EFFECT OF EXTENDING HOT-PRESS, UREA-RESIN GLUE WITH RYE FLOUR ON STRENGTH AND DURABILITY OF THE GLUE JOINTS

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EFFECT OF EXTENDING HOT-PRESS, UREA-RESIN GLUE WITH RYE FLOUR ON STRENGTH AND DURABILITY OF THE GLUE JOINTS

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Introduction

Shortly after the introduction of synthetic resins of the urea type as hot-press glues for woodworking, it was discovered that they could be mixed with fillers or extenders that did not destroy the desirable working characteristics or dry strength of the glue and that served to decrease the cost of the glue line considerably. One of the extenders commonly used at first was rye flour and its use persisted to some extent although it was later replaced by wheat flour in many cases.

Experiences in commercial practice and early tests demonstrated that joints made with highly-extended, urea-resin glues by hot pressing possessed high dry strength and appeared suitable for use in many plywood operations. The question was raised, however, as to the effect of the addition of an extender, like flour, on the durability of joints that might be exposed to severe service conditions. Previous tests had demonstrated that joints made with pure urea-resin glues by hot pressing were highly resistant to exposures involving water at ordinary temperatures, to dampness, and to conditions favoring the development of micro-organisms. The addition of fillers of an organic type that might be affected by water and micro-organisms, however, might decrease the resistance of the resin-glue joints to these agencies. This report, therefore, covers empirical tests to determine the effect of rye flour filler in a hot-press, urea-resin glue on the dry strength, initial water resistance, and durability of the joints under severe conditions of exposure.

Experimental Procedure

Six glue mixtures were used to glue plywood panels from which test specimens were taken. In each case, the ratio of catalyst (or hardener) to resin remained the same, but the amount of extender varied in the different mixtures from none to twice the amount of resin. The extender used was rye flour and the glue was of the urea-formaldehyde type (Uformite 430 from the Resinous Products & Chemical Company) marketed in a liquid form in which the concentration of the resin was reported to be approximately 70 percent. This reported concentration was not checked but flour extender was added on the assumption that 100 pounds liquid resin contained approximately 70 pounds dry resin. The mixture of resin as received, plus the amount of hardener recommended, was of a suitable consistency for spreading. When flour was

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added, the mixture was thickened and addition of more water was required to reduce the mixture to a suitable spreading consistency. In all the mixtures containing flour, the attempt was made to add such an amount of water that the consistency of the mixture was approximately the same as that of the straight glue mixture containing no flour.

Following is a tabulation of the proportions used in preparing the glues:

	Constituents	Parts by weight	Percent flour in the dry glue line (based on amount of resin)
(1)	Liquid resin	100	0
	Hardener	8	
(2)	Liquid resin	100	10
	Hardener	8	
	Flour	7	
	Water	7	
(3)	Liquid resin	100	25
	Hardener	8	
	Flour	17.5	
	Water	20	
(4)	Liquid resin	100	50
	Hardener	8	
	Flour	35	
	Water	35	
(5)	Liquid resin	100	100
	Hardener	8	
	Flour	70	
	Water	70	
(6)	Liquid resin	100	200
	Hardener	8	
	Flour	140	
	Water	150	

Of these glues, numbers 2 to 6 were prepared and used in February 1939, while number 1 was prepared and used in January 1938. Glues number 2 to 6, therefore, are all from the same shipment of resin while glue number 1 was from a prior shipment of the same resin.

The preparation of veneer for gluing followed the same procedure whether the gluing was done in 1938 or 1939. A quantity of 1/16 inch yellow birch veneer, selected for smoothness, firmness, and straightness of grain, was conditioned to approximate equilibrium with 30 percent relative humidity at 80° F. (about 6 percent moisture content) before gluing. Eighteen 3-ply panels, 12 by 12 inches in size, were made with each glue and, after gluing, the panels were stickered and again conditioned to approximate equilibrium with 30 percent relative humidity before they were cut into test specimens.

