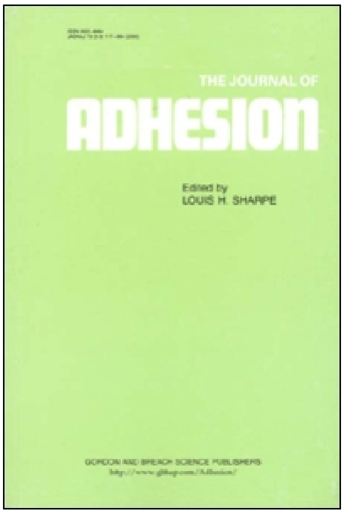

The Journal of Adhesion
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713453635

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To cite this Article Leitheiser, Robert H. , Bogner, Ben R. , Grant-Acquah, Frank C. , Johns, William E. and Plagemann, Walter(1982) 'A Water-Dilutable Furan Resin Binder for Particleboard', The Journal of Adhesion, 14: 3, 305 — 313

To link to this Article: DOI: 10.1080/00218468208073210

URL: http://dx.doi.org/10.1080/00218468208073210

J. Adhesion, 1982, Vol. 14, pp. 305–313
0021-8464/82/1404-0305 $06.50/0

A Water-Dilutable Furan Resin Binder for Particleboard

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(*Received December 5, 1981; in final form, May 10, 1982*)

A water-dilutable furan resin made from agricultural residues can be used as a binder for particleboard. Board properties obtained were comparable to those obtained with commercial phenolic binders. Board costs can be significantly reduced by using low cost ammonium lignin sulfonate as a resin extender.

INTRODUCTION

The forest products industry in the United States consumes approximately one billion pounds annually of urea-formaldehyde and phenol-formaldehyde resins. Scarcity of quality round wood for the manufacture of lumber and plywood is forcing greater usage of composite wood products, such as structural flakeboard, Comply® panels and Comply® lumber. This is especially true for exterior grade applications which will require greater quantities of resin binders with better moisture resistance in the future.

The oil embargo of 1974 and the recent rapid escalation of crude oil prices by OPEC nations, as well as the current political instability in many of the oil producing countries, encourage the development of alternate resinous binders

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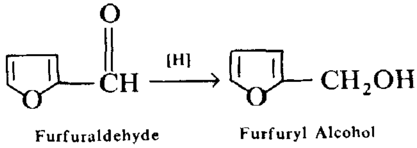
Presented at the First Annual International Symposium on Adhesion and Adhesives for Structural Materials, Washington State University, Pullman, WA 99164, U.S.A., Sept. 29–Oct. 1, 1981.

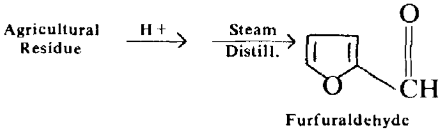
† To whom inquiries should be addressed.

not dependent on crude oil. The objective of this work was to demonstrate the suitability of newly developed resole-type furan resins, which are derived from renewable agricultural resources, as exterior grade adhesives.

FURAN RESIN CHEMISTRY

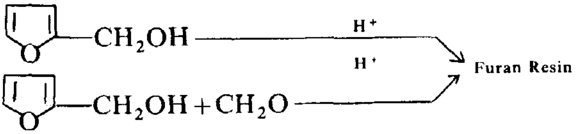
The starting material for furan resins is furfuraldehyde which is hydrogenated to furfuryl alcohol.





Furfuraldehyde is prepared from agricultural residues such as corncobs, rice hulls, oat hulls or sugarcane bagasse by digestion with acid followed by steam distillation.

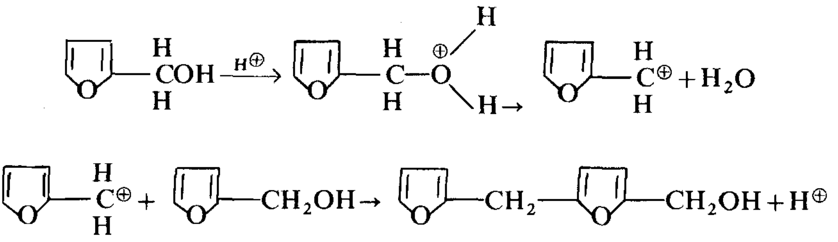
Furan resins are prepared by the homopolymerization of furfuryl alcohol or by the copolymerization of furfuryl alcohol with formaldehyde under mildly acidic conditions (pH 2.0–2.5).



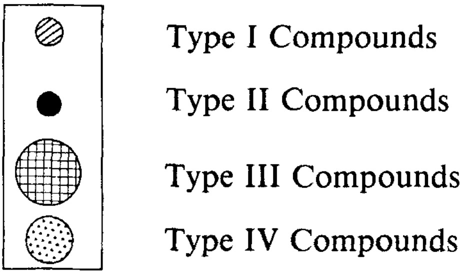
Note: Under strongly acidic conditions, furfuryl alcohol can polymerize with explosive violence.

