

A Formaldehyde-Free Flame Retardant Wood Particleboard System Based on Two-Component Polyurethane Adhesive

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ABSTRACT: To address the problem of formaldehyde-free flame retardation of wood particleboard, a novel phosphorus-containing compound, di(2,2-dimethyl-1,3-propanediol phosphate) urea (DDPPU) was synthesized. DDPPU was used as flame retardant for wood particleboard. The flammability of treated wood particleboard systems consisted of wood particles, polyurethane (PU) adhesive, and different flame retardant formulations were investigated by limiting oxygen index (LOI). The results of LOI indicate that DDPPU could improve the flame retardancy of wood particleboard. However, when H_3BO_3 was used as the second flame retardant component and combined with DDPPU, the flame retardant wood particleboard could obtain the highest LOI

value (46.0) in these experiments. Thermogravimetric analysis shows that treated wood particleboard can decrease the initial decomposition temperature, and that at higher temperatures the degradation rate are lower than the untreated wood particleboard. Furthermore, wood particleboard treated with DDPPU/ H_3BO_3 has a higher yield of residue char at 600°C than that treated with other flame retardant systems. The ability of char formation of these samples agrees with the order of LOI values. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 108: 1216–1222, 2008

Key words: flame retardance; wood particleboard; intumescence; polyurethane; adhesive

INTRODUCTION

Recently, there is a growing tendency in using wood particleboard and wood-based products in various industries such as building and transportation. Wood particleboard is manufactured from lignocellulosic-wood material, in the form of discrete particles, combined with synthetic resin and bonded together under heat and pressure in a hot press. Adhesives based on urea-formaldehyde (UF) and phenol-formaldehyde (PF) are commonly used for wood particleboard,^{1,2} but they are sensitive to hydrolysis and stress scission. The adhesives also produce health hazards because of the formaldehyde they release.³ To resolve such problems, scientists are trying to develop new polymeric adhesives. Polyurethane (PU) is a class of polymer that has found widespread applications not only in several industries but also in daily life such as furniture, adhesives, fibers, paints, elastomers, coatings for automobiles, and synthetic skins. As per literature survey,

PU adhesive has developed a reputation for reliability, high performance, and no formaldehyde release.^{4–6} So it is necessary to use PU adhesive instead of UF or PF adhesives for wood particleboard.

Wood particleboards currently used in the furniture construction and transport industries are in the form of boards and panels. However, one of the main limitations for the use of wood particleboard is its flammability.^{7–9} The lowered flammability of wood particleboard-based panels enables them to be used in high-performance applications, which places emphasis on research into improving their fire retardant properties. Therefore, the study on flame retardant treatment of wood materials has attracted considerable industrial and scientific interest in recent years. The most usual method to improve the fire performance of wood particleboard is the chemical treatment with flame retardants.¹⁰ There are three main methods of applying flame retardants to wood and wood products: impregnation of wood with the solution of flame retardants, incorporation of flame retardants into the glue system, and surface treatment of product.¹¹ Some inorganic chemicals such as H_3BO_3 , Na_2SO_3 , Na_2HPO_4 , melamine, and so on were developed to be flame retardants for wood materials. However there are still some problems, for example, the high-additive amount, which is a characteristic of most inorganic flame retardants,

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will cause the wood particleboards higher sensitivity to moisture, etc.^{12–14}

To our best knowledge, now most flame retardant wood particleboards were still based on UF and PF adhesives. The flame retardancy of wood particleboard based on polyvinyl alcohol-polyarylate-poly-methylene-isocyanate (PVA-PAPI), a two-component PU adhesive, which is extremely positive in the environment sense because of its formaldehyde-free character, has hardly been addressed. Therefore, the objective of this study was to synthesize a highly effective phosphorus-containing flame retardant di(2,2-dimethyl-1,3-propanediol phosphate) urea (DDPPU), and investigate the flame retardant behavior of DDPPU and its synergist with some traditional flame retardants in the wood particleboard based on PU adhesive.

