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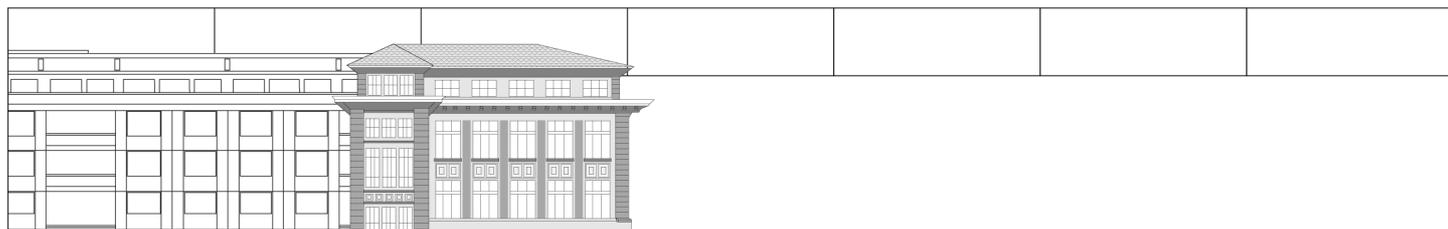
Forschungsbericht

Schlussbericht

Assemblage bois-bois par vibration linéaire

Welding of wood using the linear vibration welding technology (LVWT)

Bericht Nr.	2640-SB-01
Auftrag Nr.	2640.DWH
Klassifizierung	Öffentlich
Gegenstand	Gegenstand
Datum	10.12.2004



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ABSTRACT

Wood grain orientation differences in the two surfaces to be bonded yield bondlines of different strength in no adhesives wood welding. Longitudinal wood grain bonding of tangential and radial wood sections yields an approximately 10% difference in strength results of the joint. Cross-grain (90°) bonding yields instead a much lower strength result, roughly half that observed for pieces bonded with the grain parallel to each other. These differences can be explained by the very marked effect that homogeneity of fibre orientation is known to have on fibre-matrix composites. Oak yields lower results than beech and maple and is more sensitive to welding conditions. Differences in both anatomical and wood constituent composition can account for this difference in performance. Contrary to the other wood species, oak always presents joint bondlines where little or no increase in density at the interface is noticed. This explains its somewhat lower

strength results. This is based on the different mode of bonding predominant in this species, while the other species present two different modes of bonding. Thus, two types of bondlines are observed by scanning electron microscopy (SEM): (i) bondlines where entangled fibre-matrix composites are formed at the interface and (ii) bondlines in which direct welding of the cell walls occurs, just by fused intercellular material or cell surface material. In this latter case the cells remain flat, without an entangled fibre-matrix composite being formed. This is the almost exclusively predominant case for oak. Both cases and even hybrid cases between the two have also been observed in beech.

Keywords: bonding modes; wood grain; wood melting; wood welding.

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1 State of the Art

1.1 State of the art at HSB

In 1993, the Research and Development (R+D) department of HSB started to investigate the adequacy of different friction welding technologies for the bonding of wood. This first research project, funded by the KTI was a feasibility study focused on the utilization of the Ultrasonic Friction Welding technique and thermoplastic adhesives. Over the years, several research projects have followed that preliminary investigation, and the HSB has gained a lot of experience on the utilization of different welding techniques (Linear vibration, ultrasonic and spin welding) for bonding of wood with both thermoplastic and duroplastic adhesives.

In the years 2002 the R+D department of the HSB has made a preliminary investigation to find alternatives to traditional glued connections. The objective of the project was the development of an **innovative assembling technique** taking into account the economical and qualitative aspects of the industrial production. This research study, has led to the discovery that the Linear Vibration Welding Technology (LVWF), which is widely used in the plastic and car industry, could also be used to induce the local fusion of wood surfaces and thus to manufacture high-quality wood joints made without the use of any adhesive or additional toxic chemicals ([1.]-[14]).

1.2 State of the art at national and international level

A literature study as well as an IP research indicates that the HSB is in a pioneer position since no publication in international refereed journals could be found in the area of welding of wood without adhesives and LVWT. However, the relevance of the topic seems to be supported by the investigations carried out by different research teams and in most cases presented during workshops and published in local journal or Internet websites. Some of these publications refer to the utilization of the Orbital Friction Welding (OFW) technique for the same purposes. Unfortunately, none of them describes in details the latest progress made in the research. Based on the available information, it seems that the OFW can be also used to produce welded joints. However, while the HSB researchers were orientated to the production of joints for non-structural applications, the above works refer to the utilization of the OFW for structural uses.

2 Objectives of the project

This research proposal deals with the discovery, that the Linear Vibration Welding Technology (LVWT) could be used to induce the local fusion of wood and thus to manufacture a new generation of high-quality environmentally friendly wood joint, made without adhesive or additional chemical source. A patent and literature study has shown that the utilization of the LVWT for these special purposes has not been investigated yet.

Description of the process: Rubbing two pieces of wood against each other produces the energy needed to change their chemical/physical state, and thus to induce the local fusion of the wooden surfaces. The linear vibration movement is parallel to the surface of the connection. The energy input is a function of the friction coefficient, the welding pressure, the type of the joint surface as well as the amplitude, the frequency and the welding time. When the fusion state of the wood is reached on the joint interface, the vibration movement will stop. The clamping pressure is held on, until the solidification of the bonded joints is guaranteed (Fig. 1).

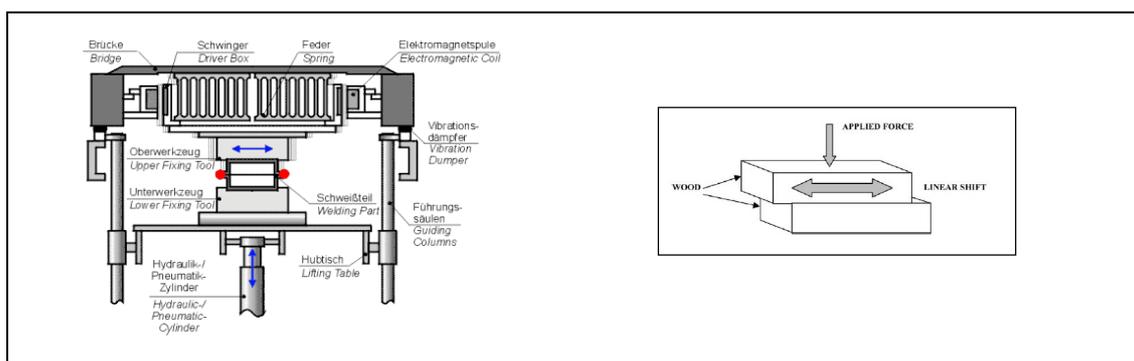


Fig. 1 Linear Vibration Welding machine (LVWM) and welding process.

2.1 Scientific Objectives

Once established that the developed technique induces the local fusion of wood and thus the production of wood joints able to meet the requirement of international standards, the principal questions that had to be answered were:

- What kind of influence the welding parameters (w. time, w. pressure, w. amplitude), and the wood properties (species, anatomy, grain orientation, moisture content etc.) have on the final strength of the welded products?
- Is it possible to increase the bonding strength of the welded joints?
- What kinds of chemical/physical mechanisms are responsible for the strengths of the welded products?

3 Research plan

Phase 1: Study of the influence of the welding parameters and wood proprieties on the final strength of the welded products.

Work package 1: Study of the influence of the welding parameters

Work package 1.1: Welding Time and amplitude

Work package 2: Study of the influence of the wood proprieties

Work package 2.1: Wood species

Work package 2.2: Grain orientation

Work package 2.3: Wood MC

Phase 2: Study of the chemical/physical mechanisms responsible for the strength of the products.

Work package 3: Physical characterization

Work package 3.1: SEM Scanning electron microscopy

Work package 3.2: X-ray micro densitometry of the welded line

Work package 3.3: TMA Thermo-mechanical analysis

Work package 4: Chemical characterization

Work package 4.1: Solid state ¹³C NMR

4 Project Organization

The project was managed by the HSB and developed in cooperation with the ENSTIB, a French school of wood engineering well knew in Europe for his experience in the field of wood adhesives and assembling technologies. However, the X-ray micro densitometry analyses were done at the INRA, France. The research team hereby to thanks for his cooperation Dr. J-M. Leban.

4.1 The research Team

For the HSB, University of applied science Bern:

Dr. F. Pichelin - Responsible of the project

Dr. M. Properzi - Project Manager

Mme S. Wieland - Project Assistant

Mr. M. Lehmann - Project Assistant

For the ENSTIB - University of Nancy 1, France:

Prof. Antonio Pizzi

Dr. Michela Zanetti

5 Material and Methods

5.1 Phase 1: Study of the influence of the welding parameters and wood properties on the final strength of the welded products.

5.1.1 Preparation of the joints by mechanically induced wood welding and their testing.

Specimens composed of two pieces of tangential, radial and cross fibre (90°) beech (*Fagus sylvatica*) wood, each of dimensions 150x20x15 mm dimensions were welded together to form a bonded joint of 150x20x30 mm dimensions by exercising a vibrational movement of a wood surface against the other at a frequency of 100 Herz. When the fusion state was reached on the joint interface, the vibration process was stopped. The clamping pressure was than briefly maintained until the solidification of the bond. The welded samples were conditioned for one week in the climatic chamber (20°C and 65% moisture content (MC)) before testing .

The parameters used for welding were those optimized previously (Gfeller et al. 2004): the welding time was varied (WT = 3, 4 and 5 s); the contact holding time maintained after the welding vibration stopped (HT = 5 s); the welding pressure exercised on the surfaces (WP = 1.3 MPa); the holding pressure exercised on the surfaces after the welding vibration stopped (HP = 2.0 MPa); the amplitude of the shift imparted to a surface relative to the other during vibrational welding (A = 3 mm). The frequency of welding was maintained at 100 Hz.

The tensile strength was measured on samples according to European standard EN 204 (2004) and method EN 205 (2003). The samples were saw cuts perpendicular to the specimens wood grain, down to the bond line, were made from each one of the specimen two surfaces. The distance between the two cuts was of 2.5 cm. The specimens were then tested in tension on an Instron model 4467 universal testing machine at a rate of 2 s/mm.

5.1.2 Testes wood species

Three species of hardwood were tested: beech (*Fagus sylvatica*), oak (*Quercus robur*) and maple (*Acer* spp.). The equilibrium MC of the samples was of 12%. Water spray was used where indicated to evaluate if it rendered bonding easier.

5.1.3 Grain orientation

The influence of the grain orientation on the final strength of the welded joints was also studied. Experimentations were carried out with specimens cut in longitudinal tangential and longitudinal radial orientation. Therefore, the sample were welded in:

- Longitudinal tangential orientation/ Longitudinal tangential orientation
- Longitudinal radial orientation/ Longitudinal radial orientation
- 90° crossed wood grains, samples cut in the longitudinal radial orientation

5.1.4 Wooden surface treatment

To observe the effect of a different surface treatment, welding was carried out on wood whose surface had been smoothed by sanding in order to compare it with previous work where planing was used.

5.2 Phase 2: Study of the chemical/physical mechanisms responsible for the strength of the products.

5.2.1 SEM Scanning electron microscopy

The SEM equipment used was a Hitachi S-520 used at magnifications of 300:1, 1000:1, 2500: 1 and 7000: 1. The equipment was used to analyze: (1) The surface of the open joints after mechanical testing, (2) The side of still closed joints and (3) the waste fibre that accumulated on the surface of the joints during welding were analyzed after metallizing with gold-palladium.

5.2.2 X-ray micro densitometry of the welded line

The X-ray micro densitometry equipment used consisted of an X-ray tube producing 'soft rays' (low energy level) with long wave characteristics emitted through a beryllium window. These were used to produce an X-ray negative photograph of approx. 2-mm-thick samples, conditioned at 12% moisture content, at a distance of 2.5 m from the tube. This distance is important to minimize blurring of the image on the film frame (18 × 24 cm) which was used. The usual exposure conditions were: 4 h, 7.5 kW and 12 mA. Two calibration samples were placed on each negative

photograph in order to calculate wood density values. The specimens were tested in this manner on equipment consisting of an electric generator (Inel XRG3000), a X-ray tube (Siemens FK60-04 Mo, 60 kV, 2.0 kW) and a Kodak film negative Industrex type M100.

