



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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| <b>(54) Title:</b> FLAME RETARDANT COMPOSITION FOR POLYMERS   |           |   |
| <b>(57) Abstract</b>  |           |   |
| <p>The present invention is a flame retardant composition, adapted to be mixed with a polymer substrate to confer flame retardancy on the substrate, which comprises: (a) a bicyclic phosphorus flame retardant compound, such as one containing one or more pentaerythritol phosphate alcohol moieties, as exemplified by bis(pentaerythritol) phosphate alcohol carbonate; (b) an intumescent flame retardant compound containing nitrogen and phosphorus, such as melamine phosphate; and (c) a monophosphate ester compound to enhance the charring and processing characteristics of the composition in the polymer substrate, such as a liquid aryl-group containing phosphate ester compound, and the monophosphate ester compound is triphenyl phosphate.</p> |           |   |

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## FLAME RETARDANT COMPOSITION FOR POLYMERS

### 5 Background of the Invention

Flame retardant compositions comprising certain bicyclic phosphorus flame retardant compounds and certain intumescent flame retardant compounds which are suitable for use in a variety of polymers, including polyolefins, are disclosed in  
10 U.S. Patent No. 4,801,625 to W.J. Parr et al. While such compositions are useful as flame retardants for such application, persons of ordinary skill in the art have desired improved flame retardant compositions which have enhanced processing characteristics with the selected polymer they are intended to flame retard. Processing improvements include less porous pellets and/or a smoother surface  
15 characteristic when extruded. The present invention is directed to addressing these needs.

### Summary of the Invention

20 The present invention relates to a flame retardant composition, adapted to being mixed with a polymer substrate to confer flame retardancy on the substrate, which comprises: (a) a bicyclic phosphorus flame retardant compound; (b) an intumescent flame retardant compound containing nitrogen and phosphorus; and (c) a monophosphate ester compound to enhance the  
25 compatibility of the composition in the polymer substrate. The bicyclic phosphorus flame retardant compound can be a compound containing one or more pentaerythritol phosphate alcohol moieties, such as bis(pentaerythritol phosphate alcohol) carbonate. The intumescent flame retardant compound containing nitrogen and phosphorus can be melamine phosphate. The  
30 monophosphate ester compound can be a liquid aryl-group containing phosphate ester compound, such as isodecyl diphenyl phosphate, or can be a

solid aryl-group containing phosphate ester compound, such as triphenyl phosphate.

### Description of the Preferred Embodiments

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The present invention's essential components are, preferably, as described herein below.

### The Bicyclic Phosphorus Flame Retardant

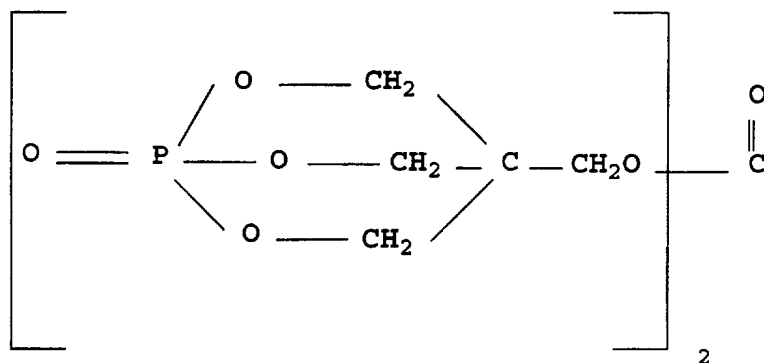
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Included within the definition of possible bicyclic phosphorus flame retardant compounds, which are solid materials, for use as one component of the flame retardant composition of the present invention are the following representative compounds:

15 Pentaerythritol phosphate alcohol itself, which forms a reagent to manufacture a number of the compounds to be described hereinafter, is one flame retardant which can be selected for use in connection with the present invention.

