

TITLE OF THE INVENTION

Thermoplastic resin composition

BACKGROUND OF THE INVENTIONField of The Invention

5 The present invention relates to a thermoplastic resin composition. More particularly, the present invention is concerned with a resin composition which is substantially the same as that produced by a method comprising the step of mixing together (A) a thermo-
10 plastic resin and (B) at least one non-polymerizable liquid substance as an additive for the thermoplastic resin (A), the non-polymerizable liquid substance (B) being in a liquid state at a temperature in the range of from 25 to 300 °C, wherein the mixing is conducted
15 at a temperature in the range of from 25 to 300 °C, at which the non-polymerizable liquid substance (B) maintains a liquid state, thereby causing the thermoplastic resin (A) to be dissolved in or swelled with the non-polymerizable liquid substance (B). The resin composition
20 of the present invention is advantageous in that, during the production of the resin composition, each of the thermoplastic resin (A) and the non-polymerizable liquid substance as the additive, which are components of the resin composition, does not experience high-
25 temperature conditions so that each does not suffer

thermal deterioration, differing from a conventional resin composition produced by a melt-extrusion method. Therefore, by molding the resin composition of the present invention, a shaped article having good appearance, excellent light stability and excellent mechanical properties can be obtained. The resin composition of the present invention can be advantageously used as a molding material for producing various shaped articles, such as parts for office automation machines, parts for household electric appliances, and electric and electronic parts. Further, when the resin composition of the present invention is used as a masterbatch for preparing a molding material, wherein the masterbatch is added to a resin material which is the same as or different from the thermoplastic resin used in the resin composition, especially great advantages are brought about. The molding material obtained by adding the resin composition of the present invention as a masterbatch to a resin material has advantages not only in that it has improved melt moldability, but also in that, by virtue of the fact that the resin in the masterbatch has not experienced high-temperature conditions, a shaped article having good appearance, excellent light stability and excellent mechanical properties can be easily

produced from the molding material. Further, in the production of the masterbatch comprised of the resin composition of the present invention, a large amount of a liquid additive can be easily incorporated in the masterbatch without lowering the productivity of the masterbatch.

Prior Art

A shaped article produced by molding a thermoplastic resin, such as a polyphenylene ether resin, has excellent heat resistance and excellent impact resistance. Therefore, thermoplastic resins are widely used as a molding material for producing various shaped articles, such as parts for automobiles, parts for household electric appliances, and parts for office automation machines.

In recent years, for improving the properties of thermoplastic resins, various types of additives therefor have been used. For example, in order to improve the processability of a thermoplastic resin, it has been attempted to add a liquid paraffin (mineral oil) thereto. Further, in order to impart flame retardancy to a thermoplastic resin, it has been attempted to add a liquid organic phosphorus compound thereto. The addition of such a liquid additive to a thermoplastic resin is generally performed by using a melt-extrusion

method. However, when the addition of a liquid additive to a thermoplastic resin having a high melting temperature or a high glass transition temperature is performed by melt extrusion, problems arise not only in that special feeding equipment is necessary for feeding the liquid additive, but also in that the handling of the liquid additive is cumbersome. In addition, when a liquid additive having a low viscosity and a thermoplastic resin having a high viscosity are melt-mixed with each other, the large viscosity difference therebetween poses problems such that the extrusion rate of the thermoplastic resin is different from that of the liquid additive, so that the extrusion operation becomes unstable, and that a portion of the high-viscosity thermoplastic resin remains unmelted during the melt extrusion, leading to the occurrence of final shaped articles having poor appearance and low mechanical properties.

In order to solve the above-mentioned problems caused by the large viscosity difference between a high-viscosity thermoplastic resin and a liquid additive, a method has been proposed in which a resin material obtained by melt-blending a polystyrene resin and a polyphenylene ether resin is further melt-mixed with a liquid additive for a polymer (see Japanese

Patent No. 2612396 and Unexamined Japanese Patent Application Laid-Open Specification No. 5-295249).

This technique has made it possible to reduce the occurrence of the problem that a portion of a poly-phenylene ether resin, which is a high-viscosity resin, remains unmelted during melt extrusion. However, in this technique, when the polyphenylene ether resin and polystyrene resin are melted at high temperatures, problems arise such that the resins suffer marked discoloration, a lowering of light stability, and a lowering of mechanical properties due to the cleavage of molecular chains.

In order to solve these problems, various proposals have been made. Examples of these proposals include a method of using a resin composition comprising a thermoplastic resin and an oil-containing resin obtained by impregnating a liquid oil into a porous, styrene-divinyl benzene copolymer (Examined Japanese Patent Application Publication No. 7-122024); a method of preventing the occurrence of the bleeding-out of an additive from a molding material upon being molded, which comprises using, as a molding material, a resin composition comprising a thermoplastic resin and an additive for resin modification, wherein the additive is obtained by impregnating a liquid additive into an

oil absorptive crosslinked polymer (Unexamined Japanese Patent Application Laid-Open Specification No. 5-214114); a method of using, as a molding material, a resin composition obtained by incorporating a specific polymer into a thermoplastic resin so that a shaped article having a matte appearance can be obtained (Unexamined Japanese Patent Application Laid-Open Specification No. 6-57007); a method of using, as a molding material, a resin composition obtained by incorporating a specific polymer into a thermoplastic resin so that a shaped article having improved impact resistance can be obtained (Unexamined Japanese Patent Application Laid-Open Specification No. 6-73190); a method of using a powdery flame retardant for a polymer, which is obtained by impregnating a phosphorus-containing flame retardant into an amorphous silica (Unexamined Japanese Patent Application Laid-Open Specification No. 7-331244); a method of using an additive for a polymer, wherein the additive comprises an inorganic carrier and, carried thereon, a phosphorus-containing flame retardant (Japanese Patent No. 2588331); a method of using a resin composition obtained by a process comprising gelating a liquid additive with a gelating agent to obtain a coagulated liquid additive and incorporating the coagulated liquid

additive into a synthetic resin (Examined Japanese Patent Application Publication No. 56-42619); a method of using a resin composition obtained by dissolving a phosphate compound in a solvent to obtain a solution and dispersing the solution in polypropylene by melt-kneading (Unexamined Japanese Patent Application Laid-Open Specification No. 4-1233); and a method of using a masterbatch comprising an oil absorptive polymer and an additive for a polymer (Unexamined Japanese Patent Application Laid-Open Specification No. 9-52956). Each of these techniques aims to improve the handling property of a liquid additive by causing it to be absorbed into or adsorbed on an oil absorptive resin so as to decrease the apparent amount of the liquid additive. Therefore, in a resin composition obtained by any of these techniques, the oil absorptive resin having a liquid additive absorbed therein or adsorbed thereon is in a powdery or non-powdery solid state, so that, when such a resin composition is melt-molded by using an extruder, a shaped article having improved appearance and excellent mechanical properties cannot be obtained.

It has been known to use a resin composition produced by a method in which a thermoplastic resin is dissolved in or impregnated with a polymerizable monomer or a crosslinkable compound, and the polymerizable

monomer or crosslinkable compound is then polymerized. Examples of such a technique include a method of using a resin composition produced by a process in which polyvinyl chloride is impregnated with an aromatic vinyl monomer and the aromatic vinyl monomer is then polymerized (Examined Japanese Patent Application Publication Nos. 61-40702 and 62-1972, and Unexamined Japanese Patent Application Laid-Open Specification No. 3-197521); a method of using a resin composition produced by a process in which a polyphenylene ether resin and a rubber polymer are dissolved together in an aromatic vinyl monomer, and the aromatic vinyl monomer is then polymerized (European Patent No. 226149 and Unexamined Japanese Patent Application Laid-Open Specification No. 7-242791); a method of using a polyphenylene ether resin solution containing an aromatic vinyl monomer for forming a coating or a film (Russian Patent No. 2069674); a method of producing a heat-cured laminate sheet for use as an electric part, in which a polyphenylene ether resin, a liquid epoxy compound, a flame retardant and a curing catalyst are mixed with each other and the resultant mixture is subjected to a reaction by heating at a high temperature (Unexamined Japanese Patent Application Laid-Open Specification No. 7-3053); and a method of using a polyphenylene oxide

composition comprising a polyphenylene ether resin, a crosslinkable flame retardant and an initiator (Unexamined Japanese Patent Application Laid-Open Specification No. 62-148564). In each of these examples, a mixture of an additive and a solution of a resin is provided and the mixture is subjected to polymerization or other reactions. In such a method, it is relatively easy to obtain a composition in which a resin and a liquid additive are homogeneously mixed with each other. However, such a method has a disadvantage in that a polymerizable monomer is likely to be grafted on the thermoplastic resin, or the thermoplastic resin is likely to undergo a crosslinking reaction, so that the properties inherent in the thermoplastic resin change.

Further, Unexamined Japanese Patent Application Laid-Open Specification No. 50-50 discloses a method for producing a shaped article, in which a resin composition produced by dissolving a polyamide and a phosphate in formic acid as a solvent is used. In such method, however, after removal of the solvent from the resin composition, the polyamide and the phosphate are not homogeneously mixed with each other, so that it is impossible to obtain a resin composition exhibiting excellent appearance and excellent light stability. Other examples of known techniques include a method of

producing a porous polymer structure, in which a polymer, such as a polyphenylene ether resin, is dissolved in diphenyl sulfone as a solvent and the resultant solution is contacted with a mixed solvent of acetone and methanol (Unexamined Japanese Patent Application Laid-Open Specification No. 8-24582); a method of producing a polyphenylene ether-containing molding material, in which a polyphenylene ether resin and another polymer are dissolved in a solvent to obtain a solution and the solvent is removed from the solution in multiple stages (Unexamined Japanese Patent Application Laid-Open Specification No. 58-34829); and a method of producing a fiber from a solution obtained by dissolving polyacrylonitrile in a solvent composed of a solution of rhodanate (Unexamined Japanese Patent Application Laid-Open Specification No. 50-20029). However, in these known techniques, additives for a polymer are not used, so that a resin composition in which a resin and a liquid additive are homogeneously mixed with each other is not obtained. Also, there is known a method of lowering the melting temperature of a polyphenylene ether resin, in which a plasticizer is dissolved in a solvent to obtain a solution, and the obtained solution is used for treating a polyphenylene ether resin (Unexamined Japanese Patent Application

Laid-Open Specification No. 52-90596). However, in this method, since the polyphenylene ether resin is not dissolved in the solution, it is impossible to obtain a resin composition usable for producing a shaped article having excellent appearance, excellent light stability and excellent mechanical properties.

Further, it is known to use a resin composition produced by a method in which a resin, such as a polyphenylene ether resin, is dissolved in an additive for use in the surface modification of a polymer. For example, it is known to use an antistatic resin composition produced from a solution obtained by dissolving a thermoplastic resin, such as polyphenylene ether or polypropylene, in an amine antistatic agent (Unexamined Japanese Patent Application Laid-Open Specification No. 50-105293 and U.S. Patent Nos. 4,210,556, 4,314,040 and 4,147,742). As another example of such a technique, a method is known in which a lubricant oil and a synthetic resin together are dissolved in a solvent to obtain a solution and the solvent is removed from the solution, thereby obtaining a resin composition in which the lubricant oil is uniformly dispersed in the synthetic resin (Examined Japanese Patent Application Publication No. 51-34536). However, the additives (that is, an antistatic agent and a lubricant) used in

the above technique are intended to modify the surface characteristics of a shaped article. Therefore, in the above technique, it is important that the additives are primarily present on the surface portion of a shaped article. For this purpose, it is necessary that the additives have an appropriately low compatibility with the resin. Therefore, when a resin composition having a high content of an additive is intended to be produced by the above technique, it is necessary to melt-knead the resin and the additive at high temperatures, which cause the resin and the additive to be inevitably degraded. Accordingly, by molding a resin composition produced by the above technique, it is impossible to obtain a shaped article having an excellent appearance, an excellent light stability and excellent mechanical properties.

SUMMARY OF THE INVENTION

In this situation, the present inventors have made extensive and intensive studies with a view toward developing a resin composition which is free from the above-mentioned problems and can be advantageously used for producing various shaped articles having good appearance, excellent light stability and excellent mechanical properties. As a result, it has unexpectedly been found that a resin composition produced by a

specific method as mentioned below is free from the problems accompanying a resin composition produced by a conventional melt extrusion method, such as the heat deterioration of a resin contained in the resin composition and the like and, therefore, the resin composition of the present invention can be advantageously used for producing shaped articles which are much improved with respect to appearance, light stability and mechanical properties. The specific method involved in the present invention comprises the step of mixing together (A) a thermoplastic resin and (B) at least one non-polymerizable liquid substance as an additive for the thermoplastic resin (A), wherein the non-polymerizable liquid (B) is in a liquid state at a temperature in the range of from 25 to 300 °C and wherein the mixing is conducted at a temperature in the range of from 25 to 300 °C, at which the non-polymerizable liquid (B) maintains a liquid state, thereby causing the thermoplastic resin (A) to be dissolved in or swelled with the non-polymerizable liquid (B). It has also been found that when the resin composition of the present invention is used as a masterbatch for preparing a molding material, wherein the masterbatch is added to a resin which is the same as or different from the resin used in the resin composition, especial-

ly great advantages are brought about. That is, the molding material obtained by adding the resin composition of the present invention as a masterbatch to a resin material has advantages not only in that it has improved melt moldability, but also in that, by virtue of the fact that the resin in the masterbatch has not experienced high-temperature conditions, shaped articles which are improved with respect to the above-mentioned desired properties can be obtained from the molding material. It has further been found that, in the production of a masterbatch comprised of the resin composition of the present invention, a large amount of a liquid additive can be easily incorporated in the masterbatch without lowering the productivity of the masterbatch. The present invention has been completed, based on these novel findings.

Accordingly, it is a primary object of the present invention to provide a resin composition which can be advantageously used for producing a shaped article having a good appearance, an excellent light stability and excellent mechanical properties.

Another object of the present invention is to provide a masterbatch which is capable of easily imparting the desired excellent properties mentioned above to a shaped article produced from a molding

material obtained by adding this masterbatch to a resin material.

The foregoing and other objects, features and advantages of the present invention will be apparent from the following detailed description and claims
5 taken in connection with the accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

In the accompanying drawings:

Fig. 1 is a diagram showing one mode of the process for producing the resin composition of the present
10 invention;

Fig. 2 is a diagram showing another mode of the process for producing the resin composition of the present invention;

15 Fig. 3(a) and Fig. 3(b) show the ^1H -NMR spectrum of the polyphenylene ether resin composition produced by a melt extrusion method in Comparative Example 3;

Fig. 4(a) and Fig. 4(b) show the ^1H -NMR spectrum of the polyphenylene ether resin composition produced
20 by the method of the present invention in Example 1 (the positions indicated by A' to D' in Fig. 4(a) and Fig. 4(b) correspond to the positions indicated by A to D in Fig. 3(a) and Fig. 3(b), respectively);

Fig. 5 is a graph showing the relationship between
25 the production temperature and the color (yellowness

index: YI) with respect to each of the {polyphenylene ether (PPE)/a flame retardant TPP} compositions produced by melt extrusion method (Comparative Examples 10 to 12) and each of the {polyphenylene ether (PPE)/a flame retardant TPP} compositions produced by the method of the present invention (Examples 2 to 6);

Fig. 6 is a graph showing the relationship between the concentration of PPE in the resin composition and the dissolution temperature, in the production of each of the {PPE/non-polymerizable liquid substance} compositions in Examples 18 to 20 and Comparative Examples 29 to 31 {wherein the solid circle indicates that a flame retardant (TPP) is used as a non-polymerizable liquid substance and the open circle indicates that an antistatic agent (A-Stat) is used as a non-polymerizable liquid substance, and wherein each of the curved lines individually indicates the lower limit of the temperatures at which PPE completely dissolves into a non-polymerizable liquid substance selected from the flame retardant and the antistatic agent and, accordingly, the region on or above the curved line is a region of complete dissolution};

Fig. 7 is a graph showing the relationship between the concentration of the monomeric aromatic phosphoric ester (in a mixture of the monomeric and oligomeric

aromatic phosphoric esters) and the dissolution temperature, in the production of each of the (PPE/the mixture of monomeric and oligomeric aromatic phosphoric esters) compositions, wherein the curved line indicates the lower limit of the temperatures at which PPE completely dissolves into the mixture of the monomeric and oligomeric aromatic phosphoric esters and, accordingly, the region on or above the line is a region of complete dissolution (this graph corresponds to Table 6);

Fig. 8 is a graph showing the relationship between the content of a white pigment (TiO_2) in the resin composition and each of the color (yellowness index: YI) and the Izod impact strength, wherein each of the open circle and the open star indicates the property of the resin composition produced by melt extrusion method and each of the solid circle and the solid star indicates the property of the resin composition of the present invention (this graph corresponds to Table 15); and

Figs. 9(a) and 9(b) show a method for determining the glass transition temperature (T_g) and the melting temperature (T_m) from a chart obtained by differential scanning calorimetry(DSC).

In Fig. 1 and Fig. 2, like parts or portions are indicated by like reference numerals.

Description of Reference Numerals

- 1: Dissolution vessel
- 2: Inlet for a material
- 3: Agitator
- 5 4: Outlet for a liquid material
- 5: Extruder-type heating mixing vessel
- 6: Inlet for a liquid material
- 7: Inlet for a material containing a thermoplastic resin
- 10 8: Outlet for a resin composition
- 9: Pump
- 10: Filter
- 11: Device for preparing liquid drops
- 12: Belt-type cooler
- 15 13: First heat exchanger
- 14: Second heat exchanger
- 15: Pelletizer
- 16: Belt
- 17: Packaging device

20 DETAILED DESCRIPTION OF THE INVENTION

According to the present invention, there is provided a resin composition which is substantially the same as that produced by a method comprising the step of mixing together:

- 25 (A) a thermoplastic resin, and

(B) at least one non-polymerizable liquid substance as an additive for the thermoplastic resin (A), the non-polymerizable liquid (B) being in a liquid state at a temperature in the range of from 25 to 300 °C, wherein the mixing is conducted at a temperature in the range of from 25 to 300 °C, at which the at least one non-polymerizable liquid substance (B) maintains a liquid state,

thereby causing the thermoplastic resin (A) to be dissolved in or swelled with the non-polymerizable liquid (B).

For easy understanding of the present invention, the essential features and various embodiments of the present invention are enumerated below.

1. A resin composition which is substantially the same as that produced by a method comprising the step of mixing together:

(A) a thermoplastic resin, and

(B) at least one non-polymerizable liquid substance as an additive for the thermoplastic resin (A), the non-polymerizable liquid substance (B) being in a liquid state at a temperature in the range of from 25 to 300 °C,

the mixing being conducted at a temperature in

the range of from 25 to 300 °C, at which the at least one non-polymerizable liquid substance (B) maintains a liquid state,

5 thereby causing the thermoplastic resin (A) to be dissolved in or swelled with the at least one non-polymerizable liquid substance (B).

2. The resin composition according to item 1 above, wherein the at least one non-polymerizable liquid substance (B) is selected from the group consisting of a
10 flame retardant, a plasticizer, a heat stabilizer and a light stabilizer.

3. The resin composition according to item 1 or 2
15 above, wherein the thermoplastic resin (A) and the non-polymerizable liquid substance (B) are mixed together with (C) a solid substance as an additive for the thermoplastic resin (A), the solid substance (C) being a crystalline solid or an amorphous solid at a temperature of 300 °C or less.
20

4. The resin composition according to any one of items 1 to 3 above, wherein the mixing is conducted at a temperature in the range of from 100 to 300 °C, at
25 which the at least one non-polymerizable liquid sub-

stance (B) maintains a liquid state.

5. The resin composition according to any one of items 1 to 4 above, wherein the mixing is conducted at a temperature in the range of from 150 to 250 °C, at which the at least one non-polymerizable liquid substance (B) maintains a liquid state.

6. The resin composition according to any one of items 1 to 5 above, wherein the weight ratio of the thermoplastic resin (A) to the at least one non-polymerizable liquid substance (B) is from 10/90 to 90/10.

7. The resin composition according to item 6 above, wherein the weight ratio of the thermoplastic resin (A) to the at least one non-polymerizable liquid substance (B) is from 30/70 to 70/30.

8. The resin composition according to any one of items 1 to 7 above, wherein the thermoplastic resin (A) comprises 1 to 100 % by weight of a polyphenylene ether resin and 0 to 99 % by weight of a polystyrene resin, the total amount of the polyphenylene ether resin and the polystyrene resin being 100 % by weight.

9. The resin composition according to item 8 above,
 wherein the thermoplastic resin (A) comprises 50 to 99
 % by weight of a polyphenylene ether resin and 1 to 50
 % by weight of a polystyrene resin, the total amount of
 5 the polyphenylene ether resin and the polystyrene resin
 being 100 % by weight.

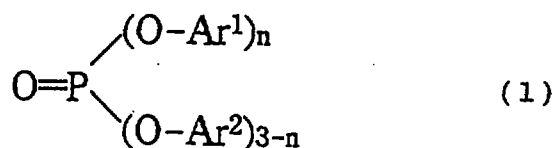
10. The resin composition according to any one of
 items 1 to 9 above, wherein the non-polymerizable
 10 liquid substance (B) is a flame retardant.

11. The resin composition according to item 10 above,
 wherein the flame retardant comprises at least one
 organic phosphorus compound.

12. The resin composition according to item 11 above,
 wherein the organic phosphorus compound is at least one
 aromatic phosphoric ester.

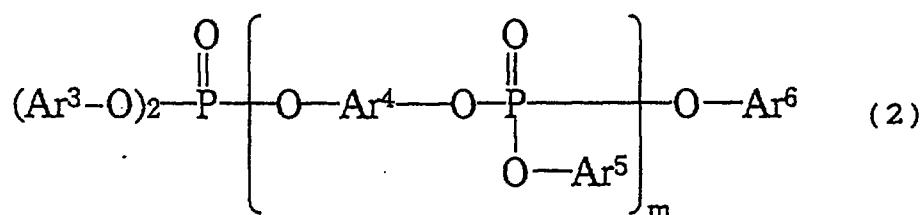
13. The resin composition according to item 12 above,
 wherein the aromatic phosphoric ester is a monomeric
 aromatic phosphoric ester represented by the following
 formula (1):

25



wherein each of Ar^1 and Ar^2 independently represents a $\text{C}_6\text{-C}_{20}$ aromatic group which is unsubstituted or substituted with at least one $\text{C}_1\text{-C}_{10}$ hydrocarbon group; and each n is identical to each other and represents an integer of from 0 to 3.

14. The resin composition according to item 12 above, wherein the aromatic phosphoric ester is an oligomeric aromatic phosphoric ester represented by the following formula (2):

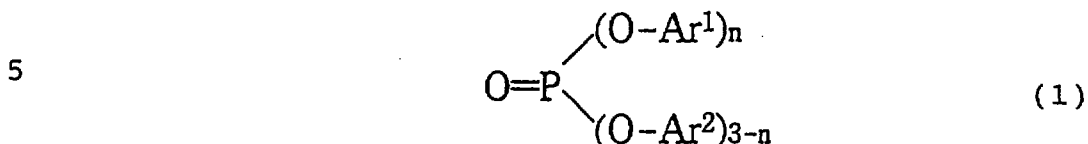


wherein each of Ar^3 , Ar^4 , Ar^5 and Ar^6 independently represents a $\text{C}_6\text{-C}_{20}$ aromatic group which is unsubstituted or substituted with at least one $\text{C}_1\text{-C}_{10}$ hydrocarbon group; and m is an integer of 1 or more.

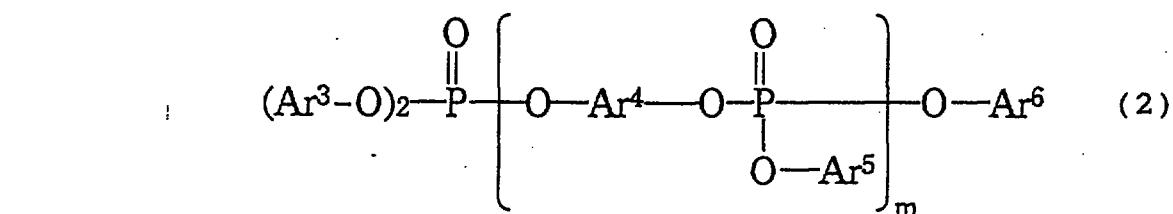
15. The resin composition according to item 12 above, wherein the at least one aromatic phosphoric ester is a

mixture of:

a monomeric aromatic phosphoric ester represented by the following formula (1):



wherein each of Ar^1 and Ar^2 independently represents a C_6 - C_{20} aromatic group which is unsubstituted or substituted with at least one C_1 - C_{10} hydrocarbon group; and each n is identical to each other and represents an integer of from 0 to 3, and an oligomeric aromatic phosphoric ester represented by the following formula (2):



wherein each of Ar^3 , Ar^4 , Ar^5 and Ar^6 independently represents a C_6 - C_{20} aromatic group which is unsubstituted or substituted with at least one C_1 - C_{10} hydrocarbon group; and m is an integer of 1 or more.

16. The resin composition according to item 15 above,
wherein the mixture contains 1 to 99 % by weight of the
monomeric aromatic phosphoric ester and 1 to 99 % by
weight of the oligomeric aromatic phosphoric ester, the
5 total amount of the monomeric aromatic phosphoric ester
and the oligomeric aromatic phosphoric ester being
100 % by weight.

17. The resin composition according to item 16 above,
10 wherein the mixture contains 1 to 50 % by weight of the
monomeric aromatic phosphoric ester and 50 to 99 % by
weight of the oligomeric aromatic phosphoric ester, the
total amount of the monomeric aromatic phosphoric ester
and the oligomeric aromatic phosphoric ester being 100
15 % by weight.

18. The resin composition according to any one of
items 3 to 17 above, wherein the amount of the solid
substance (C) is from 0.000001 to 100 parts by weight,
20 relative to 100 parts by weight of the thermoplastic
resin (A).

19. The resin composition according to item 18 above,
wherein the amount of the solid substance (C) is from
25 0.1 to 10 parts by weight, relative to 100 parts by

weight of the thermoplastic resin (A).

20. The resin composition according to any one of items 3 to 19 above, wherein the solid substance (C) is at least one additive selected from the group consisting of an adhesion-preventive agent, an oil absorber, a heat stabilizer and a light stabilizer.

21. The resin composition according to any one of items 1 to 20 above, wherein the thermoplastic resin (A) comprises an aromatic vinyl polymer and optionally at least one compound selected from the group consisting of an aromatic vinyl monomer, an aromatic vinyl dimer and an aromatic vinyl trimer, the amount of the at least one compound being 1 % by weight or less, based on the weight of the resin composition.

22. The resin composition according to item 1 above, wherein the mixing of the thermoplastic resin (A) and the non-polymerizable liquid substance (B) together is performed while heating at a temperature not exceeding 300 °C, and wherein, after the mixing, the resultant is cooled, followed by pelletization, whereby the resin composition is produced in the form of pellets.

23. The resin composition according to item 1 above, wherein the mixing is performed in the presence of (D) a solvent, and, after the mixing, the solvent (D) is removed.

