Performance of *Cunninghamia lanceolata / Uncaria* Composite Particleboard

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Particleboards were prepared with thinning wood of Cunninghamia lanceolata (Chinese fir) and wood shavings of waste Uncaria sinensis branches by using melamine-urea-formaldehyde resin as the adhesive. Influences of particle mass ratio of C. lanceolata to Uncaria, adhesive loading, and target density on internal bonding strength (IB), modulus of rupture (MOR), modulus of elasticity (MOE), thickness swelling rate of water absorption (TS), and water absorption (WA) of the composite particleboard were investigated. Results showed that: (1) IB, MOR, MOE, 2h-TS, and 2h-WA of pure C. lanceolata particleboard met service requirements of type-P2 furniture particleboard of GB/T 4897-2015. (2) With the increase of Uncaria particles, IB of composite particleboard decreased gradually, while MOR, MOE, TS, and WA first increased and then decreased. The best comprehensive performances were achieved when the mass ratio of C. lanceolata to Uncaria was 75:25, manifested by 1.5 MPa of IB, 21.3 MPa of MOR, 2140 MPa of MOE, 2.4% of 2h-TS and 12.3% of 2h-WA. These performances basically meet the service requirements on type-P3 of the standard. (3) It can make C. lanceolata fir/Uncaria particleboard meet requirements on type-P4 heavy-load particleboard of the standard by increasing target density and adhesive loading appropriately. (4) The C. lanceolata/Uncaria composite particleboard showed higher thermostability and fire resistance.

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INTRODUCTION

China is a country with relatively sparse forest resources. The 9th national forest resource survey data showed that the forest area in China is 220 million ha, the forest coverage is 22.96%, and the forest stock volume is 17.56 billion m³. The forest area per capita is only 1/4 of the global average level per capita, and the forest stock volume per capita is only 1/7 of the global average level per capita (Kauppi *et al.* 2018; Cho *et al.* 2023; Zeng and Yang 2023; Yong *et al.* 2024). Wood and woodware have been highly appreciated by people. These materials, which range from toothpicks to furniture and indoor decorations, are closely related with human life.

Wood has become an indispensable material in national economic development. In 1990s, the natural large-diameter materials decreased continuously, and the national policy of "natural forest protection project" was implemented. These intensified the imbalance between the increasing demands for high-quality wood and the shortage of high-quality forest resources. Raw material shortage has been a challenge against artificial board industrial development in China (Yang *et al.* 2018; Jin 2022; Yan *et al.* 2022; Wang *et al.* 2024). Making full use of low-quality timber, small-sized timber, artificial fast growing wood, and crop leftovers (*e.g.*, reed, corn stalk, bamboo, and straw), especially increasing the comprehensive utilization of three kinds of forest leftovers and small sub-fuelwood, is an effective measure to solve wood supply-demand imbalance (Lykidis *et al.* 2014, 2016; Zhang *et al.* 2019; Gendvilas *et al.* 2021; Zafeiroudis *et al.* 2024).

Cunninghamia lanceolata (Chinese fir) is a unique fast-growing commercial tree species in China. With characteristics of fast growth, high yield, and excellent properties, C. lanceolata is a kind of good raw material for architecture, furniture, papermaking, and other industries (Wang et al. 2019; Zhong et al. 2024; Li et al. 2023, 2024). Several intermediate cuttings are needed before the final felling of high-quality C. lanceolata forest, thus causing abundant thinning wood. The thinning wood accounts for about 72% of total timber production throughout the cultivation of C. lanceolata (Deng et al. 2009; Zhou et al. 2015; Chen et al. 2023; Liu et al. 2024). Carbonizing treatment is a simple and feasible way to increase utility value of thinning wood of C. lanceolata, which has been extensively applied (Tian et al. 2021; Wu et al. 2021a). Urea resin filling in thinning wood of C. lanceolata can increase physical properties of thinning wood effectively (Dunky 1998; Xin et al. 2010; Wang et al. 2012). Flame retardant treatment with dicyandiamide, phosphoric acid, and boride is employed to thinning wood of C. lanceolata, and the bonding strength of the C. lanceolata laminated board is improved by hot-pressing technology to meet the national standards (Ye et al. 2018; Wu et al. 2021b). These studies have laid some theoretical foundation for high-efficiency use of thinning wood of C. lanceolata.