EXPERIMENTAL

Materials

All reagents and solvents were of reagent grade or were purified by standard methods before use. Phosphoryl chloride and 2,2-dimethyl-1,3-propanediol were obtained from Ruitejin Chemical Reagent (Tianjin, China) and No.1 Reagent Company of Shanghai (Shanghai, China), respectively. 1,2-Dichloroethane, H₃BO₃, Na₂SiO₃, melamine, and urea were purchased from Kelong Chemical Engineering Reagent Company (Chengdu, China). PU adhesive was obtained from Skybamboo Chemical Reagent Factory (Nanjing, China).

Measurement

The structure of DDPPU was characterized by ¹H NMR, ³¹P NMR, and ¹³C NMR which was performed on a FT-80A NMR by using chloroform as a solvent. The FTIR spectra of 2,2-dimethyl-1,3-propanediol phosphoryl chloride (DPPC) and DDPPU were recorded with KBr powder by using a Nicolet FTIR 170SX infrared spectrophotometer. Melting point was measured with a XRC-1 melting point analyzer.

The limiting oxygen index (LOI) of all samples was tested on an oxygen index instrument at room temperature. The LOI value was measured on a HC-2C oxygen index meter (Jiangning, China) with sheet dimensions of 130 mm × 6.5 mm × 3 mm according to ASTM D 2863-97.

Thermogravimetric analysis (TGA) was conducted on Q500 V6.4 Build 193 thermal analyzer at heating rate of 10°C/min. Samples were heated in the temperature range from room temperature to 600°C under air at a flow rate of 60 mL/min. The weight of samples was kept with 3–5 mg.

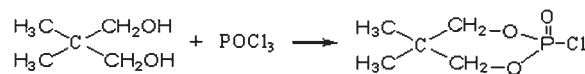
Scanning electronic microscopy (SEM) observed on a JEOL JSM-5900LV was used to investigate the residues of wood particleboards. The residue samples for SEM were obtained after combustion in their limiting oxygen concentration.

Preparation of 2,2-dimethyl-1,3-propanediol phosphoryl chloride^{15,16}

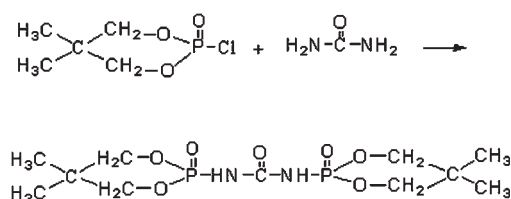
A 250-mL four-necked round bottom flask was equipped with a mechanical stirrer, reflux condenser, thermometer, addition funnel, and aqueous NaOH trap. The flask was charged with 30-mL dichloroethane and 10.4 g 2,2-diethyl-1,3-propanediol (0.1 mol). The mixture was stirred and heated. When the reaction temperature reached 55°C, 9.6 mL POCl₃ was added within about 1.5 h. Thereafter the mixture was heated to 83°C and kept under reflux for about 3 h. The reaction was kept at the same temperature until no HCl released. Successively, the reaction mixture was cooled slowly to room temperature and removed the dichloroethane solvent. The white solid was washed once with 30-mL hexane and twice with 30-mL ether. The powdery product was dried at 70°C under vacuum to a constant weight. The yield of white solid product was 92 wt % and melting point of purified product was 94–96°C. Then DPPC was demonstrated by FTIR (KBr) analysis, which showed absence of free —OH group absorption around 3590–3650 cm⁻¹. The FTIR spectrum exhibited absorption at 1312 cm⁻¹ (P=O); 1180, 973 cm⁻¹ (P—O—C); 550 cm⁻¹ (P—Cl). The reaction route of DPPC is shown in Scheme 1.