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24. The resin composition according to item 23 above, wherein the amount of the solvent (D) is 0.1 to 100 parts by weight, relative to 100 parts by weight of the thermoplastic resin (A).

10

25. The resin composition according to item 23 or 24 above, wherein the solvent (D) has a boiling point which is higher than the temperature at which the mixing is performed and which is not higher than 300 °C.

15

26. A method for producing an shaped article, which comprises subjecting to melt extrusion:

(i) a thermoplastic resin composition which is substantially the same as that produced by a method comprising the step of mixing together:

20

(A) a thermoplastic resin, and

25

(B) at least one non-polymerizable liquid substance as an additive for the thermoplastic resin (A), the non-polymerizable liquid substance (B) being in a

liquid state at a temperature in the range of from 25 to 300 °C,

the mixing being conducted at a temperature in the range of from 25 to 300 °C, at which the at least one
5 non-polymerizable liquid substance (B) maintains a liquid state,

thereby causing the thermoplastic resin (A) to be dissolved in or swelled with the at least one substance (B); and

10 (ii) a thermoplastic resin which is the same as or different from the thermoplastic resin (A).

27. The method according to item 26 above, wherein the thermoplastic resin (ii) comprises at least one thermo-
15 plastic elastomer.

28. The method according to item 26 or 27 above, wherein the resin composition (i) and the thermoplastic resin (ii) are subjected to melt extrusion together
20 with (E) at least one additive selected from the group consisting of a flame retardant, an auxiliary agent for a flame retardant, a light stabilizer, a mold release agent, a fluidity-improving agent, a heat stabilizer, an anti-static agent, a coloring agent and a foaming
25 agent, the at least one additive (E) being the same as

or different from the non-polymerizable liquid substance (B).

5 29. A shaped article produced by the method according to any one of items 26 to 28 above.

30. A polyphenylene ether resin composition comprising:

10 (A') a polyphenylene ether resin comprising a plurality of polymer chains, each comprising recurring oxyphenylene units, the polyphenylene ether resin (A') containing hydroxyl groups in an amount of not more than 0.5 mole, relative to 100 moles of the recurring oxyphenylene units, and

15 (B') at least one additive for the polyphenylene ether resin (A').

20 31. The resin composition according to item 30 above, wherein the at least one additive (B') is selected from the group consisting of a flame retardant, a plasticizer, a heat stabilizer and a light stabilizer.

25 32. The resin composition according to any one of items 30 and 31 above, wherein the weight ratio of the polyphenylene ether resin (A') to the at least one

additive (B') is from 10/90 to 90/10.

33. The resin composition according to any one of items 30 to 32 above, wherein the at least one additive (B') is a flame retardant.

34. The resin composition according to item 33 above, wherein the flame retardant comprises at least one organic phosphorus compound.

35. The resin composition according to item 34 above, wherein the organic phosphorus compound is at least one aromatic phosphoric ester.

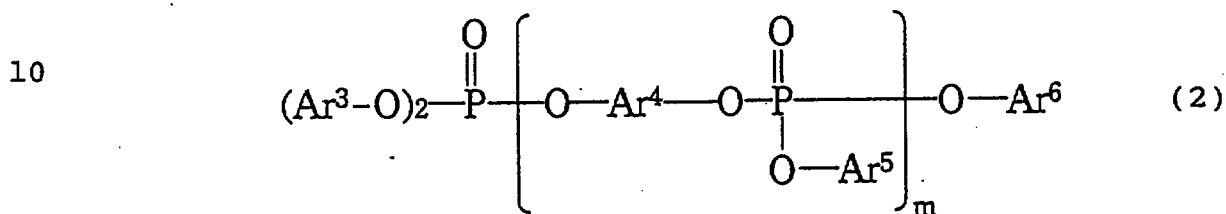
36. The resin composition according to item 35 above, wherein the aromatic phosphoric ester is a monomeric aromatic phosphoric ester represented by the following formula (1):



wherein each of Ar^1 and Ar^2 independently represents a C_6 - C_{20} aromatic group which is unsubstituted or substituted with at least

one C₁-C₁₀ hydrocarbon group; and each n is identical to each other and represents an integer of from 0 to 3.

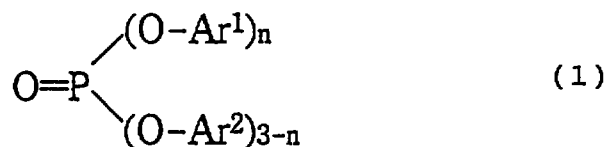
5. 37. The resin composition according to item 35 above, wherein the aromatic phosphoric ester is an oligomeric aromatic phosphoric ester represented by the following formula (2):



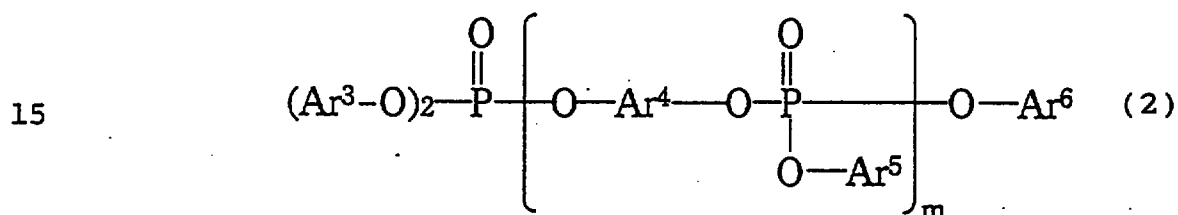
- 15 wherein each of Ar³, Ar⁴, Ar⁵ and Ar⁶ independently represents a C₆-C₂₀ aromatic group which is unsubstituted or substituted with at least one C₁-C₁₀ hydrocarbon group; and m is an integer of 1 or more.

- 20 38. The resin composition according to item 35 above, wherein the at least one aromatic phosphoric ester is a mixture of:

a monomeric aromatic phosphoric ester represented by the following formula (1):



5 wherein each of Ar^1 and Ar^2 independently
represents a $\text{C}_6\text{-C}_{20}$ aromatic group which is
unsubstituted or substituted with at least
one $\text{C}_1\text{-C}_{10}$ hydrocarbon group; and each n is
identical to each other and represents an
10 integer of from 0 to 3, and
an oligomeric aromatic phosphoric ester represent-
ed by the following formula (2):



wherein each of Ar^3 , Ar^4 , Ar^5 and Ar^6 inde-
pendently represents a $\text{C}_6\text{-C}_{20}$ aromatic group
20 which is unsubstituted or substituted with at
least one $\text{C}_1\text{-C}_{10}$ hydrocarbon group; and
 m is an integer of 1 or more.

39. The resin composition according to item 38 above,
25 wherein the mixture contains 1 to 99 % by weight of the

monomeric aromatic phosphoric ester and 1 to 99 % by weight of the oligomeric aromatic phosphoric ester, the total amount of the monomeric aromatic phosphoric ester and the oligomeric aromatic phosphoric ester being 100 % by weight.

40. The resin composition according to item 39 above, wherein the mixture contains 1 to 50 % by weight of the monomeric aromatic phosphoric ester and 50 to 99 % by weight of the oligomeric aromatic phosphoric ester, the total amount of the monomeric aromatic phosphoric ester and the oligomeric aromatic phosphoric ester being 100 % by weight.

41. A shaped article produced from the resin composition of any one of items 1 to 25 above, which is a film, a sheet or a foamed article.

42. A shaped article produced from the resin composition of any one of items 1 to 25 above, which is a part for an office automation machine, a part for an electric appliance, a part for an electronic appliance or a part for an automobile.

43. Use of the resin composition of any one of items 1

to 25 above as a material for injection molding.

44. The shaped article according to item 29 above, which is a film, a sheet or a foamed article.

5

45. The shaped article according to item 29 above, which is a part for an office automation machine, a part for an electric appliance, a part for an electronic appliance or a part for an automobile.

10

46. The shaped article according to item 29 above, which is a pellet for use as a material for injection molding.

15

47. A shaped article produced from the resin composition of any one of items 30 to 40 above, which is a film, a sheet or a foamed article.

20

48. A shaped article produced from a resin composition of any one of items 30 to 40 above, which is a part for an office automation machine, a part for an electric appliance, a part for an electronic appliance or a part for an automobile.

25

49. Use of the resin composition of any one of items

30 to 40 above as a material for injection molding.

Hereinbelow, the present invention is described in detail.

In one aspect of the present invention, there is
5 provided a resin composition comprising: (A) a thermoplastic resin; (B) at least one non-polymerizable liquid substance as an additive for the thermoplastic resin (A); and optionally (C) a crystalline or
10 amorphous solid substance as an additive for the thermoplastic resin (A).

In the resin composition of the present invention, the above-mentioned thermoplastic resin (A) serves as a resin component, and each of the above-mentioned non-polymerizable liquid substance (B) and
15 solid substance (C) is used to impart the thermoplastic resin (A) with desired properties, such as flame retardancy and fluidity.

In the resin composition of the present invention, the thermoplastic resin (A) is dissolved in or swelled
20 with at least one non-polymerizable liquid substance (B). The resin composition of the present invention is produced, for example, in the form of a pellet (such as a cylindrical pellet or a spherical pellet) having a diameter of from 0.01 to 10 mm. Such a pellet may be
25 subjected to extrusion molding to obtain a sheet or a

film or injection molding to obtain a shaped article having a desired configuration. Further, as described below in detail, it is especially advantageous to use the resin composition of the present invention as a masterbatch for preparing a molding material, wherein the masterbatch is added to a resin material which is the same as or different from the thermoplastic resin (A) used in the resin composition.

The resin composition of the present invention can be produced without a need for a step of subjecting the thermoplastic resin (A) to melt processing to cause the resin to be in a molten state. The "molten state" means the state of a thermoplastic resin having a temperature which is higher than the softening point of the thermoplastic resin, wherein the resin is molten and flowable. Generally, in the production of a conventional (resin/additive) composition used as a molding material or a conventional shaped resin article, the melt processing is conducted. With respect to the temperature conventionally employed for the melt processing (such a temperature is, hereinafter, frequently referred to simply as a "conventional melt processing temperature"):

(1) in the case of the production of a resin composition containing an amorphous polymer or the

production of a shaped resin article using an amorphous polymer, the conventional melt processing temperature is about 50 to more than 100 °C higher than the glass transition temperature (T_g) of the polymer; and

5 (2) in the case of the production of a resin composition containing a crystalline polymer or the production of a shaped resin article using a crystalline polymer, the conventional melt processing temperature is about 50 to more than 100 °C higher than the
10 melting temperature (T_m) of the polymer.

When a polymer is in a molten state, the high temperature of such a polymer in a molten state not only causes the cleavage of the polymer chains but also causes polymer chains (which are present in a close
15 positional relationship) to suffer side reactions, such as a rearrangement reaction, which leads to the deterioration of the polymer. Therefore, for avoiding the above-mentioned disadvantages, so as to obtain a shaped resin article having excellent appearance, light
20 stability and mechanical properties, the way to impart the thermoplastic resin used as a molding material with a satisfactory fluidity at a temperature as low as possible, during the production of a resin/additive composition or a shaped resin article, is very important.
25 Therefore, it is desirable that the production of

a resin-additive composition used as a molding material or of a shaped resin article is conducted at a temperature which is lower than the conventional melt processing temperature. Specifically, it is desirable:

5 that, in the case of the production of a resin composition containing an amorphous polymer or the production of a shaped resin article using an amorphous polymer, the production process is conducted at a temperature which is lower than the conventional melt
10 processing temperature and which is selected within the range of from 50 to not higher than 100 °C higher than the glass transition temperature (T_g) of the polymer, most preferably at a temperature which is 90 °C or less higher than the T_g of the polymer [i.e., production
15 temperature $\leq T_g + 90$ °C]; and

 that, in the case of the production of a resin composition containing a crystalline polymer or the production of a shaped resin article using a crystalline polymer, the production process is conducted at a
20 temperature which is lower than the conventional melt processing temperature and which is selected within the range of from 50 to not higher than 100 °C higher than the melting temperature (T_m) of the polymer, most
25 preferably at a temperature which is 90 °C or less higher than the T_m of the polymer [i.e., production

temperature $\leq T_m + 90\text{ }^{\circ}\text{C}$].

In the case of the resin composition of the present invention, the production of the resin composition can be conducted at a temperature which is lower than the conventional melt processing temperature by employing a method comprising the step of mixing together:

(A) a thermoplastic resin, and

(B) at least one non-polymerizable liquid substance as an additive for the thermoplastic resin (A),

wherein the non-polymerizable liquid substance (B) is in a liquid state at a temperature in the range of from 25 to 300 $^{\circ}\text{C}$, preferably from 100 to 300 $^{\circ}\text{C}$, most preferably from 150 to 250 $^{\circ}\text{C}$, and

wherein the mixing is conducted at a temperature in the range of from 25 to 300 $^{\circ}\text{C}$, preferably from 100 to 300 $^{\circ}\text{C}$, most preferably from 150 to 250 $^{\circ}\text{C}$, at which the at least one non-polymerizable liquid substance (B) maintains a liquid state,

thereby causing the thermoplastic resin (A) to be dissolved in or swelled with the at least one non-polymerizable liquid substance (B).

When, with respect to a resin composition comprising the thermoplastic resin (A) and the non-polymerizable liquid substance (B), the non-polymerizable liquid substance (B) is adsorbed onto or

absorbed by the thermoplastic resin (A), such a resin composition is disadvantageous because a part of the thermoplastic resin (A) is present in the resin composition in a powder form, so that a shaped resin article produced from such a resin composition has a rough surface, and the mechanical properties of a shaped resin article produced from such a resin composition are lowered. In the present invention, the mixing of the thermoplastic resin (A) and the non-polymerizable liquid substance (B) {to cause the thermoplastic resin (A) to be dissolved in or swelled with the non-polymerizable liquid substance (B)} may be conducted either in a continuous manner or a batchwise manner at a temperature within the above-mentioned range.

With respect to the difference between the resin composition of the present invention in which the thermoplastic resin (A) is dissolved in or swelled with the at least one non-polymerizable liquid substance (B) and the resin composition in which the non-polymerizable liquid substance (B) is adsorbed onto or absorbed by the thermoplastic resin (A) (such a resin composition is hereinafter, frequently, referred to simply as an "adsorption/absorption composition"), an explanation is made below.

In the case of the resin composition of the pres-

ent invention, when the resin composition is placed in a container and the container is inclined, the resin composition flows along the inner surface of the container. On the other hand, in the case of the adsorption/absorption resin composition, the resin composition is in a powder form or in the form of a solid mass. Therefore, even when such a resin composition is placed in a container and the container is inclined, the resin composition does not flow.

On the other hand, in the resin composition of the present invention, the non-polymerizable liquid substance (B) may be in such a state that it is adsorbed onto or absorbed by the thermoplastic resin (A) at a temperature which is lower than the temperature at which the mixing of the thermoplastic resin (A) and the non-polymerizable liquid substance (B) is performed as long as the thermoplastic resin (A) is dissolved in or swelled with the at least one non-polymerizable liquid substance (B) at the mixing temperature which is selected within the range of from 25 to 300 °C. For example, when a molding material comprising the adsorption/absorption composition as a masterbatch, in which a polyphenylene ether resin (PPE) is used as the thermoplastic resin (A), and a resin material (which may be the same as or different from the polyphenylene ether

resin used in the resin composition) is subjected to molding, the resultant molded product has many discrete segments of PPE that have not experienced melting, so that the appearance of the molded product becomes poor.

5 Further, when the resin composition undergoes the above-mentioned molten state during the molding, the resultant molded product has a disadvantageously high yellowness index (YI) and has hydroxyl groups in an amount of more than 0.5 mole, relative to 100 moles of
10 recurring oxymethylene units of the polyphenylene ether resin. On the other hand, when a molding material comprising the resin composition of the present invention as a masterbatch {which is produced by a method in which a polyphenylene ether resin as the thermoplastic
15 resin (A) is caused to be dissolved in or swelled with the non-polymerizable liquid substance (B)} and a resin material (which may be the same as or different from the polyphenylene ether resin used in the resin composition) is subjected to molding, it is possible to
20 obtain a molded product having an excellent appearance without suffering the above-mentioned disadvantageous phenomena. It should be noted that, in the present invention, when the temperature at which the mixing of the thermoplastic resin (A) and the non-polymerizable
25 liquid substance (B) is performed exceeds 300 °C, each

of the thermoplastic resin (A) and the non-polymerizable liquid substance (B) suffers thermal deterioration, so that the resultant resin composition has poor mechanical strength, light stability and color.

5 As already mentioned above, in the present invention, the non-polymerizable liquid substance (B) as an additive for the thermoplastic resin (A) is a substance which is in a liquid state at a temperature in the range of from 25 to 300 °C. The non-polymerizable
10 liquid substance (B) functions to impart the thermoplastic resin (A) with desired properties. When a resin composition of the present invention is used to obtain a shaped resin article, generally, most of the non-polymerizable liquid substance (B) used in the
15 production of the resin composition of the present invention remains in the obtained shaped resin article. Substances which do not have the above-mentioned functions, such as a solvent which only functions to dissolve the thermoplastic resin (A), are not included in
20 the non-polymerizable liquid substance (B) as defined in the present invention.

 In the present invention, it is preferred that the non-polymerizable liquid substance (B) has a compatibility with the non-polymerizable liquid substance.
25 In the present invention, the compatibility is defined

as follows. 50 Parts by weight of particulate thermoplastic resin (A) having an average particle diameter of from 15 μm to 1 mm and 50 parts by weight of the non-polymerizable liquid substance (B) are mixed with each other, followed by stirring at a temperature of 200 °C for 1 hour. The resultant mixture is subjected to filtration using a glass filter having a pore size of 120 μm , while maintaining the temperature of the mixture at 200 °C. Then, the weight of the mixture remaining on the glass filter is measured. The compatibility of the non-polymerizable liquid substance (B) with the thermoplastic resin is defined as the ratio (filtration-remaining ratio) (%) of the weight of a component (A)/component (B) mixture remaining on the glass filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration. The lower the filtration-remaining ratio, the higher the compatibility of the non-polymerizable liquid substance with the thermoplastic resin (A). In the present invention, it is preferred that the filtration-remaining ratio is 50 % or less, more preferably 30 % or less, still more preferably 10 % or less.

In the present invention, it is not preferred to use as the non-polymerizable liquid substance (B) the additives used in the surface modification of a

thermoplastic resin (hereinafter, frequently, referred to simply as "surface modifiers") because such additives are poor with respect to the compatibility as defined above. A specific reason for this is as follows. With respect to the surface modifier, it is very important for the surface modifier to bleed during the molding so that the surface modifier is primarily positioned at the surface portion of the resultant shaped article so as to exert the surface modification effect. Therefore, the surface modifier is required to have a low compatibility with the resin. When such a surface modifier is used to obtain a resin composition, it is required to conduct a high-temperature melt processing, thereby deteriorating the resin and the additive. Further, when it is intended to obtain a resin composition containing the surface modifier in a high content, it becomes necessary to conduct the melt processing at especially high temperatures. Therefore, as mentioned above, it is not preferred to use the surface modifier as the non-polymerizable liquid substance (B). However, such surface modifier may be used in combination with another non-polymerizable substance (B) so as to avoid the above-mentioned disadvantages accompanying the use of the surface modifier. In this case, it is preferred that the surface modifier is used in an

amount such that the compatibility between the thermoplastic resin (A) and the mixture of the surface modifier and other non-polymerizable liquid substance (B) is 50 % or less, in terms of the above-mentioned filtration-remaining ratio. Specifically, it is preferred that the amount of the surface modifier is 30 weight % or less, more preferably 20 % by weight or less, still more preferably 10 % by weight or less, based on the weight of the other non-polymerizable liquid substance (B).

On the other hand, conventional additives for use in improving the flame retardancy, fluidity or the like of a thermoplastic resin have satisfactorily high compatibility with the thermoplastic resin (A) so that the thermoplastic resin (A) can be dissolved at a relatively low temperature. Therefore, when such additives are used as the non-polymerizable liquid substance (B), the thermal deterioration of the thermoplastic resin can be suppressed. As another yardstick for evaluating the compatibility of the non-polymerizable liquid substance (B) with the thermoplastic resin (A), there can be mentioned a solubility parameter (SP) value. In the present invention, it is preferred that the difference between the SP value of the thermoplastic resin (A) and the SP value of the non-polymerizable

liquid substance (B) is 3 or less, more preferably 2 or less, most preferably 1 or less.

5 In the present invention, the above-mentioned solid substance (C) is a substance which is a crystalline solid or an amorphous solid at a temperature of 300 °C or less. As in the case of the non-polymerizable liquid substance (B), the solid substance (C) functions to impart the thermoplastic resin (A) with desired properties. The solid substance (C) is used in an amount such that the satisfactory compatibility
10 between the non-polymerizable liquid substance (B) and the thermoplastic resin (A) is not adversely influenced.

In the present invention, the resin composition of
15 the present invention may be produced using (D) a solvent capable of dissolving or swelling the thermoplastic resin (A) so as to improve the compatibility of the non-polymerizable liquid substance (B) with the thermoplastic resin (A). Specifically, in this case,
20 the resin composition of the present invention can be produced by a method in which the above-mentioned mixing of the thermoplastic resin (A) and the non-polymerizable liquid substance (B) is conducted in the presence of the solvent (D), and after the mixing, the
25 solvent (D) is removed. With respect to the amount of

the solvent (D), it is preferred to use solvent (D) in an amount of from 0.1 to 100 parts by weight, more preferably from 1 to 50 parts by weight, still more preferably from 5 to 20 parts by weight, relative to 100 parts by weight of the thermoplastic resin (A).
For achieving the desired compatibility of the non-polymerizable liquid substance (B) with the thermoplastic resin (A), it is preferred that each of the difference in SP value between the thermoplastic resin (A) and the solvent (D) and the difference in SP value between the non-polymerizable liquid (B) and the solvent (D) is 3 or less, more preferably 2 or less, still more preferably 1 or less. Further, it is preferred that the solvent (D) has a boiling point which is higher than the temperature at which the mixing of the thermoplastic resin (A) and the non-polymerizable liquid substance (B) is performed and which is not higher than 300 °C.

In the present invention, it is preferred that the mixing of the thermoplastic resin (A) and the non-polymerizable liquid substance (B) is conducted in the absence of polymerizable compounds, such as a vinyl monomer. When the mixing is conducted in the presence of the polymerizable compound, such a compound is likely to be grafted on the thermoplastic resin or to

cause a crosslinking reaction, leading to disadvantages, such as a change in the inherent properties of the thermoplastic resin (A) or a lowering of the thermoplasticity of the thermoplastic resin (A).

5 The resin composition of the present invention can be advantageously used as a masterbatch for preparing a molding material for melt extrusion, wherein the masterbatch is added to a resin material which is the same as or different from the thermoplastic resin (A) used
10 in the resin composition. Such a molding material can be advantageously used for producing various shaped articles, such as injection molded articles (e.g., a casing or an internal part for an office automation machine, household electric appliances or the like), a
15 film, a sheet and a foamed article.

20

25

Hereinbelow, a detailed explanation will be made on the components of the composition of the present invention.

5 In the present invention, it is desired that the thermoplastic resin (A) exhibits high compatibility with the non-polymerizable liquid substance (B). Examples of thermoplastic resins (A) include a polyphenylene ether resin, a polystyrene resin, a polyester resin, a polyphenylene sulfide resin, a polycarbonate resin, a polyolefin resin, a polyamide resin, a poly-
10 methacrylate resin, a polyacetal resin, a polyarylate resin, a polysulfone resin, a polyether imide resin, a polyether sulfone resin, a polyether ether ketone, and the like. In the present invention, examples of thermoplastic resins (A) also include a thermoplastic elastomer having the same polymer skeleton as that of
15 any of the above-mentioned thermoplastic resins.

With respect to a yardstick for selecting a thermoplastic resin preferred as the thermoplastic resin
20 (A), when an amorphous polymer is used as the thermoplastic resin (A), a glass transition temperature (T_g) can be used as the yardstick, and when a crystalline polymer is used as the thermoplastic resin (A), a melting temperature (T_m) can be used as the yardstick.
25 It is preferred that the glass transition temperature

(T_g) of an amorphous polymer or the melting temperature (T_m) of a crystalline polymer is 85 °C or more, more preferably 100 °C or more. When an amorphous polymer having a glass transition temperature (T_g) of less than 85 °C or a crystalline polymer having a melting temperature (T_m) of less than 85 °C is used as the thermoplastic resin (A), the advantages of the present invention over the conventional melt-extrusion methods are not large, because thermoplastic resins having such a low glass transition temperature (T_g) or such a low melting temperature (T_m) can be melt-processed or melt-molded at low temperatures. On the other hand, when an amorphous polymer having a glass transition temperature (T_g) of 85 °C or more or a crystalline polymer having a melting temperature (T_m) of 85 °C or more is used as the thermoplastic resin (A), the advantages of the present invention over the conventional melt-extrusion methods are remarkable, because thermoplastic resins having such a high glass transition temperature (T_g) or such a high melting temperature (T_m) can be melt-processed or melt-molded only at high temperatures. That is, when a resin composition containing a thermoplastic resin having such a high glass transition temperature (T_g) or such a high melting temperature (T_m) is subjected to a conventional melt-extrusion method, each of

the thermoplastic resin and an additive therefor inevitably suffers thermal deterioration. As mentioned above, in the present invention, there is no need for a melt-processing at high temperatures and, therefore, there is no danger of thermal deterioration.

Preferred examples of thermoplastic resins (A) include a polyphenylene ether resin, a polystyrene resin and a polycarbonate resin. Examples of polystyrene resins include rubber-modified styrene polymers. An especially preferred example of thermoplastic resins (A) is a polymer blend of a polyphenylene ether resin and a polystyrene resin, since it exhibits excellent solubility in the non-polymerizable liquid substance (B).

It is preferred that the thermoplastic resin (A) used in the present invention is a resin which has not experienced a melt processing. For example, after completion of the production of a polyphenylene ether resin, the polyphenylene ether resin is obtained not in the molten state, but in the form of a solution or slurry containing the polyphenylene ether resin. If a thermoplastic resin produced in such a non-molten state is used as the thermoplastic resin (A), it is preferred that, before the use, the thermoplastic resin does not experience a melt processing in which the thermoplastic

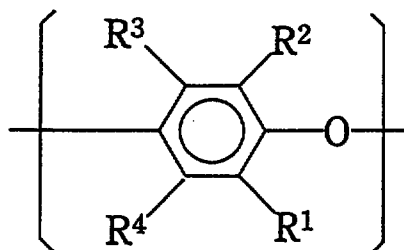
resin is brought into the molten state as defined above. On the other hand, after completion of the production of a polystyrene resin, the polystyrene resin is obtained in the molten state; however, the
5 production process in which a thermoplastic resin is produced in the molten state is excluded from the category of the melt processing mentioned herein.

