Waste Tetra Pak and Wood Shavings Composites: Flame Retardant Modification

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This study aimed to achieve the value-added utilization of waste Tetra Pak (WTP) and to alleviate the shortage of wood resources by partially replacing wood shavings with WTP and using phenol-formaldehyde resin (PF) to prepare composites. Flame-retardant modification was conducted by introducing single additives-boric acid/borax (BA/Brx), ammonium polyphosphate (APP), and disodium octaborate tetrahydrate (DOT)-as well as the combined systems (BA/Brx/APP, BA/Brx/DOT, and BA/Brx/APP/DOT). Their effects on flame retardancy, mechanical properties, and thermal stability were investigated. It was found that all six systems improved flame retardancy, among which the DOT-modified composite specimen (Z3) achieved a limiting oxygen index (LOI) of 34.3%, representing a 25.6% increase compared with the control composite specimen (Z0), reaching the flame-retardant grade. The mechanical properties of Z3 met the requirements of GB/T 4897 (2015) for general particleboard. Fourier transform infrared spectroscopy (FTIR) indicated that the flame retardants interacted with the matrix through hydrogen bonding and functional composite specimen reactions. Thermogravimetric analysis (TG) showed that the char yield of Z3 reached 32.0%, which was 4.16 times higher than that of the control composite specimen, indicating a significant improvement in thermal stability. This study provides a feasible pathway for WTP recycling and the preparation of flame-retardant composites.

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INTRODUCTION

With the rapid development of the food packaging industry, Tetra Pak cartons, as a multilayer composite packaging material (Polyethylene [PE]/paper/Low-Density Polyethylene [LDPE]/aluminum foil/organic adhesive layer/PE), have been widely used in the packaging of liquid food products, such as milk and beverages, due to their excellent sealing properties and long shelf life (Ma 2018). However, the increasing consumption of Tetra Pak cartons has resulted in a growing environmental burden. Statistical data indicate that recycling one metric ton of “brick-type” Tetra Pak cartons can save approximately 11 ten-year-old trees, 0.3 tons of petroleum, or 7.5 tons of bauxite. Nevertheless, more than 80% of discarded Tetra Pak cartons are still disposed of by landfilling or incineration, leading to a significant waste of paper, aluminum, and plastic resources, and potentially causing soil and air pollution (Ye *et al*. 2022). Therefore, achieving the resource utilization and high-value recycling of waste Tetra Pak cartons has become an urgent issue to be addressed.

Wood-based composites, such as particleboard, are widely used as essential materials in the furniture and construction industries (Li *et al*. 2020). China faces a relative shortage of wood resources, and the inherent flammability of wood-based materials limits their broader application (Li *et al*. 2023; Albert and Liew 2024). Substituting part of the wood shavings with discarded Tetra Pak cartons, combined with flame-retardant modification to enhance material safety, could not only alleviate the shortage of timber resources but also enable the recycling and high-value utilization of waste packaging materials. This approach simultaneously achieves “resource circulation” and “performance optimization”, thereby aligning with the principles of green development and the national “dual-carbon” goals (Jian *et al*. 2019).

At present, the recycling and reuse of discarded Tetra Pak cartons can generally be classified into two categories: separation-based recycling and integrated recycling. Among these, direct regeneration technology has attracted increasing attention due to its simple process and low production cost. Bekhta *et al*. (2016) mixed Tetra Pak cartons with food packaging films and fabricated composite boards through hot pressing without using any adhesive; the resulting panels met the minimum requirements of the European particleboard standard. García *et al*. (2020) utilized Tetra Pak flakes and high-density polyethylene(HDPE) as raw materials to produce composite boards suitable for furniture and construction applications. Ebadi *et al*. (2016) reported that composites containing 30% Tetra Pak and 3% maleic anhydride grafted polyethylene (MAPE) exhibited the best mechanical performance. Furthermore, Xu *et al*. (2015) developed electromagnetic shielding boards reinforced with Tetra Pak and iron fibers, expanding the high-value application potential of waste Tetra Pak materials. However, most existing studies have primarily focused on optimizing a single property, such as mechanical strength, while research on the flame-retardant modification and underlying mechanisms of Tetra Pak–wood shavings composite systems remains limited.

Flame-retardant modification of wood-based materials is typically achieved through methods such as impregnation, flame-retardant bonding, or surface coating (Popescu and Pfriem 2020). For example, Yan *et al*. (2019) synthesized magnesium phosphate ester (MPEA) flame retardants and incorporated them with amino resins to prepare transparent intumescent fire-resistant coatings, which exhibited excellent synergistic flame-retardant performance. Jier *et al.* (2023) utilized recycled plastics and soybean residues as the matrix, and by adding an intumescent flame retardant (IFR), the resulting composite achieved a limiting oxygen index (LOI) of 31.0% while reducing the heat release rate by 40%. However, these studies have primarily focused on flame-retardant modification of single-component matrices (such as pure wood or pure plastics). Research concerning the flame-retardant mechanisms and performance regulation of Tetra Pak-wood shavings composite systems remains limited. Moreover, the influence of flame retardants on the interfacial bonding behavior and mechanical properties of such composites has not yet been fully elucidated.