Synthesis of di(2,2-dimethyl-1,3-propanediol phosphate) urea

DDPPU was prepared from urea and DPPC. The reaction route is shown in Scheme 2. A 250-mL four-necked round bottom flask was equipped with a mechanical stirrer, reflux condenser, thermometer, addition funnel, and aqueous NaOH trap. DPPC (0.1 mol) dissolved in 30-mL dichloroethane was added into the 250-mL reaction vessel, the system was slowly heated to 60°C and kept for 30 min. Then 3.6 g urea was added with stirring and kept at 60°C until HCl evolution subsided. Thereafter, the mixture was gradually heated to 83°C and refluxed until no HCl gas was emitted. Successively, the reaction mixture was cooled slowly to room temperature and the dichloroethane was removed, then the raw product obtained was filtrated, and washed twice by



Scheme 1 The synthesis of DDPC.



Scheme 2 The synthesis of DDPPU.

50-mL water. The product was dried to constant weight at 70°C in vacuum. The yield of white solid product was 85 wt % and melting point of purified product was 185–187°C. The white solid product was demonstrated by FTIR (KBr) analysis (shown in Fig. 1), which showed absence of free P–Cl absorption around 550 cm⁻¹. The FTIR spectrum exhibited absorption at 1308 cm⁻¹ (P=O); 1168, 974 cm⁻¹ (P–O–C); 1680 cm⁻¹ (C=O); 963, 709 cm⁻¹ (P–N); 3430 cm⁻¹ (–NH). Figure 2 shows the ¹H NMR spectra of DDPPU. Because of the space effect, methyl or methylene is separated two types. The very strong peaks at about 0.90 and 1.3 ppm should be assigned to the two types of methyl. The peaks of two types of methylene were observed at 4.0 and 4.5 ppm. The proton of –NH group could combine with P=O groups as hydrogen bonding, so it can not appear in 11.0–13.0 ppm clearly in the ¹H NMR spectra when chloroform is the solvent. Furthermore, the ³¹P NMR and ¹³C NMR spectra of DDPPU are shown in Figures 3 and 4, respectively, and the adscription of P and C have signed in the figures. All these facts allowed us to conclude that the target product was synthesized successfully.

Flame retardant treated particleboard¹⁷

Ordinary wood particleboard was prepared on a laboratory scale. Adhesive loading was 12 wt % calcu-

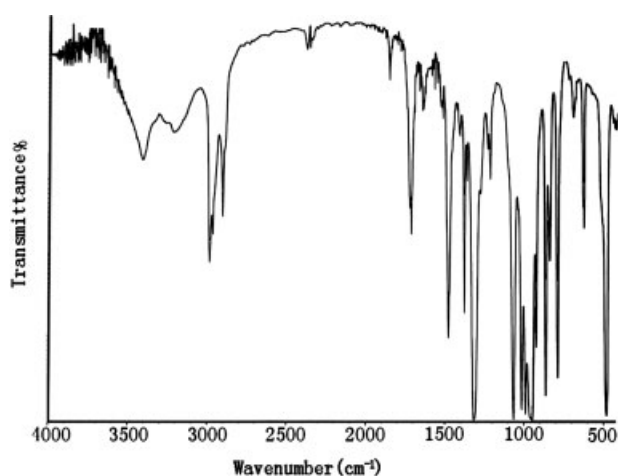


Figure 1 The FTIR of DDPPU.

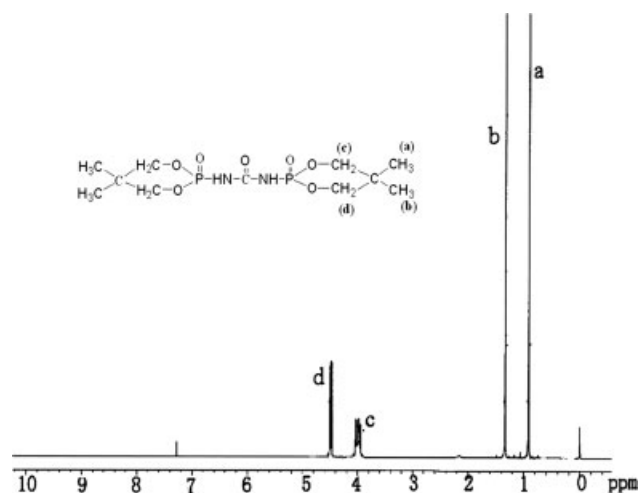


Figure 2 The ¹H NMR of DDPPU.

