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EFFECTS OF VARYING REFINER PRESSURE ON THE MECHANICAL PROPERTIES OF LOBLOLLY PINE FIBRES

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SUMMARY

Loblolly pine chips, separated into mature and juvenile portions, were refined at three pressures (4, 8, and 12 bar) in a single disc refiner at the BioComposites Centre. Fibres were dried in a flash drier to a moisture content of approximately 12 percent. The mechanical properties of single fibres from each refining pressure were determined using a tensile strength tester. The tensile tester is a custom-built screw-driven cross-head fitted with a 5 Newton-capacity load cell. Fibres were tested at an elongation rate of 80 microns/minute. Load-elongation traces were converted to stress-strain curves with tensile span and cross-sectional area. Tensile span was measured as cross-head movement and was approximately 1mm. The cross-sectional area of each tested fibre was measured using confocal microscopy. Fibre modulus of elasticity and ultimate tensile stress were then determined from the stress-strain curves. Fibre stiffness and strength were further studied to evaluate their effect on structural fibreboard panels. Small MDF panels were prepared with a urea-formaldehyde adhesive at an addition level of 12 percent solids to determine this effect. The dimensions of the mini-panels were 100 x 125 x 3mm The panels were tested for MOE, MOR and internal bond strength. Correlations were then drawn between fibre and subsequent panel properties.

INTRODUCTION

It has long been accepted by the forest products community that the mechanical properties of wood-based composites are strongly influenced by the mechanical properties of the components. This is especially true for composites made up of large components such as glue-laminated beams and laminated veneer lumber. This tenet of composite-component association has been more difficult to evaluate for composites made up of wood fibres due to the experimental difficulty of assessing fibre mechanical properties.

Researchers (Jayne, 1959; Jayne, 1960; Ehrnrooth and Kolseth, 1984) in the 1960's developed techniques to determine individual wood fibre mechanical properties. Though the data was extremely useful in estimating fibre stiffness and strength, the techniques proved too slow and tedious to be of practical use. Subsequent modification of techniques and research by Page and colleagues proved the significance of fibre mechanical properties to hand-sheet properties (Page *et al.*, 1977; Page and Seth, 1980).

Kersavage (1973) developed a tensile technique that used a ball-and-socket type assembly for determination of individual wood fibres. The technique eliminated stress concentrations at the grips and dramatically increased the sampling rate of specimens. This technique was later modified (Mott, 1995) for rapid tensile testing of black spruce and loblolly pine fibres. Due to these advances, mechanical property evaluation of individual wood fibres is becoming more routine and has been applied to fundamental questions of failure mechanisms and property variation within and between trees (Mott *et al.*, 1996). Also, the opportunity now exists to more accurately assess the basic relationships between fibre properties and panel performance.

In addition to fibre mechanical properties, other variables specifically related to stress transfer between fibres are also important in the structural performance of wood fibre-based

composites. Atomic force microscopy (AFM), widely used in assessing surface morphology, has been shown by Pesacreta *et al.* (1997) to be a valuable tool in determination of fibre roughness. New technologies and techniques regarding wood fibre surface energy are being established (Boras and Gatenholm, 1999; Gardner *et al.*, 1999) to evaluate the availability of bonding sites and their impact on transferring stresses within the fibre network.

The primary objective of the work presented in this paper was to assess the effect of refining pressure on wood fibre properties and as a consequence panel properties. The work was conducted using both juvenile and mature wood portions of loblolly pine.

MATERIALS AND METHODS

The raw material used for the work was loblolly pine (*Pinus taeda* L.) harvested from a conventional plantation in southern Arkansas (USA). The juvenile and mature portions of the bole were separated manually: 'juvenile' in this paper refers to wood comprising growth rings one to ten, whilst 'mature' refers to wood material comprising the 30th growth ring and higher. The transition wood between these zones was discarded.

Refining: Refining was conducted at the BioComposites Centre pilot plant, Bangor, Wales, UK. Chips were fed into the pilot plant and screw fed into a pressurised vessel. Pressure was maintained by means of a wood plug generated with an auger screw. The pressurised chips were fed into the refiner at a rate of approximately 40 kg per hour, with retention times in the cooker of approximately 4 minutes. Pressure was maintained at a constant 4, 8, or 12 bar for both the juvenile and mature chips. These conditions were selected to be below, at and above the glass transition temperature (T_g) of lignin, respectively.

Determination of Fibre Properties: The mechanical properties of individual fibres were ascertained on the 6 fibre types (juvenile or mature; 4-, 8-, or 12-bar pressure). In addition, unrefined portions of juvenile and mature chips were macerated in acetic acid and hydrogen peroxide. Fibres (ca. 100 for each condition) were then tested in tension to determine the modulus of elasticity (MOE) and ultimate tensile stress (UTS) of the furnish. A detailed explanation of the maceration technique can be found in Panshin and deZeeuw (1970). Additional information on the mechanical property determination methods are available in Groom *et al.* (1996).

Mini-panels: Miniature MDF panels measuring 100 x 125 x 3mm were constructed to investigate the relationship between fibre and panel properties. Mini-panels were prepared with a urea-formaldehyde resin at a constant resin content of 12 percent solids. A total of 45 mini-panels were constructed. Three replicates were evaluated for each refiner pressure (4, 8, and 12 bar) at varying ratios of mature (0, 25, 50, 75, or 100 percent) versus juvenile fibres. After conditioning, the mini-panels were tested in 3-point bending for modulus of elasticity (MOE) and modulus of rupture (MOR). Internal bond (IB) stress was also determined according to standard methods.

Fibre physical properties: The surface morphology of the treated fibres was investigated using a JEOL scanning electron microscope (SEM) and a Nanoscope IIIa atomic force microscope (AFM) (Digital Instruments, Santa Babara, Calif. USA). For the AFM analysis three 5μm scans, located in the middle and quarter-points of 10 individual fibres were collected from each treatment. Unrefined fibres macerated in a solution of peroxide/acetic acid were used for comparison. Fibres were oriented with the long axis parallel to the raster scan direction. Images were obtained in intermittent-contact mode (Tapping mode ^{TM}) at a

scan rate of 1 Hz. Three data channels, height, amplitude and phase shift were monitored during the image acquisition. In total 240 images, taken at a resolution of 512 x 512, were collected. Statistical analysis using the height data was carried out on each 5 μm image and representative areas of 2.5μm^2 and 1.25μm^2 selected from each image to quantify surface roughness (rms).

