

# Improvement of Flame Retardancy in Phenolics and Paper-Sludge/Phenolic Composites

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**ABSTRACT:** Paper-sludge/phenolic composites were fabricated using a novolac-type phenolic resin and paper sludge. A phosphate flame retardant containing halogen (tris 2-chloroethyl phosphate) and inorganic flame retardant (aluminum trihydroxide) were introduced into neat phenolics and paper-sludge/phenolic composite in order to improve their flame retardancy. In addition, magnesium hydroxide and halogenated flame retardant were added into the paper-sludge/phenolic composite. The flame retardancies were estimated with the UL 94 test. To study the flame retardant mechanism, thermal analysis of the phenolics and the paper-sludge/phenolic composites were carried out using a thermogravimetric analyzer and a differential scanning calorimeter. The phosphated flame retardant and inorganic flame retardant both showed the flame retardant effect on the phenolics. However, the flame retardancy of the paper-sludge/phenolic composite was enhanced only by phosphated and halogenated flame retardants. This result is attributed to the fact that the flame retardancies of the phenolics and paper-sludge/phenolic composite depend on their heat capacities and decomposition behaviors. © 1998 John Wiley & Sons, Inc. *J Appl Polym Sci* 69: 2043–2050, 1998

**Key words:** paper sludge; phenolics; flame retardance

## INTRODUCTION

Phenolic resins have been used in various applications largely because of their thermal stability, high char yield, and moderate flame retardancy.<sup>1,2</sup> The introduction of a cellulosic material into phenolic resins improves their mechanical properties and reduces the cost due to the partial replacement of the phenolic resin.<sup>3</sup> Paper sludge, one of the cellulosic materials, is a by-product during paper manufacture. It is mainly composed of paper pulp and inorganic materials. Especially from the viewpoint of recycling, the application of paper sludge to phenolic composites is very important. The paper-sludge/phenolic

composite can be applied to construction materials such as home furnishings and domestic and industrial buildings. The mechanical properties of various boards available commercially are compared with that of paper-sludge/phenolic composite in Table I. This data shows that the flexural strength of paper-sludge/phenolic composite is superior to that of other boards. However, because paper sludge is very sensitive to flame, a paper-sludge/phenolic composite has flame retardancy inferior to that of phenolics. Therefore, the improvement of the flame retardancy in paper-sludge/phenolic composites is of great importance, in spite of the good flame retardancy of phenolic resin.

In general, the ideal flame retardant polymer system has been described as having a high resistance to ignition and flame propagation, low rate of combustion, low rate and amount of smoke gen-

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**Table I Comparison of Mechanical Properties of Various Panel Boards**

	Density (g/cm <sup>3</sup> )	Flexural Strength (kgf/cm <sup>2</sup> )
Hansol board	1.15	222.0
Cement-fiber board	1.60	235.2
Paper-sludge board	1.44	466.0

eration, low combustibility, and toxicity of product gases. In addition, retention of flame retardancy during use, acceptability in appearance and properties for specified end uses, no environmental or health safety impact, and little or no economic penalty are needed for the ideal flame retardant polymer system.<sup>4</sup>

The factors associated with polymer flammability have been recently reviewed by Pearce et al.<sup>5</sup> Fundamentally, the a four-step burning process is considered to be involved in polymer flammability: preheating, decomposition, ignition, and combustion and propagation. Flame retardancy involves the disruption of the burning process at one or more stages so that the process is terminated within an acceptable period of time, preferably before ignition actually occurs. When dealing with polymeric materials, the following general approaches to flame retardancy are available: design of the basic polymer so that exposure to heat and oxygen will not produce combustion, modification of existing polymers so that they exhibit satisfactory performance upon exposure to fire, and incorporation of flame retardants so that the resulting polymeric materials exhibit satisfactory performance upon exposure to fire.<sup>5</sup> The third approach was adopted in this study because it was the most expeditious means to provide flame retardancy.

