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Full Length Research Paper

Effect of mixture components on the properties of MUF resin

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The effect of disodium tetraborate decahydrate (DTD) on the properties of melamine-urea-formaldehyde (MUF) resin was studied. The leachability from the MUF resin into solvents was investigated. The solvents chosen in this research were distilled water, 10% ethanol, and 30% ethanol. The main constituents that contribute to the leaching in water are melamine, additional urea, and DTD concentrations. Based on the statistical analysis using Response Surface Methodology, it was found that melamine and DTD should be reduced and additional urea required to get the least leach out in water. For 10% ethanol, the components that contribute to the leaching were melamine, initial urea. Therefore, these should be reduced to get the least leach out in 10% ethanol. The similar trend was also observed in the case of 30% ethanol solvent. On the other hand, addition of DTD resulted in longer resin's shelf-life and better water solubility. However, DTD gave higher resin viscosity and pH.

Key words: Melamine-urea-formaldehyde resin, disodium tetraborate decahydrate, leachability, shelf-life, viscosity.

INTRODUCTION

Resins are manufactured throughout the world for a variety of applications. Adhesives are for variety of applications. Among of these amino resins, the MUF resin is mostly used adhesives for exterior and semi exterior wood panels and for the preparation and bonding of low and high-pressure paper laminates and overlays. Melamine-urea-formaldehyde (MUF) resins are also used for panels that are evaluated in damp conditions. Compared to the price of MF resin, MUF resin is cheaper. MUF resins have been cheapened by addition of greater or lesser amount of urea, are also often used. So, we can state that MUF resins are intermediate between MF and UF resins with durability proportional to MF content (Shields, 1984). With the widespread use of MUF resin and its economical importance, MUF resin is chosen in this research in order to develop the formulation of MUF resin with the combination of the DTD pesticide. In order to preserve plywood or veneer from being attack by termites, fungi and other microorganism, in this research, MUF resin is developed and added with pesticide to produce wood adhesive with the capability of killing wood deteriorating agents. However, resin is non-water soluble and non polar, whilst pesticide is polar, thus, we need to produce water soluble resin that can accept pesticide.

The pesticide used in this research is disodium tetraborate decahydrate (DTD). This type of pesticide was chosen because it is less toxic to human. DTD has been used to control the insect's stomach and thus the insect cannot absorb nutrients and it will starve to death (Kroschwitz and Grant, 1992). Nevertheless, it is important to study the effect of the pesticide on the resin to produce a safe killing pesticide. Residual life of DTD is indefinite if the treated wood is protected from running water.

The aim for this research was to study the application of Response Surface Methodology (RSM) in the determination of the designed experiments to observe the reaction between disodium tetraborate decahydrate (DTD) pesticide and melamine-urea-formaldehyde (MUF) resin.

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Table 1. Constraint of the component proportion

Components, X_i	Constraint (%)	
	Lower limit, L_i	Upper limit , U_i
Melamine	25	30
Formaldehyde	50	60
Initial urea (Urea 1)	4	10
Additional urea (Urea 2)	6	15
DTD pesticide	1	10

MATERIALS AND METHODOLOGY

Experimental Design

The experimental settings are performed by statistical mixture experimental design. The experimental design of five-component system is conducted by using Design Expert (version 6.10, Stat-Easy Inc., Minneapolis, USA). A set of candidate points in the design space is selected using the D-optimal criterion. In D-optimal criterion, there are restrictions on the component proportions that take the form of lower and upper constraint.

The constraint of the component proportion as shown in Table 1 is adapted from the experimental results of a previous study (Awang et al., 2006).

Synthesis of resin

Formalin, melamine, urea (U1) and DTD were charged into the 500 ml flask, while stirring. The pH of the mixture was adjusted with 40% w/w NaOH solution in the range of pH 8.8 - 11 to prevent the polymerization process happening rapidly. The resin temperature was raised slowly for every 5 min until it reached 80°C to optimize the polymerization process.

