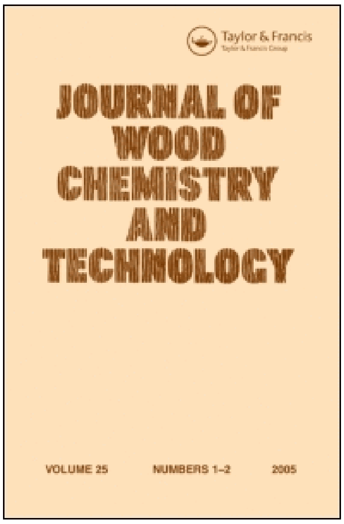

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CHEMICAL MODIFICATION OF WOOD PRODUCTS WITH GASEOUS ALKYLENE OXIDES

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ABSTRACT

A procedure for alkoxylation of wood with gaseous alkylene oxides was developed, using solid wood samples and ethylene oxide catalyzed with trimethyl amine. The procedure consisted of an initial reaction step with wood, trimethyl amine and ethylene oxide, followed by evacuation of excess trimethyl amine. Several small additions of ethylene oxide were then made, followed by evacuation and steaming. Solid wood ethoxylated by this procedure was found to be less degraded by wood destroying fungi than was unmodified wood.

Chips for particle board production were alkoxylated, according to the procedure, using ethylene oxide and propylene oxide, respectively. Particle boards were made from alkoxylated chips and evaluated with respect to swelling in water. As a comparison boards were also made from chips alkoxylated with liquid propylene or butylene oxide. Boards made from chips modified with gaseous alkylene oxides performed better than boards made from unmodified chips only when wood chips with a moisture content above that of oven-dried wood were alkoxylated. Chips alkoxylated with liquid alkylene oxides resulted in much better boards than chips alkoxylated with gaseous alkylene oxides.

INTRODUCTION

Chemical modification of wood has been shown to improve the dimensional stability and to reduce the susceptibility of the wood to biological degradation. Alkylene oxides are one class of chemicals that can be used for chemical modification of wood. Modification with liquid propylene or butylene oxide has been shown to be effective in reducing swelling and shrinkage, and biodegradation of wood.[1-5]

For uniform distribution of chemical within the wood, reactions with liquid reagents require the wood to be impregnated to a much higher total uptake of chemical than the amount actually consumed in the reaction. As a carrier of the reagent into the wood, either a solvent or an excess of the reagent itself can be used. In both cases the carrier must be recovered from the wood after the reaction. A gaseous reagent needs no such carrier, but can be added either in a small excess or in the exact amount needed for the reaction. Thereby the need for recovery of chemicals is simplified, or even eliminated.

Chemical modification of wood using gaseous alkylene oxides has previously been investigated.[6-11] Morén[6] used gaseous ethylene oxide either uncatalyzed or catalyzed with trimethyl amine. All ethylene oxide was added on one occasion in the beginning of the reaction. McMillin[7] used gaseous ethylene oxide catalyzed with trimethyl amine. A constant pressure procedure was used, or a procedure where all the chemical was added at one time. An antishrink efficiency (ASE) of 65% was reported. Barnes et al.[9] obtained extensive dimensional distortion, with separation between the annual rings, when using a constant pressure procedure. When oscillating pressure was used during the ethoxylation, no such distortion was observed, and 55% ASE was reported. Leachability of the chemicals introduced into the wood was not determined in any of these studies, and antishrink efficiencies were reported only for one water soaking cycle.

This study was aimed at determining whether alkoxylation using gaseous alkylene oxides could enhance wood properties as effectively as alkoxylation with liquid alkylene oxides. A procedure for alkoxylation with gaseous reagents was developed, using solid wood samples and ethylene oxide catalyzed with trimethyl amine. The procedure was then applied to wood chips for particle board production, using either ethylene or propylene oxide, and particle boards were made from the chips. For comparison, boards were also made from chips modified with liquid propylene or butylene oxide. Dimensional stability of the boards was evaluated. Resistance of the ethoxylated wood to biological degradation was also investigated. In all experiments the stability of the modification in repeated water soaking tests was studied, to determine how much of the added chemical had been permanently bonded to the wood and how much had formed water soluble glycols. This is of course an important consideration for a process aimed at improving the water resistance of wood.

EXPERIMENTAL PROCEDURES

Wood Material

Samples of pine (Pinus sylvestris) sapwood were prepared in the dimensions 20x20x100 mm or 70x5x850 mm.