5.2.3 TMA Thermo-mechanical analysis

The wood fusion joints obtained were cut progressively into joints of dimensions suitable for testing by thermo mechanical analysis. Thus, samples of 21 x 6 x 1.2 mm were obtained. Triplicate samples of these beech wood joints for a total sample of 21 x 6 x 1.2 mm were tested in non isothermal mode between 40 and 220°C at a heating rate of 10°C/min with a Mettler 40 TMA apparatus in three points bending on a span of 18 mm exercising a force cycle of 0.1/0.5N on the specimens with each force cycle of 12s (6s/6s). The classical mechanics relation between force and deflection, $E = \frac{L^3}{4bh^3} \frac{F}{\Delta f}$ allows the calculation of Young's modulus E for each case tested. Since the deflections Δf obtained were proven to be constant and reproducible, and these are inversely related to the values of the modulus, the values reported in the tables are often the values of the deflection (in micrometers).

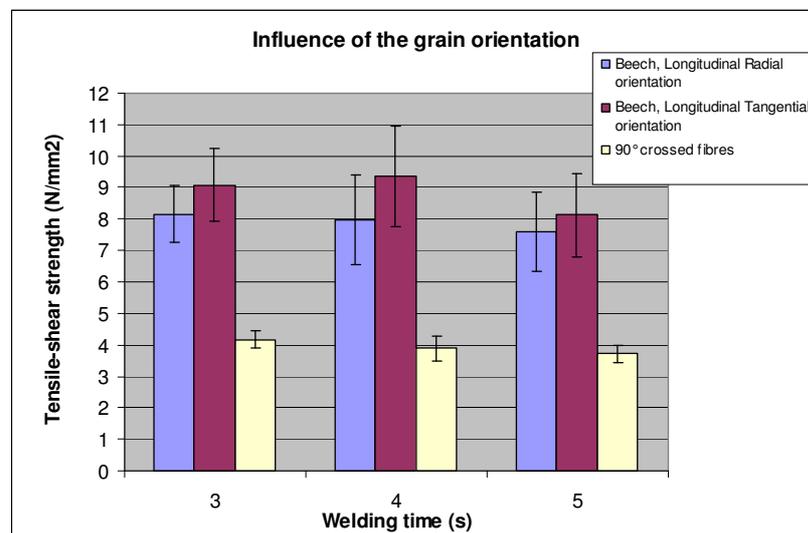
5.2.4 Solid state ¹³C NMR

The surface of the welded joints obtained was analyzed by solid state CP MAS ¹³C NMR. The equipment was used to analyze: (1) the surface of the open joints after mechanical testing, (2) the surface of joints after that one of the wood surfaces had been microtomed away as well as (3) the side of still closed joints were tested. Furthermore, (4) the waste fibre that accumulated on the surface of the joints during welding was also analyzed. The spectra were obtained on a Bruker MSL 300 FT-NMR spectrometer. The solid state CP-MAS ¹³C NMR spectra of the bonded interface reduced to powder by sanding, of the control beech wood powdered by either sanding or grinding and of the ground excess fibre waste coming out at the joints sides were obtained at a frequency of 75.47 MHz and at sample spin of 4.0 kHz. The impulse duration at 90° was of 4.2 microseconds, contact time was 1 ms, number of transients of about 1000, the decoupling field was of 59.5 kHz. Chemical shifts were calculated relative to TMS for NMR control. The spectra were accurate to 1 ppm.

6 Results - Phase 1

6.1 Influence of the wood species, grain orientation, surface treatment, and welding parameters on the final strength of the joints.

The results shown in Tab. 1 and Graph. 1 indicate that, for bonding along the longitudinal wood grain, slightly better results are obtained for the tangential versus the radial section. This is only a trend because the differences noted, although consistent, are not statistically significant. Both 3-s and 4-s welding times yield the best results. At the 5-s welding time, the results start to become lower, confirming previous findings. Statistically significant differences do, however, exist between these specimens and those bonded at 90° crossed wood grains of the two surfaces (Graph. 1 Fig. 2 Fig. 3). The crossed wood grain specimens (typical example shown in Fig. 2) yield much lower bond strengths. They also present much lower variability. For cross-grain-bonded specimens, the bond strength decreases slightly with lengthening welding time, as expected. The best results are obtained for the 3-s welding time. European standard EN 204 (2004) and testing method EN 205 (2003) for thermoplastic adhesives were chosen for the test rather than EN 301 (1993) and EN 301-1 (1993) for thermosetting adhesives. This was done because wood welding is exclusively used for interior furniture and joinery, hence as a substitute for PVAc, a thermoplastic adhesive.



Graph. 1 Beech specimens: influence of the grain orientation on the final strength of the welded specimens.

			Welding time		
			3 s	4 s	5 s
			(N mm ⁻²)°	(N mm ⁻²)°	(N mm ⁻²)°
Beech	radial section	fibres longitudinal	8.2±0.8	8.0±1.0	7.6±1.2
	tangential section	fibres longitudinal	9.0±1.3	9.4±1.6	8.2±1.4
	radial cut	90° crossed fibres	4.2±0.4	3.9±0.4	3.7±0.3
	radial section	planed	8.2±1.9	8.0±2.9	7.6±2.7
Oak	radial section	planed	7.5±1.6	5.8±3.0	3.1±2.0
Maple	radial section	planed	10.7±2.4	10.6±1.5	9.2±2.0

Tab. 1 Tensile shear strength mean values and standard deviations tests according to European standard EN 204 (2004) and method EN 205 (2003) of mechanically induced wood fusion welding (each result is the average of 10 tests).

The bondline has been observed as being composed of entangled fibres immersed in a matrix of molten compounded middle lamella material. The comparative results of radial, tangential, and cross-fibre strength in Tab. 1 and Graph. 1 indicate that grain angle and fibre orientation are important in wood welding. Passing from the standard bonding of radial sections to bonding tangential sections, grain angle and hence fibre orientation of the two surfaces change slightly.

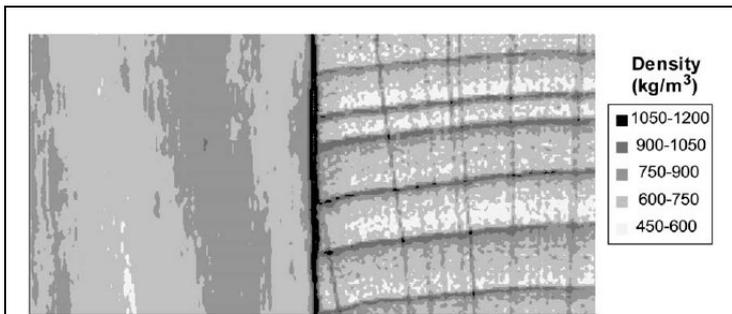


Fig. 2 X-ray micro densitometry-obtained density map of welded joint from cross-fibres of beech specimens.

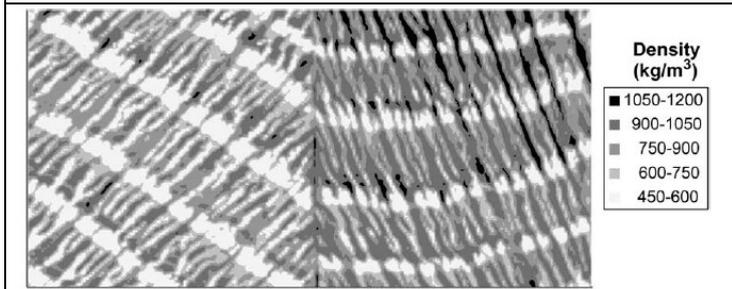
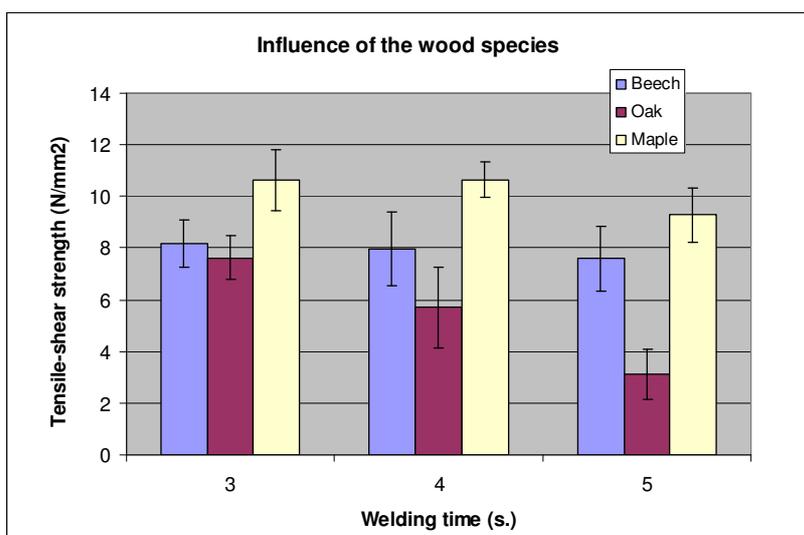


Fig. 3 X-ray micro densitometry-obtained density map of welded joint from radial section of oak specimens.

microscopy: (i) bondlines in which higher density entangled fibre–matrix composites are formed and for which photographic evidence has already been given ; (ii) bondlines in which direct welding of the cell walls of a surface to the cell walls of the other surface by the molten cell surface or inter-cellular material is observed. In these, the wood cells remain flat, and an entangled fibre–matrix composite does not constitute the bondline. Thus, Fig. 4A,B shows oak welded bondlines that have been separated and where quite evidently the cell walls of a surface have bonded to the fused material of the cell wall of the second surface, without any entanglement. In the case of oak, this shallower case is almost exclusively the predominant one. This explains the small or no increase in interface density observed and the lower bond strength obtained. In the case of beech, cases (i) and (ii) are equally present; whenever a small anatomical gap between the two surfaces occurs, then case (i) predominates. Hybrid cases also occur, in which the cells of a surface remain flat and their walls bond to the entangled fibre–matrix composite.

As a consequence, fibre orientation in the interfacial high-density composite also changes slightly. The orientation of the fibres in relation to the testing force applied to it has a very marked influence on the strength of a fibre–matrix composite. Even very minor variations in fibre orientation angle cause very marked strength differences in the composite. Thus, the very small change in fibre orientation passing from bonding radial sections to bonding tangential sections yields some improvement in the strength results. Equally, in the cross-fibre case, because relative fibre orientation in the composite is considerably more affected and less homogeneous, the mechanical resistance of the joint is lower Tab. 1.

Two types of morphology of the welded bondlines are observable by scanning electron



Graph. 2 Influence of the wood species and welding time on the final strength of the welded products.

Several parameters appear to be in direct relation to the bond strength obtained. These are the interfacial composite density as measured by X-ray micro densitometry, the evenness of the bondline, and the evenness of the grain of the wood species used. This can be seen from the comparison of the bond strength results of planed radial sections of beech, oak, and maple in Table 1. Maple specimens, the most evengrained of the three species, give a smoother bondline than the equivalent beech specimens. Conversely, while the bondline in oak (typical example in Fig. 3) appears to be equally smooth, the bondline density is much lower (Fig. 3; no

extensive dark line can be observed at the interface). This indicates a lower density composite at the interface, consistent with the lower oak bond strength in Tab. 1.

However, for oak, contrary to beech and maple, the dependence of the bond strength on the welding time is considerable and statistically significant (Tab. 1 Graph. 2). For oak, a 3-s welding time gives the best bond strength result, and the bond strength markedly decreases with lengthening welding time. The poorer welded strength results for oak indicate that the anatomy of the wood species, hence its microroughness, may have an influence on the friction coefficient. Thus, it has an influence on the maximum temperature that can be reached during welding and consequently on the quality of welding. Oak specimens always give the impression of low or no friction and of the two surfaces just sliding against each other without wood fibres detaching from the surfaces.

