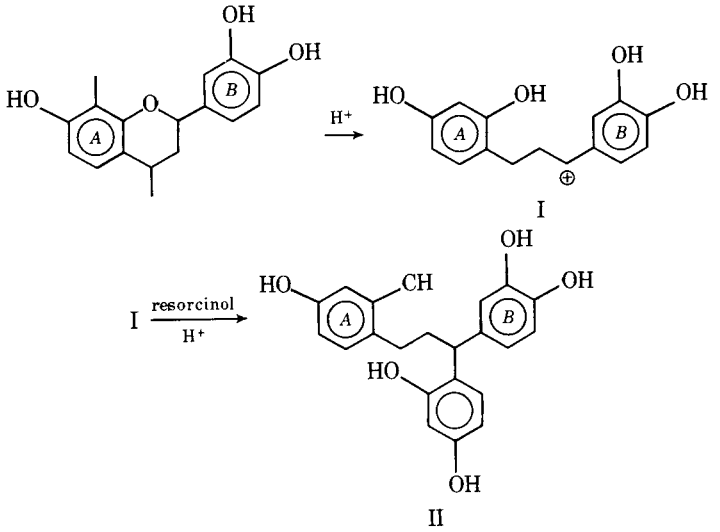
Resorcinol/ Wattle Flavonoids Condensates for Cold-Setting Adhesives

A new method to produce wattle-based cold-setting adhesives was devised following the finding from Roux and co-workers[1] that the etherocyclic ring of flavonoid units can be easily opened by acid hydrolysis in water solutions, similarly to what was already found by Brown and co-workers[2] for 4′-methoxyflavan in acid/ethanolic solutions, with the formation of a carbocation able to react with any other nucleophile present, including the phenolic rings of other flavonoid units present. As no usable adhesive could be produced by this system when only the flavonoid monomers and polymers were present, because of the formation of condensation products similar to those formed by Freudenberg and co-workers[3] between 4′-methoxy flavan and 7-hydroxyflavan and delucidated by Brown and co-workers,[4] it was decided to allow part of the carbocations formed by this method to react with resorcinol so as to form the following structure II isolated by Roux and coworkers[1] when using pure catechin for the experiment. Brown and co-workers[2] already reported similar flavonoid/phenol condensations, but without opening of the etherocyclic ring.



Structure II alone or mixed with untreated flavonoid polymers reacts easily with formaldehyde, at ambient temperature, to give cured resins. A few advantages are introduced with this system. (1) The resorcinol is grafted onto the flavonoid unit without any formaldehyde. (2) The resorcinolic *A* rings of the flavonoid units presents less steric hindrance to the reaction with formaldehyde during resin curing. (3) The disappearance of the etherocyclic ring causes better solubility of the wattle-based adhesive and eliminates skinning of the adhesive on the open glue lines.

EXPERIMENTAL

We added 2 g of a commercial defoamer and 169 g of resorcinol to 677 g of an aqueous 53% wattle-extract solution and the mixture was warmed until the resorcinol was completely dissolved. We then added 42 g of trichloroacetic acid and the mixture was refluxed for 90 min at 86°C, then cooled and stored. The pH was adjusted to 7.41 using 33% caustic soda and to the total mixture we added 24 g of 180 mesh wood flour, 34 g of 200 mesh coconut shell flour, 82 g of a high reactivity paraformaldehyde powder, and enough water to adjust the viscosity to 2500–2800 cP. The usable pot life of this glue mix, at ambient temperature, was of 2¼ hr.

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TABLE I
Strength and Percent Wood Failure Results

	Dry	24 hr cold soak	6 hr boil
Shear strength, psi	528	532	583
% wood failure	36	25	74

This glue mix was used to glue beech strips according to British Standard[5] BS 1204 part 2 for synthetic resin adhesives and the results obtained are shown in Table I.

DISCUSSION AND CONCLUSIONS

The system used is suitable to give cold-setting adhesives for wood passing the relevant British Standard specification, though the results obtained are somewhat lower than those given by previous formulations.[6] A saving in the amount of formaldehyde used is achieved as well as a considerable improvement in the skinning of the adhesive in open glue lines during long assembly times.

References

1. J. J. Botha, D. Ferreira, and D. G. Roux, CSIR unit for flavonoid chemistry, reports, 1975–1976.
2. R. Brown and W. Cummings, *J. Chem. Soc.*, 4302 (1958).
3. K. Freudenberg and J. M. A. DeLama, *Justus Liebig Annalen der Chemie* (*LEMGO*), **612,** 78 (1958).
4. R. Brown, W. Cummings, and J. Newbould, *J. Chem. Soc.*, 3677 (1961).
5. British Standard 1204–1965 part 2.
6. A. Pizzi and D. G. Roux, *J. Appl. Pol. Sci.*, in press.

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