

Shear Strength of Wood to Wood Adhesive Based on Palm Kernel Oil

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Received 8 June 2005; accepted 22 August 2005

DOI 10.1002/app.23015

Published online 23 January 2006 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: A polyurethane adhesive system was prepared by reacting a resin consisting of palm kernel oil-based polyester and dimethyl cyclohexanediamine with an aliphatic adduct based on 2,4-diphenylmethane diisocyanate (MDI). Brushing technique was used for applying the adhesive (of thickness 0.05–0.10 mm) onto the wood substrate. Shear strength test for substrates that have been exposed to moisture and various degrees of heat was carried out. Collected data indicated that the adhesive exposed to heat at 70°C has the highest shear strength. At this point, the shear force was at the maximum of 2562 N with strength of 2.65

MPa. However, at higher testing temperature, there is a decrease in the shear force and strength of the adhesive. The presence of moisture, however, does not affect much on the shear strength. The morphological observations via optical microscope were made to explain the relationship of heat and moisture with the shear strength of the adhesive. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 100: 1759–1764, 2006

Key words: shear strength; RBD palm kernel oil-based polyamide; polyurethane adhesive

INTRODUCTION

The selection of wood panel as a substrate is due to its importance as a structural material. It consists of cellulosic structure with natural porosity. This contributes to its hygroscopic property. Woody materials face expansion or shrinkage problems when exposed to changing environment. As such, the type of adhesive used on this kind of substrate must have higher strength than the wood itself. This is required for stronger adhesion and to avoid defects.¹ Wood is used widely in the construction field, especially for housing development. Application of adhesive such as the polyurethane (PU) can be used to glue the wood tiles or parquets to the cement-type of flooring. The utilization of Plaster de Paris as an adhesive is not suitable, as it is easily broken when pressure is applied on the surface.² It is also widely being used in the furniture industry for wood joints.

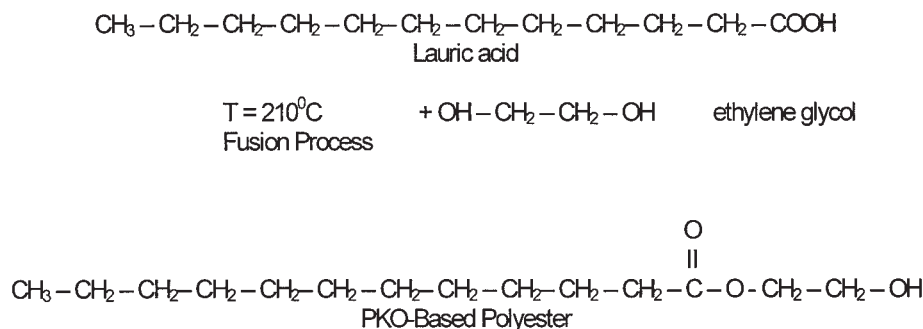
Adhesion is a phenomenon that allows two substrates to be bonded together. Only internal force or thermal movement could separate two substrates from the bonding force.³ The performance or the behavior of a wood adhesive system is dependent on a

wide range of variables, such as surface smoothness of wood substrate, presence of wood extractives, pH, and amount of debris present, which are related to the environment, such as the level or the rate of change in both temperature and relative humidity.⁴ The bonding mechanism of adhesive is due to the complex chemistry of the cellulosic substrate, such as the hydrogen bonding in some adhesives and weak van der Waal forces with others.⁵ Sometimes, mechanical interlocking could also be one of the factors interfering in the bonding properties.