The gluing conditions were held nearly constant throughout the gluing with the 6 different mixtures. The amount of glue spread varied from 19 to 23 grams per square foot with most of the surfaces receiving 20 grams per square foot (44 pounds per 1,000 square feet). The assembly period varied over the range from 10 to 30 minutes, but in most cases the panels were pressed 15 minutes after they were spread. A gluing pressure of 200 pounds per square inch was used throughout. The temperature of the platens was approximately 280° F. and the time in the press was 5 minutes.

From each panel, 30 plywood shear test specimens of conventional design were obtained. Of these, 5 specimens were tested dry at a moisture content of approximately 6 percent, and 5 were tested wet after soaking in water for 48 hours. The results of these tests were examined, all specimens from the panels containing defective joints were discarded and specimens for further tests were taken only from those panels in which the joints were well made. The test values of specimens of panels shown to be suitable for further testing were tabulated according to the glue mixture used and averages were determined for both wet and dry tests. These average values then served as controls and are shown in the first two rows of Table 1. Even though the specimens glued with the straight resin were prepared about a year prior to the remainder, the test values compare reasonably well with those from the specimens glued with 10 percent extension. The average wet test values of the two groups are well within experimental error of duplicate tests on the same glue. In dry test values, those specimens glued with the straight resin averaged 24 pounds per square inch higher in strength and 20 percent lower in wood failure than those specimens glued with 10 percent extension. Even these variations are no greater than often occur in successive tests on the same glue mixture. Since the control averages show no discrepancies, specimens glued with the straight resin may be used as a basis for comparison even though they were glued prior to the others.

The remaining specimens of the panels selected for further tests were arranged in sets according to the glue mixture used. Holes (3/8 inch) were then bored near one end of each specimen. From each set of specimens glued with one glue mixture, 4 groups of 80 specimens each were selected at random and each group strung on metal rods to facilitate handling through the exposure cycles (Fig. 1). Spacers were used between specimens to provide for circulation of air and prompt change in moisture content in the cyclic tests.

One such group of specimens from each glue mixture was then soaked continuously in water at room temperatures. A second group from each glue mixture was exposed continuously in a room where the temperature was maintained at 80° F. and the relative humidity at 97 percent. A third group from each glue mixture was exposed to a repeating cycle that consisted of 2 days soaking in water at room temperatures followed by 12 days drying in an atmosphere maintained at 80° F. and 30 percent relative humidity, and a fourth group was subjected to a repeating cycle that consisted of 2 weeks exposure at 80° F. and 97 percent relative humidity followed by 2 weeks at 80° F. and 30 percent relative humidity.

At the end of each interval of 2, 4, 6, 12, 18, 24, 30, and 36 months, 5 specimens were removed from each group and tested in the plywood shear testing machine. The test values of each such set of 5 specimens

were averaged and the results are recorded in the body of Table 1. Table 2 shows the same results calculated as a percentage of the dry test values for the joints made with unextended glue. The testing schedule was so arranged that, in the case of the "cyclic" tests, the testing was done at the end of the "dry half" of the cycle. The specimens that were soaked continuously were tested promptly upon removal from the water before they had time to dry out. Specimens exposed in 97 percent relative humidity were likewise tested promptly upon removal from the conditioning room before the moisture content of the samples changed appreciably.

Results and Discussion

This report summarizes the results up to the end of the 36th month. Sufficient specimens were provided to continue the test over a period of some 7 years if the joints proved sufficiently durable. Consequently the tests are being continued and further results will be available. At this period of 36 months, however, certain significant trends are apparent that can be changed but little regardless of the future course of the results.

Continuous exposure to 97 percent relative humidity

The effect of extending this hot-press, urea-resin glue with rye flour was most pronounced in the tests involving continuous exposure to 97 percent relative humidity and 80° F., which is a condition favoring the development of molds and wood-destroying fungi.

Glue joints made with the urea-resin glue extended with 200 percent (based on the weight of the dry resin) of rye flour barely had strength to hold the plies together at the end of the second month of exposure in 97 percent relative humidity (last column of Table 1). By the end of the 18th month the last specimen in the group had failed completely. This comparatively low resistance to attack by dampness and micro-organisms is slightly lower than shown by casein glue joints in previous tests under similar conditions.[1]

Glue joints made with the urea resin extended with 100 percent of rye flour had lost nearly all their strength by the end of the 12th month. While several of the remaining specimens were still hanging together at the end of the 18th month, the strength was so low that, of the 5 specimens selected for test at that period, none was strong enough to give a reading on the testing machine. As compared to casein glue joints under similar conditions,[1] joints made with this mixture of equal parts of urea resin and flour were somewhat superior but not to a marked degree. Under the conditions of the test, yellow birch veneer usually showed marked evidence of rot in about two years and casein glue joints usually failed before the wood had developed much evidence of decay.