Surface Energy: Column wicking experiments were carried out to determine the contact angle of the refined fibres with the following probe liquids: α-bromonaphthalene, ethylene glycol, formamide and water. The pore size of the packed columns was first estimated using methanol and hexane. The rate of rise was determined for six replicates of each fibre/probe set, and contact angles calculated with the Washburn equation (Washburn, 1921). Surface energy for the fibres was calculated from the average contact angles using the Chang model (Qin and Chang, 1996). Gardner *et al.* (1999) have previously outlined the experimental details.

RESULTS AND DISCUSSION

Fibre Physical Properties: The refining condition had a pronounced effect on the physical appearance of individual wood fibres as well as their surface characteristics. Figure 1 shows scanning electron microscope (SEM) images of macerated juvenile and mature fibres as well as their corresponding refined counterparts. The surfaces of the macerated fibres were smooth and the fibres were intact with little or no fines. Physical observation of the refined fibres in the SEM showed a marked difference as compared to macerated fibres: the surface features of refined fibres were torn and irregular, with the surface characteristics becoming increasingly complex with increased refining pressure. The 12-bar fibres were fragmented in length and contained many globular features, presumably deposited during the refining process.

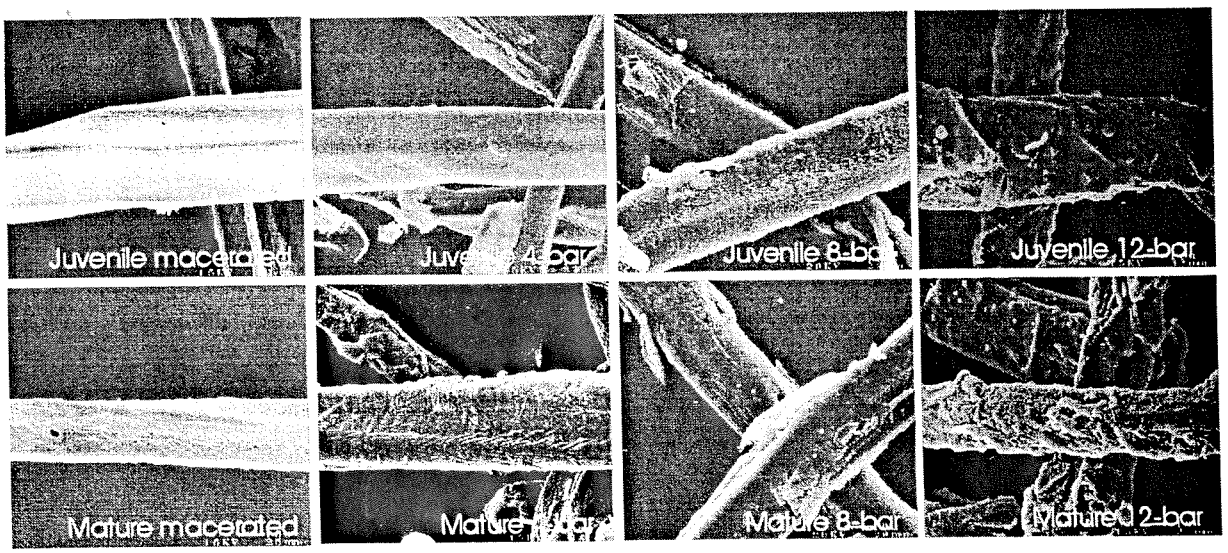


Figure 1: Scanning electron micrographs of juvenile and mature loblolly pine fibres macerated or refined at pressures of 4, 8, or 12 bar

The differences in fibre surfaces can be better characterised with the AFM. Figure 2 shows 5-micron scans of the 8 various fibre types as imaged with the AFM. Microfibrils can be seen over the entire surface of the macerated fibres indicating that the primary layer has been exposed. For the 4-bar fibres microfibril angle was at 30 degrees to the fibre axis indicating that the S2 layer had been revealed. Microfibrils were less evident at the higher refining pressures and the fibres appeared to have a smooth deposit on the surface. Surface profiles of

juvenile fibres are show in Figure 3. A line trace was taken at approximately 60 degrees to the fibre axis for each treatment. The scans show that surface roughness increased with increasing refining pressure. For the highest pressure there was a greater variation in height but the surface had a smaller frequency of oscillations when compared to that of the lower pressures. These results are reflected in the frequency distribution of surface roughness (rms) shown in Figure 4 for the juvenile fibres. There is a wide range of rms values for each treatment. However, macerated and 4-bar fibres had a high frequency of small rms values whereas 8- and 12-bar fibres had a greater proportion of large rms values.

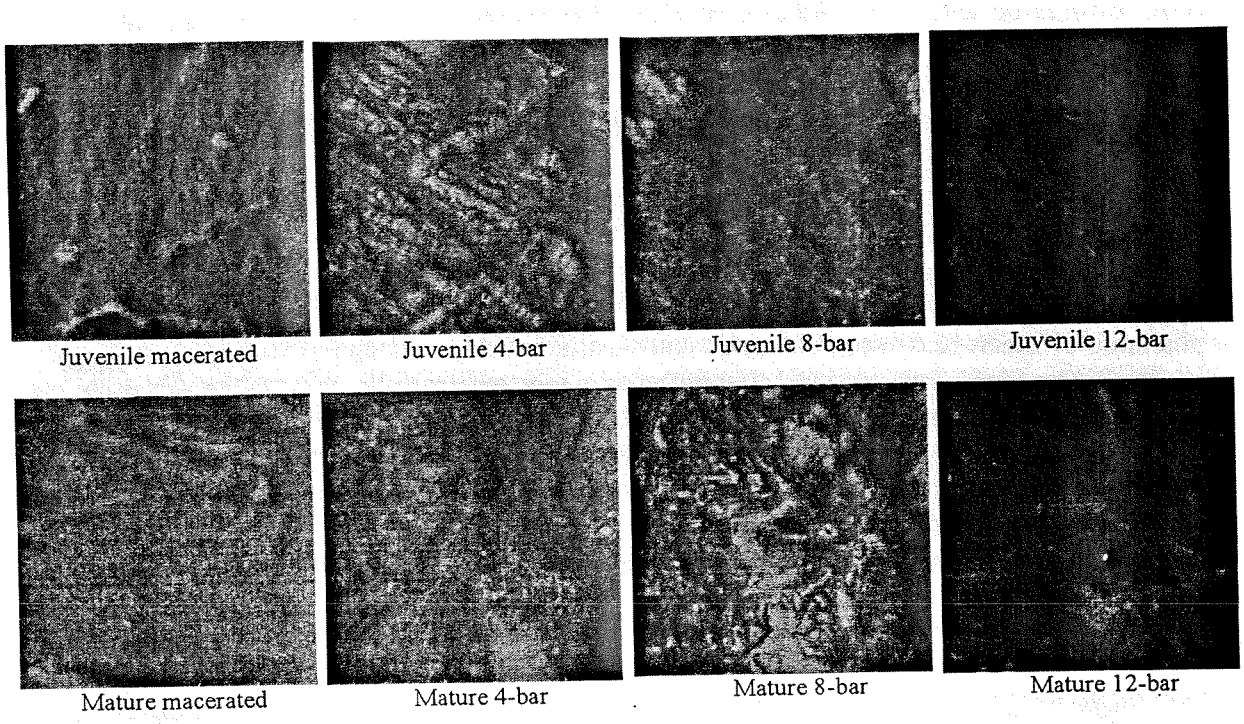


Figure 2: 5-micron scans of loblolly pine fibres observed with an atomic force microscope (tapping mode) generated by maceration or by refining at pressures of 4, 8, or 12 bar