The major adhesives used for bonding wood are adhesives based on urea formaldehyde and phenol formaldehyde; but, some of these adhesives are sensitive to hydrolysis and stress scission. They also produce health hazards, because of the formaldehyde they release. Polyacrylate-type adhesives are also used as wood adhesives, but it has some disadvantages such as shorter pot life, higher cost, and limited durability. To overcome such problems, scientists are trying to develop new polymeric adhesives. Sandip et al.⁴ discovered that the shear strength of the potato starch/natural oil-based PU adhesive performed better than the commercial adhesives for wood joints. Lower NCO/OH ratios (below 1.3) showed better strength because of the formation of a more highly crosslinked structure. This factor was also confirmed by Somani,⁶ when castor oil-based PU is used. Other factors affecting shear strength of the adhesive are types of isocyanate adducts (aromatic isocyanate performed better than aliphatic isocyanate), chain length

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Contract grant sponsor: Universiti Kebangsaan Malaysia; contract grant number: ST-0014–2003.



Scheme 1 Synthesis of PKO-based polyester.

of glycols (polyether polyol gave poor performance compared with polyester polyol), concentration of aromatic and aliphatic groups, type of solvent used in casting the films, and crystallinity of the polyols.

Extensive research work on synthesis of palm kernel oil (PKO)-based PU is reported in literature,^{7,8} but not much is said about its usage as an adhesive. Hence, this effort to synthesize PKO-based polyol is utilized in the PU adhesives. Somani⁶ found that PU adhesive synthesized from castor oil (extract of *Ricinus communis* L.) has good properties in terms of electrical insulation and hydrolytic stability. It is contributed by the presence of hydroxyl groups. PKO has a reactive site, the carboxyl group linking the acid to the glycerol. The present work describes the development of PU adhesives by reacting the PKO-based polyester with an aliphatic isocyanate adduct. The PU adhesive was characterized by FTIR spectroscopy. These PU adhesives were applied onto the wood substrates. Lap shear strength was measured using a standard method, and was determined upon exposure to various conditions, i.e., heat and moisture.

EXPERIMENTAL

Materials

Palm kernel oil (PKO) (Lee Oil Mills, Klang, Malaysia) is used without further purification. Ethylene glycol and dimethyl cyclohexanediamine (DMCHA) (Merck) were used as received. Aliphatic isocyanate adduct 2,4-diphenylmethane diisocyanate is obtained from Cosmopolyurethane (Port Klang, Malaysia), with NCO content of 31%, specific gravity of 1.24, and viscosity of 115 cps.

Preparation of PKO-based polyester

A dry process (fusion process) was employed for the synthesis of the polyester, in which a kettle equipped with a mechanical stirrer and a provision for nitrogen flushing was charged with PKO and ethylene glycol (1 : 1 molar ratio). The temperature was raised slowly

with constant stirring, and was maintained at 210°C for 2 h. The water formed during the reaction was removed from the top of condenser by vacuum evaporation. The acid values were measured periodically. A gradual decrease in the acid value showed the progress of reaction. The reaction was finally stopped at an acid value near to zero. Finally, the PKO-based polyester was cooled to room temperature. The reaction sequence is shown in Scheme I. The properties of the polyester such as moisture content, viscosity, light absorption, and color are determined.

Polyurethane adhesive preparation

A two-pack polyurethane (PU) adhesive system is prepared by mixing PKO-based polyester resin (base component) with isocyanate adduct (hardener component). The two components along with DMCHA of 0.10% (w/w) of total composition were mixed at the time of application on the tested substrates.

Preparation of wood specimen

Wood strips (conforming to ASTM D906-82 (1987) requirements) with size of 25 × 300 × 3 mm³ were polished with sandpaper No. 10 (250 μm) to remove surface imperfections. Wood dust was removed by wiping with a linen rag. Panels were dried to remove traces of moisture, allowed to return to room temperature, and then brushed (using a 30-mm wide brush) with the adhesive system without delay to a thickness of 0.05–0.10 mm. Two brushed strips were placed together by placing a load of 1 kg on the joint for 24 h to allow for complete drying.⁴ Before and after the application of adhesive, both strips were measured for thickness, using the Mitutoyo thickness gauge. The difference in thickness was taken as the thickness of the adhesive. Acceptable specimens should be the specimens with adhesive thickness ranging from 0.05 to 0.10 mm. They were then conditioned at room temperature (30°C) and (50 ± 5)% relative humidity for 24 h, before the shear test was carried out on an

