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Lignin as binder in particle boards using high frequency heating

Properties and modulus calculations

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This paper reports the results of a study exploring the possibility of employing unmodified Kraft (sulphate) lignin as a binder in particle boards manufactured using high frequency (27 MHz) heating. The mechanical parameters of the samples varied substantially with their density. For instance, the modulus of elasticity increased from 2.5 to 6.1 GPa when density changed from 700 to 1200 kg/m^3. Similar results were obtained when replacing the lignin component with some thermoplastic binders (PVCAC, PVC, LDPE, PA 11, and an acrylic copolymer).

The water resistance of the samples was relatively low. For instance, a sample containing 20% lignin and having a density of 700 kg/m^3 exhibited after 2 h exposure to water a thickness swelling of 40%. With a similar sample containing PVCAC as binder, the corresponding value was 13%. It was possible to reduce the degree of swelling with hydrophobizing additives or by using acetylated wood flakes. An addition of 5% of a polyethylene wax thus reduced the swelling of lignin bound samples to below 5%. Similar results were obtained with PVCAC as binder. Supplementary experiments were carried out with adding hollow microspheres (Expancel) to the material. Reduced swelling was recorded also in this case.

Predictions of the modulus values based on the Halpin-Tsai concept produced a fair degree of agreement with the experimental data, provided the density and the binder content were sufficiently high.

Lignin als Bindemittel für Spanplatten unter Verwendung von Hochfrequenz-Erhitzung. Eigenschaften und Berechnung des Moduls

Unmodifiziertes Lignin aus dem Kraft-(Sulfat)-Aufschluß wurde zur Herstellung von Spanplatten mit Hilfe einer Hochfrequenzerhitzung bei 27 MHz verwendet. Die mechanischen Eigenschaften der Proben variierten sehr stark entsprechend ihrer Dichte. Z.B stieg der E-Modul von 2,5 auf 6,1 GPa, wenn die Dichte von 700 auf 1200 kg/m^3 erhöht wurde. Ähnliche Ergebnisse wurden erzielt, nachdem die Lignin-Komponente durch einige thermoplastische Harze wie PVCAC, PVC, LDPE, PA 11 und ein Acryl-Copolymer ersetzt wurden. Die Feuchtebeständigkeit der Proben war relativ gering. So wies eine Probeplatte mit 20% Ligninanteil und einer Dichte von 700 kg/m^3 eine Dickenquellung von 40% auf. Bei einer ähnlichen Probe, die PVCAC als Bindemittel enthielt betrug die Dickenquellung nur 13%. Die Quellungseigenschaften konnten deutlich verbessert werden durch Zusatz von Hydrophobierungsmitteln. Die Zugabe von 5% Polyethylen- Wachs reduzierte die Quellung der ligningebundenen Platten auf unter 5%. Ähnliches gilt für PVCAC. Ergänzende Untersuchungen wurden unter Zugabe von Microballoons (Expancel) durchgeführt. Auch damit konnte die Quellung herabgesetzt werden. Die Berechnung der Festigkeits-eigenschaften nach dem Halpin-Tsai-Modell führte zu einer annehmbaren Übereinstimmung mit den experimentellen Daten, wenn die Dichte und Harzgehalt ausreichend hoch waren.

1
Introduction

This communication presents the results of an experimental study concerning the possibility of using high frequency (HF) heating in the production – on a laboratory scale – of particle board and similar wood fibre based materials. The wood flakes employed in this investigation are normally used in the top layer of conventional particle boards.

Although boards can be formed by pressing the flakes in an HF field, such products are found to possess inferior strength and swelling properties. As in normal production, it is therefore necessary to use suitable adhesives. This work is primarily concerned with the use of unmodified Kraft (sulphate) lignin as the bonding agent. Apart from lignin, some thermoplastic binders, in the first place PVC, were employed in order to improve the water resistivity of the wood/lignin composites. Hydrophobic additives were used for the same purpose, as were also acetylated wood flakes. Although the literature contains a relatively large number of reports on the use of sulphonated lignin and spent sulphite liquor as binders, there appear to be no studies describing the use of unmodified Kraft (sulphate) lignin. When such lignin is being employed, it is always as a reactive component replacing phenol in phenol-formaldehyde resins.

The possibility of using HF heating in the manufacture of particle boards bonded with conventional thermosetting resins (UF or PF type) is well documented. However, the practical significance of this technique is limited. HF has been used for pre-heating the boards in order to shorten the pressing time and increase the press capacity (Fahey 1976; Stevens and Woodson 1977). Some patents cover the use of HF as the sole heat source during the pressing operation, preferably with phenol-formaldehyde resin as the bonding agent (Swed. Pat. Appl. 1978, 1986). In other areas of wood technology, the use of HF heating is a well established technology (Yavorsky 1954; Mann 1954; Estep 1954; Miller and Cole 1957, Miller and George 1965). This is, for instance, the case with gluing of veneers (Mann 1954).

It is known that cellulose can, in principle, attract sufficient amount of energy to attain temperatures where it begins to decompose. This also applies to different wood species. The dielectric parameters determining the extent of heat dissipation in a wood sample are amply documented in the literature. In general, the power N dissipated in a unit volume is given by the formula (Kollmann 1955; Menges et al. 1980)

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$$N = 0.556\, E^2 f \varepsilon \tan\delta \times 10^{12}\ \mathrm{W/cm^3} \qquad (1)$$

where E is the effective value of the AC voltage, f the frequency, ε the relative dielectric constant, and $\tan\delta$ the loss factor. Given the voltage and frequency, the dissipated power is thus determined by the product $\varepsilon \tan\delta$. The system electrodes/wood can be considered as equivalent to a circuit consisting of a condenser in parallel with a resistance. The current through the system is the sum of the currents flowing through the two parts of the circuit; its magnitude is determined by the quantities entering Eq. (1).

In wood, ε and $\tan\delta$ depend on the humidity content (Lin 1967; James 1977; Nanassy 1970; Kröner and Pungs 1952; Trapp and Pungs 1956). For pine wood (Pinus silvestris), for instance, values of ε ranging from about 8 to 15 have been reported when humidity content was increased from 15% to 45% (Kollmann 1955). These values relate to a frequency of 10 MHz. For dry birch wood, ε amounting to 2.2 has been found at the same frequency increasing to 2.6 at 10.7% humidity content. On the whole, ε values around 3 are normally recorded for various wood species at 10 MHz and 0–15% RH (Kröner and Pungs 1952).