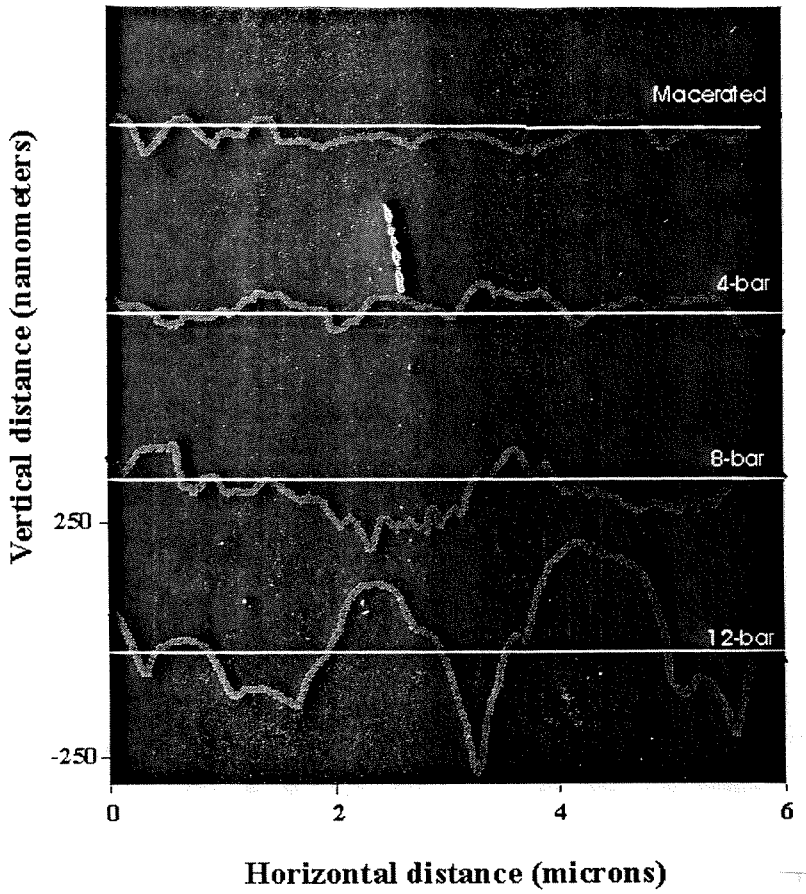


Figure 3: Section analyses of 5-micron AFM scans of loblolly pine juvenile fibres macerated or refined at pressures of 4, 8, or 12 bar

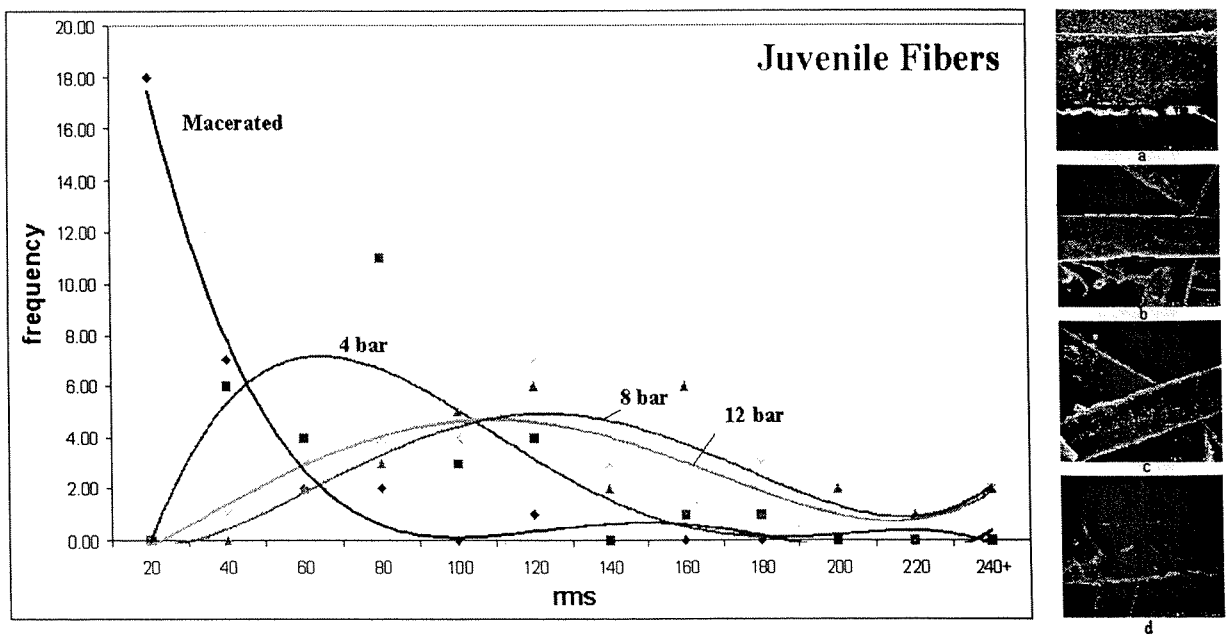


Figure 4: Frequency distributions of fibre roughness (rms) as measured with an atomic force microscope for juvenile, loblolly pine fibres that have been (a) macerated or refined at pressures of (b) 4 bar, (c) 8 bar, or (d) 12 bar

This trend is also seen in the phase images shown in Figure 5 where uniform areas of colour indicate areas of similar surface composition. For the 4 bar fibre surface there were mainly lots of smaller 'blobs' whereas for the 8 and 12 bar the areas of uniform composition increased in size. The results indicate that at the lower refining pressures a substance appeared to be deposited on to the fibre surface in very fine particles. At the higher pressures the deposit had 'flowed' together to form a larger and more homogenous coating.

