THERMAL ANALYSIS OF ORIENTAL BEECH WOOD TREATED WITH SOME BORATES AS FIRE RETARDANTS

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ABSTRACT

Wood is widely used as a construction material for many applications. To have knowledge about the thermal degradation characteristic of wood could be crucial for indoor and outdoor applications. Chemical treatments could improve the resistance of wood against fire and heat. In this study, to investigate the thermal properties of Oriental beech (Fagus orientalis) wood treated with 0,25%; 1% and 4,70% aqueous solutions of boric acid, borax, disodium octaborate tetrahydrate and mixtures of and in different proportions, thermogravimetric analysis, differential-thermogravimetry, and differential-thermal analysis were performed.

Our results showed that borate treatment decreased the $T_{\text{max}}$ (maximum degradation temperature) and increased residual char amount. Higher concentration levels of borates resulted in higher char content of Oriental beech wood specimens. Residual char content of borate treated wood samples at fixed concentration of 4,70% ranked as in the following order: disodium octaborate tetrahydrate > boric acid + borax (7:3) > boric acid + borax (1:1) > boric acid > borax > Control. The highest residual char content was achieved for the sample impregnated with 4,70% disodium octaborate tetrahydrate among the all treated samples.

Keywords: Borates, differential-thermogravimetry, differential-thermal analysis, oriental beech, residual char, thermogravimetric analysis.

INTRODUCTION

Among the construction materials which are used by people wood holds a special place because of its impressive range of attractive properties, including low thermal extension, low density, and high enough mechanical strength (Bektha and Niemz 2003, Fabiyi and Ogunleye 2015, Elaieb et al. 2015). However, wood and wood composites being a carbon based material can burn when subjected to flame source or direct heat in suitable conditions (Yalinkılıç et al. 1998). In order to reduce flammability and provide safety, wood is treated with fire-retardant chemicals (Nussbaum 1988, Ellis and Rowell 1989, Mitchell 1993). It is well known that boron compounds work efficiently as fire retardant chemicals for cellulosic materials. The fire resistance properties of boron containing compounds were surveyed by many scientists and proven to be effective since the late 19th century. The fire retardant effectiveness of boron compounds is a physical mechanism achieved by the formation of a coating or protective layer on the wood surface at high temperature, and by forming glassy films that may inhibit

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mass transfer of combustible gases (LeVan and Tran 1990, Wang et al. 2004). Borates have several advantages as wood preservative in addition to imparting flame retardancy, they can provide protection against wood destroying organisms and have a low mammalian toxicity and low volatility. Moreover, they are colorless and odorless (Hafizoglu et al. 1994, Murphy 1990, Yalinkilic et al. 1999, Drysdale 1994, Chen et al. 1997). Boric acid (BA) and borax (BX) are the most common boron compounds which have found many application areas in the wood preservation industry (Baysal 2002). Boric acid and borax mixtures have some efficacy in retarding flame spread on wood surfaces. In addition to the usual char-forming catalytic effect, they have a rather low melting point and form glassy films when exposed to high temperatures in fires. Borax tends to reduce flame spread but can promote smoldering or glowing. On the other hand, boric acid suppresses smoldering but has little effect on flame spread. Therefore, these compounds are normally used together (LeVan and Tran 1990).

Wood and treated wood with flame were subjected to thermal degradation using differential thermal analysis (DTA) and thermogravimetry (TG) techniques and the values of different kinetic parameters of wood and modified wood can be obtained from degradation study (Gao et al. 2004). Thermal degradation of wood products is separated in three stages. Firstly; below 200 °C, water and volatile components of the wood evaporate. Secondly; main constituents of the wood (hemicelluloses, celluloses, and lignin) degrade between 200–378 °C. Finally, nonvolatile and noncombustible part of the wood are converted into tar and char, up to 600 °C, in third stage (Beall and Eickner 1970, Tomak et al. 2012).