lated on the dry wood particles weight. The moisture content of wood particles was 3 wt %. The blend of wood particles, flame retardants, and adhesive was pressed at 50°C for 5 min, followed by hot pressing at 90°C under a pressure of 6–7 MPa for 30 min. The amount (wt %) of flame retardant was calculated from the following equation:

$$FR\% = \frac{W_t - W_w - W_p}{W_t} \times 100\%$$

where W_t , W_w , and W_p represent the weight of flame retardant particleboard, wood particles, and PU adhesive, respectively, and all of them were dried to constant weight.

RESULTS AND DISCUSSION

Flammability

LOI is a parameter for evaluating flame retardancy and flammability of polymeric materials in the same

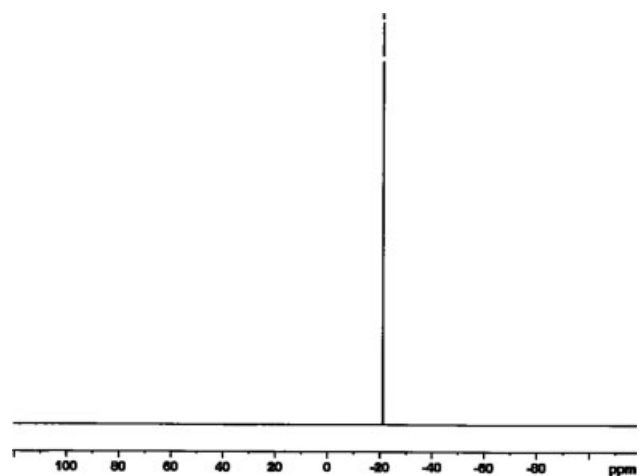


Figure 3 The ³¹P NMR of DDPPU.

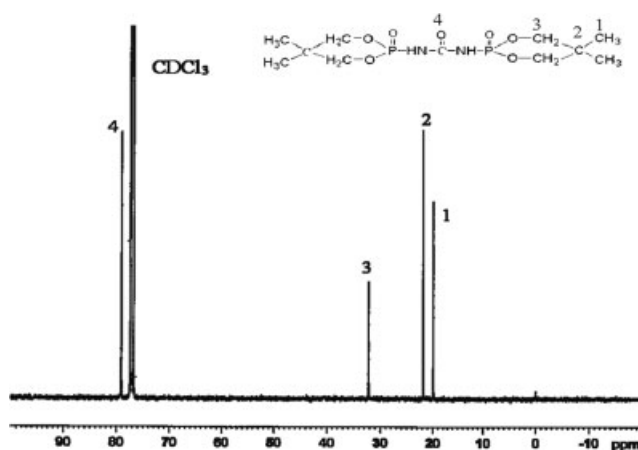


Figure 4 The ^{13}C NMR of DDPPU.

conditions. It denotes the lowest volume concentration of oxygen sustaining candle-like burning of materials in mixing gases of nitrogen and oxygen.¹⁸ The higher the LOI values, the more effective is the flame-retardant treatment. Table I presents the LOI test results of DDPPU-treated particleboard with different loading level of additives. Table II presents the LOI test results of particleboard treated with DDPPU combined with other conventional flame retardants. As we know, when intumescent flame retardants are exposed to fire, they form a non-flammable, multicellular char layer on the surface of polymers. This layer provides an efficient shield and insulation for the underlying material against direct contact with fire, oxygen, and heat. As a result, the structure and formation of char layer is a critical factor for the flame retardancy of intumescent flame retardants.¹⁹

From Tables I and II we can conclude that wood particleboard is easily flammable and its LOI value is only 22.3, because wood particles and PU adhesive are easily flammable materials. DDPPU is an effective flame retardant for wood particleboard. It obviously enhanced the flame retardancy of wood particleboard, and the LOI value of the wood particleboard increased from 22.3 to 39.7 as the loading of DDPPU increased from 0 to 18 wt %, as shown in Table I. When the loading of DDPPU increased from 12 to 18 wt %, no obvious increase in LOI values was observed. Therefore, we decreased the loading of DDPPU in the wood particleboard, and combined DDPPU with other traditional flame retardants as the second flame retardant component to enhance the flame retardancy and reduce the cost of the non-flammable wood particleboard.