The loss factor $\tan\delta$ also increases with humidity. As an example, we mention that $\tan\delta$ values for 30 different kinds of wood increase from about 0.03 to about 0.08 when the humidity content is raised from zero to 16% (measurements perpendicular to fibre direction, 15 MHz) (Kollmann 1955). The above values are given here as illustrations only; it is not possible to depict the complicated interplay of the various factors affecting the dielectric parameters in terms of a few numerical figures.

Apart from the humidity, also the temperature plays an important role. At constant humidity, the loss factor decreases significantly at elevated temperatures, for birch wood at 10 MHz from about 0.06 to about 0.01 in the range 20–140°C at 10% constant humidity content (Kollmann 1955). Another parameter influencing the dielectric behavior is the density. The value of ε for instance, increases linearly with this quantity, the slope of ε vs. density lines depending on the humidity (Kollmann 1955).

The frequency of the HF field influences the dissipated power in two ways, directly as evident from Eq. (1), and indirectly via its influence on the values of ε and $\tan\delta$. As known for other solids, ε decreases with increasing frequency. In ε-$\log f$ diagrams, this decrease, when covering sufficiently large f ranges, has a sigmoid shape characteristic of a dispersion region. In accordance with this, $\tan\delta$ shows a maximum. It may be noted that according to literature data on spruce wood and cellulose this maximum is located in the vicinity of the frequency used in the present case (27 MHz) (Kollmann 1955).

Lignin behaves somewhat differently, since its $\tan\delta$ increases less than for wood with frequency (from 0.008 to 0.02 between 300 Hz and 10 MHz). On the other hand its ε value is somewhat higher than that of wood and cellulose, at least in the examples given by Kollmann (1955) and Venkateswaran (1972).

The product $\varepsilon \tan\delta$ determining the material dependent part of the dissipated power falls with increasing temperature (from 0.21 to 0.036 between 20° and 140°C for birch veneer). For a dry phenol-formaldehyde film, used in gluing veneers, it shows a maximum (0.55) at about 100°C (0.09 at 20°C, 0.19 at 140°C).

It thus follows that normal wood species and their constituents appear to have sufficiently high values of the product $\varepsilon \tan\delta$ in order to enable adequate heating of various composites, also when the adhesive agent used remains relatively inert towards the HF field, especially at higher initial humidity contents.

In a separate section, we present the results of theoretical calculations of the modulus of elasticity based on the Halpin–Tsai concept. At sufficiently high binder contents and densities, the predictions showed a fair degree of agreement with measured modulus values.

2
Experimental

2.1
The press

A schematic picture of the hydraulic press used is shown in Fig. 1. The HF electrodes were mounted on the press platens using a number of cylindrical insulators (glass reinforced bakelite). The electrodes, made of aluminium, had a rectangular shape with the dimensions of the sample (95 × 45 mm). They were preheated to 80°C in order to reduce the temperature gradient in the board samples. The frequency of the HF generator (FIAB System Co., type FS) was 27 MHz.

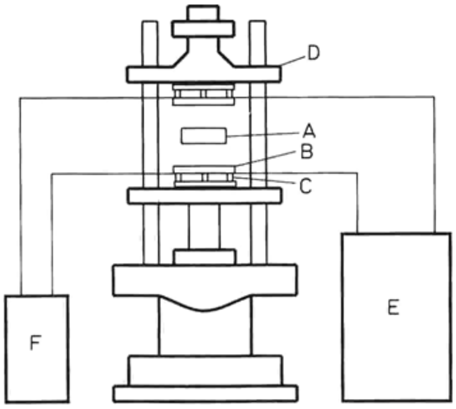

Fig. 1. Schematic picture of the hydraulic press. A – mould of PPS, B – HF electrodes, C – insulators, D – press, E – HF generator, F – heating unit circulating oil.

Bild 1. Schema der hydraulischen Presse A – Werkzeug aus PPS, B – HF Elektroden, C – Isolatoren, D – Presse, E – HF-Generator, F – Heizung für den Ölkreislauf

2.2
The mould

The mould was designed for compression moulding of rectangular plates, 95 × 45 mm, with varying thickness and density (depending on the pressure used). The frame of the mould was made of a high-temperature engineering plastic (polyphenylene sulphide, PPS, Ryton). Its inner surfaces were covered with 5 mm thick, hardened glass plates. The same cover was used for the electrode surfaces facing the sample. The role of the glass plates was to reduce the cooling of the sample during the HF heating thus diminishing the corresponding temperature gradients.

2.3
Materials used

The various materials used were as follows:

Cellulosic components

Wood flakes (60% spruce, 30% pine, 10% birch, with bark), average size 3 × 0.3 mm, Byggelit AB, Sweden.
Wood flakes, spruce, laboratory made, average size 3 × 0.5 mm.
Wood flour, spruce, sieving mesh 80, Lignitverken AB, Sweden.
Acetylated spruce flakes, average size 3 × 0.5 mm, laboratory made, 16.6% weight increase.

Bonding agents

Kraft lignin, Indulin AT, av. particle size 4 μm, Westwaco Corp., USA.

Kraft lignin, KL2, KL88, and Curan 100, av. size 16 μm, Holmens Bruk AB, Sweden.
PVC, emulsion type, Pevikon PE 710, av. size 5 μm, Norsk Hydro, Sweden.
PVC-copolymer (PVCAC), 17% vinylacetate, av. size 4 μm, Aldrich Chem. Co., England.
LDPE, type 017.040, density 924 kg/m^3, av. size 380 μm, Atochem, France.
PA 11, Rilsan powder ES natural, av. size 30 μm, Atochem, France.
Acrylic copolymer latex, Mowilith DM 774, av. size 0.1 μm, Hoechst-Perstorp AB, Sweden.

All the bonding agents used, except the acrylic latex, were particulate solids; they were dry-blended with the cellulosic component. Optical inspection showed that they provided an even coverage of the wooden flakes.

Hydrophobing agents
Paraffin wax, T_m 58–60°C, laboratory grade.
Paraffin oil, purum.
Stearic acid, T_m 55–56°C.
Vestovax, type SH 105, av. size 20 μm, Veba-Chemie AG, Germany.

Expanding agent
PVDC microspheres, Expancel 461 DU, av. size 10 μm (ca. 40 μm expanded), Expancel AB, Sweden.

This agent was intended as a means to change the density of the samples. Contrary to the expectations, it did not influence the density or the mechanical parameters, the pressure being the primary factor in this context. However, the expanding agent did have an effect on the swelling behaviour.

2.4
Compounding and sample preparation

The components were conditioned one day at 50% RH, whereafter they were dry-blended. In some experiments, the wood component was mixed with a latex emulsion, and the mixture dried at 100°C. When using the paraffin oil, this was added first to the cellulose phase, the components being added afterwards. Paraffin wax was melted onto the cellulose particles at 70°C before adding the remaining substances.