Quality and quantity of volatile compounds from thermal degradation can also be used as an analytical tool for characterizing different woods. The composition of volatiles reveals the effect of inorganic salts used for fire protection. Therefore, the analytical results are useful for basic burning studies and the development of fire retardants (Faix et al. 1990). The study of the pyrolysis process of fire-retardant treated wood may aid to understanding of the mechanism of fire-retarding. Basic studies employing thermal analysis of fire-retardant treated wood indicated that some fire retardant chemicals promote wood decomposition at lower temperatures; thus, direct the composition to carbon dioxide and water instead of intermediate flammable tar formation (Hirata and Kawamoto 1990). Because of the lowering of the decomposition temperature, there is a possibility that the charring rate of wood treated with certain fire-retardant chemicals might be greater than of untreated wood. An increased amount of char could provide additional surface insulation and also inhibit volatilization of the wood beneath during exposure (Baysal 2002). Yunchu et al. (2000) investigated TG-DTA of Cunninghamia lanceolata wood untreated and treated with flame-retardants. Results showed that upon lowering the pyrolysis temperature, wood weight loss was reduced and char yield increased in the charring phase of the pyrolysis. Karastergiou and Philippou (2000) studied TGA of particleboards treated with some fire retardants. They found that fire retardants altered the reaction of thermal decomposition of particleboard. More specifically, retardant chemicals increased the amount of char residuals and reduced the volatile, combustible vapors. Tomak et al. (2012) investigated thermal behavior of borate treated Scots pine wood. They found that weight loss of wood was reduced while char yield was increased and pyrolysis temperature was reduced in the boron preservative treated wood. Baysal (2002) found that decomposition started earlier for BA impregnated Scots pine than untreated wood and proceeded at lower temperature levels.

Beech wood is economically valuable and important natural source for many counties. So, the improvement of the thermal properties of the beech wood and its product are crucial for many applications. In literature, the effect of concentration of wood preservatives including borates on thermal degradation and the degradation mechanism of treated Oriental beech wood have not been investigated yet. The aim of this research is to increase the knowledge of effects of borates concentration on thermal properties of Oriental Beech wood. For the purpose of evaluation of the thermal behavior of Oriental beech wood impregnated with 0,25%; 1%, and 4,70% aqueous solutions of DOT, BA, BX, and mixtures of BA and BX (1:1 and 7:3;weight:weight) wood preservatives done by thermogravimetry analysis (TGA), differential thermogravimetry (DTG), and differential thermal analysis (DTA). The thermal properties of Oriental beech wood treated with (0,25%; 1%, and 4,70%) chemicals including boron were compared with the non-treated Oriental beech wood called as control and the literature.
MATERIALS AND METHODS

Materials

Boric acid (BA), borax (BX) and disodium octaborate tetrahydrate (DOT) were obtained from Eti Mine Works General Management in Turkey. Sapwood of Oriental beech (Fagus orientalis L.) timber free of knots, excessive cross-grain and other obvious defects was obtained from Yucel Wood Products, Mugla located in the South West region of Turkey. Oriental beech timber was machined into narrow strips. The strips were carefully chosen for having the same annual ring width and then cut into small pieces prior to milling. Wood flour was prepared by grinding the small wood pieces in a Wiley to pass a 50 mesh screen. Before impregnation, the specimens were conditioned at 65% relative humidity and 20 °C for two weeks.

Impregnation method

Aqueous solutions of the wood preservatives having concentration of 0,25%; 1%, and 4,70% were prepared using distilled water for the impregnation procedure. Wood flour samples of approximately 100 g were immersed in the solutions at 60 °C for 2 h. The treated samples were subsequently dried at 60 °C until they had the unchanged weight. Similar impregnation procedure of wood flour and wood samples is described in TG and DTA studies on fire retardant treated wood by Jiang et al. (2010) and Yunchu et. al. (2000). The treated samples were then moisture conditioned for two weeks at 20 °C and 65% relative humidity.

Thermal analysis

Differential thermal analysis (DTA), thermogravimetry analysis (TGA), and differential thermogravimetry (DTG) were carried out under argon at a heating rate of 10 °C/min and a purge rate of 50 mL/min using a LABSYS TG-DTA analyzer (France). The temperature was heated from the room temperature up to 600 °C. During the heating and pyrolysis of about 10 mg of sample, the weight loss was monitored continuously. Onset and inflection temperatures of the pyrolysis were recorded by the analyzer for each treatment group. The rate of weight loss as a function of time was derived from TG curve resulting in a derivative TG curve.

