A STUDY ON THE PROPERTIES OF SYNTHETIC ADHESIVES AVAILABLE IN PENINSULAR MALAYSIA

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CHEW, L.T., WONG, W.C., MOHD SHUKARI MIDON & MATSUMOTO, T. 1991. A study on the properties of synthetic adhesives available in Peninsular Malaysia. The properties of various types of urea-formaldehyde (UF), phenol-resorcinol formaldehyde (PRF) and polyvinyl acetate (PVAc) adhesives available in Peninsular Malaysia were determined by methods specified by Japanese Industrial Standards and a local adhesive manufacturer. The adhesives have different physical and chemical properties.

Key words: Synthetic adhesives - properties - methods of determination - adhesive formulation

Introduction

Malaysia is an important exporter of tropical hardwood plywood as well as chipboard, blockboard, laminated decking and furniture. For the production of such items, adhesives of various types are required. Peninsular Malaysia has three adhesive producers with a combined capacity exceeding the local demand of the adhesives of urea-formaldehyde (UF), phenol-formaldehyde (PF) and phenol-resorcinol formaldehyde (PRF) for plywood and laminated decking production. There are also local producers of polyvinyl acetate adhesive (PVAc), although some of it is also imported.

This study was undertaken to determine the properties of locally available adhesives and to introduce the various techniques for their determination. The results of this study would be used to determine appropriate adhesive formulations for use in subsequent glue-lamination research.

Experimental

Raw materials

Adhesive samples as well as the specifications were collected from the local manufacturers and from the sole importer.

Determination of adhesive properties

Solid content

A measured quantity of adhesive (*ca.* 1 - 2 *g*) was dried in an oven at 105 ± 1.5°*C* for 3 *h*, cooled and reweighed for determining the solid content of the adhesive.

Viscosity

About 200 *ml* of the adhesive was taken and warmed up to 25°*C*. A Brookfield Synchro-Lectric Viscometer was used to determine the viscosity of the adhesive.

pH

A pH meter was used to determine the pH of the adhesive at 25 ± 1°*C*.

Specific gravity

A simplified method was used. The weights of the same volume of water and adhesive were taken and the specific gravity evaluated.

Storage stability

The methods used were in accordance to JIS K 6801-5.3(1973a) for UF and JIS K 6 802-5.3(1973b) for PF. In these methods, about 10 *g* of the adhesive were placed in a test tube whose mouth was then covered with a plastic film. The test tube was allowed to stand in a beaker and then placed in the oven at 60 ± 2°*C* for PF and PRF and at 70 ± 2°*C* for UF. The time for the adhesive to be gelatinised was measured.

Gelation time

A measured quantity of adhesive was mixed with the specific hardener. About 10 *g* of the adhesive mix was put in a test tube which was then immersed in a constant temperature bath at 25 ± 0.5°*C* or 30 ± 0.5°*C*. The gelation time taken by the sample was recorded.

Free formaldehyde

Two methods were used. The method based on JIS K 6801(1973a) and the method adopted by Norsechem (Malaysia) Berhad are given in Appendix I.

Results and discussion

The various properties of locally available adhesives were evaluated and given in Tables 1 to 4.

Table 1. Physical and chemical properties of adhesives

Kind of adhesives	Trade name	Solid content (non-volatile content) (%)*1	Specific gravity at 25°C	Viscosity at 25°C (P)*2	pH	Supplied for:
UF	N-50	54.5 (54.3-54.7)	1.225	6.1 (2/30)	8.6	plywood
UF	N-150	63.7 (63.3-64.1)	1.270	4.9 (2/30)	7.1	particleboard
UF	N-100	53.7 (53.6-53.7)	1.224	8.5 (2/30)	8.1	plywood (less free CH_2O)
UF	UL-150	50.0 (49.7-50.5)	1.191	2.3 (2/30)	7.5	plywood
PF	PL-60M	41.4 (41.3-41.4)	1.186	1.2 (2/60)	12.1	plywood
PRF	N-45	54.9 (54.6-55.3)	1.162	8.2 (2/30)	7.7	wood lamination
PVAc	PVAc40	41.3 (41.1-41.4)	1.066	770 (4/6)	4.2	wood working
PVAc	PVAc5000	49.9 (49.9-50.0)	1.068	510 (4/6)	4.3	wood working

*1: Average of three tests and minimum and maximum in parentheses; *2: Figures in parentheses show rotor number/rotation speed in rpm

Table 2. Storage stability test*

Sample		Test condition	
		Time required until the sample is gelatinized and does not flow (G) (*h*)	
		70°C (JIS K 6801)	60°C (JIS K 6802)
UF	N-50	27 < G < 40	
UF	N-150	24 < G < 27	
UF	N-100	20	
UF	UL-150	44 < G	
PF	PL-60M		28 < G < 44
PRF	N-45		120 < G

*: JIS K 6801: Urea Resin Adhesives for Wood (The value must be 10 or more); JIS K 6802: Phenolic Resin Adhesives for Wood (The value must be 30 or more)

The solid contents of the various adhesives tested were similar to those in the instruction pamphlets given by the adhesive manufacturers to the users. UF adhesive for particleboard has higher solid content and viscosity as compared with UF adhesive for plywood production. The adhesive manufacturers have different clienteles and they produce adhesives with properties that suit their specific requirements. Hence, the results given in Tables 1 to 4 show that similar adhesives produced by different manufacturers have different values in solid contents, specific gravity, viscosity, pH, gelation time, free formaldehyde and storage stability.