The mixture was refluxed for an hour until it reached end point. End point was determined by dropping the resin solution into a beaker at the temperature of 40°C. If the droplet solution forms a whitish streak (polymerized), it means that the end point has reached. Whereby, if the droplet solution dissolved in the water without any trace, the end point has yet been reached.

The pH reading for the mixture within the end point period reduced naturally to pH 7.0 or acidic condition. After sufficient refluxing, the solution was added with few drops of 40% w/w NaOH solution to increase the pH range again, between pH 9.0 - 10.0. The mixture was gradually cooled down to 65°C, and then additional urea (U2), which acts as free formaldehyde scavenger, was introduced into the reaction mixture.

The pH of the mixture was kept constant at the pH range of 9.0 - 10.0 by adding 40% w/w NaOH solution during the post-refluxing process. Finally, the mixture was cooled to room temperature or 40°C and then transferred into the storage container for testing.

Concentration of DTD

20 g of MUF-DTD resin was mixed with ammonium chloride (1% of resin's weight, which was 0.2 g). The mixture was mixed in polyethylene (PE) cups. Then the PE cups was placed in oven at 105°C until the resin hardened and solidified.

For leachability testing, resins based on the designed experiments were hardened by ammonium chloride, and then the resin solids will be soaked in 100 mL of distilled water, 10 and 30% ethanol solvents. Eight solid matters were prepared for each experiment because every week, one solid matter was taken out and the solvents were tested until 8 weeks reached. The same was done for the other solvents. The resin solid sample leached out into distilled water and ethanol solvent with time. The solution containing DTD/ethanol was analyzed by using quinalizarine method, the absorbance and concentration of which can be measured spectrophotometrically at the wavelength of 620 nm (A. O. A. C, 1990)

The leaching results were taken every week, until the 8th week. The 8 weeks results were then analyzed.

Determination of degree of acidity

100 g of liquid resin was filled into a 250 ml beaker. The resin temperature was adjusted to 30°C by immersing the beaker into a water bath. The pH meter probe was immersed into the liquid resin until the reading was stable. The reading of the pH meter was taken and analyzed.

Determination of degree of rheological and storage life properties

The viscosity of resin was determined using the Cole-Parmer 98936-15 viscometer. The temperature of 200 g liquid resin was adjusted to 30°C by using a water bath. The viscometer spindle was then submerged into the resin until the resin level reached the minimum mark on the spindle. The viscometer speed was adjusted to 100 r.pm. After the reading was stable, the viscosity of the resin was taken. The resin was stored at ambient room temperature. Viscosity of the resin was measured every 1-2 days and the graph of days vs. viscosity (cP) was plotted. The storage life of the resin was judged based on its viscosity. The storage life (days) was taken when the viscosity of the resin was more than 400 cP. The initial viscosity reading and the storage life were analyzed.

RESULT AND DISCUSSION

MUF resin formulation

Twenty-five MUF resin with DTD pesticide formulations were prepared in a laboratory scale in accordance with the composition percentage as suggested by the mixture design. Table 2 shows the design layout in terms of actual factor values.

Effects of mixture components on the leaching of pesticide

In this study, the DTD pesticide that was added in the resin was leached out into several selected solvents, that is water, 10 and 30% ethanol. The effects of mixture components on the leaching of the pesticide are shown in Figure 1 to 3.

Figure 1 shows the effect of pesticide leaching from resin into water, where the resin is in solid form and water acts as a solvent medium. The main materials that contribute to the leaching of DTD in water are, melamine, additional urea, and DTD. Melamine and DTD should be reduced and additional urea should be increased to get the least DTD leach-out in water as solvent. Additional urea and formaldehyde can also be increased to get the least DTD leach-out. Figure 2 shows the effect of pesti