RESULTS AND DISCUSSION

The temperature of the initial weight loss of pyrolysis (T_i), maximum degradation temperature (T_max), and residual char are given in Table 1. Figures 1-5 show the TG curves, first derivative of TG curves (DTG) and DTA curves of BX, BA, BA+BX (1:1), BA+BX (7:3) and DOT impregnated Oriental beech wood.

As seen in Figures, for the untreated Oriental beech wood, below 200 °C, there were no significant changes in the TG and DTG curves, but from the DTA curves, nearly at 110 °C endothermic peaks corresponds to drying of the wood by the evaporation of physically adsorbed water was observed. For the untreated sample, between 231–350 °C sharp decrease in TG curves could be observed because of degradation of cellulose, hemicelluloses and lignin (Sinha et al. 2000). It was reported that the hemicelluloses decomposed easily to form gaseous products (CO, CO2, condensable vapors, and etc.) between 200-280 °C (Sinha et al. 2000). Degradation of the hemicelluloses also causes the organic acid especially acetic acid formation. Acetic acid formation accelerates the decomposition of polysaccharides by means of acting as depolymerization catalyst (Brosse et al. 2010, Esteves and Pereira 2008). In temperature range between 250-300 °C, the other main components of the wood products (lignin and cellulose) are degraded and gas, tar and char is produced during the pyrolysis (Pétrissans et al. 2014). As seen in Table 1, while residual char content was calculated as 33% for untreated sample, from DTG curves, the T_max was found as 353 °C for the untreated sample.
Table 1. $T_i$ (the temperature of the initial weight loss of pyrolysis), $T_{\text{max}}$ (maximum degradation temperature) and residual char of borates impregnated Oriental beech wood.

<table>
<thead>
<tr>
<th>Concentration (%)</th>
<th>$T_i$ (°C)</th>
<th>$T_{\text{max}}$ (°C)</th>
<th>Residual char (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control</td>
<td>-</td>
<td>231</td>
<td>353</td>
</tr>
<tr>
<td>BX</td>
<td>0,25%</td>
<td>251</td>
<td>332</td>
</tr>
<tr>
<td></td>
<td>1%</td>
<td>255</td>
<td>327</td>
</tr>
<tr>
<td></td>
<td>4,70%</td>
<td>255</td>
<td>313</td>
</tr>
<tr>
<td>BA</td>
<td>0,25%</td>
<td>237</td>
<td>331</td>
</tr>
<tr>
<td></td>
<td>1%</td>
<td>276</td>
<td>322</td>
</tr>
<tr>
<td></td>
<td>4,70%</td>
<td>294</td>
<td>315</td>
</tr>
<tr>
<td>BA+BX (1:1)</td>
<td>0,25%</td>
<td>240</td>
<td>324</td>
</tr>
<tr>
<td></td>
<td>1%</td>
<td>251</td>
<td>323</td>
</tr>
<tr>
<td></td>
<td>4,70%</td>
<td>255</td>
<td>342</td>
</tr>
<tr>
<td>BA+BX (7:3)</td>
<td>0,25%</td>
<td>235</td>
<td>328</td>
</tr>
<tr>
<td></td>
<td>1%</td>
<td>237</td>
<td>287</td>
</tr>
<tr>
<td></td>
<td>4,70%</td>
<td>262</td>
<td>330</td>
</tr>
<tr>
<td>DOT</td>
<td>0,25%</td>
<td>244</td>
<td>324</td>
</tr>
<tr>
<td></td>
<td>1%</td>
<td>249</td>
<td>325</td>
</tr>
<tr>
<td></td>
<td>4,70%</td>
<td>267</td>
<td>332</td>
</tr>
</tbody>
</table>