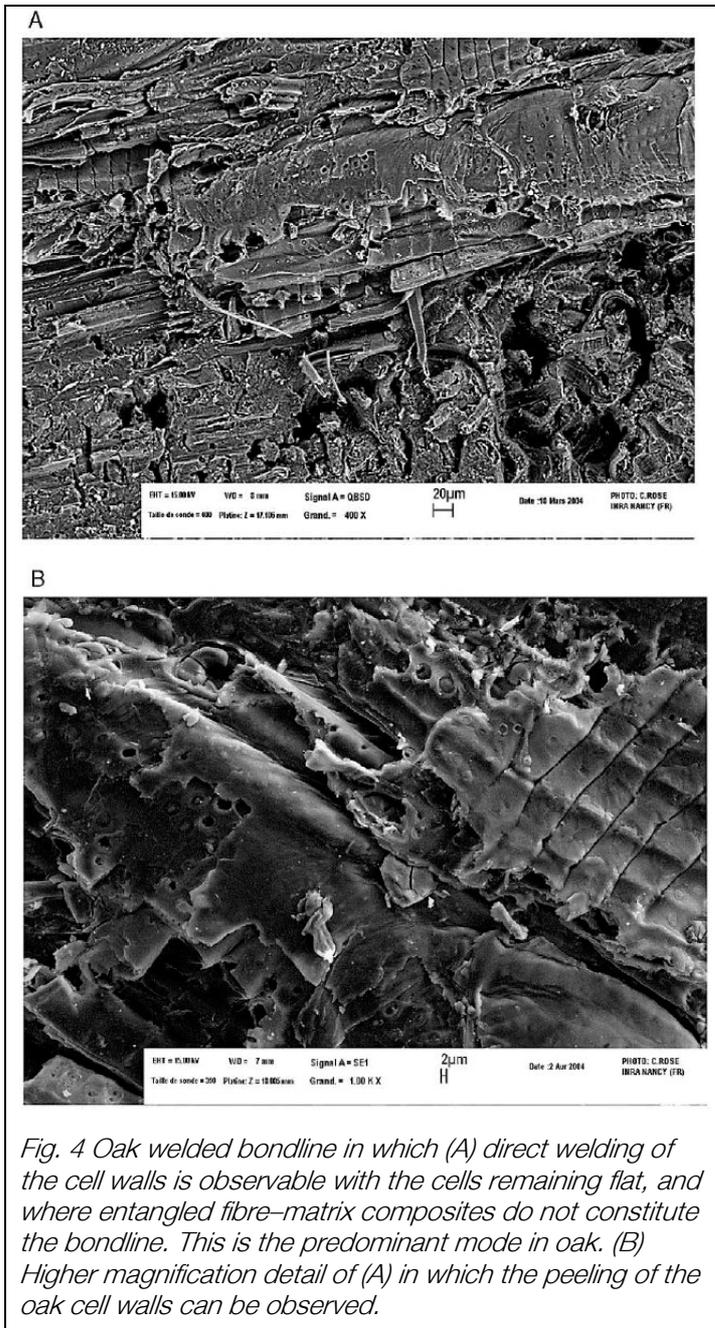


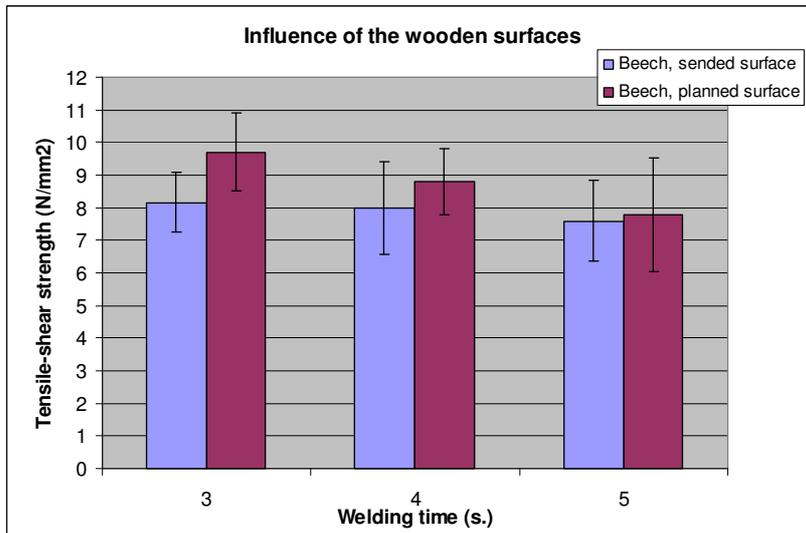
Fig. 4 Oak welded bondline in which (A) direct welding of the cell walls is observable with the cells remaining flat, and where entangled fibre-matrix composites do not constitute the bondline. This is the predominant mode in oak. (B) Higher magnification detail of (A) in which the peeling of the oak cell walls can be observed.

Fig. 3 shows that the oak interface (bondline) is not any denser than the two bonded wood pieces. The effect cannot be due to oak being more sensitive to degradation following more extensive and rapid charring at the interface. Rather, it seems to be due to the extreme shallowness of the interface composite formed. Once formed, to prolong the welding time means only to disturb and destroy the small amount of fibre entanglement giving the bond. This leads to the marked decrease in bond strength shown in Tab. 1. The differences leading to this could be anatomical or chemical.

The poorer welded strength results for oak indicate that the anatomy of the wood species, hence its micro-roughness, may have an influence on the friction coefficient. Thus, it has an influence on the maximum temperature that can be reached during welding and consequently on the quality of welding. Oak specimens always give the impression of low or no friction and of the two surfaces just sliding against each other without wood fibres detaching from the surfaces. Alternatively, the lignin/hemicelluloses of oak's middle lamella may not flow as easily. This implies perhaps a higher molecular weight or a different type of lignin; hence, oak middle lamella presents a lower proportion of material that can flow. It can also imply that heat transfer is poorer in denser wood, thus flowing is not as rapid as in other species.

That the composition of the middle lamella has an effect on the results in Table 1 is evident by the typical analysis values for the three wood species. Typical results obtained for lignin and polyosis analysis of the three types of wood show that the typical level of lignin in maple is 22.8%. This is comparable to the lignin levels reported for beech at 22.2%, 22.8%, and 23.8%. The values reported for oak are instead much higher at 24.9% and 29.6%. Conversely, for the total polyoses, the typical values reported by the same authors for the three wood species are, respectively,

26.5% for maple; 50.8%, 44.4%, and 36.5% for beech; and finally 38.8% and 22.2% for oak. Although these values refer to the whole timber, it is noticeable that some correlation trend exists between the total amount of lignin and the results in Tab. 1. Thus, beech and maple give the best results with a comparable amount of lignin, and oak gives a lower result with a higher level of lignin. No obvious correlation between polyosis level and Tab. 1 results is noticeable.



Graph. 3 Influence of the wooden surface treatment on the final strength of the welded joints.

The effect of the surfaces treatment on the final strength of beech joints is shown in

Graph. 3. It can be noted that the planned surfaces give slightly better results than sanded surfaces, for welding times of 4 and 5 s. It seems to be possible that the sanding treatment reduces the wood roughness and thus the friction coefficient of the welding surfaces. In these conditions, the chemical physical modifications leading to the microstructure development, probably do not happen in optimal thermal conditions.

6.1.1 Conclusions

Mechanically induced vibration welding of wood can yield bond lines of different strength according to the wood grain orientation in the two surfaces to be bonded. Thus, longitudinal grain bonding of tangential and radial wood sections gives different strength results of the joint, although such a difference is not excessive and is limited to roughly 10%. Cross-grain bonding, hence bonding of radially cut sections, with the longitudinal wood grain of the two surfaces at a 90° angle to each other, yields much lower strength. This is about half that observed for pieces bonded with the grain parallel to each other. These differences can be explained by the law of fibre–matrix composites and by the very marked effect that homogeneity of fibre orientation is known to have on such composites. Oak has been found to yield lower results than beech and maple and to be more sensitive to welding conditions. Differences in both anatomical and wood constituent composition can account for this difference in performance. Furthermore, oak always presents joint bondlines where little or no increase in density at the interface is noticed. This is based on the different mode of bonding predominant in this species, while the other species present two different modes of bonding. Thus, both entangled fibre–matrix composite bondlines and direct welding of flat cell to flat cell occur. The latter is the predominant case for oak. Both cases and even hybrid cases between the two have also been observed in beech.

7 Results - Phase 2

Once established that mechanically-induced wood fusion welding can and does yield bonding results satisfying the relevant standard, the question that remained was: why? Additional analyses of the joints by scanning electron microscopy (SEM), by solid-state ^{13}C -NMR, by FT-IR and by thermomechanical analysis (TMA) were carried out to understand what occurred.

7.1 Solid state ^{13}C NMR

In Fig. 5 and Fig. 6 the CP-MAS ^{13}C NMR spectra of the wood of the bondline (less than 0.5 mm on each side of the bondline) of a wood fusion-welded beech joint welded for 3 s and held for 5 s (Fig. 5) compared to controls (Fig. 6) are reported. The bondline sample was obtained by sanding. The spectrum of the bondline Fig. 5 must be compared with the general CP-MAS ^{13}C -NMR spectra of beech controls (Fig. 6), one powdered by sanding and one ground into powder. It must also be compared with the CP-MAS ^{13}C -NMR spectrum of the considerable amount of excess fibres which are expelled from the joint sides during welding (Fig. 5). The spectra shown in Fig. 5 and Fig. 6 indicate that not much difference as regards the wood carbohydrates appears to exist in the 4 samples. As regards the bondline sample in relation to the controls one notes a slightly higher proportion of amorphous carbohydrates: the 83.29 ppm shoulder is more pronounced than in the two controls. Conversely, in the excess fibres expelled from the joints the 88.47 ppm peak belonging to the C4 of crystalline carbohydrates is clearly visible, different from the two controls, indicating a relatively higher proportion of crystalline cellulose in the excess fibres. The main differences between the four spectra are, however, in the smaller signals in the region 120–175 ppm, characteristic of some of the signals of lignin.

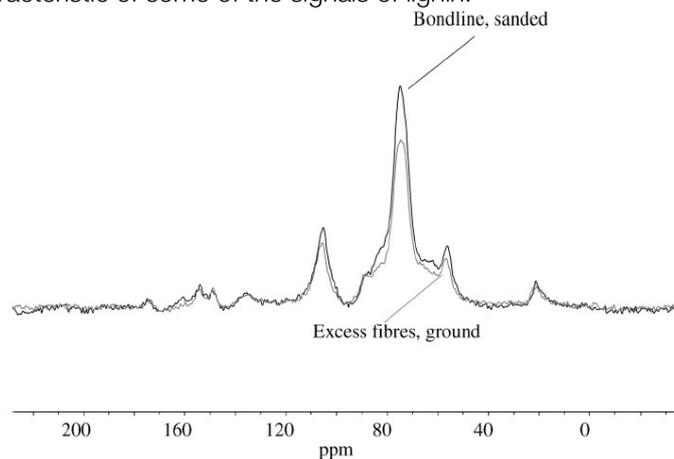


Fig. 5 Comparison by superposition of CP-MAS ^{13}C -NMR spectra of beech wood material in the bondline and excess fibres expelled from the bondline during welding.

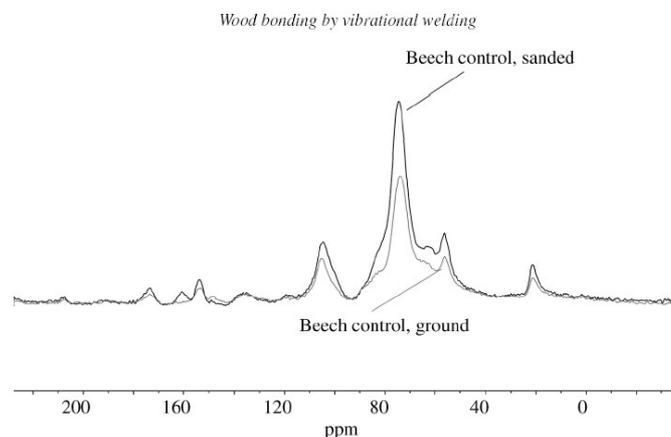


Fig. 6 Comparison by superposition of CP-MAS ^{13}C -NMR spectra of control beech wood samples. Samples were powdered by fine sanding or by fine grinding.