The mould cavity was filled with 20 g of the preblended material, whereafter appropriate pressure, depending on the density to be attained, was applied. The stamps of the mould, which also acted as electrodes, were preheated to 80°C. An anode current of 3 mA was employed in the experiments, with the pressing time varying between 60 and 120 s, depending on the melting characteristics of the bonding agent. After the HF field had been switched off, the sample was allowed to cool for 2 min at 80°C before releasing the pressure. Thereafter, the sample was taken out of the mould cavity.

In the high density range, typical sample densities were around 1200 kg/m^3, while corresponding values for the low density samples were about 700 kg/m^3.

2.5
Testing

The samples were cut to test bars and conditioned at 20°C and 50% RH for 48 h. The bars were subjected to conventional tensile tests (Instron, model 1193) at a cross-head speed of 5 mm/min, corresponding to a strain rate of 1.7×10^{-3} s^{-1}. The effective length of the tensile test bars was 50 mm, their width 10 mm. The thickness varied between 3.4 and 8.0 mm, depending on density.

From the stress-strain curves, the initial modulus, E, and stress and strain at rupture, σ_b and ε_b were evaluated. The impact strenght, IS, was determined using a pendulum tester (Frank, model 565 k) according to BS 2782. The water absorption and swelling were measured after immersion in water at RT, by determining the weight and thickness increase, after 1 h, 2 h, 24 h, and 1 week.

3
Results

3.1
Preliminary experiments with wood flour composites

In a preliminary series of experiments, various cellulose and wood flour based samples containing PVC as the binder were prepared. Due to initial difficulties in optimizing the experimental conditions, relatively low values of the mechanical parameters were obtained. It may therefore suffice to exemplify these results with some data obtained with pressing the 80 mesh wood flour with PVC, Kraft lignin and PA 11 as bonding additives, Table 1. With PVC, modulus values increasing with the wood content up to about 80% were recorded. At higher wood contents the modulus fell markedly. The stress and strain at rupture decreased within the entire wood content range, thus indicating an overall increase in brittleness.

Using lignin as binder in the above experiments yielded somewhat better results. Especially interesting was the finding that the σ_b values retained their relatively high level up to 90% wood content, Table 1. With polyamide 11, the strength parameters were somewhat improved, as compared to samples containing PVC and lignin as binders. On the other hand, the modulus was slightly lower, and decreasing with increasing woodflour content, Table 1. The low IS values at more that 80% wood content were probably due to incomplete coverage of the wood particle surface by the PA 11 binder. This appears also to be the case with PVC.

3.2
Properties of flake board samples

In these experiments, the properties of samples based on wood flakes with varying amounts of thermoplastic binder were measured. The results are shown in Figs. 2–4. The binders used were PVCAC, Kraft lignin, and LDPE. In the following, the results obtained with each of the binders will be briefly commented.

PVCAC
This binder is an amorphous copolymer of vinyl chloride and acetate, the content of the latter component amounting to 17%.

Table 1. Mechanical properties of high density boards from wood flour with lignin, PVC, and PA 11 as binder. Density 1300 (± 100) kg/m^3. IS-impact strength.

Wood flour Content, %	E GPa	σ_b MPa	ε_b %	IS kJ/m^2
Lignin binder				
20	3.0	10	0.4	2.2
40	4.1	18	0.6	2.9
60	4.5	28	1.8	4.0
80	4.3	30	2.0	4.0
90	4.1	28	1.8	4.8
PVC binder				
20	2.6	38	3.7	6.2
60	4.1	31	1.9	5.8
80	4.2	26	2.0	5.1
90	2.6	12	0.9	-
PA 11 binder				
70	2.5	39	2.0	5.8
80	1.9	39	1.7	5.8
90	1.5	11	0.4	4.0

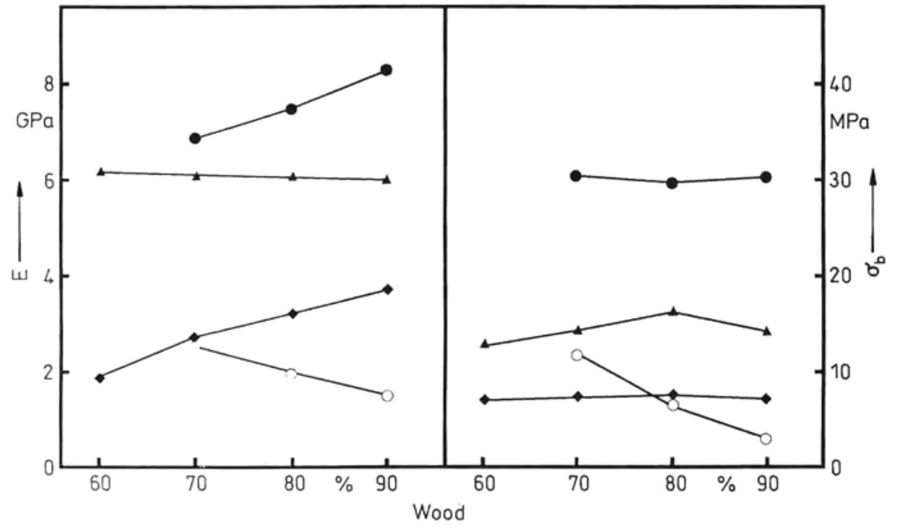

Fig. 2. E-modulus (E) and tensile strength (O_b) vs. wood flake content. Binder: Δ - lignin; ◌ - PVCAC; ◊ - LDPE. Filled symbols - high density (1200 kg/m³), unfilled - low density (700 kg/m³)

Bild 2. Abhängigkeit des E-Moduls (E) und der Zugfestigkeit (O_b) von Spananteil und folgenden Bindemitteln: Δ - Lignin, ◌ - PVCAC, ◊ - LDPE. Augefüllte Symbole: hohe Dichte (1200 kg/m³), nicht ausgefüllte: niedrige Dichte (700 kg/cm³)