From Table II, it can be seen that when the total amount of flame retardant in the wood particleboard was maintained at 15 wt %, both Na_2SiO_3 and melamine could not further improve the flame retardancy of the wood particleboard treated with DDPPU. In

comparison with the flame retardant particleboard with only 15 wt % DDPPU, the LOI value of the sample with 10 wt % DDPPU and 5 wt % Na_2SiO_3 as the second flame retardant component decreased from 38.1 to 35.4; while 5 wt % melamine was used as the second flame retardant component, the LOI value of the sample decreased to 35.1. From the phenomenon of burning we can see that Na_2SiO_3 can restrain the smoke release well, so it is also important to applying Na_2SiO_3 in the wood particleboard as a flame retardant and smoke suppressant. However, when H_3BO_3 was used as the second flame retardant component and combined with DDPPU, the flame retardant wood particleboard with 10 wt % DDPPU and 5 wt % H_3BO_3 obtained the highest LOI value (46.0), which increased about 8 units compared with the LOI value (38.1) of the sample treated with only 15 wt % DDPPU, and about 7 units compared with the LOI value (39.2) of the sample treated with only 15 wt % H_3BO_3 . So we can say that the incorporation of H_3BO_3 to the flame retardant wood particleboard system with DDPPU is the most effective method to improve the flame retardancy of wood particleboard in this study. In other words, DDPPU and H_3BO_3 show a remarkably synergistic flame retardant effect to wood particleboard, which means that the addition of H_3BO_3 offers the possibility of decreasing the level of DDPPU in the particleboard matrix.

Thermogravimetric behaviors

The associated TGA and DTG curves of Samples A–E carried out in dynamic air atmosphere from ambient temperature to 600°C are shown in Figures 5 and 6. Table III shows the experimental data for particleboard and the particleboard treated with flame retardant such as DDPPU, DDPPU/ H_3BO_3 , DDPPU/ Na_2SiO_3 , and DDPPU/melamine from the TGA and DTG.

The TGA and DTG curves of Sample A show three main stages of degradation. There is a mass loss of 51.9% in the temperature range from 240 to 330°C at the first stage. Of the four components in wood particleboard, the thermal degradation of PU adhesive and hemicellulose occurs first, then cellulose rapidly decompose at higher temperatures via

TABLE I
Flame Retardance of DDPPU-Treated Particleboard

Samples	DDPPU (wt %)	LOI (%)
A1	0	22.3
A2	5	28.7
A3	9	32.4
A4	12	36.0
A5	15	38.1
A6	18	39.7

TABLE II
Flame Retardance of DDPPU Combined with Other Flame Retardants-Treated Particleboard

Samples	DDPPU (wt %)	Na ₂ SiO ₃ (wt %)	H ₃ BO ₃ (wt %)	Melamine (wt %)	LOI (%)
A	0	0	0	0	22.3
B	15	0	0	0	38.1
C	10	5	0	0	35.4
D	10	0	5	0	46.0
E	10	0	0	5	35.1
F	0	0	15	0	39.2

the formation of laevoglucose, which can be further decomposed into volatile and flammable products. On the other hand, lignin decomposes at lower rates in a broad temperature range by char oxidation. So the decomposition of PU adhesive or dehydration of hemicellulose occurs at the first stage.^{20,21} At the second stage, 20.0% of mass loss occurs in the temperature range from 330 to 440°C. The peak of DTG at 437°C is because of the flaming combustion of cellulose.²⁰ After the two main decomposition stages, mass loss rates of the residual materials of wood particleboard are found to be quicker in the temperature range from 448 to 508°C. The TG curves show a mass loss of 18.3%. This is because of the oxidation of char derived from cellulose and lignin.^{22–24}

For the thermal analysis curves of wood particleboard, it is evident that the main decomposition of wood particleboard occurs at high temperature (>280°C), and the largest mass loss rate occurs in the temperature range from 280 to 330°C, which can be seen from the sharp peaks of DTG in Figure 4.