To facilitate observation of the differences in this region of the spectra their details are compared in Fig. 7. In Fig. 7 one can see a main peak, composed of three signals at 153.4, 153.9, 154.1 ppm, characteristic of the carbon of an aromatic ring to which is bonded a -O-, be it an -OH or an -OCH₃. The main lignin aromatic carbon to which a -OCH₃ methoxy group is bonded is noticeable at 161.2 ppm. The absence of this latter peak in the excess fibre and in the ground beech control indicates that lignin in the glue-line does not appear to be too heavily demethoxylated, while the fibres expelled during welding contain heavily demethoxylated lignin.

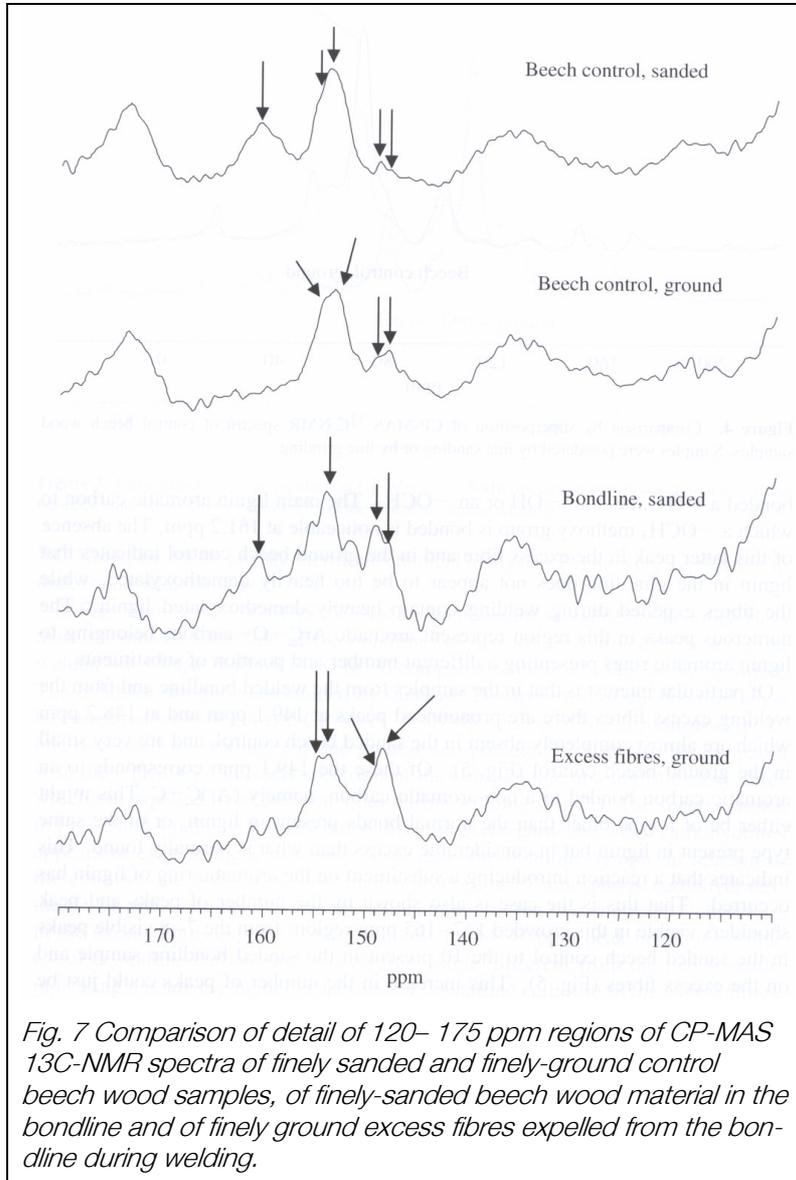
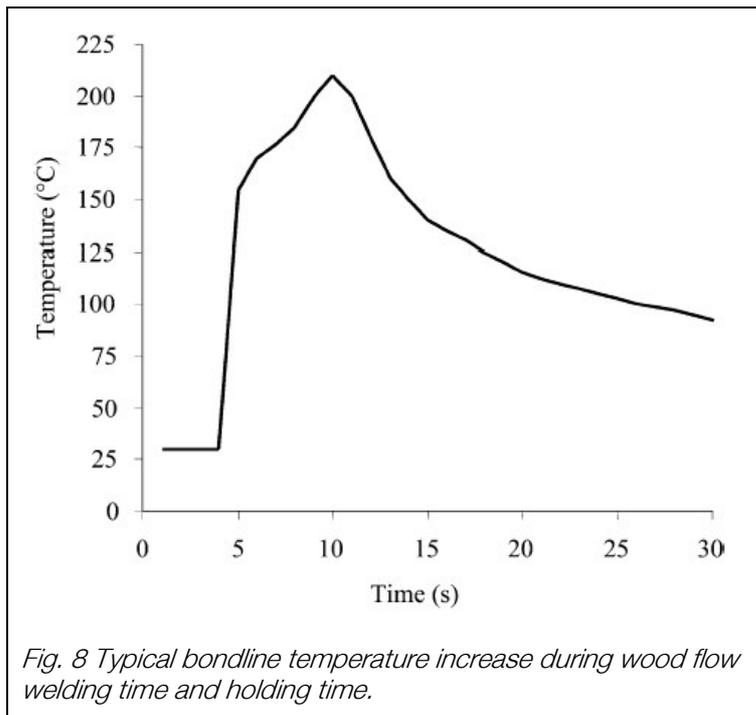


Fig. 7 Comparison of detail of 120–175 ppm regions of CP-MAS ¹³C-NMR spectra of finely sanded and finely-ground control beech wood samples, of finely-sanded beech wood material in the bondline and of finely ground excess fibres expelled from the bondline during welding.

the sanded bondline sample and on the excess fibres (Fig. 7). This increase in the number of peaks could just be due to part of the lignin being demethoxylated. Or, it could be due to peaks shifts rendering them more visible.

Most interesting is the peak at 148.2 ppm characteristic of the carbons of a furan ring (Fig. 7). Furfural, methyl furfural and other furanic materials are well known to be obtained from wood carbohydrates, possibly from hemicelluloses, by thermal treatment. The high temperature reached during welding (Fig. 8) indicates that the formation of furfural in the bondline may well be possible and is even quite likely and the presence of the 148.2 ppm peak appears to indicate its presence both in the bondline and in the excess fibres. The fact that this peak is more marked in the fibre samples, which have undergone a more drastic thermomechanical treatment, indicates that this is most likely to be the peak of the C5 of a furan. A peak of the same order of size should, however, show up in the 152–155 ppm region for the furan C2. Indeed, a number of peaks fulfilling this requirement are present although the most likely candidate peak is slightly more marked, as it is likely to be a superposition of the furan C2 signal on another signal.

The numerous peaks in this region represent aromatic ArC-O- carbons belonging to lignin aromatic rings presenting a different number and position of substituents. Of particular interest is that in the samples from the welded bondline and from the welding excess fibres there are pronounced peaks at 149.1 ppm and at 148.2 ppm which are almost completely absent in the sanded beech control, and are very small in the ground beech control (Fig. 7). Of these the 149.1 ppm corresponds to an aromatic carbon bonded to a non-aromatic carbon, namely (Ar)C-C. This might either be of a type other than the normal bonds present in lignin, or of the same type present in lignin but in considerable excess than what is normally found. This indicates that a reaction introducing a substitute on the aromatic ring of lignin has occurred. That this is the case is also shown by the number of peaks and peak shoulders visible in this crowded 147–165 ppm region: from the 7–8 visible peaks in the sanded beech control to the 10 present in



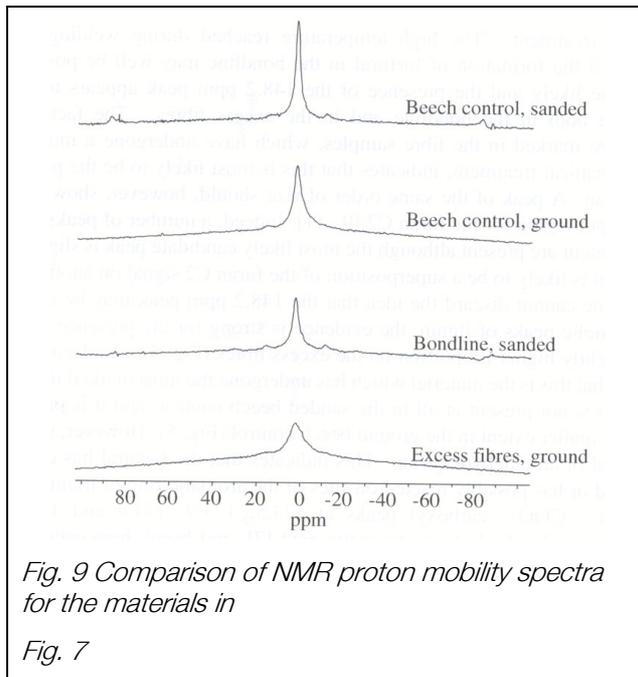
Although one cannot discard the idea that the 148.2 ppm peak may be due just to shifted phenolic peaks of lignin, the evidence is strong for the presence of a furan ring. Its slightly higher proportion on the excess fibres (Fig. 7) is understandable if one thinks that this is the material, which has undergone the most marked mechanical treatment. It is not present at all in the sanded beech control, and it is present to a very much smaller extent in the ground beech control (Fig. 7). However, the spectra lack a signal of an aldehyde group. This indicates that the furfural has either selfpolymerised or has possibly reacted on sites of the aromatic ring of lignin.

The clear $-\text{COO}-$ carboxyl peaks at 173.5, 173.9, 174.9 and 176.9 ppm belonging to acids, the last one to acetic acid, and beech hemicelluloses can be observed in the two controls, and decrease markedly in the bondline and the excess fibres samples. The number of peaks in the 170–180 ppm region, which is low for the two control samples, increases considerably for the bondline and excess fibres. In these cases among the multiple smaller peaks observed some of them have the shift characteristic of levulinic acid, known to be obtained from furfural by continuation of the reaction. This indicates that welding has heavily deacetylated the hemicelluloses and that it is this free acid that has helped to catalyze at higher temperature the formation of furanic compounds during wood welding.

In short, the results of the NMR analysis indicate that:

- A certain amount of lignin demethoxylation has occurred during welding.
- The proportion of amorphous carbohydrates appears to slightly increase during welding.
- An auto condensation reaction of lignin involving its aromatic rings appears to have occurred.
- Furfural appears to have been produced during welding and to have self polymerized or reacted with lignin aromatic nuclei, or both. The evidence for the existence of a furan ring might be weak, hence the idea that the 148.2 ppm peak may be due just to superposition of shifted phenolic peaks of lignin cannot be discarded. In either case cross-linking reactions subsequent to the initial welding are shown to occur.
- Some deacetylation of hemicelluloses appears to have occurred during welding, the free acetic acid possibly contributing to the formation of furanic compounds.

Notwithstanding the indication that some self-condensation and/or cross-linking reactions involving lignin and furfural clearly appear to occur, their limited extent does not appear to justify joint bond strengths in excess of 10 MPa as obtained in wood fusion welding. These reactions are surely a contributory factor, but they are unlikely to be the main cause of the bonding results obtained in mechanically-induced wood fusion welding. A first indication that the main cause of wood fusion welding might be a physicochemical effect rather than just a chemical cross-linking reaction comes from the proton mobility NMR spectra of the four main samples shown in Fig. 9.



The NMR proton signals show weak interactions of water with the wood. The intensity of the interaction increases as the peak becomes less sharp. Thus, for the first three cases in Fig. 9 the intensity of the water /wood interaction is the weakest for the sanded beech control, and the strongest for the bondline. The interaction of water with the excess fibres is even stronger as shown by the much broader proton peak in Fig. 9. The higher is the water interaction the lower is the T_g of hemicelluloses and lignin and the easier is to melt these two materials or for these two materials to flow. This means that particularly in the excess fibres and, to a lesser extent, in the bondline, melting of hemicelluloses and lignin appears to have taken place during welding, an occurrence that has clearly not taken place in the two controls.