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[1] Exposure tests on plywood. Don Brouse. Mech. Engr. 60, 852-56, Nov. 1938.

Joints made with the urea resin extended with 50 percent of rye flour showed greater resistance to attack by micro-organisms than the joints made with more extended mixtures, but at the end of 24 months the joint strength was very low although most of the specimens were still holding together.

The drop in strength of joints with 25 percent extension was gradual throughout the test period and slower than with any of the higher dilutions. By the end of the 30th month, however, the test values were low and the joints were no longer reliable although nearly all the specimens were holding together.

The specimens glued with a mixture containing 10 parts of flour, appeared comparatively clean and free from mold in contrast to the specimens glued with more extended mixtures which were supporting considerable mold growth on the surface (Fig. 1). At the end of 36 months, the glue diluted with 10 parts of flour showed somewhat lower test values than were shown by the undiluted glue. Apparently the 10 percent dilution did not reduce to a significant degree the resistance of the glue to continuous high humidity.

Specimens glued with the unextended urea resin decreased gradually in joint strength and at the end of 36 months the average test value was reduced to some 28 percent of the original dry test values (Table 2), with zero wood failure. The resistance shown was considerably less than shown by hot-press phenolic glues in previous tests,[1] which, at the end of 36 months, averaged about 60 percent of the original dry joint strength with more than 90 percent of the failure in the wood. Joints made with hot-press, paraformaldehyde-blood glues[1] averaged about 35 percent of the original dry test values, with nearly all the failure in the wood.

Exposure to a repeating cycle of 2 weeks in 97 percent relative humidity followed by 2 weeks in 30 percent relative humidity

This exposure combines mechanical stress on the glue joint (when the plies tend to swell or shrink with moisture changes) with exposure to mold attack during the time the specimens are under conditions favoring development of micro-organisms. If the glue line is completely resistant to attack by micro-organisms, this test is not so severe as a soaking-drying cycle, but if the glue is not so completely resistant to mold attack, it may prove more severe than a soaking-drying cycle in which test molds and fungi have little chance to develop. In the case of joints made with hot-press, urea-resin glues highly extended with rye flour, this exposure proved more severe than the soaking-drying cycles. In many cases the specimens failed completely and almost as rapidly as those exposed continuously to 97 percent relative humidity.

Specimens whose joints were glued with a mixture containing 200 percent of rye flour had failed completely in this cycle at the end of the 6th month. This rate of failure is even more rapid than has been observed in previous tests[1] with joints made with casein glue.

Joints made with a mixture extended with 100 percent of rye flour failed completely in about 24 months. In previous tests, joints made with

water resistant casein glues have held together for some 12 to 15 months under the same exposure cycle.

As compared with results on other glues in previous and similar tests, the behavior of the joints with the 50 percent extension, with 25 percent extension, and with 10 percent extension was somewhat better than that of joints made with casein glues, but not so good as joints made with hot-pressed paraformaldehyde-blood albumin glue.

The behavior of the joints made with the unextended resin glue resembled that of joints made with hot-press paraformaldehyde-blood glue in previous tests in the same exposure. They did not equal the average behavior of joints made with hot-press, phenolic glues in the same exposure in previous tests.

Exposure to a repeating cycle of 2 days soaking in water followed by 12 days drying in 30 percent relative humidity and 80° F.

The effect of extension of the glue on the resistance of the joints to this soaking-drying cycle was less pronounced than the effect on resistance to exposure to conditions favoring mold growth. In spite of individual irregularities and inconsistencies, however, the resistance of the joints to wetting and drying exposure apparently decreased slightly as the amount of extender was increased.