Nevertheless, the above research methods have very limited utilization of the tremendous quantities of the available thinning wood of *C. lanceolata*. The low utilization can be attributed to poor physical and mechanical properties, loose structure, low strength and hardness, and unstable size. The production raw materials of particleboard have extensive adaptability, so it is a feasible and effective way to prepare particleboard with thinning wood of *C. lanceolata*. *Uncaria rhynchophylla* (Miq.) Miq. ex Havil is a kind of non-wood biomass material and has extensive distributions in China. It will produce abundant branches during harvest of *Uncaria*. According to statistics, it will produce at least 10 kg of wastes (*e.g.* branches and leaves) per 1kg *Uncaria*, resulting in tremendous production (Geng *et al.* 2019; Guo *et al.* 2019).

Based on the above analysis, particleboards were prepared using thinning wood of *C. lanceolata* and *Uncaria* branches in this study. Key attention was paid to investigate influences of mass ratio of particle of *C. lanceolata* to *Uncaria*, adhesive loading and target density on internal bonding strength (IB), modulus of rupture (MOR), modulus of elasticity (MOE), thickness swelling rate of water absorption (TS), and water absorption (WA) of particleboard. The objective for this study was to improve comprehensive utilization of three kinds of forest leftovers and small sub-fuelwood.

EXPERIMENTAL

Materials

Melamine-urea-formaldehyde (MUF) resin with viscosity of 103.2 mPa·s and solid content of 55% was prepared in the laboratory (Li *et al.* 2023). *C. lanceolata* shavings and *Uncaria* branches shavings with moisture content of 5% and the size of length (18 to 24) mm × width (0.3 to 0.5) mm × thickness (4 to 6) mm were provided by Qian Dong Nan Institute of Forestry of Guizhou Province.

Preparation of Particleboard and Testing

Before the preparation of particleboard, 1.5% curing agent $(NH_4)_2SO_4$ was added to MUF resin and thoroughly stirred. The particles were put into the mixer to stir at a speed of 500 r/min, and then the high-pressure spraying device sprayed the modulated MUF at a rate of 200 to 600 mL/s. After spraying, the stirring and mixing of the particles were continued for 30 min. Then a single particleboard (200 mm × 200 mm × 10 mm) was prepared under a hot–pressing temperature 140 °C and hot-pressing pressure 33 kg /cm². The adhesive loading was 10% on bone-dry wood particles. The panels were pressed with a three-stage hot pressing cycle (2 min-4 min-2 min). Then the particleboards with the target densities of 0.5, 0.6, 0.7, and 0.8 g/cm³ were obtained.

Testing and Characterization

After surface sanding, the prepared particleboard was measured with reference to GB/T 4897-2015 for internal bonding strength (IB), modulus of rupture (MOR), modulus of elasticity (MOE), 2 h and 24 h water absorption (WA), and thickness swelling in the course of water absorption (TS). The average value of 10 specimens was used as the final data. The data were processed in Excel and Origin software and were expressed as the mean \pm standard deviation. A one-way analysis of variance was applied to determine significant differences (P < 0.05). The Vertical Density Profile (VDP) of the board was tested by their thickness scan at a rate of 0.4 mm/s using a DAX 6000 profile densitometer (GreCon, Alfeld, Germany). The section of the board was observed using a Hitachi S–3400N scanning electron microscope (Tokyo, Japan). Thermogravimetric analysis of resin was conducted using a TG 209 F3, NETZSCH, Germany, with the test conditions of N₂ protection, temperature range of 30 to 600 °C, and the heating rate of 10 °C/min.