In this study, motivated by the goals of sustainable utilization and environmental protection, composite materials were fabricated using discarded Tetra Pak cartons as a partial replacement for wood shavings, with phenol-formaldehyde resin serving as the adhesive. Both single-component and combined flame-retardant systems were introduced to modify the composites. The limiting oxygen index (LOI) and physical–mechanical properties were evaluated, while Fourier transform infrared spectroscopy (FTIR) and thermogravimetric analysis (TG) were employed to characterize the flame-retardant mechanisms. The effects of different flame-retardant systems on the flame retardancy and mechanical performance of the composites were systematically investigated, and the interfacial interactions and thermal degradation behaviors between the flame retardants and the matrix were elucidated. The aim of this research was to develop flame-retardant Tetra Pak/wood shavings composites that meet national performance standards, thereby providing theoretical and technical support for the high-value recycling of waste Tetra Pak cartons and the flame-retardant modification of wood-based composite materials.

EXPERIMENTAL

Materials

The waste Tetra Pak used in this study was collected from milk product packaging within three months of use. The collected Tetra Pak materials were manually cleaned, dried in a convection drying oven, and then cut into 5 mm × 5 mm pieces using a paper-cutting machine. Wood shavings were obtained by planing Chinese fir (*Cunninghamia lanceolata*) logs with a wood shaver. The shavings were screened to select particles of similar size to the Tetra Pak fragments, and the thickness of the resulting wood shavings was approximately 1 mm. Phenol-formaldehyde resin (solid content: 56%) was purchased from Guangdong Aiktair New Materials Co., Ltd. Waste polyethylene was derived from discarded transparent polyethylene plastic bags with a width of 30 cm and a height of 45 cm. Emulsified wax (58# paraffin, solid content: 37.2%) was purchased from Gu’an Shenghui Flame Retardant Materials Co., Ltd. The main reagents required for the experiments included sodium hydroxide (AR grade) from Aladdin Biochemical Technology Co., Ltd. (Shanghai, China), boric acid (AR grade) and borax (AR grade) from Tianjin Damao Chemical Reagent Co., Ltd., ammonium polyphosphate (AR grade) from Macklin Biochemical Technology Co., Ltd. (Shanghai, China), disodium octaborate tetrahydrate (AR grade) from Titan Technology Co., Ltd. (Shanghai, China), anhydrous ethanol (AR grade) from Jiangsu Aikang Biomedical R&D Co., Ltd., potassium bromide (AR grade) from Jizhi Biochemical Technology Co., Ltd. (Shanghai, China), and deionized water (purified water) from Lianlong Bohua Pharmaceutical Chemical Co., Ltd.

The main instruments used in the experiments included a precision convection drying oven (RPG-9140A, Shanghai Yiheng Scientific Instruments, China), an automatic hot press (CARVER 3895, Carver Inc., USA), and a bone saw machine (BSM-400, Tianjin Zhihua Mechanical & Electronic Co., Ltd., China). Other instruments included a magnetic stirrer (DF-101SZ, Shanghai Yushen Instruments, China), a differential scanning calorimeter (PerkinElmer DSC 8000, PerkinElmer Inc., USA), a simultaneous thermal analyzer (SDT650, Waters Technology Shanghai Co., Ltd., China), and a universal testing machine (Instron 3369, Instron Inc., USA). An electronic analytical balance (JY3001, Shanghai Precision Scientific Instruments Co., Ltd., China) was also used, along with a Fourier transform infrared (FTIR) spectrometer (ThermoFisher Scientific IS5, Thermo Fisher Scientific Inc., USA). A constant temperature and humidity chamber (RTH-408ST, Rongxin Litian Experimental Equipment Co., Ltd., China) and a digital thermostatic water bath (HH-6, Shanghai Lichen Bangxi Instrument Technology Co., Ltd., China) were employed in the study. Additionally, an oxygen index tester (KS-653B, Shanghai Jinsen Testing Equipment Co., Ltd., China) was used. Auxiliary equipment included 250 mL/500 mL beakers, a 100 mL graduated cylinder, glass rods, and a vernier caliper.

Preparation of Flame Retardants

The six systems developed based on the preliminary experimental results are presented below. The mass fraction of each flame retardant corresponded to its maximum solubility in deionized water at 25 °C (*i.e.*, the system remained stable without precipitation after magnetic stirring for 2 h).

Table 1. Formulations of the Six Flame-retardant Systems

No.	Type of Flame Retardant	Mass Fraction of Reagents (wt%)
Z0	No treatment	--
Z1	Boric acid / Borax (BA/Brx)	20 : 20
Z2	Ammonium polyphosphate (APP)	2
Z3	Disodium octaborate tetrahydrate (DOT)	10
Z4	BA/Brx/APP	20 : 20 : 2
Z5	BA/Brx/DOT	20 : 20 : 10
Z6	BA/Brx/APP/DOT	20 : 20 : 2 : 10

Preparation of Composites

The target density of the recycled Tetra Pak/wood shavings composite board was 720 kg·m^{-3}. The procedure was to weigh a certain amount of wood shavings and place them in a blast drying oven. Drying was at 105 °C for 24 h. After drying, the wood shavings were immersed in a 10% sodium hydroxide solution and sonicated for 6 hours. Afterward, the wood shavings were removed and rinsed with deionized water until the wash liquid became neutral. Finally, the treated wood shavings were dried to obtain alkali-treated wood shavings. The wood shavings treated with alkali, waste Tetra Pak particles, and waste transparent polyethylene particles were processed to approximately 5 mm × 5 mm in size. The Tetra Pak particles and alkali-treated wood shavings were mixed thoroughly in a 6:4 mass ratio, and then 30% PE (based on the total mass of wood shavings and Tetra Pak) was added. The mixture was placed in a convection drying oven and dried until the moisture content reached 5% to 8%.