With respect to the morphology of the thermoplastic resin (A) used in the present invention, there is
10 no particular limitation. However, from the viewpoint of improving the solubility of the thermoplastic resin (A), it is preferred that the thermoplastic resin (A) is a powder having a number average particle diameter of from 0.01 to 10,000 μm , more preferably from 0.1 to
15 1,000 μm , still more preferably from 1 to 100 μm . When the number average particle diameter of the thermoplastic resin (A) is less than 0.01 μm , an agglomeration of the thermoplastic resin particles tends to occurs, so that the handling properties and quality of the thermoplastic resin become low. On the other hand, when the
20 number average particle diameter of the thermoplastic resin (A) is more than 10,000 μm , the rate of the dissolution of the thermoplastic resin (A) into the non-polymerizable liquid substance (B) becomes low, so
25 that a portion of the thermoplastic resin particles

(i.e., discrete segments of the resin) is likely to remain as such in the resin composition, thus causing a lowering of the appearance and mechanical properties of an ultimate shaped article.

5 In the present invention, a polyphenylene ether (hereinafter, frequently referred to simply as "PPE"), which can be used as the thermoplastic resin (A), is a homopolymer and/or a copolymer comprising a plurality of polymer chains, each comprising recurring oxyphenylene units represented by the following formula (3):

10



20

wherein each of R^1 , R^2 , R^3 and R^4 independently represents a hydrogen atom or an unsubstituted or substituted hydrocarbon group.

The R^1 , R^2 , R^3 and R^4 may be the same or different.

25

Specific examples of polyphenylene ethers include

poly(2,6-dimethyl-1,4-phenylene ether), a copolymer of 2,6-dimethyl phenol and 2,3,6-trimethyl phenol, and the like. Of them, poly(2,6-dimethyl-1,4-phenylene ether) is preferred. The method for producing such a PPE is not particularly limited. For example, a PPE can be readily produced by the method described in U.S. Patent No. 3,306,874, in which, for example, 2,6-xylenol is subjected to oxidative polymerization, using as a catalyst a complex of copper (I) salt and an amine. Further, a PPE can also be readily produced by other methods described, for example, in U.S. Patent No. 3,306,875, U.S. Patent No. 3,257,357, U.S. Patent No. 3,257,358, Examined Japanese Patent Application Publication No. 52-17880, and Unexamined Japanese Patent Application Laid-Open Specification No. 50-51197. The reduced viscosity η_{sp}/c (as measured in a 0.5 g/dl chloroform solution at 30 °C) of the PPE used in the present invention is preferably in the range of from 0.20 to 0.70 dl/g, more preferably from 0.30 to 0.60 dl/g. As a method for achieving the above-mentioned range of reduced viscosity of the polyphenylene ether, a method in which the amount of a catalyst used in the production of the polyphenylene ether is appropriately chosen can be mentioned.

Examples of polystyrene resins usable as the

thermoplastic resin (A) include rubber-modified styrene polymers.

5 A rubber-modified styrene polymer comprises an aromatic vinyl polymer as a continuous phase and rubber particles dispersed therein as a dispersion phase. A rubber-modified styrene polymer can be obtained by graft-polymerizing an aromatic vinyl monomer and optionally a vinyl comonomer copolymerizable with the aromatic vinyl monomer, on a rubber polymer, using a
10 customary method, such as a bulk polymerization method, a bulk suspension polymerization method, a solution polymerization method or an emulsion polymerization method.

Examples of rubber-modified styrene polymers
15 include the so-called high impact polystyrene (hereinafter, frequently referred to as "HIPS"), ABS resin (acrylonitrile/butadiene/styrene copolymer), AAS resin (acrylonitrile/butyl acrylate rubber/styrene copolymer), AES resin (acrylonitrile/ethylene-propylene
20 rubber/styrene copolymer) and the like.

With respect to the above-mentioned rubber polymer to be used in the production of a rubber-modified styrene polymer, it is preferred that the rubber polymer has a glass transition temperature (T_g) of -30°C or
25 lower. If the rubber polymer has a glass transition

temperature higher than -30°C , the improvement in the impact resistance may be unsatisfactory in some cases.

5 Examples of suitable rubber polymers include diene rubbers, such as polybutadiene, poly(styrene-butadiene) and poly(acrylonitrile-butadiene); saturated rubbers obtained by hydrogenating the diene rubbers mentioned above; an isoprene rubber; a chloroprene rubber; acrylic rubbers, such as polybutyl acrylate; and an ethylene/propylene/diene terpolymer (EPDM). Diene rubbers
10 are particularly preferred.

 Preferred examples of aromatic vinyl monomers which are graft-polymerizable with the rubber polymer include styrene, α -methylstyrene, p-methylstyrene, p-chlorostyrene, p-bromostyrene, and 2,4,5-tribromostyrene.
15 Styrene is most preferred, but styrene can be used for copolymerization in combination with other aromatic vinyl monomers mentioned above.

 As mentioned above, at least one vinyl comonomer copolymerizable with the aromatic vinyl monomer can be
20 introduced into the rubber-modified styrene polymer. For obtaining a rubber-modified styrene copolymer having excellent oil resistance, as a vinyl comonomer copolymerizable with the aromatic vinyl monomer, use can be made of an unsaturated nitrile monomer, such as
25 acrylonitrile or methacrylonitrile. Also, for lowering

the melt viscosity of the aromatic vinyl monomer, use can be made of an acrylate comonomer having an alkyl group having 1 to 8 carbon atoms. Further, for improving the heat resistance of a final resin composition, other comonomers, such as α -methylstyrene, acrylic acid, methacrylic acid, maleic anhydride and N-substituted maleimide or the like can be used.

When the aromatic vinyl monomer is used in the form of a monomer mixture with a comonomer, the amount of a vinyl monomer copolymerizable with the aromatic vinyl monomer in the monomer mixture is generally in the range of from 0 to 40 % by weight.

In the present invention, the content of the rubber polymer in the rubber-modified styrene polymer is preferably in the range of from 5 to 80 % by weight, more preferably from 10 to 50 % by weight. The content of comonomer units copolymerized with the aromatic vinyl monomer units in the rubber-modified styrene polymer is preferably in the range of from 95 to 20 % by weight, more preferably from 90 to 50 % by weight. When the contents of the rubber polymer and the comonomer units in the rubber-modified styrene polymer are within the above-mentioned ranges, a good balance of impact resistance and rigidity can be achieved with respect to an ultimate shaped article. The diameter of

the rubber particles in the rubber-modified styrene polymer is preferably from 0.1 to 5.0 μm , more preferably from 0.2 to 3.0 μm . When the rubber particle diameter is within the above-mentioned range, the impact resistance is particularly enhanced.

With respect to the rubber-modified styrene polymer, the reduced viscosity $\eta_{\text{sp}}/\text{c}$ (as measured in a 0.5 g/dl solution at 30 °C) of the solvent soluble matters, which is a yardstick of the molecular weight, is preferably in the range of from 0.30 to 0.80 dl/g, more preferably from 0.40 to 0.60 dl/g, wherein, when the styrene polymer is a polystyrene resin, toluene is used as the solvent and, when the styrene polymer is an unsaturated nitrile/aromatic vinyl copolymer, methyl ethyl ketone is used as the solvent. In the production of the rubber-modified styrene polymer, the reduced viscosity $\eta_{\text{sp}}/\text{c}$ can be controlled by selecting the type and amount of the initiator, the polymerization temperature and the amount of the chain transfer agent.

The aromatic polycarbonate usable as the thermoplastic resin (A) in the present invention can be selected from aromatic homopolycarbonates and aromatic copolycarbonates. Examples of methods for producing an aromatic polycarbonate include a phosgene method in which phosgene is blown into a bifunctional phenolic

compound in the presence of caustic alkali and a solvent, and a transesterification method in which, for example, a bifunctional phenolic compound and diethyl carbonate are subjected to transesterification in the presence of a catalyst. A preferred range of the viscosity average molecular weight of the aromatic polycarbonate is from 10,000 to 100,000.

Examples of bifunctional phenolic compounds include 2,2'-bis(4-hydroxyphenyl)propane, 2,2'-bis(4-hydroxy-3,5-dimethylphenyl)propane, bis(4-hydroxyphenyl)methane, 1,1'-bis(4-hydroxyphenyl)ethane, 2,2'-bis(4-hydroxyphenyl)butane, 2,2'-bis(4-hydroxy-3,5-diphenyl)butane, 2,2'-bis(4-hydroxy-3,5-dipropylphenyl)propane, 1,1'-bis(4-hydroxyphenyl)cyclohexane, and 1-phenyl-1,1'-bis(4-hydroxyphenyl)ethane. 2,2'-bis(4-hydroxyphenyl)propane (i.e., bisphenol A) is particularly preferred. In the present invention, bifunctional phenolic compounds can be used individually or in combination.

The non-polymerizable liquid substance (B) used in the present invention is a component which serves as an additive for the thermoplastic resin (A) for imparting the thermoplastic resin (A) with desired properties, such as flame retardancy. In the present invention, it is requisite that the non-polymerizable liquid sub-

stance (B) maintain a liquid state and be fluid at a temperature in the range of from 25 to 300 °C. With respect to viscosity of the non-polymerizable liquid substance (B), it is preferred that the viscosity is 1,000,000 centistokes or less, more preferably 100,000 centistokes or less, most preferably 10,000 centistokes or less, as measured at a temperature of 100 °C. When a non-polymerizable liquid substance (B) having a viscosity of 10,000 centistokes or less is used, the thermoplastic resin (A) can be well dissolved into the non-polymerizable liquid substance. For achieving the viscosity of 10,000 centistokes or less, it is preferred that the non-polymerizable liquid substance (B) is a compound having a molecular weight of 1,000 or less. The non-polymerizable substance may be either a polymeric or an oligomeric compound.

As mentioned above, the non-polymerizable liquid substance (B) used in the present invention serves as an additive, such as a plasticizer, a heat stabilizer, a light stabilizer, a flame retardant, a coloring agent, a foaming agent, a lubricant, a perfume, a smoking inhibitor, an adhesiveness-imparting agent, an antiseptic agent, a heat retaining ability-imparting agent, an impact strength improver, a repellent, a wetting agent, a surfactant, an emulsifier, a destabi-

lizer, a coagulating agent, an anti-foaming agent, an anti-freezing agent, a creaming agent, a thickening agent, a heat sensitizer and a frothing agent. In the present invention, it is preferred that, as the non-
5 polymerizable liquid substance (B), a plastizer, a heat stabilizer, a flame retardant or a light stabilizer is used in view of the high compatibility of these additives with the thermoplastic resin (A).

With respect to the amount of the non-polymerizable liquid substance (B) used in the resin composition of the present invention, it is preferred that the amount of the non-polymerizable substance (B) is from 1 to 99 % by weight, more preferably from 10 to 90 % by weight, still more preferably from 20 to 80 % by
15 weight, most preferably from 30 to 70 % by weight, based on the weight of the resin composition.

Examples of plastizers usable as the non-polymerizable liquid substance (B) include phthalic acid esters, such as dimethyl phthalate, diethyl phthalate and diisobutyl phthalate; phthalic acid-containing
20 esters, such as butylbenzyl phthalate; aliphatic dibasic acid esters, such as diisodecyl succinate and dioctyl adipate; glycol esters, such as diethylene glycol dibenzoate; aliphatic acid esters, such as butyl
25 oleate and methyl acetylricinoleate; epoxy compounds,

such as an epoxidized soy bean oil and an epoxidized linseed oil; trioctyl trimellitate; ethylphthalylethyl glycolate; butylphthalylbutyl glycolate; tributyl acetyl citrate; chlorinated paraffins; polypropylene adipate; polyethylene sebacate; triacetin; tributyrin; 5 toluenesulfonamide; alkylbenzenes; biphenyls; partially hydrogenated terphenyls; and camphor.

Examples of heat stabilizers usable as the non-polymerizable liquid substance (B) include a metallic soap, a lead stabilizer, an organotin stabilizer, a 10 composite stabilizer and an epoxy compound.

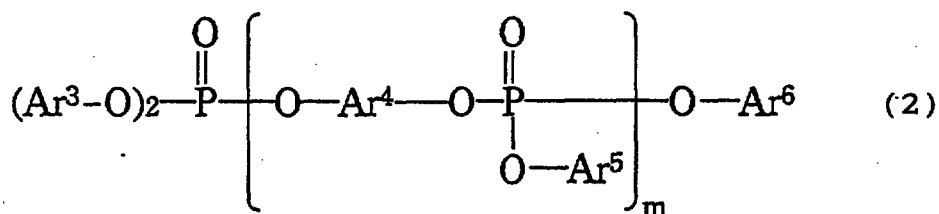
Examples of light stabilizers usable as the non-polymerizable liquid substance (B) include an ultraviolet ray absorber, a hindered amine light stabilizer, an antioxidant, an active species-capturing agent, a 15 sunproofing agent, a metal inactivator and a light quenching agent.

Examples of flame retardants usable as the non-polymerizable liquid substance (B) include a halogen-containing flame retardant and a phosphorus-containing flame retardant. With respect to the phosphorus-containing flame retardants, organic phosphorus compounds 20 are especially preferred.

Examples of organic phosphorus compounds include a 25 phosphine, a phosphine oxide, a bisphosphine, a phos-

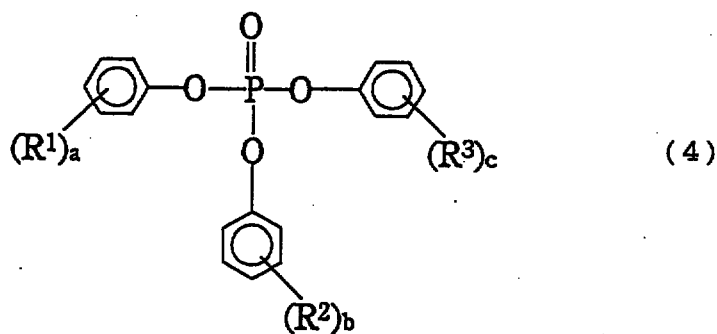
phonium salt, a phosphinic acid salt, a phosphoric ester and a phosphorous ester. More specific examples of organic phosphorus compounds include triphenyl phosphate, methylneopentyl phosphite, pentaerythritol diethyl diphosphite, methyl neopentyl phosphonate, phenyl neopentyl phosphate, pentaerythritol diphenyl-diphosphate, dicyclopentyl hypodiphosphate, dineopentyl hypophosphite, phenylpyrocatechol phosphite, ethylpyrocatechol phosphate and dipyrocatechol hypodiphosphate.

With respect to the organic phosphorus compound as the non-polymerizable liquid substance (B), from the view point of the compatibility with a thermoplastic resin, it is preferred to use a monomeric aromatic phosphoric ester represented by the following formula (1) and an oligomeric aromatic phosphoric ester represented by the following formula (2):



wherein each of Ar^1 , Ar^2 , Ar^3 , Ar^5 and Ar^6 independently represents a C_6 - C_{20} monovalent aromatic group (such as a phenyl group) unsubstituted or substituted with at least one C_1 - C_{10} hydrocarbon group; Ar^4 represents a C_6 - C_{20} bivalent aromatic group; each n is identical to each other and represents an integer of from 0 to 3; and m represents an integer of 1 or more.

With respect to the above-mentioned monomeric aromatic phosphoric ester of formula (1), especially preferred are hydroxyl group-containing monomeric aromatic phosphoric esters, such as those having at least one phenolic hydroxyl group (e.g., tricresylphosphate and triphenyl phosphate) and a monomeric aromatic phosphoric ester represented by the following formula (4):



wherein each of a , b and c independently represents an integer of from 1 to 3, and

each of R^1 , R^2 and R^3 independently represents a hydrogen atom or a C_1 - C_{30} alkyl group.

When a plurality of different compounds of formula (4) are used, it is preferred that the sum of the respective numbers of carbon atoms of the substituents R^1 , R^2 and R^3 is from 12 to 30, more preferably from 15 to 30, still more preferably from 20 to 30, most preferably from 25 to 30, on the average, wherein the average is a sum of the values obtained by multiplying the respective weight percentages of the different compounds of formula (4) by the respective total numbers of the carbon atoms of R^1 , R^2 and R^3 in the different compounds of formula (4).

Specific examples of substituents R^1 , R^2 and R^3 include a nonyl group, a butyl group (e.g., a t-butyl group), a t-amyl group, a hexyl group, a cyclohexyl group, a heptyl group, an octyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl group, a heptadecyl group, an octadecyl group, a nonadecyl group and an octadodecyl group. With respect to the substitution of the compound of formula (4), one or more substituents may be bonded to the aromatic ring of the compound of formula (4) at any one or more posi-

tions selected from the group consisting of an ortho-position, a meta-position and a para-position. Of these, preferred is a para-position. In the compound of formula (4), it is preferred that the sum of the
5 respective carbon atoms of the substituent alkyl groups is in the range of from 12 to 30. As compared to a compound of formula (4) having only one of the aromatic rings which is substituted with only one long-chain alkyl group, the compound of formula (4) having a
10 plurality of aromatic rings, each of which is substituted with an alkyl group (the number of carbon atoms of which is smaller than that of the above-mentioned long-chain alkyl group) is excellent with respect to the heat resistance and the water resistance. For
15 example, with respect to octadecylphenyldiphenyl phosphate and bis(nonylphenyl)phenyl phosphate, in each of which the sum of the number of carbon atoms of the alkyl group or the alkyl groups is 18, the latter is preferred from the viewpoint of high heat resistance.

20 In the present invention, among the monomeric aromatic phosphoric esters represented by formula (4), especially preferred are those in which at least one of R^1 , R^2 and R^3 is a nonyl group, and most preferred are those in which each of R^1 , R^2 and R^3 is a nonyl group
25 [i.e., tris(nonylphenyl)phenyl phosphate], from the

viewpoint of high fluidity and low volatility.

Especially when a flame retardant used as the non-polymerizable liquid substance (B) contains the compound of formula (4) in an amount of 50 % by weight or more, based on the weight of the flame retardant, such a flame retardant exhibits an excellent ability to impart a flame retardancy to the thermoplastic resin. Further, the compound of formula (4) has a high dripping-preventive property, and exhibits an especially excellent self-extinguishing property among flame retardants which are at the "V-2" level in the flame retardancy evaluation in accordance with UL-Subject 94.

Further, with respect to a flame retardant comprising a single compound of formula (4) or a flame retardant comprising a plurality of different compounds of formula (4), when the sum of the respective numbers of carbon atoms of R^1 , R^2 and R^3 or the average value of the sums is within the above-mentioned respective ranges, the flame retardant exhibits low volatility.

With respect to the flame retardant comprising a plurality of different compounds of formula (4), when the content of the compounds of formula (4), in which the sum of the respective numbers of carbon atoms of R^1 , R^2 and R^3 is less than 12, is 1 % by weight or less, such a flame retardant exhibits especially excellent low

volatility.

For achieving a satisfactory thermal stability of a flame retardant comprising at least one compound of formula (4) (especially, in respect of the resistance to heat discoloration), the content of the impurities in the compound of formula (4) is as low as possible. Specifically, it is preferred that the acid value of the flame retardant is 1 mgKOH/g or less, more preferably 0.5 mgKOH/g or less, as measured in accordance with JIS-K6751 (the acid value is used as a yardstick for the amount of an acidic substance remaining in the flame retardant) and/or that the content of an alkylphenol in the flame retardant is 1 % by weight, more preferably 0.5 % by weight. Further, it is preferred that the content of each of aluminum, magnesium, sodium and antimony in the flame retardant is 1,000 ppm or less. Further, especially when the flame retardant contains a hindered phenol type antioxidant in an amount of 1 to 1,000 ppm by weight, based on the weight of the flame retardant, such a flame retardant exhibits a remarkably improved thermal stability.

In the flame retardant comprising at least one compound of formula (4), all of substituents R^1 , R^2 and R^3 are alkyl groups. When an aryl group is used as substituents R^1 , R^2 and/or R^3 instead of an alkyl

group, the light stability of the flame retardant disadvantageously becomes low. With respect to the alkyl group used in the compound of formula (4), it is preferred that the amount of the branched structure is as small as possible. It is especially preferred that the alkyl group has a straight chain structure or a chain structure having only one branch.

With respect to the compound of formula (4), it is preferred that each of the aromatic rings has only one substituent bonded thereto. When a plurality of substituents are bonded to the aromatic ring, the viscosity of the compound of formula (4) becomes high. The larger the number of the substituents bonded to the aromatic ring, the higher the viscosity of the compound of formula (4). When the viscosity of the compound of formula (4) becomes too high, various disadvantages are likely to be caused, such that the handling of the compound becomes difficult, and that the purification of the compound also becomes difficult, so that the content of the above-mentioned impurities in the compound of formula (4) becomes high, thereby adversely affecting the properties of the compound of formula (4), such as the light stability and the resistance to heat discoloration.

Specific examples of flame retardants comprising a

plurality of different compounds of formula (4) include a flame retardant comprising tris(nonylphenyl) phosphate (TNPP) as a main component and a small amount of bis(nonylphenyl)phenyl phosphate (BNPP), wherein the sum of the respective numbers of carbon atoms of the substituents R^1 , R^2 and R^3 is from 20 to 27, preferably from 25 to 27, more preferably from 26 to 27, most preferably from 26.5 to 27, on the average. For obtaining such a flame retardant, for example, it is recommended that the content of BNPP in the flame retardant is from 78 to 0 % by weight, preferably from 22 to 0 % by weight, more preferably from 11 to 0 % by weight, most preferably from 5 to 0 % by weight, and that the content of TNPP is from 22 to 100 % by weight, preferably from 78 to 100 % by weight, more preferably from 89 to 100 % by weight, most preferably from 95 to 100 % by weight. The use of the above-mentioned flame retardant comprising TNPP and BNPP in the resin composition of the present invention is advantageous not only for imparting an excellent fluidity to the resin composition, but also for obtaining an ultimate shaped resin article having an excellent balance of various properties, such as flame retardancy, thermal resistance, impact strength, water resistance, gloss retention property and surface hardness. Further, the use

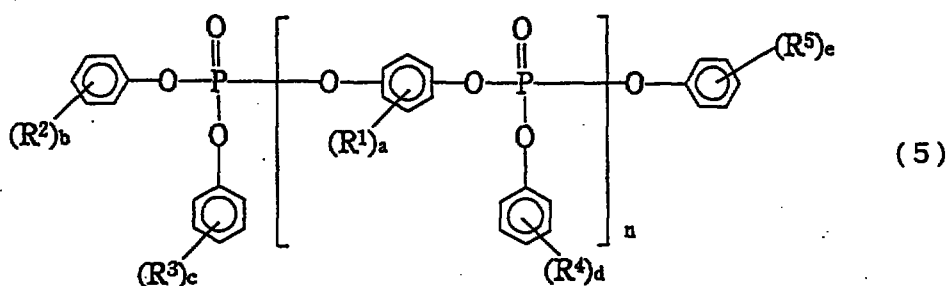
of TNPP is advantageous not only in that it has low volatility, but also in that it can impart the ultimate shaped resin article with excellent thermal resistance, water resistance and gloss retention property (the achievement of excellent gloss retention property is ascribed to the symmetrical structure of TNPP). Therefore, TNPP is especially preferred as the non-polymerizable liquid substance (B) in the resin composition of the present invention.

With respect to the method for producing the compound of formula (4), various known methods can be mentioned, for example, the methods disclosed in Unexamined Japanese Patent Application Laid-Open Specification No. 1-95149 and Unexamined Japanese Patent Application Laid-Open Specification No. 3-294284, respectively. Specifically, the compound of formula (4) can be produced by, for example, a method in which an alkylphenol is reacted with a phosphorus oxychloride in the presence of an aluminum chloride anhydride as a catalyst while heating, and a method in which a phosphonic triester is subjected to oxidation using oxygen, to thereby convert the phosphonic triester to a corresponding aromatic phosphoric ester.

With respect to the above-mentioned oligomeric aromatic phosphoric esters represented by formula (2),

preferred are compounds of formula (2) in which Ar^4 is a phenyl group or a bivalent group derived from a diarylalkane, from the viewpoint of achieving high resistance to hydrolysis and thermal stability. Specific examples of such compounds of formula (2) include bisphenol A bis(diphenylphosphate) and bisphenol A bis(dicresylphosphate).

Further preferred examples of compounds of formula (a) include an oligomeric aromatic phosphoric ester represented by the following formula (5):



wherein each of a, b, c, d and e independently represents an integer of from 0 to 3; each of R^1 to R^5 independently represents a C_1 - C_{10} hydrocarbon group; and n represents an integer of from 1 to 3.

With respect to the oligomeric aromatic phosphoric esters represented by formula (5), it is preferred that a is 0; each of b, c, d and e is 2; and the substitution positions of each of the di-substituted aromatic rings are 2- and 6-positions. Such a compound of



formula (5) having di-substituted aromatic rings is in a solid state at room temperature; however, since the crystallization rate of the compound is relatively small, the compound is rendered liquid by adding there-
5 to another non-polymerizable liquid substance which is in a liquid state at room temperature, such as TNPP. The compound of formula (5) having di-substituted aromatic rings can be produced by known methods, such as a method disclosed in Unexamined Japanese Patent
10 Application Laid-Open Specification No. 5-1079. Specifically, the compound of formula (5) having di-substituted aromatic rings can be produced by, for example, a method in which a monofunctional phenol, which is substituted at the 2- and 6-positions thereof with
15 C₁ to C₁₀ hydrocarbon groups, is reacted with an oxyhalogenated phosphorus in the presence of a Lewis acid catalyst to obtain a diarylphosphoro halide and, then, the obtained diarylphosphoro halide is reacted with a
20 bifunctional phenol in the presence of a Lewis acid catalyst.

Examples of foaming agents usable as the non-polymerizable liquid substance (B) include azo foaming agents, N-nitroso foaming agents and sulfonylhydrazide.

25 Examples of lubricants usable as the non-polymerizable liquid substance (B) include hydrocarbon lubric-

ants, such as a liquid paraffin; fatty acid lubricants; fatty amide lubricants; alcohol lubricants; and metallic soaps.

Further, in the present invention, even if the
5 non-polymerizable liquid substance (B) is in the form of a powder or a solid mass at any temperature in the range of from 25 to 300 °C, it may be used as the non-polymerizable liquid substance (B) as long as it is soluble in the non-polymerizable liquid substance (B)
10 so that it is rendered liquid at the above-mentioned range of temperature.

In the present invention, for enhancing the compatibility between the thermoplastic resin (A) and the non-polymerizable liquid substance (B), either another
15 non-polymerizable liquid substance which is different from the above-mentioned non-polymerizable liquid substance (B) or a surface active agent which is soluble in or has an interaction with the non-polymerizable liquid substance (B) can be employed as a compatibility
20 agent. Even a solid surface active agent, which does not satisfy the requirements for the non-polymerizable liquid substance (B), can be employed as long as it is soluble in the non-polymerizable liquid substance (B).

In the present invention, when the thermoplastic
25 resin (A) (such as polyphenylene ether and the like) is

dissolved in an oligomer type non-polymerizable liquid substance which is a di-or higher-mer (such as an aromatic phosphoric ester condensation product or the like) selected among various non-polymerizable liquid substances (B), a resin composition which has excellent appearance, light resistance and mechanical properties can be obtained by the co-use of a monomer type non-polymerizable liquid substance (such as the above-mentioned TNPP and the like) serving also as a compatibility agent. The content of the monomer type non-polymerizable liquid substance in the non-polymerizable liquid substance (B) is preferably from 0.01 to 50 % by weight, more preferably from 0.1 to 30 % by weight, still more preferably from 1 to 20 % by weight, most preferably from 2 to 10 % by weight.