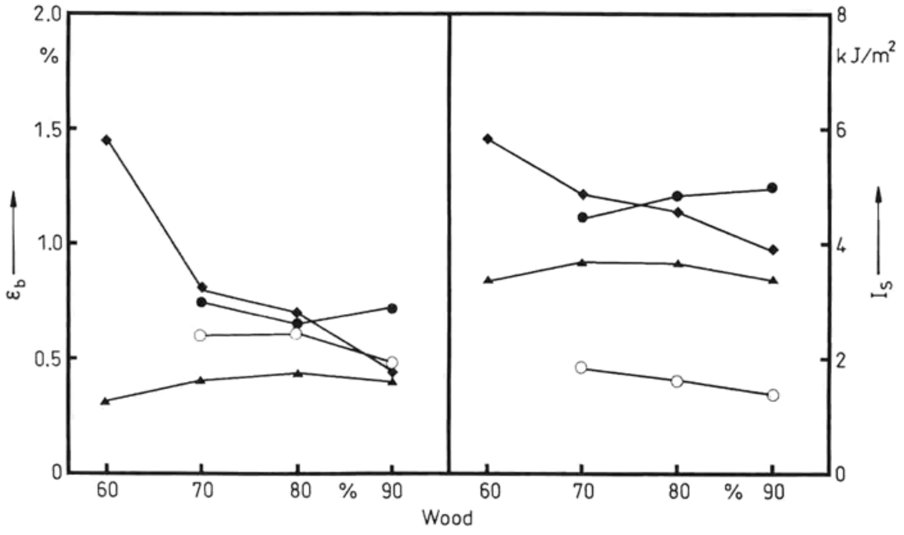

Fig. 3. Elongation at break and impact strength (I_s) vs. wood flake content. Symbols as in Fig. 2

Bild 3. Abhängigkeit der Dehnung bei Bruch (ε_b) und der Schlagbiegefestigkeit (I_s) vom Spangehalt. Symbole wie Bild 2

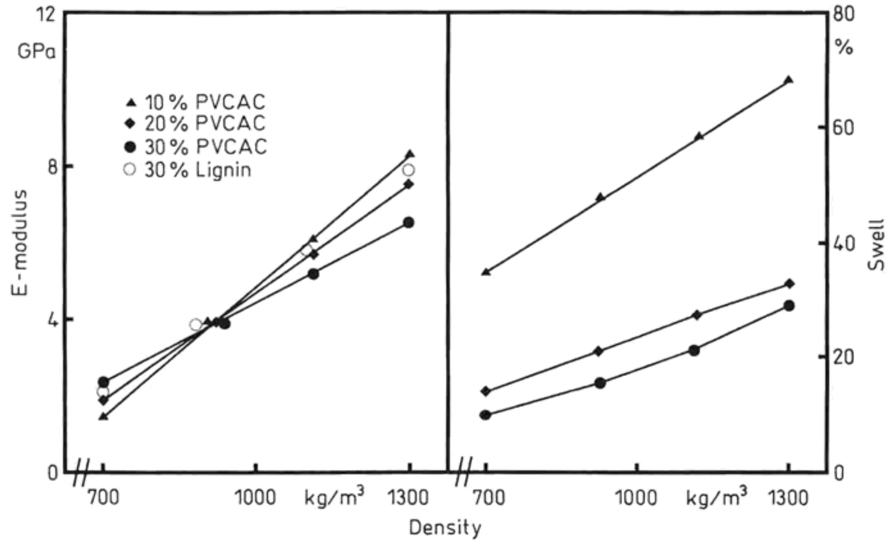

Fig. 4. E-modulus (left hand) and swell after 1 week in water (right hand) vs. density of wood flake boards for varying contents of PVCAC and for lignin at 30% concentration

Bild 4. Abhängigkeit des E-Moduls (links) und der Quellung (rechts) von der Plattendichte für verschiedene Harzgehalte und für 30% Ligninanteil

In comparison with PVC it has an improved heat stability; it can also be expected to provide better bonding to cellulose than unmodified PVC. It was therefore chosen instead of PVC which had been used in the preliminary experiments with wood flour, Table 1.

High density board samples (1300 kg/m³) based on this binder have some appealing mechanical properties. For instance, both modulus, Fig. 2, and impact strength, Fig. 3, increase with the wood content, while σ_b and ε_b remain relatively stable.

Figs. 2–3 also show corresponding data for samples with lower density 700 kg/m³). The density apparently is an important factor in this context, determining not only the absolute values of the various mechanical parameters, but also the fashion in which the properties change with the composition. One finds, for instance, that both E and σ_b are reduced when the proportion of wood flakes in the samples is increasing. The IS and ε_b values indicate a high level of brittleness. The differences between the high and low density samples are summarized in

Table 2. Mechanical properties of wood flake board samples using lignin and PVCAC as binder

Binder %	Density kg/m³	E GPa	σ_b MPa	ε_b %	IS kJ/m³	Swell. in water, %	
						2 h	168 h
Lignin 30%	700	2.5	10.5	0.6	2.0	30	Disintegrates
PVCAC 30%	700	2.6	10.5	0.6	1.9	7	10
Lignin 30%	1200	6.1	14	0.8	3.5	35	Disintegrates
PVCAC 30%	1300	6.9	30.5	0.7	4.5	12	28

Table 2, which also shows corresponding values of the mechanical parameters and thickness swelling for lignin bonded samples.

The increase in E with density is somewhat higher than proportional. All data in Table 2 relate to a wood content of 70%. The density dependence of the modulus is further illustrated in Fig. 4, where results relating to samples with varying wood content are shown. At densities in excess of 1000 kg/m³, the highest E values are obtained for samples with the lowest binder content. This is to be expected since the modulus of the wood component is higher than that of the binder. At low densities, on the other hand, a higher binder concentration is required in order to provide sufficient cohesion of the flakes. This explains the reduction in E at low binder contents for samples with relatively low densities. With regard to impact strength, a similar dependence on density and binder content is found.

Kraft lignin (Indulin AT)

With this binder, the mechanical parameters are nearly independent of the wood content within the composition range covered by the experiments. The E and IS values are somewhat lower than with PVCAC, while σ_b and ε_b only amount to about half the corresponding PVCAC levels, Figs. 2–3.

The variation of the modulus with density and binder content is similar to that found for PVCAC. This follows also from Table 2 which shows the relevant parameter values for samples at two density levels. Lignin binders can be expected to show a fair degree of affinity to cellulose.

Some complementary results were carried out with three types of kraft lignin from Holmens Bruk AB (Sweden). No major difference in the results (mechanical parameters, swelling) were noted. In the following, only the results obtained with Indulin AT are presented.

LDPE

This binder has the advantage of a low melting point and a relatively low cost. On the other hand, its affinity to cellulose is limited. In comparison with the other binders, LDPE produced relatively low E and σ_b values. On the other hand, it improved the ductility, as reflected in the IS and ε_b levels attained, Figs. 2–3.

3.3
Water uptake

In this section, the characterization of the mechanical parameters of the various samples given above is supplemented with data relating to their interaction with water as reflected in measurements of water uptake and swelling.

Wood flour samples

The variation of the water uptake with time of the wood flour/PVC samples produced in the preparatory experiments is shown in Fig. 5 for 20%, 40% and 60% wood content. As can be seen, the water uptake increases in that order. Corresponding lignin based samples showed a similar behaviour, although the uptake increased with time even faster in this case. Also the amount of water measured in the samples was higher (80–90% for samples containing 40% and 60% wood after about 100 h immersion in water).