The alkali-treated wood shavings, Tetra Pak scraps, and PE were mixed thoroughly after drying. During the mixing process, the prepared flame retardant was added uniformly. The flame retardant-modified mixture was placed in the drying oven at 103 ± 2 °C and dried until the moisture content reached 5% to 8%.

Phenol-formaldehyde resin (PF) was added to the mixture at a concentration of 12% based on dry mass, and emulsified wax were added to the impregnated substrate, and the mixture was stirred at high speed for 10 min to ensure uniform coating of the adhesive.

The adhesive-coated material was manually laid as 150 mm × 200 mm × 6 mm panel blanks. After pre-pressing, the material was placed into an automatic hot press and hot-pressed at 170 °C and 1.6 MPa for 17.5 min. The pressed panels were then removed and cooled to room temperature.

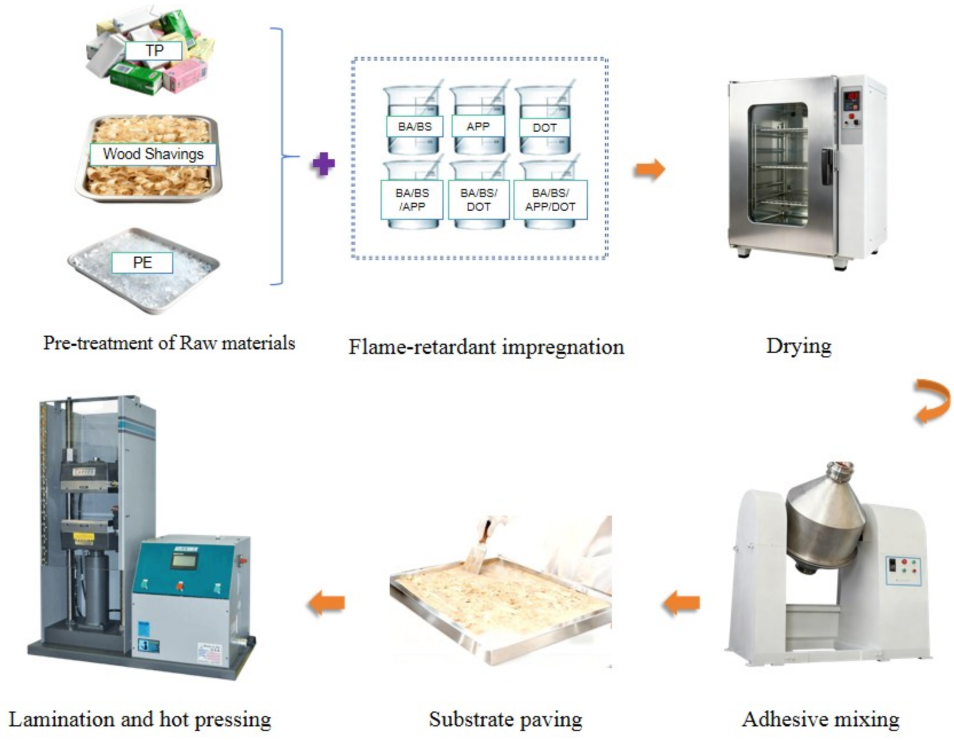


Fig. 1. Flowchart of the composite preparation process

Testing of Physical and Mechanical Properties

In accordance with GB/T 17657 (2022), all specimens were conditioned to constant mass in an environment of 20 ± 2 °C and 65 ± 5% relative humidity prior to testing. The modulus of rupture (MOR), modulus of elasticity (MOE), internal bond strength (IB), and 24-h thickness swelling(TS) were measured separately. According to GB/T 4897 (2015), the number of samples examined for each experiment was as follows: 12 samples for both MOE and MOR, 8 samples for IB, and 8 samples for TS.

The flexural properties of the specimens were measured using the three-point bending method. Specimens with dimensions of 200 mm × 50 mm × 6 mm were tested, and the MOE and MOR at the maximum failure load were recorded. The values were determined with an accuracy of 10 MPa for MOE and 0.1 MPa for MOR.

For IB, specimens with dimensions of 50 mm × 50 mm × 6 mm were used. The specimens were bonded to the clamps with hot-melt adhesive and tested at a loading speed of 10 mm/min. The IB was calculated according to Eq. 1, with values determined to an accuracy of 0.01 MPa,

$$\boldsymbol{\sigma}_{\perp} = \frac{\boldsymbol{F_{max}}}{\boldsymbol{l \times b}} \tag{1}$$

where $\boldsymbol{\sigma}_{\perp}$ is the internal bond strength of the specimen (MPa) and $F_{\max}$ represents the maximum failure load (N), while l and b represent the length (mm) and width (mm), of the specimen, respectively.

For 24-h thickness swelling (TS), specimens with dimensions of 50 mm × 50 mm × 6 mm were immersed in deionized water at 20 °C for 24 h. The TS was calculated according to Eq. 2, with values determined to an accuracy of 0.1%,

$$T = \frac{t_2 - t_1}{t_1} \times 100 \tag{2}$$

where T represents the thickness expansion rate (%), t_1 and t_2 represent the thickness (mm) of the specimen before and after the water absorption test, respectively.