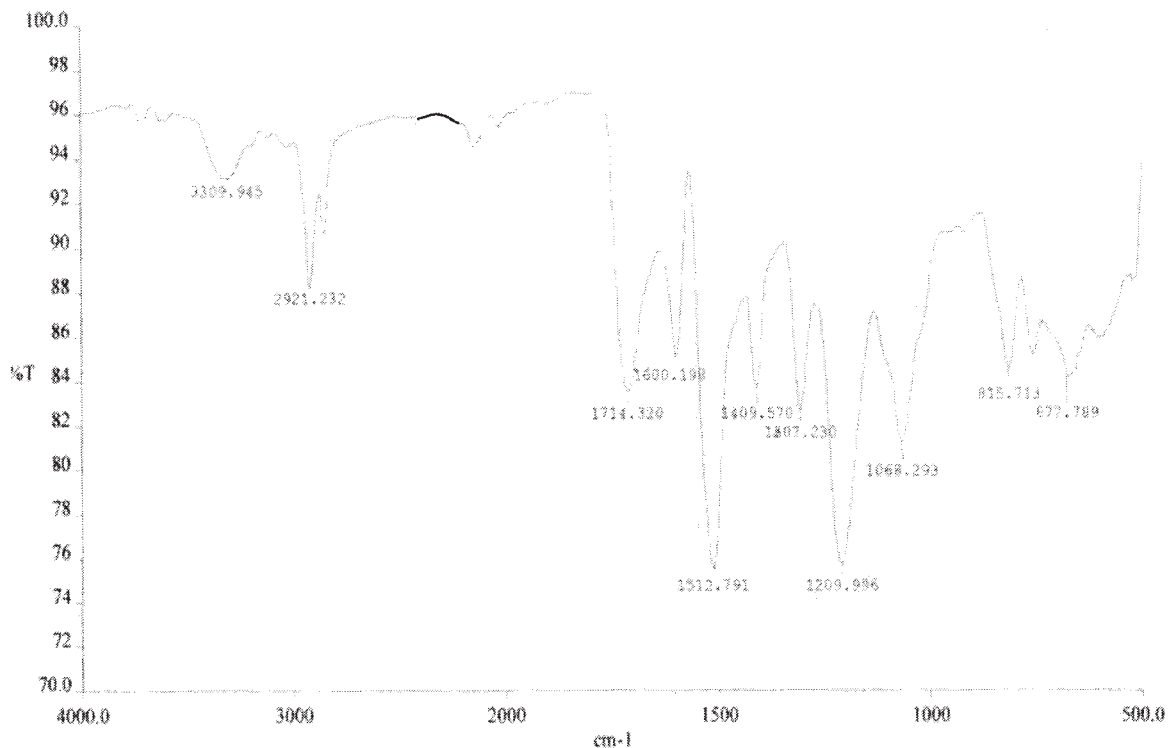


Figure 1 FTIR spectrum of the PU adhesive.

Instron Universal Test Machine model 5567 as per ASTM D906–82 (1987).⁹

Spectroscopy analysis

Fourier transform infrared (FTIR) spectroscopy analysis of the PU adhesive was carried out using Perkin-Elmer model BX with diamond attenuation total reflectance (DATR) method at wavelength of 4000–700 cm^{-1} . Identification of functional groups was carried out.

Green strength

The specimens were placed in an oven at four different temperatures: 70, 80, 90, and 100°C for 24 h. The observations at room temperature and under the influence of moisture were also included for comparison. The specimen was also tested for its dimensional stability in observing the shrinkage and expansion

problems. The changes in the specimen's thickness were measured.

RESULTS AND DISCUSSION

Physical properties of the polyester

The polyester is a golden yellow liquid with 0.02% moisture content and viscosity of 246 cps. The light absorption determined using spectrophotometer model Genysis gives 96.5% transmittance at 650 nm wavelength.