Examples of compatibility agents which can be used for enhancing the compatibility between the thermoplastic resin (A) and the non-polymerizable liquid substance (B) include an aliphatic hydrocarbon, a higher fatty acid, a higher fatty amide and a higher fatty alcohol. The compatibility agent is contained in the non-polymerizable liquid substance (B) in an amount of from 0.01 to 20 % by weight, preferably from 0.1 to 10 % by weight, more preferably from 1 to 5 % by weight, based on the total weight of the non-polymerizable

liquid substance (B) and the compatibility agent.

Examples of aliphatic hydrocarbons as the compatibility agent include liquid paraffin, natural paraffin, microwax, polyolefin wax and synthetic paraffin; partial oxidation products thereof; fluorides thereof; and chlorides thereof.

Examples of higher fatty acids include saturated fatty acids; and unsaturated fatty acids, such as ricinoleic acid, ricinoelaidic acid, and 9-oxy-12-octadecenoic acid.

Examples of higher fatty amides include saturated fatty monoamides, such as phenylstearamide, methylolstearamide, methylolbehenamide; N,N'-disubstituted monoamides, such as coconut oil fatty diethanolamide, lauric diethanolamide, oleic diethanolamide; saturated fatty bisamides, such as methylenebis(12-hydroxyphenyl)stearamide, ethylenebis(stearamide), ethylenebis(12-hydroxyphenyl)stearamide, hexamethylenebis(12-hydroxyphenyl)stearamide; and aromatic bisamides, such as m-xylylenebis(12-hydroxyphenyl)stearamide.

Examples of higher fatty alcohols as the compatibility agent include monohydric alcohols, such as stearyl alcohol and cetyl alcohol; polyhydric alcohols, such as sorbitol and mannitol; polyoxyethylene dodecy-

lamine; polyoxyethylene octadecylamine; allylated ethers having a polyalkylene ether unit, such as polyoxyethylene allylated ether; polyoxyethylene alkyl ethers, such as polyoxyethylene lauryl ether, polyoxyethylene dodecyl ether, polyoxyethylene cetyl ether, polyoxyethylene stearyl ether, and polyoxyethylene oleyl ether; polyoxyethylene alkylphenyl ethers, such as polyoxyethylene octylphenyl ether and polyoxyethylene nonylphenyl ether; dihydric alcohols having a polyoxyethylene unit, such as polyepichlorohydrin ether, polyoxyethylene bisphenol A ether, polyoxyethylene ethylene glycol ether, polyoxypropylene bisphenol A ether and polyoxyethylene polyoxypropylene glycol ether.

The solid substance (C) used in the present invention, which is a crystalline solid or an amorphous solid at a temperature of 300 °C, imparts desirable properties to the resin composition. The solid substance (C) is dispersed in, swelled with or dissolved in the thermoplastic resin (A) and the non-polymerizable liquid substance (B). With respect to the type of the solid substance (C), there is no particular limitation as long as they satisfy the above-mentioned requirements. Examples of solid substances (C) include an anti-block agent, an oil absorptive compound, a heat

stabilizer and a light stabilizer.

The amount of the solid substance (C) is preferably from 0.000001 to 100 parts by weight, more preferably from 0.01 to 20 parts by weight, most preferably from 0.1 to 10 parts by weight, relative to 100 parts by weight of the thermoplastic resin (A).

The anti-block agent as the solid substance (C) is a component which is employed for improving the handling characteristics (such as melt-adhesion preventive properties) of the resin composition which is composed of the thermoplastic resin (A) and the non-polymerizable liquid substance (B). Examples of anti-block agents include organic acid metal salts, such as metal soaps; metal oxides; inorganic acid salts; waxes; minerals, such as silica, alumina, talc and diatomaceous earth; metals, such as iron; fibers, such as cotton and pulp. These solid substances may be employed either individually or in combination. Among these solid substances, it is preferred to use those powdery substances which have a number average particle diameter of from 0.01 to 300 μm . When a powdery substance having a number average particle diameter of less than 0.01 μm is used, agglomeration of these powdery substances is likely to disadvantageously occur. When a powdery substance having a number aver-

age particle diameter of more than 300 μm is used, the rate of the dissolution of the thermoplastic resin (A) into the non-polymerizable liquid substance (B) is likely to be decreased.

5 Examples of organic acid metal salts as the anti-block agent include lithium salts, copper salts, beryllium salts, magnesium salts, calcium salts, strontium salts, barium salts, zinc salts, cadmium salts, aluminum salts, cerium salts, titanium salts, zirconium
10 salts, lead salts, chromium salts, manganese salts, cobalt salts and nickel salts of organic carboxylic acids as mentioned below: straight chain saturated fatty acids, such as butyric acid, n-caproic acid, pelargonic acid, n-capric acid, undecanoic acid, lauric
15 acid, myristic acid, palmitic acid, stearic acid and arachic acid; straight chain unsaturated fatty acids, such as oleic acid, linoleic acid and linolenic acid; branched fatty acids, such as isostearic acid; hydroxyl group-containing fatty acids, such as ricinoleic acid
20 and 12-hydroxystearic acid; benzoic acid; naphthenic acid; abietic acid; and resin acids of rosin, such as dextropimaric acid.

 Examples of metal oxides as the anti-block agent include zinc oxide, alumina (aluminum oxide), aluminum
25 silicate, silica (silicon oxide), calcium oxide, calci-

um carbonate, titanium oxide, iron oxide, barium oxide, manganese dioxide and magnesium oxide. Especially, alumina and silica which are already mentioned above as minerals are preferred.

5 Examples of inorganic acid salts as the anti-block agent include barium titanate, barium sulfate, manganese carbonate, magnesium carbonate and calcium carbonate.

10 Examples of waxes as the anti-block agent include organosiloxane wax, polyolefin wax and polycaprolactone.

15 As the anti-block agent, hydrophobic metal oxides, such as hydrophobic silica and hydrophobic alumina are especially preferred. For producing hydrophobic metal oxides, the surfaces of metal oxides are chemically or physically treated with a silane coupling agent, such as an alkylalkoxysilane or an alkyl halogenated silane; a polysiloxane, such as dimethylsiloxane; synthetic wax or natural wax, to thereby inactivate the surfaces of the metal oxides.

20 The oil absorptive compound as the solid substance (C) is a component which is employed for improving the dissolution of the thermoplastic resin (A) into the non-polymerizable liquid substance (B). It is preferred to use an oil absorptive compound having a

25

number average particle diameter of from 0.001 to 1000 μm from a viewpoint of the effect of improving the dissolution characteristics of the thermoplastic resin (A) into non-polymerizable liquid substance (B).

5 Examples of oil absorptive compounds include powdery substances or porous powdery substances, such as those which are made of a thermoplastic resin, a thermosetting resin, a metal, a metal oxide, flame retardants, reinforcing agents and ceramics.

10 As one of the oil absorptive compounds as the solid substance (C), there can be mentioned a thermoplastic resin having a substituent which has an affinity to the non-polymerizable liquid substance (B). The thermoplastic resin preferably has a number average
15 particle diameter of from 0.001 to 1000 μm . It is preferred that such thermoplastic resins comprise at least one monomer (containing a polymerizable unsaturated group) selected from the group consisting of: unsaturated carboxylic acid esters, such as alkyl
20 (metha)acrylate; (metha)acrylamides which have a hydrocarbon group, such as a dialkyl (metha)acrylamide; α -olefins, such as 1-hexene; alicyclic vinyl compounds, such as vinylcyclohexane; allyl ethers which have a hydrocarbon group, such as dodecyl allyl ether; vinyl
25 esters which have a hydrocarbon group, such as vinyl

caproate; vinyl ethers which have a hydrocarbon group, such as butyl vinyl ether; and aromatic vinyl compounds, such as styrene.

5 The above-mentioned thermoplastic resin as the oil absorptive compound may comprise a small amount of a crosslinkable monomer having at least two polymerizable unsaturated groups as long as it holds thermoplasticity. Examples of such crosslinkable monomers include (metha)acrylates of a divalent alcohol, such as ethylene
10 glycol di(metha)acrylate; (metha)acrylates of a polyhydric alcohol, such as glycerol or tetramethylolmethane; and divinylbenzene.

Examples of thermosetting resins as the oil absorptive compound include a copolymer of an aromatic
15 vinyl monomer (e.g. a styrene-divinylbenzene copolymer) and a crosslinkable monomer having a plurality of unsaturated bonds; a copolymer of (metha)acrylate and the above-mentioned crosslinkable monomer; a phenolic resin; a melamine resin; an epoxy resin; an unsaturated
20 polyester resin; a furan resin; a urea resin; a xylene-formaldehyde resin; a silicone resin; a diallyl phthalate resin; and an aramid.

In the present invention, for improving the compatibility between the thermoplastic resin (A) and the
25 non-polymerizable liquid substance (B) during the

mixing therebetween, it is preferred that the mixing between components (A) and (B) is performed in the presence of a solvent (D) which has high compatibility with both components (A) and (B), and, after the mixing, the solvent (D) is removed from the mixture in which component (A) is dissolved in or swelled with component (B).

Examples of solvents (D) include hydrocarbons, halogenated hydrocarbons, alcohols, phenols, ethers, acetals, ketones, fatty acids, acid anhydrides, esters, nitrogen compounds, sulfur compounds, and inorganic solvents. It is preferred that the amount of the solvent (D) is from 0.1 to 100 parts by weight, more preferably from 1 to 50 parts by weight, most preferably from 5 to 20 parts by weight, relative to 100 parts by weight of the thermoplastic resin (A).

Of these solvents, preferred are those which are selected from ethers, acetals and hydrocarbons and which have a boiling point in the range of from 200 to 300 °C. Examples of ethers include diphenyl ether and butyl phenyl ether. Examples of acetals include 1,1-diethoxyethane and 2,2-diethoxypropane. Examples of hydrocarbon solvents include pentane, 2-methyl butane, hexene, 2-methyl pentane, 2,2-dimethyl butane, 2,3-dimethyl butane, heptane, octane, 2,2,3-trimethyl

pentane, isooctane, nonane, 2,2,5-trimethyl hexane,
decane, dodecane, unsaturated aliphatic hydrocarbons,
benzene, toluene, xylene, ethylbenzene, amylbenzene,
isopropylbenzene, cumene, mesitylene, naphthalene,
5 tetralin, butylbenzene, p-cymene, cyclohexylbenzene,
diethylbenzene, pentylbenzene, dipentylbenzene, dode-
cylbenzene, biphenyl, cyclopentane, methylcyclopentane,
cyclohexane, methylcyclohexane, ethylcyclohexane, p-
menthane, bicyclohexyl, cyclohexene, α -pinene, dipen-
10 tene, decalin, petroleum ether, petroleum benzine,
petroleum naphtha, ligroin, industrial gasoline, kero-
sine, solvent naphtha, camphor oil, turpentine oil,
pine oil and the like. Of these, preferred are those
which have a boiling point in the range of from 200 to
15 300 °C, such as amylbenzene, cyclohexylbenzene, dode-
cylbenzene, pine oil and biphenyl.

When the resin composition of the present inven-
tion is intended to be produced in the form of pellets,
it is preferred to use a method in which the non-polym-
20 erizable liquid substance (B) is heated, and the ther-
moplastic resin (A) and the non-polymerizable liquid
substance (B) are mixed together while heating, and the
resultant is cooled, followed by pelletization. In
this method, it is necessary that the heating be per-
25 formed at a temperature in the range of from 25 to

300 °C.

It is preferred that the mixing of the thermoplastic resin (A) and the non-polymerizable liquid substance (B) while heating is performed by using a dissolution vessel having a stirrer, or an extruder. With respect to the mixing method using an extruder, this method is different from a conventional melt-extrusion method in that the thermoplastic resin which has been dissolved in or swelled with the liquid additive is fed to an extruder. With respect to the method for cooling and pelletizing the resin composition comprising components (A) and (B), there can be employed a method in which the resin composition is passed through a vessel containing cooling water and the cooled resin composition is pelletized by means of a pelletizer having a rotary cutter (see Fig. 1), and a method in which the pelletization is performed by using a conveyor belt type cooler equipped with a cooling system (developed by Sandvik Co., Sweden) (see Fig. 2).

With respect to the extruder used for mixing the thermoplastic resin (A) and the non-polymerizable liquid substance (B), there is no particular limitation, and a conventional extruder for melt-extrusion can be employed as long as components (A) and (B) can be mixed together under an appropriate shearing force

for an appropriate residence time so as to cause component (A) to be dissolved in or swelled with component (B) without bringing any of components (A) and (B) into the molten state (as defined in the present invention).

5 Another preferred embodiment of the method for producing the resin composition of the present invention is a method in which the heat-melting of the non-polymerizable substance (B) for rendering liquid is performed simultaneously with the heat-mixing of the
10 thermoplastic resin (A) and the non-polymerizable liquid substance (B). Specifically, in this preferred embodiment, the non-polymerizable substance (B) is charged in a vessel and heated at a temperature in the range of from 25 to 300 °C and the thermoplastic resin
15 (A) is gradually added thereto while stirring so as to cause component (A) to be dissolved in or swelled with component (B), and the resultant is cooled, followed by pelletization. It is preferred that the mixing of components (A) and (B) is performed in the presence of
20 (D) an organic solvent, and, after the mixing, the organic solvent (D) is removed.

 The thermoplastic resin composition of the present invention as such can be used as a molding material for use in, for example, an injection molding. Alterna-
25 tively, the resin composition can also be used as a

masterbatch for preparing a molding material, wherein the masterbatch is added to a thermoplastic resin material which is the same as or different from the thermoplastic resin (A) and the resultant mixture is
5 subjected to melt extrusion at a temperature, for example, in the range of from 100 to 300 °C. In this instance, various liquid additives and solid additives can also be added along with the masterbatch, wherein these liquid additives and solid additives may include
10 those which are the same as the non-polymerizable liquid substance (B) and the solid substance (C).

With respect to the yardstick for selecting an appropriate combination of the thermoplastic resin (A) used in the masterbatch and the thermoplastic resin
15 material to be melt-extruded together with the masterbatch, the selection can be made according to the glass transition temperature (T_g) thereof in case of an amorphous polymer, and the selection can be made according to the melting temperature (T_m) thereof in case
20 of a crystalline polymer. The T_g or T_m of the thermoplastic resin (A) used in the masterbatch is preferably 100 °C or more, more preferably 140 °C or more, most preferably 200 °C or more. On the other hand, the T_g or T_m of the thermoplastic resin material to be melt-
25 extruded together with the masterbatch is preferably in

the range of 85 to 200 °C, more preferably from 85 to 120 °C. When both the thermoplastic resin (A) used in the masterbatch and the thermoplastic resin material to be melt-extruded together with the masterbatch satisfy the above-mentioned respective preferred ranges of T_g or T_m, the advantageous effects of the present invention are most effectively exhibited.

With respect to the thermoplastic resin to be melt-extruded together with the masterbatch comprising the resin composition of the present invention, there is no particular limitation. Examples of thermoplastic resins include a polystyrene resin, a polyphenylene ether resin, a polyolefin resin, polyvinyl chloride resin, a polyamide resin, a polyester resin, a polyphenylene sulfide resin, a polycarbonate resin, a polymethacrylate resin, a polyacetal resin, a polyarylate resin, and a polysulfone resin. These thermoplastic resins can be used individually or in combination. Of these thermoplastic resins, a polystyrene resin and a polycarbonate resin are preferred. Examples of polystyrene resins include rubber-modified styrene polymers.

In the present invention, various solid substances can be added to the masterbatch comprising the resin composition of the present invention. Examples of

solid substances include a flame retardant other than the non-polymerizable liquid substance (B), a flame retardant auxiliary, a thermoplastic elastomer, an antioxidant, a heat stabilizer (such as a tin compound type heat stabilizer), a light resistance improver, a fluidity improver, a lubricant, a filler, a reinforcing agent (such as a glass fiber), a colorant (such as a dye or a pigment), a foaming agent, an antistatic agent, and a mold release agent. Hereinbelow is given a detailed explanation on these solid substances which can be added to the masterbatch.

Examples of flame retardants other than the non-polymerizable liquid substance (B) include a halogen-containing flame retardant, a phosphorus-containing flame retardant and an inorganic flame retardant.

Specific examples of halogen-containing flame retardants include a bisphenol halide, an aromatic halide, a polycarbonate halide, an aromatic vinyl polymer halide, a cyanurate halide-containing resin and a polyphenylene ether halide. Of these, decabromodiphenyloxide, tetrabromobisphenol A, an oligomer of tetrabromobisphenol A, a bisphenol bromide-containing phenoxy resin, a bisphenol bromide-containing polycarbonate, polystyrene bromide, crosslinked polystyrene bromide, polyphenylene oxide bromide, polydibromopheny-

lene oxide, a condensation product of decabromodiphenyl oxide bisphenol, a halogen-containing phosphate, a fluororesin and the like are preferred.

5 Examples of phosphorus-containing flame retardants include red phosphorus, an inorganic phosphate and the like.

10 Examples of red phosphorus flame retardants include not only an ordinary red phosphorus but also a red phosphorus product which has been coated with a film of at least one metal hydroxide selected from the group consisting of aluminum hydroxide, magnesium hydroxide, zinc hydroxide and titanium hydroxide. As another red phosphorus product, there can be mentioned those which have been coated with a film composed of a mixture of a metal hydroxide (selected from the group consisting of aluminum hydroxide, magnesium hydroxide, zinc hydroxide and titanium hydroxide) and a thermosetting resin. As still another red phosphorus product, there can be mentioned those which have been coated with a double-film layer comprising an inner film composed of a metal hydroxide (selected from the group consisting of aluminum hydroxide, magnesium hydroxide, zinc hydroxide and titanium hydroxide) and an outer film composed of a thermosetting resin.

25 A representative example of inorganic phosphates

usable as the phosphorus-containing flame retardant is ammonium polyphosphate.

Examples of inorganic flame retardants include aluminum hydroxide, magnesium hydroxide, dolomite, hydrotalcite, calcium hydroxide, barium hydroxide, basic magnesium carbonate, zirconium hydroxide, a hydrate of an inorganic metal compound (such as tin oxide hydrate), zinc borate, zinc metaborate, barium metaborate, zinc carbonate, magnesium carbonate, calcium carbonate, barium carbonate and the like. These compounds can be used individually or in combination. Of these, as an inorganic flame retardant which has not only excellent flame retardancy but is also commercially advantageous, there can be employed at least one compound selected from the group consisting of magnesium hydroxide, aluminum hydroxide, basic magnesium carbonate and hydrotalcite.

With respect to the amount of the flame retardant other than the non-polymerizable liquid substance (B), it is preferred that the amount is from 1 to 100 parts by weight, more preferably from 1 to 50 parts by weight, still more preferably from 3 to 20 parts by weight, most preferably from 5 to 15 parts by weight, relative to 100 parts by weight of the masterbatch comprising the resin composition of the present inven-

tion.

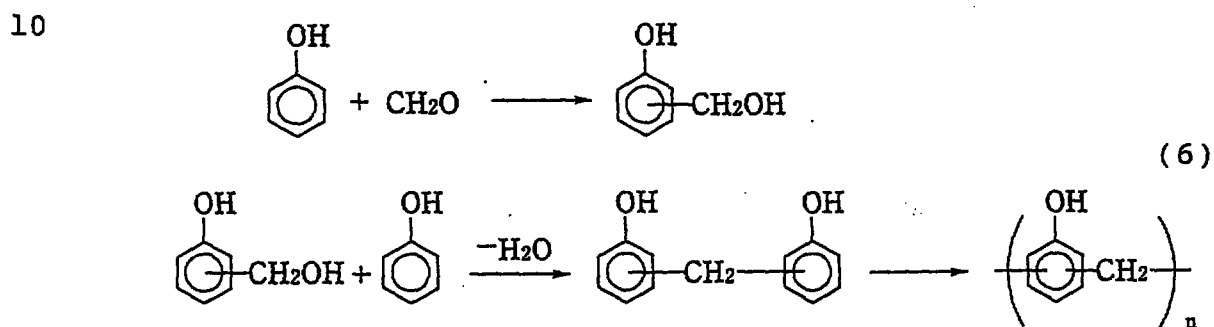
Further, if desired, at least one flame retardant auxiliary may be added to the masterbatch comprising the resin composition of the present invention, which
5 is selected from the group consisting of a novolak resin, a triazine skeleton-containing compound, a silicone resin, a silicone oil, a fluororesin, an aramid fiber and a polyacrylonitrile fiber.

With respect to the amount of the above-mentioned
10 flame retardant auxiliary, it is preferred that the amount is from 0.001 to 40 parts by weight, more preferably from 1 to 20 parts by weight, most preferably from 5 to 10 parts by weight, relative to 100 parts by weight of the masterbatch.

15 When the above-mentioned novolak resin is used in combination with the above-mentioned aromatic phosphoric ester, the novolak resin also acts to improve the melt fluidity and heat resistance of the resin composition of the present invention; however, the compatibility
20 between the thermoplastic resin (A) and the aromatic phosphoric ester (B) is slightly lowered. The novolak resin is a thermoplastic resin which can be obtained by subjecting a phenol and an aldehyde to a condensation reaction in the presence of an acid catalyst,
25 such as sulfuric acid or hydrochloric acid. For

example, the method for obtaining a novolak resin is described at pages 437 to 455 of "Kobunshi Jikkengaku 5, Jushukugo to Jufuka (Experimental Polymer Chemistry, Series 5, Condensation Polymerization and Polyaddition)" (which was published by Kyoritsu Shuppan Co., Ltd., Japan).

A scheme (6), illustrating a typical example of the production of novolak resins, is shown below.



Specific examples of phenols used for the production of novolak resins include phenol; o-cresol; m-cresol; p-cresol; 2,5-dimethyl-, 3,5-dimethyl-, 2,3,5-trimethyl-, 3,4,5-trimethyl-, p-t-butyl-, p-n-octyl-, p-stearyl-, p-phenyl-, p-(2-phenylethyl)-, o-isopropyl-, p-isopropyl-, m-isopropyl-, p-methoxy- and p-phenoxy-phenols; pyrocatechol; resorcinol; hydroquinone; salicylaldehyde; salicylic acid; p-hydroxybenzoic acid; methyl p-hydroxybenzoate; p-cyano- and o-cyanophenols;

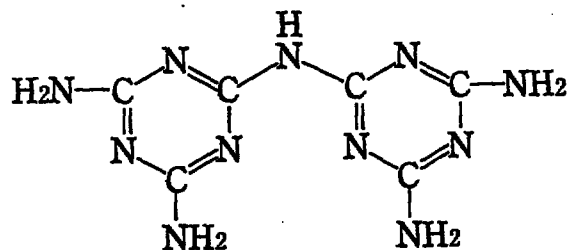
p-hydroxybenzenesulfonic acid; p-hydroxybenzenesulfon-
amide; cyclohexyl p-hydroxybenzenesulfonate; (4-hydroxy-
phenyl)phenylphosphinic acid; methyl (4-hydroxyphenyl)
phenylphosphinate, 4-hydroxyphenylphosphonic acid;
5 ethyl 4-hydroxyphenylphosphonate; and diphenyl 4-hy-
droxyphenylphosphonate.

Specific examples of aldehydes used for the pro-
duction of novolak resins include formaldehyde, acetal-
dehyde, n-propanal, n-butanal, isobutyl aldehyde, 3-
10 methyl-n-butanal, benzaldehyde, p-tolylaldehyde and 2-
phenylacetoaldehyde.

The above-mentioned triazine skeleton-containing
compound acts as a flame retardant auxiliary for en-
hancing the flame retarding effects of a phosphorus
15 flame retardant. Specific examples of triazine skele-
ton-containing compounds include melamine, melam repre-
sented by formula (7), melem represented by formula
(8), mellon (a product obtained by deammonium reaction
of three molecules of melem at 600 °C or higher, gener-
20 ating three molecules of ammonia), melamine cyanurate
represented by formula (9), melamine phosphate repre-
sented by formula (10), succinoguanamine represented by
formula (11), adipoguanamine, methylglutaroguanamine, a
melamine resin having a structure represented by formu-
25 la (12), and a BT resin having a structure represented

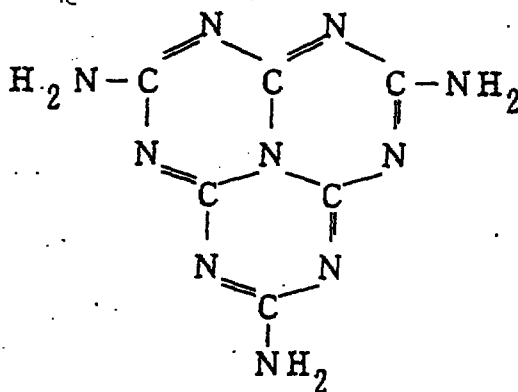
by formula (13). Of these, melamine cyanurate is preferred from the viewpoint of the resistance to volatilization.