The performance evaluation of waste Tetra Pak/wood shavings composites was conducted with reference to GB/T 4897 (2015), according to the requirements for "general type particleboard under dry conditions" (Table 2): MOR ≥ 11.5 MPa, MOE not specified (≥ 1900 MPa for furniture type), IB ≥ 0.30 MPa, and TS not specified (≤ 22.0% for load-bearing type).

Table 2. Performance Requirements for Particleboard

Type	Modulus of Rupture (MPa)	Modulus of Elasticity (MPa)	Internal Bond (MPa)	24 h Thickness Swelling (%)
General	≥ 11.5	—	≥ 0.30	—
Furniture	≥ 12.0	≥ 1900	≥ 0.45	—
Load-bearing	≥ 15.0	≥ 2200	≥ 0.45	≤ 22.0
Heavy-duty	≥ 20.0	≥ 3100	≥ 0.60	≤ 16.0

Limiting Oxygen Index Test

The limiting oxygen index (LOI) of the samples was measured using a KS-653B oxygen index apparatus. Specimens with dimensions of 100 mm × 3 mm × 6 mm were ignited from the top surface. A N_2/O_2 mixed gas flow of 20 L/min was maintained during the test. The LOI values were determined using Dixon's ascending-descending method (Eq. 3) and are reported with a precision of 0.1%,

$$\boldsymbol{LOI = c_1 + kd} \tag{3}$$

where C_1 is the last recorded oxygen concentration during the measurement, expressed as a volume fraction (%) and reported to one decimal place, d is the difference between the oxygen concentrations used and controlled during the test, also expressed as a volume fraction (%) and reported to one decimal place, and k is a coefficient used to calculate the oxygen index when applying Dixon's ascending-descending method.

Fourier Transform Infrared Analysis

Pellets were prepared by the KBr method (sample:KBr = 1:80). Approximately 2 mg of sample was thoroughly mixed with 160 mg of dry KBr and pressed into a transparent disk. Spectra were collected over 4000 to 500 cm^{-1} with a resolution of 4 cm^{-1} and 16 scans. Changes in functional composite specimens were interpreted from the resulting spectra.

Thermogravimetric Analysis

The TG measurements were performed on a PerkinElmer DSC 8000 instrument. Samples (5 to 10 mg) were analyzed under N_2 atmosphere (flow rate: 50 $mL\cdot min^{-1}$) at a heating rate of 10 $°C\cdot min^{-1}$ over 50 to 600 °C. The TG and DTG curves were recorded to determine the onset decomposition temperature (T_0), the temperature at the maximum decomposition rate (T_P), and the char yield at 600 °C.

RESULTS AND DISCUSSION

Impact of Flame-retardant Systems on Composite Flame Retardancy

The LOI values of the waste Tetra Pak/wood-shavings composites without and with various flame retardants are shown in Table 3. As can be seen, the unmodified Z0 specimens exhibited an LOI of 27.3%, which can be attributed to the phenol-formaldehyde resin adhesive used during fabrication, a polymer material that is intrinsically flame-resistant, high-temperature tolerant, low-smoke emitting, and low-toxicity (Wang *et al*. 2025). After the addition of different flame retardants, the LOI values of the composites increased compared to the Z0 specimens, indicating that these flame retardants can enhance the flame retardancy of the waste Tetra Pak/wood-shavings composites to a certain extent.

Table 3. Flame Retardant Test Results of Waste Tetra Pak/Wood Shavings Composites

No.	Flame Retardant Composition	LOI (%)	Improvement Rate *vs* Z0 (%)
Z0	Blank	27.3	-
Z1	Boric acid/Borax	30.7	12.5
Z2	Ammonium polyphosphate (APP)	29.2	7.0
Z3	Disodium octaborate tetrahydrate (DOT)	34.3	25.6
Z4	Boric acid/Borax/Ammonium polyphosphate	33.8	23.8
Z5	Boric acid/Borax/Disodium octaborate tetrahydrate	32.3	18.3
Z6	Boric acid/Borax/Ammonium polyphosphate/Disodium octaborate tetrahydrate	32.4	18.7

The Z1 composite specimens had an LOI of 30.7%, which was a 12.5% increase compared to the Z0 composite specimen. This is because, at high temperatures, boric acid and borax (BA/Brx) undergo a dehydration reaction, forming a glassy coating that adheres to the material surface. The coating effectively isolates oxygen from the material and prevents it from reaching the combustion zone, thus suppressing combustion. Additionally, boric acid and borax release bound water at the burning temperature, providing a cooling and heat-absorbing effect, which further slows or prevents combustion (Wang *et al*. 2014). This result is consistent with the findings of other researchers, such as Wu and Xu (2018), who modified wood/HDPE (high-density polyethylene) composites (WF-HDPE) with 12% boric acid/borax mixture. The experimental results showed that, compared to the unmodified samples, the modified samples exhibited a 17.7% reduction in heat release, a 10.7% reduction in mass loss, and a 97.4% increase in pyrolysis residue, showing significant flame-retardant and smoke-suppressing effects. Similarly, Cui *et al*. (2016) found that the use of boric acid/borax (3:2 mass ratio) for flame-retardant treatment of Yunnan pine improved its LOI, providing excellent flame retardancy and smoke suppression effects.