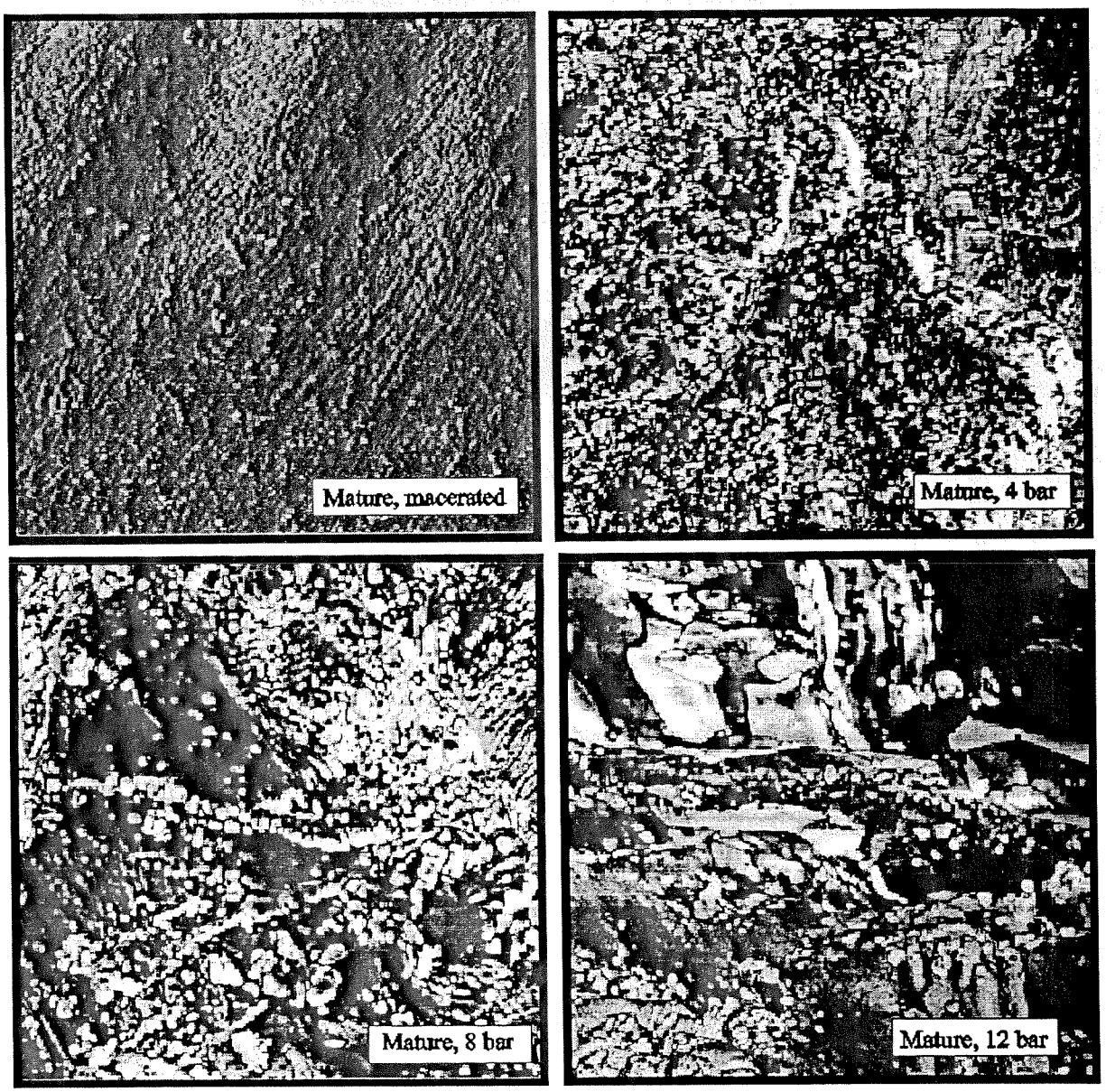


Figure 5: AFM phase shift images of mature, loblolly pine fibres that have been macerated or refined at pressures of 4, 8, or 12 bar. The lighter regions refer to material deemed as 'sticky'

Chemical composition data for the refined fibres are summarised in Table 1. The greatest effect of refining occurred with the hemicellulose component. As the severity of the refining process increased, hydrolysis of the hemicellulose occurred which led to increased solubility in water. As a result, a significant carbohydrate fraction was lost in the process water, and the

total hemicellulose content of the fibres was reduced. Interestingly, there was a corresponding increase in extractives as the refining pressure was raised. Subsequent analysis of the soluble material by NMR showed that it was comprised primarily of carbohydrates and hydrocarbon components (e.g., fatty acids). Although the relative contents of cellulose and lignin appeared to increase with higher refiner pressures, this increase was due primarily to the reduction in the hemicelluloses and thus their absolute content actually remained fairly constant. The mature fibres were higher in cellulose and lower in lignin than the corresponding juvenile fibres.

Table 1: Chemical composition of juvenile and mature loblolly pine fibres refined at pressures of 4, 8, and 12 bar

Fibre Type	Pressure (bars)	Extractives (Percent)	Lignin (Percent)	Alpha-cellulose (Percent)	Hemicellulose (Percent)
Juvenile	4	9.7	29.3	42.3	23.7
Juvenile	8	11.2	30.6	46.3	20.9
Juvenile	12	14.8	32.4	49.3	17.2
Mature	4	7.8	26.3	47.9	22.2
Mature	8	9.5	27.7	48.0	21.5
Mature	12	11.0	27.3	51.5	18.8

Surface energy: The effect of refining pressure on the surface energy of the juvenile and mature fibres is presented in Figure 6. Surprisingly, very different behaviour was observed for the two wood types. Increased refining pressure had very little influence on the surface energy of the mature fibre. In contrast, the total surface energy increased substantially for the juvenile fibres from 39 to 44 mJ/m^2. Consistent with previous reports, the total surface energy of the fibres was dominated by dispersive force contributions. The variation in the polar component with refining pressure was very different for the two wood types. At 8 bar, the mature fibres exhibited a maximum of 0.7 mJ/m^2 while the juvenile fibres reached a minimum of 0.24 mJ/m^2. This reflects the composition and distribution of chemical constituents on the surface. Additional work is required to fully interpret the implications of this observation; however, it is likely that these differences will influence the wettability and bonding by the adhesive resin.

