

Damage Mode and Failure Mechanism of Starch-based Aqueous Polymer Isocyanate Plywood Bonded Structure

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Aging means that a polymer material's performance gradually deteriorates, with the loss of use value due to the comprehensive effect of internal and external factors. In this work, a starch-based aqueous polymer isocyanate (API) adhesive joint structure of crosslinking in bonding interface was investigated. The compression shear strength was recognized as a key evaluation index, and the hygrothermal aging experiment tests were accelerated to study the damage mode and failure mechanism of the glue joint structure. The results showed that the adhesive properties of fracture were ductile fracture, and with an increase of aging time, the damage mode of the bonding was transformed from a cohesion damage mode to a cohesion damage with interface damage mode. In the early stages of the aging tests, the effect of temperature on the compression shear strength was most important; however, with the increase of aging time, the effect of humidity became most important.

Keywords: Bonding; Accelerated aging; Fracture; Damage mode

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INTRODUCTION

With recent improvements in standards of living, the demand for environmentally friendly wood-based panels has grown. The aging resistance of starch-based formaldehyde-free wood adhesives directly affects the life of such products. Research on the effects of aging-resistant performance factors to improve the durability of man-made boards has important theoretical significance and practical application value. However, research on modification of wood adhesives should focus on improving aging resistance and increasing the adhesive product's service life. Artificial board adhesives are high polymer materials and are therefore affected by environmental factors such as light, oxygen, temperature, chemical, and biological active media, as well as material factors such as chemical composition, phase structure, molecular structure, and functional group (Bierwagen *et al.* 2000). These can cause a change in the material's surface, the physical and chemical properties, or the mechanical properties, resulting in a loss of performance and failure of the material (Bullions *et al.* 2003). To make highly reliable products, it is necessary to study the environmental conditions, which usually takes years and is difficult to achieve. Therefore, accelerated aging tests are particularly important.

EXPERIMENTAL

The adhesive used for laminated wood cementation was a starch-based aqueous polymer isocyanate adhesive system, which is composed of a starch-based API main agent

and polymerized isocyanate as the cross-linking agent, provided by Jilin Chenlong Biomass Materials Co. Ltd, according to the Japanese industrial standard (JIS K6806).

The birch specimen used in this experiment had a moisture content of 8 to 10% and dimensions of 30 mm × 25 mm × 10 mm (Fig. 1). The adhesive surface was smooth, and the fiber direction and the specimen axis were parallel. Specimens were taken from 3 different blocks. From each block, 3 specimens were prepared to compile a set.

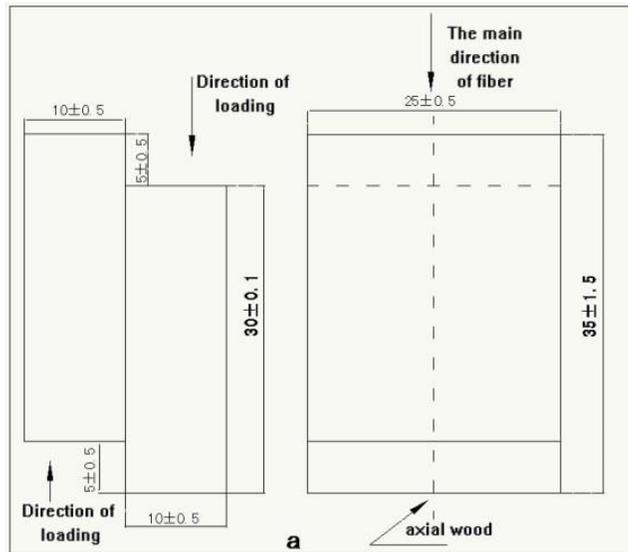


Fig. 1. Shape and dimensions of test block for compression and tension shear strength

Preparation of Specimens

After weighing the test piece, the two pieces were assembled to make them one specimen. The main agent and cross-linking agent were mixed evenly in proper proportion, applied to the adhesive surface of the two specimens, and gelatinized to $125 \pm 25 \text{ g/m}^2$. The two specimens were laminated in the same fiber direction, and the storage time did not exceed 5 min. The pressure (1.0 to 1.5 MPa) was released after 24 h at 20 to 25 °C; then, the sample was placed under the same temperature for 72 h. Twelve specimens were created, and the length and width of the adhesive surfaces were measured.