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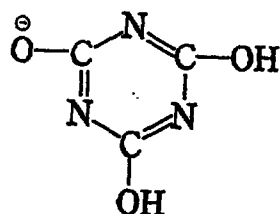
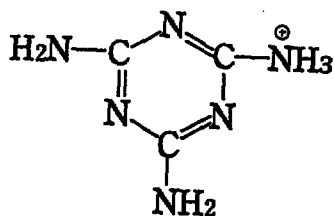
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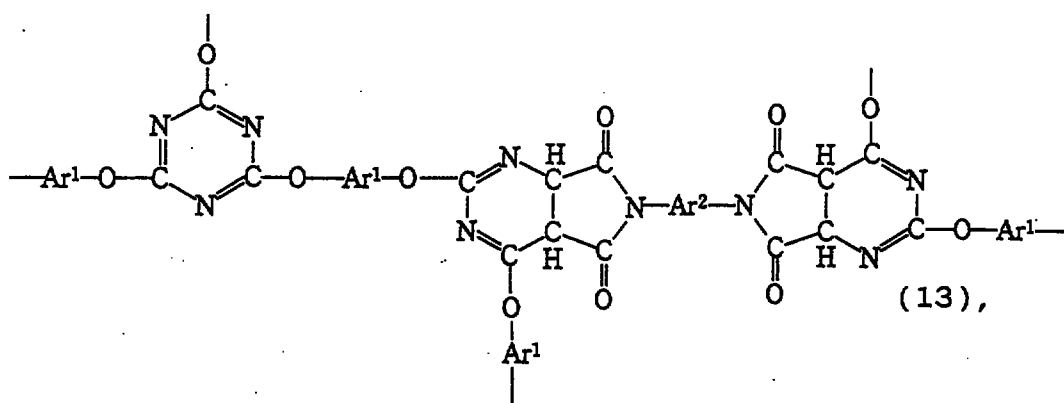
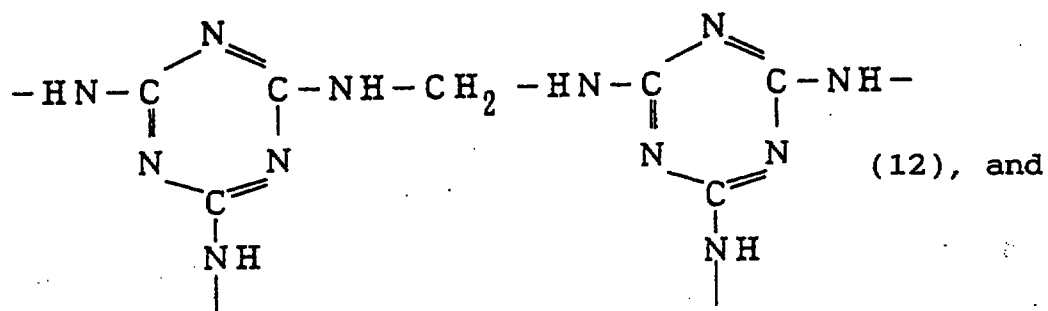
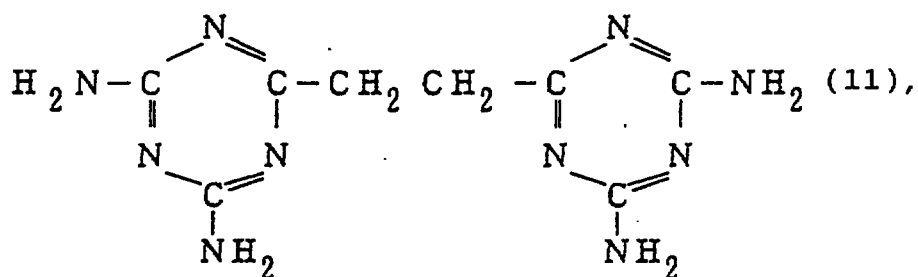
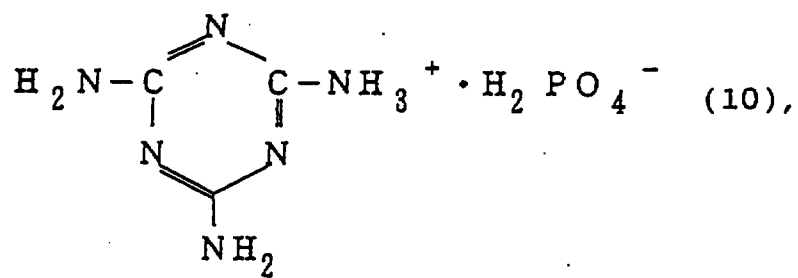
(8),

20



(9),

25



wherein each Ar^1 represents

, and

each Ar^2 represents

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5

The above-mentioned silicone resin used as a flame retardant auxiliary is one which has a three-dimensional network structure which is obtained by combining the structural units SiO_2 , $\text{RSiO}_{3/2}$, RSiO and $\text{RSiO}_{1/2}$. In these structural units, each R independently represents an alkyl group, such as a methyl group, an ethyl group or a propyl group, an aromatic group, such as a phenyl group or a benzyl group, or a vinyl group-containing substituent group obtained by introducing a vinyl group to the above-mentioned alkyl group or the above-mentioned aromatic group. Especially, a silicone resin, in which R is the above-mentioned vinyl group-containing substituent group, obtained by introducing a vinyl group to the above-mentioned alkyl group or the above-mentioned aromatic group, is preferred.

20

The above-mentioned silicone resin can be obtained by hydrolyzing organohalosilanes corresponding to the above-mentioned structural units and subsequently polymerizing the hydrolyzed products.

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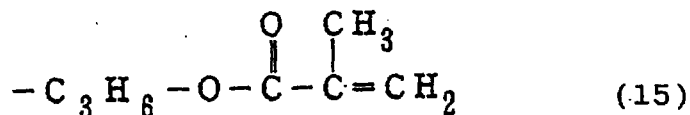
A silicone oil used as a flame retardant auxiliary



is a polydiorganosiloxane comprising a structural unit represented by the following formula (14):



In formula (14) above, each R independently represents a C₁-C₈ alkyl group, a C₆-C₁₃ aryl group, a vinyl group-containing group represented by formula (15) or a vinyl group represented by formula (16).



As a silicone oil used as a flame retardant auxiliary, especially preferred is a silicone oil comprising structural units of formula (14) containing as R at least one substituent selected from the group consisting of the vinyl group-containing group of formula (15) and the vinyl group of formula (16).

With respect to the viscosity of such a vinyl group-containing silicone oil, it is preferred that the

viscosity is from 600 to 1,000,000 centistokes (as measured at 25 °C), more preferably 90,000 to 150,000 centistokes (as measured at 25 °C).

5 The above-mentioned fluoro-resin used as a flame retardant auxiliary is a resin containing fluorine atoms. Specific examples of fluoro-resins include polymonofluoroethylene, polydifluoroethylene, polytri-
fluoroethylene, polytetrafluoroethylene and a tetra-
fluoroethylene-hexafluoropropylene copolymer. If
10 desired, a fluorine-containing monomer which is a monomer selected from monomers contained in the above-mentioned fluoro-resins may be optionally polymerized or copolymerized with a monomer which is copolymerizable with the fluorine-containing monomer.

15 With respect to the method for producing a fluoro-resin, reference can be made to U.S. Patent Nos. 2,393,697 and 2,534,058. For example, a powdery polytetrafluoroethylene can be obtained by a method comprising polymerizing tetrafluoroethylene in an aqueous
20 medium at a temperature of from 0 to 200 °C under a pressure of from 7 to 70 kg/cm² using a radical initiator (such as ammonium persulfate or potassium persulfate), and coagulating or precipitating the resultant reaction mixture (in the form of a suspension, disper-
25 sion or emulsion).

For effectively preventing melt dripping of a molded product, it is preferred to use as the flame retardant auxiliary a fluororesin having a specific higher-order structure, which is obtained by melt-kneading the fluororesin together with a thermoplastic resin (the type and function of which are described below) at a temperature which is not less than the melting temperature of the fluororesin. For example, a polytetrafluoroethylene is preferably melt-kneaded together with such a thermoplastic resin at a temperature of 300 to 350 °C. By melting the fluororesin together with the thermoplastic resin, while applying a shearing force, at a temperature which is not less than the melting temperature of the fluororesin, the fluororesin is highly fibrillated, oriented and crystallized in the thermoplastic resin. As a result, a fluororesin having a specific higher-order structure can be obtained. Illustratively stated, the obtained fluororesin is in a branched fibril form, wherein the branched fibril comprises an oriented trunk fiber and branch fibers branched from the trunk fiber. Such a fluororesin in a branched fibril form is three-dimensionally entangled with the thermoplastic resin, so that melt dripping of a molded product can be prevented. As the thermoplastic resin to be melt-kneaded together with

the fluoro-resin, a rigid resin having a high melt viscosity (e.g., polyphenylene ether) is more preferred than a rubber-modified resin (e.g., a rubber-modified polystyrene) from the viewpoint of ease in applying a high shearing force to the fluoro-resin.

As a method for producing the above-mentioned fluoro-resin having a specific higher-order structure, for example, the following two methods can be mentioned. In the first method (two-step process), a fluoro-resin, a thermoplastic resin, and optionally a dispersant are melt-kneaded together at a temperature which is not less than the melting temperature of the fluoro-resin, thereby obtaining a masterbatch. Subsequently, the obtained masterbatch is melt-kneaded together with a thermoplastic resin and a flame retardant. In the second method (one-step process), use is made of an extruder which has a first extrusion zone and a second extrusion zone having a side feed opening. A fluoro-resin, a thermoplastic resin and optionally a dispersant are melt-kneaded together at a temperature which is not less than the melting temperature of the fluoro-resin in the first extrusion zone and then, further melt-kneaded in the second extrusion zone at a temperature which is lower than the temperature in the first extrusion zone while feeding a flame retardant

through the side feed opening.

With respect to the above-mentioned aramid fiber as the above-mentioned flame retardant auxiliary, it is preferred that the fiber has an average diameter of from 1 to 500 μm and an average fiber length of from 0.1 to 10 mm. The aramid fiber can be produced by a method in which isophthalamide or polyparaphenylene terephthalamide is dissolved in a polar solvent containing an amide, or sulfuric acid, and the resultant solution is subjected to dry spinning or wet spinning.

With respect to the above-mentioned polyacrylonitrile fiber as the above-mentioned flame retardant auxiliary, it is preferred that the average diameter is 1 to 500 μm and the average fiber length is 0.1 to 10 mm. With respect to the method for producing a polyacrylonitrile fiber, there can be mentioned a dry spinning method in which an acrylonitrile polymer is dissolved in a solvent (such as dimethylformamide), and the resultant solution is subjected to a spinning under the flow of air at 400 °C, and a wet spinning method in which an acrylonitrile polymer is dissolved in a solvent (such as nitric acid), and the resultant solution is subjected to spinning in water.

As examples of the above-mentioned thermoplastic elastomers, which may optionally be contained in the

resin composition of the present invention, there can be mentioned a polystyrene elastomer, a polyolefin elastomer, a polyurethane elastomer, a 1,2-polybutadiene elastomer, a polyvinyl chloride elastomer and the like. The thermoplastic polystyrene elastomer is particularly preferred. The amount of the thermoplastic elastomer is preferably from 1 to 30 parts by weight, more preferably from 2 to 20 parts by weight, most preferably 2 to 10 parts by weight, relative to 100 parts by weight of the above-mentioned masterbatch.

The above-mentioned thermoplastic polystyrene elastomer is selected from a block copolymer comprised of aromatic vinyl compound units and conjugated diene compound units and a hydrogenated block copolymer obtained by partially hydrogenating the conjugated diene moiety of the above block copolymer. Examples of aromatic vinyl monomers usable for producing the above-mentioned block copolymer include styrene, α -methylstyrene, p-methylstyrene, p-chlorostyrene, p-bromostyrene, 2,4,5-tribromostyrene and the like. Styrene is most preferred, but styrene may be copolymerized with other aromatic vinyl monomers mentioned above.

As examples of conjugated diene monomers usable for producing the above-mentioned block copolymer, 1,3-

butadiene, isoprene and the like can be mentioned.

With respect to the block configuration of the above-mentioned block copolymer, the block copolymer is preferably a linear block copolymer having a configuration of, for example, SB, S(BS)_n or S(BSB)_m, or a star-shaped block copolymer having a conjugated diene at the linkage center and having a configuration of (SB)_pX, wherein S represents a polymer block comprised of aromatic vinyl compound units, B represents a polymer block comprised of conjugated diene compound units and/or a partially hydrogenated product thereof, X represents a coupling agent residue (e.g., silicon tetrachloride, tin tetrachloride, a polyepoxy compound), n represents an integer of from 1 to 3, m represents an integer of 1 or 2, p represent an integer of from 3 to 6. Among them, linear block copolymers having a 2-block configuration "SB", a 3-block configuration "SBS", and a 4-block configuration "SBSB" are preferred.

The resin composition of the present invention may optionally contain at least one mold release agent selected from the group consisting of a higher saturated aliphatic carboxylic acid and a metal salt thereof, a carboxylic ester wax, an organosiloxane wax, a polyolefin wax and a polycaprolactone wax.

The amount of the mold release agent is preferably from 0.01 to 5 parts by weight, more preferably 0.1 to 5 parts by weight, most preferably 0.3 to 1 part by weight, relative to 100 parts by weight of the above-mentioned masterbatch.

Among the above-mentioned mold release agents, one or more compounds selected from the group consisting of higher saturated aliphatic carboxylic acids and metal salts thereof are preferred.

As preferred examples of higher saturated aliphatic carboxylic acids, there can be mentioned C₁₂-C₄₂ straight chain saturated monocarboxylic acids, such as lauric acid, myristic acid, palmitic acid, stearic acid, behenic acid, montanic acid and the like.

Preferred examples of metal salts which are those of the above-mentioned higher saturated aliphatic carboxylic acids include lithium salts, sodium salts, potassium salts, magnesium salts, calcium salts, aluminum salts, zinc salts and the like. Among these, zinc stearate, magnesium stearate, calcium stearate and aluminum stearate are preferred.

The resin composition of the present invention may optionally contain at least one fluidity improver selected from the group consisting of a copolymer comprising aromatic vinyl compound units and acrylic

ester compound units, an aliphatic hydrocarbon, a higher fatty acid, a higher fatty acid ester, a higher fatty acid amide, a higher aliphatic alcohol and a metallic soap.

5. The amount of the fluidity improver is preferably from 0.1 to 20 parts by weight, more preferably 0.5 to 10 parts by weight, most preferably 1 to 5 parts by weight, relative to 100 parts by weight of the above-mentioned masterbatch.

- 10 Examples of aromatic vinyl monomer units in the above-mentioned aromatic vinyl-acrylic ester copolymer as a fluidity improver include those derived from styrene, α -methylstyrene, p-methylstyrene, p-chlorostyrene, p-bromostyrene, 2,4,5-tribromostyrene and the like. Styrene is most preferred, but styrene may be
15 copolymerized with other aromatic vinyl monomers mentioned above.

- As examples of acrylic ester monomer units in the above-mentioned aromatic vinyl-acrylic ester copolymer
20 as a fluidity improver, there can be mentioned those derived from acrylates which have an alkyl group having 1 to 8 carbon atoms, e.g., methyl acrylate, butyl acrylate and the like.

- The acrylic ester compound unit content of the
25 above-mentioned aromatic vinyl-acrylic ester copolymer

as a fluidity improver is preferably from 3 to 40 % by weight, more preferably from 5 to 20 % by weight. It is preferred that the above-mentioned copolymer has a solution viscosity (which is a yardstick of the molecular weight) of from 2 to 10 cp (centipoises) as measured in a 10 % by weight solution in methyl ethyl ketone at 25 °C. If the solution viscosity is less than 2 cp, the impact strength of the resin composition is lowered. On the other hand, when the solution viscosity is larger than 10 cp, the improving effect on the fluidity is reduced.

The aliphatic hydrocarbon as a fluidity improver can be selected from liquid paraffin, natural paraffin, microwax, polyolefin wax, synthetic paraffin, and partial oxidation products thereof, fluorides thereof, chlorides thereof and the like.

The higher fatty acid as a fluidity improver can be selected from saturated fatty acids other than those which are mentioned above as mold release agents. Specifically, as a fluidity improver, there can be used ricinoleic acid, ricinoelaidic acid, 9-oxy-12-octadecenoic acid; and the like.

The higher fatty acid ester as a fluidity improver can be selected from monohydric alcohol esters of fatty acids, e.g. methyl phenylstearate and butyl phenylstea-

rate; monohydric alcohol esters of polybasic acids, e.g. diphenylstearyl phthalate; sorbitan esters, e.g. sorbitan monolaurate, sorbitan monostearate, sorbitan monooleate, sorbitan sesquioleate, sorbitan trioleate, polyoxyethylene sorbitan monolaurate, polyoxyethylene sorbitan monopalmitate, polyoxyethylene sorbitan monostearate, polyoxyethylene sorbitan monooleate; fatty acid esters of glycerin monomer, e.g. stearic acid monoglyceride, oleic acid monoglyceride, capric acid monoglyceride, behenic acid monoglyceride; fatty acid esters of polyglycerin, e.g. polyglycerin stearic acid ester, polyglycerin oleic acid ester, polyglycerin lauric acid ester; fatty acid esters having a polyalkylene ether unit, e.g. polyoxyethylene monolaurate, polyoxyethylene monostearate, polyoxyethylene monooleate; neopentyl polyol esters of fatty acids, e.g. neopentyl polyol distearate; and the like.

The higher fatty acid amide as a fluidity improver can be selected from monoamides of saturated fatty acids, e.g. phenylstearamide, methylol stearamide, methylol behenylamide; N,N'-disubstituted monoamides, e.g. coconut oil fatty acid diethanolamide, lauric acid diethanolamide, oleic acid diethanolamide; saturated fatty acid bisamides, e.g. methylene bis(12-hydroxyphenyl) stearamide, ethylene bisstearamide, ethylene

bis(12-hydroxyphenyl) stearamide, hexamethylene bis(12-hydroxyphenyl) stearamide; aromatic bisamides, e.g. m-xylylene bis(12-hydroxyphenyl) stearamide; and the like.

5 The higher aliphatic alcohol as a fluidity improver can be selected from monohydric alcohols, e.g. stearyl alcohol, cetyl alcohol; polyhydric alcohols, e.g. sorbitol, mannitol; polyoxyethylene dodecylamine; polyoxyethylene octadecylamine; allyl ethers having
10 polyalkylene ether units, e.g. polyoxyethylene allyl ether; polyoxyethylene alkyl ethers, e.g. polyoxyethylene lauryl ether, polyoxyethylene tridodecyl ether, polyoxyethylene cetyl ether, polyoxyethylene stearyl ether, polyoxyethylene oleyl ether; polyoxyethylene
15 alkylphenyl ethers, e.g. polyoxyethylene octylphenyl ether, polyoxyethylene nonylphenyl ether; dihydric alcohols having polyalkylene ether units, e.g. polyepichlorohydrin ether, polyoxyethylene bisphenol A ether, polyoxyethylene ethylene glycol, polyoxypropylene
20 lene bisphenol A ether, polyoxyethylene polyoxypropylene glycol ether; and the like.

 The resin composition of the present invention may optionally contain at least one light resistance improver selected from the group consisting of an ultraviolet
25 violet light absorber, a hindered amine light stabiliz-

er, an antioxidant, an active species capturing agent, a sunproofing agent, a metal inactivating agent, and a light quenching agent.

5 The amount of the light resistance improver is preferably from 0.05 to 20 parts by weight, more preferably 0.1 to 10 parts by weight, most preferably 0.1 to 5 parts by weight, relative to 100 parts by weight of the above-mentioned masterbatch.

10 The ultraviolet light absorber as a light resistance improver is a component which absorbs light energy and releases the absorbed light energy harmlessly as heat energy due to the transformation thereof into a keto form through intramolecular proton transfer (in the case of benzophenones and benzotriazoles) or due to
15 cis-trans isomerization (in the case of cyanoacrylates). Specific examples of UV light absorbers include 2-hydroxybenzophenones, e.g. 2,4-dihydroxybenzophenone, 2-hydroxy-4-methoxybenzophenone, 2-hydroxy-4-octoxybenzophenone, and 5,5'-
20 methylenebis(2-hydroxy-4-methoxybenzophenone); 2-(2'-hydroxyphenyl)benzotriazoles, e.g. 2-(2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-5'-t-octylphenyl)benzotriazole, 2-(2'-hydroxy-3',5'-di-t-butylphenyl)benzotriazole, 2-(2'-hydroxy-3',5'-di-t-butylphenyl)-5-chlorobenzotriazole, 2-(2'-hydroxy-3'-t-

25

butyl-5'-methylphenyl)-5-chlorobenzotriazole, 2-(2'-hydroxy-3',5'-dicumylphenyl)benzotriazole, and 2,2'-methylenebis(4-t-octyl-6-benzotriazolyl)phenol; benzoates, e.g. phenylsalicylate, resorcinol monobenzoate, 2,4-di-t-butylphenyl-3',5'-di-t-butyl-4'-hydroxybenzoate, and hexadecyl-3,5-di-t-butyl-4-hydroxybenzoate; substituted oxanilides, e.g. 2-ethyl-2'-ethoxyoxanilide and 2-ethoxy-4'-dodecyloxanilide; and cyanoacrylates, e.g. ethyl- α -cyano- β,β -diphenylacrylate and methyl-2-cyano-3-methyl-3-(p-methoxyphenyl)acrylate.

The hindered amine light stabilizer as a light resistance improver is a component which decomposes hydroperoxides produced by light energy into stable N-O $\cdot$ radicals, N-OR or N-OH, thereby providing light stability. Specific examples of hindered amine light stabilizers include 2,2,6,6-tetramethyl-4-piperidylstearate, 1,2,2,6,6-pentamethyl-4-piperidylstearate, 2,2,6,6-tetramethyl-4-piperidylbenzoate, bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)sebacate, tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-butanetetracarboxylate, tetrakis(1,2,2,6,6-pentamethyl-4-piperidyl)-1,2,3,4-butanetetracarboxylate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)-di(tridecyl)1,2,3,4-butanetetracarboxylate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)-2-butyl-2-

(3',5'-di-t-butyl-4-hydroxybenzyl)malonate, 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4-piperidinol/diethyl succinate polycondensate, 1,6-bis(2,2,6,6-tetramethyl-4-piperidylamino)hexane/dibromoethane polycondensate,
 5 1,6-bis(2,2,6,6-tetramethyl-4-piperidylamino)hexane/
 2,4-dichloro-6-t-octylamino-s-triazine polycondensate,
 1,6-bis(2,2,6,6-tetramethyl-4-piperidylamino)hexane/
 2,4-dichloro-6-morpholino-s-triazine polycondensate,
 and the like.

10 The antioxidant as a light resistance improver is
 a component for stabilizing peroxide radicals, such as
 a hydroperoxide radical, which is generated at the time
 of heat molding or at the time of light exposure, and
 for decomposing a generated peroxide, such as a hydro-
 15 peroxide. Examples of antioxidants include hindered
 phenol type antioxidants and peroxide decomposers. In
 preventing autoxidation, the hindered phenol type
 antioxidant acts as a radical chain inhibitor, and the
 peroxide decomposer decomposes a generated peroxide
 20 into a stable alcohol.

Specific examples of hindered phenol type antioxi-
 dants include 2,6-di-t-butyl-4-methylphenol, styrenated
 phenol, n-octadecyl-3-(3,5-di-t-butyl-4-hydroxyphenyl)
 propionate, 2,2'-methylene bis(4-methyl-6-t-
 25 butylphenol), 2-t-butyl-6-(3-t-butyl-2-hydroxy-5-meth-

ylbenzyl)-4-methylphenyl acrylate, 2-[1-(2-hydroxy-3,5-di-t-pentylphenyl)ethyl]-4,6-di-t-pentylphenyl acrylate, 4,4'-butylidene bis(3-methyl-6-t-butylphenol), 4,4'-thio-bis(3-methyl-6-t-butylphenol), alkylated bisphenol, tetrakis[methylene-3-(3,5-di-t-butyl-4-hydroxyphenyl) propionate]methane, 3,9-bis[2-{3-(3-t-butyl-4-hydroxy-5-methylphenyl)-propionyloxy}-1,1-dimethyl-ethyl]-2,4,8,10-tetraoxyspiro[5.5]undecane.

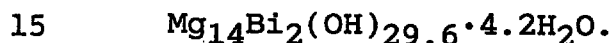
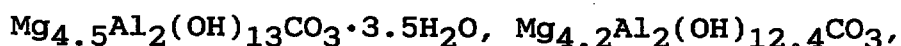
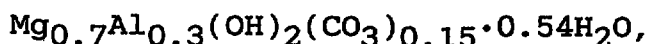
Specific examples of peroxide decomposers as the above-mentioned antioxidant include organic phosphorus type peroxide decomposers, such as trisnonylphenyl phosphite, triphenyl phosphite and tris(2,4-di-t-butylphenyl)phosphite; and organic thio type peroxide decomposers, such as dilauryl-3,3'-thiodipropionate, dimyristyl-3,3'-thiodipropionate, distearyl-3,3'-thiodipropionate, pentaerythrityltetrakis(3-laurylthiopropionate), ditridecyl-3,3'-thiodipropionate and 2-mercaptobenzimidazole.

The active species-capturing agent as the light resistance improver is a component used for capturing active species, such as free halogen radicals produced during heat molding or light exposure. Specific examples of active species-capturing agents include basic metal salts, such as calcium stearate and zinc stea-

rate; hydrotalcite; zeolite; magnesium oxide; organotin compounds; and organic epoxy compounds.

The hydrotalcite as the above-mentioned active species-capturing agent can be selected from basic carboxylates of metals, such as magnesium, calcium, zinc, aluminum and bismuth, and these basic carboxylates may or may not contain water. Either natural or synthetic products can be used. As a natural hydrotalcite, one having the structure of the formula:

$Mg_6Al_2(OH)_{16}CO_3 \cdot 4H_2O$ can be mentioned. Examples of synthetic hydrotalcites include:



As the above-mentioned zeolite, an A type zeolite represented by the formula: $Na_2O \cdot Al_2O_3 \cdot 2SiO_2 \cdot xH_2O$, and a zeolite, which is substituted with at least one metal selected from the group of metals belonging to Groups II and IV of the Periodic Table, can be mentioned. Examples of substituent metals include Mg, Ca, Zn, Sr, Ba, Zr, and Sn. Of these, Ca, Zn and Ba are especially preferred.

The organic epoxy compound as the above-mentioned active species-capturing agent can be selected from

epoxidized soy bean oil, tris(epoxypropyl) isocyanurate, hydroquinonediglycidyl ether, diglycidyl terephthalate, 4,4'-sulfobisphenol-polyglycidyl ether, and N-glycidylphthalimide; and alicyclic epoxy compounds, such as hydrogenated bisphenol A glycidyl ether, 3,4-epoxycyclohexyl-methyl-3,4-epoxycyclohexane carboxylate, 2-(3,4-epoxycyclohexylspiro[5,5]-3,4-epoxy)-cyclohexane-m-dioxane, bis(3,4-epoxycyclohexylmethyl) adipate, vinylcyclohexenedioxide, 4-vinylepoxy-cyclohexane, bis(3,4-epoxy-6-methylcyclohexylmethyl) adipate, 3,4-epoxy-6-methylcyclohexyl-3,4-epoxy-6-methylcyclohexane carboxylate, methylenebis(3,4-epoxycyclohexane), dicyclopentadiene epoxide, di(3,4-epoxycyclohexylmethyl)ether of ethyleneglycol, ethylenebis(3,4-epoxycyclohexane carboxylate), dioctyl epoxyhexahydrophthalate, and di-2-ethylhexyl epoxyhexahydrophthalate.

The sunproofing agent as the light resistance improver is a component used for preventing light from penetrating into the interior of a polymer. Specific examples of sunproofing agents include titanium oxide having a rutile structure (TiO_2), zinc oxide (ZnO), chromium oxide (Cr_2O_3) and cerium oxide (CeO_2).

The metal inactivating agent as the light resistance improver is a component used for inactivating

heavy metal ions in the resin through the formation of a chelate compound. Specific examples of metal inactivating agents include an acid amine derivative, and benzotriazole and derivatives thereof.

5 The light quenching agent as the light resistance improver is a component used for inactivating photoexcited functional groups, such as hydroperoxides and carbonyl groups, formed in the polymer. The inactivation occurs due to the energy transfer from the functional groups. Organic nickel compounds and the like
10 are known as the light quenching agents.

 In the present invention, when a polymer blend of a plurality of different thermoplastic resins is used as the thermoplastic resin (A), it is preferred to use
15 as a compatibility agent a copolymer having a non-uniformity in distribution with respect to the proportions of component monomers constituting the copolymer so as to impart a further improved impact strength to the ultimate shaped resin article. Especially when a
20 polymer blend comprising a styrene polymer is used as the thermoplastic resin (A), the use of such a compatibility agent is effective.