Bicyclic phosphate ether, ester, and carbonate flame retardants as described in U.S. Patent No. 4,801,625 to W.J. Parr et al., which is incorporated herein by

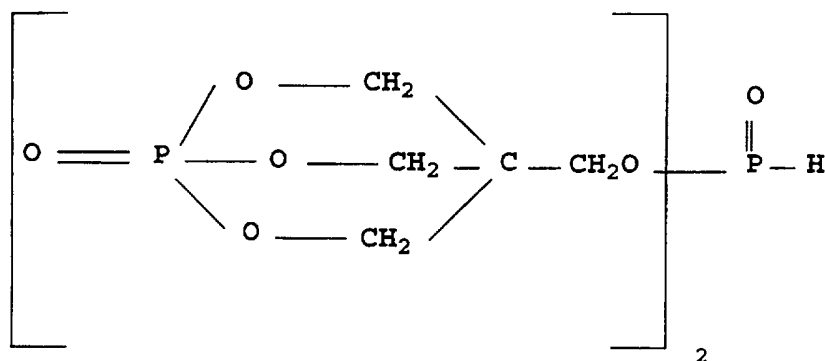
20 reference, can be employed and form a preferred class of such component(s) herein. A preferred species is bis(pentaerythritol phosphate alcohol) carbonate which is of the formula:



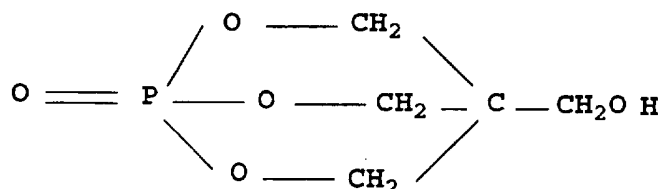
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Bis(pentaerythritol phosphate alcohol) hydrogen phosphonate may be used as the pentaerythritol phosphate alcohol-derived flame retardant additive in accordance with the present invention, and it is of the following formula and is described in U.S. Patent No. 5,420,326:

5



- One process for forming the above-described type of compound is by the transesterification of a diphenyl phosphite with pentaerythritol phosphate alcohol which has the formula:



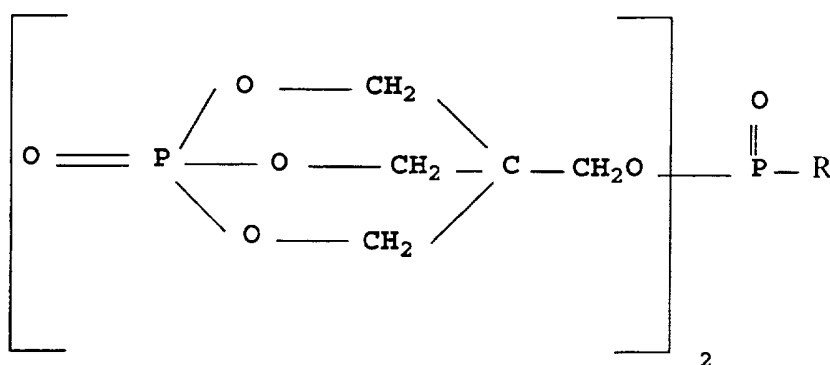
- The phosphite reagent is of the formula  $(\text{ArO})_2\text{P}(\text{O})\text{H}$ , where Ar is substituted or unsubstituted phenyl. The transesterification reaction is advantageously conducted at elevated temperature (e.g., a temperature of from about  $120^\circ\text{C}$  to about  $250^\circ\text{C}$ , preferably in a high boiling organic solvent, such as an aryl phosphate solvent (as described in U.S. Patent No. 5,237,085), using an appropriate transesterification catalyst (e.g., magnesium dichloride, sodium phenoxide, or the like). The reaction mixture will contain the desired crude product with a phenolic by-product. The crude product can be triturated with a solvent such as acetonitrile or

methanol to give the desired, purified product.

Bis(pentaerythritol phosphate alcohol) alkylphosphonate

compounds can also be used in the present invention as the pentaerythritol phosphate alcohol-derived flame retardant component. These compounds,

5 which are described in U.S. Patent No. 5,362,898, are of the formula:

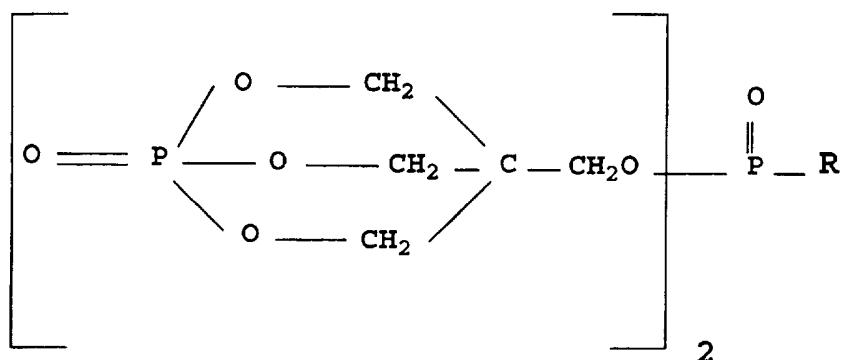


where R is alkyl, for example alkyl of from one to four carbon atoms, preferably  
 10 methyl. One process for forming the above described compounds is by the transesterification of a diphenyl alkyl phosphonate carrying the desired alkyl group with pentaerythritol phosphate alcohol which has been previously depicted above. The phosphonate reagent is of the formula (ArO)<sub>2</sub>P(O)R, where Ar is substituted or unsubstituted phenyl and R is alkyl as previously  
 15 described. The transesterification reaction is advantageously conducted at elevated temperature (e.g., a temperature of from about 170°C to about 200°C) in a high boiling organic solvent, such as an aryl phosphate solvent (as described in U.S. Patent No. 5,237,085), using an appropriate transesterification catalyst (e.g., magnesium dichloride, sodium phenoxide, or  
 20 the like). The reaction mixture will contain the desired crude product with a phenolic by-product. The crude product can be triturated with a solvent such as acetonitrile or methanol to give the desired, purified product. An alternative way of preparing these compounds is by the reaction of pentaerythritol phosphate alcohol, a trialkylamine, such as triethylamine, and an alkylphosphonic dihalide,  
 25 such as methylphosphonic dichloride, in an appropriate solvent, such as

acetonitrile, under cooling at essentially ambient temperature (e.g., 20°C to about 30°C).

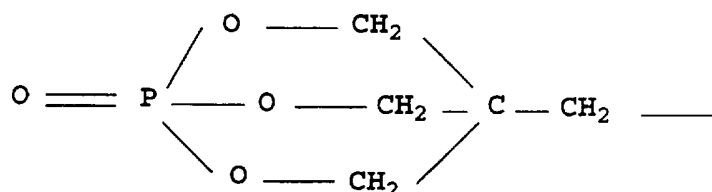
Another type of compound that can be used has the following formula and is bis(pentaerythritol phosphate alcohol) pentaerythritol phosphate alcohol

5 phosphonate and has the following formula:



where R in the above formula is derived from pentaerythritol phosphate alcohol

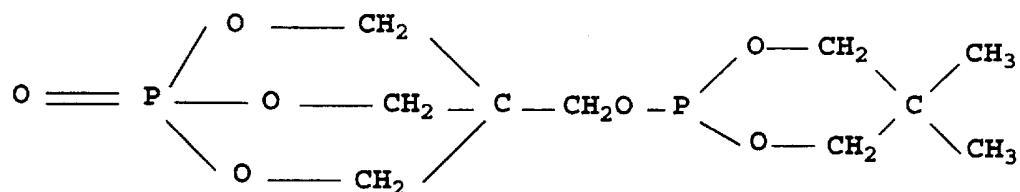
10 and is of the formula:



This compound can be formed by a known pentaerythritol phosphate alcohol-triaryl phosphite transesterification reaction followed by Arbuzov rearrangement.

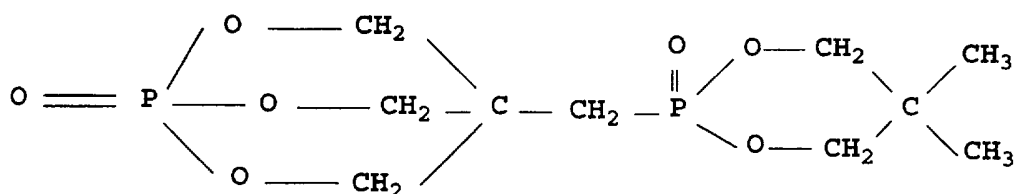
15 This invention also contemplates use, as the pentaerythritol phosphate alcohol-derived flame retardant, of the (pentaerythritol phosphate alcohol) (cyclic neopentyl glycol) phosphite compound of the formula:

6

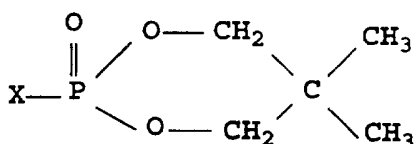


and the (pentaerythritol phosphate alcohol) (cyclic neopentyl glycol)

5 phosphonate compound of the formula:



10 One process for forming the above-described phosphite compound is by the reaction of a neopentylene glycol halophosphite with pentaerythritol phosphate alcohol which has been previously depicted hereinabove. The neopentylene glycol halophosphite reagent is of the formula:



15

where X is a halogen atom, such as chlorine. The reaction can be conducted at room temperature in an appropriate organic solvent, such as acetonitrile using an acid acceptor, such as a trialkylamine. Once the phosphite compound has been synthesized, it can be converted to the phosphonate compound of the present invention by heating, for example, in a high boiling organic solvent, such as an aryl phosphate solvent.

20



The above described type of flame retardant component can be used in the present invention of from about 1 % to about 30 %, by weight of the composition, preferably from about 5 % to 12 %.

#### 5 The Intumescent Flame Retardant

The type of intumescent flame retardant component which also forms a part of the present invention includes the intumescent phosphorus acid salts of amines or ammonia. They contain both phosphorus and nitrogen atoms. Included  
10 within this category of useful reagents for use herein are such phosphates as melamine phosphate (which is preferred), ethylenediamine phosphate, and cyanoguanidine phosphate, such polyphosphates as ammonium polyphosphate, and such pyrophosphates as ammonium pyrophosphate.

This type of intumescent flame retardant additive can be used in amounts  
15 ranging from about 5% to about 30 % by weight of the composition, preferably from about 15% to about 20%.

#### The Monophosphate Ester

20 The novel additive for the flame retardant composition of the present invention as compared to U.S. Patent No. 4,801,625 to W.J. Parr et al. is a monophosphate ester compound to enhance the compatibility of the known two-component flame retardant in the selected polymer to be flame retarded. Included as useful additives of this type are the aryl group-containing  
25 monophosphate esters which can be liquid (such as the alkyl diaryl phosphates, as exemplified by isodecyl diphenyl phosphate), or solid (such as triphenyl phosphate).

The amount of this monophosphate additive which can be used in the compositions of the present invention can range from about 1% to about 30%,  
30 by weight of the composition, preferably from about 5% to about 15%.

The compositions of the present invention can be formed by mixing the foregoing additives in such conventional mixing equipment as: single or multiple screw extruders; internal mixers, such as the Banbury mixer; or other melt blending plastic compounding devices.

- 5** The type of polymers to which the foregoing type of flame retardant can be added include the polyolefins, the polyamides, the polycarbonates, the polyacrylates, the polyesters, and the polyphenylene oxides. The weight amount of such flame retardant in such polymer can, in general, range from about 10% to about 30%, by weight of the polymer.
- 10** In addition to the flame retardant composition of the present invention, the selected polymer to be flame retarded can include one or more other functional additives known to persons of ordinary skill in the art of polymer compounding and processing, including fillers, antioxidants, lubricants, stabilizers, pigments, rheology modifiers, impact modifiers, and antistatic agents.

**15**

The present invention is further illustrated by the Examples which follow.

## EXAMPLES

The Examples which follow demonstrate that bicyclic phosphorus flame retardant materials in polypropylene (PP) composites have shown surprising significance to low level additions (less than or equal to about 2.5%) of a phosphate ester plasticizer (for example, isopropylphenyl diphenyl phosphate). Char forming effectiveness was enhanced an additional ten percent (as seen in cone calorimetry evaluations: ASTM-1354-90). Other flammability tests such as the UL-94/V-0 protocol, showed marked improvement from a dripping V-2 rating up to V-0 with composites containing the phosphate ester plasticizer (see Tables 1 and 2). This behavior was especially striking in the bis(pentaerythritol phosphate alcohol) pentaerythritol phosphate alcohol phosphonate additive ("BP<sup>3</sup>" in the Tables given below. The abbreviation "PEPA" standing for pentaerythritol phosphate alcohol). Combined with up to twenty five percent (by weight) of the isopropylphenyl diphenyl phosphate, the FR effectiveness was improved to the higher levels of performance.

