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Reducing formaldehyde emission of urea formaldehyde-bonded particleboard by addition of amines as formaldehyde scavenger

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A B S T R A C T

Particleboard is one of the building materials that contribute to the emittance of formaldehyde in enclosed area. In order to reduce the formaldehyde emission from particleboard, amines were added into the urea formaldehyde (UF) resin as formaldehyde scavenger. The amines used were methylamine, ethylamine and propylamine. 0.5, 0.7 and 1% of each type of amine were added into UF resin and the mixtures were used to produce particleboard from rubberwood particles. The properties of the UF resin after addition of amines such as gelation time, viscosity, pH, free formaldehyde content and thermal stability were evaluated. The physical, mechanical properties and formaldehyde emission of the produced boards were also assessed. The results revealed that fully cured amine-containing UF resin possesses higher thermal stability compared to control UF resin. Amine-containing UF resin also had longer gelation time due to higher pH value. Nevertheless, both physical and mechanical properties of the resultant particleboard were negatively affected. Particleboard made from amine-containing UF resin had higher thickness swelling and water absorption. In addition, lower bending strength and internal bonding strength were also recorded. Insufficient pressing time for fully cured of resin might be the reason for such phenomenon. Particleboard with F*** emission level ($0.5 \leq x \leq 1.5$ mg/L) as specified in Japanese Industrial Standard (JIS) or European's E0 class equivalent were achieved when ethylamine and propylamine were added, regardless of dosage used. This study showed the feasibility of using amines as formaldehyde scavenger. However, optimisation of processing parameters is needed to enhance the physico-mechanical properties of the particleboard.

1. Introduction

As one of the wooden materials for buildings applications, particleboard is classified as reconstructed panels that are mainly used to manufacture furniture as well as for thermal and acoustic insulation [1]. Particleboard is one of the important major timber products in Malaysia. In the year 2017, the total revenue from the exportation of Malaysian major timber products was RM 23.2 billion [2]. Particleboard has contributed 1.88% of the total export value in 2017, which accounted for RM 437 million. The local production line in Asian countries, particularly Malaysia, is continuously influenced by the Japanese trends as Japan is a main and vital for demand of particleboard. Japanese Industrial Standard (JIS) has the most stringent standards in the world where only wood panels with emission level of F**** ($\leq$0.3 mg/L) could be used unrestrictedly within the room, while the F*** ($\leq$0.5 mg/L) and F** ($\leq$1.5 mg/L) panels are only allowed provided that the room is spacious and have good ventilation [3]. According to Athanassiadou and Ohlmeyer [4], the respective emission level of F****, F*** and F** are more or less equivalent to European standard's SE0, E0 and E1.

Sick House Syndrome, a term derived from Sick Building System that was first recognised in the year of 1983 by World Health Organization as a medical condition, has been reported in residential houses and educational facilities throughout the world. The occupants experience various symptoms such as headache, nose and throat irritation and fatigue [5]. Formaldehyde, acetaldehyde, acetone and 2-ethyl-1-hexanol are the main indoor pollutants that were detected in buildings and are closely related to the occurrence of mucosal symptoms among users [6]. The formaldehyde levels present in indoor air are highly dependent on the formaldehyde sources, temperature, humidity and air exchange rate in the building. The main sources of indoor formaldehyde emission in the residential houses and educational facilities nowadays include wood floor finishes, wood-based products such as plywood, particleboard and medium density fiberboard, wallpaper and paints as well as cigarette smoke [7].

Urea formaldehyde is a major aminoplastic resins used for the

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fabrication of interior wood-based products due to its low cost and high reactivity [8]. A study by He et al. [9] revealed that urea formaldehyde (UF) resin is the main source that contributes to the formaldehyde emitted from wood-based panels. Urea and formaldehyde are highly reactive and could react rapidly to form a strong bond. Nevertheless, the reaction is reversible and therefore provides potential for long-term formaldehyde release [10]. Formaldehyde emits from formaldehyde-based adhesive bonded particleboard is mainly caused by the existence of unreacted free formaldehyde in the board. However, this type of release lasts only for a short period of time after manufacture. Another release mechanism that could continue throughout the entire working life of the board is through the hydrolysis of the aminoplastic bond when exposed to elevated temperature and relative humidity [11].

In the past decades, great effort in reducing formaldehyde emission from particleboard such as lowering the formaldehyde to urea (F/U) molar ratio in UF resin has been made. However, lowering F/U ratio inevitably lower the UF reactivity and subsequently, reduced the properties of the resulted panels [12]. In addition, lowering of F/U ratio has reached its limit when Maminski et al. [13] reported that the strength of joints made with UF resin with F/U ratio of 0.85 is 20% lower than the resin with an F/U ratio of about 1.1. To make matters worse, no significant reduction of formaldehyde emission was recorded. An additional amount of 15–20% of resin is needed in order to fulfill the performance standards. Although lowering F/U ratio is the most direct and economic method, other methods known to reduce formaldehyde emission including incorporation of formaldehyde catcher or scavenger, optimisation of processing parameters, and coating with nanoparticles modified water based varnish have also been adopted by several researchers [14–17]. Recently, a study by Jiang et al. [18] has proved that particleboard thermally treated at mild temperature (50 or 60 °C) displayed significant reduction in formaldehyde emission. Ayrilmis et al. [19] incorporated microfibrillated cellulose (MFC) into different grades of urea formaldehyde (UF) resins (SE0, E0 and E1) and the formaldehyde emission of produced laminated veneer lumber (LVL) were determined. The results revealed that the modification by MFC only showed significant effect on SE0 grade UF resin in terms of formaldehyde emission reduction, while E0 and E1 grade UF resin did not indicate the same observation.