7.2 SEM Scanning electron microscopy and TMA Thermo-mechanical analyses

If melting of some of the polymeric wood constituents occurs, this should be observed by SEM.

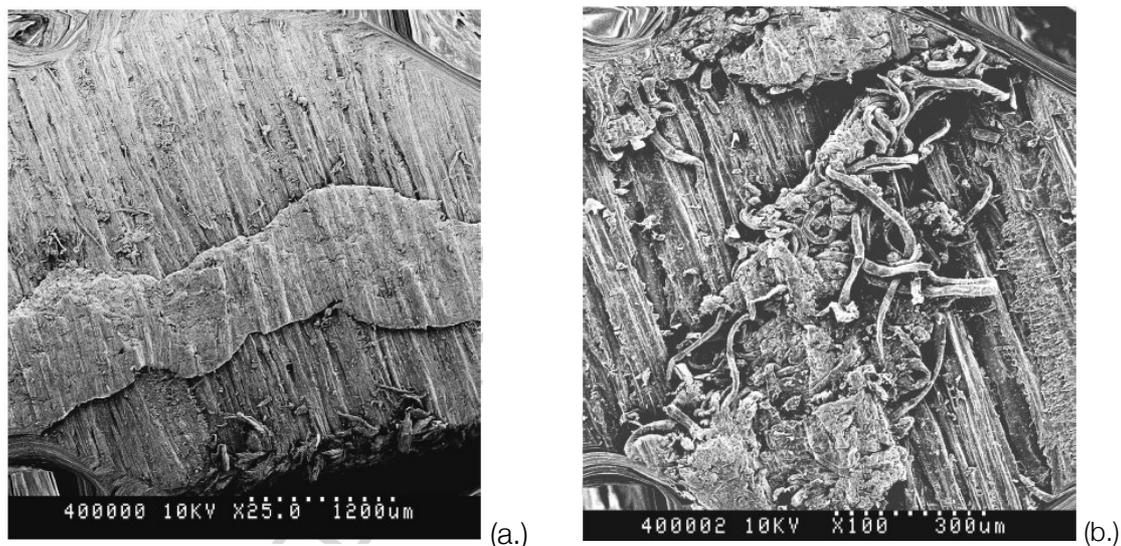


Fig. 10 Scanning electron microscopy images of lignin fusion band with cellulose fibers and wood cells (tracheids) immersed in it obtained during welding at (a) magnification = 25 \times showing general appearance of the band and (b) detail of the same at 100 \times magnification showing the entangled and detached tracheids, a fused intercellular lignin mass and tracheids and fibers immersed in the fused lignin matrix.

In Fig. 10a is shown one of the characteristic bands on the surface of fusion welded wood. One can see fibres, long wood cells or tracheids immersed in a mass of molten polymer. As the cells do not appear to be greatly damaged it means that melting has occurred mainly in the intercellular connecting tissue or *middle lamella*. Wood *middle lamella* is particularly rich in lignin, more than any other anatomical feature of wood. A more magnified detail in Fig. 10b of the same type of surface confirms the presence of a mass of entangled long wood cells immersed in a matrix of amorphous, fused intercellular material, almost certainly composed of lignin, possibly including some hemicelluloses. Figure 8a and b shows confirm this. In Fig. 11a, b the fibres are clearly undamaged, long wood cells (tracheids). This confirms that the molten matrix encasing them can only be formed of cell-interconnecting material, hence mainly from the lignin from the wood *middle lamella*. The amorphous material that is seen in patches on the surface of the undamaged tracheids

in Fig. 11b can only be part of the cells interconnecting material, after it has melted. Thus, the SEM observation clearly establishes that it is the wood *middle lamella* that melts, and this is mainly composed of lignin.

The SEM investigation thus confirms what was partly inferred from the CP-MAS ^{13}C -NMR spectra. The mechanism of mechanically-induced vibrational wood fusion welding is due mostly to the melting and flowing of the amorphous polymer material which interconnects wood cells in the structure of wood. This is mainly composed of lignin, but also some hemicelluloses. This causes partial detachment, the 'unglueing' of long wood cells (tracheids), that form an entanglement network drowned in a matrix of molten material, which then solidifies. In short, a wood cells entanglement network composite having a molten lignin matrix is formed. During the welding period some of the detached wood cells which are no longer held by the interconnecting material are pushed out of the joint as excess fibres.

This is not, however, the complete story. The CP-MAS ^{13}C -NMR spectra may indicate that some chemical cross-linking reaction of lignin and of carbohydrate derived furan also occurred.

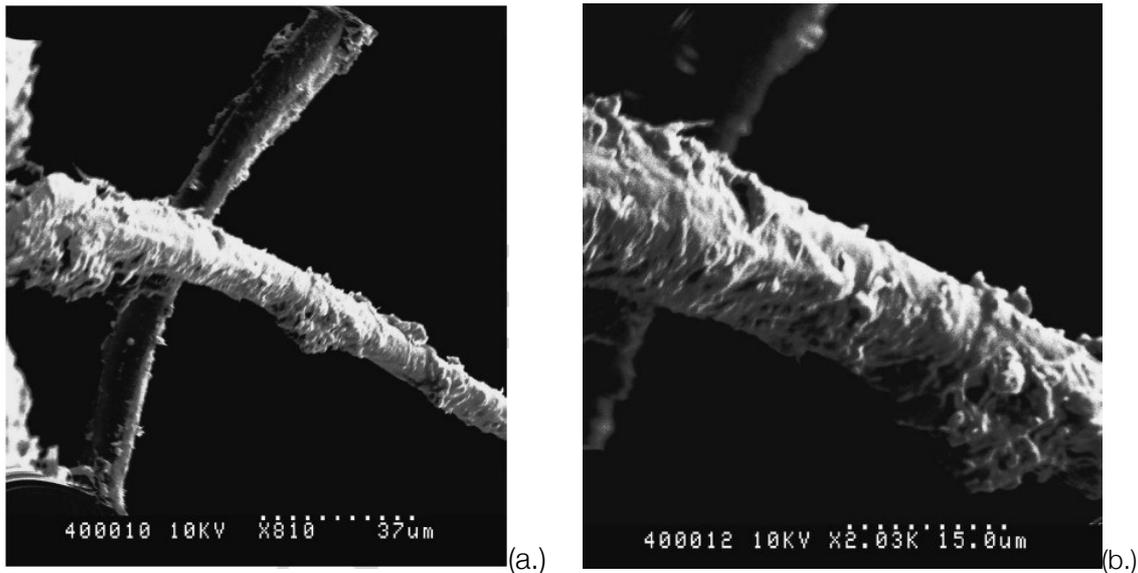


Fig. 11 Scanning electron microscopy images of a single filament of a detached wood cell (tracheid) from the excess fibres expelled during welding from the bondline at (a) 810 \times magnification and (b) at 2000 \times magnification. Note the residual fused amorphous material clinging to the tracheid's surface. This is the residue of the intercellular bonding material holding wood cells together.

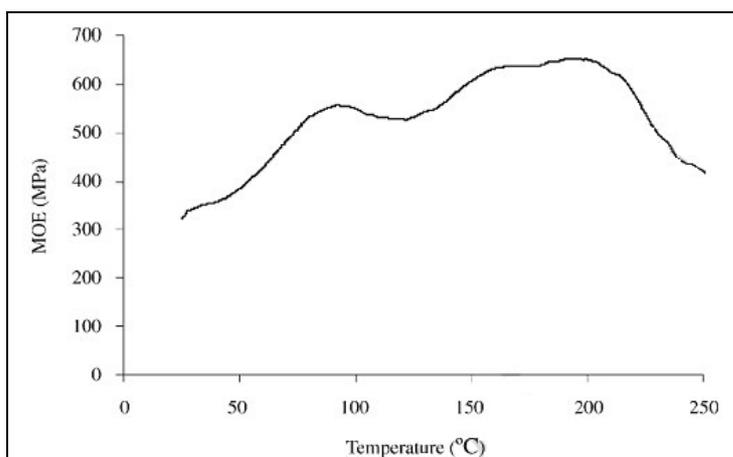


Fig. 12 An example of the increase of the Modulus of Elasticity (MOE) as a function of temperature obtained by thermomechanical analysis, at a constant heating rate of 10.C/min, of a beech wood joint obtained by mechanically-induced wood fusion welding. The MOE increases with the temperature indicating continuation of cross-linking reactions after welding.

These contributed to the mechanical resistance of the joint, although to a minor extent. In this regard, the results of constant heating rate thermomechanical analysis (TMA) of welded bondline joints yield a marked increase of the modulus of elasticity (MOE) of the joint at increasing temperature (Fig. 12). The increase occurs in two distinct steps, indicating a distinct series of chemical reactions, which go to completion only after welding has occurred. Fig. 12 shows that the distinct series of chemical cross-linking reactions contribute further to the strength of the joint, but their contribution comes to bear only after welding has finished. This might well be one reason why the longest holding time under pres-

sure after the end of welding contributes so markedly to the formation of a good bond. These cross-linking reactions might well be due to further polymerization and cross-linking of lignin and/or furfural formed during the initial welding period.

7.2.1 Conclusions

Interior grade solid wood joints have been obtained by vibrational welding without the use of any adhesive, opening the possibility for wood bonding without adhesive for furniture and for interior joinery. Melting mainly of lignin is one of the main causes of the bonding observed. The entanglement network encased in a matrix of molten intercellular material, mainly lignin, that has subsequently solidified on cooling constitutes the joint's bondline. Some of the excess wood cells that are no longer held by the interconnecting material are pushed out of the joint as excess fibres during welding. Some cross-linking chemical reactions appear to occur mainly after the welding action is finished, explaining why relatively longer holding times under pressure improve the joint strength. Polymerization and cross-linking of lignin and of carbohydrates-derived furfural appear to be the reactions involved.

7.3 X-ray micro densitometry of the welded line

In Fig. 13A and B X-ray micro densitometry images of two joints are shown, one of beech wood mechanically well welded (Fig. 13A) and the other of Norway spruce poorly welded (Fig. 13B). These images show clearly why spruce performs relatively poorly in relation to beech wood when mechanical wood welding is used. Beech joints are smooth when welded, while welded spruce joints have a very irregular interface. A similar irregular interface is always observed in spruce after any mechanical action, such as, for example, vacuum-pressure impregnation with wood preservatives, due to the phenomenon of wood cells collapse, which is characteristic of this wood species. It is evident that in the case of spruce wood either mechanical welding is not possible to any great extent or the conditions of welding have to be markedly changed from those used for a hardwood such as beech. The three-dimensional map in Fig. 13C represents the beech joint in Fig. 13A. Fig. 13C clearly shows that a marked increase in density has occurred at the mechanically welded interface. This is one of the main causes of the joint's high strength. Thus, from a density between 700 and 800 kg/m³ of normal beech wood the welded interface reaches density values of 1000 kg/m³, and even higher in some specimens. Wood strength is strongly density-dependent. The loss of the cellular structure in wood during mechanical welding with the consequent loss of the empty cellular spaces is the cause for this density increase. The increase in density at the welded wood interface and the wood density map of a joint are then measures of how good the joint is likely to be.