The Z2 composite specimen specimens had an LOI of 29.2%, which was a 7.0% increase compared to the Z0 composite specimen. This can be attributed to the fact that ammonium polyphosphate (APP) is a highly phosphorus- and nitrogen-rich intumescent flame retardant with advantages such as low cost, low smoke, low toxicity, and low corrosion (Pan *et al*. 2020). When heated, the decomposition of APP releases non-flammable gases, such as ammonia and water vapor, which dilute the concentration of combustible gases generated by the burning material. The APP also promotes the rapid dehydration and carbonization of oxygenated organic compounds, forming a char layer that prevents heat from transferring into the material. Theses effects slow the thermal decomposition and combustion (Chen *et al*. 2024). This result was confirmed by Mao *et al.* (2023), who used a "core-shell-core" layering method to apply 12.5% APP to large particleboards. This procedure effectively improved the gas-phase flame-retardant effect of the boards, without largely affecting the condensed-phase flame-retardant performance.

The Z3 composite specimen specimens (with disodium octaborate tetrahydrate, DOT) had the highest LOI value of 34.3%, representing a 25.6% increase compared to the Z0 composite specimen, showing the best flame-retardant performance. This is because the boron oxide produced by the thermal decomposition of disodium octaborate tetrahydrate captures the free radicals produced during combustion, interrupting the chain reaction and effectively preventing the spread of combustion, thus considerably increasing the composite's LOI.

The composite flame-retardant modified Z4, Z5, and Z6 composite specimens showed LOI values of 23.8%, 18.3%, and 18.7%, all of which were higher than the Z0 composite specimen, respectively. This suggests that the action mechanisms of different flame retardants vary. The APP acts through a condensed-phase flame-retardant mechanism, DOT combines both condensed-phase and gas-phase flame retardancy, while BA/Brx utilizes both mechanisms (Yu *et al*. 2024). The use of blended flame retardants may result in synergistic effects, further enhancing flame retardancy (Yang *et al*. 2020; Zhang *et al*. 2021). However, the experimental results indicate that, under the conditions of this study, modification with DOT alone provided the best improvement in the flame-retardant performance of the composites.

Impact of Flame-retardant Systems on Composite Properties

To evaluate whether the differences among flame-retardant systems were statistically significant, a one-way ANOVA followed by Tukey's HSD test (α = 0.05) was performed using SPSS 27.0. The analysis confirmed that the flame-retardant treatments induced significant variations in both the mechanical properties and dimensional stability of the composites. The flame-retardant modification significantly affected the mechanical properties of the waste Tetra Pak/wood-shavings composites to varying degrees. Figure 2 presents the mechanical performance test results for six flame-retardant modified waste Tetra Pak/wood-shavings composites and the unmodified composites.

Regarding the elastic modulus, according to GB/T 4897 (2015), the MOE of dry particleboard should reach 1900 MPa. The Z0 composite specimen had an MOE of 1954 MPa, which meets the requirements for furniture-grade particleboard. After flame-retardant modification, only the Z3 composite specimen (1968 MPa) showed a slight increase compared to Z0, while the other composite specimens experienced a decrease ranging from 4.2% to 22.6%, failing to meet the national standard. Regarding static bending strength, all modified composite specimens had lower static bending strength compared to the Z0 composite specimen, with reductions ranging from 17.2% to 35.9%. Only the Z1 composite specimen had an MOR less than 11.5 MPa, failing to meet the standard for general-purpose particleboard under dry conditions, while the MOR of Z0 and Z3-Z6 composite specimens was greater than 12.0 MPa, meeting the requirements for furniture-grade particleboard. For internal bonding strength, the reduction for modified samples ranged from 22% to 47.5%, with only the Z3 and Z5 composite specimens still meeting the national standard for internal bonding strength in dry conditions for general particleboard (⩾0.30MPa).

The Z1 composite specimen (BA/Brx) treated samples showed a substantial decrease in mechanical strength, as the boron compounds may aggregate on the fiber surface, weakening the interface bonding strength of the composite. Additionally, the addition of boric acid/borax might interfere with the curing process and effectiveness of the adhesive, reducing the bonding strength and negatively affecting the overall mechanical properties of the composite (Wang *et al*. 2019; Chen *et al*. 2020). The mechanical performance of the Z2 composite specimen (APP) decreased due to the acidic nature of ammonium polyphosphate, which contains a large amount of phosphate ions in its molecular structure. These ions may react with phenol-formaldehyde resin, leading to incomplete curing and a crosslinked structure, preventing the adhesive from fully curing. Moreover, the poor interface compatibility between APP and the composite material resulted in a reduction in mechanical strength. In the Z3 composite specimen (DOT), the mechanical performance also declined due to the insufficient bonding strength between disodium octaborate tetrahydrate and the composite, which led to poor interface adhesion. The modified composite composite specimens (Z4, Z5, and Z6) exhibited varying degrees of changes in mechanical properties, which can be attributed to the interactions between different flame-retardant components and between the flame-retardant components and the composite material.