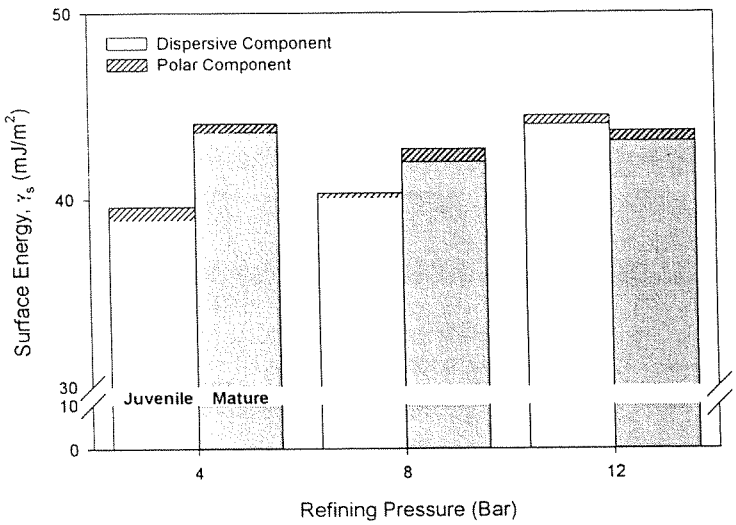
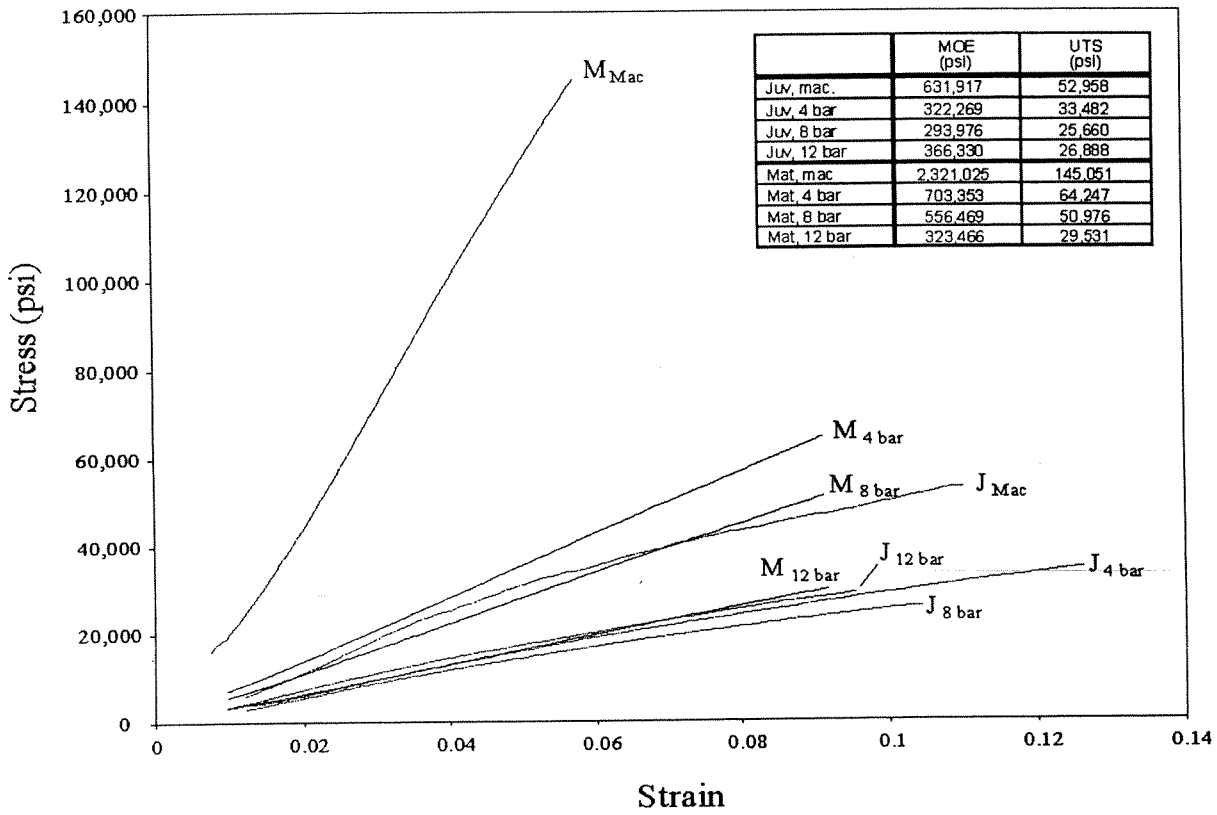


Figure 6: Surface energy of fibres refined at pressures of 4, 8, or 12 bar

Fibre mechanical properties: The results of the tensile testing of individual wood fibres are tabularly and graphically represented in Figure 7. The mature, macerated fibres were much stiffer and stronger than the corresponding juvenile fibres. This is in part due to the higher alpha-cellulose content in the mature fibres (Table 1) but primarily due to the inherent lower microfibril angle of mature fibres as compared to juvenile fibres.



	MOE (psi)	UTS (psi)
Juv, mac.	631,917	52,958
Juv, 4 bar	322,269	33,482
Juv, 8 bar	293,976	25,660
Juv, 12 bar	366,330	26,888
Mat, mac	2,321,025	145,051
Mat, 4 bar	703,353	64,247
Mat, 8 bar	556,469	50,976
Mat, 12 bar	323,466	29,531

Figure 7: Stress-strain curves and resulting mechanical properties of macerated loblolly pine fibres (J = juvenile; M = mature) and their corresponding response to refining at pressures of 4, 8, or 12 bar

The refining process had a dramatic effect on the mechanical properties of mature fibres and very little effect on the juvenile fibres (Figure 7). Initial refining at 4 bar reduced the stiffness and strength of the mature loblolly pine fibres by about 40 percent. Subsequent 4 bar incremental increases in refiner pressures decreased MOE and ultimate tensile stress (UTS) each time by approximately 25 percent. Refining also reduced the mechanical properties of juvenile fibres. However, the decrease of juvenile mechanical properties relative to the macerated fibres was approximately half, regardless of the refining level.

Panel Mechanical Properties: The effect of fibre maturity on the stiffness and strength of MDF panels is shown in Figure 8. At all refining pressures, MDF panel MOE and MOR increased with the addition of mature fibres. The extent of the increase in panel stiffness and strength was most pronounced at higher pressures. It was also found that the optimal refining level for development of MDF panel stiffness and strength resides between 4 and 12 bar and in the proximity of 8 bar (Figure 9). This is presumably a reflection of the T_g of lignin. Optimal refining will most likely exist above the T_g of lignin, thus minimising structural

damage of the fibre by directing failure into a specific plane or region of the cell wall. However, pressures must be kept low enough to minimise hydrolysis and thus the breakdown of the carbohydrate component.