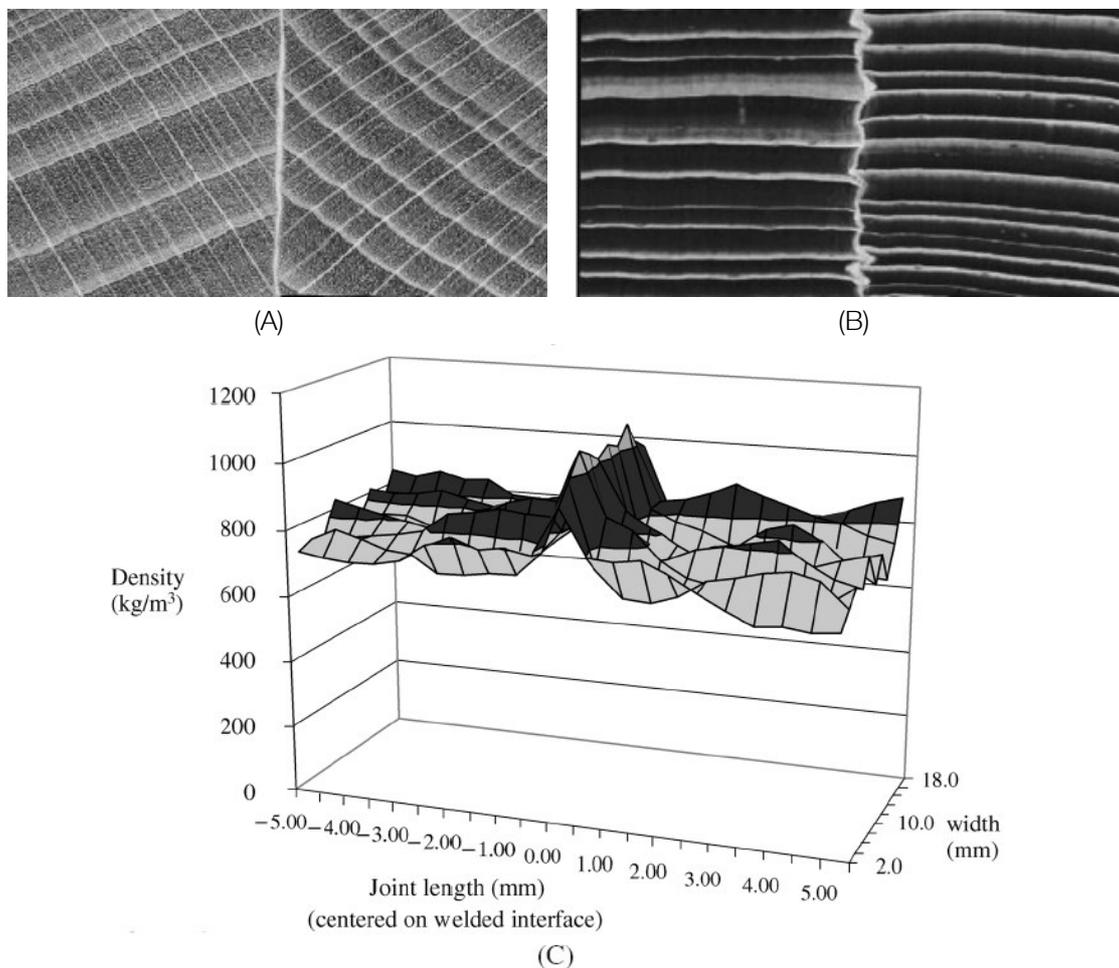
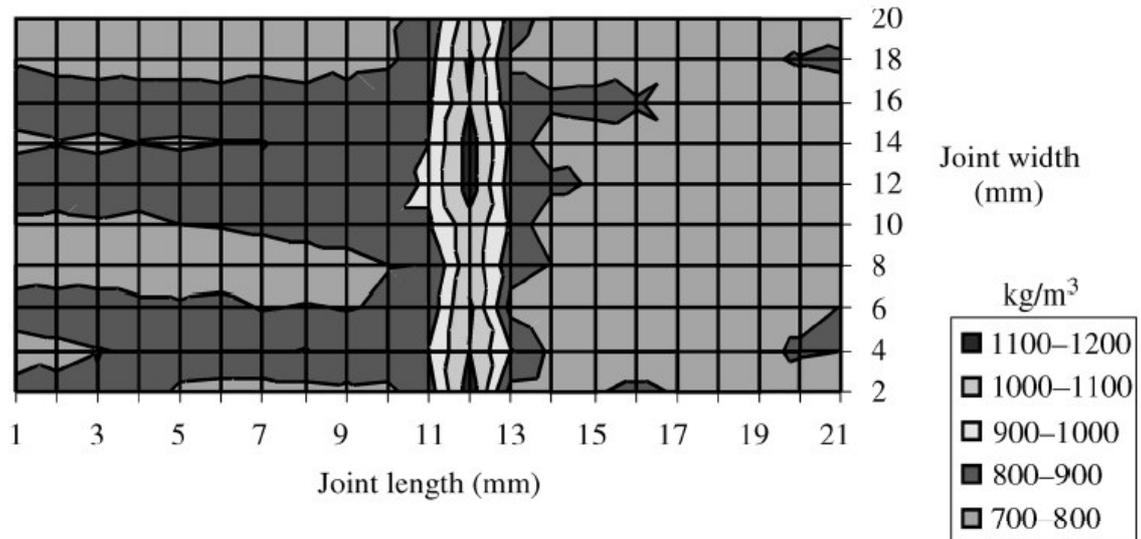


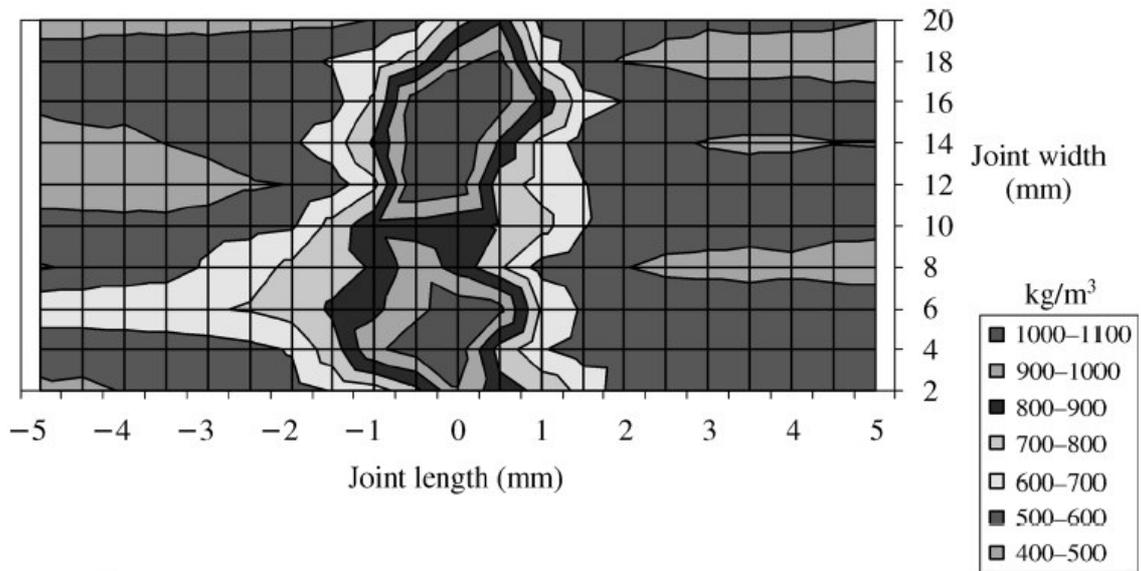
Fig. 13 X-ray micro densitometry photographs of (A) well-bonded vibration-welded beech wood and (B) poorly-bonded vibration-welded Norway spruce wood. Note the finger jointing effect in (B), due to spruce cellular collapse as a consequence of the wide difference in density of the latewood and early wood growth rings of this species. (C) Three-dimensional wood density map of the well-bonded vibration-welded beech wood in (A).

For each joint section, the density values were measured by centering one regular network of lines on the welded interface. This network consisted of 10 horizontal lines and 100 vertical lines. The average wood-density value is calculated at each intersection point between the above-mentioned vertical and horizontal lines. The horizontal resolution is, therefore, 0.3 mm while the vertical one is 2 mm. These wood-density values calculated by the CERD software are printed in one flat ASCII data file and can be displayed as illustrated in Fig. 13C. In this figure the vertical axis represents the wood density expressed in kg/m³ while the two other axes represent the positions in the section as represented in Fig. 13A and Fig. 13B. In the middle of the sample, i.e. exactly on the welded bond, one can notice a strong increase in wood density value.

This increase in density is the result of the welding process itself and the assumption has been made that this increase in wood density can be seen as one quality criterion of the welding process. In order to illustrate the results, the wood density maps for two examples of beech and spruce have been reported in Fig. 14A and Fig. 14B.



(A)



(B)

Fig. 14 Maps of actual density values in kg/m³ of (A) well-bonded vibration-welded beech wood in Fig. 3 A and (B) the poorly-bonded vibration-welded Norway spruce wood in Fig. 13B.

For each species the sections obtained are analyzed in terms of wood density increase around the welded bond in order (i) to display the area where the densification process occurs and (ii) how constant it is along the sample. Figure Fig. 14A and Fig. 14B are top views of the wood-density variations as they are represented in Fig. 13C. The welded bond is represented in the mid-

dle of each figure. For the beech samples the increase in density is limited to one small area close to the bond (interface is about 0.6 mm thick). The average wood density of the three sections is about 760 kg/m³ for beech while the maximum wood density attained in the bond is about 1100–1200 kg/m³. The ratio between these two values is about 1.58 and expressed in mm of bond thickness it is about 2.63 mm.

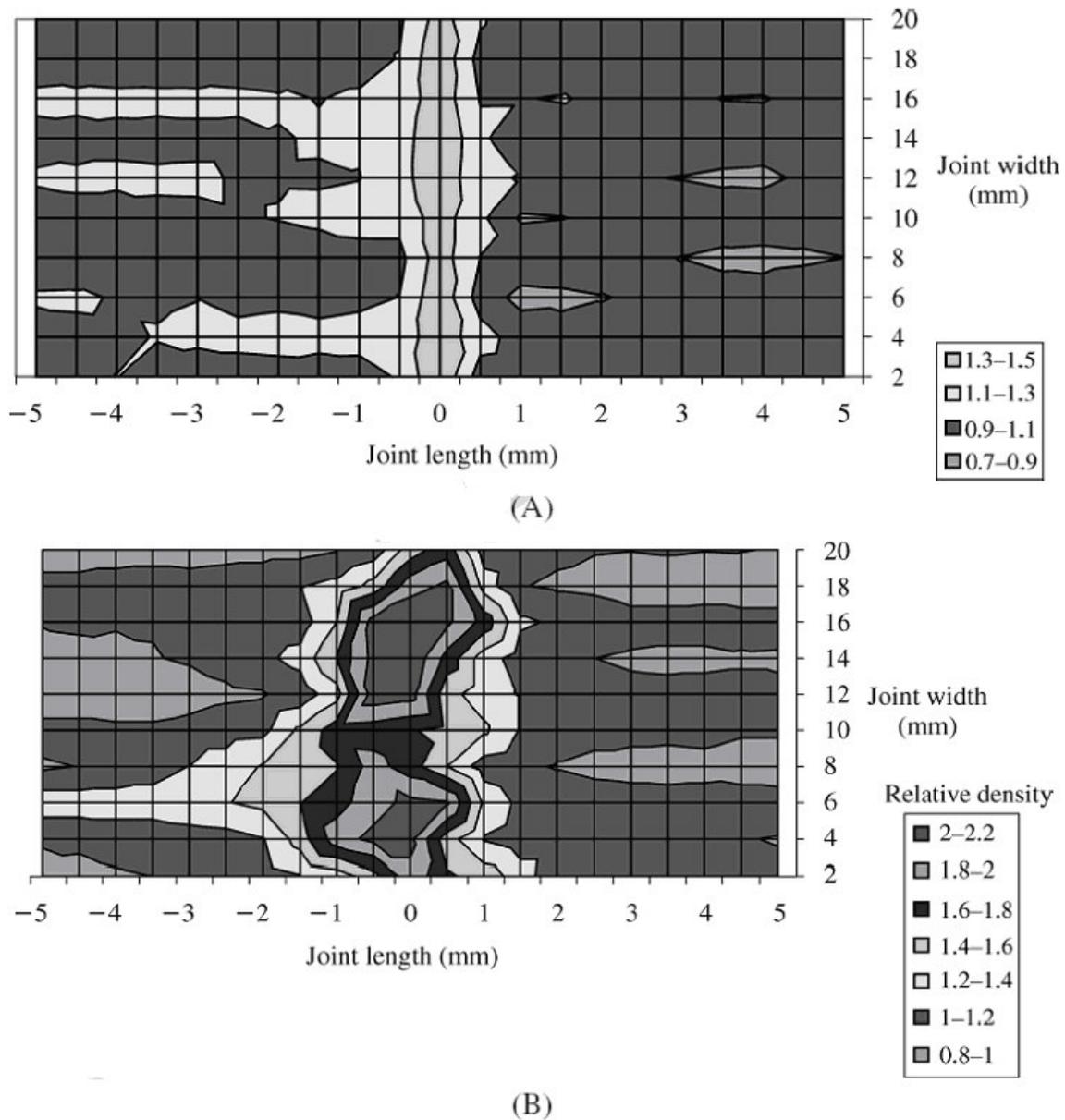


Fig. 15 Relative density maps of (A) well-bonded vibration-welded beech wood in Fig. 1A and (B) poorly-bonded vibration-welded Norway spruce wood in Fig. 1B.

