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2, 4, 3',

5'-Tetrahydroxystilbene as

RESEARCH CORPORATION OF THAILAND

MISCELLANEOUS INVESTIGATION NO. 15

CHEMICAL TRANSFORMATION OF 2, 4, 3', 5'- TETRAHYDROXYSTILBENE

REPORT NO. 2

2, 4, 3', 5'- TETRAHYDROXYSTILBENE AS REINFORCING AGENT
FOR TANNIN FORMALDEHYDE ADHESIVES

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YAACOV AMIEL

BERNARD PH. ESSELINK

EVELYN BOONMEEPRASERT

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F O R E W O R D

This investigation evolved from an examination of Thai plant materials used in the production of pharmaceuticals under Research Programme No. 17 during the course of work on Research Project No. 17/6: "Pharmaceuticals from Artocarpus lakoocha".

It aims at exploring the possibility of using 2,4,3',5'-tetrahydroxystilbene which occurs in considerable amounts (about 10%) in the heartwood of Artocarpus lakoocha or mahat, a large tree abundantly available in Thailand. The first report in this series covered preliminary investigations on the possibility of producing useful chemicals from this material. The present report covers work on the possibility of using this stilbene as a reinforcing agent for tannin formaldehyde adhesives for marine phywood.

2,4,3',5'-TETRAHYDROXYSTILBENE AS REINFORCING AGENT
FOR TANNIN FORMALDEHYDE ADHESIVES

By Yaacov Amiel,* Bernard Ph. Esselink,*
and Evelyn Boonmeeprasert*

SUMMARY

This is an attempt to incorporate 2,4,3',5'-tetrahydroxystilbene into the tannin formaldehyde adhesives as a substitute for the reinforcing resin. Initial evaluations, however, show that although tetrahydroxystilbene tends to reinforce the tannin formaldehyde adhesive, it is still below the accepted standard.

I. INTRODUCTION

Reinforced tannin formaldehyde adhesives (RTF) are used in Australia for the manufacture of marine plywood. The reinforcing agent is a resorcinol-phenol-formaldehyde resin. This report describes an attempt to substitute resorcinol with 2,4,3',5'-tetrahydroxystilbene for the local plywood industry. Since tetrahydroxystilbene is a compound of two resorcinol molecules (linked to the terminal positions of an ethylene molecule), it is plausible that the tetrahydroxy compound may undergo similar chemical interactions and may replace resorcinol in reinforced tannin formaldehyde adhesives. Resorcinol has to be imported into the country and is by no means cheap whereas tetrahydroxystilbene could easily and cheaply be obtained in high yields by extracting heartwood of Artocarpus lakoocha Roxb.

II. MATERIALS AND METHODS

2,4,3',5'-tetrahydroxystilbene dihydrate was obtained by ether extraction of "puakhat", a powder extract from the heartwood of Artocarpus lakoocha Roxb. The ether was evaporated off and the tetrahydroxystilbene was recrystallized twice from ethanol/water.

* Industrial Chemistry Group, Technological Research Institute, ASRCT.

The other materials used, such as tannins, phenol, formaldehyde, resorcinol, veneer, and fillers have been detailed in Report No. 2 on Research Project No. 35/4*. The test methods, which are based on industrial practice, are also given in that report.

III. EXPERIMENTAL AND RESULTS

Preliminary experiments showed that the 2,4,3',5'-tetrahydroxystilbene reacted too vigorously under the conditions used for the preparation of resorcinol-phenol-formaldehyde resin. Hence, to slow down the reaction, the percentage of 2,4,3',5'-tetrahydroxystilbene as well as the temperature of the reaction and the pH of the reaction mixture were modified.

The final method of preparing the resin was to stir phenol (1 mole) with 40 per cent formaldehyde solution (1 mole) using sodium hydroxide as catalyst (1.45% of the weight of phenol, as a 35% solution). The mixture was heated for 60 minutes at 90°C, was then cooled to 60°C and added to a mixture of 2,4,3',5'-tetrahydroxystilbene and 40 per cent formaldehyde solution; the resulting mixture was heated at 60°C for one hour with stirring.

Four such resins were prepared, each having a different percentage of tetrahydroxystilbene. The shelf life of each resin was observed. Results are expressed in the following tabulation.

Percentage by weight of phenol	Shelf life
50% tetrahydroxystilbene	set immediately to a hard mass
20% tetrahydroxystilbene	set to a jelly after half an hour
10% tetrahydroxystilbene	has a shelf life of 1 week and longer
1% tetrahydroxystilbene	has a shelf life of more than 3 months

*"Evaluation of the properties of reinforced tannin-formaldehyde adhesives for plywood" by Shannan Deetesna and Bernard Ph. Esselink. Report No. 2 on Research Project No. 35/4 (Development of water-resistant glues from tannin). ASRCT unpublished report.