Aging tests

The specimens were put into the hygrothermal aging machine (Fig. 2). The relative humidity and temperature were controlled by the computer system. The relative temperature and humidity were respectively set to 85 °C and 95%; 75 °C and 95%; 85 °C and 85%; and 75 °C and 85%.



Fig. 2. The hygrothermal aging machine

Compression and Shear Strength Test Specimens

The following method was used to test specimen shear strength. The test data are reported as the average value of 3 groups of specimens (each group has 10 specimens).

1. The specimens were placed indoors at a temperature of 23 ± 2 °C and relative humidity of $50 \pm 5\%$ for 48 h.
2. The specimens were then placed in boiling water for 3h. Afterwards, they were immediately cooled for 10 min in room-temperature water.
3. For the compression shear strength test, the shear plane of the specimens was placed parallel with the load shaft and the maximum load was recorded.
4. The compression shear strength of the specimens was calculated using the following formula:

$$\sigma = \frac{p}{L_a \times L_b} \quad (1)$$

In the formula, δ is the compression shear strength (MPa), p is the maximum damage load (N), L_a is the length of the specimen adhesive (mm), and L_b is the width of the specimen adhesive (mm) (Chun *et al.* 2008).

Four groups of compression shear specimens were put in a box at constant temperature and relative humidity. The temperature and humidity were respectively set to 85 °C and 95%; 75 °C and 95%; 85 °C and 85%; and 75 °C and 85%. Accelerated aging tests were performed under these four conditions, and the compression shear strength was tested at different accelerated aging times. Origin Pro 7.5 software was used to fit the compression shear data.

Scanning Electron Microscope Analysis

A JEOL scanning electron microscope (SEM; Japan) was used to observe the compression shear fracture after aging treatment. Low-magnification microscopic observations were used to determine the failure mode of the glue joint structure as well as the change in failure mode. High-magnification microscopic observations were used to determine the fracture properties of the bonding structure, the failure mode, and the fracture properties of the cementing structure to study the failure mechanism.

RESULTS AND DISCUSSION

Analysis of Shear Strength of Compression

The relationship between the compressive shear strength and aging time with the same humidity and different temperatures is shown in Fig. 3. The figure shows that under the same humidity, the compression shear strength decreased more with higher temperatures. With increasing aging time, the compression shear strength maintained a fairly constant value, and the values at high temperatures were less than those at low temperatures.

The relationship between compression shear strength and aging time at the same temperature but different humidities is shown in Fig. 4. Under the same temperature, the decrease in compression shear strength was more obvious with higher humidities. With increasing aging time, the compression shear strength maintained a fairly constant value, and the values at high humidities were less than those at low humidities.

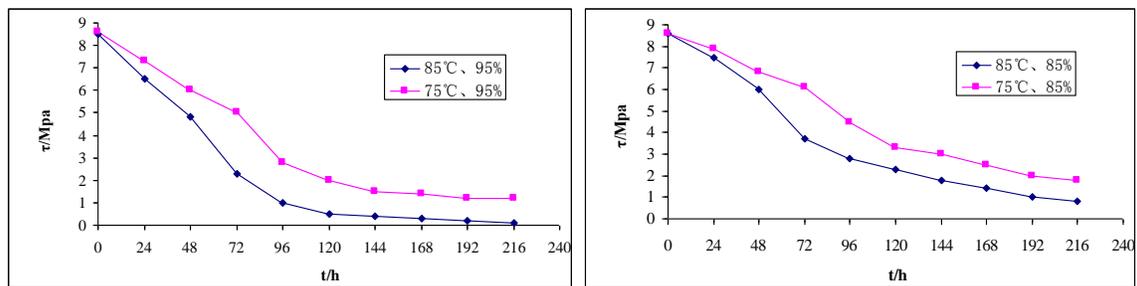


Fig. 3. Relationship between compression shear strength and aging time with the same RH and different temperatures

