to the pulpmill. For this reason, then, it is necessary to know something of its structure and behaviour in comparison with normal wood and, most important, its influence on the properties of the pulp made from wood containing it.

Tension wood and compression wood are quite different in detailed structure, although they appear to perform a similar function and have some properties in common. In the following discussion, when possible they will be treated together, but for the most part it will be necessary to refer to each separately.

Structure and properties of reaction wood

Macroscopic appearance

Compression wood is generally much darker in colour than normal wood of the same species, varying from a definite brown to a dark reddishbrown and, because of this, together with the wider growth rings, can often be readily detected on a freshly cleaned log cross-sections (Fig. 1). In those species with distinct growth rings, the late wood in the compression wood areas is wider than normal, but there is a lack of contrast between it and the early wood of the same ring. In species without distinct growth rings such as certain of the softwoods from tropical areas (for example, *Araucaria klinkii, Cupressus macrocarpa*), compression wood may appear as a solid band of darker coloured wood extending over a wide area of growth.

Varying degrees of compression wood may be observed ranging from that with characteristics outlined above to that which can be distinguished only with difficulty from normal wood. Thus, it is not always possible to be certain of the presence of compression wood on the basis of macroscopic features alone and it is necessary to study its anatomy microscopically. However, when it is not easily detectable, the amount present is unlikely to cause any difficulty in utilisation as pulpwood.

Tension wood is not always as conspicuous as compression wood in log cross-sections. Admittedly, eccentricity of growth and wider growth rings are often indications of its presence, but not always so. As with compression wood, definite bands have been observed in the case of a number of Australian and tropical hardwoods. These bands are darker in colour. harder and denser than neighbouring normal wood (Fig. 2); however, in a large number of species, both from temperate and tropical regions, tension wood, although present, is not easily detectable macroscopically on crosscut surfaces. Sometimes, as in beech,⁽²⁾ it appears as a silvery zone, rather more lustrous than the remainder of the surface. $Clarke^{(2)}$ showed that certain stains such as phloroglucinol in hydrochloric acid could be used to make it more clearly visible. Although these stains were satisfactory for beech, they have not been found satisfactory in the case of a number of species known to contain tension wood. In most cases, its presence is revealed by the extreme woolliness of sawn longitudinal surfaces of green timber. This feature can be extremely troublesome in sawing, because the fibrous mass may choke the saw cut and cause overheating of the saw (Fig. 3). The woolliness of veneer surfaces is also indicative of its presence. Another feature that often aids its macroscopic detection is its tendency to collapse on drying (Fig. 4). This collapse is of the non-recoverable type.

Anatomy

Compression wood — The most characteristic features are the typical rounded appearance of the fibres* in cross-section (Fig. 5), the consequent presence of numerous intercellular spaces between individual cells and the numerous radial discontinuities in the cell wall, which appear as striations in longitudinal sections (Fig. 6). These striations parallel the micellar orien-

^{*} In this paper, both the tracheids of softwoods and the fibres of hardwoods have been referred to as fibres.

tation in the cell wall and are especially apparent in the regions of bordered pits. As might be expected from our knowledge of the development of reaction wood, there are all gradations between severe compression wood (100 per cent. compression wood) and normal wood. When mild compression wood is present, the only anatomical evidence is the presence of the checks, referred to above, extending from the pit apertures. These are readily seen on microscopic examination of radial longitudinal sections or of isolated fibres. By means of them, compression wood fibres in a pulp can be easily detected (Fig. 7).

Fibres from compression wood are on the average shorter than those from comparable normal wood. This has been shown in the examination of fibre length variation through successive growth rings from the pith⁽³⁾ in cases when bands of definite compression wood were present. In this work, last formed late wood from each growth ring was used. Although normally there is an increase in cell length through a growth ring from first formed early wood to last formed late wood, when compression wood is present the cell length decreases.⁽⁴⁾ This is related to the great increase in the radial growth rate and to the fact that compression wood is most often found in the late wood zones.

Detailed information on the fine structure of the cell wall of compression wood fibres in comparison with that of normal wood fibres is given below.

Tension wood — The main anatomical features are fibres with a characteristic thick, highly refractive, inner layer often termed a gelatinous layer, which gives a cellulose reaction with various stains; marked reduction in size and number of vessels; and higher average density than normal wood. Tension wood fibres, however, are not always easy to detect, even microscopically, particularly where normally the fibres are thick-walled, but this may be achieved by the use of selective staining techniques. We have used, in general, safranin followed by light green for this purpose; the unlignified cell wall lavers of the tension wood fibres stain green and the lignified layers red. Using this method, it has been found that the amount and distribution of tension wood varies considerably. In certain cases, tension wood fibres are present in definite bands (compact type, Fig. 8); in other cases, they are mixed more or less indiscriminately with normal wood fibres (diffuse type) and may constitute from a few per cent. to well over 50 per cent. of the wood (Fig. 9). This latter type of distribution appears to be normal in timbers of certain families, including Leguminosae, Anacardiaceae, Combretaceae, Lauraceae, Moraceae, Salicaceae and others. Two genera of particular interest from the pulping angle — namely, *Populus* and *Acacia* — show this more or less irregular distribution of tension wood fibres.

There has been no general agreement in the literature on the length of tension wood fibres compared with those of comparable normal wood fibres. Our work has shown, however, that this may be greater, equal to or less than that of normal wood fibres and that these differences may be correlated with the rate and duration of seasonal growth at the time of the tension wood formation.^(5,6)

The appearance of isolated tension wood fibres is very little different from that of normal wood fibres; however, staining reactions for cellulose (for example, iodine followed by sulphuric acid) can be used to detect them in a pulp.

Cell wall organisation

For the more complete understanding of the behaviour of reaction wood and of fibres isolated from it, knowledge of cell wall organisation is essential. From the standpoint of development, two distinct structures can be recognised in the cell wall of mature fibres of both hardwoods and softwoods, namely, the primary wall and the secondary wall. The former is present during the phase of surface enlargement of the cell in its differentiation from the cambium and the latter develops after surface enlargement has ceased, at least locally, in the cell. In the mature fibre, the primary wall is usually heavily lignified, the cellulose framework occupying a relatively small fraction of the volume. It is not proposed here to discuss the primary wall in any detail, but to record only that its organisation appears to be the same in both normal and reaction wood fibres. After pulping, it separates readily from the fibres on slight beating and appears as an extremely tenuous membrane, which contributes to the fine debris of beaten pulps.

The secondary wall is a structure of great complexity and here there is considerable divergence between normal fibres and the fibres of reaction wood. The fine structure of each has been discussed in some detail below, but, in the first instance, reference has been made to the cell wall organisation of normal wood fibres.