As with the mechanical parameters, the moulding pressure plays a central role here. This is illustrated in Fig. 5 where data for a sample (20% wood flour) moulded at somewhat lower pressure is included. The rate of water uptake was extremely high in this case; after about 50 h the sample disintegrated (low density, LD).

Flake based samples

The rate of water uptake for this type of material is shown in Table 3.

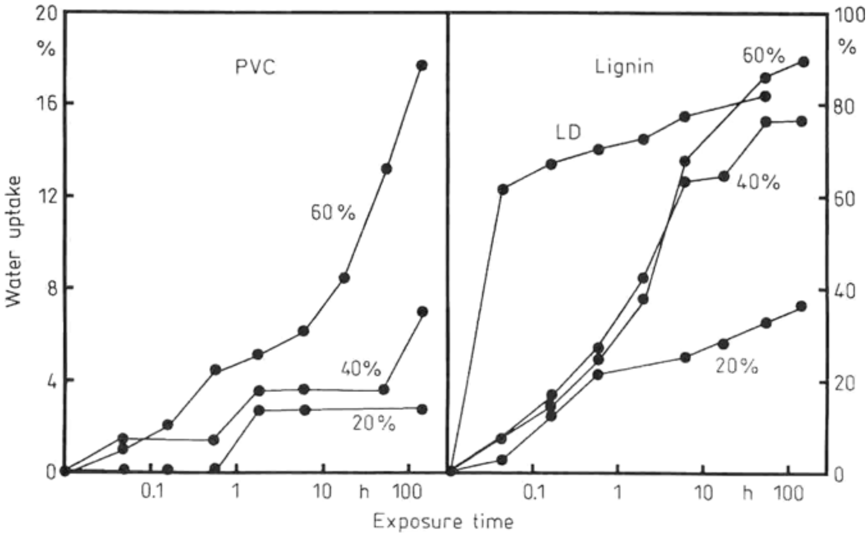

Fig. 5. Water uptake with time of wood flour/PVC and wood flour/lignin composites. Wood content 20%, 40% and 60%; density 1200 kg/m³. Curve marked LD denotes a low density sample (700 kg/m³; wood content 20%)

Bild 5. Wasseraufnahme von PVC- und ligningebundenen Bodenplatten mit Holzanteilen von 20%, 40% und 60% sowie einer Dichte 1200 kg/m³. Die mit LD bezeichnete Kurve beschreibt das Verhalten einer Probe mit geringer Dichte (700 kg/m³) und 20% Holzanteil

Table 3. Water uptake (%) of different wood flake board compositions. DIS = Disintegrated

Density Kg/m^3	Binder %	Exposure to water			
		1 h	2 h	24 h	168 h
1200	lignin, 40%	14	20	71	DIS
1200	” 30%	16	25	87	DIS
1200	” 20%	21	36	113	DIS
1200	” 10%	30	56	114	DIS
1200	LDPE, 40%	6	8	26	DIS
1200	” 30%	12	15	31	DIS
1200	” 20%	13	20	49	DIS
1200	” 10%	15	80	91	DIS
700	PVAC, 30%	25	31	48	67
700	” 20%	53	60	72	83
700	” 10%	81	88	105	127
1300	” 30%	2	3	11	25
1300	” 20%	2	3	12	26
1300	” 10%	8	11	32	57

3.4
Swelling

The significant amounts of water taken up by the wood based composites are also reflected in marked swelling effects. In this section we report data relating to swelling in the thickness direction upon immersion in water. Apart from samples based on the binders treated above, some additional agents were used. The swelling data are supplemented with results obtained with acetylated flakes.

Figure 6 shows the variation of the swelling degree with the wood content after a weeks exposure to water for the samples discussed above. A sample of the wood flour/PA11 composites has also been included. Among the high density samples those bonded with PA11 exhibit the lowest swelling. However, low density wood flour/PA11 samples, not shown in Fig. 6, suffered from unusually high swelling levels, showing at the same time inferior mechanical properties. There was no significant difference between flake based samples containing LDPE and PVCAC. As expected, the degree of swelling increases with the wood content for all sample types. Lignin bonded samples, not included in Fig. 6, showed such a high swelling level that they disintegrated spontaneously after a few days immersion in water.

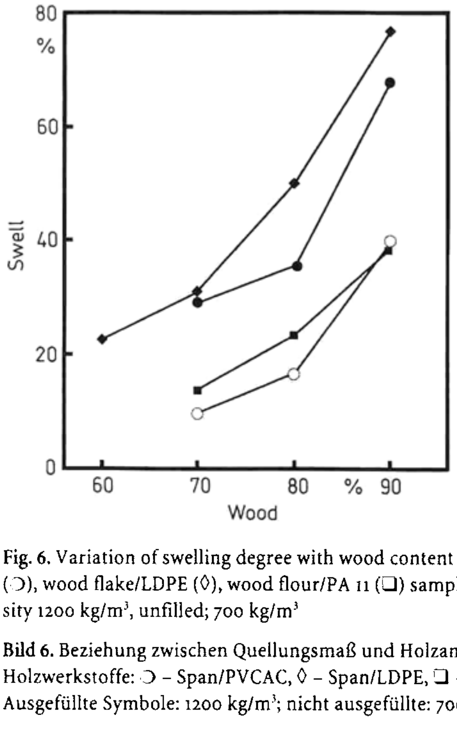

Fig. 6. Variation of swelling degree with wood content for wood flake/PVCAC (○), wood flake/LDPE (◊), wood flour/PA 11 (❑) samples. Filled symbols; density 1200 kg/m^3, unfilled; 700 kg/m^3

Bild 6. Beziehung zwischen Quellungsmaß und Holzanteil für die Holzwerkstoffe: ○ – Span/PVCAC, ◊ – Span/LDPE, ❑ – PA 11 – Bodenplatte. Ausgefüllte Symbole: 1200 kg/m^3; nicht ausgefüllte: 700 kg/m^3

Interesting was the finding that low density samples exhibited lower swelling degrees, as illustrated in Fig. 4 (right part) for wood-flake/PVCAC. This is probably due to the large amount of free space in such materials, allowing the fibres to expand without influencing their surroundings to the same degree as in a densely packed structure.

Figure 4 shows the density dependence of the thickness swelling for the flake/PVCAC samples. In contrast to the mechanical parameters, lowering the density thus improves the response to water exposure. Reducing the binder content from 20% to 10% results in a threefold increase in swelling.