Halogenated compounds, phosphorus compounds, inorganic compounds, and so on are used as additive-type flame retardants.<sup>6</sup> Halogenated flame retardants are postulated to function primarily by a vapor-phase flame inhibition mechanism through radical reaction. Phosphorus compounds are a typical example of flame retardants that increase the conversion of polymeric materials to a char residue during pyrolysis and reduce the formation of flammable, carbon-containing gases (condensed-phase mechanism). Especially because the char residue of the sample is measured by thermogravimetric analysis, the flame retardant mechanism of phosphorus compounds can be studied.<sup>4</sup> Inorganic compounds (aluminum

trihydroxide and magnesium hydroxide) are believed to lower the polymer surface temperature and retard pyrolysis of the polymer by their endothermic decomposition.

In relation to the burning process, inorganic compounds are effective in stage 1 (heating) and stage 2 (decomposition) rather than in later stages, increasing the possibility of arresting the burning process at earlier stages. On the contrary, halogenated flame retardants and phosphorus compounds are especially effective in stage 3 (ignition) and stage 4 (combustion and propagation) with earlier stages.<sup>5</sup>

In the light of preceding discussion, the object of this article was to study the effects of various flame retardants on the flammability of phenolics and the paper-sludge/phenolic composite in relation to the burning process. The UL 94 test was carried out for the measurements of flammability, and thermal analysis was performed for the study of the flame retardant mechanism.

## EXPERIMENTAL

### Materials

Novolac-type phenolic resin (Kangnam Hwasung Co., Phenolite KC-3060), which contained hexamethylenetetramine (HMTA) as a curing agent, was used in this study. Aluminum trihydroxide (British Aluminium Chemicals Ltd., BACO FRF 10) and magnesium hydroxide (Martinswerk, MAGNIFIN H5 GV) were used as the inorganic flame retardants. Tris 2-chloroethyl phosphate (TCEP) was used as a phosphate flame retardant and contained halogen (Samsung General Chemicals). It contains 9.5 wt % P and 32.5 wt % Cl. As halogenated flame retardants, ethylenebis(penta-bromodiphenyl) (EBPBD, SAYTEX 8010) was purchased from Asano Chemical. EBPBD containing 82 wt % Br has a 345°C melting point and a 370°C decomposition temperature. Paper sludge was supplied by Hansol Paper Co. Ltd. The physical properties and the composition of the paper sludge are shown in Tables II and III. Acetone (Oriental Chemical Industries) was used as a solvent without further purification.

### Preparing of Phenolics

A mixture of novolac-HMTA and flame retardant additives was milled by a hand-rolling technique. The phenolic resins with various proportions of

**Table II Physical Properties and Composition of Paper Sludge**

Properties	Content (wt %)
Water	65 ( $\pm 5$ )
Solid	35 ( $\pm 5$ )
Pulp in solid	64 ( $\pm 3$ )
Inorganic compounds in solid	36 ( $\pm 3$ )
Specific gravity ( $\text{g}/\text{cm}^3$ )	0.76 ( $\pm 0.06$ )
Fiber length (mm)	0.23 ( $\pm 0.05$ )

flame retardants were cured by compression molding. The curing profile of the phenolic resin is given in Figure 1. The total curing time of the phenolic resin was 55 min. After curing, the sample was cooled to room temperature at a given pressure.

### Preparation of Paper-Sludge/Phenolic Composites

A mixture of novolac-HMTA and aluminum trihydroxide was milled by a hand-rolling technique. This mixture was introduced into the paper sludge. In the TCEP, a solution of TCEP in acetone solvent was dispersed into the paper sludge. The resulting paper sludge was vacuum dried at 30°C to remove acetone and then mixed with novolac-HMTA.

Composites of paper-sludge/novolac-HMTA (in 3 : 1 wt : wt proportion) with various proportions of flame retardants were manufactured by compression molding similar to the curing profile of the phenolic resins.

### Measurements

Flammabilities of the phenolics and the paper-sludge/phenolic composite were assessed by UL 94 vertical burn test. The sample's dimension was  $125 \times 12.7 \times 3$  mm. The thermal properties of the phenolics and paper-sludge/phenolic composite were investigated with a TGA-7 Perkin–Elmer thermogravimetric (TG) analyzer in air. Sample weight was approximately 10 mg and heating rate was 20°C/min. The heat capacity of sample was

determined by a DSC-7 Perkin–Elmer differential scanning calorimeter.