Normal wood fibres — In transverse section, normal fibres show threelayered optical heterogeneity when viewed between crossed nicols (Fig. 10). This reflects the orientation of the microfibrils in the three layers here designated the first or outer layer (S1); second or middle layer (S2); and third or inner layer (S3) of the secondary wall (Fig. 11). The outer layer S1 possesses an organisation of considerable complexity; in many respects it











Fig. 11—Diagrammatic representation of the organisation of the secondary cell wall in—(a) normal wood fibres; (b) compression wood fibres; (c), (d) and (e) various types of tension wood fibres S1—S4=successive layers of the secondary wall G=gelatinous layer A line below the designation of any layer indicates that it is lignified

shows the characteristics of a simple helix oriented at a large angle to the longitudinal cell axis.⁽⁷⁻¹⁰⁾ In other respects, however, it appears to possess some kind of crossed fibrillar organisation.⁽¹¹⁻¹⁶⁾ The structure illustrated in Fig. 12 taken from Wardrop⁽¹⁷⁾ appears to reconcile these conflicting aspects, especially as the microfibril bundles of S1 adjacent to layer S2 may be elaborated to form a continuous lamella. The layer S2 possesses a simple helical organisation, the microfibrils being normally at a small angle to the longitudinal cell axis. This layer is lamellated as shown by Bailey,⁽¹⁸⁾ the individual lamellae being of the order of thickness of a microfibril (Wardrop and Dadswell ⁽¹⁰⁾). The inner layer S3 also appears to be helical, the microfibrils making a large angle to the longitudinal cell axis. There does, however, seem to be some dispersion about this direction as shown in the electron micrographs of Hodge and Wardrop,⁽¹¹⁾ Fischbein,⁽¹⁹⁾ Liese⁽²⁰⁾ and Harada.⁽²¹⁾

Compression wood fibres — The cell wall organisation of typical compression wood fibres can be considered as a modification of that of normal wood cells and is illustrated in Fig. 11(b). The inner layer S3 is absent, so that the normal three-layered appearance is not seen between crossed nicols and there are some other differences in organisation. The layer S1 is usually wider than in normal cells and the layer S2, while possessing a helical organisation, shows numerous regular radial discontinuities. The organisation of the layer S2 may be visualised as consisting of a series of helically wound ribbons attached edgewise to the inside of the layer S1. In longitudinal section, this gives the cell wall the appearance of being conspicuously striated (see reference earlier). This interpretation of the structure of S2 in compression wood has been reported by Jaccard and Frey,⁽²²⁾ Münch,⁽²³⁾ Wardrop and Dadswell⁽³⁾ and more recently by Harada.⁽²⁴⁾ It has been recognised that the width of the radial discontinuities varies greatly with the species for example, coarse in *Pinus*, but extremely fine in *Araucaria*.

The organisation of the layer S1 is similar to that of normal wood as shown in Fig. 12. Some evidence of this may be gained from Fig. 13 and 14, the former showing an isolated part of the layer S1 with typical intersecting bands of microfibrils and the latter showing a longitudinal section with two displaced segments of the layer S2 and the inner surface of the layer S1.

The distribution of lignin in the cell wall of compression wood has been studied qualitatively by Bailey and Kerr,⁽²⁵⁾ Dadswell and Ellis⁽²⁶⁾ and Wardrop and Dadswell,⁽³⁾ quantitatively by Lange.⁽²⁷⁾ It is of interest that between the layers S1 and S2 there appears to be a considerable concentration of lignin as shown in the ultra-violet photomicrograph (Fig. 5). The texture of the radial discontinuities is also shown in this figure and is reflected in the lignin pattern obtained when cross and longitudinal sections are treated with 72 per cent. sulphuric acid (*see* Plate II, Fig. 1 - 4 from Wardrop and Dadswell⁽³⁾).

The cell wall organisation described above is reflected in the behaviour of the fibres on pulping and beating. The removal of lignin from between the layers S1 and S2 results in the loosening of the layer S1, which then lies as a sleeve around the fibre (Fig. 15). On severe beating, the ribbon-like arrangement of S2 becomes unravelled into very coarse fibrils (Fig. 16). Where compression wood fibres fail in tension, the unwinding of the layer S2 often occurs (Wardrop⁽²⁸⁾).

Tension wood fibres — In these fibres, the cell wall organisation differs from the typical structure of the normal cell by the presence of the unlignified, so-called gelatinous layer, which is present in addition to layers S1, S2 and S3 or replaces layer S3 or replaces both S2 and S3. In this additional layer, the microfibrils are axially oriented. Possible variations in cell wall organisation of tension wood fibres as described by Wardrop and Dadswell⁽²⁹⁾ are shown in Fig. 11 (that organisation illustrated by Fig. 11(d) was first described by Jaccard and Frey⁽²²⁾). In these diagrams, a line under the designation of any particular layer indicates that it is lignified; it should be further noted that the axially oriented layer is in every case adjacent to the lumen. The latter point is interesting because in a recent paper by Jutte⁽³⁰⁾ tension wood fibres have been illustrated, apparently corresponding to the type (c) or (d) of Fig. 11, but with a lignified layer inside the gelatinous layer.

Thus, in transverse section between crossed nicols, certain tension wood fibres show the typical three-layered optical heterogeneity with an additional dark layer adjacent to the lumen (type (e)), while others may show only a bright outer layer with a dark layer adjacent to the lumen (types (c) and (d)). It has been noted that the particular type of organisation present in some instances reflects the time of the annual growth cycle at which the cells have been differentiated. For example, in *Eucalyptus gigantea* and *Eucalyptus regnans*, both sources of pulp in Australia, tension wood formed in the early part of the growing season may have the organisation of type (c) and that formed in the last part of the season the organisation of type (d). It has been shown⁽²⁹⁾ that the layer S1 possesses a crossed fibrillar type of organisation similar to that observed in normal cells. Evidence of this may be gathered from Fig. 17, in which the fine grid adjacent to the primary wall can be seen and from Fig. 18, where it appears that this grid is overlain by a coarser grid; at the right of the figure, only one layer of microfibril bundles is present, but at the left both sets can be seen.

The distribution of lignin in the cell wall of tension wood has been studied qualitatively^(29, 31) and is illustrated in Fig. 19. Lange⁽²⁷⁾ has made more detailed investigations using ultra-violet microscopy. From his investigations and our own work, the most striking feature is the comparative absence of lignin in the axially oriented inner gelatinous layer of tension wood fibres. This is in direct contrast to compression wood, in which the cell wall is highly lignified. Layers S1 and S2 may also possess a low degree of lignification in certain types of tension wood. The gelatinous layer consists largely of cellulose, which is highly crystalline, as shown by X-ray diffraction patterns of tension wood in comparison with normal wood of the same tree, ^(29,31,32) by the hydrolysis characteristics of the two types of wood (31, 33) and by density and moisture regain determinations. The high crystallinity reflects the high degree of lateral order of the micelles, which appear to be surrounded by a paracrystalline phase to an extent similar to that in normal wood. This is consistent with the low rate of hydrolysis of tension wood compared with that of normal wood. Viewed in the electron microscope, the gelatinous layer shows the extent of the aggregation of the microfibrils that has occurred on drying, so that individual microfibrils are difficult to recognise (Fig. 20). It is considered that this aggregation is largely irreversible and this may account for the irreversible shrinkage associated with the presence of tension wood. Further, the highly crystalline nature of the gelatinous fibres and the consequent low degree of swelling are reflected in the slow beating rate (as shown by damage to the fibre) of tension wood fibres.

