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Pesticide Based Melamine-Urea-Formaldehye (MUF) Resin with Disodium Tetraborate Decahydrate (DTD)

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Abstract: Amino resin is widely used as binder for various wood products such as plywood, particleboard, fibreboard and decorative laminate. Wood products are liable to be attacked by wood deteriorating agents. In this research, Melamine-Urea-Formaldehyde (MUF) resin was developed with a pesticide DTD) to produce wood adhesive to control pests. The interaction between the MUF resin and the DTD pesticide was found. The results show that MUF resin with and without DTD were chemically and spectrally very similar and were distinguished only by O-H stretch functional group, at 3380 cm^{-1} of the spectra. The absorbance of this functional group increased when the concentration of DTD in MUF resin increased. The difference in the absorbance is a strong indication that the O-H functional group of MUF resin has reacted with borate ions in DTD. It was found that the concentration of DTD leached out during the first 5 weeks linearly increased but after that the amount of DTD leached out was consistent. These results show that, a small amount of DTD can react with MUF resin.

Key words: Binding, MUF resin, DTD pesticide, leaching

INTRODUCTION

Melamine-urea-formaldehyde (MUF) resin formed by condensation polymerisation of formaldehyde, melamine and urea (Emery, 1986) exhibits remarkable stability by symmetrical triazine ring made these products resistant to chemical change once the resin cured to the insoluble, cross linked state (Diem and Matthias, 1976). MUF resin was made cheap by addition of a greater or lesser amount of urea were also often used. MUF resins are intermediates between MF (Malamine Formaldehyde) and UF (Urea formaldehyde) resins with durability proportional to MF content. Notwithstanding their widespread use and economical importance, the literature on MF and MUF resin is only a small fraction that dedicated to urea formaldehyde resin (Shields, 1984).

Amino resin are manufactured throughout the world for a variety of applications especially as wood adhesives. Therefore, the cost of adhesives has an extensive implication on some wood based product such as medium density fibreboard, particleboards and other panel products (Ratnasingan, 2001). Malaysia is one of the major countries exporting tropical wood-based products, therefore it must be ensured that the quality of timber and timber products is maintained (Razak, 2001). However, at present most of the timber and timber products are liable to be attacked by wood deteriorating agents such as borers, termites and wood-rotting fungi.

In this research, MUF resin was developed with a pesticide to produce wood adhesive to kill the wood deteriorating agents. DTD was used as a pesticide in this research and it is less toxic to human. It was used to control insects by interfere with insects' metabolic systems and effectively kill them. This pesticide kills the microorganisms that produce enzymes in the insects' stomach and thus the insects cannot absorb nutrients and it starves to death (Kroschwitz and Grant, 1992). To produce a safe killing pest, it is important to study the leachability of the pesticide from the MUF resin. However, there is no study regarding the determination of leachability and effect of pesticide in resin industry.

MATERIALS ANDMETHODS

Variables and experimental set-up: In this research, MUF resin synthesis was used as reported (Bono *et al.,* 2003; Siambun *et al.*, 2003). This process was divided into three main stages i.e. initial process, polymerisation or refluxing

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process and post-refluxing process. The mole ratio of formaldehyde to urea and melamine in each stage are $F:U_1 = 9$, $F:M_1 = 2.91$ and $F:U_3 = 3.55$.

During the initial process, formalin, melamine and part of urea (U_1) were charged into the flask, while stirring. The pH of the mixture adjusted with NaOH to between 8.5 and 9.0 to prevent the polymerisation process happened rapidly (Pizzi, 1994). The resin temperature was raised slowly for every 5 min until it reached 80°C, to optimise the polymerisation process. During the polymerisation process, the mixture was refluxed for an hour until it reached the end point. End point was determined by dropping the solution into a beaker of water with 40°C temperature. If the droplet solution forms a whitish streak (polymerised), it means that the end point has reached. Whereas if the droplet solution dissolved in the water without any trace, this means that the end point has not yet reached (Pizzi, 1994). The pH reading for the mixture within the end point period reduced naturally to 7.0 or acidic condition. After sufficient refluxing, the solution is added with few drops of NaOH to increase the pH range to between 8.5 and 9.0 again. Additional urea (U_2), which acts as free formaldehyde scavenger, is introduced into the reaction mixture and then cooled to 60°C. The pH of the mixture is to be kept constant at 8.5-9.0 by adding NaOH during the post-refluxing process. Finally, the mixture was cooled down to room temperature and transferred into the storage container, for FTIR and leachability testing.