Commercial chips of pine wood (Pinus sylvestris), used for particle board production, and containing a small amount of bark, were used. The chips were about 1 mm thick and the maximum length was 25 mm. Before use, the chips were passed over a 3x3 mm screen to remove fines.

Apparatus

The alkoxylations were done in reactors of stainless steel. A small reactor (0.6 dm^3) was used for ethoxylations of solid wood samples. This reactor had a thermocouple for measuring the internal wood temperature. The wood chips were alkoxylated in a larger reactor (16 dm^3). Both reactors were equipped with an external electrical heating coil and an internal cooling coil. Normally, the reactors were cooled by airflow through the cooling coil, but if the temperature exceeded the set point by more than 15°C, water-cooling was automatically turned on. The reactors were equipped with safety valves. All parts in contact with epoxides were made of stainless steel or teflon. The reactors were connected to cylinders containing ethylene oxide, propylene oxide, and trimethyl amine, respectively, via flexible teflon tubes. The cylinders could be placed on a balance for weighing the additions of chemicals to the reactors.

The reactors were equipped with thermocouples and pressure gauges, and both temperature and pressure could be recorded continuously.

Alkoxylation with Gaseous Alkylene Oxides

A number of reaction conditions for alkoxylation with gaseous reagents were used. The initial experiments were made on solid wood using ethylene oxide. When satisfactory reaction conditions had been established, the experiments were extended to wood chips using ethylene as well as propylene oxide.

Pine sapwood samples (20 x 20 x 100 mm, tangential x radial x longitudinal or radial x longitudinal x tangential) were prepared. A 3 mm hole was drilled lengthwise into the samples, perpendicular to the 20 x 20 mm surface. The hole was drilled either in the centre of the 20 x 20 mm surface, or, for some of the samples that were cut 20 x 20 x 100 mm radially x longitudinally x tangentially, 5 mm from the end grain surface. Before ethoxylation the wood was either oven-dried, or conditioned in constant relative humidity, and weighed.

A 3 mm thermocouple was placed in the drilled hole, and the sample, which weighed approximately 20 g, was placed in the 0.6 dm^3 reactor. The reactor was evacuated and purged with nitrogen several times. All tubes connected to the reactor were also purged with nitrogen. The reactor was heated to the desired reaction temperature (100-110^{o}C), before pulling a vacuum for 30 min, followed by addition of 1 g of trimethyl amine and, initially, 1 g of ethylene oxide. When the pressure had decreased to a constant value another addition of ethylene oxide was made. The ethylene oxide was added either as 1 g additions, or as one 5 g addition followed by one 1 g addition. Pressure and internal wood temperature were allowed to decrease to nearly constant values between the ethylene oxide additions. When the last charge of ethylene oxide had reacted, the reactor was evacuated for 30 min and the wood was taken out, oven-dried and weighed. The weight gain was calculated as a percentage based on the weight of o.d. unmodified wood.

In a number of experiments the reactor was evacuated for 5 min after the trimethyl amine and the first addition of ethylene oxide had reacted to constant pressure, and before further 1 g ethylene oxide additions were made.

When wood with a moisture content above that of oven-dried wood was alkoxylated, the first evacuation and the initial additions of chemicals were made at room temperature, to prevent the wood from drying to an unknown moisture content during the vacuum period at an elevated temperature.

Pine sapwood samples, 70 x 5 x 85 mm (tangential x radial x longitudinal) were also treated in the small (0.6 dm^3) reactor. Approximately 60 g of wood was ethoxylated at the same time. After nitrogen purging and evacuation, 1 g of trimethyl amine and 1 g of ethylene oxide was added. When the pressure had decreased to a constant value, the reactor was evacuated for 15 min, after which more ethylene oxide was added, in charges of 4 g. When the last ethylene oxide addition had reacted until only a very low pressure remained, the reactor was evacuated for 30 min, after which 200 ml of water was added at the bottom of the reactor. After 2 h the reactor was evacuated again, and the wood was taken out, oven-dried, and weighed.

The same procedure was used for alkoxylation of batches of 400 g of pine wood chips in the 16 dm^3 reactor at 120°C. Initially 6 g of trimethyl amine and 6 g of ethylene or propylene oxide were added. After pulling a vacuum for 15 min, the epoxide was added as 65 g additions. When the last epoxide addition had reacted, the reactor was evacuated for 30 min, 400 ml of water was added and was allowed to remain for 2 h, after which the reactor was again evacuated and the chips were removed, oven-dried, and weighed.