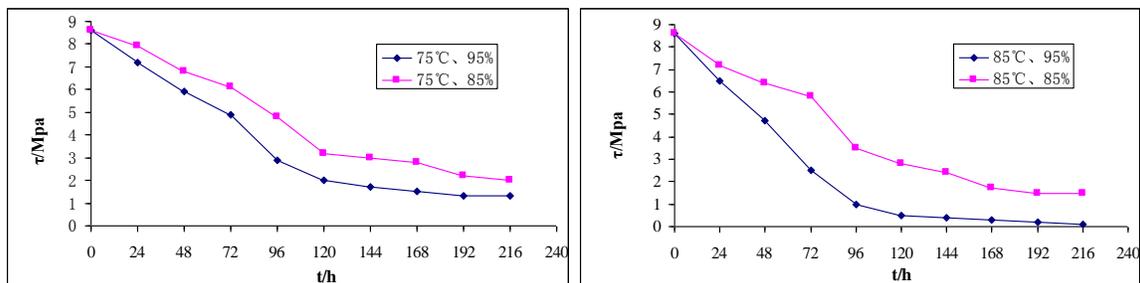


Fig. 4. Relationship between compression shear strength and aging time at the same temperature and different humidities

Analysis of Fracture Failure Mode of Bonding Structure

Generally speaking (Junyou *et al.* 2007), there are three types of failure mode of bonding destruction: cohesion damage, interface destruction, and mixed damage. Cohesion damage is divided into cohesive failure of the adhesive and glued cohesion damage. In the former, the adhesive layer itself is destroyed, and in the latter, sticking failure occurs. Interface destruction is a kind of failure where the adhesive and adherend are completely removed at the interface. Mixed damage is also called alternate damage. It is partly cohesion damage and partly interface damage; specifically, the destruction alternately occurs in the adhesive interface.

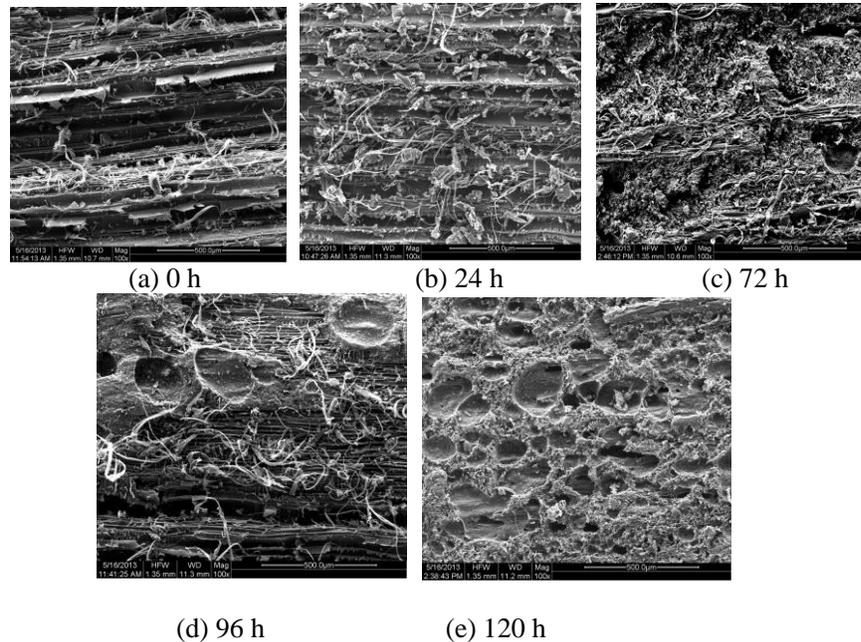


Fig. 5. Micro-appearance of the fracture surfaces of compression shear specimens after different aging times

Figure 5 shows cross-sections of shear specimens after different aging times under different conditions of compression. It is clear that the compression shear damage mode changed from cohesion damage to interface damage. With increasing aging time, the area of the interface damage increased.

The damage mode was the same for all combinations of relative humidity and temperature, and the damage increased with increasing aging time for all specimens. From the macroscopic and microscopic observation, the compression shear damage mode changed from cohesion damage to interface damage, and the interface fracture area increased with increasing aging time.