Table 2. Design layout of the MUF resin with DTD pesticide formulation

Exp.	Melamine (%)	Formalin (%)	Urea 1 (%)	Urea 2 (%)	DTD (%)
1	25.00	50.00	9.00	15.00	1.00
2	30.00	50.00	10.00	9.00	1.00
3	30.00	50.00	4.00	10.50	5.50
4	29.50	59.50	4.00	6.00	1.00
5	25.00	50.00	10.00	10.00	5.00
6	29.50	59.50	4.00	6.00	1.00
7	25.00	50.00	4.00	15.00	6.00
8	27.50	50.00	6.50	6.00	10.00
9	25.00	50.00	4.00	11.00	10.00
10	30.00	50.00	10.00	9.00	1.00
11	30.00	50.00	4.00	15.00	1.00
12	25.00	54.00	10.00	10.00	1.00
13	30.00	50.00	4.00	6.00	10.00
14	30.00	56.00	7.00	6.00	1.00
15	25.00	60.00	8.00	6.00	1.00
16	25.00	60.00	4.00	6.00	5.00
17	27.50	55.50	10.00	6.00	1.00
18	25.00	55.00	4.00	6.00	10.00
19	25.00	60.00	4.00	10.00	1.00
20	26.84	54.29	5.32	9.27	4.27
21	25.00	50.00	10.00	6.00	9.00
22	25.00	55.00	4.00	15.00	1.00
23	25.00	50.00	9.00	15.00	1.00
24	30.00	50.00	4.00	6.00	10.00
25	30.00	50.00	4.00	15.00	1.00

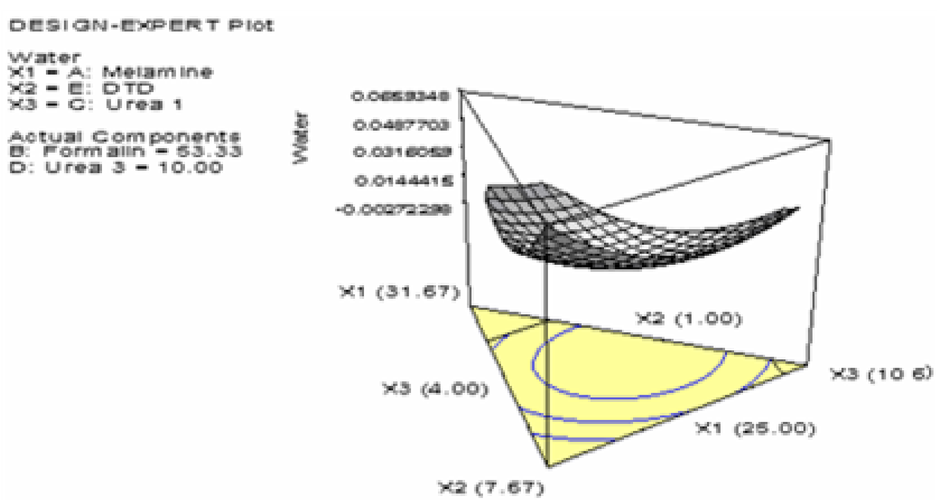

Figure 1. Effect of mixture component on viscosity.

cide leaching from resin into 10% ethanol solvent. The main materials that contribute to the leaching of DTD in 10% ethanol solvent are melamine, initial urea, and DTD. Thus, the content of melamine, initial urea and DTD