## RESULTS AND DISCUSSION

### Flammability Studies by UL 94 Test

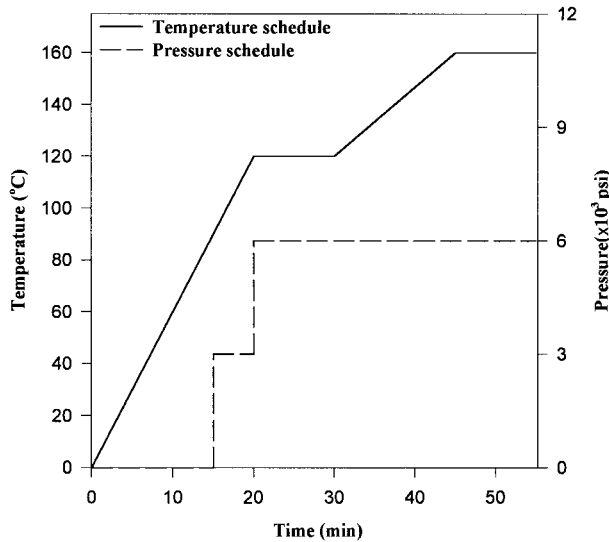
Flammability tests are classified according to various characteristics of fire response. Among the various flammability tests, the UL 94 test is carried out to estimate the degree of flame spread of the sample.<sup>5</sup> The degree of flame spread can be defined as the travel rate of the flame front under a given burning condition. Considering the burning process, the UL 94 test gives the information of whole stages in the burning process of the test specimen.

Neat phenolics have good flame retardancy and give a high char yield and good thermal property due to the aromatics of phenolic resins.<sup>2</sup> The effects of various flame retardant elements on the flammability of phenolic resins have been reviewed by Sunshine.<sup>7</sup> The results of the UL 94 test for the phenolics with the types and contents of flame retardants are represented in Table IV. In this article the burning time is defined as the sum of the burning time of five samples. The flammability of neat phenolics is classified as V-1. (UL 94 ratings are classified as V-0, V-1, and V-2 in order of increasing flammability.) Flame retardants decreased the burning time of neat phenolics below 40 s, which means enhanced flame retardancy (V-0 class). In the case of aluminum trihydroxide, the data show a drastic decrease of burning time at 5 wt %, but only slight changes of burning time above that value. In the case of TCEP, the increase of TCEP content gave a continuous decrease of burning time up to 10 wt %. These UL 94 test results indicate that TCEP and aluminum trihydroxide both have an effect on the flame retardancy of the phenolics.

Figure 2 shows the results of the UL 94 test for the paper-sludge/phenolic composites with the variation of the flame retardant content as burning time. The control paper-sludge/phenolic composite represents burning times longer than 300

**Table III Component of Inorganic Compounds in Solid (wt %)**

SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	Na <sub>2</sub> O	K <sub>2</sub> O	MnO	P <sub>2</sub> O <sub>5</sub>
51.42	31.83	2.69	1.23	2.51	8.15	0.95	0.48	0.06	0.65



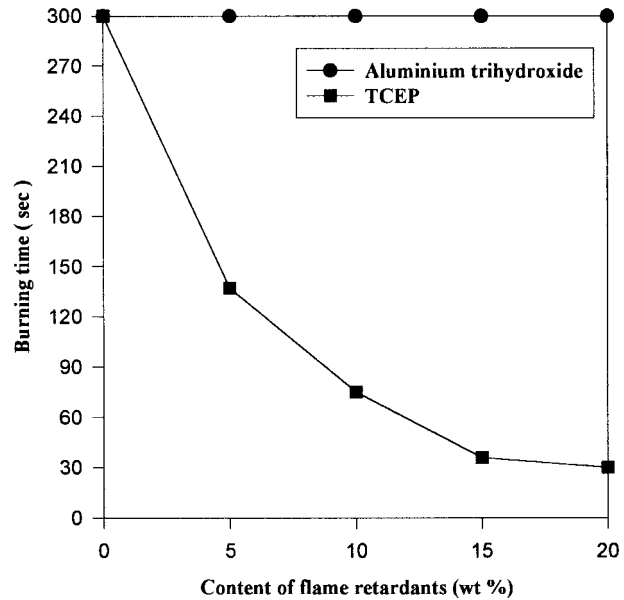
**Figure 1** Curing cycle in the preparations of the phenolics and the paper sludge/phenolic composite.