RESULTS AND DISCUSSION

Internal Bonding Strength Analysis

Internal bonding strength (IB) is an important index to evaluate performances of particleboards. The value of IB reflects the bonding quality among wood shavings. If IB value is too low, one can expect cracking and layering of boards. The influencing relationship between mass ratio of particles of *C. lanceolata* to *Uncaria* and IB of particleboard is shown in Fig. 1. With the increase of mass ratio of *Uncaria* particle, the IB value decreased gradually. Effects of mass ratio of particles of *C. lanceolata* to *Uncaria* on IB presented significant differences. When the *Uncaria* particle content increased from 0% to 75%, the IB value decreased by 41.5% from 1.64 MPa to 0.96 MPa. Thus, increasing *Uncaria* particle content was disadvantageous for the IB of particleboards. However, all

particleboards had relatively high internal bonding strength. The lowest IB value (0.89 MPa when the *Uncaria* particle content is 100%) was far higher than service requirements of national standard GB/T 4897-2015 for furniture particleboard under dry state (type-P2, \geq 0.28 MPa), load-bearing particleboard under dry state (type-P3, \geq 0.45 MPa), and heavy-load particleboard under dry state (type-P4, \geq 0.60 MPa).

The internal bonding strength of particleboards is determined by bonding strength among wood shavings, while bonding strength is also influenced by properties of raw materials. Raw material extractive content is one of the important factors that influence bonding performances of particleboards. It might influence curing of adhesives, thus resulting in the poor internal bonding strength of particleboards. Nemli and Aydin (2007) found that pinecone contains a lot of benzenol extract, which contains plenty of resin, thus weakening bonding performances of particleboards. *Uncaria* is an important medicinal plant. Major chemical components of *Uncaria* branches include alkaloids, triterpenes, and flavonoids (Geng *et al.* 2019; Guo *et al.* 2019), which influence curing speed and curing quality of MUF resin adhesive. This also can explain why pure wood shavings of waste *Uncaria* branches exhibited the lowest internal bonding strength.

Additionally, the flowing, diffusion, and penetration behaviors of water absorption and adhesive between particles of *C. lanceolata* and *Uncaria* on their surfaces may be different. It is easy to make adhesive penetrate excessively toward pores of one material, resulting in adhesive shortage and decreasing the bonding strength. However, the uneven diffusion of vapor during hot-pressing treatment also may decrease crosslinking degree of adhesives, thus causing failure of bonding interface. This also will decrease the IB value.



Fig.1. Effects of mass ratio of particle of *C. lanceolata* to *Uncaria* on internal bonding strength of particleboards

When the mass ratio of particle of *C. lanceolata* to *Uncaria* was 75:25, the relationship of adhesive loading and density with IB of particleboards are shown in Fig. 2. When the adhesive loading was 11%, the IB of particleboards increased from 0.84 to 2.15 MPa as density increased from 0.50 to 0.80 g/cm³. Given a low density, the superficial

structure was loose, and there were large pores and low crosslinking strength among wood shavings. All specimen failures occurred on the surface. With the gradual increase of density, the failure positions of specimens became transferred to the center. This is because during preparation of particleboards, the pressing volume differs due to the uneven spreading and uneven heating of surface layer and core layer. The stress decreases gradually from surface layer to internal layers of particleboards, and a large density gradient is formed within the thickness profile. The surface layer of particleboards undertakes the maximum stress, and the density is the highest. In contrast, the core layer bears the minimum stress. Moreover, particleboard thickness recovers after the removal of stress due to plasticity of the material, resulting in the minimum density of the core layer (Xi et al, 2019). Hence, IB is decided by the weakest part of the particleboards. The density of the core layer can affect internal bonding strength of particleboards greatly, and the density gradient of core layer decides IB of particleboards directly. Due to low porosity among wood shavings, the compactness increases by increasing density of particleboards, while density gradient of core layer decreases, thus increasing internal bonding strength accordingly. When the adhesive loading was 13%, the IB of particleboards was further increased by 14.3%, 20.8%, and 14.0% at the densities of 0.5, 0.6, and 0.7 g/cm³, while IB decreased by 17.8% at the density of 0.8 g/cm³. This indicated that within the density of 0.5 to 0.7 g/cm³, increasing adhesive loading can increase adhesive coverage and make bonding of wood shavings tighter, thus further increasing bonding strength. Given a high density, increasing adhesive loading is an easy way to form a dense pre-curing layer on the particleboard surface, thus lowering the IB value.