Alkoxylation with Liquid Alkylene Oxides

Oven-dried pine wood chips were alkoxylated with liquid propylene oxide/triethyl amine or butylene oxide/triethyl amine (95/5, v/v) at 120°C at a pressure of 1 MPa.[1]

Particle Board Preparation

Particle boards were prepared on a laboratory scale as described earlier.[13] A melamine/urea/formaldehyde adhesive was added (12% based on o.d. wood) as a 59% solution during 6 min to batches of o.d. wood chips (190 g). The chips were pressed in a laboratory press to 175 x 175 mm boards with a thickness of 10 mm, and a density of approximately 700 kg/m^3. Samples (50 x 50 mm) were cut out of the boards.

Water Soaking Tests

Dimensional stability of solid wood was determined as antishrink efficiency (ASE).[12] Ethoxylated samples and untreated controls were oven-dried, weighed, and measured in the tangential and radial (in some cases only in the tangential) directions. They were water soaked in distilled water, which was changed every day, for 7 days. After every change of water a vacuum was applied for 30 min. After the 7 days the samples were measured again and then oven-dried, followed by a repetition of the procedure.

The swelling was calculated either as a volumetric swelling, using the tangential and radial measurements and neglecting the very small longitudinal swelling, or as a tangential swelling. The antishrink efficiency was calculated as:

$$ASE = \frac{S_{ut}-S_t}{S_{ut}} \cdot 100\% \qquad (1)$$

where: S_{ut} = swelling of untreated sample
S_t = swelling of treated sample

ASE_1 is the antishrink efficiency value obtained in the first water soaking cycle, and ASE_2 is the value obtained in the second cycle.

The weight of the o.d. solid wood samples was determined before and after water soaking in order to determine the weight loss. The weight gain that remained after two water soaking cycles was calculated from the weight of the water leached, o.d. sample based on the weight of the o.d. sample before modification. The bound fraction, defined as the ratio between the weight gain that remained after water soaking and the initial weight gain, was also calculated.

Alkoxylated chips were water soaked for four days in order to determine weight loss and bound fraction.

The thickness swelling of particle board samples (50 x 50 mm) was measured as a function of time during 5 days of water soaking, and in a repeated water soaking and oven-drying test.[13] The samples were oven-dried and their thickness measured, after which they were water soaked for 5 days, and their thicknesses measured again. This was repeated in 5 cycles.

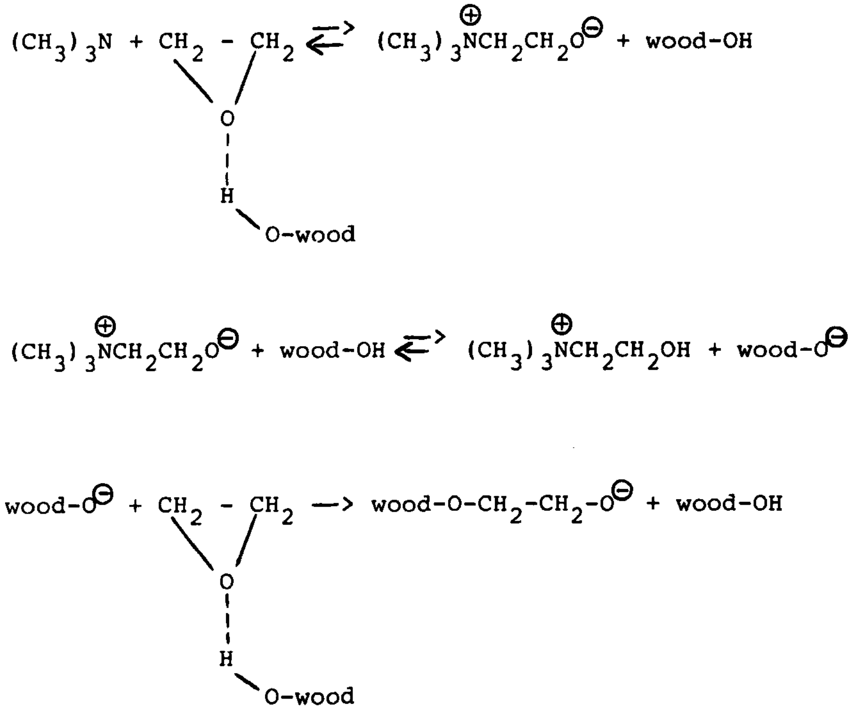
Microbiological Testing

The resistance of ethoxylated solid wood to biological degradation was tested in a 11 weeks soil block test using the brown rot fungus Poria placenta, and in a fungus cellar test. The wood was leached before testing. The fungus cellar test consisted of incubating the samples, together with untreated control samples, in moist unsterile soil at approximately 25°C. The soil contained both white-, brown- and soft-rot fungi, and tunneling bacteria. The samples were inspected after 12 months and the degree of attack was rated, both in a macroscopic and microscopic evaluation.