s. The introduction of TCEP into the composite gave a steep decrease in burning time. In the case of 5 and 10 wt % of TCEP, the paper-sludge/phenolic composites were classified as V-1. Above 15 wt % TCEP, the paper-sludge/phenolic composite yielded the flame retardancy of the V-0 class. For aluminum trihydroxide, burning times above 300 s were regarded as identical values because longer burning times than 300 s are of no value in the UL 94 test standard. Aluminum trihydroxide scarcely decreased the burning time of the paper-sludge/phenolic composites in comparison to the result of the TCEP.

The UL 94 test results for the paper-sludge/phenolic composite with magnesium hydroxide and EBPED are shown in Figure 3. The contents of the flame retardants were fixed at 20 wt %. The effects of these flame retardants were compared with those of aluminum trihydroxide and TCEP. Magnesium hydroxide represents a lower burning

**Table IV** Effect of Aluminum Trihydroxide and TCEP on Burning Time of Phenolics

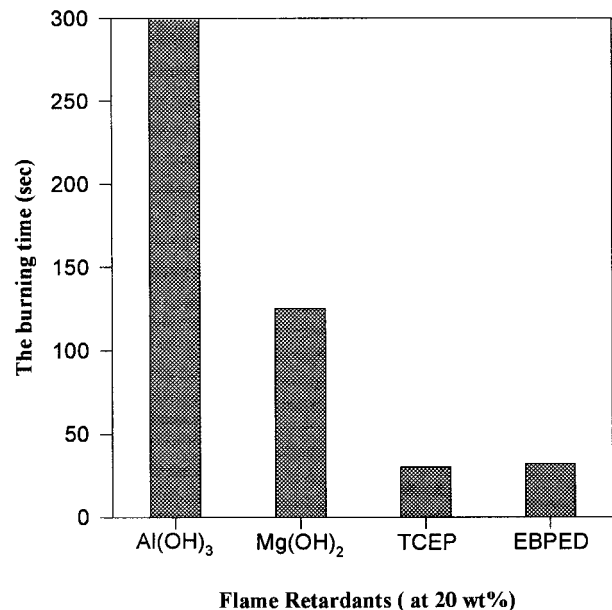
Additive Content (wt %)	Burning Time (s)	
	Al(OH) <sub>3</sub>	TCEP
0	56 (V-1)	56 (V-1)
5	13 (V-0)	32 (V-0)
10	10 (V-0)	9 (V-0)
15	3 (V-0)	—
20	7 (V-0)	—



**Figure 2** The effect of flame retardant content on the burning time of the paper-sludge/phenolic composite.

time than aluminum trihydroxide, but this UL 94 result corresponds to a V-1 class (125 s). However, the burning time of the paper-sludge/phenolic composite was decreased to 31 s by EBPED. This data was nearly similar to that of TCEP (V-0 class).

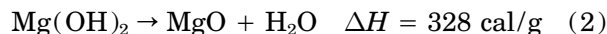
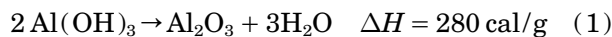
In the view of the results of the UL 94 test, the



**Figure 3** The comparison of flame-retardant effect on the paper-sludge/phenolic composite with 20 wt % flame retardant.

flame retardancy of the phenolics was improved by both the inorganic flame retardant and TCEP. However, in the paper-sludge/phenolic composite, TCEP and EBPED showed a flame-retardant effect. The inorganic flame retardant conclusively showed lower a flame retardant effect on the paper-sludge/phenolic composite than halogenated and phosphate flame retardants. The better flame retardant effects of TCEP and EBPED than inorganic flame retardants can be explained by their flame retardancy mechanism. That is to say, the flame retardancy of paper-sludge/phenolic composite was improved by both the condensed-phase and vapor-phase mechanisms due to phosphorus and halogen.