Cell wall deformations

The occurrence of numerous slip planes and minute compression failures in the cell walls of tension wood has been reported in earlier papers. Not only are these deformations found in much of the tension wood itself, but they are found commonly in the wood on the pith side of the tension wood. On the other hand, they are absent or practically so from compression wood, although often observed in the cell walls of wood on the pith side of the compression wood bands and on the opposite side of the tree. The important point is that wood containing these failures is 'acid susceptible'⁽³⁴⁾ and pulps prepared from such wood under acid pulping conditions are likely to contain numerous 'broken fibres'. These are directly traceable to the

minute compression failures referred to and, as may be expected, have a deleterious effect on pulp strength properties. The formation of minute compression failures in tension wood and on the pith side of it, as well as on the pith side of compression wood bands, can be readily understood, if we accept the idea that development of reaction wood involves definite reorientation movements within the stem. In the case of hardwoods, any movement of the stem towards the vertical in response to a stimulus throws the wood of the outer layers of the upper side (that is, just where tension wood has been laid down) into compression. Similarly, in softwoods, the recovery of a stem associated with the formation of compression wood throws the wood on the inside of the compression wood bands into compression again, resulting in the development of minute compression failures.

Wood properties

In spite of the very great differences in cell wall organisation between compression wood and tension wood, both these forms of reaction wood have certain properties in common. Both are higher in density than normal wood, an average ratio being 4:3. Both show abnormally high longitudinal shrinkage, although in this respect compression wood is much worse than tension wood, values as high as 10 per cent. being recorded in this laboratory for samples dried from the green condition to the oven-dry. On the other hand, the maximum longitudinal shrinkage in tension wood recorded here under the same conditions has been approximately 1.5 per cent.

From Fig. 11, it will be seen that the micellar orientation of the gelatinous layer in tension wood fibres is nearly axial, while that in the other layers remains much the same as would be found in normal wood. Thus, on the basis of the usually accepted theories, it is difficult to explain the longitudinal shrinkage. However, it is of interest that, in fibres such as ramie, with a somewhat similar chemical composition to that of the gelatinous layer of tension wood fibres, shrinkage in the direction of the length is about 1 per cent. The lack of lignification in cell walls of tension wood, together with the highly crystalline nature of the cellulose may be important factors determining the longitudinal shrinkage, though these views do not take account of the layer S1, which may also be important. In this layer, the angle of micellar orientation varies $50^{\circ} - 90^{\circ}$ to the cell axis and should therefore greatly favour shrinkage in the longitudinal direction. Normally, the layer SI is narrow in relation to S2 and here the angle of orientation of the microfibrils is generally small in respect to the cell axis and so the component of longitudinal shrinkage is small. Thus, the layer S2 tends to restrain and nullify any longitudinal shrinkage of S1. In tension wood, however, this restraint is probably greatly diminished because of the lack of lignification and the shrinkage may be attributed to the nature of the gelatinous layer, together with the effect of S1.

In the case of compression wood where the longitudinal shrinkage is much greater, the layer S1 is somewhat wider than in normal wood, has a micellar orientation nearly transverse to the longitudinal cell axis and apparently is not highly lignified (Fig. 5). Therefore, it could be expected to exert considerable influence on longitudinal shrinkage and, in this case, such shrinkage is not greatly restrained by the layer S2, which, in compression wood, consists of numerous separate helical ribbons in which the micellar orientation to the longitudinal cell axis is large (Fig. 11). This type of structure with its radial discontinuities would not be expected to exercise nearly as much restraint on the shrinkage of S1 as the normal organisation of S2.

The characteristic collapse in tension wood observed in drying has been referred to earlier. This is also associated with cell wall shrinkage and the lack of cell wall lignification. The irreversibility of this shrinkage is probably due to some degree of hydrogen bonding between lamellae in the cell wall. Such hydrogen bonding could not take place in normal wood where the lamellae of the layer S2 are separated by lignin and other substances. What may be analogous to irreversible collapse has been observed in another unlignified fibre — cotton. This fibre has a circular cross-section when removed from the boll, but on drying collapses into a ribbon-like form and does not recover on soaking in water or boiling.

In strength properties, compression wood is normally weaker than would be expected for its density. This is particularly so in the case of modulus of elasticity, impact strength and tensile strength. Tests in this laboratory have not shown it to be any stronger in compression than normal wood of the same density (*see* also Pillow and Luxford⁽³⁵⁾). In the green state, tension wood is much lower in compression strength than might be expected for wood of the same density, but the difference is much less marked in dry material. The presence of minute compression failures makes tension wood particularly weak in impact strength.

Chemical composition

The chemical composition of reaction wood has been investigated by numerous workers. It is extremely difficult to obtain comparable normal wood and it has long been recognised that the wood on the side of the tree opposite the reaction wood is not normal in either properties or chemical composition. In our investigations, we have usually taken wood from the same growth rings, but where possible at rightangles to the reaction wood zone or, alternatively, before the reaction wood has formed. It has been found that the wood from the same growth rings on the opposite side of the tree has lower lignin content and higher cellulose content than the side wood.

Early studies by Johnson and Hovey⁽³⁶⁾ on balsam fir and Dadswell and Hawley⁽³⁷⁾ on a number of North American softwoods showed that compression wood had substantially higher lignin content and lower cellulose content that comparable normal wood. Similar results have been obtained on a wide range of other softwoods by later workers. Staining techniques and ultra-violet microscopy (see Lange ⁽²⁷⁾ and Fig. 5) have indicated that the extra lignin in compression wood is located in the secondary cell wall. Some results recently obtained with various zones taken from the *Pinus radiata* sample shown in Fig. 1 are given in Table 1.

Not nearly as much information on the chemical composition of tension wood is available in the literature. Wardrop and Dadswell⁽³¹⁾ published data on tension wood of *Eucalyptus regnans* and Jayme and Harders-Steinhauser⁽³⁸⁾ have reported results for the tension wood in various species of poplar. It would appear that tension wood is much higher in cellulose and lower in lignin and pentosan contents than is comparable normal wood. Recent results obtained with different eucalypt samples are given in Table 2. As would be expected from the studies of cell wall organisation by X-ray diffraction and electron microscopy (*see* reference above), the tension wood is much higher in alpha-cellulose content.

It would seem that in reaction wood there is a general pattern throughout, whether it be tension wood or compression wood and, in any one cross-section, the highest lignin content and lowest cellulose content are on the lower side.