As the 50 per cent and 20 per cent resins were not suitable for mixing, the 10 per cent and 1 per cent resins were tested for their effect on the bonding strength of the tannin adhesive. It was further thought desirable to use pure tetrahydroxystilbene powder and also as alcoholic solution of tetrahydroxystilbene to introduce the tetrahydroxystilbene into the tannin adhesive.

Tetrahydroxystilbene dihydrate was incorporated in the tannin formaldehyde adhesives as follows:

1. Pure tetrahydroxystilbene dihydrate powder. The powder was mixed in the glue, percentage 3-12 per cent on dry tannin powder.
2. 40 per cent pure tetrahydroxystilbene dihydrate in alcohol. The solution was mixed in the glue, percentage 2.5-10 per cent pure tetrahydroxystilbene on dry tannin powder.
3. 10 per cent tetrahydroxystilbene dihydrate-phenol-formaldehyde resin. A tetrahydroxystilbene-phenol-formaldehyde resin was prepared with 10 per cent tetrahydroxystilbene dihydrate on phenol (by weight). This resin was mixed in the glue, percentage 7.5-30 per cent resin on dry tannin powder.
4. 1 per cent tetrahydroxystilbene dihydrate-phenol-formaldehyde resin. This resin contains 1 per cent by weight of tetrahydroxystilbene dihydrate on phenol. This resin was mixed in the glue, percentage 7.5-30 per cent resin on dry tannin powder.

To evaluate the effect of tetrahydroxystilbene a comparison was made with the following adhesive mixtures:

- (1) Pure tannin formaldehyde adhesives.
- (2) Tannin formaldehyde adhesives reinforced with phenol-formaldehyde resin.
- (3) Tannin formaldehyde adhesives reinforced with resorcinol-phenol formaldehyde resin.

With the glue mixtures plywood test panels were prepared.

Veneer conditions

Kind: yang
Thickness: 1.6 mm
Moisture content: 10-11%
Construction: yang/yang/yang

Pressing conditions

Glue spread: 180 g/m² (single glue line)
Cold press: 10 kg/cm² (10 min)
Assembly time: 45 min
Temperature: 130-140°C
Pressure: 200 kg/cm
Pressing time: 6 min

Each prepared panel was conditioned for 24 hours and then tested on:

- (1) Dry strength and knife test (British Standard 1203: 1963).
- (2) Waterboil Proof (WBP) test and knife test (British Standard 1203: 1963). The WBP test is especially important as it indicates the water and weather resistance of the plywood. Plywood of WBP strength above 200 lb/in² is considered to be satisfactory.

The results of the strength tests on the various formulations are shown in the following table:

	Tetrahydroxystilbene on dry tannin (%)	Dry strength (lb/in ²)	WBP (lb/in ²)
1.1 Pure tetrahydroxy- stilbene dihydrate	3	-	183
	6	-	164
	9	-	174
	12	-	170
1.2 40% tetrahydroxy- stilbene dihydrate in alcohol	2.5	354	207
	5	329	189
	7.5	323	140
	10	324	180
	2.5	378	187
	2.5	351	179

	Resin on dry tannin (%)	Dry strength (lb/in ²)	WBP (lb/in ²)
1.3 10% tetrahydroxy-stilbene dihydrate-phenol-formaldehyde resin	7.5	-	158
	15	-	195
	22.5	-	202
	30	-	202
	22.5	381	189
	22.5	330	200
	22.5	293	203
	22.5	276	158
	22.5	314	176
1.4 1% tetrahydroxy-stilbene-phenol-formaldehyde resin	7.5	-	181
	15	-	195
	22.5	-	210
	30	-	182
	Percentage resin (%)		
2.1 Pure tannin	-	310	161
	-	293	151
2.2 Reinforced with phenol-formaldehyde	22.5	471	261
	-	354	220
	-	435	229
	-	300	188
	-	377	235
	30	388	220
	-	302	192
	-	358	197
	-	354	196
-	314	174	
2.3 Reinforced with 20% resorcinol-phenol-formaldehyde resin	7.5	-	261
	15	-	242
	22.5	-	222
	30	-	244

IV. CONCLUSIONS AND RECOMMENDATION

From the results presented it appears that tetrahydroxystilbene, incorporated in a tannin formaldehyde plywood adhesive either directly or as a tetrahydroxystilbene-phenol-formaldehyde resin, does have some effect as a reinforcing agent. However, under the conditions tested the effect was not as great as that of phenol-formaldehyde or resorcinol-phenol-formaldehyde resins. Further, the WBP results obtained with the tetrahydroxystilbene additions usually failed to attain 200 lb/in², the lowest level considered to be adequate.

It must therefore be concluded that a basis for using tetrahydroxystilbene as a reinforcing agent for tannin formaldehyde plywood adhesives has not been established. As the adoption of tannin formaldehyde adhesives by the Thai plywood industry remains highly problematical, it is recommended that further work on the use of tetrahydroxystilbene for this purpose be postponed until the status of tannin formaldehyde adhesives has been clarified.