There are many theories that proposed to explain the mechanism of fire retardancy of chemicals. These theories are classified into 6 categories: Barrier theories, thermal theories, dilution by noncombustible gases theories, free radical trap theories, increased char/reduced volatiles theories and increased char/reduced volatiles theories. Generally, fire retardancy mechanism of the chemicals could operate by the combination of these theories (LeVan and Rowell 1984). The most acceptable theory which is called as chemical theory suggests that the retardant chemicals operates by altering the pyrolysis of wood, increasing of char amount and causing the formation of less amount of volatile and combustible vapors (LeVan and Winandy 1990). The pyrolysis reactions generally occur at low temperatures for the treated wood than for the untreated wood (LeVan and Winandy 1990, Holmes 1977). Borax (BX) and boric acid (BA) are commonly used fire-retardant compounds which melt at lower temperature and form glassy films on surface of the wood during the pyrolysis. Figures 1 and 2 show the TG curves, first derivative of TG curves (TGA) and DTA curves of BX and BA impregnated Oriental beech wood with different concentrations (0,25%; 1%, and 4,70% ) respectively. As seen in Figure 1, for the BX impregnated samples, there was no weight loss under 200 °C observed in TG patterns similarly with the untreated samples. From Figure 1 (c), it was easily seen that the peaks correspond to evaporation of water was observed for all BX treated samples between 100-150 °C. As listed in Table 1, $T_i$ values of BX treated sample were higher than that of controls. Pyrolysis of BX impregnated samples started nearly at 255 °C. In contrary, the maximum degradation temperature of the BX treated samples ($T_{\text{max}}$) was lower than that of controls. The peak which appears at 353 °C in DTG (Figure 1b) curves of control sample shifted to the left for the BX treated samples. These results confirmed that the pyrolysis reaction was occurred at lower temperature for BX impregnated samples. Same results were also obtained in literature by Tomak et al. (2012) on the Scots pine treated with ammonium pentaborate (APB), ammonium tetrafluoroborate (AFB) and sodium tetrafluoroborate (SFB). $T_{\text{max}}$ values of BX impregnated samples were decreased with the increase of the BX concentrations. Also as listed in Table 1, residual char content of samples treated with BX increased proportionally with the raise in BX concentration. From the DTA curves (Figure 2c), between 110 and 150 °C endothermic peak related with desorption of water was observed. In addition to this peak, for the 4,70% BA impregnated sample, endothermic peak was observed at 157 °C. This peak might be related with the first dehydration of boric acid. As reported in literature, boric acid could be degraded even below 150 °C. Degradation of
BA could occur in two steps (Eq 1 and 2).

\[ 2H_3BO_3 \rightarrow 2HBO_2 + 2H_2O \] (1)

\[ 2HBO_2 \rightarrow B_2O_3 + H_2O \] (2)

When boric acid dehydrate is below 150 °C, the boric acid is transformed into metaboric acid (HBO₂). After this point, all water content of metaboric acid is removed and it transforms into boron oxide (B₂O₃). Although the crystalline form of boron oxide melts at 450 °C, amorphous boron oxide does not melt at a specific temperature. Also it was reported that the amorphous boron oxide softens at 325 °C (Sevim et al. 2006). Little peaks in DTA curves appeared between 300-400 °C may indicate the transition of amorphous B₂O₃ and reaction of amorphous B₂O₃ with the surface for BA impregnated samples. From DTG curves of BA impregnated samples (Figure 2b), it was easily seen that the degradation of the impregnated samples began at higher temperatures than the control sample. The temperature degree increased with the increase in the BA concentration. \( T_i \) values of the BA impregnated samples are listed in Table 1. Parallel results were obtained for BA impregnated samples with the BX impregnated samples for the residual-char values and \( T_{max} \) values. Residual char content raised with the increasing of BA concentration and \( T_{max} \) values decreased inversely proportional with the increasing of BA concentration. In literature, it is concluded that the amount of char produced during TG-DTA analysis of wood samples has good agreement with the degree of retardancy against fire (Tomak et al. 2012, Qu et al. 2011). The BA and BX treated Oriental beech produced considerably more char than the untreated samples. According to our findings, decomposition products of BA and BX formed protective layers on surface of the wood samples and increased the char content.