Various amine-based compounds such as urea, ammonia, melamine, dicyandiamide, and polyamides have been incorporated into formaldehyde-based resin to reduce its formaldehyde emission [20]. Nevertheless, studies on the addition of primary alkyl amines as formaldehyde scavenger are very limited. A study by Boran et al. [21] reported on the effectiveness of adding different amine compounds in the reduction of formaldehyde emission of medium density fiberboard bonded with urea formaldehyde (UF) resin. Another study by Ghani et al. [22] revealed that the addition of 1% propylamine into UF resin could reduce the formaldehyde emission of the particleboard from 0.7 mg/L to around 0.3 mg/L. Nevertheless, physical and mechanical properties of the produced particleboard were adversely affected. This study aims to produce UF-bonded particleboard with lower formaldehyde emission using three primary alkyl amines, namely methylamine, ethylamine and propylamine. The effects of incorporating different amines and dosages on the properties of urea formaldehyde resin were investigated. In addition, the mechanical, physical properties and formaldehyde emission of the resultant particleboard were also evaluated.

2. Materials and methods

2.1. Preparation of materials

Rubberwood particles were obtained from a local particleboard plant, HeveaBoard Berhad, which is located in Gemas. The binding agents used in this study, urea formaldehyde (UF) resin type E1, was supplied by Aica Chemicals (M) Sdn. Bhd from Senawang. Three different types of amines, namely, methylamine, ethylamine and propylamine which were used as formaldehyde scavenger om this study were purchased from Evergreen Engineering & Resources. The hardener used in this study was ammonium sulphate and wax was applied as water repellent.

2.2. Resin properties after addition of amines

Several properties such as acidity (pH), viscosity of the resin, gelatin time and free formaldehyde content were tested in the UF resin after addition of different dosage of amines. The Mi105 pH/temperature professional portable meter was calibrated with buffer solutions at pH 4 and 10 before testing begins. The resin/amine mixtures were cooled to 30 °C. Following that, the pH meter electrode was immersed into the mixtures and pH reading was recorded. For viscosity measurements, 75 ml UF resin was poured into a 100 ml beaker. The viscosity of the mixture was measured with an AMETEK Brookfield rotational viscometer & rheometer at 20 °C with a spinning rate of 1 rpm. As for pH determination, mixtures of UF resin and amines were poured into a beaker and stirred well. Then, 6.5 g of the mixture was poured into a test tube which was immersed (below water line) in a 100 °C water bath. Immediately, the content was stirred continuously and the time (in seconds) required for resin mixtures to cure was recorded.

For free formaldehyde content determination, 10 g UF resin was weighed and poured into a 250 ml Eerlenmyer flask and 50 ml of dimethyl sulphoxide solution was added. Rapidly, within 5 s whilst stirring, 30 ml of 0.1 M HCl and Na_2SO_3 were added. Next, to ensure complete reaction of the formaldehyde with sulphite, the mixture solution was cooled in ice cubes for 3 min. Then, 1 ml 0.1% thymolphthalein solution was added. The excess acid was immediately titrated with 0.1 M NaOH solution until it changed to blue color. Volume of the 0.1 M NaOH used was recorded as V_1. The blank test under the same condition but without the UF resin was also carried out and the volume of 0.1 M NaOH used was recorded as V_2. The free formaldehyde content was calculated using Equation (1).

$$\text{Free formaldehyde (\%)} = ((V_1 - V_2) \text{ x } M \text{ x } 3.002) / W \quad [1]$$

where; V_1 = volume of 0.1 M NaOH solution for resin, mlV_2 = volume of 0.1 M NaOH solution for blank, mlM = molarity of NaOH solutionW = weight in grams for resin, g.

The experiment was repeated for the UF resin admixed with different dosage of amines. Two replicate measurements for each sample were made.

2.3. Fourier transform infrared (FT-IR) spectroscopy analysis

A FT-IR spectrometer was used to determine any differences occurring to the functional group on pure formaldehyde sample and after the formaldehyde was mixed with different amounts of amine compounds. FT-IR spectra tests were run at ambient temperature using pure samples within the wave number range of 4000 to 400 cm^{-1} and at a resolution of 4 cm^{-1}. The infrared spectra of the samples were measured on a Perkin-Elmer FT-IR (model spectrum 100 series, USA).

2.4. Thermal stability of UF resin and amine-containing UF resins

Samples of cured control UF resin and amine-containing (methylamine, ethylamine and propylamine) UF resins were tested for thermogravimetric analysis (TGA) using Thermal Gravimetric Analyzer, TA Instrument Q500 model. About 8 mg samples were placed in alumina crucible. An empty alumina crucible was used as reference. All the samples were heated from ambient temperature to 600 °C in a 50 mL min $^{-1}$ flow of nitrogen at 10 °C min $^{-1}$ heating rate.