Infrared spectroscopy

The expected structure of PU was confirmed by FTIR spectroscopy. Figure 1 shows a characteristic carbonyl stretching at 1714 cm^{-1} , indicating the presence of a urethane linkage. The absorption peaks resulting from the —NH stretching and bending vibrations were ob-

TABLE I
The Shear Force and Shear Strength of the Adhesive at Various Conditions

Parameter	Control	Humid	–15°C	70°C	80°C	90°C	100°C
Max. shear force (N)	1879	1857	2237	2652	2456	2456	1335
Shear strength at break (MPa)	1.58	1.76	2.15	2.25	2.36	2.38	1.23
Shear strength (MPa)	1.88	1.86	2.24	2.65	2.46	2.46	1.34

Both control and humid conditions are at room temperature.

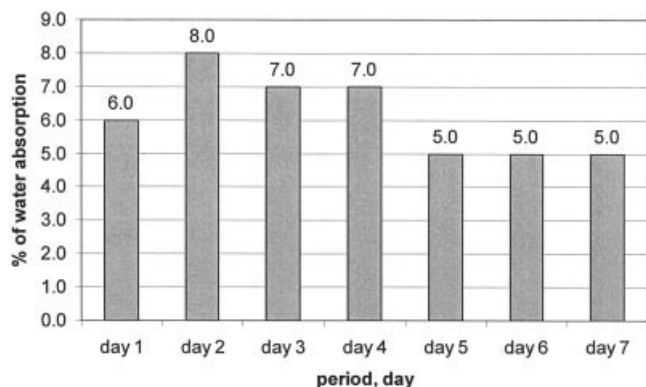


Figure 2 Average percentage of water absorption on a day-to-day basis.

served at 3309 and 1512 cm^{-1} , respectively. The OH stretching band at 3400 cm^{-1} indicated the presence of free hydroxyl groups. The band observed at 2921 cm^{-1} is due to the —CH stretching. The formation of PU is clearly indicated due to the absence of an —OH absorbance band associated with the polyester and the presence of a —NH band of PU.

Gel time

The gel time or pot life is important for an adhesive system in its application. Gel time is the maximum length of time the system remains in sufficiently fluid condition to be applied to a substrate. The gel time is predetermined before application on the substrate by the exposure to air at 30°C. The gel time obtained is 110 s, a much sufficient time for brushing application.

Green strength: effect of testing conditions (temperature and humidity) on the shear strength of the PU adhesive

The shear force and the strength of the specimens were determined, and the results are shown in Table I. It is observed that both the shear force and the strength increase with increasing temperature up to 70°C. The shear force and strength decreased at testing temperature of 80°C to almost half the value at 100°C. Higher temperature increased the strength because of the curing effect caused by the heat. The heat absorbed increased the kinetic of the reaction to an extent. However, too much heat resulted in the hydrolysis of the PU to amines, carbon dioxide, carbon monoxide, etc.,² deteriorating the adhesive strength, as observed at 100°C exposure. This resulted in an unstable adhesive system because of the changes in the cellular structure and the chemical structure.¹⁰

Table I also shows the shear stress at break for all specimens. This is the point where the specimens encountered the splitting phenomenon without breaking

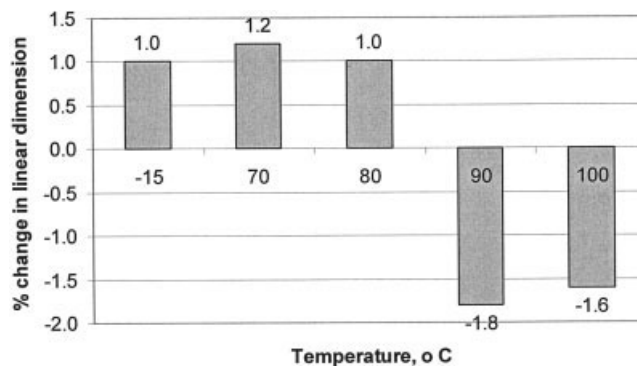


Figure 3 Average percentage of dimensional changes of the PU adhesive.