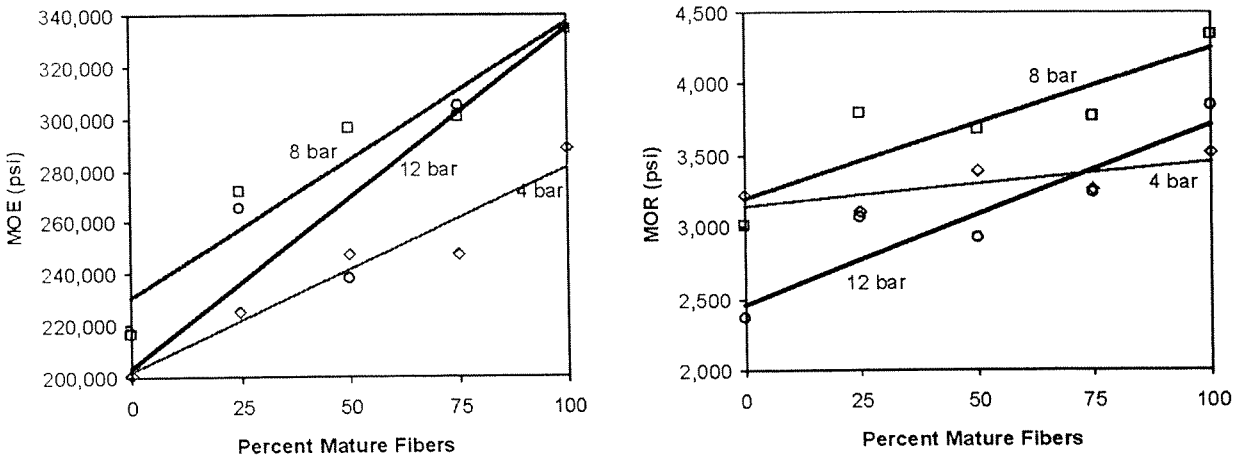


Figure 8: Modulus of elasticity and modulus of rupture of MDF mini-panels comprised of varying proportions of juvenile and mature fibres

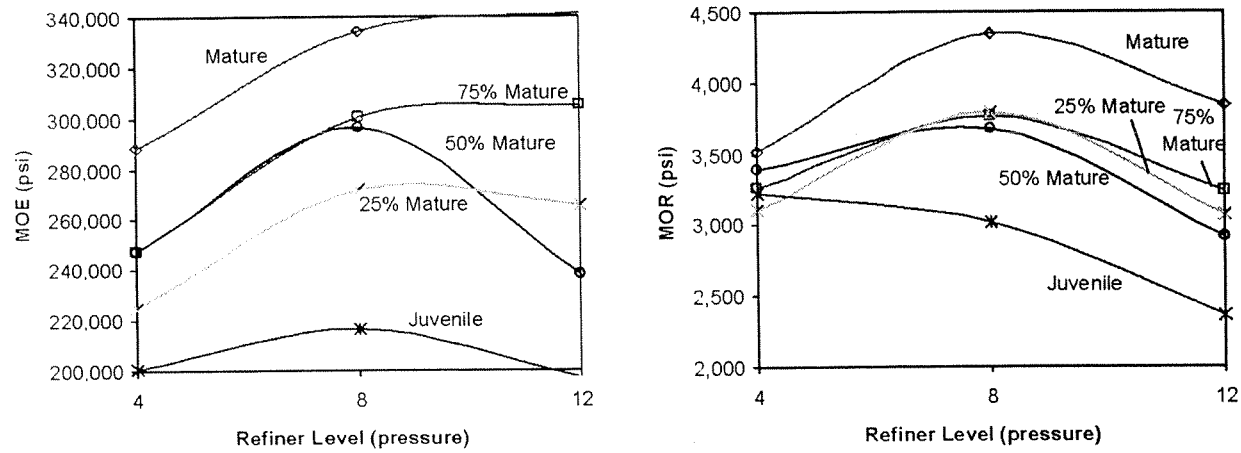


Figure 9: Modulus of elasticity and modulus of rupture of MDF mini-panels comprised of varying proportions of juvenile and mature fibres and plotted as a function of refiner level

The internal bond strength of MDF panels was directly related to the refining pressures (Figure 10). The internal bond consistently increased as the refiner pressure was raised. Unlike the mechanical bending properties of MDF panels, the maximum internal bond strength did not appear to be associated with the T_g of lignin. The continued increase in internal bond strength with refining pressures suggests a strong dependence on mechanical disruption of the fibres' cellulose network. These microfibril disruptions make the fibre more compliant and increase the number of intimate fibre-fibre contacts. This enhances the resistance to loading normal to the plane of the MDF panel.

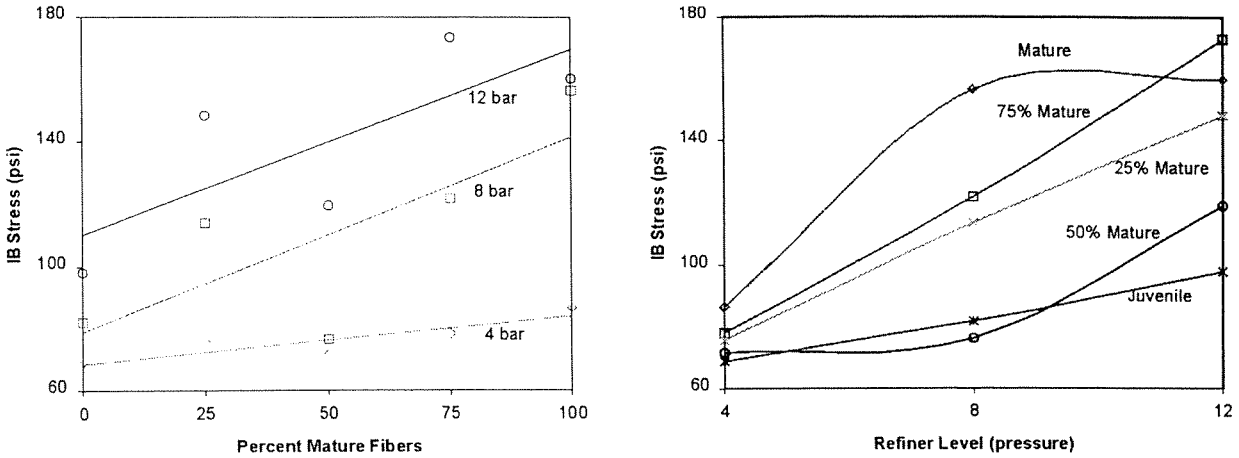


Figure 10: Internal bond strength of MDF mini-panels plotted as a function of ratio of juvenile to mature fibres and refining pressure