RESULTS AND DISCUSSION

Ethoxylation of Solid Pine Wood

Fig. 1 shows the pressure and internal wood temperature of an ethoxylation of o.d. pine wood. The oscillation in both pressure and wood temperature is explained by the temperature regulator turning on and off the airflow through the cooling coil. The first chemicals added (1 g of trimethyl amine and 1 g of ethylene oxide) reacted slowly, as shown by the rather slow drop in pressure, and the moderate increase in internal wood temperature. This is believed to be due to a slow initial formation of phenolate and alcoholate ions, that are further able to react with ethylene oxide. A possible mechanism for the reaction is:



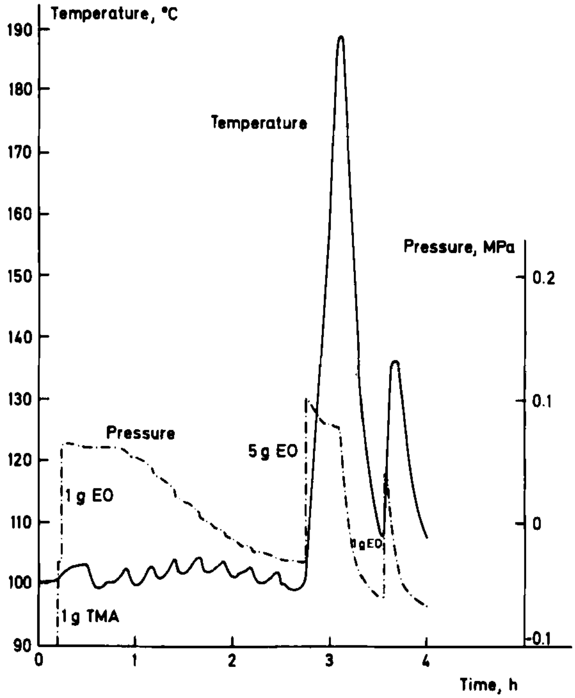


FIGURE 1. Pressure and internal wood temperature during ethoxylation of o.d. pine wood (20 x 20 x 100 mm) at 100°C gas phase temperature. Temperature measured 5 mm from end grain surface.

The addition of one large quantity of ethylene oxide (5 g) caused a fast increase in wood temperature to a very high value. In the given example (fig. 1), where the temperature was measured 5 mm from the wood end grain surface, 190°C was recorded, but when the temperature was measured in the center of the 20 mm thick sample, temperatures above 200°C were reached (cf. Table 1).

As can be seen in fig. 2 the temperature increase was not quite so dramatic when wood with a 10% moisture

TABLE 1

Weight Gain, Bound Fraction, and Increase in Internal Wood Temperature for Ethoxylation of Pine Sapwood (20 x 20 x 100 mm, Radial x Longitudinal x Tangential) at 100°C Gas Phase Temperature.

Moisture content %	Thermocouple distance from end grain mm	Temperature increase at addition of EO[1] °C		Weight gain %	Weight gain after leaching %	Bound fraction %
		5g	1g			
0	10	114	15	25.1	8.2	32.8
0	5	88	28	30.4	8.1	26.8
10.2	10	72	4	29.4	5.7	19.3
10.2	5	53	10	29.4	5.7	19.3

[1] Ethylene oxide added as a 5 g addition followed by a 1 g addition. Temperature increase at initial addition of 1 g of trimethyl amine and 1 g of ethylene oxide omitted.

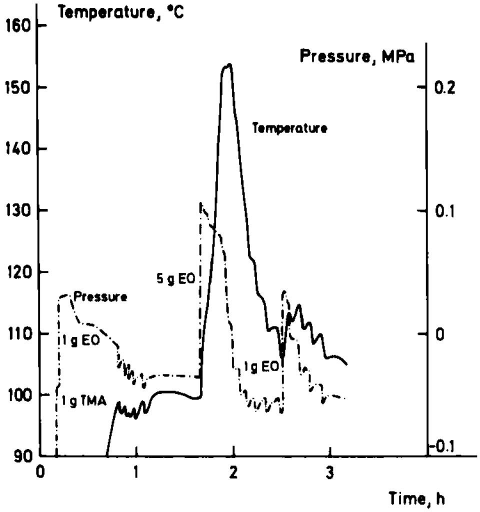


FIGURE 2. Pressure and internal wood temperature during ethoxylation of pine wood (20 x 20 x 100 mm) with 10.2% moisture content at 100°C gas phase temperature. Temperature measured 5 mm from end grain surface.

content was ethoxylated. There are two possible explanations for this; better heat conductivity in moist wood and possible evaporation of water from the heated wood, giving a cooling effect. The shorter total reaction time, and especially the shorter time needed for the consumption of the initial addition of ethylene oxide, was probably attributable to the faster reaction between water and ethylene oxide than between wood and ethylene oxide.