Determination of DTD effect on MUF resin: For FTIR testing, the synthesised MUF resin was added with different percentage of DTD pesticide (1-3% wt). The MUF resin (with and without pesticide) was then hardened using ammonium chloride. Ammonium chloride widely used as catalyst in forest products industry (Pizzi, 1994). The FTIR spectroscopy was used to determine any reactions or decomposition occurred between the functional groups of MUF resin and DTD after the addition of pesticide. A comparison of the spectra before and after DTD had been added to MUF resin was made. The sample was made into KBr pellet and analyzed with FTIR instrument (Derrick, 1989).

Determination of DTD leachability from MUF resin: For leachability testing, the synthesised MUF resin was added with different percentage of DTD pesticide (1-3% wt) and left aside for a varying period of time (one-four weeks). The sample was leached out with water and time. Then, the solution containing DTD/water was vacuum distilled to recover DTD. The solution containing DTD/water was analysed by using quinalizarine method, the absorbance and concentration were measured spectrophotometrically at 620 nm (Association of Official Analytical Chemist, 1990).

RSULTS AND DISCUSSION

Determination of DTD effect on MUF resin: The strongest bands in the MUF resin are generally the hydroxyl, carbonyl and hydrocarbon stretching frequencies, whereas the strongest band in the DTD spectra Fig. 1 is B-O stretching, near the 1490-1310 cm^{-1} (Rao, 1963). The B-O bond has considerable double-bond character and the positions of the band and depends strongly on the conjugative ability of the constituents due to the importance of resonance form (Werner *et al.*, 1955).

Based on Fig. 1, the spectra of MUF resin with 1 wt% DTD and MUF resin with 3 wt% DTD were chemically and spectrally very similar and distinguished only by stretch at 3380 cm^{-1} of the spectra. This is due to different concentration (1-3 wt %) of DTD was added to MUF resin and thus reflects the changes in absorbance of the spectra. It indicates that, the higher the concentration of DTD in MUF resin, the higher the absorbance of in the range of 3200-3400 cm^{-1} in the spectra. Besides, the difference in absorbance of these spectra is a strong indication that the O-H functional groups of MUF resin reacted with borates ion in DTD. This means that there might be a few path ways for this reaction because MUF resin is the intermediate of MF and UF resin (Pizzi, 1994). This reaction occurs as shown in Fig. 2 and 3.

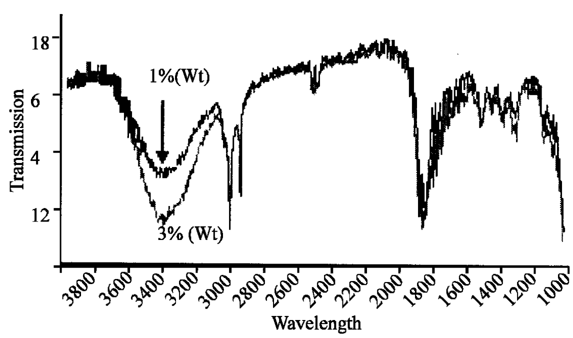

Fig. 1: Comparison of IR spectrum for MUF res with 1% DTD and MUF resinwith 3% DTD