Bland (unpublished) has examined in this laboratory the nature of reaction wood lignins and has shown that they give the same principal oxidation products and therefore have the same basic structure as normal wood lignins. Tension wood lignin and compression wood lignin differ, however, in that such lignin appears to have slightly modified side chain structure, which is also different from that in normal wood lignin. With reference to the carbohydrate fraction, Schwerin (unpublished) has shown that reaction wood from *Eucalyptus goniocalyx* and *Pinus radiata* has five to six times as much galactose as comparable normal wood. In these species, all the galactose in the normal wood, half the galactose in the compression

wood and one quarter of the galactose in the tension wood, remained in the alpha-cellulose — that is, for both species, the non-cellulosic polysaccharides of reaction wood contained substantial amounts of galactose, whereas those from comparable normal wood contained no galactose.

Chemical analysis also shows up the gradation between normal and reaction wood and variations in composition between the two extremes are possible in the one tree. These differences are correlated with the severity of reaction wood development in the tree.

Pulping characteristics of reaction wood

The effect of reaction wood on pulping and pulp strength properties has been examined by a number of workers; for example, it has been reported⁽⁴⁰⁻⁴²⁾ that the presence of compression wood results in low yields of pulp when wood containing it is pulped by acid or alkaline processes. Moreover, the pulps themselves have lower strength properties and are difficult to bleach. The presence of compression wood also adversely affects the strength properties of groundwood. In recent years, some work has been done on the effect of tension wood and this, too, has been shown to have an adverse effect on pulp strength properties (see below). Generally speaking, these various investigations have been carried out on material that was a mixture of normal wood and reaction wood and consequently it is difficult to determine from the results whether chemical composition or cell wall organisation -or both-are correlated with pulp strength properties. Therefore, of some interest are results of our work in which what might be termed 100 per cent. reaction wood has been treated by various processes in comparison with other wood zones from the same tree in each case.

In the case of compression wood, material from the tree, of which a cross-section is shown in Fig. 1, was used. Various zones, including the area that was shown to be all compression wood, were cooked separately by the sulphate process, using 20 per cent. total alkali and well-cooked, shive-free pulps were obtained from all samples. The yield from the compression wood was low (Table 1) and the pulp had a high lignin content. Evaluation of pulps in the standard manner revealed the lower strength properties of that from the compression wood (Table 3). It was considered that this might be related to difficulties in developing strength in a pulp of such high lignin content. However, when the lignin content was reduced considerably either by bleaching the pulp or by cooking the wood with a high percentage total alkali (25 per cent.), there was little change in pulp strength properties.

Thus, it seemed clear that the lignin content could not account for the lower strength observed. On the other hand, it seemed probable that the shorter, stiffer, thicker-walled compression wood fibres were associated with the low strength properties, especially as it was observed that these fibres

Yield of sulphate pulp Average Lignin content Pentosan content (prepared by length of Specimen cooking whole fibres with 20⁵ in pulp, total alkali), Pulp, Wood, Pulp, %⁽²⁾ mm. Wood, %(²) %(1) %(1) %⁽¹⁾ 2.76 7.2 Compression wood ... 40 34.4 11.6 8.3 Normal wood (formed before development of compression 48 3.85 24.2 2.8 10.0 8.7 wood) . . Opposite wood 48 3.51 25.8 5.4 8.8 7.7 . . Side wood ... 50 3.55 25.5 4.9 8.9 7.9 . .

CHEMICAL COMPOSITION OF WOOD AND PULP FROM VARIOUS ZONES IN ONE TREE OF PINUS RADIATA

TABLE 1

(1) Results calculated on an oven-dry wood basis

(2) Results calculated on an oven-dry pulp basis

TABLE 2

CHEMICAL COMPOSITION OF TENSION WOOD FROM DIFFERENT ZONES OF VARIOUS EUCALYPT SPECIES IN COMPARISON WITH THAT OF NORMAL WOOD OF THE SAME SPECIES

Species	Type of wood	Soda pulp yield, %	Lignin content of wood, %	Pentosan content of wood, %
E. goniocalyx	Tension	67.2	10.3	11.2
E. goniocalvx	Normal	50.0	25.2	19.7
E. regnans Tree 1	Tension	58.8	15.8	9.3
E. nitens	Tension		14.6	11.0
E. nitens	Normal	-	21.3	16.7
E. regnans Tree 2	Tension	68.1	12.4	9.7
E. regnans Tree 2	Normal	55.1	21.4	17.3

Results calculated on an oven-dry wood basis

did not fibrillate as readily on beating as did the non-compression wood fibres of the same tree (Fig. 21 and 22).

Lower yields of pulp comparatively high in lignin were also obtained from compression wood by the sulphite process. With this method of cooking, the zones opposite the compression wood also gave pulps of low strength properties. This has been associated with the presence in these zones of

 TABLE 3

 STRENGTH PROPERTIES OF SULPHATE PULPS MADE FROM COMPRESSION WOOD AND FROM OTHER ZONES OF P. RADIATA, A CROSS-SECTION OF WHICH IS SHOWN IN FIG. 1 (Pulps beaten in Lampén mill)

Wood zone	Type of pulp	Beat- ing, rev.	Bulk	Break- ing length, km.	Stretch	Burst factor	Tear factor	Air resist- ance, sec.	Fold (Köh- ler- Molin)	Free- ness, C.S.F.
Com- pression wood	Sulphate, 20% total alkali	0 4 500 9 000 18 000 36 000	1.96 1.59 1.53 1.47 1.41	3.9 6.6 7.6 7.8 8.4	3.4 4.7 4.6 4.3 4.4	34 54 59 67 69	164 129 113 110 99	<0.5 <0.5 0.9 5.8 150	221 830 902 946 1274	725 720 683 505 218
Com- pression wood	Sulphate, 25% total alkali	0 4 500 9 000 18 000 36 000	1.75 1.57 1.50 1.45 1.40	3.7 6.0 7.2 7.0 8.2	4.3 4.5 4.2 4.5 3.9	45 54 62 62 62	165 144 119 106 98	<0.5 <0.5 5.8 4.6 58.0	947 648 916 1116 1226	707 708 652 464 222
Com- pression wood	Sulphate, 20% total alkali, bleached	0 4 500 9 000 18 000 36 000	1.59 1.44 1.45 1.42 1.46	4.8 7.6 8.0 8.1 9.0	6.1 4.9 5.1 4.4 3.4	51 72 72 71 71	196 152 142 137 118	1.2 1.2 1.6 3.0 15.6	2003 2010 2680 2080 1623	692 690 632 515 535
Opposite wood	Sulphate, 20% total alkali	0 18 000	1.72 1.35	6.4 10.5	3.6 4.6	57 89	209 115	3.0 80.8	2032 2427	680 430
Side wood	Sulphate, 20% total alkali	0 4 500 9 000 18 000	1.76 1.46 1.42 1.38	6.4 9.6 10.1 10.9	3.7 4.2 4.2 4.7	56 84 92 98	212 138 129 111	3.2 4.0 9.8 49.1	1440 1183 1686 2525	702 694 644 445
Normal wood	Sulphate, 20% total alkali	0 4 500 9 000 18 000	1.84 1.53 1.46 1.40	5.8 10.0 11.0 10.9	2.4 3.4 3.8 3.7	40 76 85 91	244 153 136 117	1.3 2.4 5.8 79.1	1782 2505 3089 2306	726 704 631 336

numerous slip planes and minute compression failures, which are particularly susceptible to acid treatment (Wardrop and Dasdwell⁽⁴³⁾). Green and Yorston⁽³⁴⁾ referred to 'acid susceptible' wood and such wood was always present on the pith side of compression wood bands, where numerous cell wall deformations are known to be present. There is little doubt that these deformations give rise to broken fibres on acid cooking, with the resultant low strength properties observed by Green and Yorston and referred to here. This is, therefore, one aspect of compression wood occurrence that must be considered, if acid pulping processes are to be used.