 For example, when a polymer blend comprising an aromatic polycarbonate and a styrene polymer is used as
25 the thermoplastic resin (A), it is advantageous to use

as a compatibility agent a copolymer which has a non-uniformity in distribution with respect to the proportions of component monomers constituting the copolymer and which, therefore, is comprised of copolymer molecules having different solubility parameter (SP) values. In this case, it is preferred that the difference in the SP value (Δ SP value) between a copolymer molecule having the largest SP value and a copolymer molecule having the smallest SP value is from 0.3 to 1.0 [(cal/cm³)^{1/2}] and the average of the SP values of respective copolymer molecules is from 10.6 to 11.0, more preferably from 10.6 to 10.9, still more preferably from 10.7 to 10.8. With respect to the Δ SP value, for achieving high impact strength of the ultimate shaped resin article, it is preferred that the Δ SP value is in the above-mentioned range of from 0.3 to 1.0 [(cal/cm³)^{1/2}], more preferably in the range of from 0.3 to 0.8 [(cal/cm³)^{1/2}], still more preferably in the range of from 0.4 to 0.6 [(cal/cm³)^{1/2}].

As mentioned above, the resin composition of the present invention can be used as a masterbatch which is to be subjected to melt extrusion together with a resin material which is the same as or different from the thermoplastic resin (A) and, optionally, together with various solid additives. When the resin composition of

the present invention is used as a masterbatch as mentioned above, it becomes possible to melt-extrude a thermoplastic resin having a high viscosity, such as polyphenylene ether, at a relatively low temperature.

5 Examples of the methods for conducting the melt extrusion include a method in which all of the materials (including the masterbatch, the thermoplastic resin material other than the thermoplastic resin (A) and optionally the solid additive) are simultaneously fed
10 to an extruder and a method in which the resin materials {including the masterbatch and the thermoplastic resin other than the thermoplastic resin (A)} and the solid additive are charged to an extruder at different inlets thereof, wherein the inlet for the solid addi-
15 tive is positioned downstream of the inlet for the resin components, as viewed in the direction of the extrusion of the extruder, so that the solid additive is added to the molten resin. With respect to the extruder used for the melt extrusion, either a single-
20 screw extruder or a twin-screw extruder may be used. When the solid additive is used, for uniformly dispersing the solid additive in the resin materials, it is preferred to use a twin-screw extruder. With respect to the twin-screw extruder, it is preferred that the
25 L/D (ratio of the length L of each screw to the inner

diameter D of a cylinder portion of the extruder) value is from 20 to 50. Further, it is preferred that the twin-screw extruder has at least two inlets including a main feeding inlet and a side feeding inlet, and a kneading zone positioned at a region of the extruder extending from one end portion of the extruder to the inlet provided at a position adjacent to the other end portion of the extruder, wherein the kneading zone has a length corresponding to 1D to 10D.

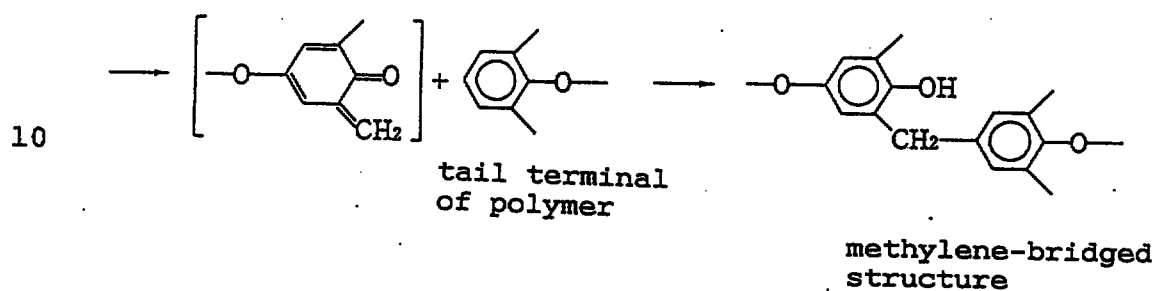
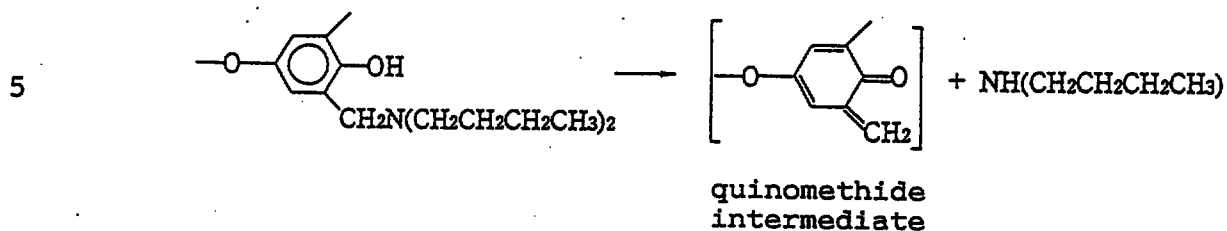
In another aspect of the present invention, there is provided a polyphenylene ether resin composition comprising (A') a polyphenylene ether resin, and (B') at least one additive for the polyphenylene ether resin (A'), wherein the polyphenylene ether resin (A') contains hydroxyl groups in an amount of not more than 0.5 mole, relative to 100 moles of the recurring oxyphenylene units of the polyphenylene ether resin. With respect to the additive (B'), there is no particular limitation, and any conventional additives for a polymer can be used. However, it is preferred to use the above-mentioned non-polymerizable liquid substance (B) as the additive (B').

In conventional melt-mixing methods for producing a composition of a polyphenylene ether resin (which has high heat resistance) and an additive, it is required

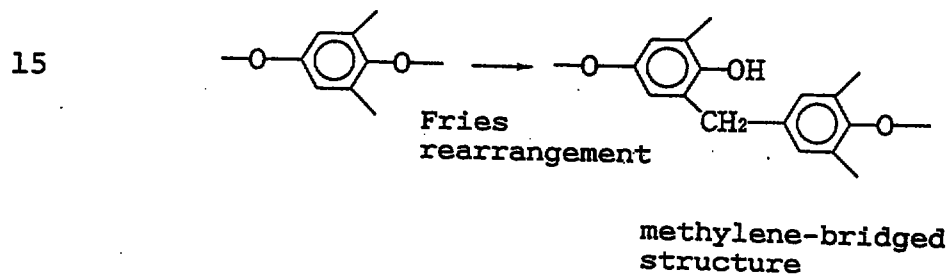
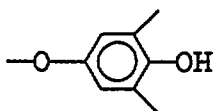
to conduct the melt-mixing at a temperature of from 300 to 350 °C. Therefore, in such conventional melt-mixing methods, the polyphenylene ether resin is caused to have a methylene-bridged structure which is formed, for example, by the reaction of a terminal group of the polymer other than the hydroxyl group as illustrated in formula (i) below {in formula (i), such a terminal group of the polymer is an aminoalkyl group} or by the Fries rearrangement as illustrated in formula (ii) below, to thereby increase the hydroxyl group content of the polyphenylene ether resin. Therefore, the polyphenylene ether resin composition produced by the conventional melt-mixing method has a problem in that, due to the high content of the hydroxyl group, a shaped resin article produced from such a polyphenylene ether resin composition suffers discoloration and has poor light stability.

Methylene-bridged structure

Reaction (i)



Reaction (ii)

20 Head terminal of polymer

The present inventors have found that the light stability and discoloration resistance of a polyphenylene ether resin can be remarkably improved by controlling the total hydroxyl group content of the polyphenylene ether resin {including not only the content of the hydroxyl groups formed due to the occurrence of the methylene-bridged structures but also the content of the hydroxyl groups which the polyphenylene ether inherently possesses (i.e., the hydroxyl group present in the head terminal of each polyphenylene ether chain)} to a level of not more than 0.5 mole, relative to 100 moles of the recurring oxyphenylene units.

The present inventors have also found that, for obtaining a resin composition comprising such an excellent polyphenylene ether resin (A') having remarkably improved light stability and discoloration resistance and an additive (B') for the polyphenylene ether resin, it is effective to use a method comprising the step of mixing together a polyphenylene ether resin (A) and a non-polymerizable liquid substance (B), wherein the mixing is conducted at a temperature in the range of from 25 to 300 °C, at which the non-polymerizable liquid substance (B) maintains a liquid state, thereby causing the polyphenylene ether resin (A) to be dissolved in or swelled with the non-polymerizable liquid

substance (B). Examples of additives as the additive (B') for the polyphenylene ether resin (A') are not limited to those which are mentioned above as the non-polymerizable liquid substance (B). As the flame retardant, which is a preferred additive, the above-mentioned specific organic phosphorus compounds are preferably used.

Still another preferred embodiment of the present invention is a shaped article produced by a method in which a masterbatch is produced by mixing together (A) from 20 to 80 parts by weight of a polyphenylene ether resin having a number average particle diameter of from 10 to 100 μm , (B) from 20 to 80 parts by weight of an aromatic phosphoric ester and (C) from 0.01 to 1 part by weight of silica, at a temperature in the range of from 150 to 250 $^{\circ}\text{C}$, thereby causing the polyphenylene ether resin to be dissolved in or swelled with the aromatic phosphoric ester, and the obtained masterbatch is melt-extruded together with a styrene polymer resin and a solid substance (such as a light resistance improver, a mold release agent or the like) to thereby obtain a molding material, and the molding material is injection-molded to obtain a shaped article having a desired shape, wherein the shaped article, such as a pellet, may further be used as a molding material.

The thus obtained shaped article has an excellent balance of various properties, such as flame retardancy, impact resistance, heat resistance and the like.

It has further been found that a more excellent flame retardancy can be achieved when the total content of the residual aromatic vinyl monomer, dimer and trimer of the resin composition is not more than 1 % by weight. When the above-mentioned total content of residual aromatic vinyl monomer, dimer and trimer is more than 1 % by weight, the flame retardancy becomes poor, because these vinyl compounds vaporize and act as flammable gas when the resin composition is burned.

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BEST MODE FOR CARRYING OUT THE INVENTION

Hereinbelow, the present invention will be described in more detail with reference to the following Examples and Comparative Examples, but they should not be construed as limiting the scope of the present invention.

In the Examples and Comparative Examples, measurements were done by employing the following methods and apparatus.

(1) Analysis of a resin composition containing a phosphoric ester flame retardant:

5 g of a resin composition is dissolved in 100 ml of methyl ethyl ketone and the resultant solution is subjected to ultracentrifugation (20,000 rpm, 1 hr) to obtain a supernatant. Then, methanol is added to the supernatant in an amount which is 2 times that of the supernatant, to thereby precipitate the resin component. The resultant mixture containing the precipitated resin component is subjected to ultracentrifugation to separate the resin component from the mixture. The resultant solution is analyzed by gel permeation chromatography (GPC) using HLC-8020 (manufactured and sold by Tosoh Corp., Japan) equipped with a differential refractometer (columns: G100HXL x 2, manufactured and sold by Tosoh Corp., Japan; mobile phase: tetrahydro-

furan; flow rate: 0.8 ml/min. ; pressure: 60 kgf/cm²;
inlet temperature: 35 °C; oven temperature: 40 °C;
temperature at differential refractometer: 35 °C;
internal volume of sample loop: 100 ml; concentration
5 of sample: 0.08 g/20 ml), and the composition and
amount of the phosphoric ester contained in the solu-
tion are determined from the proportions of the respec-
tive areas (which are determined from the chromatogram)
for the components of the solution. On the other hand,
10 the above-mentioned resin component is dissolved in
deuteriochloroform to obtain a sample solution. With
respect to the obtained sample solution, the propor-
tions of the integrated values of aromatic protons and
aliphatic protons and the chemical shifts are deter-
15 mined using a Fourier transform nuclear magnetic reso-
nance (proton-FT-NMR) apparatus (DPX-400, manufactured
and sold by Bruker Analytik GMBH, Germany), and the
composition and content of the resin component (thermo-
plastic resin component, such as a rubber-modified
20 styrene resin, a polyphenylene ether resin and/or a
polycarbonate in the resin component) are determined.

(2) Analysis of hydroxyl groups contained in a
polyphenylene ether resin:

5 g of a resin composition is dissolved in 100 ml
25 of methyl ethyl ketone and the resultant solution is

subjected to ultracentrifugation (20,000 rpm, 1 hr), followed by evaporation to obtain a residue. The obtained residue is dissolved in methylene chloride to obtain a methylene chloride solution having a solids content of 5 % by weight. The methylene chloride solution is allowed to stand in a refrigerator having a temperature of -15 °C for 24 hours to thereby obtain a precipitate. Then, the precipitate is obtained by filtration under cooled temperature conditions. The precipitate is washed with methylene chloride, followed by drying in vacuo, to thereby obtain a polyphenylene ether. The obtained polyphenylene ether is dissolved in deuteriochloroform to obtain a sample solution, which is then subjected to the following analysis.

Using a Fourier transform nuclear magnetic resonance (proton-FT-NMR) apparatus, the amount of hydroxyl groups present in the methylene bridged structures of the polyphenylene ether resin and the amount of hydroxyl groups present in the head terminals of the polymer chains of the polyphenylene ether resin are determined. In the NMR spectra shown in Figs. 3(a), 3(b), 4(a) and 4(b), peaks respectively ascribed to the hydrogen atoms bonded to the aromatic rings, the hydrogen atoms in the methylene group and the hydrogen atoms in the hydroxyl group, which are present in the methylene bridge struc-

tures, are indicated as A or A', D or D' and B or B', respectively, and a peak ascribed to hydrogen atoms in the hydroxyl group present in the head terminals of the polymer chain is indicated as C or C'. Generally, the
 5 peak ascribed to the hydrogen atoms present in a hydroxyl group appears in the range of from 4.2 to 4.4 ppm.

(3) SP value (δ) (solubility parameter) and an average SP value:

10 The SP value is calculated in accordance with the following Fedors Formula, which is described in Polymer Engineering and Science, 14, (2), 147 (1974), using the data of Δe_1 and Δv_1 of functional groups of polymers, which data are also described therein:

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$$\delta = \sqrt{\{\Sigma(\Delta e_1)/\Sigma(\Delta v_1)\}}$$

wherein Δe_1 represents the cohesive energy per unit functional group, and Δv_1 represents the molecular volume per unit functional group, and the unit for δ is $(\text{cal}/\text{cm}^3)^{1/2}$.

20 The average SP value of a copolymer or a blend of copolymers is calculated from the SP values of the different constituent monomer units of the copolymer or the SP values of the different constituent copolymers of the copolymer blend by the proportional allocation
 25 of the respective SP values of the different constitu-

ent monomer units or the different constituent copolymers in accordance with the respective weight ratios of the constituent monomer units or the constituent copolymers, on the assumption that the additivity rule can be applied. For example, the average SP value of an acrylonitrile-styrene copolymer is calculated from the SP value of polyacrylonitrile (14.39) and the SP value of polystyrene (10.52) by the proportional allocation of the respective SP values of the polyacrylonitrile and the polystyrene in accordance with the respective weight ratios of the acrylonitrile and styrene in the copolymer.

(4) Glass transition temperature (T_g) and melting temperature (T_m):

The glass transition temperature (T_g) and the melting temperature (T_m) are measured by the differential scanning calorimetry (DSC) method described in "Polymer Handbook" (edited by J. Brandrup, A Wiley-Interscience Publication, John Wiley & Sons, New York, U.S.S., 1975). In accordance with the differential scanning calorimetry (DSC) method, the glass transition temperature (T_g) is determined by observing the change in the specific heat capacity, and the melting temperature (T_m) is determined as a temperature corresponding to the position of a peak of a melting curve. Specifi-

cally, the measurements are conducted as follows. The measurements are conducted using a thermal analysis apparatus (Model DT-40, manufactured and sold by Shimadzu Corporation, Japan), in which 5 mg of a sample is heated at a rate of 10 °C/ min. under the flow of nitrogen gas. As shown in Fig. 9(a), T_g is defined as a temperature corresponding to the intersection of the base line with a straight line drawn from the first step of the line of the specific heat capacity. As shown in Fig. 9(b), T_m is defined as a temperature corresponding to the intersection of the base line with a straight line drawn from the rising line of the heat absorption peak.

The determination as to whether a polymer is a crystalline polymer or an amorphous polymer is done by observing whether or not a peak of the heat absorption is observed. The area of the peak of the heat absorption corresponds to the heat of crystallization (Cal/g), and is used as a criterion of the crystallinity of the polymer.

The "Polymer Handbook" describes that the glass transition temperatures (T_g 's) of poly(2,6-dimethyl-1,4-phenylene oxide), polycarbonate of Bisphenol A, polystyrene and poly(vinyl chloride) are 209 °C, 145 °C, 100 °C and 81 °C, respectively.

(5) Reduced viscosity η_{sp}/c of a rubber-modified styrene resin and a polyphenylene ether resin:

The viscosity η_{sp}/c of a rubber-modified styrene resin is measured as follows. To 1 g of a rubber-modified styrene resin is added a mixed solvent of 18 ml of methyl ethyl ketone and 2 ml of methanol, followed by shaking at 25 °C for 2 hours. The resultant mixture is centrifuged at 18,000 rpm at 5 °C for 30 minutes to obtain a supernatant. The obtained supernatant is taken out, and the resin component is deposited from the supernatant with methanol, followed by drying.

0.1 g of the thus obtained resin is dissolved in a solvent (when a rubber-modified styrene resin is a rubber-modified polystyrene, toluene is used as a solvent; and when a rubber-modified styrene resin is a rubber-modified acrylonitrile/styrene copolymer, methyl ethyl ketone is used as a solvent), so as to obtain a solution having a resin concentration of 0.5 g/dl. 10 ml of the obtained solution is put in a Cannon-Fenske viscometer, and the falling time T_1 (sec) of the solution is measured at 30 °C. Further, the respective falling times T_0 's (sec) of pure toluene and pure methyl ethyl ketone are measured using the same viscometer as mentioned above. The reduced viscosity of the

rubber-modified styrene resin is calculated by the following formula:

$$\eta_{sp}/c = (T_1/T_0 - 1)/C$$

wherein C represents the concentration (g/dl) of the rubber-modified styrene resin in the solution.

With respect to a polyphenylene ether resin, the reduced viscosity η_{sp}/c thereof is measured as follows. 0.1 g of the polyphenylene ether resin is dissolved in chloroform, to thereby obtain a solution having a polyphenylene ether resin concentration of 0.5 g/dl, and the determination of the reduced viscosity is conducted in substantially the same manner as mentioned above.

(6) Color (yellowness index):

The color of a resin composition is measured, using an SM color computer Model SM-5 (manufactured and sold by Suga Test Instruments Co. Ltd., Japan), in accordance with JIS-Z-8722.

(7) Izod impact strength:

The Izod impact strength of a 1/4 inch-thick (unless otherwise specified), V-notched specimen is measured at 23 °C in accordance with ASTM-D256.

(8) Volatility (thermogravimetric analysis; TGA method):

The volatility is evaluated using a thermogravimetric analyzer DT-40 (manufactured and sold by Shimadzu Corporation, Japan) by elevating the heating temperature at a rate of 40 °C/min under the flow of nitrogen gas. The temperature at which a 1 % weight loss occurred is used as a criterion of the volatility.

(9) Vicat softening temperature:

The Vicat softening temperature is measured substantially in accordance with ASTM-D1525, and used as a criterion of the heat resistance.

(10) Melt flow rate (MFR):

The melt flow rate is measured substantially in accordance with ISO-R1133, and used as a criterion of melt fluidity. That is, the melt flow rate is determined from the extrusion rate (g/10 min) of a resin composition measured at a melting temperature of 200 °C under a load of 5 kg.

(11) Flame retardancy:

The flame retardancy of a 1/8 or 1/16 inch-thick specimen is evaluated in accordance with the Vertical Burning Method which is described in UL-Subject 94. With respect to the method described in UL-Subject 94, reference can be made to, for example, U.S. Patent No. 4,966,814.

(12) Light resistance of a shaped article:

The light resistance is evaluated using ATLAS CI35W Weatherometer (manufactured and sold by ATLAS Electric Devices Co., U.S.A.) as a light resistance testing device, in accordance with JIS K7102. Exposure conditions are as follows: an internal temperature of testing device of 55 °C, a humidity of 55 %, no rain, and exposure to a xenon light (wavelength: 340 nm, energy: 0.30 W/m²) for 300 hours. The color difference ΔE between a pre-exposed shaped article and a post-exposed shaped article is measured, using an SM color computer model SM-3 (manufactured and sold by Suga Test Instruments Co., Ltd, Japan) in accordance with the L.a.b. method, to evaluate the color tone difference. The smaller the color tone difference, the higher the light resistance.

(13) Evaluation of the compatibility between a thermoplastic resin (A) and a non-polymerizable liquid substance (B):

50 parts by weight of a powdery thermoplastic resin (A) having an average particle diameter of from 15 μ m to 1 mm and 50 parts by weight of a non-polymerizable liquid substance (B) are mixed while stirring at 200 °C for 1 hour, to thereby obtain a thermoplastic resin composition. Subsequently, the obtained resin composition is filtered through a glass filter having

an average pore diameter of 10 μm , while maintaining a temperature of the resin composition at 200 °C under a reduced pressure of around 50 mmHg, and the weight of a component (A)/component (B) mixture remaining on the filter is measured.

The weight ratio of a component (A)/component (B) mixture remaining on the filter to the thermoplastic resin (A) used in the resin composition is used as a criterion of the compatibility between a thermoplastic resin (A) and a non-polymerizable liquid substance (B). The lower the filtration-remaining ratio, the higher the compatibility between a thermoplastic resin (A) and a non-polymerizable liquid substance (B).

The number average particle diameter of the powdery thermoplastic resin (A) is determined by using an optical microscope or an electron microscope. The average particle diameter of the powdery thermoplastic resin is obtained as the arithmetical mean value of the diameters of 100 particles, wherein the arithmetic mean value of the long axis and short axis of the particle is taken as the diameter of each particle, irrespective of the shape of the particles (such as a spherical shape or a pellet shape).

(14) Evaluation of the stability of production (by melt extrusion) of a resin composition:

Production of a resin composition is conducted by using a twin-screw extruder having a main feeder and a side feeder (ZSK-40; nominal maximum extrusion rate: 100 kg/hr; L/D = 46) (manufactured and sold by Werner Pfleiderer GmbH, Germany). Specifically, a thermoplastic resin (A) is fed from the main feeder, followed by melt kneading at the melting temperature of the thermoplastic resin (A) {e.g., at about 310 °C when a polyphenylene ether resin (PPE) is used}. Then, a non-polymerizable liquid substance (B) is fed from the side feeder into the melt-kneaded PPE, followed by melt extrusion at a temperature lower than the melting temperature of the thermoplastic resin (A) (e.g., at about 230 °C when a PPE is used) at an extrusion rate of 30 kg/hr (process II-1).

The evaluation of the stability of production (by melt extrusion) is made according to the following criteria.

Production is impossible: the non-polymerizable liquid substance (B) spouts out of a die, so that the production of a shaped article is impossible.

Unstable extrusion: the production of a shaped article is possible; however, the fluctuation of extrusion rate during the production is ± 20 % or more, relative to the predetermined rate, and the wetting of

the surface of the produced shaped article with the non-polymerizable liquid substance (B) is observed.

5 Slightly unstable extrusion: the fluctuation of extrusion rate during the production is from $\pm 5\%$ to less than $\pm 20\%$, and the wetting of the surface of the produced shaped article with a non-polymerizable liquid substance (B) is not observed.

Good: the fluctuation of extrusion rate during the production is less than $\pm 5\%$.

10

The materials used in the Examples and Comparative Examples are as follows.

(a) Thermoplastic resin (A)

15 (1) Production of a polyphenylene ether (PPE)

The production of a PPE was conducted using a stainless steel reactor provided with an inlet for feeding oxygen at a bottom thereof, a cooling coil and agitating blades. The reactor was purged with nitrogen gas. Then, 54.8 g of cupric bromide, 1,110 g of di-n-butylamine, and a solution which had been obtained by dissolving 8.75 kg of 2,6-xylenol in a mixed solvent of 20 liters of toluene, 16 liters of n-butanol and 4 liters of methanol were added to the reactor. A polymerization reaction was performed for 90 minutes while

25

stirring and continuously feeding oxygen into the reactor having an internal temperature controlled to 30 °C. After completion of the reaction, a deposited polymer was collected by filtration. To the collected polymer was added a mixture of methanol and hydrochloric acid to decompose the catalyst remaining in the polymer. Further, the polymer was washed well with methanol to obtain a powdery polyphenylene ether (hereinafter, frequently referred to as "PPE-1"). The obtained PPE-1 had a reduced viscosity η_{sp}/c of 0.41 dl/g.

Polyphenylene ethers having different reduced viscosities (η_{SP}/C) were produced in substantially the same manner as mentioned above, except that the amount of the catalyst and the polymerization time were changed. Results are shown in Table 12. Each of the obtained polyphenylene ethers had a number average particle diameter of 20 μm , and an SP value of 11.2.

(2) non-rubber-modified polystyrene (general purpose polystyrene) (GPPS):

A commercially available polystyrene (weight average molecular weight : 200,000, manufactured and sold by Asahi Chemical Industry Co., Ltd., Japan) (hereinafter, frequently referred to as "GPPS") was used. The SP value of GPPS was 10.5.

(3) Rubber-modified styrene polymer (HIPS)

Polybutadiene (a weight ratio of cis-1,4 bond/trans-1,4 bond/vinyl-1,2 bond = 95/2/3) (trade name: Nipol 122 OSL, manufactured and sold by Nippon Zeon Co., Ltd., Japan) was dissolved so as to form a homogeneous solution having the following composition.

	Polybutadiene	10.5 % by weight
	Styrene	74.2 % by weight
	Ethylbenzene	15.0 % by weight
10	α -methylstyrene dimer	0.27 % by weight
	t-butylperoxyisopropyl carbonate	0.03 % by weight

The thus obtained solution was continuously fed to a reactor having four different reaction regions connected in series and each equipped with an agitator. Polymerization was performed at 126 °C and 190 rpm in the first region; at 133 °C and 50 rpm in the second region; at 140 °C and 20 rpm in the third region; and at 155 °C and 20 rpm in the fourth region. The resultant polymerization reaction mixture (solids content: 73 %) was transferred to a degassing apparatus to remove unreacted monomers and the solvent. Thus, a rubber-modified styrene polymer (hereinafter, referred to as "HIPS-1") was obtained. The obtained rubber-modified styrene polymer was subjected to analysis. As a re-

sult, it was found that the rubber content was 12.1 % by weight, the weight average particle diameter of the rubber particles was 1.5 μm , the reduced viscosity $\eta_{\text{sp}}/\text{c}$ was 0.53 dl/g, and the SP value was 10.3.

5 Rubber-modified styrene polymers having different viscosities ($\eta_{\text{sp}}/\text{c}$) were produced in substantially the same manner as mentioned above, except that the amount of the polymerization initiator, the polymerization temperature and the amount of the chain transfer agent
10 were changed. Results are shown in Table 11.

HIPS used in the Examples and Comparative Examples are as follows (see Table 11 also).

HIPS-1: Rubber component: polybutadiene rubber; rubber
15 content of HIPS-1: 12.1 % by weight; weight average particle diameter of the polybutadiene rubber: 1.5 μm ; and reduced viscosity $\eta_{\text{sp}}/\text{c}$: 0.53 dl/g.

HIPS-2: Rubber component: polybutadiene rubber; rubber
content of HIPS-2: 12.1 % by weight; weight average
20 particle diameter of the polybutadiene rubber: 1.5 μm ; and reduced viscosity $\eta_{\text{sp}}/\text{c}$: 0.79 dl/g.