Thus, density and adhesive loading have comprehensive influences on IB of particleboards. Appropriate sizing pressure and adhesive loading can make adhesive form uniform coverage on the wood shaving surface, which is crucial to improve mechanical performances of particleboards.



Fig. 2. Effects of density and adhesive loading on internal bonding strength of particleboards

Modulus of Rupture Analysis

The modulus of rupture (MOR) is an important index to determine mechanical strength of particleboards. Boards with high MOR are difficult to be broken and deformed. The relationships between mass ratio of particle of *C. lanceolata* to *Uncaria* and MOR of particleboards are shown in Fig. 3.



Fig. 3. Effects of mass ratio of particle of C. lanceolata to Uncaria on MOR of particleboards

With the increase of *Uncaria* particle content, the IB value first increased and then decreased gradually. The MOR of particleboards reached the maximum (21.3 MPa) when *Uncaria* particle content was 25%. As *Uncaria* particle content increases from 25% to 100%, MOR of particleboards decreased gradually. The lowest MOR (15.6 MPa at 100% of *Uncaria* particle) still exceeded the regulated value of national standard GB/T 4897-2015 for furniture particleboard under dry state (type-P2, \geq 11.0 MPa) and load-bearing particleboard under dry state (type-P3, \geq 15 MPa). According to theory of mechanics of materials, the upper surface and the lower surface of the board are the compression faces under stresses, and both bear the maximum loads. With the increase of *Uncaria* particle content, the maximum density of surface layer declined and MOR presented a decreasing trend accordingly.



Fig. 4. Stress-strain curves in anti-bending test

Figure 4 shows that during the MOR test, the bearing capacity of specimens increased first to the maximum and then the boards broke. However, boards still were able to bear some loads. The bearing loads presented a "step-wise" reduction with respect to time rather than drop sharply to the bottom. The failure process showed "unsteady-state" expansion. The failure process of specimens is divided into three stages, namely, linear elastic deformation stage, nonlinear deformation stage and ductile fracture stage (Zhang *et al.* 2022). This is because layers among wood shavings crosslink together after hot pressing and they also break upon compressive failure of the boards, showing ductile fracture failures.

When the mass ratio of particle of *C. lanceolata* to *Uncaria* was 75:25, the relationships of adhesive loading and density with MOR of particleboards are shown in Fig. 5. Upon bending stress-induced deformation of particleboards, failures often are initiated from pores. Decreasing pores increases the stress bearing area accordingly, thus increasing MOR. Therefore, compactness of particleboards increases with the increase of density and there is more material per unit volume to bear compressive stresses. In Fig. 5, the MOR of particleboards increased from 9.5 to 31.9 MPa as density increased from 0.50 to 0.80 g/cm³ when the adhesive loading was 11%. The MOR of particleboards further increased by 26.3%, 14.2%, 8.5% and 12.5% at the density of 0.5, 0.6, 0.7 and 0.80 g/cm³, respectively. To sum up, the impact trends of density and adhesive loading on MOR of particleboards were basically consistent with that of IB. Appropriate sizing pressure and adhesive loading are crucial to improve MOR of particleboards.



Fig. 5. Effects of density and adhesive loading on MOR of particleboards

Modulus of Elasticity Analysis

The modulus of elasticity (MOE) is also an important index to measure performances of particleboards. A higher MOE indicates the greater stress for boards to develop elastic deformation, the higher rigidity and hardness of the board. Particleboards with insufficient rigidity will undergo great deformation during service, which also may affect their services. Hence, particleboards must meet some requirements of rigidity. The relationship between mass ratio of particle of *C. lanceolata* to *Uncaria* and MOE of particleboards is shown in Fig. 6.