TABLE 4	
STRENGTH PROPERTIES OF SODA PULPS MADE FROM TENSION WOOD AND NO OF <i>E. REGNANS</i> (TREE NO. 2)	ORMAL WOOD

Beating, rev.	Bulk	Burst factor	Stretch %	Breaking length, km.	Tear factor	Air re- sistance, sec.	Fold (Köhler- Molin)	Free- ness, C.S.F.
Tension wood 0 1 125 4 500 9 000 18 000 36 000	2.35 2.18 2.00 1.76 1.69 1.65	6 14 18 22 25 30	0.6 1.0 1.7 1.9 2.3 2.7	2.1 3.5 4.0 4.5 4.8 5.3	26 45 56 68 71 76	<0.5 <0.5 <0.5 <0.5 1.2 7.6	1 2 4 5 9 16	672 589 560 527 420 197
Normal wood 0 1 125 4 500 9 000 18 000	1.85 1.69 1.57 1.53 1.44	30 40 52 61 66	1.7 2.2 2.6 2.7 2.9	5.5 7.5 8.5 9.5 10.4	96 108 124 130 116	3.2 5.0 6.9 9.0 17.1	22 45 85 186 454	544 470 434 390 322

TABLE 5

STRENGTH PROPERTIES OF MECHANICAL PULPS MADE FROM TENSION WOOD AND NORMAL WOOD OF E. REGNANS AND E. NITENS

Species and type	Bulk	Burst factor	Stretch, %	Break- ing length, km.	Tear factor	Air resist- ance, sec.	Fold (Köhler- Molin)	Free- ness, C.S.F.
E. nitens —tension wood	3.18	9	1.4	1.6	36	8	2	220
E. nitens —normal wood		Sheet	s could n	ot be co	uched f	rom she	etmachin	e
E. regnans—tension wood	2.90	11	1.8	1.7	59	3	4	506
E. regnans—normal wood		Sheet	s could n	ot be co	uched f	rom she	etmachin	e

Jayme and co-workers^(44, 45) have investigated the influence of tension wood on the pulping characteristics of poplar, using the sulphate process. They have shown that the yield of pulp increased as the percentage of tension

wood increased from 22 per cent. to 71 per cent. Watson has recently published results obtained in the investigation of the influence of tension wood on the pulping of eucalypt timbers.⁽⁴⁶⁾ Here, the soda process was used and, in the case of specimens from trees of several different species of eucalypts, 100 per cent, tension wood was pulped separately from normal wood of the same tree. Pulp yields were much higher for the tension wood than for the normal wood and the tension wood pulps had much inferior strength properties (see results for material from one tree of *Eucalyptus regnans* in Table 4). It is particularly noticeable that the tension wood pulps are deficient in those properties associated with fibre bonding and this is related to the low pentosan content of these pulps. The gradations of development of tension wood are reflected both in the structure of the fibres and the chemical composition of the wood. As might be expected, where the most severe development has occurred (tree 2), the lowest lignin and lowest pentosan contents are found (for example, E. regnans tree 1-20.5 per cent. and 12.3 per cent., respectively; tree 2-12.2 per cent. and 9.5 per cent., respectively). Furthermore, the most severe tension wood gives the chemical pulp with the lowest strength properties.

On the other hand, tension wood appears to grind much more readily than normal wood, giving a pulp from which handsheets can be made. In our experiments, pulps from tension wood and normal wood have been prepared by means of the laboratory Bauer refiner, using green chips. In these experiments, both *Eucalyptus nitens* and *Eucalyptus regnans* were used and here the ability to form suitable handsheets and the best pulp strength properties were associated with the most severe type of tension wood as determined by microscopic examination and chemical composition (Table 5). The ease with which his type of tension wood could be separated into fibres by mechanical treatment is possibly associated with its lower lignin content or possibly with lower amounts of a lignin-carbohydrate complex associated with fibre bonding. The tension wood being less stiff and more 'rubbery' in nature would tend to rub into fibres, whereas the normal wood with its full component of stiffening material (lignin) would tend to grind into smaller pieces.

It would appear that, whereas in the case of compression wood the pulp strength properties are a function of the dimensions and cell wall organization of the fibre, in the case of tension wood the pulp strength properties are more related to the chemical composition.

Practical considerations

The whole question from the point of view of the pulp and paper industry is whether the presence of reaction wood in the raw material will seriously affect the properties of the pulp and, therefore, of the paper manufactured from it. It must be accepted that, whether the raw material be hardwood or softwood, a considerable amount of reaction wood is likely to be present. In Fig. 23 can be seen the type of stand where the proportion of crooked stems is quite high. With the present day integration of sawmilling and pulping industries, all the straight logs and those without obvious defects should go to the sawmill, while the crooked stems and larger branches should automatically go to the pulp mill. Therefore, there is a great need for the pulp and paper industry through its technical men to assess the effects of such reaction wood as will be present on the quality of the pulp being produced. It is for this reason that we have summarised present knowledge of the structure and properties of reaction wood, given methods for its recognition and discussed the work that has been done to determine its influence on pulping and on pulp properties. The conclusions that can be drawn from these various investigations are as follows.

Both types of reaction wood can be pulped satisfactorily under the same conditions as are used for normal wood of the same species. This is particularly important in commercial pulping, as it avoids the necessity of attempting to segregate reaction wood and normal wood during pulping. However, although reaction wood can be cooked without difficulty to give a shive-free pulp, it is quite clear from the results that both compression wood and tension wood adversely influence the pulp from a papermaking viewpoint.

The deleterious effects of compression wood are most serious in acid pulping processes or in mechanical processes. In the case of the sulphate process, its presence causes some lowering of yields, but the actual reduction in pulp strength is not great. Experiments have been carried out with a 50 : 50 mixture of severe compression wood pulp from Pinus radiata with a commercial sulphate pulp of the same species. In one case, the pulps were beaten together and, in the other, they were beaten separately and mixed before sheetmaking. In both cases, the strength properties of the mixture were intermediate to those of the constituent pulps, but were such that satisfactory grades of paper could be obtained. On the other hand, if the sulphite process is being used, the presence of compression wood has a much greater effect, particularly because of the association with it on the pith side-and sometimes on the opposite side-of the stem of acid-susceptible wood. The tendency for the formation of broken fibres derived from the cell wall deformations (Fig. 24) causes a marked reduction in pulp strength properties. This effect of the cell wall deformation is not observed to any extent with alkaline pulps.