the wood strips. A consistent increment in the shear stress at break was observed at increasing temperature. The maximum reading of 2.38 MPa was observed at 90°C, and the lowest at 100°C with a reading of 1.23 MPa, almost half the maximum value. At higher temperature of 100°C, the adhesive encountered changes in its cellular structure because of the hydrolysis process. The cell walls ruptured, deteriorating the shear strength. Furthermore, extra heat provided to the adhesive system produced a brittle surface of adhesive on the substrates. As such, an occurrence of weak points was observed.

On the other hand, moisture did not affect much the shear strength of the adhesive when comparing the specimens that have been immersed in water for 24 h with the control specimen. The adhesion between both wood strips was so strong that the interface areas were not affected by the presence of water. Since this type of PU adhesive is not water-soluble, there is less tendency of hydrolysis to occur. However, because of the hygroscopic nature of the wood strips, an increment in the percentage of water absorption was observed on day 2 (8%) of the testing period, as shown in Figure 2.



Figure 4 The micrograph of the tested specimens at the temperature of 70°C.

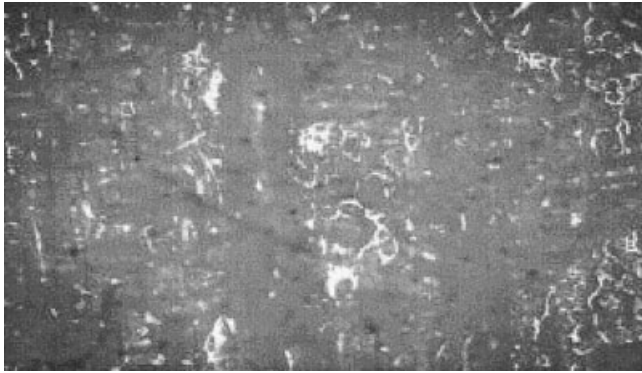


Figure 5 The micrograph of the tested specimens at the temperature of 80°C.

Immersion period had little effect on the water absorption of the adhesive (thickness change of about 0.03 mm). Since the water vapor produced during hydrolysis is a function of the surrounding humidity, the highest percentage of water absorption it can reach was 5%.¹⁰

Dimensional stability test involved a 24-h conditioning at temperature of -15 , 70, 80, 90, and 100°C. The results are as plotted in Figure 3. An expansion as high as 1.2% was observed when specimens were put at -15 , 70, and 80°C. However, at 90 and 100°C, a reduction in the thickness was observed. During hydrolysis, some volatile compounds are produced and caused a reduction in the mass of the adhesive. The decrease in the mass resulted in a reduction in the thickness of the whole specimens. Moreover, the cellular networks in the PU adhesive contain carbon dioxide, which at higher temperature, diffused out easily. This is due to the low pressure inside the adhesive system, compared with that in the surrounding.

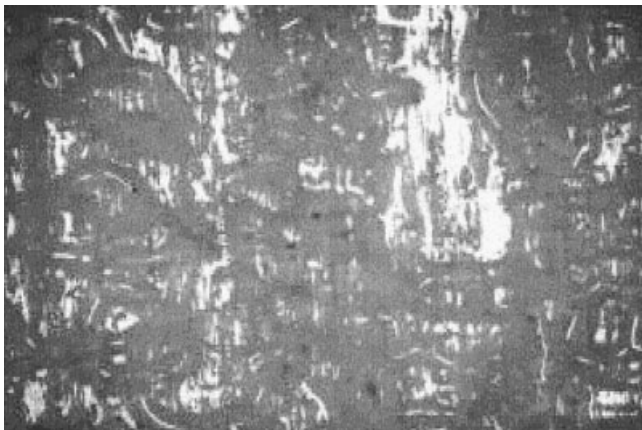


Figure 6 The micrograph of the tested specimens at the temperature of 90°C.