Relationship between Fibre and MDF properties: A comparison of fibre mechanical properties and the bending properties of MDF panels is shown in Figure 11. Although the refining process decreases fibre strength by 40 to 70 percent, fibre strength still far exceeds the strength of the resulting MDF panels by a factor of between 10 and 20. The trends of fibre and MDF panel strength are mirrored with stiffness results; however, the differences between fibre and MDF panel stiffness are not so pronounced. The stronger, stiffer mature fibres produced MDF panels that were also stiffer and stronger. The elevated refining levels resulted in fibres with diminished mechanical properties but subsequent MDF mechanical properties that seemed more closely aligned with the T_g of lignin. This effect of refining on fibres and MDF panels is better illustrated in Figure 12. Figure 12 plots the product of stiffness and strength and normalises these values against the MDF panels made from mature fibres refined at 8 bar. The refined fibres are also plotted in Figure 12 but on a different scale. Figure 12 illustrates the decrease in fibre mechanical properties with refining as well as the peak of MDF panel mechanical properties at 8 bar.

Although some of the MDF panel properties are explainable by fibre mechanical properties, there appear to be important contributors other than fibre stiffness and strength. Figure 13 illustrates the relationship between fibre surface properties as ascertained by the AFM and MDF panel mechanical properties. Macerated fibres were also analysed and included. Roughness is simply an accumulation of the deviation of every pixel in relation to a 2-dimensional plane of average height. Spectral density also evaluates the height data but focuses on the size and frequency of deviations. Phase shift reflects the 'stickiness' of a surface by impeding the vibrational characteristics of the tapping tip: greater impedance signifies a stickier surface. The phase shift denoted here is a measure of the accumulated stickiest 20 percent of the fibre surface divided by the least sticky 20 percent. The AFM-determined fibre surface properties reflect the shape of the corresponding panel mechanical properties plot, peaking in the vicinity of 8 bar. However, the magnitude of the mechanical property traces seem to be more closely related to the fibre mechanical properties. Fibre roughness appears to be the most closely related with MDF properties. However, phase shift and spectral density both indicate that surface properties of fibres are affected by the refining process.

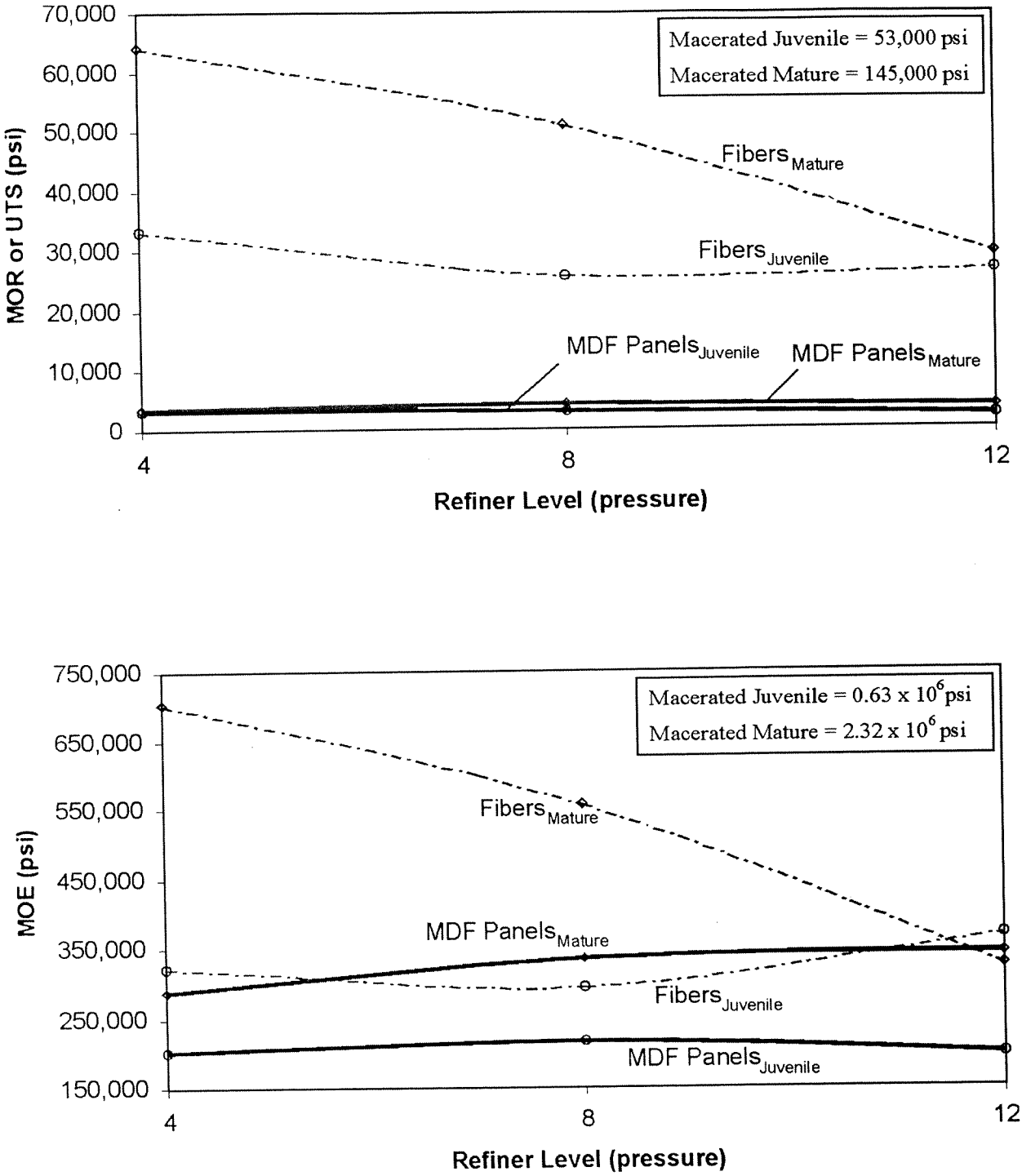


Figure 11: The strength (top) and stiffness (bottom) of individual wood fibres and subsequent MDF mini-panel properties. Fibre mechanical properties are shown as a dashed line; panel properties are shown as solid lines