The addition of the plasticizer improved the ductility and the tensile elongation of the composite. Also improved was the dispersion of additives in the composites. Surprisingly, plasticizer migration was not observed despite exposure to accelerated testing conditions (150° C) for over thirty days.

The Tables in the following Examples demonstrate the physical properties and flammability performance.

A series of flammability trials were performed, examining the effects of low level additions of an isopropylphenyl diphenyl phosphate, which is also termed "isopropylated phosphate" or "isopropyl diphenyl phosphate" in the tables which follow. Initial trials of bis(PEPA) PEPA phosphonate in polypropylene looked at the variation of FR adjuvants:

Table 1

| Formulations:                                    | #1   | #2   | #3   | #4   | #5   | #6   |
|--|------|------|------|------|------|------|
| PP-6323, polypropylene (Himont)                  | 66.2 | 68.2 | 71.2 | 68.2 | 66.2 | 61.2 |
| BP <sup>3</sup> / isopropylated phosphate (< 5%) | 13.5 | 11.5 | 11.5 | 13.5 | 11.5 | 11.5 |
| Melamine Phosphate                               | 18   | 18   | 15   | 15   | 21   | 21   |
| THEIC*   | 2    | 2    | 2    | 3    | 1    | 3    |
| Calcium Stearate                                 | 0.3  | 0.3  | 0.3  | 0.3  | 0.3  | 0.3  |
|  |      |      |      |      |      |      |
| UL-94 (1/16")                                    | Fail | Fail | Fail | Fail | Fail | Fail |

\*tris (hydroxyethyl) isocyanurate

5

Despite the high loading levels and significant amounts of phosphorus (up to about 2.9%) contributed from the intumescent flame retardant, little to no FR efficacy was seen.

10 Several levels of the isopropylphenyl diphenyl phosphate (PHOSFLEX 41P brand) were incorporated into BP<sup>3</sup> and were compounded again into polypropylene with the same adjuvants with the following results:

Table 2

| Formulations:   | #1        | #2   | #3   | #4   |
|---|-----------|------|------|------|
| PP-6323, polypropylene (Himont)                         | 66.2      | 68.2 | 71.2 | 68.2 |
| BP <sup>3</sup> /isopropylated phosphate<br>(about 15%) | 13.5      | -    | 13.5 | -    |
| BP <sup>3</sup> /isopropylated phosphate<br>(about 25%) | -         | 13.5 | -    | 13.5 |
| Melamine Phosphate                                      | 18        | 18   | 18   | 18   |
| THEIC   | 2         | 2    | 2    | 3    |
| Calcium Stearate  | 0.3       | 0.3  | 0.3  | 0.3  |
|   |           |      |      |      |
| UL-94 (1/16") Evaluation #1                             | V0/<br>V1 | V2   | -    | -    |
| UL-94 (1/16") Evaluation #2                             | -         | -    | V0   | V0   |

5 Tested in two concurrent evaluations, both studies showed marked flame retardant improvement over non-plasticized composites.

For further testing of the flame resistant effect from varying plasticizer dosage levels, the following formulations were prepared and evaluated in a cone calorimeter apparatus (ASTM 1354-90):

Table #3

| Formulations:                           | #1   | #2   | #3   | #4   | #5   |
|---|------|------|------|------|------|
| PP-6323 (Himont)                        | 69.8 | 69.8 | 69.8 | 69.8 | 69.8 |
| (BPC) <sup>1</sup>                      | 10   | -    | -    | -    | -    |
| (BPC-15% 41P) <sup>2</sup>              | -    | 10   | -    | -    | -    |
| (BPC-25% 41P) <sup>3</sup>              | -    | -    | -    | 10   | -    |
| (BP <sup>3</sup> -18% 41P) <sup>4</sup> | -    | -    | 10   | -    | -    |
| (BP <sup>3</sup> -25% 41P) <sup>5</sup> | -    | -    | -    | -    | 10   |
| Melamine Phosphate                      | 18   | 18   | 18   | 18   | 18   |
| Tris hydroxyethyl isocyanurate          | 2    | 2    | 2    | 2    | 2    |
| Calcium Stearate                        | 0.2  | 0.2  | 0.2  | 0.2  | 0.2  |