With the increase of *Uncaria* particle content, MOE increased and then declined gradually. It reached the maximum (2142.9 MPa) when *Uncaria* particle content was 25%. As *Uncaria* particle content further increased from 25% to 100%, MOE of particleboards

decreased. Only particleboards prepared with 0%, 25% and 50% of *Uncaria* particle content met the regulations of MOE in national standards GB/T 4897-2015 for load-bearing particleboards under dry state (type-P3, \geq 1800 MPa).

Effects of adhesive loading and density on MOE of particleboards when mass ratio of particle of *C. lanceolata* to *Uncaria* is 75:25 are shown in Fig. 7. Clearly, when the adhesive loading was 11%, MOE of particleboards increased from 934.0 to 2906.8 MPa when the density increased from 0.50g/cm³ to 0.80 g/cm³. When the adhesive loading was 13%, MOE of particleboards increased by 4.4%, 19.8%, 10.8%, and 4.9% to 974.7, 1445.2, 2375.4 and 3048.3 MPa at the densities of 0.5, 0.6, 0.7, and 0.8 g/cm³, respectively. Generally speaking, MOE of particleboards was positively related with MOR, and influencing factors of MOR showed similar impacts on MOE.



Fig. 7. Effects of density and adhesive loading on MOE of particleboards

Water Absorption and Thickness Swelling Analysis

Effects of mass ratio of particle of *C. lanceolata* to *Uncaria* on TS and WA of particleboards are shown in Fig. 8. With the increase of *Uncaria* particle content, TS value decreased and then increased. All particleboards showed 2h-TS<8%, thus meeting the regulations of national standard GB/T 4897-2015 for furniture particleboards under dry state (type-P2, 2h-TS≤8%). All particleboards showed 24h-TS<9%, meeting the regulations of national standard GB/T 4897-2015 for load-bearing particleboards under dry state (type-P3, 24h-TS≤19%) and heavy-load particleboards under dry state (type-P4, 24h-TS≤16%). The variation trends of WA and TS were consistent. This is because MUF resin fully reacts with hydrophilic groups after it penetrates cell walls of wood shavings, thus decreasing the number of hydrophilic groups and providing effective water resistance to boards. When the *Uncaria* particle content was 25%, 2h-TS and 24h-TS reached the lowest values, indicating that the adhesive was able to disperse fully among *C. lanceolata* and *Uncaria* particles. If *Uncaria* particle content is increased, the *Uncaria* particle extracts might influence curing speed and curing quality of MUF resin adhesive, resulting in poor bonding strength of particles and thereby increasing water absorption.

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Fig. 8. Effects of mass ratio of C. lanceolata to Uncaria on TS and WA of particleboards

Effects of adhesive loading and density on TS and WA of particleboards when mass ratio of particle of *C. lanceolata* to *Uncaria* is 75:25 are shown in Fig. 9. Based on above analysis, bonding points decreased to some extent and porosity was relatively high when the density was relatively low, although internal stress was low, resulting in the strong water absorption.



Fig. 9. Effects of density and adhesive loading on TS and WA of particleboards

With the increase of density, there were few pores among particles and compactness increased, thus strengthening water resistance. However, compression rate was too high when the density is relatively high and the locking internal stress at hot pressing is also high. Stresses are released through working after being immersed in water and water absorption increases accordingly. With certain adhesive loading, the coverage of adhesive increases and bonding of wood shavings is tighter, thus decreasing water absorption. However, given a high density, increasing adhesive loading is easy to form a dense precuring layer on particleboard surface, thus increasing water absorption of particleboards.

VDP and SEM Analyses

Section of particleboard with a density of 0.70 g/cm^3 , an adhesive loading of 11%, and mass ratios (100:0 and 75:25) of particle of *C. lanceolata* to *Uncaria* were chosen for VDP and SEM tests. Results are shown in Figs. 10 and 11.