As compared to previous similar tests on other adhesives, the entire group falls between the hot-pressed phenolic resins and the hot-pressed paraformaldehyde-blood glues. Joints made with mixtures containing 200 percent flour were slightly better than joints made with paraformaldehyde-blood glue and joints made with the straight urea resin were not quite so resistant as joints made with hot-pressed, phenolic-resin glue.

Continuous soaking in water at room temperatures

In general, continuous soaking in water proved to be the least severe of the exposure tests used in this experiment, but the results were reasonably consistent in indicating that the resistance of the joints to continuous soaking decreased gradually as the amount of filler added to the glue increased. The moisture resistance was high, however, even for the highest dilution with starch.

At the end of 36 months of continuous soaking, joints made with the unextended resin, with the mixtures containing 10 percent extension, 25 percent extension, and 100 percent extension, were all sufficiently strong to cause a high percentage of failure in the wood although the actual strength test values were only some 50 to 62 percent of the original dry test values of the controls. The behavior of these joints compares well with that of joints made with hot-press phenolic glues and with hot-press, paraformaldehyde-blood albumin glue in similar exposure in previous tests. Joints made with mixtures containing 50 percent of flour and 200 percent of flour in

the dry glue line were still strong enough to cause considerable failure in the wood when tested at the end of 36 months of continuous soaking. The joints made with 200 percent starch appeared considerably less resistant to continued soaking than the joints made with the less extended mixtures, but they were still more resistant than joints made with casein glues proved to be in earlier tests.[1]

Dry strength and initial water resistance

The effect of extension with starch on initial water resistance of the joints was more regular than its effect on the resistance of the joints to continuous soaking in water. This is shown in the first two lines of entries in Tables 1 and 2. As the amount of extension was increased, the initial water resistance of the joints fell off slowly and gradually up to an extension of 100 percent. With 200 percent flour, however, the decrease in water resistance was considerably greater.

The effect of extension on the dry strength of the joints was the least pronounced of any of the effects investigated in this series of tests. While the averages of the dry strengths showed a slow and gradual decrease as the amount of extension increased, the differences between the averages as the extension increased were small and even the mixtures containing 200 parts flour showed high strength values and high wood failures. So far as dry strength was concerned, good joints resulted even when the glue mixture was highly extended.

Summary

From the results of the tests up to and including the 36th month of exposure the following indications were noted:

1. The resistance of the glue joints to mold attack was most markedly affected by extension with rye flour. Joints made with a mixture containing 200 parts of flour and 100 parts of resin proved scarcely equal to joints made with casein glues in resistance to mold attack. The effect of 10 percent flour was noticeable, although of minor importance, when the joints were subjected to conditions favoring mold growth. Joints made with unextended, urea-resin glues did not prove so resistant to these conditions of exposure as joints made with hot-press, phenolic-resin glues.

2. The effect of extension on the resistance of the joints to wetting and drying cycles was less marked than the effect on resistance to conditions favoring mold growth but, in general, the resistance fell off gradually as the amount of extension was increased. The reduction caused by 10 percent of flour was slight and even the glue with 200 percent showed higher resistance than would be expected of a casein glue.

3. Continuous soaking in water did not prove to be a severe exposure for these urea-resin glue joints. Extensions of 10 percent and 25 percent of

flour caused no decrease (as compared with unextended glue) that could be detected by joint tests during 36 months. Joints made with mixtures containing 200 percent of flour were definitely lower in their resistance to continuous soaking, but still were more resistant than joints made with casein glue in previous tests under the same exposure.

4. As the amount of extension increased, the initial water resistance of the joints decreased somewhat more regularly than the resistance to continuous soaking in water.

5. Within the range of extensions used in this experiment, the dry strength of the glue joints decreased very slowly as the amount of extension increased. While even a 10 percent extension caused a slight decrease, joints made with mixtures containing twice as much flour as resin were still strong enough to develop a high percentage of wood failure on test.

6. Where maximum resistance to severe exposure is essential, particularly where the exposures involve conditions favorable to mold growth, the extension of hot-pressed, urea-resin glues with flour should not exceed 10 percent and, preferably, should be avoided.

7. If the service conditions require only good dry strength and a moderate degree of water resistance, the use of comparatively large amounts of flour extender is permissible.