Tension wood has a more adverse effect on pulp properties depending on fibre bonding for strength development. This is due to the lower amount of non-resistant polysaccharides in the tension wood pulp and to the thickerwalled fibres. On the other hand, pulp yields are much higher than with comparable normal wood and the pulps so obtained would have some advantages in the production of high alpha-pulp. Cell wall deformations are present in considerable number in tension wood and these render acid pulping out of the question. The presence of tension wood, particularly of the most severe type, appears to be an advantage as far as mechanical pulping is concerned. We have shown that, with the laboratory Bauer refiner at least, quite fair pulp can be made from green tension wood, but not from green normal wood.

Finally, it must once again be emphasised that there are all gradations from normal wood to the most severe reaction wood. Some of the milder forms of reaction wood are no great disadvantage and for certain purposes may even be an advantage. When the most severe forms of reaction wood are present, however, such that definite bands of it can be detected macroscopically, some loss of strength properties must be expected when alkaline pulping processes are used and a considerable loss with acid pulping processes.

Although it would appear from the pulping viewpoint that the occurrence of reaction wood in the raw material need not, in general, be viewed with too great alarm, it should be emphasised that in an integrated industry care must be taken to segregate those logs containing such reaction wood so that as little as possible goes into sawn timber where its presence might be very troublesome in subsequent utilisation.



Fig. 1—Cross-section of a log of *Pinus radiata* taken from a tree that had been blown over after approximately 10 years of normal growth— note the excessive development of compression wood on the lower side



Fig. 2—Cross-section from a small tree of *Eucalyptus* regnans that had developed a very definite zone of tension wood on the upper side of the stem



Fig. 3—An example of the excessive 'woolliness' that is encountered on the sawing of logs containing bands of tension wood



Fig. 4-A band of tension wood in Eucalyptus regnans that has collapsed



Fig. 6.—A radial longitudinal section of compression wood in *Pinus ponderosa* (\times 125)—note the characteristic striations in the cell wall and the checks extending from the pit apertures

Fig. 7—Compression wood fibres as observed in pulp prepared from *Pinus* radiata compression wood (\times 110)— note the characteristic striations



Fig. 8—A cross-section of *Eucalyptus* regnans showing a band of tension wood fibres at the end of a growth ring $(\times 63)$ Fig. 9—A cross-section of *Nothofagus* menziesii showing the irregular distribution of tension wood fibres $(\times 70)$



Fig. 5—A cross-section of the compression wood from the log illustrated in Fig. 1, showing the characteristic rounded nature of the cells, the intercellular spaces and the lignin distribution—this photomicrograph was taken by means of ultra-violet illumination and the dark coloured zones are indicative of lignin, which absorbs the ultra-violet note the distinct outer layer of the cell (S1) with an apparent concentration of lignin in the outer portion of layer S2 (refer Fig. 11 (b)), also the evidence of radial discontinuities in the layer S2 (\times 660) Fig. 10— An unstained cross-section of *Pinus* radiata, viewed by transmitted light (upper) and between crossed nicols (lower) (×418)



Fig. 12—A wooden model of the first layer of the secondary wall (S1) in wood fibres: this layer consists of two grid systems, the lower (fine) one adjacent the primary wall being overlain by a coarser (upper) grid; the uppermost set of microfibrils in the coarse grid is frequently elaborated into a complete lamella In this model, the cell axis would be approximately diagonal to the grid



Fig. 13—Electron micrograph of part of the outer layer of the secondary wall of a compression wood tracheid of *Pinus radiata* after delignification (uranium-shadowed)

Fig. 14—Electron micrograph of a delignified longitudinal section of a compression wood tracheid of *Pinus radiata* showing two displaced parts of the helically wound layer S2 overlying the layer S1 (uranium-shadowed) (cf. Fig. 12)



Fig. 15—A delignified compression wood tracheid of *Pinus radiata* after mild beating, showing the first layer of the secondary wall (S1) lying as a loose sleeve around the helically wound layer S2: *top* —at lower focus on the layer S1; *bottom*—at higher focus on the layer S2 (aluminiumshadowed) (×784)

Fig. 16—A delignified tracheid of compression wood of *Pinus* radiata after beating, showing the coarse tangled fibrils originating from the layer S2 (aluminium-shadowed) (×706)



Fig. 17—Part of the outer layer of the secondary wall of a fibre from Eucalyptus regnans, showing the fine grid system adjacent the primary wall (uranium-shadowed)



Fig. 19—Cross-section of tension wood from *Eucalyptus elaeophora* taken in ultra-violet light—note heavy absorption in the intercellular zone and the weak absorption in the cell wall, particularly in the layer G ($\times 2$ 000)



Fig. 18—As Fig. 17, with part of the coarse grid system overlying the fine grid (uranium-shadowed)



Fig. 20—Part of the gelatinous layer G (Fig. 11) from a tension wood fibre of *Eucalyptus goniocalyx*



Fig. 21—Normal wood fibres from *Pinus radiata* prepared by the sulphate process and beaten in the Lampen mill for 72 min. (×120)—note swelling and development of external fibrillation

Fig. 22—Compression wood fibres from *Pinus radiata* prepared by the sulphate process and beaten in the Lampén mill for 72 min. $(\times 120)$ —cf. Fig. 21



Fig. 23—A stand of *Pinus radiata* in which numerous leaning and crooked stems are evident



Fig. 24—Pulp prepared by the sulphite process from the heartwood zone of the log illustrated in Fig. 1, photographed between crossed nicols (\times 65)—note number of broken fibres

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DISCUSSION

MR. J. BOS: I want to make some comments on the degree of polymerisation (D.P.) of reaction wood cellulose.

The occurrence of reaction wood in hardwood has been the subject of investigations from different viewpoints. The papermaking properties of pulps produced by the sulphate process from this abnormal wood have been determined especially by Jayme and his co-workers, as well as recently by Watson.

As there is also rather a quantity of poplar pulp produced by the sulphite process, we thought it useful to investigate the influence of reaction wood in poplar on this kind of pulp.

On a number of trunks of *Populus eur. serotina*, the Wood Institute T.N.O. at Delft determined microscopically the distribution of the reaction wood. From these, we selected four with heavy occurrence of reaction wood throughout their length for the pulping experiments. Under standard conditions, with a maximum temperature of 130° c, we prepared an easy bleaching paper grade sulphite pulp from the part of each trunk with reaction wood, as well as from the opposite part without reaction wood. The yield ranged 49 — 51 per cent. for the normal wood on a wood basis and 57.5 — 64.5 per cent. for the reaction wood.

The properties of handsheets of the unbeaten pulps differed considerably. As an example, the breaking length for the pulps from the normal wood was $5\ 000\ -6\ 000\ m$. and $2\ 100\ -3\ 250\ m$. for the pulps from the reaction wood.