For the inorganic flame retardants, the flame retardant effect of magnesium hydroxide is superior to that of aluminum trihydroxide. This can be explained by the difference in their flame retardant mechanism. Generally, the flame retardant mechanism of inorganic flame retardants is the endothermic decomposition reactions as described in eqs. (1) and (2).

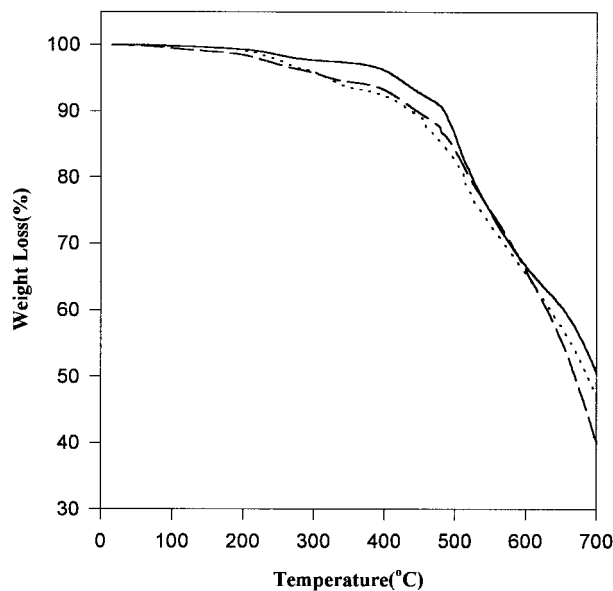


From the viewpoint of the heats of endothermic reaction ( $\Delta H$ ), magnesium hydroxide shows a higher value than aluminum trihydroxide. In addition, the decomposition reaction of aluminum trihydroxide occurs at 200–300°C and that of magnesium hydroxide occurs at 300–400°C. Therefore, the flame retardant effect of magnesium hydroxide was slightly higher than that of aluminum trihydroxide.

This conflicting flame retardant effects of aluminum trihydroxide on the phenolics and the paper-sludge/phenolic composite are described by the difference of burning progress rates between two materials. In paper-sludge/phenolic composite, the burning progress rate becomes rapid due to paper sludge, so the burning state rapidly reaches stages 3 and 4. Therefore, TCEP and EBPED, which have flame retardant mechanisms containing interference with stages 3 and 4, showed better a flame retardant effect on the paper-sludge/phenolic composite than inorganic flame retardants. Thermal studies will explain this flame retardant mechanism in detail.

### Thermal Studies

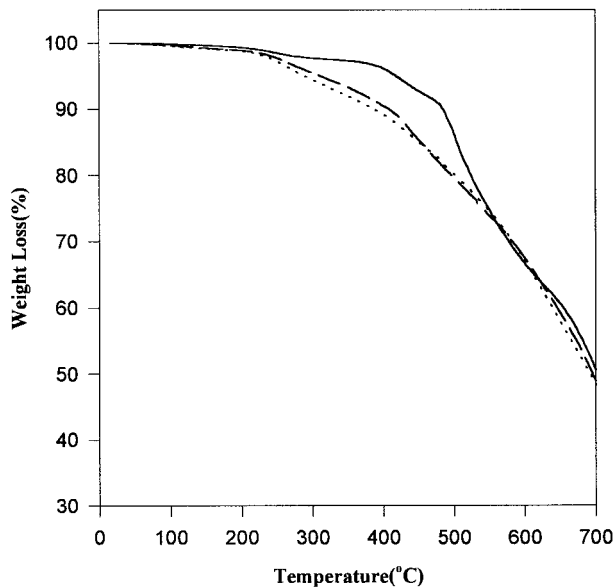
Figure 4 shows the TG curves of the phenolics as a function of aluminum trihydroxide content. The