HIPS-3: Rubber component: polybutadiene rubber; rubber
content of HIPS-3: 12.1 % by weight; weight average
particle diameter of the polybutadiene rubber: 1.5 μm ;
25 and reduced viscosity $\eta_{\text{sp}}/\text{c}$: 0.60 dl/g.

HIPS-4: Rubber component: polybutadiene rubber; rubber content of HIPS-4: 12.1 % by weight; weight average particle diameter of the polybutadiene rubber: 1.5 μm ; and the reduced viscosity $\eta_{\text{sp}}/\text{c}$ of HIPS-4: 0.58 dl/g.

5 HIPS-5: Rubber component: polybutadiene rubber; rubber content of HIPS-5: 12.1 % by weight; weight average particle diameter of the polybutadiene rubber: 1.5 μm ; and reduced viscosity $\eta_{\text{sp}}/\text{c}$: 0.40 dl/g.

10 HIPS-6: Rubber content of HIPS-6: 12.1 % by weight; weight average particle diameter of the polybutadiene rubber: 1.5 μm ; and reduced viscosity $\eta_{\text{sp}}/\text{c}$: 0.35 dl/g.

(4) Aromatic polycarbonate (PC)

15 A commercially available bisphenol A type polycarbonate [Novarex 7025A, manufactured and sold by Mitsubishi Chemical Industries, Ltd., Japan (hereinafter, frequently referred to as "PC")] was used. The SP value of PC was 11.3.

(5) ABS resin (ABS)

20 A commercially available ABS resin [acrylonitrile/butadiene/styrene = 26/14/60 (weight ratio)] (hereinafter, frequently referred to as "ABS") was used.

25 (6) Compatibility agent: acrylonitrile-styrene copolymer (AS)

A mixture of 3.4 parts by weight of acrylonitrile, 81.6 parts by weight of styrene, 15 parts by weight of ethylbenzene and, as a polymerization initiator, 0.03 part by weight of 1,1-bis(t-butylperoxy)-3,3,5-trimethylcyclohexane was continuously fed to a plug flow type reactor having three regions connected in series, and each equipped with an agitator at a rate of 0.7 liter/hr. Polymerization was performed at 126 °C and 100 rpm in the first region; at 135 °C and 20 rpm in the second region; and at 147 °C and 10 rpm in the third region. Subsequently, the resultant polymerization reaction mixture was transferred to a degassing apparatus having a temperature of 230 °C, to remove unreacted monomers and the solvent. Thus, a random copolymer (hereinafter, referred to as "AS") was obtained. The obtained copolymer was subjected to analysis. As a result, it was found that the copolymer contained 6 % by weight of the acrylonitrile monomer unit and 94 % by weight of the styrene monomer unit (as measured by infrared adsorption spectrophotometry), and had an average SP value of 10.75. Further, the distribution of the proportions of the component monomers of the copolymer was measured by liquid chromatography (according to a method described in International Patent Application Publication No. WO-95-35346). As a

result, it was found that the proportion of the acrylonitrile monomer units was varied in the range of from 0 to 12 % by weight in the copolymer, and the maximum SP value was 11.0, the minimum SP value was 10.5, and the Δ SP value was 0.5.

(7) Thermoplastic elastomer:

A commercially available styrene-butadiene block copolymer {styrene block/butadiene block = 40/60 (weight ratio), trade name: Tufprene 125, manufactured and sold by Asahi Chemical Industry Co., Ltd., Japan} (hereinafter, frequently referred to as "TPE") was used.

(8) Polyamide

A commercially available polyamide 6 (trade name: UBE Nylon 1013B, manufactured and sold by Ube Industries, Ltd., Japan) (hereinafter, frequently referred to as "PA6") was used.

(b) Non-polymerizable liquid substance (B)

(1) Production of tris(nonylphenyl) phosphate (TNPP)

431.0 parts by weight (molar ratio: 3.0) of nonylphenol and 0.87 part by weight (molar ratio: 0.01) of aluminum chloride were placed in a flask. Then, 100 parts by weight (molar ratio: 1.0) of phosphorus oxych-

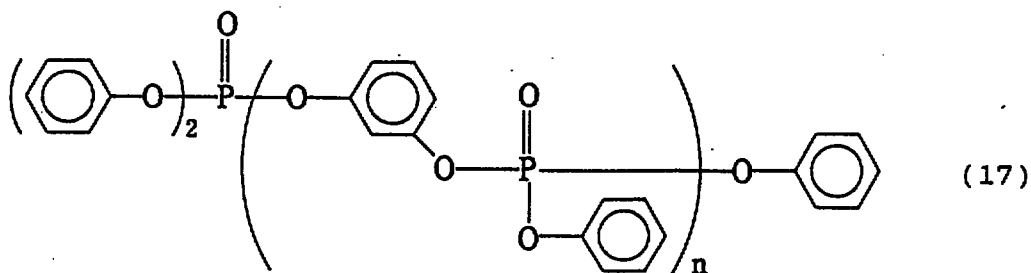
loride were dropwise added to the flask at 90 °C over 1 hour. The temperature in the flask was gradually elevated to 180 °C to thereby complete the esterification reaction. Then, the resultant reaction product was cooled and washed with water to remove the catalyst and chlorine values to obtain a mixture of tris(nonylphenyl)phosphate (hereinafter, frequently referred to as "TNPP").

The average value of the sum of the respective numbers of carbon atoms of the substituent groups was 27.0.

(2) 1,3-phenylene-bis(diphenylphosphate) (FR-1):

A commercially available oligomeric aromatic phosphoric ester derived from resorcinol (manufactured and sold under tradename of "CR733S" by Daihachi Chemical Industry Co., Ltd., Japan) (hereinafter, frequently referred to as "FR-1") was used.

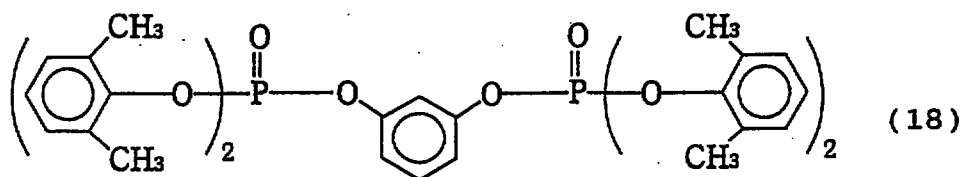
According to the analysis by GPC, it was found that FR-1 is composed of TPP dimer ($n=1$) and TPP oligomers ($n \geq 2$) in a weight ratio of 65/35, which are represented by the following formula (17).



(3) 1,3-phenylene-bis{di(2,6-dimethylphenyl)phosphate} (FR-2)

244 Parts by weight of 2,6-xylenol, 20 parts by weight of xylene and 1.5 parts by weight of magnesium chloride were placed in a reactor and mixed while heating to thereby perform a reaction. The temperature of the reaction mixture was gradually elevated, and after the temperature of the reaction mixture reached 120 °C, 153 parts by weight of phosphorus oxychloride were dropwise added to the reaction mixture over 2 hours. The hydrochloride gas being formed during the reaction was led into a water scrubber. After completion of the addition of phosphorus oxychloride, the temperature of the reaction mixture was gradually elevated to 180 °C over 2 hours to complete the reaction. The yield of the resultant intermediate {di(2,6-xylyl)phosphochloride} was 99.7 %. Then, 45 parts by weight of the intermediate, 55 parts by weight of resorcinol and 1.5 parts by weight of aluminum chloride were placed in a reactor and mixed together while heating. The temperature of the resultant reaction mixture was gradually elevated to 180 °C over 2 hours, to thereby perform a dehydrochlorination reaction. Then, the resultant reaction mixture was aged at 180 °C

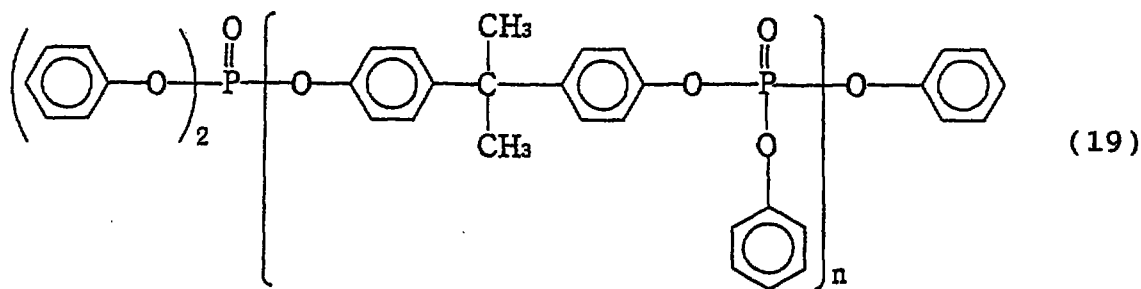
for 2 hours and then, further aged at 180 °C under a reduced pressure of 200 mmHg, to thereby complete the dehydrochlorination reaction. To the resultant reaction mixture were added 500 parts by weight of xylene and 200 parts by weight of a 10 % aqueous hydrochloride solution to remove the remaining catalyst and then, the resultant was repeatedly washed with water, to thereby obtain a purified reaction mixture. The obtained purified reaction mixture was cooled while stirring to effect a crystallization. Then, crystals were obtained by filtration and washed with methanol, followed by drying at 100 °C under reduced pressure to thereby obtain 1,3-phenylene-bis-(di-2,6-dimethyl-phenylphosphate) (hereinafter, frequently referred to as "FR-2") represented by the following formula (18).



(4) Bisphenol A-bis(diphenylphosphate) (FR-3)

A commercially available oligomeric aromatic phosphoric ester produced from bisphenol A (trade name:

CR 741, manufactured and sold by Daihachi Chemical Industry Co., Ltd., Japan) (hereinafter, frequently referred to as "FR-3") was used. According to the analysis by GPC, it was found that FR-3 is composed of TPP-A-dimer, TPP-A-oligomers and trisphenylphosphate (TPP) in a weight ratio of 84.7/13.0/2.3, wherein TPP-A-dimer ($n=1$) and TPP-A-oligomer ($n \geq 2$) have a structure represented by the following formula (19):



(5) Triphenyl phosphate (TPP):

A commercially available monomeric aromatic phosphoric ester (hereinafter, referred to as "TPP") (trade name: TPP, manufactured and sold by Daihachi Chemical Industry Co., Ltd., Japan) was used.

(6) Antistatic agent

A commercially available amine-containing antistatic agent N,N-bis-(2-hydroxyethyl)tallow amine

(hereinafter referred to as "A-stat") (trade name: Armostat 310, manufactured and sold by Armak Company, U.S.A.) was used.

(7) Lubricant

5 A commercially available lubricant, SAE#30 engine oil (hereinafter, referred to as "Lb") was used.

(8) Bromine-containing flame retardant

A commercially available brominated epoxy resin (hereinafter, referred to as "Br-EP") (trade name: Pratherm EC-14, manufactured and sold by Dainippon Ink & Chemicals, Inc., Japan) was used.

(9) Stabilizer

A commercially available phosphorus-containing stabilizer {tris(nonylphenyl) phosphite (hereinafter referred to as "TNP") (trade name: TNP, manufactured and sold by Asahi Denka Kogyo K.K., Japan)} was used.

(10) Mineral oil

A commercially available mineral oil (hereinafter referred to as "MO") (trade name: Kaydol, manufactured and sold by Witco Corp., U.S.A.) was used.

(11) Flame retardant containing bromine and phosphorus

A commercially available tris(2-chloro-3-bromopropyl) phosphate (hereinafter, referred to as "PB") (manufactured and sold by Daihachi Chemical Industry

Co., Ltd., Japan) was used.

(c) Solid substance (C): Anti-block agent

(1) Silicon compound

5 A commercially available silica (hereinafter, referred to as "SI") (trade name: Nipsil VN3LP, manufactured and sold by Nippon Silica Industrial Co., Japan) was used.

(2) Oil-absorptive material

10 A commercially available porous styrene-divinylbenzene copolymer (hereinafter, referred to as "SDV") (trade name: Diaion MP-A, manufactured and sold by Mitsubishi Chemical Corporation, Japan) was used. SVD has a number average particle diameter of 10 μm and a
15 number average pore diameter of 0.01 μm .

(d) Solvent (D)

 Commercially available xylene, methylene chloride, methanol and formic acid (hereinafter, referred to as
20 "XY, MCl, MeOH, FA", respectively) were used.

(e) Other solid substances

(1) Phenol novolak resin (softening temperature:
120 °C)

25 A commercially available novolak resin (hereinaft-

er, referred to as "NK") (trade name: SP1006N, manufactured and sold by Asahi Organic Chemicals Industry Co., Ltd., Japan) was used.

(2) Ultraviolet light absorber

5 A commercially available Benzotriazole-containing ultraviolet light absorber (hereinafter, referred to as "UVA") (trade name: Tinuvin P, manufactured and sold by CIBA-GEIGY, Switzerland) was used.

10 (3) Hindered piperidine-containing light stabilizer

A commercially available hindered piperidine-containing light stabilizer (hereinafter, referred to as "HALS") (trade name: Tinuvin 770, manufactured and sold by CIBA-GEIGY, Switzerland) was used.

15 (4) White pigment (Sunproofing agent)

A commercially available powdery titanium oxide having an average particle diameter of 0.2 μm (hereinafter, referred to as " TiO_2 ") (trade name: TIPAQUE PF-691, manufactured and sold by Ishihara Sangyo Kaisya, Ltd., Japan) was used.

20 (5) Organic epoxy compound

A commercially available epoxidized soybean oil (hereinafter, referred to as "EP") (trade name: O-130P, manufactured and sold by Asahi Denka Kogyo K.K., Japan) was used.

25

(6) Fluororesin

A commercially available polytetrafluoroethylene (hereinafter, referred to as "PTFE") (trade name: Teflon 6J, manufactured and sold by Du Pont-Mitsui Fluorochemicals Co., Ltd., Japan) was used.

Example 1 and Comparative Examples 1 to 4

Each of five types of resin compositions shown in Table 1 was individually produced by a process selected from four processes as described below, and evaluated with respect to various properties, such as color (yellowness index: YI). Results are shown in Table 1.

Process I

Using a batch type mixing vessel equipped with an agitator, the thermoplastic resin (A) was dissolved into the non-polymerizable liquid substance (B) at a temperature indicated in Table 1 while stirring. The resultant was introduced to a belt-conveyer type drying cooler (belt cooler), to thereby obtain pellets.

Process II-1

A twin-screw extruder having a main feeder and a side feeder was used. The thermoplastic resin (A) was fed from the main feeder and the non-polymerizable liquid substance (B) was fed from the side feeder, followed by melt-extrusion at a temperature indicated

in Table 1, to thereby obtain pellets.

Process II-2

5 A twin-screw extruder having a main feeder and a side feeder was used. Both the thermoplastic resin (A) and the non-polymerizable liquid substance (B) were simultaneously fed from the main feeder, followed by melt-extrusion at a temperature indicated in Table 1, to thereby obtain pellets.

Process III

10 The non-polymerizable liquid substance (B) was portionwise added to the thermoplastic resin (A) so as to be adsorbed on the thermoplastic resin (A), to thereby obtain an adsorption mixture of the thermoplastic resin (A) and the non-polymerizable liquid substance (B). The mixture was introduced to a belt-conveyer type drying cooler (belt cooler), to thereby obtain pellets.

20 From Table 1, it can be seen that, with respect to the resin composition produced by process I (present invention), in which PPE-1 as the thermoplastic resin (A) was dissolved at a temperature lower than 300 °C, so that the formation of OH groups in PPE unit was suppressed (see Figs. 3 and 4), the resin composition not only had superior appearance, color and light stability to those of the other resin compositions

25

produced by melt extrusion, but also could be stably produced. On the other hand, in each of processes II-1 and II-2, the extrusion of the resin composition became unstable when the content of the non-polymerizable liquid substance (B) in the resin composition was at the level of 60% by weight. Further, the resin composition produced by process II-1 (which was conducted under conditions wherein PPE-1 experienced melting) does not contain any discrete segments of resin that have not experienced melting and, accordingly, has a good appearance; however, it is poor with respect to color and light stability (see Comparative Example 3). It is observed that the resin composition produced by process II-2, in which both the thermoplastic resin (A) and the non-polymerizable liquid substance (B) were simultaneously fed from the main feeder followed by melt-extrusion, contains many discrete segments of PPE-1 that have not experienced melting and, therefore, has a poor appearance. Especially in the case of process III, the resin composition produced by process III, in which the non-polymerizable liquid substance (B) was adsorbed on the thermoplastic resin (A), contains a large number of discrete segments of PPE-1 that have not experienced melting and, therefore, has a very poor appearance.

Examples 2 to 17 and Comparative Examples 5 to 28

Resin compositions were prepared using the materials and methods (Process I or II) as described in Tables 2 to 4, wherein PPE-1, PC, GPPS and HIPS-1 were individually used as the thermoplastic resin (A). The obtained resin compositions were evaluated with respect to various properties, such as color (yellowness index: YI). Results are shown in Tables 2 to 4 and Fig. 5.

From Tables 2 to 4 and Fig. 5, it is apparent that the resin composition obtained by the method of the present invention, in which the production of the resin composition is conducted by Process I (dissolution) at a temperature of 300 °C or less, is excellent with respect to appearance and color as compared to the resin composition obtained by a conventional melt-extrusion method.

Examples 18 to 34 and Comparative Examples 29 to 32, and 52 through 55

Resin compositions were prepared using the materials and methods (Process I or II) as described in Tables 5 to 7, wherein PPE-1 and GPPS were individually used as the thermoplastic resin (A). The obtained resin compositions were evaluated with respect to various properties, such as color (yellowness index:

YI). Results are shown in Tables 5 to 7 and Figs. 6 and 7.

From Tables 5 to 7 and Figs. 6 and 7, it can be observed:

5 that when the production of the resin article is conducted by Process I (dissolution) using a surface modifier, such as an antistatic agent or a lubricant, instead of the non-polymerizable liquid substance (B), since such a surface modifier has poor compatibility
10 with the thermoplastic resin (A), it is necessary to conduct the dissolution of the thermoplastic resin (A) into the surface modifier at high temperature, resulting in the acceleration of deterioration of the thermoplastic resin (A) and the surface modifier (Comparative
15 Examples 29 to 32);

 that the compatibility of an oligomeric aromatic phosphoric ester with PPE-1 (among flame retardants which generally have good compatibility with the thermoplastic resins, the oligomeric aromatic phosphoric
20 ester has slightly inferior compatibility with the thermoplastic resins, such as PPE-1) can be remarkably improved by the use of a monomeric aromatic phosphoric ester or GPPS in combination with the oligomeric aromatic phosphoric ester; and

25 that with respect to Process II-1 (melt

extrusion), the extrusion can be stably conducted when the amount of the non-polymerizable liquid substance (B) is not more than 20 % by weight, based on the total amount of (A) and (B); however, the extrusion becomes
5 unstable when the amount of the non-polymerizable liquid substance (B) exceeds 20 % by weight.

Examples 35 to 50 and Comparative Examples 33 to 34

Resin compositions were prepared using the materials and method (Process I) as described in Tables 8 and
10 9. The obtained resin compositions were evaluated with respect to various properties, such as color (yellowness index: YI). Results are shown in Tables 8 and 9.

From Tables 8 and 9, it can be observed:

15 that when the PPE-1/non-polymerizable liquid substance (B) (flame retardant) weight ratio is in the range of from 30/70 to 70/30, it is possible to obtain a shaped resin article which is excellent with respect to the ease in handling and properties;

20 that when the solid substance (C), such as an oil-absorptive material or an anti-block agent, is added to the component (A)/component (B) mixture having a weight ratio within the above-mentioned range in an amount such that the mixture remains in a liquid state, the
25 resultant resin composition is free of blocking; and

that when the component (A)/component (B) mixture (which is in a liquid state) having the solid substance (C) added thereto is non-uniform or has poor fluidity, the appearance of the produced resin composition becomes extremely poor. (Comparative Example 34)

Examples 51 to 53 and 108, and Comparative Examples 35 to 38, 50 and 51

Resin compositions to be used as masterbatches were prepared using the materials and methods as described in Table 10, wherein the dissolution of PPE-1 into the non-polymerizable liquid substance (B) was conducted in the presence of the solvent (D) which functions to improve the compatibility of the component (B) with PPE-1, and the resultant resin composition was introduced to a belt conveyer type cooling drier (a belt cooler). Using a twin-screw extruder (ZSK-40 mm ϕ , manufactured and sold by Werner Pfleiderer GmbH, Germany), the obtained masterbatch and HIPS-1 were subjected to melt extrusion according to formulations and conditions for producing the final products, which are indicated in Table 10, while removing the solvents under a vacuum of 0.1 mmHg, to thereby obtain a final product in the form of pellets. Using an injection molding machine (Model JSW J100EP manufactured and sold

by The Japan Steel Works, Ltd., Japan), the obtained pellets were subjected to injection molding at a cylinder temperature of 230 °C and at a mold temperature of 60 °C to obtain a specimen. The obtained specimen was evaluated with respect to various properties, such as color (yellowness index: YI). Results are shown in Table 10.

From Table 10, it can be observed:

that the compatibility of the oligomeric aromatic phosphoric ester with PPE-1 can be remarkably improved by the addition of a small amount of the solvent (D) (among various flame retardants which generally have good compatibility with PPE-1, the oligomeric aromatic phosphoric ester has relatively poor compatibility with PPE-1);

that, however, when the amount of the solvent (D) exceeded 100 parts by weight, relative to 100 parts by weight of the thermoplastic resin (A), it becomes difficult to maintain the above-mentioned vacuum condition, so that the solvent (D) is caused to remain in the resin composition to thereby lower the Vicat softening temperature of the produced resin composition (this indicates that the role of the solvent (D) is auxiliary and that when the compatibility between thermoplastic resin (A) and the non-polymerizable

liquid substance (B) is relatively low, the addition of a small amount of the solvent (D) renders it possible to improve the compatibility satisfactorily);

5 that when a lubricant oil is used as the non-polymerizable liquid substance (B), which has no compatibility with the thermoplastic resin (A), it becomes necessary to use a disadvantageously large amount of the solvent (D) for achieving a satisfactory compatibility between the component (A) and the component
10 (B) and therefore it becomes difficult to achieve the above-mentioned vacuum condition, thereby causing the above-mentioned disadvantages (Comparative Examples 35 and 36);

15 that when the resin composition is produced by a method in which the non-polymerizable liquid substance (B) is dispersed into the thermoplastic resin (A), since the thermoplastic resin (A) is not dissolved in the non-polymerizable liquid substance (B), the appearance of the produced resin composition becomes
20 extremely poor (Comparative Examples 37 and 38);

25 that when polyamide (PA) is used as the thermoplastic resin (A) and phosphoric ester (PB) is used as the non-polymerizable liquid substance (B), in which the compatibility between the components (A) and (B) is very poor, the addition of a large amount of a solvent

(FA) renders it possible to uniformly dissolve PA in PB; however, when the resultant mixture is used as a masterbatch in the second step, a phase separation occurs during the removal of the solvent (Comparative Examples 50 and 51).

Examples 54 to 69 and Comparative Examples 39 to 42

Resin compositions having the compositions shown in Tables 11 and 12 were respectively produced by three production processes described below, wherein HIPS's and PPE's, which have different reduced viscosities η_{sp}/c as shown in Tables 11 and 12, were individually used as the thermoplastic resin (A). Each of the obtained resin compositions was subjected to injection molding according to the same formulations and conditions as indicated in Table 10 to thereby obtain specimens. The specimens were evaluated with respect to flame retardancy, MFR, Izod impact strength, Vicat softening temperature and color (yellowness index: YI). Results are shown in Tables 11 and 12.

From Tables 11 and 12, it can be observed:

that when the production process (a) (using the resin composition of the present invention as a masterbatch) is employed, the resultant final resin composition exhibits excellent properties as compared to those

of the resin compositions produced by the production processes (b) and (c);

that, when HIPS having a reduced viscosity η_{sp}/c within the range of from 0.4 to 0.6 is employed as the thermoplastic resin (A), the produced resin composition exhibits an excellent balance of fluidity, Izod impact strength and flame retardancy;

that, especially when PPE having a reduced viscosity η_{sp}/c within the range of from 0.3 to 0.6 is employed, the produced resin composition exhibits an excellent balance of fluidity, heat resistance, Izod impact strength and flame retardancy.

(production processes)

(a): Using a batch type mixing vessel equipped with an agitator, the thermoplastic resin (A) was dissolved into the non-polymerizable liquid substance (B) at 200 °C while stirring. The resultant was introduced to a belt-conveyer type cooling drier (belt cooler) to thereby obtain a resin composition to be used as a masterbatch. Then, the masterbatch and HIPS were subjected to melt extrusion at 230 °C by means of a twin-screw extruder ZSK-40 mm ϕ (manufactured and sold by Werner Pfleiderer GmbH, Germany).

(b): A twin-screw extruder having a main feeder and a side feeder was used. The thermoplastic resin

(A) was fed from the main feeder and melted at 350 °C. Then the non-polymerizable liquid substance (B) was fed from the side feeder and kneaded at 230 °C to thereby obtain a resin composition to be used as a masterbatch, followed by melt extrusion in the same manner as in the production process (a).

(c): The non-polymerizable liquid substance (B) was portionwise added to thermoplastic resin (A) to cause the component (B) to be adsorbed on thermoplastic resin (A). The resultant adsorption mixture of the component (A) and the component (B) was mixed with HIPS to thereby obtain a mixture, followed by melt extrusion at 230 °C in the same manner as in the production process (a).

The above-mentioned production processes (a), (b) and (c) were used in the following Examples and Comparative Examples.

Examples 70 to 107 and Comparative Examples 43 to 49

Resin compositions having the compositions shown in Tables 13 to 17 were prepared using the materials and methods {production process (a) or (b)} as described in Tables 13 to 17}. The obtained resin compositions were evaluated with respect to flame retardancy, MFR, Izod impact strength, Vicat softening temperature, light stability, temperature at which a 1 %

weight loss occurred, and color (yellowness index: YI). Results are shown in Tables 13 to 17.

5 Table 13 shows the relationship between the type of the aromatic phosphoric ester and the properties of the resin composition.

10 Table 14 shows the influence of the amount of the styrene monomers and styrene oligomers remaining in the resin composition on the flame retardancy of the resin composition and the effect of addition of novolak resin and fluororesin.

15 Table 15 shows the effect of addition of a white pigment and a light resistance improver. From Table 15, it is apparent that, especially when it is intended to obtain a shaped resin article having a light color, since the resin composition of the present invention can be colored by the use of a small amount of a white pigment, a shaped article obtained using the resin composition of the present invention does not suffer lowering of mechanical properties (such as Izod impact strength) which is a problem accompanying a shaped resin article obtained using a conventional resin composition produced by a conventional melt-extrusion method (Comparative Examples).

20
25 Tables 16 and 17 show the effects of the addition of an aromatic phosphoric ester, a compatibility agent

(AS), a thermoplastic elastomer, TPE, and PPE-1 on the properties (such as Izod impact strength) of PC/HIPS type resin compositions and the influence of the ratio of PC to HIPS on the properties of the resin composition.