Resins prepared in this fashion are novolac in nature since they contain low levels of hydroxyl groups (~6%). These resins are relatively nonreactive and require a cross-linker such as hexamethylenetetramine or furfuraldehyde to effect cure.

The commonly accepted mechanism for the polymerization of furfuryl alcohol is initial protonation of the furfuryl alcohol followed by the splitting off of a molecule of water to give a carbonium ion. Displacement of a proton from a second molecule of furfuryl alcohol by this carbonium ion would give a homolog of furfuryl alcohol. Repetitive displacement would give furan resins.



This reaction scheme is too simplistic, as is shown by a TLC (Thin Layer Chromatographic) plate of a conventional furan resin.



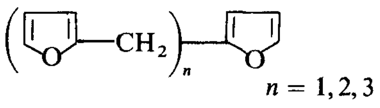
TLC plate of conventional furan resin.

By using a Kontes Model K-4 95000 Densitometer, the amount of the various types of compounds can be determined to be as follows:

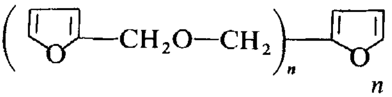
Type I	10%
Type II	5%
Type III	45%
Type IV	40%

The various types of compounds were separated by column chromatography and each type fractionated by gel permeation chromatography. Individual oligomers were identified by using NMR, IR and other analytical techniques. Identification of Type I through IV is as follows:

Type I Homologs of Difurfurylmethane

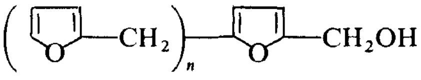


Type II Homologs of Difurfuryl Ether



$n = 1, 2, 3$

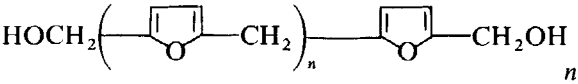
Type III Homologs of Furfuryl Alcohol



$n = 0, 1, 2, 3$

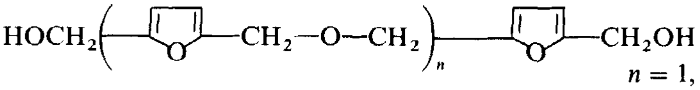
Type IV Polyfunctional Resin

The Type IV material did not migrate in the chromatographic column, hence could not be separated into components for characterization. Based on NMR, IR and other data, it is believed to consist primarily of polymer terminated on both ends by hydroxyl groups such as:



$n = 1, 2, 3 \ldots.$

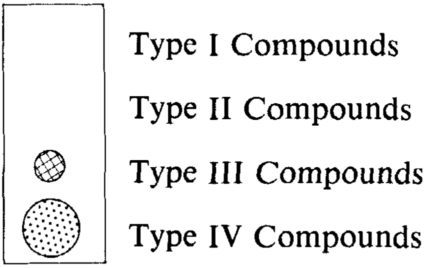
or



$n = 1, 2, 3 \ldots.$

These results confirm the work of Wewerka, Loughran and Walters[1] who separated and identified compounds of Type I, II and III from acid-polymerized furan resins.

Recently, a proprietary procedure was developed for making a reactive, self-polymerizable furan resin which contains approximately 16% hydroxyl. This resin is quite different in composition from a conventional furan resin as is illustrated by the following TLC chromatogram.



TLC plate of water-dilutable furan resin.

There is an absence of non-functional, non-polar Type I and Type II compounds and only 5–15% of the monofunctional furfuryl alcohol homologs. The majority of the resin (85–95%) did not move from the origin of the TLC plate, hence would appear to consist of highly polar, polyfunctional Type IV compounds.

The high hydroxyl content imparts a unique property to this type of resin, being offered commercially as FaRez® B-260,† of ready dilutability with water to give a stable, apparently true, solution. The resin itself is not soluble in water; however, up to 50% water by weight can be stirred into the resin, resulting in a dramatic reduction in viscosity as shown by the data in Figure 1.

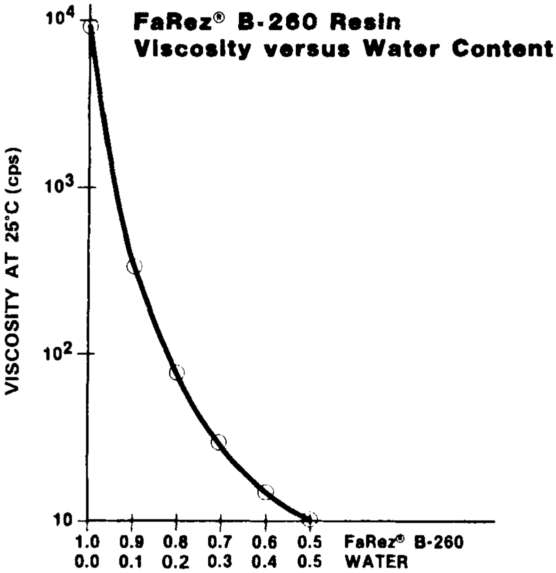


FIGURE 1 Viscosity reduction resulting from water dilution.