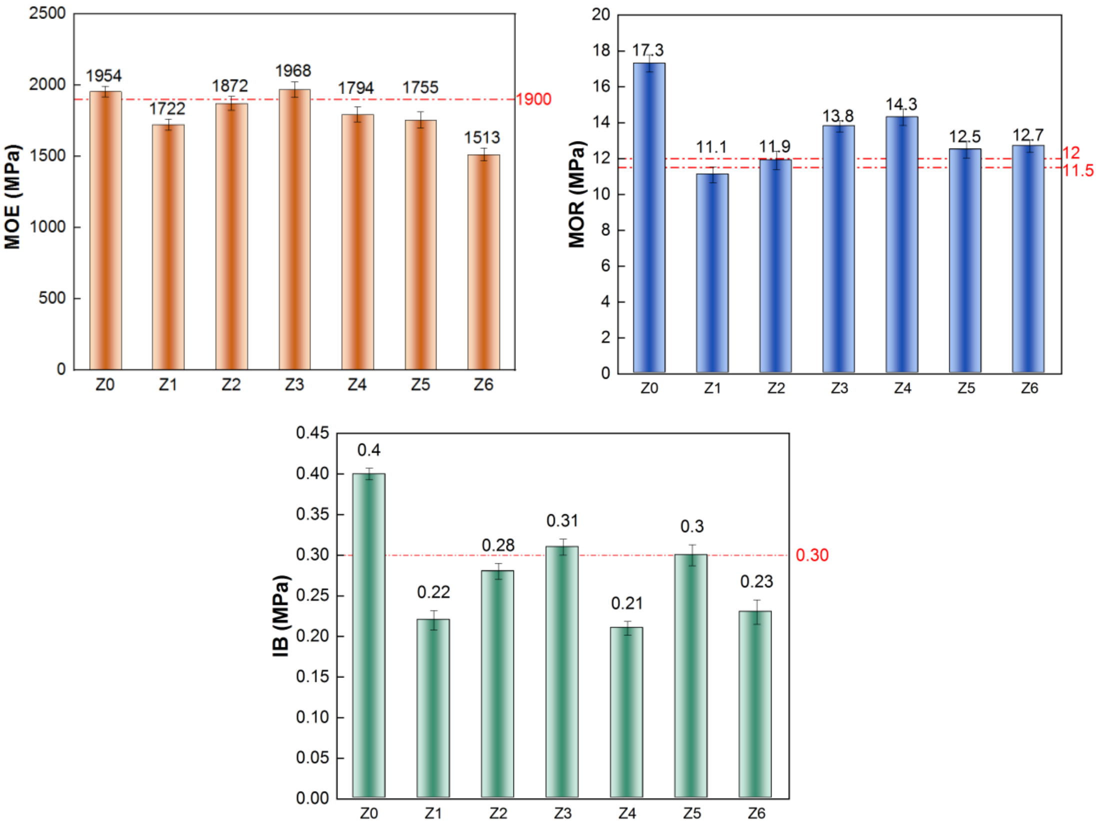


Fig. 2. Effect of different flame-retardant treatments on mechanical properties of composites (MOE, MOR, and IB)

The 24-h thickness swelling (TS) after water immersion of the composites under different flame-retardant systems is shown in Fig. 3. As can be observed, all flame-retardant treatments increased the 24-h thickness swelling values of the composites, with improvements ranging from 11.6% to 22.4%. This can be attributed to the presence of hydrophilic composite specimens (such as hydroxyl groups) in the molecular structures of boric acid/borax and disodium octaborate tetrahydrate, which can form hydrogen bonds with water molecules (Li *et al*. 2024). These hydrophilic groups facilitate water penetration during immersion, thereby increasing the extent of thickness swelling (Adebisi *et al*. 2023). Additionally, ammonium polyphosphate contains a large number of polar composite specimens that can easily attract water molecules. Furthermore, ammonium polyphosphate is prone to hydrolysis, and the resulting hydrolysis products further increase the material's hydrophilicity. The 24-h thickness swelling rates of the samples modified with hybrid flame retardants (Z4, Z5, and Z6) varied depending on the type and content of the flame retardants used.

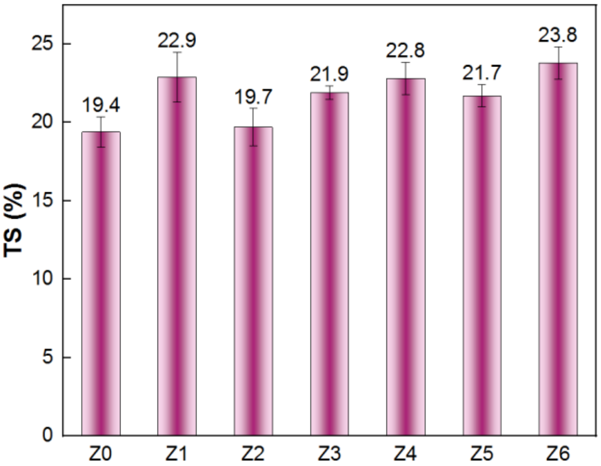


Fig. 3. Effect of different flame-retardant treatments on 24-h thickness swelling after water immersion

Evaluation of Flame-retardant Composites Based on LOI and Mechanical Properties

According to the flame-retardant classification standard (LOI ≥ 30%), four modified composite composite specimens- Z3 (DOT), Z4 (BA/Brx/APP), Z5 (BA/Brx/DOT), and Z6 (BA/Brx/APP/DOT)- met the flammability requirements, while Z1 (BA/Brx), Z2 (APP), and the unmodified composite specimen (Z0) did not meet this level.