Bonding Structure Fracture Analysis

For the analysis of the structural adhesive fracture properties, specimens under the four humidity/temperature conditions were magnified 500 times using the SEM to observe the bonding structure fracture. The morphology is shown in Fig. 6. As can be seen from the image, the fracture showed torn edge characteristics, namely ductile fracture. With short aging times, temperature played a dominant role; however, the humidity played a dominant role with longer aging times.

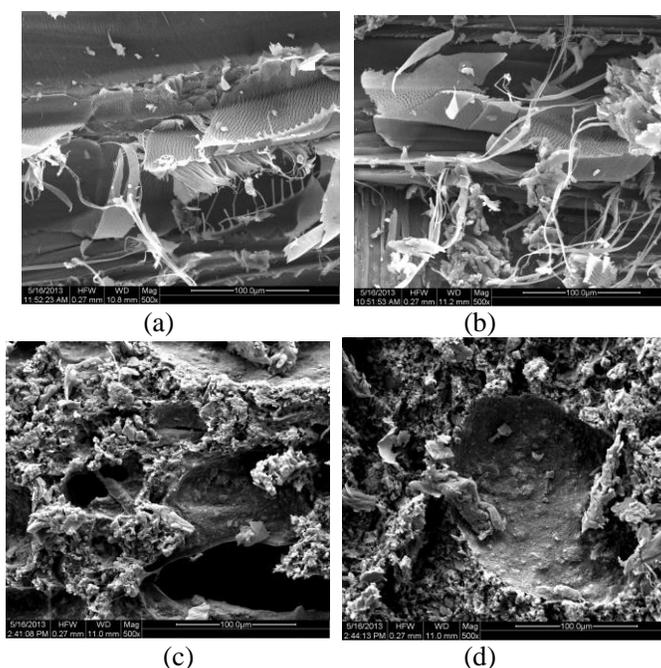


Fig. 6. Micro-appearance of the fracture surface of compression shear specimens

Adhesive Failure Mechanism Analysis

Figures 3 and 4 show the compression shear strength tests under the four conditions of accelerated aging. With respect to conditions of 75 °C and 95% humidity and 85 °C and 85% humidity, the compression shear strength values of the latter initially declined at a faster rate than those of the former. With increasing aging time, the rate of decline reversed.

The reason for the reversal in the rate of decline is that at the beginning of the cementing structure aging test, temperature played the primary role in the compression shear strength (Wang *et al.* 2009). This is because as the adhesive is heated, physical and chemical changes occur after physical deformation under the action of an external force. The chemical changes are primarily characterized by thermal decomposition, and oxidation cracking will occur. With increasing aging time, the effect of humidity on the properties of the bonding structure is greater because of the effect of water on the bonding structure. On the one hand, a large number of water molecules along the surface of the hydrophilic glued object quickly permeate the entire adhesive interface, replacing the adhesive molecules in the original wood surface, causing the cementing strength to decline dramatically (Ying *et al.* 2011). On the other hand, water can penetrate in almost all polymers. Water molecules can damage the hydrogen bonding and other secondary bonding of the polymer molecules, resulting in a plasticizing effect on the polymer, which can decrease the mechanical and other physical properties (Zanni-Deffarges and Shanahan 1995). Water can also cause the polymer chemical bond to fracture, leading to the chemical degradation of the polymer.

CONCLUSIONS

1. The connection structure of adhesive fracture was observed by scanning electron microscopy as a function of increasing aging time. The failure mode of the four aging conditions was found to change from cohesive failure mode to cohesive failure and interface failure. The interfacial failure area also increased.

2. In the fracture morphology observation at high magnification, it is found that in the adhesive part of the bonding structure, the characteristic fracture of the four aging conditions was tearing. Therefore, the fracture properties of the bonding structure included toughness fractures.
3. By observing the compression shear strength when alternately varying the temperature and humidity, it was discovered that the decrease in the compression shear strength was more obvious at higher temperatures and humidities.
4. The compression shear strength degradation rate and fracture microscopic image observation results shows that temperature played the dominant role in the early stages of aging, and humidity played the primary role later on.

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