As shown in Table 1 the increase in temperature was largest in the center of the sample, which is what can be expected if transport of chemical is faster than the consumption of ethylene oxide. The increase in internal wood temperature started immediately after the addition of the chemical, which also shows that the chemical was transported very fast the 10 mm along the grain. Also in experiments where the wood sample was cut at a different angle, and the temperature was measured 10 mm from radial and tangential surfaces, the increase in temperature was very rapid.

The weight gain in these experiments was about 30%. After leaching, only a fraction of the weight gain remained, approximately 1/3 for samples that had been ethoxylated when o.d., and approximately 1/5 of the original weight gain for samples that were ethoxylated at a 10% moisture content.

In experiments where the same total amount of ethylene oxide (7 g) was added as 1 g additions, the internal wood temperature never was higher than 125°C. This is important, since too high a temperature causes degradation of the wood. Also for safety reasons it is important to control the temperature. At 560°C, which could possibly be reached in a larger block of wood, an explosive break down of ethylene oxide occurs. However, the lower temperature did not affect the bound fraction of the weight gain.

When excess trimethyl amine was removed by means of evacuation, after the first addition of ethylene oxide had reacted to constant pressure, the bound fraction increased to approximately 50% for o.d. wood (cf. Table 2). This is probably because there was less formation of leachable substances consisting of ethoxylated amine.

As can be seen in Table 2, a higher antishrink efficiency was, unexpectedly, found in the second water soaking cycle than in the first. This was due to the fact that the volume of the wood when it was oven-dried after the first water soaking cycle was larger than directly after the ethoxylation, even though most of the weight loss took place during the first water soaking

TABLE 2
Ethoxylation of o.d. Pine Sapwood (20 x 20 x 100 mm) at 100°C Gas Phase Temperature, Using the Procedure 1 g TMA[1] + 1 g EO[2] Followed by 5 min Vacuum and 1 g Additions of EO.

Weight gain %	Weight gain after leaching %	Bound fraction %	ASE_1 %	ASE_2 %
3.6	0	0	-	-
5.6	1.1	20.2	-	-
13.9	6.4	45.9	-	-
21.1	11.6	55.1	14.7	27.9
27.7	15.5	55.8	18.4	32.8

1) Trimethyl amine.
2) Ethylene oxide.

cycle. Obviously, the expected bulking of the wood did not take place during the ethoxylation. The swelling due to ethoxylation was very low, e.g. 2.4% volumetric swelling at 28% weight gain. During the first water soaking cycle, when the wood was swelled by water, a rearrangement of glycol chains into the cell walls may have taken place, giving the desired bulking, which in turn resulted in a higher ASE_2-value.

Based on these results a series of experiments was carried out aimed at incorporating the rearrangement into the ethoxylation procedure. After the completed reaction, the wood was steamed with water vapor while still in the reactor. The steaming had the extra advantage of yielding an epoxide free wood product, since water reacts with any epoxide left in the wood.

As shown in Table 3 the bound fraction was 50-60% for wood ethoxylated when o.d., and 30% for wood ethoxylated at 7.2% moisture content. The initial dimensional

TABLE 3
Ethoxylation of Pine Sapwood (70 x 5 x 85 mm), Tangential x Radial x Longitudinal) Using the Procedure 1 g TMA[1] + 1 g EO[2] Followed by 15 min Vacuum and Several 4 g Additions of EO, Followed by 30 min Vacuum and 2 h Water Vapor Treatment

Gas phase temperature °C	Moisture content %	Weight gain %	Weight gain after leaching %	Bound fraction %	ASE_1 %	ASE_2 %
100	0	30.2	16.0	53.1	40.4	18.2
110	0	29.1	17.8	61.1	25.9	10.6
100	7.2	28.2	8.5	30.3	63.1	27.6
110	7.2	26.4	8.1	30.6	51.2	17.1

1) Trimethyl amine
2) Ethylene oxide

stability (ASE_1) was increased by the steaming to 40% and 26%, for the experiments performed at 100°C and 110°C gas phase temperature, respectively. However, the dimensional stability in the second water soaking cycle remained low. When wood with a 7.2% moisture content was ethoxylated, a bound fraction of 30% was obtained. The initial antishrink efficiency was high, 50-60%, and in the second water soaking cycle the dimensional stability was also higher than for wood ethoxylated when o.d. Hence, if the wood was ethoxylated in a partly swollen state, i.e. with a moisture content above that of o.d. wood, the glycol chains were deposited in the cell walls to a larger extent, giving the desired bulking. Even the low weight gain remaining after leaching, 8%, resulted in a higher ASE_2 value than the 16-18% weight gain remaining in the wood that was ethoxylated when o.d.