Table 3. Gelation time of different adhesive mixes

Kind of adhesives	Trade name	Hardener	Amount of hardener per 100 parts of sample	pH at 20 *min* after mix hardener	Gelation time (*h:min*)	
					25°C	30°C
UF	N-50	20%NH_4Cl	3.0	4.5	0:45	0:30
		"	1.0	-	2:50	1:20
		H-75	2.5	-	3:15	2:30
		H-75	2.0	-	4:50	2:50
		20%NH_4Cl	1.0	-	1:05	1:00
		H-75	2.0			
UF	N-150	20%NH_4Cl	3.0	5.3	7<T<20	4:50
		H-75	2.5	-	T<48	5<T<20
UF	N-100	20%NH_4Cl	3.0	6.1	5:30	4:10
UF	UL-150	20%NH_4Cl	3.0	3.9	1:10	0:50
		MH-6	2.0	-	4<T<20	3:20
		MH-6	3.0	-	5:00	2:45
PRF	N-45	H-110	20	-	2:00	1:15
			15	7.5	2:05	1:30
			10	-	3:30	2:05

Table 4. Free formaldehyde in UF adhesives (values are shown in percentage)

Trade name	Method 1 (JIS K 6801)		Method 2 (Norsechem method)			
			Without methanol		With methanol	
	Measured	A & R	Measured	A & R	Measured	A & R
N-50	1.15	1.16	1.69	1.52	1.85	1.60
	1.10		1.65		1.69	
	1.23	R=0.13	1.22	R=0.47	1.26	R=0.59
N-150	0.82	0.82	0.54	0.74		
	0.81		0.80			
	0.82	R=0.01	0.87	R=0.33		
N-100	0.48	0.48	0.30	0.30		
	0.45		0.29			
	0.51	R=0.06	0.30	R=0.01		
UL-150	1.39	1.46	1.69	1.52		
	1.57		1.65			
	1.43	R=0.18	1.22	R=0.47		

A & R - Average and (maximum - minimum) Range

The results in Table 1 also indicate that for the same type of adhesive (Samples 1 to 4), that with higher solid content also had higher specific gravity. However, this relationship was less pronounced in the case of PVAc adhesives.

The storage stability values of locally produced adhesives are generally high exceeding the values given in JIS K 6801 (G > 10 for UF) and JIS K 6802 (G > 30 for PF) as given in Table 2.

The gelation times of the different adhesive mixes are given in Table 3. With

higher temperature, the gelation times were shorter. It is possible to change the gelation time of an adhesive mix by using a greater quantity of the specific hardener. Gelation times could also be changed with the use of different hardeners. Local manufacturers do not normally change the basic production parameters for making adhesive but they adjust the properties of the adhesive mix to suit the need of their customers through changing the components or the quantity of the individual components of hardeners (local adhesive mills personal communication).

Furthermore, with the addition of hardener, the pH of the adhesive mix changed with time (Figure 1). This change in pH for the UF adhesive mix accelerated the condensation reaction to facilitate the curing of the adhesive.

The two methods used for the determination of free formaldehyde in UF adhesives yielded different results (Table 4). With Method 1, the measured values were higher in three of the four adhesives tested. Moreover, there were less deviations in values among the readings for each particular sample with the use of Method 1 because the end point of the titration could be determined more easily than in Method 2.