Referencing the dry state usage requirements from GB/T 4897 (2015), the performance of each composite specimen was as follows: Only the Z3 composite specimen met the elastic modulus requirement for general-purpose particleboard, while the other flame-retardant modified composite specimens did not meet this standard. For static bending strength, all composite specimens except Z1 met the requirements for general-purpose particleboard, and Z3, Z4, Z5, and Z6 further met the performance requirements for furniture-grade particleboard. For internal bonding strength, only Z3 and Z5 met the requirements for general-purpose particleboard. Regarding the 24-h TS, Z2, Z3, and Z5 met the requirements for load-bearing particleboard, while the other composite specimens did not meet this standard.

Considering the comprehensive flame-retardant performance, mechanical properties, and hydrophobicity, the Z3 composite specimen treated with disodium octaborate tetrahydrate exhibited the best performance and met the main physical and mechanical property requirements for general-purpose particleboard in dry conditions.

Infrared Spectral Analysis

The infrared spectra of the Z2, Z3, Z4, and Z5 composite specimens, which exhibited superior flame-retardant and physical-mechanical properties, as well as the unmodified samples, are shown in Fig. 4. The absorbance peak at 3429 cm^{-1} corresponds to the bending vibration of O-H. In the Z4 composite specimen, boric acid contains hydroxyl groups, and the cellulose, hemicellulose, and other components in the waste Tetra Pak/wood-shavings composites also contain a large number of hydroxyl groups. During the flame-retardant modification, the hydroxyl groups in boric acid may form additional hydrogen bonds with the hydroxyl groups in the composite material or increase the strength of the existing hydrogen bonds. The formation of these hydrogen bonds alters the stretching vibration frequency of the hydroxyl groups, resulting in an increase in the intensity of the absorbance peak corresponding to the O-H stretching vibration near 3429 cm^{-1} for the Z4 composite specimen. The absorbance peak near 2920 cm^{-1} corresponds to the C−H bond, where reactions may occur between boric acid, borax, ammonium polyphosphate, and the organic components in the waste Tetra Pak/wood-shavings composites, generating new composite specimens or structures containing more C-H bonds. The peak near 1605 cm^{-1} is typically associated with bending vibrations of C=C, C=O, and N-H bonds. In the Z4 composite specimen, the hydroxyl composite specimens in boric acid may form hydrogen bonds with some polar composite specimens in the wood cell wall components (such as C=O, N-H), which can change the electron cloud distribution of the C=O or N-H bonds, leading to a shift in their bending vibration frequencies and an increase in the absorbance peak intensity.

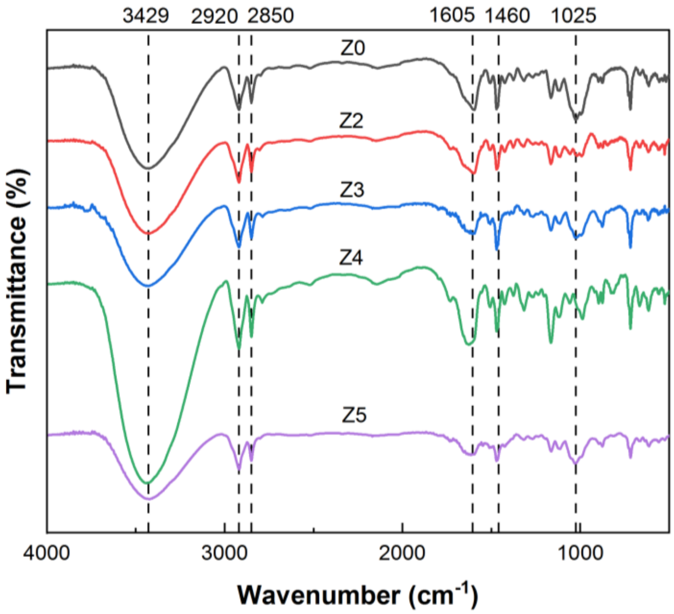
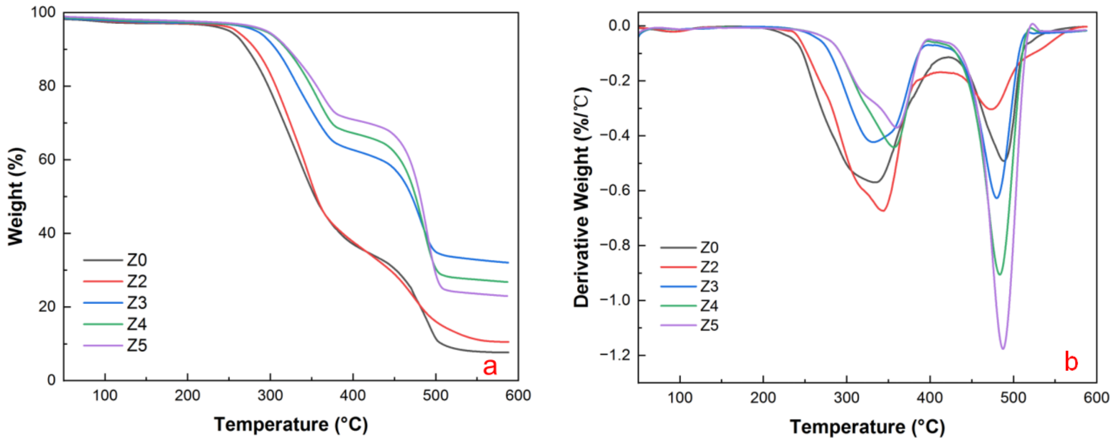


Fig. 4. Infrared spectrogram of unmodified and flame-retardant modified samples

In the Z5 composite specimen, the components may react with the active composite specimens in the phenol-formaldehyde resin, causing changes in the substituents on the benzene ring, which results in a decrease in the absorbance peak intensity of the benzene ring skeletal vibration and C=O bond stretching vibration. The characteristic peaks in the Z2 and Z4 composite specimens around 1100 to 1050 cm^{-1} are caused by the asymmetric stretching vibration of PO_4^{3-} in ammonium polyphosphate.