The results from the microbiological testing are shown in Table 4. The ethoxylated wood was attacked by the brown rot fungus, but to a much lower extent than the unmodified control samples. No correlation could be seen between weight loss in the brown rot test and weight gain, or moisture content prior to ethoxylation. In the fungus cellar test the control samples could no longer be found after 12 months incubation time. No attack could be seen macroscopically in samples that had been ethoxylated at a moisture content of 7%, whereas samples that had been ethoxylated when o.d. were slightly to moderately attacked. Evidently the 8-10% weight gain remaining after leaching in the samples ethoxylated at 7% moisture content was more efficient in preventing microbiological attack than the 17% weight gain remaining in the samples that had been ethoxylated when o.d. Microscopic examination showed that the samples that did not appear attacked to the naked eye, were also somewhat degraded, primarily by tunneling bacteria.

The ethoxylation method using an initial reaction step with trimethyl amine, evacuation of excess amine followed by several small epoxide additions has certain similarities to the oscillating pressure method described by Barnes et al.,[9] which consists of cycles of 6 min of elevated pressure and 3 min of atmospheric pressure. Using this procedure they obtained high antishrink efficiency values. If, on the other hand, a constant pressure procedure was used, the growth rings separated during water soaking. In this study no such growth ring separation could be observed, either for samples ethoxylated using the procedure with one large addition of ethylene oxide or for those modified in the procedure

TABLE 4
Weight Loss in Soil Block Test using *Poria placenta* and Results from Fungus Cellar Test on Ethoxylated Pine Wood Samples.

Moisture content when ethoxylated %	Weight gain %	Weight gain after leaching %	Weight loss *Poria placenta* %	Fungus cellar rating[1] Macroscopic	Microscopic
control	0	-	57.1	Disintegrated	
0	32.3	17.3	23.1	3	SR 4, TB
0	31.2	17.8	13.7	2	SR 3
7.2	26.8	6.7	34.9	1	SR 3, WR 2
7.2	26.5	8.8	20.9	0	TB 2
7.2	29.8	9.5	24.2	0	TB 2, SR 1, WR 2

[1] Rating system 0-5, with 0 meaning no attack and 5 that the specimen is destroyed. SR = soft rot, TB = tunneling bacteria, WR = white rot.

with several small additions of ethylene oxide. The advantage of adding the ethylene oxide in several small charges was that it became possible to keep the internal wood temperature at a lower level. The bound fraction, a parameter not evaluated by Barnes et al., was increased by the removal of excess trimethyl amine by means of evacuation.

Particle Boards Made from Alkoxylated Chips

The preferred alkoxylation procedure described was applied to pine wood chips. Both oven-dried chips and chips conditioned to a 9.5% moisture content were used and alkoxylated with either ethylene or propylene oxide. For comparison, chips were also modified with liquid propylene and butylene oxide, respectively.

Fig. 3 shows swelling versus time in water soaking of particle boards made from alkoxylated chips. After 5 days boards made from chips modified when o.d., with gaseous ethylene oxide, swelled more than a control board made from unmodified chips (fig. 3A). Boards made from chips ethoxylated at 9.6% moisture content to 11% weight gain swelled to the same extent as the control board (fig. 3B). At 25% weight gain there was a considerable decrease in swelling, from 53% for a control board after 5 days, to 31% for the board made from ethoxylated chips.

Boards made from chips modified when o.d., with gaseous propylene oxide, swelled more than the control board (fig. 3C). When chips with a 9.5% moisture content were modified with propylene oxide, the resulting boards swelled less than the control board, e.g. after 5 days 37% for a board made from chips with 18% weight gain, as compared to 53% for the control board.