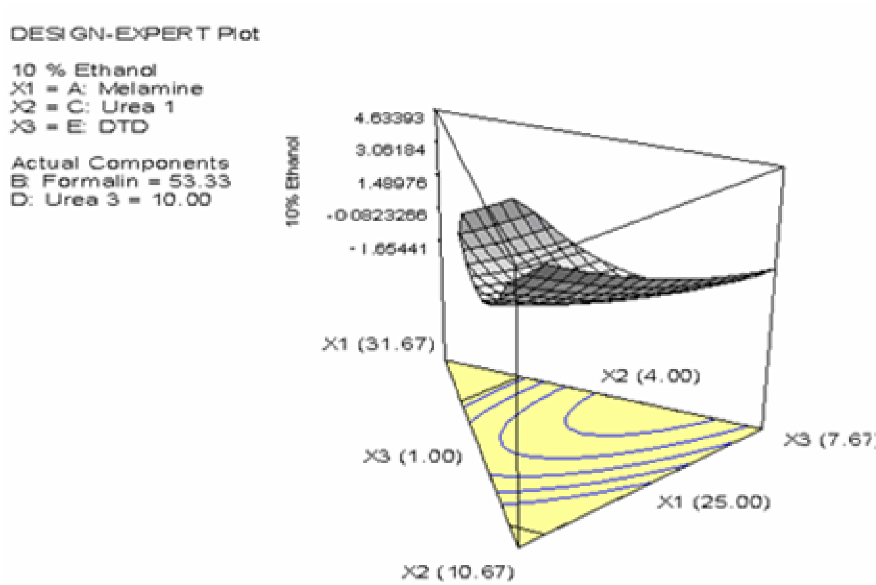

Figure 2. Effect of mixture component on shell life.

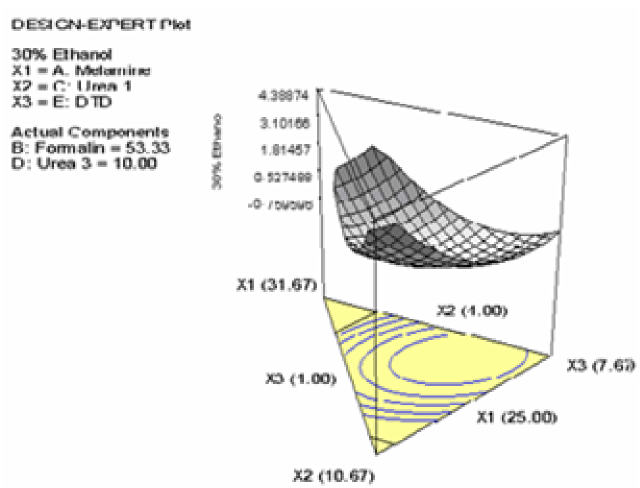

Figure 3. Effect of mixture component on alkalinity.

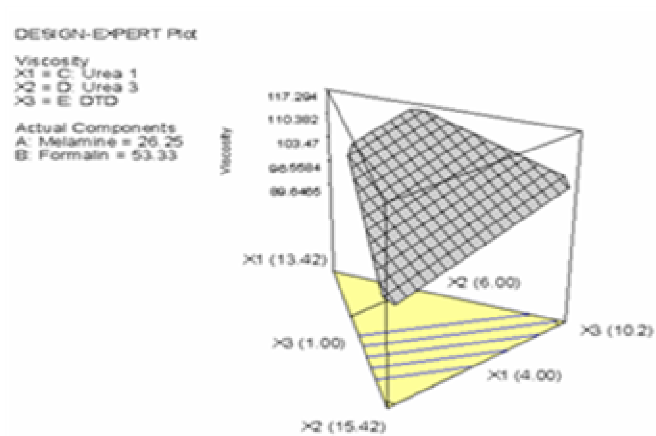

Figure 4. Effect of mixture component on pesticide leaching from resin into water as solvent.

should be reduced to get the least DTD leach-out in 10% ethanol solvent.

Figure 3 shows the effect of pesticide leaching from resin into 30% ethanol solvent. The main materials that contribute to the leaching of DTD in 30% ethanol solvent are melamine, initial urea, and DTD. Therefore, the content of melamine, initial urea and DTD should be reduced to get the least DTD leach-out in 30% ethanol solvent.

Effects of mixture components on the viscosity

Figure 4 shows the three dimensional response surface plot of viscosity. The diagram describes the variation on response as a function of the factor. In order to represent the response evaluation in a bi-dimensional system, two of the variables were to be kept constant. In this case, melamine and formaldehyde composition were kept constant since these components have the less effect on viscosity. The figure shows that, the viscosity reduced with an increase in additional urea and DTD composition. On the other hand, an increase in initial urea enhances the viscosity.