Thermogravimetric Analysis

The TG (a) and DTG (b) curves of the waste Tetra Pak/wood-shavings composites after different flame-retardant treatments, as well as the unmodified composites, are shown in Fig. 5. It can be observed that the thermogravimetric curve of the waste Tetra Pak/wood-shavings composites was approximately divided into three stages. Stage 1 (50 to 250 °C): This stage corresponds to the loss of water and other small molecules from the composite material, with hemicellulose in the wood shavings beginning to degrade. Stage 2 (250 to 350 °C): Hemicellulose in the wood shavings is nearly completely degraded, and cellulose, lignin, and other components start to undergo thermal decomposition, generating low molecular weight compounds such as sugars, acids, and alcohols. The paper fibers in the Tetra Pak also begin to decompose, producing volatile organic substances. Stage 3 (350 to 500 °C): Polyethylene plastic begins to decompose, and lignin and cellulose further degrade, generating a series of easily volatile carbonyl compounds.



(a) TG Curve **(b)** DTG Curve

Fig. 5. TG curves and DTG curves of unmodified and flame-retardant modified samples

The thermogravimetric characteristics of the waste Tetra Pak/wood-shavings composites before and after different flame-retardant treatments are presented in Table 4. It can be observed that all flame-retardant treatments improved the thermal stability of the waste Tetra Pak/wood-shavings composites. The initial decomposition temperature (T_0), the temperature at which the second stage of decomposition occurs most rapidly (T_{P2}), and the char residue at 600 °C for the Z2 composite specimen were all higher than those of the unmodified composites (Z0). This is because the added flame retardant APP began to decompose around 200 °C, and the acidic substances produced during decomposition reacted with the hydroxyl composite specimens in the cellulose molecules, thereby accelerating the dehydration and carbonization of cellulose, while releasing large amounts of ammonia and other non-flammable gases that suppress the combustion reaction.

The Z3 composite specimen exhibited the highest char residue at 600 °C, at 32.0%, which was 4.16 times that of the unmodified composites. The presence of DOT substantially increased the char residue of the composites, indicating that DOT promotes the formation of char. The multiple synergistic effects between the components of the hybrid flame retardants substantially improved the T_0, T_{P2}, T_{P3}, and char residue at 600 °C for the Z4 and Z5 composite specimens.

Table 4. Flame Retardant Test Results of Waste Tetra Pak/Wood Shavings Composites

Sample	T_0 (°C)	T_{P2} (°C)	T_{P3} (°C)	Char Residue at 600 °C (%)
Z0	238.8	335.2	486.6	7.7
Z2	242.3	343.3	474	10.5
Z3	272.6	333.1	480.5	32
Z4	288.4	356.9	484.3	26.8
Z5	289.8	360.4	487.3	23

Note: T_0 = Initial decomposition temperature; T_{P2} = Temperature at which the second stage of decomposition occurs most rapidly; T_{P3} = Temperature at which the third stage of decomposition occurs most rapidly)

CONCLUSIONS

1. The composite material prepared by replacing 30% of wood with discarded Tetra Pak cartons met the physical and mechanical requirements of GB/T 4897 (2015) for general-purpose particleboard, even without the addition of flame retardants. This confirms that discarded Tetra Pak cartons can serve as a usable additive or substitute to reduce the amount of fresh wood needed in wood-based composite manufacturing.
2. When various flame-retardant modification systems were applied to the discarded Tetra Pak/wood shaving composites, all six flame-retardant systems increased the LOI. Among them, formulations Z1 (BA/Brx), Z3 (DOT), Z4 (BA/Brx/APP), Z5 (BA/Brx//DOT), and Z6 (BA/Brx/APP/DOT) achieved LOI > 30%, reaching the flame-retardant level. The Z3 composite specimen modified with disodium octaborate tetrahydrate (DOT) exhibited the most effective flame-retardant performance.
3. Under the present experimental conditions, the Z3 (DOT) system demonstrated the most balanced overall performance, with an LOI of 34.3% (25.6% higher than the unmodified control). Its modulus of elasticity (1,968 MPa), modulus of rupture (9.5 MPa), internal bond strength (0.32 MPa), and 24-h TS (20.8%) all complied with the requirements of GB/T 4897 (2015) for general-purpose particleboard.
4. The FTIR and TG analyses clarified the flame-retardant mechanism. The DOT formed stable structures with the composite matrix through hydrogen bonding and B-O bond formation, thereby increasing the initial decomposition temperature to 272.6 °C and resulting in a char residue of 32.0% at 600 °C. Considering the trade-off between flame retardancy and mechanical performance, future research should optimize the formulation ratio of DOT with boron- and phosphorus–nitrogen-based flame retardants to achieve balanced performance and promote industrial application.

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