EXPERIMENTAL AND RESULTS

Particleboard screening studies

The ready dilutability and chemical reactivity of this resolic type resin suggested its use as a particleboard binder. Initial screening studies showed that this resin could be easily applied from a water solution by spraying onto chips in a rotating drum. The resultant chips can be molded into a board

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† Available from The Quaker Oats Company.

with acceptable internal bond strength and water boil resistance (see Table I). All boards in this study, unless otherwise noted, were prepared using Douglas fir Pallman flakes dried to a moisture content of less than 7%. A resin level of 7% based on weight of oven dried wood was used with a press temperature of 350°F, a press close cycle of one minute, and a press time of 7 minutes.

Initial boards, made without the addition of catalyst, gave poor results. Insufficient cure was suspected; hence, a number of latent catalysts were studied. The catalysts of choice, from a handling and performance point of view, were 50% water solutions of either maleic anhydride or oxalic acid. As can be seen from Table I, IB (Internal Bond), MOR (Modulus of Rupture), Wet MOR (2 hour water boil) and percent swell were comparable to the phenolic resin controls. One item to note is the last furan resin example in Table I where properties, especially water boil resistance, were dramatically improved as a result of better resin distribution on the chips achieved by increasing air atomization pressure. The effect of efficiency of resin application needs further study.

Initial screening studies at WSU† demonstrated that addition of 10% ammonium lignin sulfonate (50% water solution of Orzan A from Crown Zellerbach) to a water dilution of the resolic furan resin gave a dramatic improvement in board properties as compared to the use of the resin without catalyst. Although subsequent studies showed that the use of acidic catalysts such as maleic anhydride and oxalic acid gave similar increases in board properties, there was continued interest in the use of ammonium lignin sulfonate (ALS) as catalyst and extender because of its low lost. Consequently, a series of boards were made using different ratios of ALS to resin. As can be

TABLE I

FaRez® B-260 boards—7% resin

Catalyst	%	IB	MOR	Wet MOR (2 hr WB)	% Swell	Data source
—	—	21	850	—	—	WSU
MA	0.2	130	1840	688	—	WSU
MA	0.5	94	2531	1067	42	QO
MA	1	124	2557	961	44	QO
Ox	0.5	95	2358	1046	39	QO
Ox	1	101	2226	992	35	QO
MA	1	135	2762	1879	20	QO
phenolic[a]		108	2381	1032	45	QO
phenolic[b]		100	2551	1027	40	QO

[a] Borden, Inc., Adhesives & Chemicals Div.
[b] Georgia–Pacific Corporation, Chemical Div.

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† Forest Products Laboratory, Washington State University, Pullman, WA 99164.

TABLE II

FaRez® B-260/ALS boards

FaRez® B-260 %	ALS %	IB	MOR	Wet MOR	% Swell
6.3[a]	0.7	120	2300	—	—
6.0[a]	1.0	147	1965	710	—
6.0[b]	1.0	97	2518	1072	46
5.5[b]	1.5	64	2364	1220	56
5.0[b]	2.0	58	2233	608	85

Data source: [a]WSU; [b]Quaker Oats.

seen in Table II, up to 14% of the resin can be replaced with ALS without any significant effect on properties; and up to 21% can be replaced with minor effects other than a reduction in IB. At 20% replacement, initial MOR shows little decrease; IB and moisture resistance are still much better than that of UF bonded boards in that the samples survived the water boil test.

The effect of press time on board properties

The enhanced chemical reactivity of this resolic type furan resin suggested the opportunity for higher production rates and lower press temperatures than can be achieved with commercial boards bonded with phenolic resins. In order to verify enhanced production rates, a series of boards were run at varying press times. Preliminary results are given in Figures 2 to 5. As in the previous studies, the press temperature was kept at 350°F, but the press time was varied from 2.5 to 6 minutes. In earlier experiments, press times were kept constant at

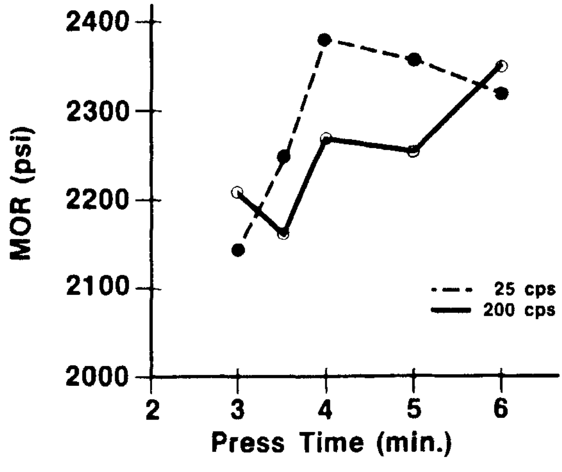

FIGURE 2 Effect of press time on modulus of rupture.