Fig. 14B and Fig. 15B for spruce exhibit very different patterns. As in the case of beech, the welded bond is located along the vertical line at the center of the specimen. Here, the area where wood density has increased is wider than for beech. The shape of this area looks different, mainly because of the heterogeneity of this softwood species, especially because of large variation of wood density between latewood and early wood. This point is very clearly illustrated in the picture for spruce in Fig. 13B. In this picture one can see that the latewood parts of one sample have compressed the early wood parts of the other sample and, as a consequence, a typical 'finger jointing' pattern is obtained. This explains why the welded bond area has an irregular shape in terms of wood density. Even if this shape is not as well defined as for beech, which is a very homogeneous hardwood, the interface is 1.8 mm thick. If we calculate as for beech the ratio of maximum wood density to average wood density, we obtain a ratio of 2.35 (1200/510) and this ratio expressed in terms of mm of thickness of the bonded area drops to 1.30 which is about 50% lower than for beech.

X-ray-derived wood-density maps can be presented in a planar projection both as absolute density maps or as relative density maps. In Fig. 14A and Fig. 14B the absolute density maps for the well-welded beech wood joint in Fig. 13A and for the poorly welded Norway spruce joint in Fig. 13B are presented. The difference in the central zone for the absolute density maps for beech and spruce in Fig. 14A and Fig. 14B clearly shows the evenness and narrowness of the high density zone of the well-welded beech joint, and the very irregular, very broad and malformed interfacial zone of the poorly-welded spruce joint. In Fig. 14B, one can see that welding in one 'spot' has occurred also for spruce but this constitutes only approx. 20% of what should have been welded. In reality, a direct comparison of different wood species is risky when using absolute wood density maps. These maps are useful because they indicate the real densities of the various components of each joint, but this does not constitute a good basis for comparison when very different species such as hardwood and softwood are compared.

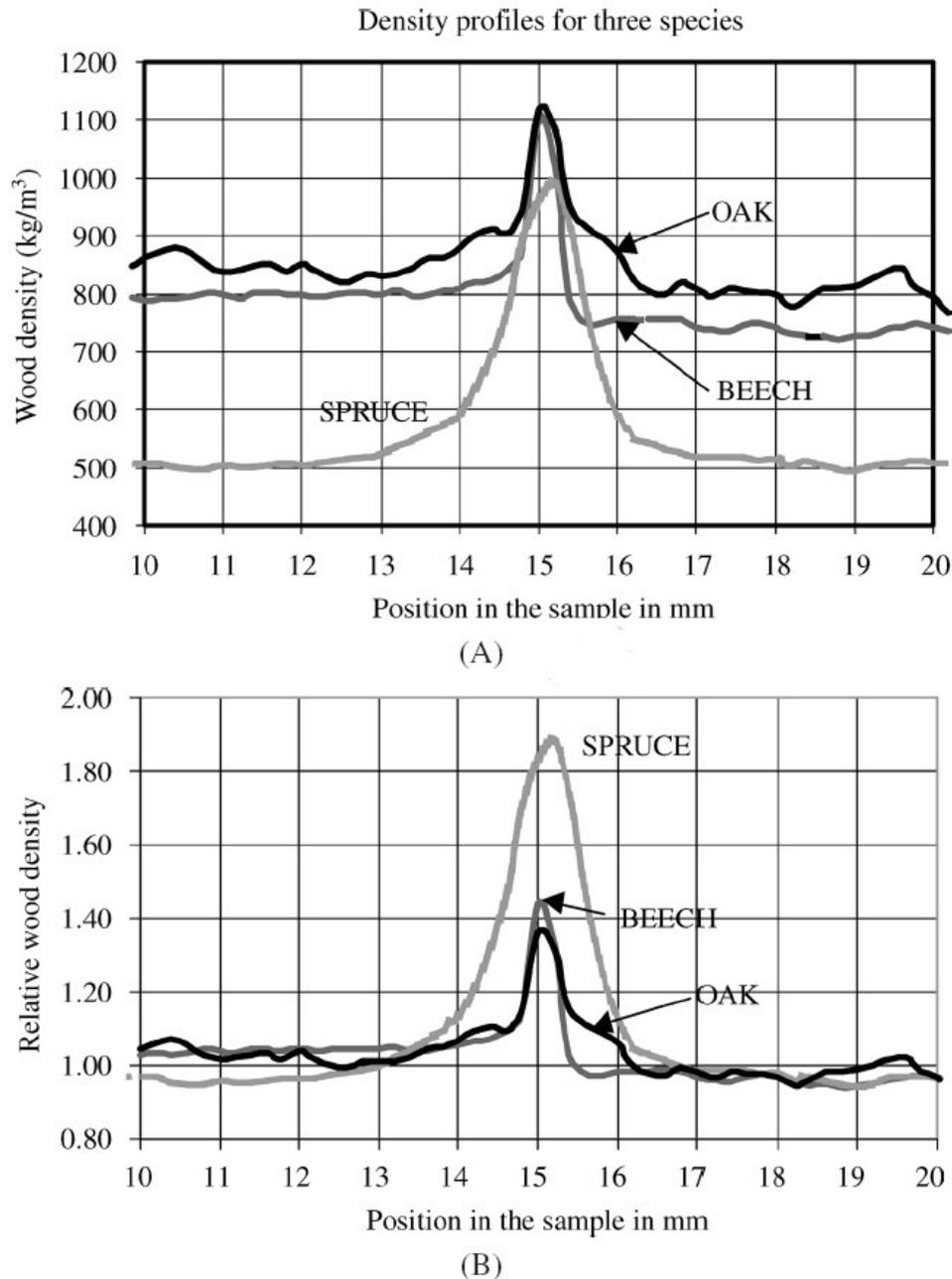


Fig. 16 Density profiles for beech, oak and spruce allowing comparison of their performances. (A) Actual density profiles, (B) relative density profiles.

Relative density maps obtained by dividing density values by the mean density of the specimen are more appropriate instead for interspecies comparison. The relative density maps corresponding to the specimens shown in Fig. 14A and Fig. 14B are shown in Fig. 15A and Fig.

15B. The regularity and irregularity of the well-welded interface and poorly welded interface, respectively, are clearly visible in these maps too. The conclusions, which can be drawn, are the same. The difference between the absolute and relative density maps is illustrated in Fig. 16A and Fig. 16B. Fig. 16A and Fig. 16B show, respectively, the absolute and relative density profiles of welded joints of beech, spruce and oak. Fig. 16A shows the real density values of the wood away from the interface and at the interface of the welded joint. Thus, oak and beech are clearly shown as higher density timbers than spruce. Fig. 16B instead reports the density of the timber to a normalized value, rendering comparison easier.

An oak welded joint is shown in Fig. 17A and Fig. 17B. The interfacial bond line is well formed and regular, although not quite as sharp and narrow as that of well-welded beech joints. One can observe from Table 1 and Fig. 15A, Fig. 15B and Fig. 17B that the relative strengths of the welded joints are in direct relation with the evenness and narrowness of the interfacial bondline. The narrower and more even the bondline, the better the mechanical performance of the joint appears to be. Thus, in Table 1 the dry strengths of the beech, oak and spruce joints shown, respectively, in Fig. 15A, Fig. 15B and Fig. 17B are reported and indicate that the narrowest and broadest bondlines correspond, respectively, to the strongest and the weakest joints.

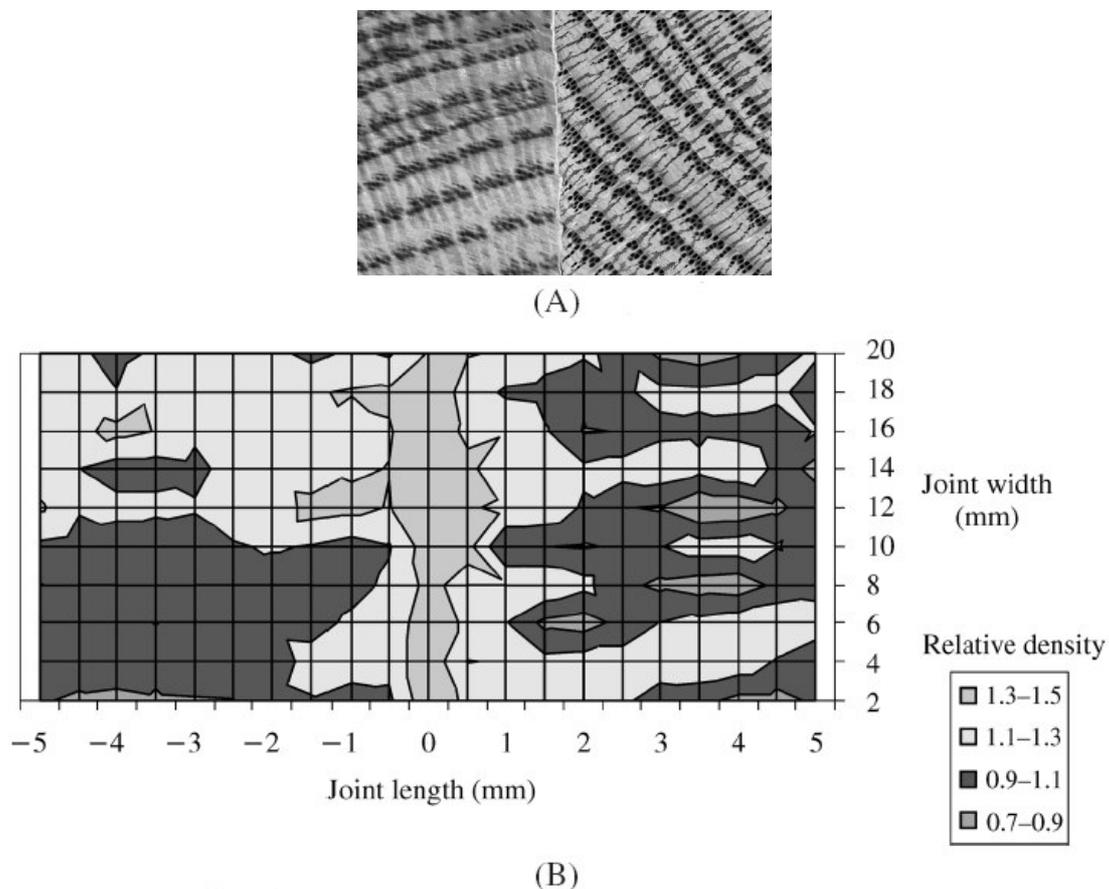
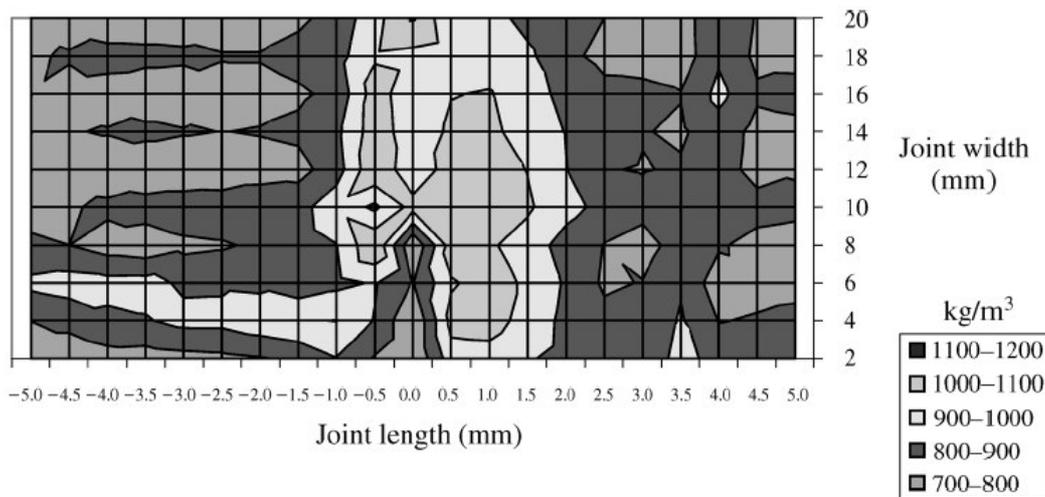


Fig. 17 X-ray microdensitometry (A) photograph of well-bonded vibration-welded oak wood and (B) its relative density map.