![Figure 1. TG curves (a), first derivative of TG curves (TGA) (b) and DTA (c) curves of BX impregnated Oriental beech (Fagus orientalis) wood with different concentrations (0.25% 1% and 4.70%).](image-url)
BA and BX were often used together for commercial application because of their synergetic effects against fire. It was reported that the BX has positive effects on inhibition of the surface flame spread but also, it has promotive effects on molding or glowing. Although, smoldering and glowing combustion can be restricted by using boric acid, it is insufficient for prevention of flame spread (LeVan and Rowell 1984). In this study, two different solutions including different proportions (1:1 and 7:3) were used to investigate the fire retardancy of mixture of BA and BX. Also, the performances of commercial compound, disodium octaborate tetrahydrate (DOT) were investigated. Figures 3-5 show the TG curves, first derivative of TG curves (DTG) and DTA curves of BA+BX (1:1), BA+BX (7:3) and DOT impregnated Oriental beech wood respectively. As seen in Figure 3 (a) and Figure 4 (a), under 200 °C, considerable weight loss was observed only for 4,70% BA and BX mixture impregnated samples in both proportions. Weight losses of the samples increased with the increasing of the proportion of BA. As mentioned before BA is decomposed even below 150 °C and form metaboric acid. These weight losses may be caused from the one mole water elimination of BA during the decomposition. From DTA curve (Figure 3c and Figure 4c) for both proportion and all concentration at temperature between 90-120 °C, endothermic peaks related with desorption of the water were seen. For the sample impregnated with 4,70% BA+BX (7:3) solutions, additional peaks were observed at 138 °C and 159 °C. It was thought that these peaks probably related with the decomposition of BA. From DTA curves of BA+BX impregnated samples in all concentration and for both proportions at temperatures between 300-400 °C, peaks related with the reactions of B2O3 and cellulosic materials during the pyrolysis and also related with the charring reactions. For BA+BX impregnated samples in all proportions and concentrations, pyrolysis started at higher temperature than the pyrolysis of control started. T1 values were found as 240, 251 and 255 °C for 0,25%, 1%, and 4,70% BA+BX (1:1) impregnated sample and 235, 237 and 262 °C for 0,25%, 1% and 4,70% BA+BX (7:3) impregnated sample, respectively. From DTG curves (Figure 3b and 4b) intensity of the peaks appeared between 230-360 °C decreased. This result was related with the decrease in degradation rate of the sample. The magnitude of decrease in degradation rate was proportional with the increase of the concentration of the solution for both, BA+BX (1:1) and BA+BX (7:3). For the samples exposed with the solutions up to 1% concentrations in both proportions, T1 values decreased with the increasing of concentration. But for the samples impregnated with 4,70% BA+BX in both proportion, T1 values shifted to higher temperature with respect to the that for samples exposed with solutions of 1% concentration. Also from DTG curves, two maxima point were obtained for the sample impregnated with 4,70% BA+BX (7:3) and the peak obtained in DTG curves of the sample impregnated with 4,70% BA+BX (1:1) was smother than that.
of others. It was thought that, these results originated from that the shading effects of multiple reaction steps were suppressed by addition of high concentration of BA+BX.

Pyrolysis of wood product is combinations of complex, multi-step and sequential elimination reactions. Some of these reactions occurred in non-treated samples was prevented and charring reactions was improved by using chemicals including borates. When the residual char content of the samples were considered, it was seen that the final char amounts increased with the increasing of concentration for both proportions. As seen in Table 1, the residual char amount increased from 33% of control samples to 56%, 57%, 64% and 67% of BA, BX, BA+BX (1:1) and BA+BX (7:3) treated samples with fixed concentrations (4.70%), respectively. This result showed that firstly, individually BA and BX impregnation on Oriental beech wood improved the resistance against fire with respect to the untreated samples, and secondly, synergistic effects of using BA and BX together advanced the retardancy against fire, with respect to the usage of BA or BX alone.

Figure 3. TG curves (a), first derivative of TG curves (TGA) (b) and DTA (c) curves of BA+BX (in proportions of 1:1) impregnated Oriental beech (Fagus orientalis) wood with different concentrations (0.25%; 1% and 4.70%).
Figure 4. TG curves (a), first derivative of TG curves (TGA) (b) and DTA (c) curves of BA+BX (in proportions of 7:3) impregnated Oriental beech (*Fagus orientalis*) wood with different concentrations (0.25%; 1% and 4.70%).