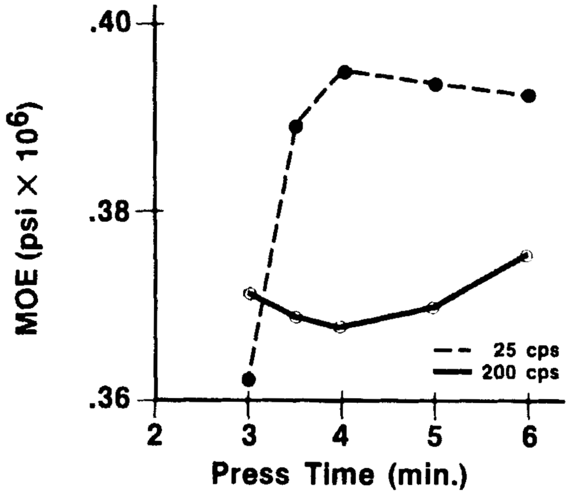

FIGURE 3 Effect of press time on modulus of elasticity.

7 minutes. Two viscosity levels (200 cps and 25 cps, achieved by dilution of the resin with water) were tested. As can be seen from the figures, significantly better results were achieved with the 25 cps resin solution, presumably because of better resin distribution on the chips. From these results a press time of 4 min would appear to be optimum with only IB increasing after 5 and 6 min and wet MOR only after 6 min in the press.

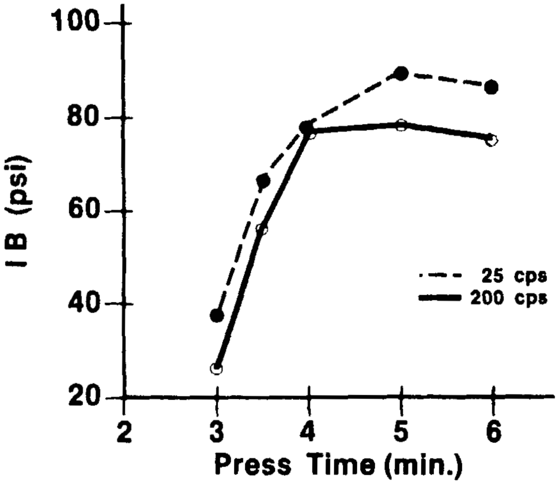

FIGURE 4 Effect of press time on internal bond.

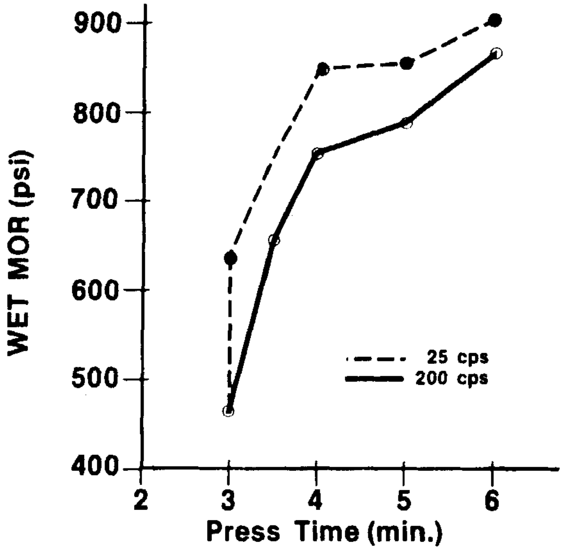

FIGURE 5 Effect of press time on wet MOR after two-hour boil.

CONCLUSIONS

Based on results obtained to date, the following conclusions were reached:

1) Furan resins can be used to make exterior grade particleboard.
2) Resolic resins seem uniquely suited for use as particleboard binders because of ready dilutability with water to give low viscosity solutions for spray application.
3) Laboratory studies indicate that a resolic furan resin gives board properties comparable to those obtained with commercial phenolic binders.
4) Ammonium lignin sulfonate is usable as a low cost catalyst/extender for resolic type furan resins. The use level of ALS will be determined by the degree of water resistance needed in the final board.
5) Resolic type furan resins are readily dilutable with water and can be easily applied using conventional equipment.
6) Results suggest that a shorter press cycle can be used with resolic type furan resins as compared to that used with phenolic binders.

Reference

E. M. Wewerka, E. D. Loughran and K. L. Walters, *J. Appl. Polym. Sci.* **15**, 1437–1451 (1971).