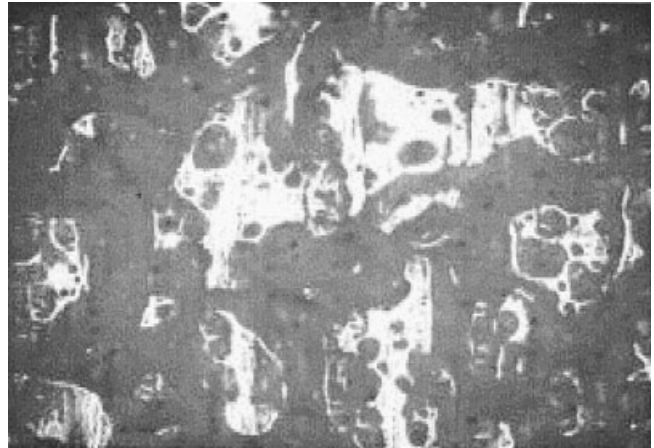


Figure 7 The micrograph of the tested specimens at temperature of 100°C.

Morphological analysis

The surface morphology for all the tested specimens was analyzed using the optical microscope with magnification of 50 \times . This is shown in Figures 4–10. The micrograph shows obvious differences from one specimen to another, and this further explained the findings on the shear strength of the specimens under various conditions. Figure 4 shows that the adhesive on the strips were still adhered to the substrate, while in Figure 5, some broken pieces (whitish spot) have been observed. At higher temperature of 90 and 100°C (Figs. 6 and 7), the broken pieces are becoming obvious. More cells were being ruptured because of the additional heat provided. At this point, hydrolysis of the PU adhesives might have already started. This comparison is supported by the shear force and strength result in Table I.

Figure 8 shows that, at low temperature (-15 °C), the cell has no tendency to be destroyed. In fact, the micrograph is identical to the control specimen (Fig.



Figure 8 The micrograph of the tested specimens at the temperature of -15 °C.

10). Comparing both specimens at room temperature and at -15°C with those at 70°C (Fig. 4), more craters appeared in the former than in the latter. The presence of the craters could be due to the premature curing time. Both the specimens at -15°C and the control system have not yet reached the ideal curing time (small reaction rate) that it created the weak points (craters). Since curing time is much related to the heat provided to the system, higher temperature showed better shear strength to an extent. These explained why the specimen at 70°C had a much higher shear strength, compared with the control specimen.

Micrograph of the specimen that has been immersed in water shows a lot of weak points (craters), compared with the control specimen. The craters (whitish spots) are well distributed. Arnoldus¹¹ mentioned that the presence of moisture in the adhesive system would reduce the adhesion between substrates because of the disturbance in the interfacial between the substrates and the adhesive. Since the PU adhesive system used no defoamer, it is suspected that the adhesive might have formed larger cellular structure (foaming). As a result, the cells absorbed water and increased the rate of hydrolysis process.

CONCLUSIONS

Urethane resins with good adhesion properties can also be synthesized successfully from palm oil-based polyester. Use of palm oil for synthesizing urethane adhesive may reduce the cost without compromising on quality. Adhesion quality, however, depends much on the adhesion between substrates, which at present is being studied extensively, especially in the area of



Figure 9 The micrograph of the tested specimens immersed in water at room temperature.



Figure 10 The micrograph of control specimen at room temperature.

formulation. PU additives, such as catalyst, play a main role in determining the kinetic of the reaction, which affect the curing time of the system. Optimum level minimized the usage, but maximized the gelling time, during brushing application. Initial findings on the correlation study of water absorption and heat stability of the urethane further justify the correct processing parameter and suitability to environmental conditions.

The authors thank the School of Chemical Sciences and Food Technology, Faculty of Science and Technology for the provided facilities.

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