There exists a strongly negative correlation between the strength properties of the pulps and the yield. This means that the strength of the pulp depends on the quantity of reaction wood fibres. I think it is not of much use now to mention all the other determinations we made on the wood and the pulp, but I wish to mention one observation that, to our knowledge, has not been published in the literature before. We determined the cuoxam viscosity of the pulps and found that the D.P. showed a correlation with pulp yield in the same way as the strength properties (Table 1, columns 6 and 7).

As it is possible that sulphite pulping causes degradation of the cellulose chains, especially with the reaction wood fibres, we determined also for tree number 6 (with the highest pulp yield from the reaction wood part) the D.P. of a holocellulose prepared from the wood by the acid chlorite method and of the same after extraction with 5 per cent. potassium hydroxide in an atmosphere of nitrogen. The results are given in Table 2.

We think these figures mean that there is, indeed, a certain degree of degradation of the reaction wood by sulphite pulping, so that the difference with normal wood is accentuated; but the results with the holocellulose indicate that it is fairly certain that the D.P. of the alpha-cellulose of the reaction wood fibres is lower than that of the normal wood fibres.

	w	ood (R) o	f four popl	ar trees			
Tree	Yield	d, %	Breaking	length, m.	DP		
number	N	R	N	R	N	R	
13 3 7 6	49.2 51.2 49.1 50.9	57.5 59.5 60.9 64.4	5 150 4 950 6 000 5 950	3 250 2 900 2 750 2 100	1 460 1 730 1 630 1 530	1 120 980 890 760	

TABLE 1Properties of sulphite pulps from normal wood (N) and reaction

TABLE 2Degree of polymerisation of holocellulose from tree No. 6

	DP				
Pup condition	Normal wood	Reaction wood			
Original holocellulose	1 080	930			
After extraction with 5% KOH under nitrogen	1 430	940			

We are continuing our investigations with the determination of the molecular weight distribution curves of the two kinds of wood from the same trunk to obtain more thorough knowledge of the chemical structure of the secondary wall of the reaction wood fibre.

DR. W. GALLAY: With reference to the second paper this morning, I would like to recall to your memory what I consider to be a classical experiment reported by Hägglund several years ago. In the preparation of sulphite

pulp, he used a rotating digester, having a perforated septum down the middle, so that two separate sulphite production experiments could be carried out under the same conditions in so far as liquor, temperature and pressure were concerned. The chips were from the same source. Major differences in strength properties were obtained by varying only the extent of the filling of the digester with wood, far higher strength being obtained when chip movement was reduced to a minimum. Many of you will doubtless recall the work.

It would appear that the explanation cannot be ascribed solely to damage done by extensive defibration of wood in the digester, since the integrity of the chips was apparently well maintained. Some explanation might well be provided by considering a continued mechanical compression and relaxation of the chips. When the latter only partially filled the container, it resulted in a condition allowing undesirable features of access of acid to portions of the structure.

It is pointed out that this is the condition brought about in acid degradation of 'acid-susceptible' wood, following compression or shearing forces. Unfortunately, ordinary chipping as at present practised introduces a great deal of this acid susceptibility, with consequent serious effect on pulp strength.

I wonder in this connection whether our expert microscopists who have reported so effectively at this meeting have ever produced pictures of an actual separation at the fibre wall of the tracheid resulting from mechanical action of this kind, which in turn would allow for access of hot acid to very sensitive elements.

DR. P. LANGE: If you take out chips during different phases of a sulphite cook, make cross-sections from them and inspect them under the microscope, they are very often seen to be split along the radial middle lamella, owing to the cutting action of the microtome knife. It is possible therefore that the chips treated as Dr. Gallay described may easily be damaged in these middle lamella regions. The reason may be that the cooking acid attacks the middle lamella via the pits and, since the pits are mostly to be found on the radial surfaces, these are weakened by the hydrolytic action of the cooking acid, resulting in a decreased resistance to mechanical action.

DR. B. G. RÅNBY: If you look upon sulphite cooking as a process, you are surprised that it is at all possible to obtain a cellulose of such a high degree of polymerisation from it: the temperature is high $(130^{\circ} - 145^{\circ}C)$ and the pH quite low (1.5 - 2) towards the end of the process. If you use ground-wood (that is, newsprint) or even cotton fibres (which are much better

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crystallised) in the sulphite process instead of the wood chips, the cellulose depolymerises very badly. The resulting pulp fibres are degraded and they give paper of low strength. I do think that the reason for this is not only a better penetration of the cooking liquid through the crushed cell walls; it lies deeper than that.

The original, undamaged pulp fibres, without sharp breaks or ruptures, have a structure with the crystalline cellulose well protected as deposited in the fibre wall together with the hemicellulose. According to Lange's measurements, the hemicellulose is distributed throughout the wood cell walls. If you remove the hemicellulose, the crystalline cellulose fibrils are left much less protected — that is, more easily accessible. Even drying the fibres after extraction of the hemicellulose considerably increases the accessibility to hydrolysis of the cellulose framework, as further described in my paper, with a lower D.P. after hydrolysis as a result. With the hemicellulose fibrils are hardly affected.

In a region where a pulp fibre is crushed or sharply bent, it seems likely that cellulose fibrils will be deformed more than their crystalline lattice can take elastically, so that slip planes develop in the lattices. The lattice will be more disordered and consequently more accessible to chemical attack at these points. Such phenomena would explain why mechanically damaged wood fibres are hydrolysed so fast at the break points that they disintegrate into fragments as often observed in practice.

This interpretation would mean that the protection of the cellulose in a sulphite cook at the high acidity and the high temperature used is due to two factors — the long extended crystalline regions inside the individual cellulose fibrils and the close packing with the incrusted hemicellulose. If the cell wall layers are ruptured, regardless of whether it is done by external forces or by shrinkage in drying, accessible points in the cellulose fibrils (slip planes in the cellulose lattice) are introduced, which opens the way to chemical degradation.

From this point of view, it was very interesting to hear about the lower D.P. values found for cellulose in tension wood. Tension wood, if the term is correct, should mean wood tissue that has been under stress. Mechanical tension could introduce slip planes in the cellulose lattice, which in themselves might degrade the cellulose chains. Slip planes in particular would make the cellulose more accessible to chemical attack during its isolation. It would be interesting therefore to know the levelling off D.P. after hydrolysis of these pulp fibres with dilute mineral acid. You would expect lower values than for normal woodpulp fibres. Are you sure, Mr. Bos, that you extracted the cellulose without further degradation?

MR. BOS: We determined the levelling off D.P. by hydrolysis of the chemical pulps with hydrochloric acid. The results should be considered as preliminary and have to be confirmed by further data.

If the D.P. is plotted against the residue, as in your paper, the slope for reaction wood is less steep than for normal wood and therefore resembles more that for cotton. The mean position of the two lines does not differ, however.