Figure 5 shows the TG curves (a), first derivative of TG curves (TGA) (b) and DTA (c) curves of DOT impregnated Oriental beech wood with different concentrations (0.25%; 1% and 4.70%). DOT is generally used for fungus and insect control. Among all other borates, it is the most soluble and it has the highest 67% boric oxide (B$_2$O$_3$) content per unit mass (Tondi *et al.* 2014, Tomak *et al.* 2012). As seen in Figure 5a, considerable weight loss under 200 °C could not be observed for DOT treated samples. As for the other samples treated with the borates, endothermic peak related with desorption of water was seen nearly at 70-130 °C in DTA curves (Figure 5c). From DTA curves (Figure 5c), peaks observed nearly at 310-390 °C was associated with the phase transformation of amorphous B$_2$O$_3$, charring reactions and degradation of cellulose for the DOT impregnated samples. Intensity of these peaks increased with the increasing of DOT concentration. As listed in Table 1, $T_i$ values for the DOT treated samples shifted to higher temperatures with respect to untreated sample and $T_{max}$ values decreased from 353 °C of untreated samples to 324, 325 and 332 °C for the DOT treated samples with the concentration of 0.25%; 1% and 4.70% respectively. As shown in DTG curves (Figure 5b), intensity of the peaks observed between 230-400 °C, decreased with the increasing of concentration of DOT. Also from DTG curves, two maxima point were obtained for the sample treated with 4.70% DOT, as the same result obtained for the samples impregnated with 4.70% BA+BX (7:3). Residual char content of DOT impregnated samples was increased with the increasing of concentration of DOT. The highest residual char content (75%) was achieved for the sample impregnated with 4.70% DOT among the all treated samples.
Fire retardancy of borates were investigated by several researchers during the last decades (Tomak et al. 2012, Tondi et al. 2014, Salman et al. 2014, Yuksel et al. 2014, Istek et al. 2013, Roth et al. 2007, Jin et al. 2014, Qu et al. 2011). Parallel to the previous studies, our results showed that the impregnation of borates (BX, BA, BA+BX (1:1), BA+BX (7:3) and DOT) enhanced the resistance against fire. In general, amount of char produced during the pyrolysis of wood has good agreement with the fire retardancy properties of the chemicals treated on wood. For each compound and their mixture, weight loss of the wood sample is inverse proportional with the concentration of solutions used for impregnation and as listed in Table 1, residual char content of borate treated wood samples at fixed concentration of 4.70% ranked as in following order: DOT > BA+BX (7:3) > BA+BX (1:1) > BA > BX > Control. When the boron amount of the compounds per unit mass was considered, it was observed that the compounds were ranked in same order. BA, BX, and DOT include 17.48%; 11.34% and 20.96% boron, respectively. For these chemicals, having higher amount of boron increases the probability of forming thicker B$_2$O$_3$ shell on surface of the wood. According to our findings, in agreement with the barrier theory (LeVan 1984), it was thought that the degradation products of borates used in this study formed B$_2$O$_3$ glassy shell on Oriental beech wood and by this way, diffusion of combustible volatile side products was reduced. This non porous barrier also did not allow the access of oxygen into inner side of the wood promoted the anaerobic oxidation inside the wood. Same findings were achieved by several researchers. Usage of borates promotes the formation of CO and decrease CO$_2$ amount during the pyrolysis (Uysal and Ozciftci 2004). Also this inorganic non-combustible sheet acts as a mantle and preserves the wood from high temperatures. Because of the reasons mentioned before the char amount of treated Oriental beech wood increased with respect to untreated sample. It was thought that the degree of retardancy against fire for fire retardants showed alteration according to their probability of forming thicker B$_2$O$_3$ sheet.
CONCLUSIONS

In this study, thermal behavior of Oriental beech wood treated with 0.25%; 1% and 4.70% aqueous solutions of BX, BA, BA+BX(1:1), BA+BX(7:3) and DOT was investigated by thermogravimetry analysis (TG), differential thermogravimetry (DTG), and differential thermal analysis (DTA) under argon atmosphere.

Our results showed that fire-retardant including boron decreased the $T_{\text{max}}$ (maximum degradation temperature) and increased residual char amount. Residual char content of the treated samples increased with the increase of concentration. Residual char content of borate treated wood samples at fixed concentration of 4.70% ranked as in following order: DOT > BA+BX(7:3)>BA+BX(1:1)>BA>BX>Control. Especially DOT showed excellent performance, when the residual char amount was taken into consideration. The residual char amount of 4.70% DOT impregnated sample increased from the 33% of control sample to 75%. The thermo-gravimetric analysis of the samples treated with fire retardant is efficient and effective way to test the performance of the chemicals against fire, but still combustion tests and/or limited oxygen index test (LOI) should be done for better understanding of exact performance of the fire-retardant chemicals. Also, advanced spectrometric and chromatographic techniques should be performed for characterizing gaseous pyrolysis products.

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