<sup>1)</sup> bis (PEPA) carbonate  
<sup>2)</sup> bis (PEPA) carbonate with 15 % isopropyl diphenyl phosphate (Phosflex<sup>®</sup> 41P)  
<sup>3)</sup> bis (PEPA) carbonate with 25 % isopropyl diphenyl phosphate (Phosflex<sup>®</sup> 41P)  
<sup>4)</sup> bis (PEPA) PEPA phosphonate with 18 % isopropyl diphenyl phosphate (Phosflex<sup>®</sup> 41P)  
<sup>5)</sup> bis (PEPA) PEPA phosphonate with 25 % isopropyl diphenyl phosphate (Phosflex<sup>®</sup> 41P)

Cone Calorimetry Evaluations of IFR/PP Composites Spiked with IP<sup>4</sup>

| Materials:                 | Heat Flux Rate kW/m <sup>2</sup> | Time of Ignit. (sec.) | Mass Loss (%) | Avg. Heat Release Rate (kW/m <sup>2</sup> ) | Avg. Peak Heat Release Rate (kW at Sec) | Peak Heat Release Rate (kW at Sec) | Effect. Heat of Combust (MJ/kg) (300 sec) | Total Heat (MJ/m <sup>2</sup> ) |
|----------------------------|----------------------------------|-----------------------|---------------|---|---|------------------------------------|---|---------------------------------|
| PP-6323 (Himont)           | 35                               | 58                    | N/A           | 297   | 484 / 180                               | 1705 / 160                         | 34.0                                      | 269                             |
| BPC, neat                  | 35                               | 110                   | 82.5          | 125   | 157 / 373                               | 340 / 176                          | 23.1                                      | 198                             |
| BPC/15% - 41P              | 35                               | 119                   | 81.1          | 128   | 159 / 373                               | 363 / 167                          | 22.3                                      | 194                             |
| BPC/25% - 41P              | 35                               | 109                   | 71.9          | 132   | 168 / 365                               | 383 / 176                          | 20.0                                      | 199                             |
| BP <sup>3</sup> /18% - 41P | 35                               | 131                   | 53.2          | 137   | 173 / 381                               | 394 / 179                          | 25.3                                      | 207                             |
| BP <sup>3</sup> / 5% - 41P | 35                               | 98                    | 50.2          | 141   | 186 / 349                               | 331 / 172                          | 30.2                                      | 209                             |

UL-94 and LOI Flammability Performance of IFR/PPs

| Materials:                       | UL-94 (1/16") | UL-94 (1/8") | LOI (O <sub>2</sub> %) |
|----------------------------------|---------------|--------------|------------------------|
| PP-6323, Polypropylene<br>Himont | -             | -            | -                      |
| BPC, Neat                        | V-0           | V-0          | 33.2                   |
| (BPC-15% 41P)                    | V-0           | V-0          | 33.4                   |
| (BPC-25% 41P)                    | V-0           | V-0          | 32.2                   |
| (BP <sup>3</sup> -18% 41P)       | V-0           | V-0          | 34.3                   |
| (BP <sup>3</sup> -25% 41P)       | V-0           | V-0          | 33.5                   |



Physical Property Performance of IFR/PPs

| Material:                       | Ten. Yield Str. (10 <sup>3</sup> psi) | Ten. Elong % | Flex. Str. 10 <sup>3</sup> psi | Flex. Mod. 10 <sup>5</sup> psi | Izod Imp. ft-lb/in | HDT 264 °C psi | MFI Cond. L |
|---------------------------------|---------------------------------------|--------------|--------------------------------|--------------------------------|--------------------|----------------|-------------|
| PP-6323, Polypropylene - Himont | 4.2                                   | 488          | 3.6                            | 1.6                            | 0.3                | 53.2           | 8.4         |
| BPC, Neat                       | 3.6                                   | 6            | 5.2                            | 2.9                            | 0.3                | 59.1           | 8.3         |
| (BPC-15% 41P)                   | 3.3                                   | 11           | 4.7                            | 2.5                            | 0.3                | 56.1           | 9.5         |
| (BPC-25% 41P)                   | 3.4                                   | 19           | 4.6                            | 2.4                            | 0.4                | 54.1           | 9.5         |
| (BP <sup>3</sup> -18% 41P)      | 3.3                                   | 13           | 4.9                            | 2.6                            | 0.3                | 69.2           | 6.5         |
| (BP <sup>3</sup> -25% 41P)      | 3.2                                   | 23           | 4.3                            | 2.4                            | 0.3                | 60.2           | 8.2         |