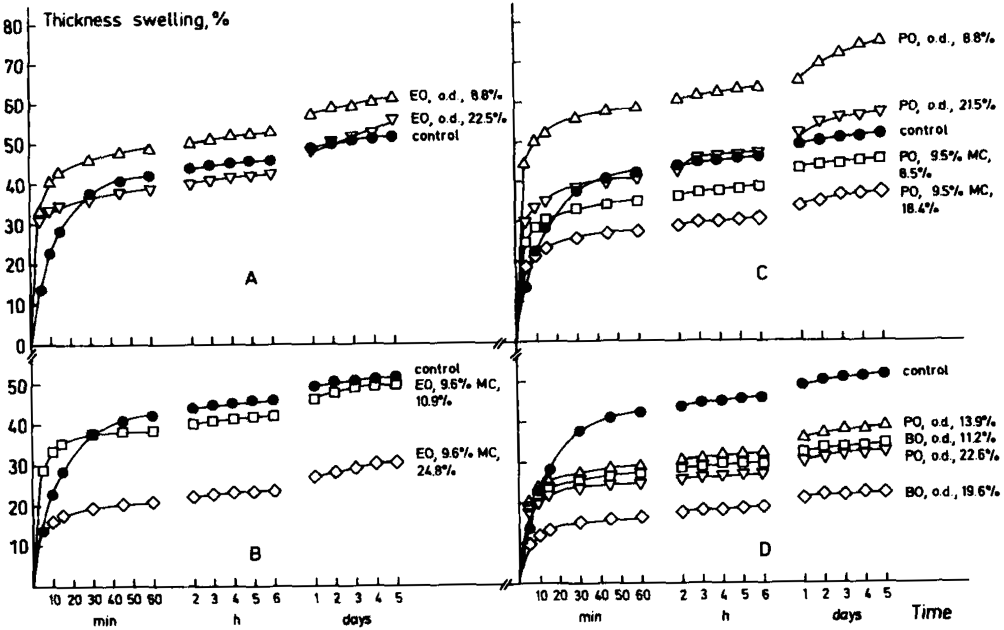


FIGURE 3. Thickness swelling versus time during water soaking (20^{o}C) of particle boards made from alkoxylated pine wood chips with varying weight gains. Chips alkoxylated when o.d. or at 9.5% moisture content (MC), with gaseous (A,B,C) or liquid (D) ethylene (EO), propylene (PO) or butylene oxide (BO).

Alkoxylation using liquid reagents gave much better reduction in swelling than did alkoxylation using gaseous reagents (fig. 3D). Modification of o.d. chips with liquid propylene oxide resulted in boards that swelled considerably less than the control board. The best results, however, were obtained for the board made from butylene oxide modified chips, with a 20% weight gain, which swelled only 23% in 5 days.

All boards made from chips alkoxylated with gaseous reagents swelled faster than the control board during the first 5-10 min of water soaking, although, for some of the boards, the final extent of swelling was less than that of a control. Boards made from chips modified with liquid alkylene oxides had approximately the same initial swelling rate as the control.

Repeated water soaking and oven-drying tests confirmed the results obtained in 5 days of water soaking (cf. fig. 4). Ethoxylation (fig. 4A) reduced swelling only when the chips were modified at a moisture content above that of o.d. wood and to a high weight gain. Chips with 11% weight gain obtained at 9.6% moisture content resulted in a board that swelled to almost the same extent as the control board after 5 cycles, whereas boards made from chips ethoxylated when o.d. swelled more than the control board. The results for modification with gaseous propylene oxide were very similar to those obtained for ethoxylation, except that boards made from propylene oxide modified chips swelled more than those made from ethoxylated chips (fig. 4B). Alkoxylation with liquid alkylene oxides (fig. 4C) resulted in boards that swelled less than the control boards. As was the case after 5 days of water soaking, the swelling of the board made from butylene oxide modified chips with 20% weight gain was half of that of the control board also after 5 water soaking cycles .