**Figure 4** Thermogravimetric traces of the phenolics with (—) neat, (---) 5 wt %  $\text{Al}(\text{OH})_3$ , and (· · ·) 10 wt %  $\text{Al}(\text{OH})_3$  in air.

weight loss of the neat phenolics is marginal up to 300°C, but its decomposition rate becomes rapid at 400°C. This result is coincident with that reported by Conley and Jackson.<sup>8,9</sup> The phenolics with aluminum trihydroxide lose materials at a faster rate than neat phenolics. The char yields for the phenolics with aluminum trihydroxide, showed lower values than that of neat phenolics, as expected. This is due to the decrease in the content of phenolic resin, which gives a high char yield. This result clearly suggests that the improvement of the flame retardancy by aluminum trihydroxide is due not to char formation but to the endothermic decomposition in stages 1 and 2.

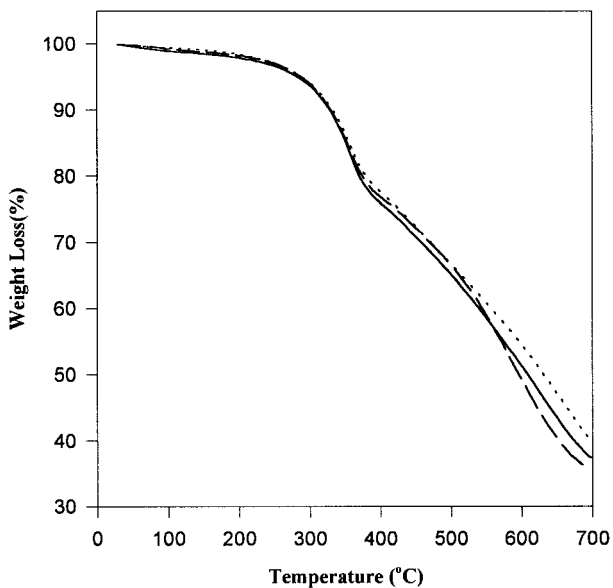
The TG curves of the phenolics modified with various TCEP contents are illustrated in Figure 5. The rapid degradation above 200°C is attributed to the initial splitting off of phosphated acid, as is commonly observed in phosphated flame retardant polymers.<sup>10</sup> At 700°C, the char yields of the phenolics with TCEP showed almost identical values with that of neat phenolics. Generally, the introduction of a phosphated flame retardant improves the char yield of the polymer.<sup>11</sup> This unexpected result may be associated with the flame retardant mechanism of chlorine contained in the TCEP.<sup>12</sup> In relation to halogen, it has been reported that the char is preferentially oxidized by some decomposition product of halogen compounds.<sup>13</sup> These flame retardant mechanisms pri-



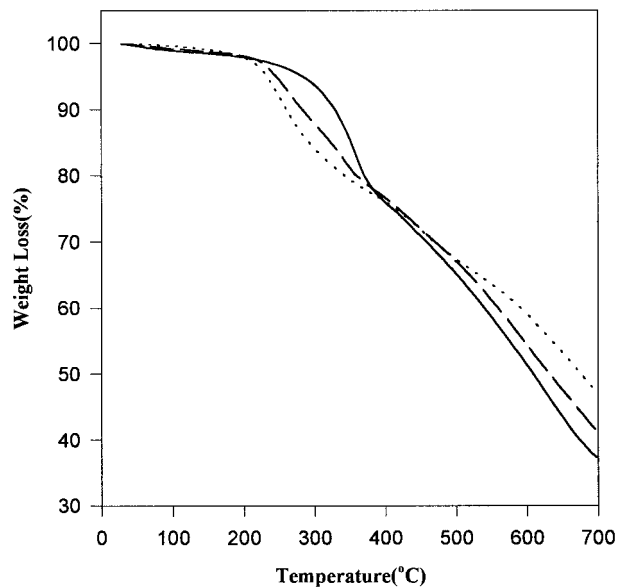
**Figure 5** Thermogravimetric traces of the phenolics with (—) neat, (---) 5 wt % TCEP, and (· · ·) 10 wt % TCEP in air.

marily contain interference with stage 2 in the burning process.<sup>5</sup>

Figure 6 shows the TG curves of the paper-sludge/phenolic composites as a function of the content of aluminum trihydroxide. In contrast to the TG curves of the phenolics, the neat composite and the composite with aluminum trihydroxide



**Figure 6** Thermogravimetric traces of the paper-sludge/phenolic composites with (—) neat, (---) 5 wt %  $\text{Al}(\text{OH})_3$ , and (· · ·) 10 wt %  $\text{Al}(\text{OH})_3$  in air.