- 5 In the preceding Tables the following conclusions can be drawn:
- All samples shown in the table displaying the cone calorimetry data exhibited exceptional flame retardancy characteristics over virgin polypropylene. Of particular note is the mass loss (%) of these composites. A lower number implies a greater degree of char formation (namely, less mass loss). Of the
- 10 three BPC-containing composites, the mass loss decreased as the dosage of the plasticizer increased. In effect, a one percent increase of plasticizer in the overall composite appeared to be responsible for a ten percent increase in char formation.
- In the table showing the UL-94 and LOI data, all listed intumescent fire
- 15 retardant composites demonstrated excellent LOI (ASTM 2863) and UL flammability results.
- In the Table illustrating the physical property performance data, it was observed that, despite the presence of plasticizer in a number of the formulations, its effect upon the heat deflection results was minimal. All plasticizer-containing
- 20 composites showed better dimensional stability than the neat polypropylene resin (PP-6323).

In the Table listed below, three additional composites were fabricated and tested in which the bis(PEPA) carbonate ("BPC") was combined with three differing phosphate ester additives:

Table

5

| Formulations:   | #1   | #2          | #3          |
|---|------|-------------|-------------|
| PP-6323 (Himont)                                      | 69.7 | 69.7        | 69.7        |
| BPC <sup>1</sup> and 2-ethylhexyl diphenyl phosphate  | 10   | -           | -           |
| BPC <sup>2</sup> and t-butylphenyl diphenyl phosphate | -    | 10          | -           |
| BPC <sup>3</sup> and triphenyl phosphate              | -    | -           | 10          |
| Melamine Phosphate                                    | 18   | 18          | 18          |
| Calcium Stearate                                      | 0.3  | 0.3         | 0.3         |
| UL-94 (1/16")   | V-O  | V-O/<br>V-1 | V-O/<br>V-2 |

The above data indicate that the alkylated diphenyl phosphate-containing composite exhibited a better degree of flame retardancy than the other two composites that were tested.

10

The forgoing Examples, which have been set forth for illustrative purposes only, should not be construed in a limiting sense for that reason. The scope of protection sought is set forth in the Claims which follow.

We Claim:

1. A flame retardant composition, adapted to be mixed with a polymer substrate to confer flame retardancy on the substrate, which comprises: (a) a bicyclic,  
5 pentaerythritol phosphate alcohol-derived phosphorus flame retardant compound; (b) an intumescent flame retardant compound containing nitrogen and phosphorus; and (c) an aryl group-containing monophosphate ester compound to enhance the processing of the composition in the polymer substrate.
- 10 2. A flame retardant composition as claimed in Claim 1 wherein the bicyclic phosphorus flame retardant compound is a compound containing one or more pentaerythritol phosphate alcohol moieties.
- 15 3. A flame retardant composition as claimed in Claim 1 wherein the bicyclic phosphorus flame retardant compound is bis(pentaerythritol phosphate alcohol) carbonate.
- 20 4. A flame retardant composition as claimed in Claim 1 wherein the intumescent flame retardant compound containing nitrogen and phosphorus is melamine phosphate.
- 25 5. A flame retardant composition as claimed in Claim 1 wherein the bicyclic phosphorus flame retardant compound is a compound containing one or more pentaerythritol phosphate alcohol moieties and the intumescent flame retardant compound containing nitrogen and phosphorus is melamine phosphate.
- 30 6. A flame retardant composition as claimed in Claim 1 wherein the bicyclic phosphorus flame retardant compound is bis(pentaerythritol phosphate

alcohol) carbonate and the intumescent flame retardant compound containing nitrogen and phosphorus is melamine phosphate.