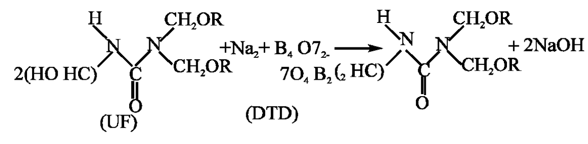

Fig. 2: UF pathway for the reaction between MUF resin and DTD

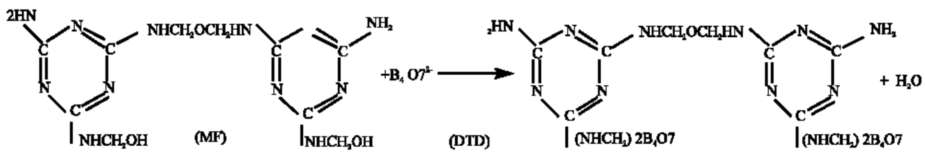

Fig. 3: MF pathway for the reaction between MUF resin and DTD

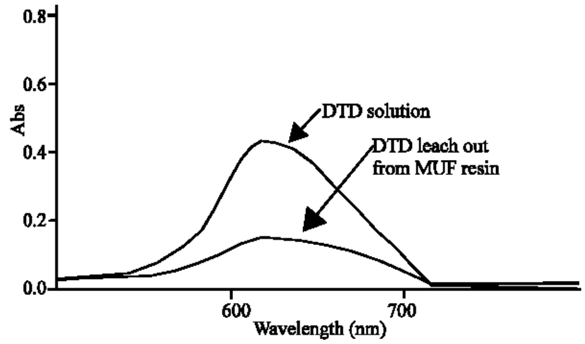

Fig. 4: Wavelength scan for DTD solution and DTD leached out from MUF resin at λ maximum which is 660 nm

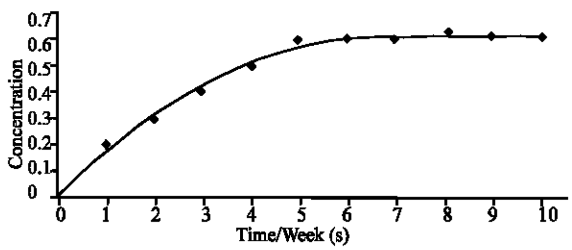

Fig. 5: Leachability of DTD from MUF resin

Determination of DTD leachability from MUF resin: The data of Fig. 4 for the wavelength scan of pesticides were obtained with a single beam spectrophotometer. Based on the diagram, both the DTD and the solution that leached out from MUF resin exhibits the same maximum absorbance value, λ, which is near 660 nm. As shown in Fig. 5, the period of leachability is proportional to the concentration of DTD in wt%. This indicates that, as the sample (MUF resin with DTD) is left aside for a longer period, the concentration of the DTD leached out increased. For instance, on the fifth weeks, the amount of DTD leached out from MUF resin is 0.6 wt% out of 1 wt% of DTD that was added to MUF resin. This means that the percentage of DTD leached out from MUF resin is 60% on the fifth week. Nevertheless, the rest of DTD binds effectively with MUF resin since there is no longer DTD leached out from the MUF resin. In a long term leachability study, Dudas (1981) observed that boron was readily leached, probably as a result of the moderate solubility of borate salts. The binding of DTD in MUF resin can be improved by adding a binder to MUF resin and DTD. The amount of DTD that leached out from MUF resin is consistent after the sixth week. This implies that only micro amount of DTD (0.4% or less) can bind effectively with MUF resin.

CONCLUSION

It was found that there is a reaction between MUF resin and DTD. The reaction occurred between O-H functional group in MUF resin and the borates ion from DTD. The higher the concentration of DTD in MUF resin, the higher the absorbance in the range of 3400-3200 cm^{-1} in the spectra. The reaction between MUF resin and DTD may happen in few pathways. This is due to MUF resin is formed by the intermediate of melamine-formaldehyde (MF) resin and urea-formaldehyde (UF) resin.

The period of leachability was proportional to the concentration of DTD w/w%. The effective amount of DTD that reacts with MUF resin found to be 0.4 w/w% or less because the excess of DTD leached out from MUF resin after a certain period of time if the amount of DTD was too high. Hence, it indicates that only micro amount of DTD can bind effectively with MUF resin.

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