As mentioned above, the lignin containing samples were especially susceptible to water, disintegrating after a few days of immersion. Lignin in itself is insoluble in water, but the bonding strength it conveys to the composite structure is apparently not sufficient to withstand the swelling pressure. Some supplementary experiments were therefore carried out in order to improve the behavior of this binder. The results, reported in Fig. 7, show that adding stearic acid (5%) does not improve the resistance to water as measured by swelling. On the other hand, short-chain hydrocarbons, such as paraffin wax (and also Vestovax), have a marked positive effect, reducing the swelling to below 20% after 2 h water exposure.

Combining a lignin content of 10% with 10% PVCAC or LDPE gives a marked swelling increase, approximately equivalent to the value obtained with 10% of either of the thermoplastic binders with no lignin present. On the other hand, combining the lignin binder with both Vestovax and Expancel has a beneficial effect on the swelling value which is only one third of that which would have been observed in a material not containing the lignin component. Low density samples bonded with PVCAC exhibit, as mentioned above, relatively limited swelling. If 5% of the binder is replaced by Vestovax, short-term swelling is reduced, cf. Fig. 8. If the replacement is for Expancel, the swelling is lower even after one week of immersion. However the use of Expancel only yields unusually high swelling values.

Chemical modification of the wood component is a rather natural, although not always technically feasible way to reduce its affinity to water. In Fig. 8 we show some results relating to acetylated spruce flakes, obtained by reaction with acetic acid. Flakes modified in this way show less than half the extent of swelling recorded with lignin or PVCAC binders.

Finally, in Fig. 8 (right) we include data for an untreated and acetylated flake material where the PVCAC binder has been replaced by an acrylate latex. Acetylation of the wood component provides also in this case a powerful means of reducing the swelling. The effect is enhanced by the high affinity of the acrylate phase to the acetyl groups of the modified flakes.

4
Modulus calculation

The usefulness of theoretical models allowing the calculation of the mechanical parameters of fibre based composites from the properties and arrangement of the components has been amply illustrated in relevant literature. Among the theoretical tools used in that area, the theory of Halpin and Tsai (Halpin 1984) stands out as especially useful, not at least due to its relative simplicity and the possibility to model composite structures with complex fibre arrangements. Basically, the Halpin–Tsai concept may appear applicable also to composite structures encountered in particle boards and related materials. However, available literature contains, to our knowledge, no mention of such an attempt. It has to be admitted that such materials have a rather unusual structure, especially with regard to the low content of

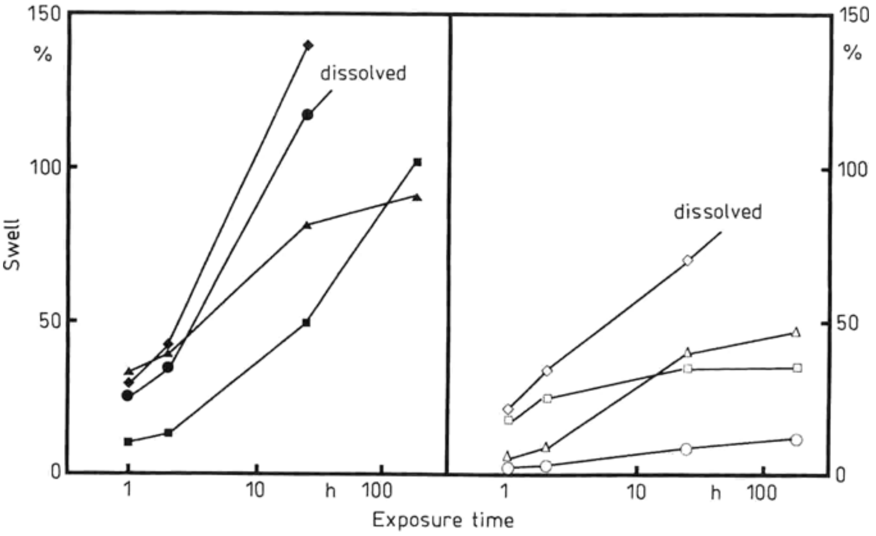

Fig. 7. Swelling after exposure to water vs. time (log. scale) for wood flake samples for different compositions of the binder. Left: density 1200 kg/m^3; right – 700 kg/m^3. Binder: ● – 20% lignin, ◆ – 15% lignin + 5% stearic acid, ■ – 10% lignin + 10% paraffin wax, ▲ – 10% lignin + 10% LDPE,) – 20% lignin + 5% Vestovax + 5% Expancel, Δ – 5% Vestovax + 5% Expancel, ⊔ – 10% lignin + 10% PVCAC, ◊ – 30% lignin

Bild 7. Dickenquellung nach unterschiedlichen Benutzungszeiten für Spanplatten, hergestellt mit verschiedenen Bindemitteln und Zusätzen. Links: hohe Dichte (1200 kg/m^3); rechts: niedrige Dichte (700 kg/m^3). Bindemittel: ● – 20% Lignin, ◆ – 15% Lignin + 5% Stearinsäure, ■ – 10% Lignin + 10% Paraffin, ▲ – 10% Lignin + 10% LDPE,) – 20% Lignin + 5% Vestovax + 5% Expancel, Δ – 5% Vestovax + 5% Expancel, ⊔ – 10% Lignin + 10% PVCAC, ◊ – 30% Lignin

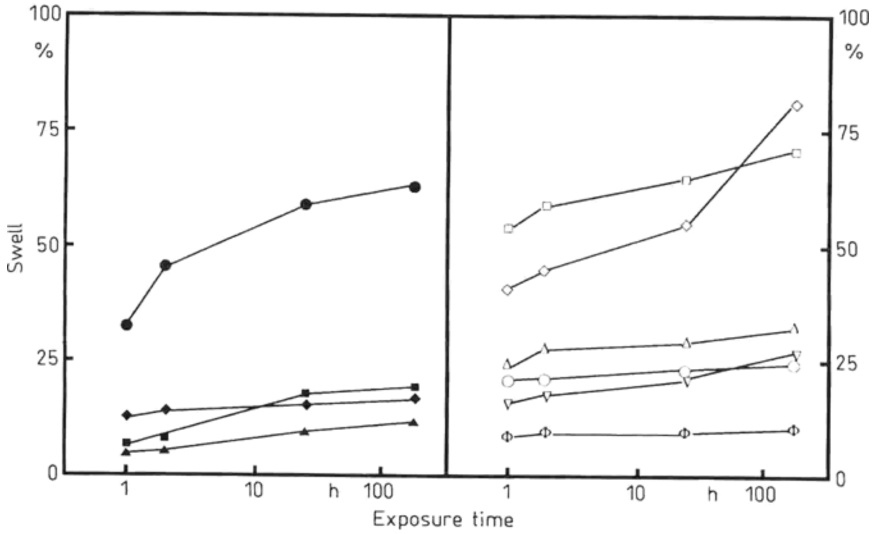

Fig. 8. Swelling degree vs. time (log. scale) for different wood flake boards. AWF = acetylated wood flakes. Left: density 700 kg/m^3; right – 900 kg/m^3. Binder: ◆ – 20% PVCAC, ▲ – 15% PVCAC + 5% Expancel, ■ – 15% PVCAC + 5% Vestovax, ● – 5% Expancel, Φ – 20% PVCAC (AWF),) – 20% PVCAC, Δ – 20% lignin (AWF), ◊ – 20% lignin, ∇ – 20% acryl latex (AWF), ⌝ – 20% acryl latex.