The TG and DTG curves of treated wood particleboard with different flame retardants (Samples B–E) are also shown in Figures 3 and 4. We can see TG curve of Sample B from Figure 3 (TG curve of Sample B), in which it can be seen that there is a mass

loss of 46.2% in the temperature range from 240 to 280°C, which is because of acid-catalyzed decomposition of hemicellulose and cellulose.²⁵ The second decomposition stage is prolonged in temperature range from 268 to 454°C, which shows a mass loss of 37.2%. The residue of Sample B is 14.0 wt % at 600°C, which is more than that of the untreated wood particleboard. From the TG and DTG curves in Figures 3 and 4, it is evident that the addition of DDPPU lowers the initial decomposition temperature at the first degradation stage. This is mainly because of acid-catalyzed dehydration and deacidification.

The thermal analysis curves of Sample D are quite different from those of Samples B, C, and E. At the first stage, there is a mass loss of 6.9% in temperature range from 170 to 258°C, which is because of the decarbonation and acid-catalyzed dehydration. At the second stage, it is evident that the main decomposition stage is divided into two stages (258–324°C and 324–429°C). There is a mass loss of 15.0% in the temperature range 258–324°C, which may be due to the limitation of released acids to catalyze decomposition of sample. The mass loss in the temperature range from 324 to 429°C is because of the decomposition of lignin and cellulose.

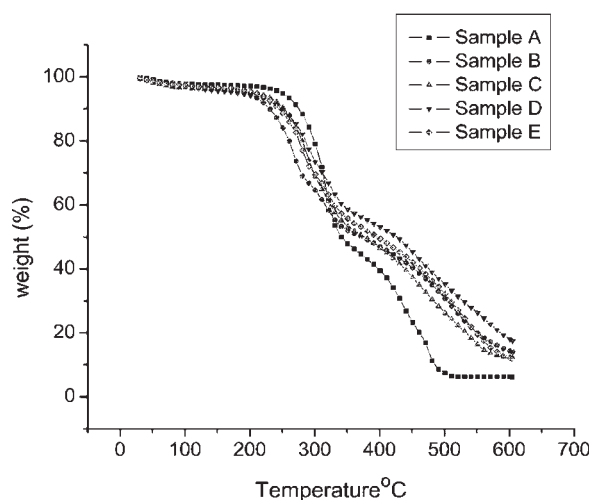


Figure 5 TG curves of samples in air.

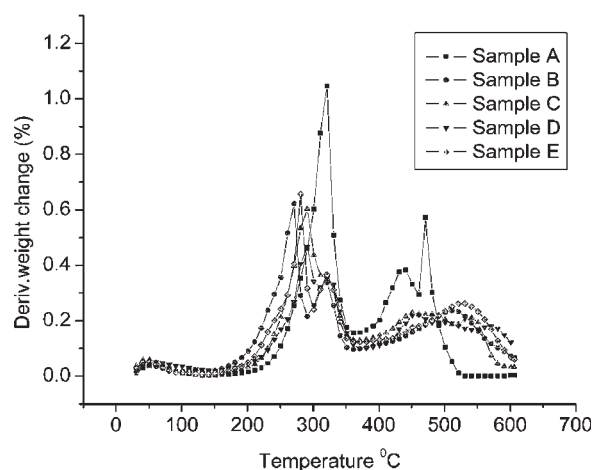


Figure 6 DTG curves of samples in air.

TABLE III
Calculated Results From TG and DTG Curves
of Various Samples

Samples	T_{on} (°C) ^a	T_{max} (°C) ^b	Y_c % ^c		
			320°C	430°C	600°C
A	288	318	53.1	26.3	6.2
B	237	268	54.9	41.7	14.0
C	255	288	57.4	39.1	12.2
D	258	287	63.2	47.3	17.6
E	252	252	59.5	43.6	11.9

^a T_{on} , the on-set degradation temperature.