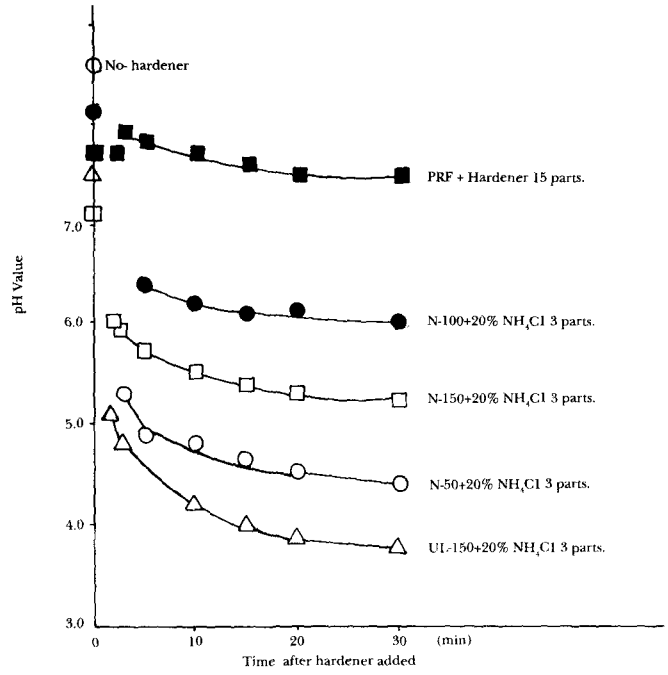

Figure 1. pH value changes after hardener added

Conclusion

The properties of the locally produced and available adhesives normally

used in the wood based panel and wood working industries were determined using the various methods. The techniques for determining the various adhesive properties, especially those based on the Japanese Standards, were successfully tried out. The basic properties of similiar adhesives manufactured by the local adhesive companies varied. Hence, knowledge of the properties of the synthetic adhesives available in Peninsular Malaysia is important, especially in the formulation of adhesive mixes for the wood lamination industry.

References

JIS. 1973a. *Urea resin adhesives for wood. JIS K 6801 - 1973.* Japan Industrial Standard.
JIS. 1973b. *Phenol resin adhesives for wood. JIS K 6802 - 1973.* Japan Industrial Standard.

APPENDIX 1

Determination of free formaldehyde in UF adhesives

Method 1 (JIS K 6801)

Apparatus:
1) Erlenmeyer flask, 250 *ml*
2) Graduated cylinder, 50 *ml*
3) Measuring pipette, 10 *ml*
4) Burette, 50 *ml*
5) Bulb pipette, 10 *ml*

Reagent:
1) N/10 sodium hydroxide solution
2) N/10 hydrochloric acid
3) Methyl red and methylene blue mixed indicator. Dissolve 0.025 *g* each of methyl red and methylene blue in 50 *ml* of 95% ethyl alcohol.
4) 10% ammonium chloride solution
5) N Standard hydrochloric acid

Procedure:
1) Weigh approximately 15 *g* of sample to the nearest 10 *mg* into a 250 *ml* Erlenmeyer flask.
2) Add 50 *ml* of distilled water and mix well.
3) Add about two drops of indicator (methyl red and methylene blue mixed) and neutralize the solution with N/10 sodium hydroxide or N/10 hydrochloric acid. (Reddish purple: acidic, greyish blue: pH 5.4, green: basic)
4) Add 10 *ml* of 10% ammonium chloride with measuring pipette and 10 *ml* of N sodium hydroxide with a bulb pipette.
5) Close the flask with a stopper. Shake the solution. Stand the solution for 30 *min* at 20°*C*, shaking the solution occasionally.
6) Titrate the solution with N hydrochloric acid until the colour changes from green to greyish blue.
7) Run a blank test also.

Calculation: Free Formaldehyde (%) = 0.045 × (B-A) × F/S × 100,

where A = the amount of N hydrochloric acid consumed by the sample (*ml*); B = the amount of N hydrochloric acid consumed in the blank test (*ml*); F = Factor of N HCl; S = Weight of the sample (*g*)

The basic chemical reaction is as follows:

$$6CH_2O + 4NH_4Cl = (CH_2)_6N_4 + 4HCl + 6H_2O$$

Method 2

Apparatus:
1) Erlenmeyer flask, 250 *ml*
2) Graduated cylinder, 50 *ml*
3) Beaker, 250 *ml*
4) Thermometer
5) Burette, 50 *ml*
6) Analytical balance
7) Beaker, 250 *ml*

Reagent:
1) 1.0 *M* sodium sulfite. Dissolve 126 *g* of sodium sulphite, anhydrous reagent grade, in about 100 *ml* of distilled water. Store in a cool dark place. New solution should be prepared every three weeks.
2) Thymolphthalein indicator. Dissolve 0.04 *g* in 100 *ml* of 50% ethyl alcohol.
3) N Standard sulphuric acid.
4) Methanol.

Procedure:
1) Weigh about 15 *g* of resin, to the nearest 10 *mg*, into a 250 *ml* Erlenmeyer flask.
2) Add 20 *ml* of distilled water and mix well. If the resin is difficult to dissolve, 20 *ml* of methanol may be added. Add about 0.5 *ml* of indicator and dilute NaOH until a slight blue colour is obtained.
3) Place 50 *ml* of N sodium sulphite solution and a few drops of thymolphtalein indicator in a 250 *ml* beaker.
4) Cool both solutions to 12-16°*C*.
5) Immediately add the sodium sulphite solution to the flask containing the resin sample.
6) Titrate within 15 *s* with N sulphuric acid to the disappearance of the blue colour.

Calculation:

The basic chemical reaction is as follows:

$$CH_2O + Na_2SO_3 + H_2O = NaSO_3CH_2OH + NaOH$$

$$\text{Free } CH_2O\ (\%) = 0.03 \times A \times F / (\text{Sample weight}) \times 100,$$

where A = the amount of N sulphuric acid consumed (*ml*); F = factor of N sulphuric acid; S = the weight of sample (*g*).