**Figure 7** Thermogravimetric traces of the paper-sludge/phenolic composites with (—) neat, (---) 5 wt % TCEP, and (· · ·) 10 wt % TCEP in air.

showed a similar tendency in curve drop. At 300°C, the decomposition rate become rapid; above 350°C, it is alleviated. Figure 7 represents the TG curves of the paper-sludge/phenolic composites as a function of the content of TCEP. In the case of the phenolics modified with TCEP, the degradation rate becomes very rapid above 200°C.

The thermal data of the above TG analysis curves are summarized in Table V. The comparison of  $T_{10}$  (temperature at 10% weight loss) between the phenolics and paper-sludge/paper composites showed that the  $T_{10}$  values of the phenolics gave higher values than those of paper-sludge/phenolic composite. This difference in the TG curve behaviors between the phenolics and the paper-sludge/phenolic composite results from the paper sludge that mainly consists of pulp and inorganic materials. The incorporation of 5 wt % aluminum trihydroxide decreased the char yield of neat paper-sludge/phenolic composite a little. The char yield of the composite modified with 10 wt % aluminum trihydroxide was slightly increased in comparison with the neat composite. This increment was ascribed not to the char formation mechanism but to the aluminum residue. Generally, when aluminum residue at char forming temperature (probably 50 wt % of aluminum content) is considered, its residue content should be subtracted from the char yield of the sample with aluminum trihydroxide. The char yield of the

**Table V Thermal Properties of Phenolics and Paper-Sludge/Phenolic Composites Containing Flame Retardants**

Flame Retardants	Phenolics		Paper-Sludge/Phenolic Composite	
	$T_{10}$ (°C)	CY (wt %) at 700°C	$T_{10}$ (°C)	CY (wt %) at 700°C
Neat	484.94	50.69	328.65	37.09
5 wt % Al(OH) <sub>3</sub>	446.67	40.15	330.39	35.39
10 wt % Al(OH) <sub>3</sub>	438.74	46.93	332.86	39.52
5 wt % TCEP	408.57	48.90	282.93	41.07
10 wt % TCEP	387.04	48.31	260.73	46.90

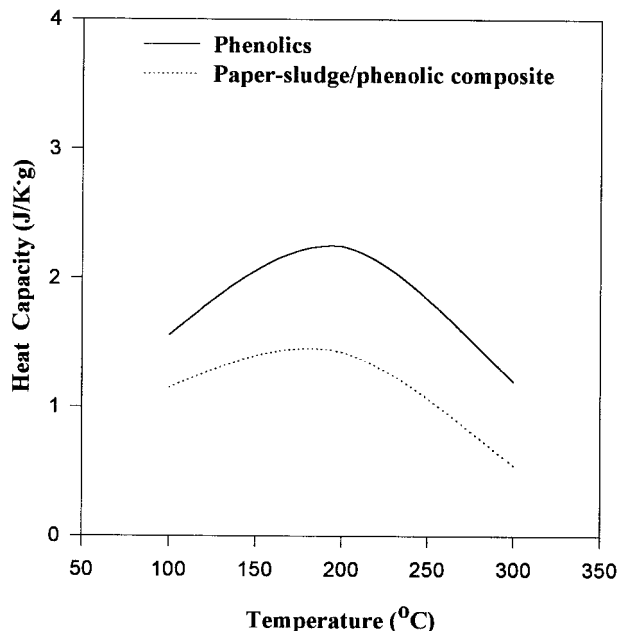
CY, char yield.