^b T_{max} , the maximum-rate degradation temperature.

^c Y_c , the char yield at different temperature.

The thermal analysis curves of Samples C and E are similar to the Sample B as shown in Figures 3 and 4, but the char yields of Samples C and E are lower than that of Sample B whose char yield is 14.0 wt % at 600°C. The char yields of Samples C and E are 12.2 and 11.9 wt %, respectively. However, both of them are much higher than that of the untreated wood particleboard.

For the thermal degradation of Samples B–E, there are some differences related to flame retardancy when compared with Sample A. The wood particleboard treated with DDPPU and traditional flame retardant lowered the initial decomposition temperature at the first stage. The influence of flame retardants on thermal degradation of wood particleboard in air is shown in Figures 3 and 4. They show similar thermal degradation behavior. In general they promote char formation, especially DDPPU combined with H_3BO_3 shows a remarkable effect. For instance, the char residue of samples A–E is 6.2, 14.0, 12.2, 17.6, 11.9 wt % at 600°C, respectively. It is generally observed that the amount of char formed during thermal degradation of samples is related to the degree of flame resistance.^{20,26} The ability of char formation follows the order: Sample D > Sample B > Sample C > Sample E > Sample A, which agrees with the order of results of LOI values for these samples. These data suggest that combustibility of the treated particleboard is reduced.

Physical and mechanical properties of particleboard with flame retardants

Table IV gives physical and mechanical properties of untreated wood particleboard and treated particleboards, whose components are based on Table II. MOR, MOE, thickness swelling, and density were compared in this study. Physical properties such as density and thickness swelling were determined in accordance with CN GB4907.

From the data as shown in Table IV, we can see that the mechanical properties of Samples C and D such as MOR and MOE have hardly decreased compared with Sample A. It can be inferred that the flame

retardants such as boric acid and melamine can react with hydroxyl of cellulose or isocyanate that is a part of PU adhesive, which may increase the adhesion and water-resistant by hot press.²⁷ The results of thickness swelling reveal that the addition of DDPPU and its combination with traditional flame retardants have little effect on the thickness swelling compared with untreated wood particleboard. The possible reason is that the density of Samples B–E is higher than that of Sample A, and water is much more difficult to be penetrated into the wood particleboard body. Furthermore, DDPPU is hydrophobic and contains imine which can react with isocyanate, so it can decrease the negative impact on properties of wood particleboards when added into the wood particleboards.

Morphology of residues

SEM micrographs in Figure 7 show the surfaces and interiors microstructures of the samples after combustion. From the SEM observations, there is an obvious difference between the morphologies of two samples' residues. From Figure 7, it can be observed that Figure 7(b) has more homogeneous and compact charred layers than Figure 7(a). The smooth and compact charred layers can form protective shields to protect effectively internal structure, inhibit the transmission of heat during contacting fire. Moreover Figure 7(c,d) shows the compact and spongy intumescent layer of interior char of Sample D. The intumescent charring layer with spongy as the effect of flame retardant DDPPU decomposed when sample was burning. The intumescent charring layer could also have a significant effect on thermal insulation and barrier properties.

CONCLUSIONS

A novel phosphorus-containing flame retardant DDPPU for wood particleboard was synthesized successfully. The flame retardant systems of DDPPU combined with some traditional flame retardant can improve the flame retardancy of wood particleboard based on two-component PU adhesive effectively. The wood particleboard containing 10 wt % DDPPU and 5 wt % H_3BO_3 had the highest LOI value of 46.0, increasing about 8 LOI units from 38.1 of LOI

TABLE IV
Properties of Samples A–E

Samples	Density (g/cm ³)	MOR (MPa)	MOE (MPa)	TS (%)
A	863	15.5	7,238	8.2
B	922	14.2	6,702	7.9
C	930	14.9	6,988	9.1
D	921	15.1	7,021	8.5
E	929	14.4	6,864	9.0