Effects of mixture components on the shelf life

Figure 5 shows the three dimensional response surface plot shelf life. The diagram describes the variation on response as a function of the factor. In this case, melamine

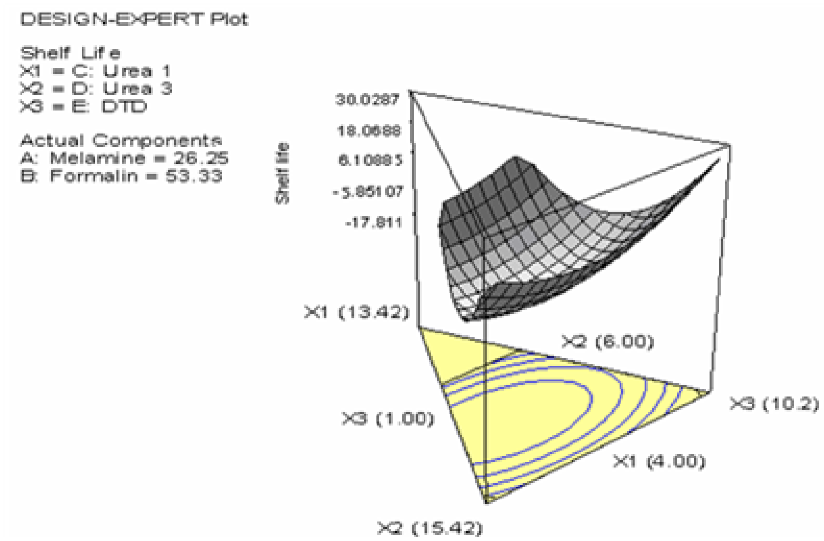

Figure 5. Effect of mixture component on pesticide leaching from resin into 10% ethanol solvent

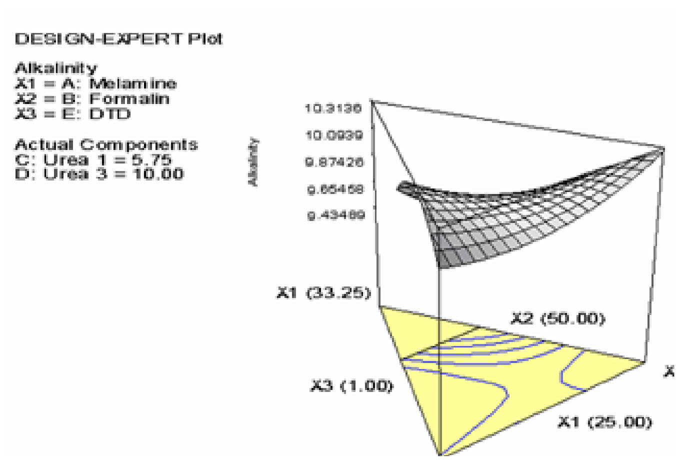

Figure 6. Effect of mixture component on pesticide leaching from resin into 30% ethanol solvent.

and formaldehyde composition were kept constant since these components have the less effect on shelf life. The figure shows that an increase in DTD, initial urea and additional urea reduce and then increase the shelf life of the resin.

Effects of mixture components on the alkalinity (pH)

Figure 6 shows the three dimensional response surface plot pH. The diagram describes the variation on response as a function of the factor. In this case, both initial and additional urea composition were kept constant since these components have the less effect on the pH. It was observed that melamine, formaldehyde, and DTD contribute on the pH of he formulated resin. The figure shows the higher amounts DTD give the higher value of pH. On the other hand, the pH of the resin reduced with the increases in formaldehyde and melamine.

Conclusions

MUF resin formulations have been prepared. The effects of the initial urea, additional urea, formaldehyde, melamine and DTD compositions on the viscosity, alkalinity, shelf life and pesticide leachability of the MUF resin have been studied. The result indicates that the properties of the MUF resin can be manipulated by changing the composition of the ingredient used in the formulation.

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