Fig.10. Profile density curves of particleboards



Fig. 11. SEM images of particleboard sections. Note: (a) and (a') were mass ratio of particle of *C. lanceolata* to *Uncaria* 100:0, (b) and (b') were mass ratio of particle of *C. lanceolata* to *Uncaria* 75:25.

When the mass ratio of particle of *C. lanceolata* to *Uncaria* was 100:0, the cross sections of particleboard were relatively rough, accompanied with pore compactness and obvious pores. These features might lead to strong water absorption. The particleboards with a mass ratio of 75:25 showed denser sections. The particleboards with a mass ratio of 75:25 showed higher uniformity of profile density. The profile density distribution curve was relatively stable, which is consistent with the high mechanical strength.

Thermal Performance Analysis

Specimen sections with mass ratios of 100:0 and 75:25 were chosen for DTG tests when density was 0.70 g/cm³ and the adhesive loading was 11%. The results are shown in Fig. 12. Pyrolysis of particleboards generally can be divided into four stages, as follows: *Stage 1: 30 to 130* °C. This is a stage of heat absorption and the pyrolysis is very slow. Water evaporation takes the dominant role and chemical composition of timbers hardly changes. *Stage 2: 130 to 300* °C. This is also a stage of heat absorption. The pyrolysis is relatively obvious, mainly manifested by decomposition of timber hemicellulose into CO₂ and CO. *Stage 3: 300 to 550* °C. Pyrolysis is strong in this stage, which releases abundant heats. The mass loss rate of pure *C. lanceolata* board and *C. lanceolata /Uncaria* particleboard were 75.2% and 71.0%, respectively. *Stage 4: 550 to 800* °C. In this stage, the mass tended to be stable and to change rather gradually with the increase of temperature.





Fig. 12. Thermogravimetry diagram of particleboards

The maximum pyrolysis peaks of pure *C. lanceolata* board and *C. lanceolata* /*Uncaria* particleboard were 351.6 and 340.7 °C, respectively, indicating that wood shavings of waste *Uncaria* branches lowered the pyrolysis temperature of particleboard and facilitate generation of combustible substances under a low ignition temperature. At the end of pyrolysis, residual weights of pure *C. lanceolata* board and *C. lanceolata* /*Uncaria* particleboard were 21.5% and 25.35%, indicating that the introduction of *Uncaria* made the particleboard generate more char, thereby providing less loss of mass at higher temperatures. This is because extracts in *Uncaria* can serve as flame retardants. They facilitate dehydration and carbonization and accelerate the charring process of timbers. Other specific flame retardant components will be analyzed deeply in the future.

CONCLUSIONS

- 1. Particleboards were prepared with thinning wood of *Cunninghamia lanceolata* (Chinese fir) and wood shavings of waste *Uncaria sinensis* branches by using melamine-urea-formaldehyde resin as the adhesive. The mechanical properties and water resistance of pure *C. lanceolata* board met the regulation of national standard GB/T 4897-2015 for type-P2 furniture particleboards under the dry state.
- 2. The particleboards achieved the optimal comprehensive performances when the mass ratio of particle of *C. lanceolata* to *Uncaria* was 75:25. They basically met service requirements of national standard GB/T 4897-2015 for type-P3 furniture particleboard under dry state.
- 3. Increasing the target density, adhesive loading, and pyrolysis temperature within appropriate ranges can make *C. lanceolata/Uncaria* particleboard meet requirements of national standard GB/T 4897-2015 for type-P4 heavy-load particleboard under dry state.

- 4. The unique components in *Uncaria* extracts can facilitate dehydration and carbonization of timbers and accelerate the charring process, thus giving particleboard some flame retardant characteristics. The flame retardant components of *Uncaria* branches will be analyzed deeply in the future.
- 5. This study also demonstrated the feasibility of preparing particleboards with thinning wood of *C. lanceolata* and waste *Uncaria* branches. It provides a new way for comprehensive use of thinning wood of *C. lanceolata* and waste *Uncaria* branches.

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