- 5 7. A flame retardant composition as claimed in Claim 1 wherein the monophosphate ester compound is a liquid aryl-group containing phosphate ester compound.
- 10 8. A flame retardant composition as claimed in Claim 1 wherein the bicyclic phosphorus flame retardant compound is a compound containing one or more pentaerythritol phosphate alcohol moieties, the intumescent flame retardant compound containing nitrogen and phosphorus is melamine phosphate, and the monophosphate ester compound is a liquid aryl-group containing phosphate ester compound.
- 15 9. A flame retardant composition as claimed in Claim 1 wherein the bicyclic phosphorus flame retardant compound is bis(pentaerythritol phosphate alcohol) carbonate, the intumescent flame retardant compound containing nitrogen and phosphorus is melamine phosphate, and the monophosphate ester compound is a liquid aryl-group containing phosphate ester compound.
- 20 10. A flame retardant composition as claimed in Claim 1 wherein the monophosphate ester compound is triphenyl phosphate.
- 25 11. A flame retardant composition as claimed in Claim 1 wherein the bicyclic phosphorus flame retardant compound is a compound containing one or more pentaerythritol phosphate alcohol moieties, the intumescent flame retardant compound containing nitrogen and phosphorus is melamine phosphate, and the monophosphate ester compound is triphenyl phosphate.

12. A flame retardant composition as claimed in Claim 1 wherein the bicyclic phosphorus flame retardant compound is bis(pentaerythritol phosphate alcohol) carbonate, the intumescent flame retardant compound containing nitrogen and phosphorus is melamine phosphate, and the monophosphate ester compound is triphenyl phosphate.
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# INTERNATIONAL SEARCH REPORT

Int: onal Application No

PCT/EP 97/02159

|  |   |                       |   |   |
|--|---|-----------------------|---|---|
| <b>A. CLASSIFICATION OF SUBJECT MATTER</b><br>IPC 6 C08K5/521 C08K5/3492 C08L23/10   |   |                       |   |   |
| According to International Patent Classification (IPC) or to both national classification and IPC  |   |                       |   |   |
| <b>B. FIELDS SEARCHED</b>  |   |                       |   |   |
| Minimum documentation searched (classification system followed by classification symbols)<br>IPC 6 C08K  |   |                       |   |   |
| Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched  |   |                       |   |   |
| Electronic data base consulted during the international search (name of data base and, where practical, search terms used)   |   |                       |   |   |
| <b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>  |   |                       |   |   |
| Category *   | Citation of document, with indication, where appropriate, of the relevant passages  | Relevant to claim No. |   |   |
| A  | EP 0 069 500 A (BORG-WARNER) 12 January 1983<br>see claims 1-5; examples 1,4,43; table 1<br>---   | 1,2,4,5               |   |   |
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| <input type="checkbox"/> Further documents are listed in the continuation of box C.  |   |                       |   |   |
| <input checked="" type="checkbox"/> Patent family members are listed in annex.   |   |                       |   |   |
| * Special categories of cited documents :  |   |                       |   |   |
| <table style="width: 100%; border: none;"> <tr> <td style="width: 50%; border: none; vertical-align: top;">                     *A* document defining the general state of the art which is not considered to be of particular relevance<br/>                     *E* earlier document but published on or after the international filing date<br/>                     *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)<br/>                     *O* document referring to an oral disclosure, use, exhibition or other means<br/>                     *P* document published prior to the international filing date but later than the priority date claimed                 </td> <td style="width: 50%; border: none; vertical-align: top;">                     *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention<br/>                     *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone<br/>                     *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.<br/>                     *&amp;* document member of the same patent family                 </td> </tr> </table> |   |                       | *A* document defining the general state of the art which is not considered to be of particular relevance<br>*E* earlier document but published on or after the international filing date<br>*L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)<br>*O* document referring to an oral disclosure, use, exhibition or other means<br>*P* document published prior to the international filing date but later than the priority date claimed | *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention<br>*X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone<br>*Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.<br>*&* document member of the same patent family |
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| Date of the actual completion of the international search<br><br><p style="text-align: center; font-size: 1.2em;">27 August 1997</p>   | Date of mailing of the international search report<br><br><p style="text-align: center; font-size: 1.2em;">05.09.97</p>   |                       |   |   |
| Name and mailing address of the ISA<br>European Patent Office, P.B. 5818 Patentlaan 2<br>NL - 2280 HV Rijswijk<br>Tel. ( + 31-70) 340-2040, Tx. 31 651 epo nl,<br>Fax: ( + 31-70) 340-3016   | Authorized officer<br><br><p style="text-align: center; font-size: 1.2em;">Engel, S</p>   |                       |   |   |

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| International Application No<br><b>PCT/EP 97/02159</b> |
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