DR. RÅNBY: Unextracted holocellulose behaves differently from chemical grade pulps and cotton fibres when hydrolysed. The initial reaction brings the holocellulose to a levelling off D.P. much higher than those of chemical grade pulps and cotton, but the continued hydrolysis causes a more marked and gradual drop in D.P. of the residual hydrocellulose from holocellulose. These data suggest then that the crystalline regions of the cellulose fibrils in the original wood are longer, but of lower general lattice order than those of cotton fibres. It is a surprising finding that cellulose from compression wood behaves more like cotton cellulose.

MR. BOS: Although it was not mentioned in the paper read by Christensen, I can add that Wardrop has produced X-ray photographs of Eucalyptus fibres and they show the degree of crystallinity of the cellulose from tension wood is much higher than for normal wood. It is of the order of that for cotton.

Recalling the earlier discussion on hemicellulose and paper strength, I think it would be useful to study further the compression wood in softwoods and the tension wood in hardwoods from the papermaker's point of view. We can say that the inner structure of the cell wall is quite different. In this way, we can have a better knowledge of the influence of the structural features of the cell wall for papermaking purposes and we can achieve faster results than by studying solely pulp fibres from normal wood.

MR. F. M. CROOK: Dr. Christensen mentioned this morning that I had some further information on Wardrop's proposed structure of the S1 layer of the secondary wall. I think it is probably appropriate for me to mention it now.

I have Wardrop's electron micrographs with me. We tried them on the episcope last night, but unfortunately they did not have enough contrast to be normally visible on the screen, so I am afraid I will have to try to draw the scheme for you on the blackboard.

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Under the primary wall with its disordered arrangement of fibrils, Wardrop proposes that the first appearance of the S1 layer is a series of fine microfibril bundles. Overlying that, approximately at rightangles, comes a second layer of fine microfibril bundles in the fashion of Fig. 17 in Wardrop's paper.

Wardrop calls this stage of the organisation his fine grid. Overlying this fine grid, there is a layer of coarse microfibril bundles in the same direction as one of these two. If you look at Fig. 18 in the paper, you will see something like that there.

The development of the S1 layer continues still further. Overlying this again comes a second series of coarse fibril bundles. Wardrop calls this a coarse grid system. This upper layer may continue to have bundles deposited in it to form a complete sheet. Wardrop considers that this may reconcile a good many conflicting views of the S1 layer. For one thing, it would agree with Asunmaa's finding that there were probably several layers of S1 wall thickening. It also confirms several ideas on cross-fibrillated structures. The existence of a complete sheet in one direction would explain some confusion about the existence of a cross-fibrillated structure and the observation of a single angle of extinction in the polarising microscope. Incidentally, the helical systems being symmetrically disposed about the long axis of the fibre, the angle at which they cross (measured wet in the polarising microscope) is about 80°. If only the fine grid system is present, the angle (measured dry in the electron microscope) remains at about 80°: but, if the coarse grid system is present, there is a change in angle to about 60°, presumably because the points of attachment in the coarse grid are quite firm and the distortion of the coarse grid is superimposed on that of the fine grid. This change in angles, he supposes, may correspond with the elongation of some wood on drying.

The fine fibril bundles are about 600 Å and the coarse bundles about 2 000 Å. If anybody is interested, I have Wardrop's pictures of *Pinus radiata* with me. It is rather interesting that I have found the same structure in some of my own work on other woods.

PROF. B. STEENBERG: The question raised by Dr. Rånby about hydrolytic attack is well worth further consideration. If I understood Dr. Rånby right, he was suggesting something that could be related to a sort of stress corrosion. You take a piece of metal, bend it and put it into a corrosive substance or liquid, you know that it will corrode very rapidly indeed. You may say that the forces of bending have decreased the energy of activation for the corrosion.

Second Discussion

The important thing in this connection is that such a break is an autocatalytic process, which means that the metal will break, not in many parts, but just in two parts and, from that moment on, there will be practically no further corrosion.

Therefore, if there is bending somewhere that introduces stresses in the chain, you will certainly have very strong hydrolytic attack at that point of stress, but the action, I think we all agree, will be autocatalytic. The stressed portion will break rapidly, the stresses will be relieved and you will have made two pieces. In order to explain that it breaks down into small pieces, either you have to assume that stresses are distributed all over the molecular chain, so that there is an equal chance of hydrolytic attack at intervals roughly about the length of the resulting pieces or you can assume according to Dr. Gallay that there will be, in the course of the chemical attack, repeated flexes and bendings of the structure that, because of their random nature may introduce a stress, now here, now there. Then the hydrolytic attack sets in at one place or another.

It is difficult with Rånby's theory to see how just one bending can be so fatal.

If we use the idea of stress corrosion, we must find a way that makes it understandable why the molecules do not break at one point but at so many points, as the decrease in D.P. shows. Another assumption is that the thermodynamic activity of the whole system is changed by swelling procedures that will, of course, influence every individual part. Let us assume that the long molecules are reinforcement in the amorphous concrete. If the whole structure is swollen, stresses will be introduced in practically all the long strand of chains and they are liable to break at several points. Swelling in undamaged fibres is restricted by the S1 structure. If S1 is broken anywhere, the swelling can occur. Comparison with an osmotic cell with and without membrane failure may give a clue.

DR. RÅNBY: Prof. Steenberg has discussed the increased accessibility of native cellulose fibrils as stress corrosion. This concept seems to be useful for the understanding and description of this phenomenon. I would like to refer to electron micrographs shown yesterday of well crystallised cellulose fibrils from animal cellulose (tunicin). Before hydrolysis, the fibrils were wavy and sometimes sharply bent, indicating the presence of stress in the lattice along the fibrils. After hydrolysis, the fibrils are straightened out and practically only rodlike fibril fragments are seen with a number of sharp breaks at various intervals. This looks like localised hydrolysis at stress

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points, allowing the fibrils to recrystallise and straighten out when a few chains are broken at these points.

Slip planes in the lattice of the fibrils do not necessarily mean stress. Their formation could simply mean a local disordering of the lattice — an opening of original hydrogen bonds and formation of new bonds of lower strength. Such an effect is easy to accept when you realise that practically all hydroxyl groups in solid cellulose are engaged in hydrogen bonds of various strength and regularity. Stress corrosion and local lattice disorder could explain the accessibility of cellulose fibrils to chemical degradation at certain points.

THE CHAIRMAN: Can anybody shed any light on the galactose question mentioned by Dr. Christensen?

DR. H. MEIER: We studied in Stockholm the carbohydrate composition of compression wood, but, unfortunately, I have no data on the galactose content. We found, however, that the mannose content was considerably lower in compression wood than in normal wood. Therefore, it seems that the carbohydrate composition in compression wood is rather different from that in normal wood.

PROF. L. G. STOCKMAN: Some years ago, an investigation on compression wood from spruce wood (*Picea abies*) showed it to contain 10 per cent. of galactose, which is in very good agreement with the Australian figure.