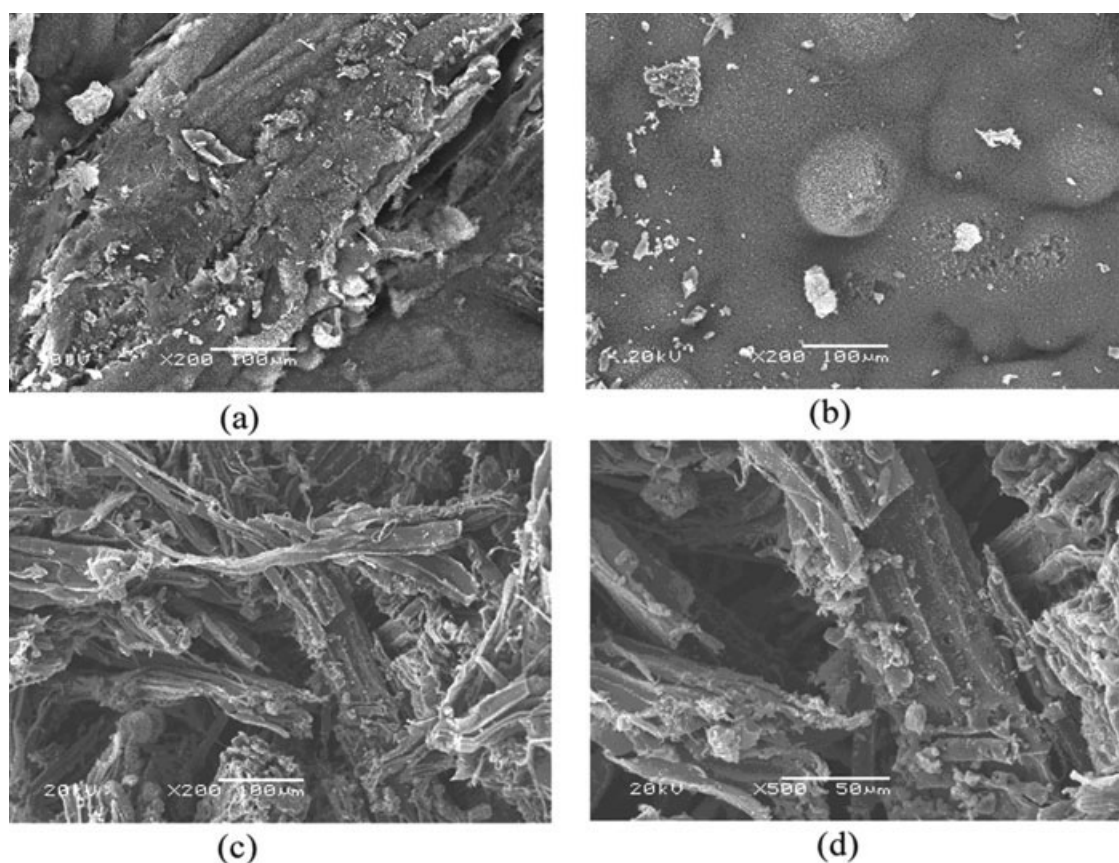


Figure 7 SEM microstructures of surfaces of residues of Sample A (a) and Sample D (b) and microstructures of interior char of Sample A (c) and Sample D (d).

value the sample treated with only 15 wt % DDPPU had, and being also higher than that of the sample treated with only 15 wt % H_3BO_3 . That is to say, DDPPU and H_3BO_3 display a remarkably synergistic flame retardant effect to wood particleboard. TGA indicates that the char yield of wood particleboard treated with 10 wt % DDPPU and 5 wt % H_3BO_3 is also higher than those of the samples treated with other flame retardant systems. The ability of char formation follows this order: Sample D > Sample B > Sample C > Sample E > Sample A, which agree with the order of LOI values. Furthermore, the flame-retardant systems of DDPPU combined with traditional flame retardants can overcome some drawbacks of conventional flame-retardant systems such as high loading level of flame retardants, high sensitivity to moisture, and so on.

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