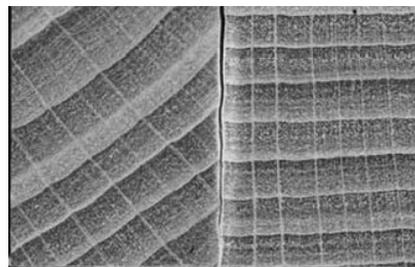
Observing Fig. 16A it is noticeable that the bondline (the peak in Fig. 16A) is broadest at the base for spruce and narrowest for beech. This indicates that the bondline formed in beech by the fusion of middle lamella material is more compact and localized than in spruce. In Fig. 18A-C a specimen of a beech joint welded at one end and unwelded at the other is shown. The density maps clearly indicate where the specimen is welded and where it is not. Thus in the planar density map (Fig. 18A) one can readily see the discontinuity due to the absence of welding which can be observed in Fig. 18B. This becomes even clearer in the three-dimensional density map (Fig. 18C), where the higher density peak at the interface indicates where the specimen is welded. Where the specimen is not welded peaks indicating a density much lower than the average of the specimen can be seen.

Table 1.
Joint strengths of vibration-welded wood joints composed of different wood species

Wood species	Average joint strength (MPa)	Best result joint strength (MPa)
Beech	8.72	11.22
Oak	5.43	7.39
Spruce	4.20	4.96
Mixed Spruce/beech	4.41	6.78



(A)



(B)

Fig. 18 X-ray micro densitometry. (A) Relative density map of partially-welded beech showing the welded and unwelded parts. (B) Photograph of partially-welded beech showing joined and not joined regions of the specimen.

Mechanical welding has also been attempted between surfaces belonging to two different wood species of different characteristics, namely a hardwood (beech) and a softwood (spruce). The results obtained are shown in Fig. 19A and Fig. 19B. In Fig. 19A one can note that spruce shows the same jagged appearance of the bondline due to the collapse of the softer early wood rings and the lack of this for the much harder latewood rings. More interesting is the appearance of the beech interface which is rather jagged (but much less than that of spruce) and is not as smooth as that in Fig. 13A characteristic of beech/beech welded joints. The indentations seen in Fig. 19A on the beech surface correspond, in general, to the pressure exercised by the higher density latewood growth rings of spruce onto the softer early wood rings of the other surface (beech). The density map in Fig. 19B further explains what occurs: the discontinuity at the bondline due to the considerable difference in average density of spruce and beech causes mainly to fuse the spruce early wood part and is densified on the beech surface. It is easy to see that at the bondline the density of the spruce interface is much higher than the density in other zones of the same timber. In short, it is the lower average density spruce that mainly crushes, fuses and welds onto the much higher density beech surface. Beech does contribute more to the interface strength (Fig. 19B) as seen by the comparison of spruce-spruce, beech-spruce and beech-beech joint strengths in Table 1. However, the process is mainly dominated by what happens to the lower-density wood, as

shown in Fig. 19B and Table 1. In Table 1 the strength of the mixed joint is better than in the case of spruce alone, but only approx 5% (average strength) and up to 35% (best result). The values obtained are lower than the value for beech joints only, confirming the limited contribution of beech to welding and to mix joints strength.

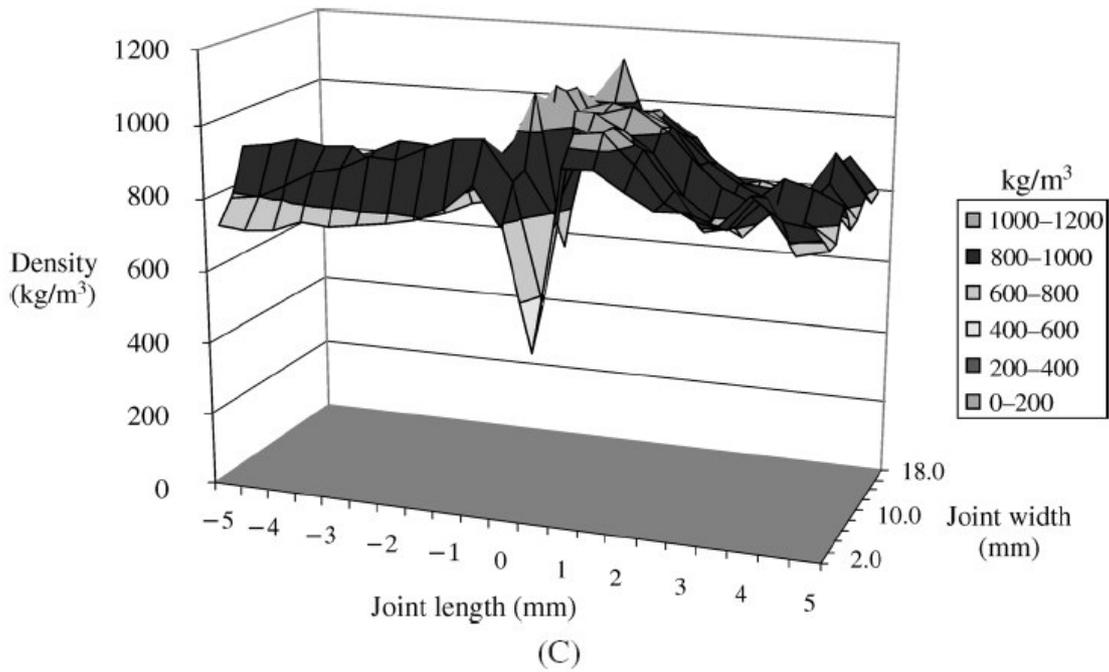


Fig. 18C-. Three-dimensional wood-density map of the same joint clearly showing high-density peaks where the wood is joined and a peak of density much lower than the average wood specimen where the joining is poor or non-existent.

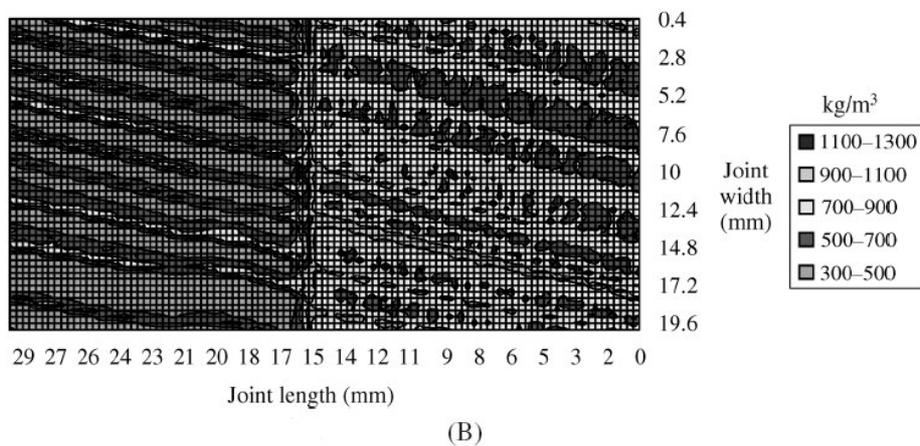
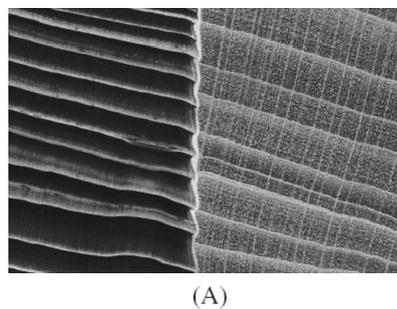


Fig. 19 ray microdensitometry results of a welded wood joint of mixed wood species (beech and spruce). (A) Photograph of vibration-welded spruce and beech. (B) Map of actual density values in g/m3 of the mixed joint.

7.3.1 Conclusions

In conclusion, X-ray micro densitometry has been shown to be a valuable technique to determine the extent of wood welding. It is capable to give an indication of both the morphology of the bondline, as well as the strength of the welded joint obtained. Analysis of welded joints using three different types of wood showed that the morphological differences observed in the final joint depend quite markedly on the density and evenness of the bondline, and that these show qualitative trends similar to those observed for the strength of the welded wood joints. Spruce was shown to give inferior welding results due to its characteristic cells collapse. Spruce contribution in a mixed hardwood/softwood joint then limits the maximum joint strength achievable. The relatively poor performance of spruce is only characteristic of this species and not of softwood in general. Work in progress at present has shown that results as good as those obtained with beech wood can also be obtained with softwood other than spruce. The wood-grain orientation of the two surfaces to be bonded appears to have an influence, although not a determinant one. Thus, the present study has been limited to the average of results of a balanced mix of different grain orientations to minimize any influence this could have. A comprehensive study on the effect of grain orientation on wood welding is in progress.

8. General conclusions

Mechanically induced wood fusion welding, without any adhesive at the interface, yields rapidly bonding joints satisfying the relevant requirements of non-structural wood joints. This technique opens the door to effective and inexpensive gluing of solid wood without adhesive for furniture and interior joinery. The mechanism of mechanically induced vibrational wood fusion welding is due mostly to the melting and flowing of amorphous cells-interconnecting polymer material in the structure of wood, mainly lignin, but also some hemicelluloses. This causes the partial detachment, the "ungluing," of long wood cells and the formation of an entanglement network drowned in a matrix of melted material which then solidifies, in short a wood cell entanglement network composite with a molten lignin matrix. During the welding period some of the detached wood cells which are not being held anymore by the interconnecting material are pushed out of the joint as excess fiber. Cross linking chemical reactions of lignin and of carbohydrates-derived furfural also occurred but are minor contributors during the very short welding period. Their contribution increases after welding has finished, explaining why relatively longer holding times under pressure after the end of welding contribute strongly to obtaining a good bond.

The results of study show also that the process can yield bondlines of different strength according to the wood grain orientation in the two surfaces to be bonded. Thus, longitudinal grain bonding of tangential and radial wood sections gives different strength results of the joint, although such a difference is not excessive and is limited to roughly 10%. Cross-grain bonding, hence bonding of radial cut sections, with the longitudinal wood grain of the two surfaces at a 90° angle to each other, yields much lower strength. This is about half that observed for pieces bonded with the grain parallel to each other. These differences can be explained by the law of fibre-matrix composites and by the very marked effect that homogeneity of fibre orientation is known to have on such composites. Oak has been found to yield lower results than beech and maple and to be more sensitive to welding conditions. Differences in both anatomical and wood constituent composition can account for this difference in performance. Furthermore, oak always presents joint bondlines where little or no increase in density at the interface is noticed. This is based on the different mode of bonding predominant in this species, while the other species present two different modes of bonding. Thus, both entangled fibre-matrix composite bondlines and direct welding of flat cell to flat cell occur. The latter is the predominant case for oak. Both cases and even hybrid cases between the two have also been observed in beech.

8 Literature

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9 Bestimmungen zu diesem Forschungsbericht

Der Bericht wurde klassiert als: Öffentlich

Dieser Bericht darf nicht ohne Genehmigung der HSB auszugsweise vervielfältigt werden. Jegliche Veröffentlichung des Berichtes oder Teilen davon bedarf der schriftlichen Zustimmung der HSB.

Ein Original dieses Berichtes wird von der HSB für 5 Jahre aufbewahrt.

Dieser Bericht ist nur mit den Unterschriften des Projektleiters und des Projektverantwortlichen gültig.

9.1 Umfang des Forschungsberichtes

Dieser Forschungsbericht besteht aus dem Titelblatt, dem Abstract und 30 Seiten inkl. Anhang.

Biel, 10.12.2004

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