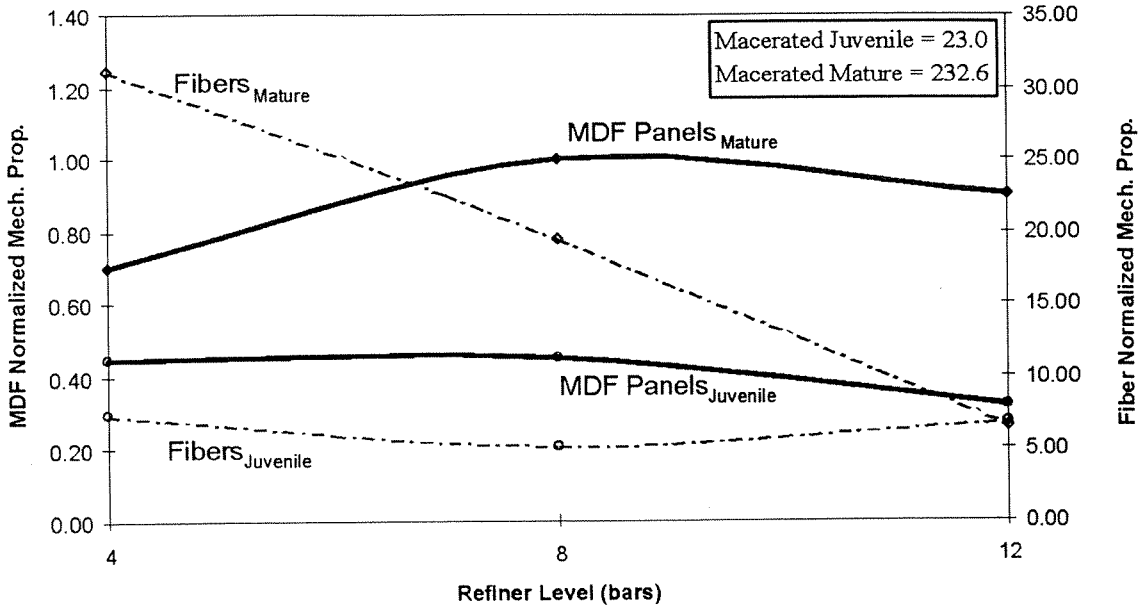
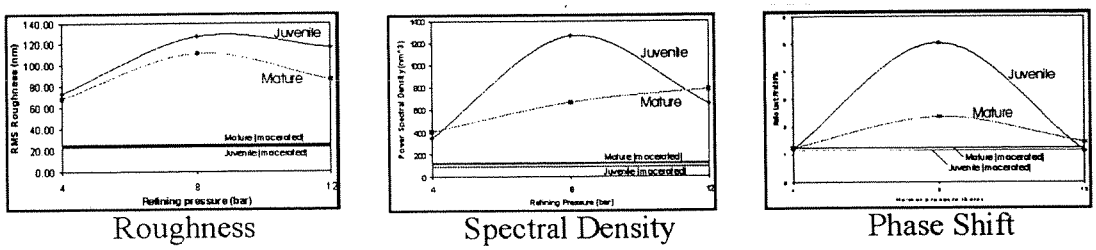


Figure 12: Normalised mechanical properties of fibres (dashed line) and MDF (solid line). All properties are normalised against MDF panel properties comprised of fibre refined at 8 bar. The scale for fibre properties is re-plotted on the rightmost y-axis due to the difference in mechanical properties magnitude.



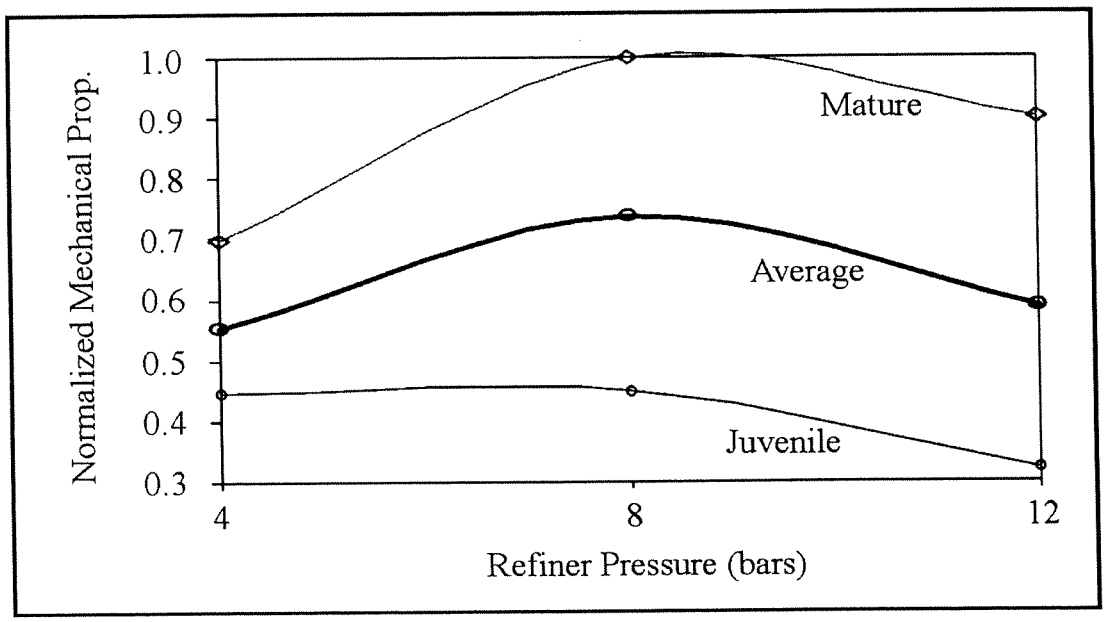


Figure 13: AFM roughness, spectral density, and phase shift of individual wood fibres and subsequent normalised mechanical properties of MDF mini-panels.

CONCLUSIONS AND RECOMMENDATIONS

The refining pressures commonly used for the manufacture of fibres for the structural fibreboard industry have a dramatic effect on the mechanical properties of the wood fibre furnish as well as corresponding MDF mechanical properties. The juvenility of the chip source also influences the final composite product. MDF panels made exclusively of mature fibres were stiffer and stronger than the juvenile fibre panel counterparts. This difference in stiffness and strength ranged from between 25 to 50 percent. The refining process was also important in the determination of the final chemical composition of the fibre furnishes, especially with respect to the hemicellulosic component. The fibre surfaces and resulting surface energetics were modified by refining pressure. These effects were reflected in the properties of the MDF panels.

This study has created as many questions as it has answered. A more comprehensive study is being conducted that compliments the findings of this study. Currently, loblolly pine chips at 4 levels of juvenility have been refined at 10 different pressures. The findings from these fibres and subsequent panels should expand our understanding of the complex relationships between refining, fibre properties, and performance of wood fibre-based composites.

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