Bild 8. Dickenquellung nach unterschiedlichen Benutzungszeiten für verschiedene Spanplattentypen. AWF = acetylierte Späne. Links 700 kg/m^3; rechts: 900 kg/m^3. Bindemittel: ◆ – 20% PVCAC, ▲ – 15% PVCAC + 5% Expancel, ■ – 15% PVCAC + 5% Vestovax, ● – 5% Expancel, Φ – 20% PVCAC (AWF),) – 20% PVCAC, Δ – 20% Lignin (AWF), ◊ – 20% Lignin, ∇ – 20% Acryl-Latex (AWF) ❏ – 20% Acryl-Latex

the binder phase which may render the matrix concept questionable. Despite these difficulties, we will attempt to apply the Halpin-Tsai formalism to the materials discussed in the present paper. As will be seen, a fair degree of agreement with experimental data will be obtained in certain cases, while in others, where the requirements of the theory connot be justified, differences will be found. Before proceeding, we present a brief summary of earlier attempts to model the elastic properties of particle boards.

Hunt (1970) applied a finite element method (FEM) to a layer of wood particles glued together with a hypothetical network of binder elements. The layer was assumed to be isotropic, with the particles being randomly oriented and having their symmetry axes lying in the sample plane. When comparing the results of the calculations with experimental data obtained with samples specially prepared in order to satisfy the requirements of the FEM model a fair degree of agreement was reached. On the whole, the differences between calulated and experimental *E* modulus were about 10% or less (board from aspen and Douglas fir). Corresponding discrepances for the shear modulus amounted to 15–20%. In another FEM analysis, Lee (1977) considered the wood particles as ellipsoids of varying thickness glued together with the binder resin. An overall orientation of the particles was allowed. The differences between predicted and measured modulus values (*E*, *G*) were dependent on the degree of orientation, amounting to 5–20%. The model consistently overestimated the elastic parameters. Harris (1977) used a simplified approach based on a number of rectangular wood segments with different orientations glued together with the binder. Several layers could be accommodated in the model. The predictions were compared with experimental data obtained with board samples having varying degrees of orientation. The modulus (*E*) showed as expected a strong orientation dependence. The differences between theory and experimental modulus (*E*) values amounted to about 70% for samples with low orientation. For more oriented samples the discrepances were somewhat smaller (about 30–40%). When taking account of the density (ρ) by considering the ratio E/ρ the theory gave a fair picture of the actual parameters at the highest orientation only (differences of about 5%). These figures apply to a one-layer model. When increasing the number of layers a better agreement was obtained (10–30% at low orientation, 5–10% at high orientation).

Table 4. Comparison of experimental and calculated modulus values for the samples referred to in Fig. 2. The apparent density of the wood flakes in the final sample, ρ_f, and their volume fraction, v_f have been calculated using Eqs. (5, 6). The wood percentages given by the figures 60–90% are by weight. Binders used: PVCAC, LDPE, and lignin

	60%			70%			80%			90%		
	Theor.	Exp.	%Diff.	Theor.	Exp.	%Diff.	Theor.	Exp.	%Diff.	Theor.	Exp.	%Diff.
PVCAC (1300 kg/m^3)												
E (GPa)				6.82	6.98	-2.3	7.47	7.70	-3.0	8.10	8.80	-8.0
ρ_f (kg/m^3)				1291			1284			1274		
v_f				0.709			0.808			0.905		
PVCAC (700 kg/m^3)												
E (GPa)				3.78	2.43	55.6	4.10	2.00	105.0	4.41	1.49	196.0
ρ_f (kg/m^3)				594			629			658		
v_f				0.841			0.896			0.949		
LDPE												
E (GPa)	2.18	1.94	12.4	2.87	2.70	6.3	4.01	3.20	25.3	5.34	3.68	45.1
ρ_f (kg/m^3)	1287			1208			1293			1156		
v_f	0.517			0.640			0.740			0.878		
Lignin												
E (GPa)	5.87	6.19	-5.2	6.14	6.15	-0.2	6.73	6.13	9.8	7.82	5.98	30.8
ρ_f (kg/m^3)	1200			1117			1130			1227		
v_f	0.636			0.745			0.832			0.911		

Both models underestimated the actual values for weakly oriented samples; at high degrees of orientation the opposite was true.

Apart from the references mentioned above the work done on modelling the mechanical properties of particle board has been centred on a number of specific problems as the role of density, binder content etc. As a rule, such studies do not allow predictions of mechanical parameters of the board material from the properties and arrangement of its components.

The Halpin-Tsai formula (Halpin 1984) is a simplification of the rigorous calculations of Hill (1965) and Hermans (1967), especially suitable for engineering applications. The formula, valid for a completely oriented orthotropic ply under plane stress conditions, reads as follows

$$\frac{E_{11}}{E_m} = \frac{1+\zeta\eta v_f}{1-\eta v_f} \qquad (2)$$

where

$$\eta = \frac{(E_{fl}/E_m)-1}{(E_{fl}/E_m)+\zeta} \qquad (3)$$

with E_{11} denoting the composite elastic modulus in the fibre (orientation) direction, E_m the matrix (binder) modulus, E_{fl} the longitudinal fibre modulus, v_f the volume fraction of fibres, and ζ a factor depending on the fibre geometry (aspect ratio). According to Halpin (1969) $\zeta=2(l/t)$ should be used with flake-formed particle reinforcement (l/t is the longitudinal aspect ratio, i.e. length/thickness). In the present study wood flakes with relatively small dimensions were used which precluded a direct measurement of their moduli in the two principal plane directions.