composite with 10 wt % TCEP at 700°C (46.9%) showed a more increased value than that for neat composite (37.1%). Thus, in the case of the paper-sludge/phenolic composite, the effect of incorporating phosphate flame retardants into the composite has remarkable TG curves (Fig. 7). This clearly shows the flame retardant effect of phosphorus by facilitating the formation of carbonaceous char (condensed-phase mechanism) on the paper-sludge/phenolic composite. This flame retardant mechanism essentially contains interference with stage 4 of the burning process.<sup>5</sup> This result is in accord with the improvements of flame retardancy of the paper-sludge/phenolic composite in UL 94 test. From the comparison of the flame retardant effects between aluminum trihydroxide and TCEP, it could be concluded that stage 4 is the primary stage in the burning process of the paper-sludge/phenolic composite.

On the other hand, it is important to know what produces the difference in the burning progress rate between the phenolics and the paper-sludge/phenolic composite. In this experiment, the involvement of paper sludge alters the thermal property of the phenolics. The two main factors affected by the addition of paper sludge are the decomposition behavior and the heat capacity of the system.<sup>5</sup>

Stage 1 depends on the flow rate of the applied heat and the heat capacity required to raise the temperature of the unit mass. Materials with high heat capacities increase more slowly in temperature than those with low heat capacities. Therefore, the burning progress rate (in stage 1) of materials with high capacity decreases. Heat capacities of the phenolics and the paper sludge/phenolic composite as a function of temperature are seen

in Figure 8. The difference between the two materials is 0.4 J/K g at 100°C and increases up to 0.8 J/K g at 200°C. This result shows that the burning state of paper-sludge/phenolic composite progresses more rapidly from stage 1 to the next stages than phenolics. As shown in Table IV, from the  $T_{10}$  values of the phenolics and the paper-sludge/phenolic composite, the decomposition phenomena of the paper-sludge composite occur more rapidly than those of the phenolics. The difference in the decomposition rate between the two materials is ascribed to the better internal ther-



**Figure 8** Heat capacities of the phenolics and the paper-sludge/phenolic composite as a function of temperature.

mal stability of the phenolic resin than that of the paper sludge. This decomposition behavior gives the different burning progress rates in stage 2 of the two materials.

These results can explain that stages 1 and 2 progress slowly in the phenolics, which have relatively high heat capacity and low decomposition rate. In contrast, due to their low heat capacity and rapid decomposition rate, the burning state of the paper-sludge/phenolic composite progresses rapidly from the front stage (stages 1 and 2) to the rear stage (stages 3 and 4). Therefore, inorganic flame retardants, which have flame retardant effects on stages 1 and 2, improved only the flame retardancy of the phenolics. Phosphate and halogenated flame retardants, which have flame retardant effects on stages 3 and 4, showed flame-retardant effects on both the paper-sludge/phenolic composite and the phenolics.

## CONCLUSION

From the UL 94 test and thermal analysis, the following conclusions were obtained. Aluminum trihydroxide showed a V-0 UL 94 test class in the phenolics, but it did not affect the flame retardancy of the paper-sludge/phenolic composite. Magnesium hydroxide represented the V-1 class in the paper-sludge/phenolic composite. In contrast to the inorganic flame retardant, TCEP increased the flame retardancies of both the phenolics and the paper-sludge/phenolic composite (V-0 class). Similarly, EBPED showed a V-0 class in the paper-sludge/phenolic composite. The conflicting effect of the inorganic flame retardant on the flame retardancies of the two materials is due to their heat capacities and decomposition behaviors. In relation to the burning process, stages 1 and 2 progress slowly in the phenolics, which have relatively high heat capacity and low decomposition rate. However, due to their low heat capacity and rapid decomposition rate, the burning state of paper-sludge/phenolic composite progresses

rapidly from the front stage (stages 1 and 2) to the rear stage (stages 3 and 4). Therefore, inorganic flame retardant, which is effective in stages 1 and 2, increased the flame retardancy of the phenolics but not that of the paper-sludge/phenolic composite. Phosphate flame retardant and halogenated flame retardant, which have flame retardant ability in stages 3 and 4, showed flame retardant effects on both the phenolics and the paper-sludge/phenolic composite.

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