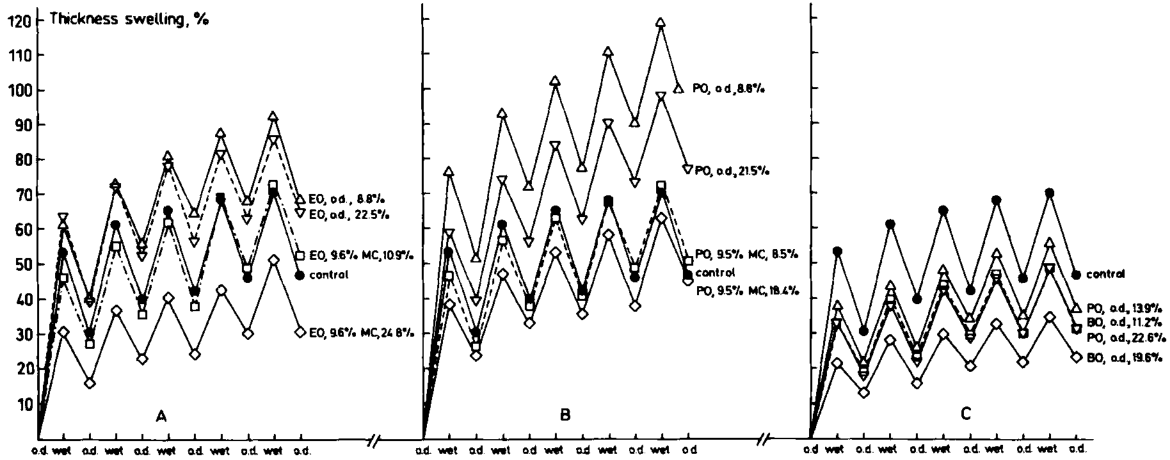


FIGURE 4. Changes in thickness in repeated water soaking (20°C) and oven-drying of particle boards made from alkoxylated pine wood chips with varying weight gains. Chips alkoxylated when o.d. or at 9.5% moisture content (MC), with gaseous (A,B) or liquid (C) ethylene (EO), propylene (PO) or butylene oxide (BO).

TABLE 5

Weight Gain and Bound Fraction for Pine Wood Chips Alkoxylated with Different Gaseous or Liquid Epoxides

Treatment	Moisture content %	Weight gain %	Weight gain after leaching %	Bound fraction %
EO[1], gas	0	8.8	4.5	50.8
"	0	22.5	12.8	57.0
"	9.6	10.9	2.5	23.4
"	9.6	24.8	12.2	49.1
PO[2], gas	0	8.8	3.9	44.4
"	0	21.5	13.6	63.3
"	9.5	8.5	2.1	24.7
"	9.5	18.4	8.7	47.3
PO, liquid	0	13.9	4.4	32.0
"	0	22.2	15.1	68.1
BO[3], liquid	0	11.2	6.9	61.3
"	0	19.6	15.2	77.4

1) Ethylene oxide
2) Propylene oxide
3) Butylene oxide

The leachability of chips used to make particle boards is shown in Table 5. The bound fraction was considerably higher for modifications made with liquid alkylene oxides than for those made with gaseous reagents, with 77% and 68% bound fraction for butylene oxide and propylene oxide modifications, respectively. A smaller bound fraction was found for the two modifications made with gaseous alkylene oxides. For wood chips that had been alkoxylated with gaseous reagents when o.d., the bound fraction was approximately 60%. When the modification had been done at 9.5% moisture content the bound fraction reached almost 50% at high weight gains.

For all modifications the bound fraction was higher at a higher original weight gain. Unmodified wood looses 1-2% of its weight during water leaching. This has not been compensated for in the calculation of the bound fraction, and so this weight loss must be considered to influence the figure obtained for bound fraction to a higher extent at low weight gains.

From the results obtained from tests on both dimensional stability and leachability it is evident that modifications made with a liquid reagent were much more efficient than those made with a gaseous reagent. The same was found to be true for acetylation of wood with liquid and gaseous reagents, respectively.[13,14] There is evidently a difference in which parts of the wood cell wall gases and liquids penetrate and are able to react with. Probably the gaseous reagents do not have enough capacity in swelling the cell wall. This is further supported by the fact that better results were obtained when wood with a moisture content above that of o.d. wood, where the cell walls are already in a partly swollen state, were alkoxylated with gaseous reagents. Experiments in which the wood was pre-impregnated with an aqueous solution of triethyl amine, oven-dried, and then modified with gaseous ethylene oxide resulted in better wood properties than when gaseous trimethyl amine was used as a catalyst in ethoxylation of o.d. wood.[11] Also in this case the wood was probably partly swollen prior to ethoxylation.

CONCLUSIONS

The preferred procedure for alkoxylation of wood with gaseous reagents consists of an initial reaction step including trimethyl amine, alkylene oxide and wood, evacuation of excess amine followed by several small additions of alkylene oxide and finally a water vapor treatment incorporated at the end of the process.

Alkoxylation with liquid alkylene oxides was found to be much more efficient in increasing the dimensional stability than was alkoxylation with gaseous reagents.

The reduction in swelling was larger for wood that had been alkoxylated at a moisture content above that of o.d. wood than for wood that had been alkoxylated when o.d., although more leachable substances were produced when alkoxylating moist wood.

The resistance to biological degradation was enhanced by gas phase ethoxylation.

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