In order to apply Eq. (2) to our experimental data a somewhat different course of action was chosen. Tsai and Pagano (1968) have shown that for a ply with fibers randomly oriented in its plane the in-plane modulus is given by

$$E = \frac{3}{8}E_{11}+\frac{5}{8}E_{22} \qquad (4)$$

with E_{11} and E_{22} denoting the longitudinal and transversal composite moduli for a completely oriented ply. In the case of wood based boards account has also to be taken of the increased density of the flakes due to their compression during the manufacturing procedure. We further assume that the binder is incompressible and that possible air inclusions are contained in the wood phase. For v_f and ρ_f we then obtain

$$v_f = \frac{w_f/\rho_f}{w_f/\rho_f+(1-w_f)/\rho_b} \qquad (5)$$

and

$$\rho_f = \frac{w_f}{1/\rho_{\text{board}}-(1-w_f)/\rho_b} \qquad (6)$$

where v_f and ρ_f are the volume fraction and density of particles in the board, ρ_{board} and ρ_b the density of the board and the binder, respectively, and w_f is the weight of particles in relation to the total weight of particles and binder. The influence of moisture has not been taken into account in these calculations. Furthermore, we assume a linear relationship between the

Table 5. Experimental and calculated modulus, ρ_f, and v_f, values for lignin-bound samples with varying density. Lignin content 30% by weight. Cf. Table 4

Lignin	Theor.	Exp.	%Diff.
700 kg/m^3			
E (GPa)	3.66	1.97	85.8
ρ_f (kg/m^3)	576		
v_f	0.850		
800 kg/m^3			
E (GPa)	4.18	2.88	45.1
ρ_f (kg/m^3)	676		
v_f	0.829		
920 kg/m^3			
E (GPa)	4.79	3.79	26.4
ρ_f (kg/m^3)	802		
v_f	0.803		
1189 kg/m^3			
E (GPa)	6.14	6.15	-0.2
ρ_f (kg/m^3)	1117		
v_f	0.745		
1255 kg/m^3			
E (GPa)	6.69	7.16	-9.6
ρ_f (kg/m^3)	1202		
v_f	0.731		

Table 6. Experimental and calculated modulus, ρ_f and v_f values for samples with varying density (underlined) containing 10, 20 and 30% by weight of PVCAC, cf. Tables 4 and 5

PVCAC	10%			20%			30%		
	Theor.	Exp.	%Diff.	Theor.	Exp.	%Diff.	Theor.	Exp.	%Diff.
	694 kg/m³			*704 kg/m³*			*743 kg/m³*		
E (GPa)	4.41	1.49	196.0	4.10	2.00	105.0	3.93	2.43	61.7
ρ_f (kg/m^3)	658			629			623		
v_f	0.949			0.896			0.835		
	741 kg/m³			*733 mg/m³*			*922 kg/m³*		
E (GPa)	4.71	2.11	123.2	4.27	2.26	88.9	4.86	4.29	13.3
ρ_f (kg/m^3)	706			658			812		
v_f	0.945			0.891			0.795		
	809 kg/m³			*770 kg/m³*			*1070 kg/m³*		
E (GPa)	5.14	2.31	122.5	4.48	2.58	73.6	5.62	4.43	26.9
ρ_f (kg/m^3)	775			695			983		
v_f	0.940			0.886			0.762		
	872 kg/m³			*851 kg/m³*			*1248 kg/m³*		
E (GPa)	5.54	2.99	85.3	4.94	3.29	50.2	6.52	6.12	6.5
ρ_f (kg/m^3)	839			779			1209		
V_f	0.935			0.874			0.723		
	919 kg/m³			*963 kg/m³*			*1338 kg/m³*		
E (GPa)	5.83	3.47	68.0	5.58	4.19	33.2	6.97	6.98	-0.1
ρ_f (kg/m^3)	888			899			1333		
v_f	0.932			0.857			0.703		
	1169 kg/m³			*1297 kg/m³*					
E (GPa)	7.40	6.08	21.7	7.47	7.70	-3.0			
ρ_f (kg/m^3)	1152			1284					
v_f	0.913			0.808					
	1281 kg/m³								
E (GPa)	8.10	8.80	-8.0						
ρ_f (kg/m^3)	1274								
v_f	0.905								

density of the flakes and their modulus, implying that the aspect ratio l/t appearing in Eqs. (2, 3) also changes with density. If the compression only amounts to a decrease in the flake thickness, it follows that l/t is proportional to density. We therefore set $l/t \sim \rho_f$. Since the elastic moduli of the flakes were not known, different E_{ll} and E_{tt} pairs (E_{tt} denoting the flake modulus in the transversal direction) had to be tried in order to fit the theoretical formulae to experimental data. Input values then were elastic moduli in longitudinal and transverse directions of a flake and its density (E_{ll} 8.5 GPa, E_{tt} 0.425 GPa, flake density 500 kg/m^3, flake dimensions 2.5 × 0.35 × 0.35 mm). Data for the binders were taken from the literature (Mackenzie 1987; Cousins 1976).

The results of our calculations are summarized in Tables 4–6. Table 4 shows the calculations and observed modulus data for samples containing varying amounts of the binders used, the observed values being those shown in Fig. 2. Apart from the modulus the table also gives data on the calculated density of the wood particles in the sample, and also their volume fraction. As can be seen, there is a fair degree of agreement between calculated and measured modulus values for the high density samples. At the highest wood contents, the theory tends to overestimate somewhat the modulus level for the LDPE and lignin-based material. On the whole, the agreement achieved through the use of the Halpin–Tsai approach can be said to parallel that attainable by the more elaborate treatment discussed above (Hunt 1970; Lee 1977; Harris 1977). For the low-density PVCAC based samples, the theory yields modulus levels far in excess of those observed. This is to be expected, in the first place due to air inclusions the effect of which is not accommodated in the formulae used.

The adverse effect of increasing porosity on the applicability of the Halpin–Tsai approach is further illustrated by the data of Table 5 relating to lignin-based samples with varying density. As the table shows, the theory gives an excellent agreement with measured data at the highest densities. When the density is decreased, a significant overestimation of the modulus is obtained. In Table 6 we reproduce data relating to the modulus vs. density relationship for PVCAC bound samples containing 10, 20 and 30% of the binder. Here again, the agreement tends to become significantly closer as the density increases. At the lowest binder content, i.e. 10%, the predicted values differ significantly from the measured ones, as the density decreases. This is not unexpected since the notion of a continuous matrix tends to loose its proper meaning. The data of Table 6 relating to 30% binder content do not challenge such a view.

The data presented above provide a strong support for the applicability of the Halpin–Tsai concept to materials consisting of flake-shaped particles held together by relatively small amounts of a binder. A natural limit to the validity range of such an approach is a sufficiently high density, since the effect of porosity is not accounted for by the theoretical formalism. Also with this limitation, the results obtained appear highly encouraging, especially considering the fact that the few attempts at modelling the mechanical parameters of materials of this type appear far too complex to be used in practice.

5
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