Table 1.--Comparative durability of joints made with a hot-pressed urea-resin glue used without extender and with different proportions of an extender of rye flour

Time of Exposure (Months)	Glue: Without extender	Extended with 10 percent rye flour	Extended with 25 percent rye flour	Extended with 50 percent rye flour	Extended with 100 percent rye flour	Extended with 200 percent rye flour
	Average test values[1]					
None (control values)						
Dry[2] [3]	507-63	483-83	458-89	439-88	440-90	429-83
Wet[2] [4]	489-96	478-94	455-90	426-71	405-60	261- 8
	Soaked continuously in water at room temperatures[5]					
2	444- 81	426- 99	317- 66	376-53	366- 79	253-12
4	441-100	423- 81	359- 79	349-51	354- 71	235-13
6	401- 96	365- 59	350-100	297-41	300- 62	182- 2
12	396-100	379- 84	350- 80	322-61	351- 22	227- 3
18	340-100	328-100	311-100	245-20	246- 52	204-19
24	316-100	295-100	311- 92	247-68	240- 80	137-22
30	298-100	277-100	267-100	275-88	238- 82	172-29
36	313- 96	264- 96	284- 90	245-48	281-100	176-64
	Exposed continuously to 97 percent relative humidity and 80°F.[5]					
2	477- 99	470- 83	408- 43	284-16	231- 21	0-0[b]
4	392- 45	403- 59	337- 23	237-20	202- 19	25-0
6	366- 4	258- 20	235- 1	215-19	149- 16	13-0
12	309- 41	290- 40	215- 0	94- 0	79- 0	0-0[b]
18	309- 0	222- 36	140- 0	90- 1	0- 0[b]	0(18)[a]
24	155- 0	225- 20	138- 0	23- 0	18- 0[b]	.....
30	198- 0	170- 20	79- 0	14- 0	0- 0[b]	.....
36	144- 0	108- 0	49- 0	32- 0	28- 0	.....
	Exposed to a repeating cycle that consisted of 2 days soaking in water at room temperatures followed by 12 days drying at 80° F. and 30 percent relative humidity[5]					
2	479- 92	376- 45	380- 76	343-39	351- 19	330-19
4	444- 44	392- 59	397- 42	410-23	378- 23	337-14
6	446- 1	460- 68	390- 22	447-57	366- 42	374-42
12	436- 12	357- 41	345- 26	308- 4	315- 24	264-20
18	511- 82	354- 28	331- 3	289-18	292- 22	310- 1
24	440- 81	345-100	214- 40	217-20	157- 0	166- 0
30	362- 0	331- 40	282- 40	312-19	246- 20	209-41
36	394- 0	299- 5	178- 19	281-20	272- 20	169-20
	Exposed to a repeating cycle that consisted of 2 weeks in 80° F. and 97 percent relative humidity followed by 2 weeks in 80° F. and 30 percent relative humidity[5]					
2	514- 80	435- 60	392- 31	419-30	325- 26	134- 0
4	524- 25	368- 49	371- 60	321-23	220- 30	0- 0[b]
6	476- 24	355- 11	329- 2	300- 0	181- 0	0(6)[a]
12	376- 21	250- 0	203- 2	124- 0	181- 20	.....
18	410- 12	290- 20	184- 0	108- 0	46- 8	.....
24	248- 0	309- 21	183- 0	111- 0	0(24)[a]	.....
30	235- 0	328- 30	124- 0	73- 0	.....	.....
36	187- 0	60- 0	91- 0	60- 0	.....	.....

[1] In each pair of values, the first represents average joint strength in pounds per square inch, the second represents wood failure in percent.

[2] Each value is an average of 100 specimens, 5 from each of 20 panels.

[3] Specimens tested when in approximate equilibrium with 30 percent relative humidity.

[4] Specimens tested wet after soaking for 48 hours in water at room temperatures.

[5] Each value is an average of five specimens.

[a] Figures in parentheses indicate the month during which the last specimen in the group failed completely.

[b] In the five specimens taken for test the joints were still holding the plies together but the strength was so low that the specimens could not be gripped in the testing machine without failure.