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Table 1

Example and Comparative Example	Composition (wt. ratio)		Compatibility between (A) and (B) (filtration-retaining ratio, %) 7)	Conditions for producing resin compositions		Stability of production	Evaluation of resin compositions				
	Thermoplastic resin (A) (PPE-1)	Non-polymerizable liquid substance (B) (TNPP)		Temperature (°C) 2)	Process 1)		Appearance	Yellowness index (YI)	Izod impact strength (J/m)	Light stability 3) (ΔE as measured after 300-hr radiation)	Amount of OH groups in PPE (mol./100 mol. of PPE units)
Ex. 1					I	good	good	41	88	12.1	0.3
Comp. 1					II-1	unstable extrusion	many discrete segments of PPE-1 5)	4)	29	4)	4)
Comp. 2	40	60	0	250	III	good	a large number of discrete segments of PPE-1 6)	4)	10		
Comp. 3					II-1	unstable extrusion	good	69	69	26.8	0.8
Comp. 4	40	60		350	II-2	unstable extrusion	many discrete segments of PPE-1 5)	4)	29	4)	4)

Note 1) Process-I (dissolution), Process-II-1 (melt extrusion), Process-II-2 (melt extrusion) and Process-III (melt extrusion) are as described herein.

2) Temperature : the temperature at which (A) is dissolved into (B) or melt-mixed with (B).

3) The smaller the ΔE value, the more excellent the light stability.

4) The surface condition is too coarse and non-uniform to measure yellowness index (YI).

5) Many discrete segments of PPE-1 that have not experienced melting are observed.

6) A large number of discrete segments of PPE-1 that have not experienced melting are observed.

7) Ratio (%) of the weight of a component (A)/component (B) mixture remaining on a filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration.

Table 2

Example and Comparative Example	Composition (wt. ratio)		Compatibility between (A) and (B) (filtration-re-maining ratio, %) 5)	Conditions for producing resin compositions		Stability of production	Evaluation of resin compositions		
	Thermoplastic resin (A) (PPE-1)	Non-polymerizable liquid substance (B) (TPP)		Temperature (°C) 2)	Process 1)		Appearance	Yellowness index (VI)	Amount of OH groups in PPE (mol./100 mol. of PPE units)
Ex. 2*				150				28	0.2
Ex. 3*				200				32	0.2
Ex. 4				250	I (dissolution)	good	good	37	0.2
Ex. 5				300				46	0.3
Comp. 5				310				56	0.6
Comp. 6				350				64	0.7
Comp. 7	30	70	0	150	II-1 (melt extrusion)	unstable extrusion	many discrete segments of PPE-1 4)	3)	3)
Comp. 8				200					
Comp. 9				250					
Comp. 10				300				65	0.8
Comp. 11				310			good	71	0.9
Comp. 12				350				78	1.0

Note * More preferred embodiment.

- 1) Process-I (dissolution) and Process-II-1 (melt extrusion) are as described herein.
- 2) Temperature : the temperature at which (A) is dissolved into (B) or melt-mixed with (B).
- 3) The surface condition is too coarse and non-uniform to measure yellowness index (VI).
- 4) Many discrete segments of PPE-1 that have not experienced melting are observed.
- 5) Ratio (%) of the weight of a component (A)/component (B) mixture remaining on a filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration.

Table 3

Example and Comparative Example	Composition (wt. ratio)		Compatibility between (A) and (B) (filtration-re-maining ratio, %) 6)	Conditions for producing resin compositions		Stability of production	Evaluation of resin compositions	
	Thermoplastic resin (A) (PPE-1)	Non-polymerizable liquid substance (B) (TPP)		Temperature (°C) 2)	Process 1)		Appearance	Yellowness index (YI)
Ex. 6*				150				9
Ex. 7*				200				10
Ex. 8*				230	I			11
Ex. 9				240	(dissolution)	good	good	17
Ex. 10				250				18
Ex. 11				300				22
Comp. 13				310				37
Comp. 14	30	70	0	350				45
Comp. 15				150			many discrete segments of PPE-1 4)	3)
Comp. 16				200			a few discrete segments of PPE-1 5)	
Comp. 17				250	II-1 (melt extrusion)	unstable extrusion		
Comp. 18				300			good	44
Comp. 19				310				50
Comp. 20				350				56

Note * More preferred embodiment.

- 1) Process-I (dissolution) and Process-II-1 (melt extrusion) are as described herein.
- 2) Temperature: the temperature at which (A) is dissolved into (B) or melt-mixed with (B).
- 3) The surface condition is too coarse and non-uniform to measure yellowness index (YI).
- 4) Many discrete segments of PPE-1 that have not experienced melting are observed.
- 5) Some discrete segments of PPE-1 that have not experienced melting are observed.
- 6) Ratio (%) of the weight of a component (A)/component (B) mixture remaining on a filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration.

Table 4

Example and Comparative Example	Composition (wt. ratio)		Compatibility between (A) and (B) (filtration-re-maining ratio, %) 3)	Conditions for producing resin compositions		Stability of production	Evaluation of resin compositions	
	Thermoplastic resin (A)	Non-polymerizable liquid substance (B)		Temperature (°C) 2)	Process 1)		Appearance	Yellowness index (YI)
Ex. 12*				150				8
Ex. 13*				180				9
Ex. 14				200	I (dissolution)			15
Ex. 15				250		good	good	16
Ex. 16				300				18
Comp. 21				310				30
Comp. 22	GPPS(30)	TPP (70)	0	350				41
Comp. 23				150				13
Comp. 24				200				17
Comp. 25				250	II-1 (melt extrusion)	unstable extrusion	good	22
Comp. 26				300				24
Comp. 27				310				36
Comp. 28				350				50
Ex. 17	HIPS-1 (30)	TPP (70)	4	200	I (dissolution)	good	good	8

Note * More preferred embodiment.

1) Process-I (dissolution) and Process-II-1 (melt extrusion) are as described herein.

2) Temperature ; the temperature at which (A) is dissolved into (B) or melt-mixed with (B).

3) Ratio (%) of the weight of a component (A)/component (B) mixture remaining on a filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration.

Table 5

Example and Comparative Example	Composition (wt. ratio)		Compatibility between (A) and (B) (filtration-re-maining ratio, %) 5)	Conditions for producing resin compositions		Stability of production	Evaluation of resin compositions		
	Thermo-Plastic resin (A)	Non-polymerizable liquid substance (B) (or comparative liquid substance)		Lower limit temperature for dissolution (°C) 2)	Process 1)		Appearance	Yellowness index (YI)	Amount of OH groups in PPE (mol./100 mol. of PPE units)
Ex. 18	PPE-1(20)	TPP (80)		109	I (dissolution)	good	good	15	0.2
Ex. 19	(50)	(50)	0	131				46	0.3
Ex. 20	(80)	(20)		172				70	0.4
Comp. 29	PPE-1(20)	A-Stat(80)		302	I (dissolution)		good	31	0.6
Comp. 30	(50)	(50)	61	329				61	0.7
Comp. 31	(80)	(20)		370				91	0.9
Comp. 32	PPE-1(20)	Lb (80)	75	Not dissolved even at 300 °C or more	I (dissolution)	many discrete segments of PPE-1 4) and unstable	many discrete segments of PPE-1 4)	3)	
Comp. 52	PPE-1(20)	TPP (80)			II-1 (melt extrusion)	TPP spouts out of the die, so that stable production is impossible.			
Comp. 53	(50)	(50)	0	310 (melting temperature) (°C)		unstable extrusion	good	73	1.1
Comp. 54	(70)	(30)				slightly unstable extrusion		88	1.2
Comp. 55	(80)	(20)				good		99	1.3

- Note 1) Process-I (dissolution) and Process-II-1 (melt extrusion) are as described herein.
 2) Resin composition is produced at the lower limit of the temperatures at which (A) is completely dissolved into (B).
 3) The surface condition is too coarse and non-uniform to measure yellowness index (YI).
 4) Many discrete segments of PPE-1 that have not experienced melting are observed.
 5) Ratio (%) of the weight of a component (A)/component (B) mixture remaining on a filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration.

Table 6

Example and Comparative Example	Composition (wt. ratio)					Compatibility between (A) and (B) (filtration-re-maining ratio, %) 3)	Conditions for producing resin compositions		Evaluation of resin compositions			
	Thermo-plastic resin (A) (PPE-1)	Non-polymerizable liquid substance (B)		Stabi-lizer (TNP)	Aromatic phosphoric ester		Lower limit tempe-rature for dissolu-tion (°C) 2)	Process 1)	Appear-ance	Yellow-ness index (YI)	Amount of OH groups in PPE (mol./ 100 mol. of PPE units)	
					Monomer (TNPP)							Oligomer (FR-1)
Ex. 21		0	50		9	200	I (disso-lution)	good	56	0.5		
Ex. 22*		5	45		0	171			51	0.4		
Ex. 23*	50	10	40	0	0	163			50	0.3		
Ex. 24		30	20		0	151			48	0.3		
Ex. 25		50	0		0	139			47	0.3		
Ex. 26*	50	0	50	0.5	9	200			50	0.4		

Note * More preferred embodiment.

1) Process-I (dissolution) is as described herein.

2) Resin composition is produced at the lower limit of the temperatures at which (A) is completely dissolved into (B).

3) Ratio (%) of the weight of a component (A)/component (B) mixture remaining on a filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration.

Table 7

Example and Comparative Example	Composition (wt. ratio)				Compatibility between (A) and (B) (filtration-re-maining ratio, %) 3)	Conditions for producing resin compositions		Evaluation of resin compositions		
	Thermo-plastic resin (A)		Non-polymerizable liquid substance (B) (SP value)	Process 1)		Lower limit temperature for dissolution (°C) 2)	Appearance	Yellow-ness index (YI)	Amount of OH groups in PPE (mol./ 100 mol. of PPE units)	
	PPE-1 SP value: 11.2	GPPS SP value: 10.5								
Ex. 27	50	0				I	200		56	0.5
Ex. 28	49	1	FR-1(50)(11.0)	9		(disso-lution)	181	good	50	0.4
Ex. 29	45	5					177		49	0.4
Ex. 30	40	10					161		47	0.3
Ex. 31			FR-2(50)(10.6)	6		I	158		46	0.3
Ex. 32			FR-3(50)(10.9)	4		(disso-lution)	140	good	45	0.3
Ex. 33	45	5	TPP (50)(10.7)	0			119		43	0.3
Ex. 34			TNPP(50) (9.4)	0			121		44	0.3

Note 1) Process-I (dissolution) is as described herein.

2) Resin composition is produced at the lower limit of the temperatures at which (A) is completely dissolved into (B).

3) Ratio (%) of the weight of a component (A)/component (B) mixture remaining on a filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration.

Table 8

Example and Comparative Example	Composition (wt. ratio)			Compat- ibility between (A) and (B) (filtra- tion-re- taining ratio, %) 3)	Conditions for producing resin compositions		Evaluation of resin compositions		
	Thermo- plastic resin (A) (PPE-1)	Non- polymer- izable liquid substance (B) (TPP)	Solid sub- stance (C)		Tempe- rature (°C) 2)	Process 1)	Appearance	Yellow- ness index (YI)	Amount of OH groups in PPE (mol./ 100 mol. of PPE units)
Ex. 35	10	90					good, however, bleeding slightly occurs	5	0.2
Ex. 36	20	80						20	0.2
Ex. 37*	30	70				I (disso- lution)		34	0.2
Ex. 38*	40	60	0		220			43	0.2
Ex. 39*	50	50						55	0.3
Ex. 40*	60	40					good	63	0.3
Ex. 41*	70	30		0				71	0.3
Ex. 42	80	20						79	0.5
Ex. 43	90	10						86	0.5
Ex. 44	10	90	0.5(SI)			I (disso- lution)	good (especially, the resin composition is free of blocking)	6	0.2
Ex. 45	20	80	0.5(SI)		220			18	0.2
Ex. 46	30	70	0.5(SI)					32	0.2
Ex. 47	20	80	0.5(SDV)					17	0.2

Note * More preferred embodiment.

1) Process-1 (dissolution) is as described herein.

2) Temperature : the temperature at which (A) is dissolved into (B) or melt-mixed with (B).

3) Ratio (%) of the weight of a component (A)/component (B) mixture remaining on a filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration.

Table 9

Example and Comparative Example	Composition (wt. ratio)			Compat-ibility between (A) and (B) (filtration-re- maining ratio, %) 7)	Conditions for producing resin compositions		State of dissolution	Evaluation of resin compositions	
	Thermo-Plastic resin (A)	Non-polymer-izable liquid substance (B)	Solid substance (C)		Temperature (°C) 2)	Process 1)		Appearance	Yellowness index (YI)
Ex. 48	GPPS(30)	Br-EP(70)	0	0	200	I (dissolution)	3)	good	9
Ex. 49	HIPS-1(30)	Br-EP(70)	0	4	200		4)		10
Ex. 50	ABS(30)	Br-EP(70)	0	5	200		pasty	many discrete segments of PPE-1 6)	12
Comp. 33	PPE-1(30)	TPP(70)	0	0	20				5)
Comp. 34	PPE-1(30)	TPP(70)	SDV(50)	0	200		pasty		

Note 1) Process-I (dissolution) is as described herein.

2) Temperature: the temperature at which (A) is dissolved into (B) or melt-mixed with (B).

3) Uniform dissolution.

4) Dissolution (for the most part) and swelling (for a part).

5) The surface condition is too coarse and non-uniform to measure yellowness index (YI).

6) Many discrete segments of PPE-1 that have not experienced melting are observed.

7) Ratio (%) of the weight of a component (A)/component (B) mixture remaining on a filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration.

Table 10

Example and Comparative Example	1st step: Production of master batch (MB)						Compat- ibility between (A) and (B)	2nd step: Melt extrusion using MB			Evaluation of resin compositions			
	Composition of MB (wt.ratio)			Conditions for producing resin compositions		State of mixing (A) and (B)		Composition of final product 2) (wt.ratio)	Conditions for producing resin compositions		Yellow- ness index (YI)	Izod impact stran- gth (J/m)	Vicat soften- ing tem- perature (°C)	Amount of OH groups in PPE (mol./ 100 mol. of PPE units)
	Thermo- plas- tic resin (A) (PPE- 1)	Non-poly- merizable liquid sub- stance (B) (or compara- tive liquid substance)	Solvent (D)	Dis- solu- tion tem- pera- ture (°C)	Process			MB	Ther- moplas- tic resin (HPS-1)	Melt extru- sion tem- pera- ture (°C)				
Ex. 108	50	50 (FR-1)	0	100		3)	9	20	80		7	80	94	0.2
EX. 51	50	50 (FR-1)	0	150		4)	9	20	80		7	88	94	0.2
Ex. 52	50	50 (FR-1)	10 (XY)	150	I (disso- lution)	5)	9	22	80		5	88	93	0.2
Ex. 53	50	50 (FR-1)	60 (XY)	100		5)	9	32	80	230	6	78	85	0.2
Comp. 35	50	50 (1b)	0	300		6)	75	20	80		Not conducted	The surface condition is too coarse and non-uniform to measure the above properties		
Comp. 36	50	50 (1b)	100 (Mcl)	40		5)	75	40	80		Conducted	Above properties cannot be measured due to phase separation		
Comp. 37	98	2 (TPP)	98 (MeOH)	20	Disper- sion	7)	0	20.2	80		Conducted	Many discrete segments of PPE-1 are observed and the surface condition is too coarse and non-uniform to measure the above properties		
Comp. 38	83	17 (MO)	0	20		7)	52	20	80		Not conducted	The surface condition is too coarse and non-uniform to measure the above properties		
Comp. 50	50 (PA6)	50 (PB)	0	150	I (disso- lution)	6)	86	20	80		Not conducted	The surface condition is too coarse and non-uniform to measure the above properties		
Comp. 51	50 (PA6)	50 (PB)	250 (FA)	150		5)	86	70	80		Conducted	Above properties cannot be measured due to phase separation		

Notes for Table 10

- 1) Ratio (%) of the weight of a component (A)/component (B) mixture remaining on a filter to the weight of the thermoplastic resin (A) in the resin composition before the filtration.
- 2) Final product was prepared so that the weight ratio of PPE-1/non-polymerizable liquid substance is 10/10.
- 3) Swelling.
- 4) Dissolution (for the most part) and swelling (for a part).
- 5) Uniform dissolution.
- 6) Phase separation.
- 7) Dispersion.

Table 11

Items		Ex.54	Ex.55*	Ex.56*	Ex.57*	Ex.58*	Ex.59	Comp.39	Comp.40			
Production process 1)		(a)										
1 st step	(A) PPE-1	20										
	(B) FR-1	14										
Composition (wt. ratio)	2 nd step	η_{sp}/c HIPS-2 -3 -4 -1 -5 -6	80	80	80	80	80	80	80			
										0.79		
										0.60		
										0.58		
										0.53		
										0.40		
Evaluation of resin composi- tions	Flame re- tardancy	Flame-out time (sec)		14.6	13.1	12.5	11.2	10.5	9.8	15.1	28.2	
		Dripping of flam- ing particles Evaluation ac- cording to UL-94 (Thickness of the test specimen: 1/8 inch)		Ob- served	Ob- served	Ob- served	Ob- served	Ob- served	Ob- served	Ob- served	Ob- served	
	MFR (g/10 minute)	3.8		4.0	5.0	5.2	7.0	8.0	4.9	12.5		
		70		67	64	62	59	40	56	13		
	Vicat softening temperature (°C)		95		95	95	95	95	96	85		
	Surface of the molded article		Good								Good	3)
	Yellowness index (YI)		19	23	18	21	20	21	53	4)		

Notes: * represents a more preferred embodiment.

1) Production processes (a), (b) and (c) are as described herein (Examples 54-69 and Comparative Examples 39-42)

2) HB means the flame retardancy of the sample which does not conform to any of the standards V-0, V-1 and V-2 of UL-94.

3) A large number of discrete segments of PPE-1 that have not experienced melting are observed.

4) The surface condition is too coarse and non-uniform to measure yellowness index(YI).

Table 12

Items		Ex.	Ex.	Ex.	Ex.	Ex.	Ex.	Ex.	Ex.	Ex.	Ex.	Ex.	Comp.	Comp.	Ex.		
		60	61*	62*	63*	64	65	66*	67*	68	(b)			(c)	(a)		
Production process 1)		(a)															
Composi- tion (wt. ratio)	(A) Polyphenylene ether (parts by weight)	5	10	20	25	30	20	20	20	20					10	35	
	Reduced viscosity η_{sp}/c	0.14	0.41	0.41	0.41	0.41	0.25	0.30	0.60	0.65	0.41			0.41	0.41		
	(B) FR-1	15														15	15
	HIPS-1	95	90	80	75	70	80	80	80	80				80	65		
Evalua- tion of resin compo- sitions	Flame-out time (sec)	15.1	12.3	9.5	8.1	5.1	8.1	8.9	10.3	12.1	16.1			27.6	3.0		
	Dripping of flaming particles	Observed	Observed	Observed	Observed	Not observed	Observed	Observed	Observed	Observed	Observed			Observed	Not observed		
	Evaluation according to UL-94 (Thickness of the test specimen: 1/8 inch)	V-2	V-2	V-2	V-2	V-1	V-2	V-2	V-2	V-2	V-2			HB 2)	V-0		
	MFR (g/10 minute)	12.3	8.1	5.5	5.4	3.9	7.9	6.1	5.0	3.8	5.0			15.1	2.0		
	Izod impact strength (J/m)	69	67	65	64	59	40	55	70	76	57			13	69		
	Vicat softening temperature (°C)	82	90	94	99	104	94	94	95	95	94			79	110		
Surface of the molded article		Good														Good	3)
Yellowness index (YI)		2	5	19	23	29	20	18	21	19	55			4)	32		

Notes: * represents a more preferred embodiment.

- 1) Production processes (a), (b) and (c) are as described herein (Examples 54-69 and Comparative Examples 39-42)
- 2) HB means the flame retardancy of the sample which does not conform to any of the standards V-0, V-1 and V-2 of UL-94.
- 3) A large number of discrete segments of PPE-1 that have not experienced melting are observed.
- 4) The surface condition is too coarse and non-uniform to measure yellowness index (YI).

Table 13

Items		Ex. 70	Ex. 71	Ex. 72	Ex. 73	Ex. 74	Ex. 75	Comp. 43
Production process 1)		(a)						
Composition (wt. ratio)	(A) PPE-1	15						15
	(B) FR-1	14	14	7	14	14	14	14
1 st step	FR-2			7				
	TNPP			7				
2 nd step	FR-3							
	TPP							
Evaluation of resin compo- sitions	HIPS-2	85						85
	Flame-out time (sec)	15.3	14.2	18.1	21.3	20.0	12.1	17.3
Flame retar- dancy	Dripping of flaming particles	Observed	Observed	Observed	Observed	Observed	Observed	Observed
	Evaluation according to UL-94 (Thickness of the test specimen: 1/8 inch)	V-2	V-2	V-2	V-2	V-2	V-2	V-2
MFR (g/10 minute)		9.8	7.3	9.3	9.5	7.2	13.2	9.9
	Izod impact strength (J/m)	65	68	72	68	52	77	57
Vicat softening temperature (°C)		92	97	97	95	95	86	92
	Temperature at which 1 % weight loss occurred (°C)	299	300	299	298	320	194	297
Yellowness index (YI)		12	13	13	14	13	12	48

Notes: 1) Production processes (a), (b) and (c) are as described herein (Examples 54-69 and Comparative Examples 39-42)

Table 14

Items		Ex. 76*	Ex. 77*	Ex. 78	Ex. 79	Ex. 80	Ex. 81	Comp. 44	Ex. 82	Ex. 83	Ex. 84	Ex. 85
Production process 1)		(a)										
Compo- sition (wt. ratio)	1 st step	(b)										
		(a)										
	(A) PPE-1	15										
	GPPS	15										
	(B) FR-1	15										
	HIPS-1	70										
	NK	0										
2 nd step	PTEK	0										
	Content of styrene monomer and styrene oligomer in the resin composition (wt.%)	0.1	1.0	1.2	2.0	0.3	0.3	0.4	0.5	0.4	0.4	0.7
Evalua- tion of resin compo- sitions	Flame-out time (sec)	9	12	20	25	10	15	13	3	5	4	2
	Dripping of flaming particles	Observed	Observed	Observed	Observed	Observed	Not observed	Observed	Observed	Not observed	Not observed	Not observed
	Evaluation according to UL-94 (Thickness of the test specimen: 1/16 inch)	V-2	V-2	V-2	V-2	V-2	V-1	V-2	V-2	V-0	V-0	V-0
	MFR (g/10 minute)	11.0	11.2	11.4	11.5	11.9	11.5	11.0	7.1	7.0	4.9	2.3
	Izod impact strength (J/m)	61	60	58	57	60	63	60	79	81	96	113
	Vicat softening temperature (°C)	93	92	91	90	99	100	93	105	106	121	138
	Surface of the molded article	Good										
	Yellowness index (YI)	12	13	14	16	13	11	46	27	28	40	51

Notes: * represents a more preferred embodiment.

1) Production processes (a), (b) and (c) are as described herein (Examples 54-69 and Comparative Examples 39-42)

Table 15

Items		Ex. 86	Ex. 87	Ex. 88	Comp.	Comp.	Comp.	Ex. 89	Ex. 90	Ex. 91	Ex. 92	
Production process 1)												
Composition (wt. ratio)	1 st step	(A) PPE-1	20			20			20			
		(B) FR-1	13			13			13			
		HIPS-1	80			80			80			
	2 nd step	White pigment (TiO ₂)	0	2	4	0	2	4				
		UVA		0			0		0.3	0.3	0.3	0.3
		HALS		0			0		0	0.2	0	0.2
		EP		0			0		0	0	0.1	0.1
		Flame-out time (sec)	10	11	13	11	12	14	11	12	13	12
		Dripping of flaming particles	Observed	Observed	Observed	Observed	Observed	Observed	Observed	Observed	Observed	Observed
		Evaluation according to UL-94 (Thickness of the test specimen: 1/8 inch)	V-2	V-2	V-2	V-2	V-2	V-2	V-2	V-2	V-2	V-2
Evaluation of resin compositions	MFR (g/10 minute)	7	7	8	7	7	8	7	8	9	8	
	Izod impact strength (J/m)	118	108	98	69	49	29	108	108	108	108	
	Vicat softening temperature (°C)	102	102	101	102	101	101	102	101	100	101	
	Surface of the molded article	Good			Good			Good				
	Yellowness index (YI)	19	14	7	54	43	36	14	14	15	14	
	Light stability (Δ E as measured after 300-hr-radiation)	9	8	6	16	14	13	6	4	2	4	

Notes: 1) Production processes (a), (b) and (c) are as described herein (Examples 54-69 and Comparative Examples 39-42)

Table 16

Items		Ex. 93	Ex. 94	Ex. 95	Ex. 96	Ex. 97	Ex. 98	Ex. 99	Ex. 100	Comp.
Production process 1)		(a)								
Composition (wt. ratio)	1 st step	62								
	(A) PC	57								
	PPE-1	0								
	(B)	14 (FR-2)	14 (TNPP)	7/7(FR- 2/TNPP)	14 (FR-3)	14 (FR-1)	14 (TPP)	0	5	5
	2 nd step	38								
Evaluation of resin compo- sitions	HIPS-1	38								
	AS	0								
	Evaluation according to UL-94 (Thickness of the test specimen: 1/8 inch)	V-0	V-2	V-0	V-0	V-0	V-0	V-0	V-0	V-0
	Evaluation according to UL-94 (Thickness of the test specimen: 1/16 inch)	V-1	V-2	V-1	V-1	V-1	V-1	V-1	V-0	V-0
	MFR (g/10 minute)	3	4	5	5	6	9	6	5	4
	Izod impact strength (J/m)	147	108	137	88	108	108	294	98	78
	Vicat softening temperature (°C)	116	114	115	111	109	99	109	115	116
	Temperature at which 1 % weight loss occurred (°C)	300	298	299	320	280	194	281	282	281
	Yellowness index (YI)	16	15	12	14	13	13	16	21	43

Notes: 1) Production processes (a), (b) and (c) are as described herein (Examples 54-69 and Comparative Examples 39-42)

Table 17

Items		Ex.101	Ex.102	Ex.103	Ex.104	Ex.105	Ex.106	Ex.107	Comp.47
Production process 1)		(a)							
Composition (wt. ratio)	1 st step	5	20	30	60	70	95	30	30
	(A) PC								
	(B) FR-1				20			20	20
	2 nd step	95	80	70	40	30	5	65	70
Evaluation of resin com- positions	HIPS-1								
	TPE				0			5	0
	Evaluation according to UL-94 (Thickness of the test specimen: 1/8 inch)	V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0
	Evaluation according to UL-94 (Thickness of the test specimen: 1/16 inch)	V-1	V-1	V-1	V-1	V-0	V-0	V-0	V-1
	MFR (g/10 minute)	12	10	8	6	3	2	8	8
	Izod impact strength (J/m)	176	167	157	137	108	98	304	108
	Vicat softening temperature (°C)	99	101	111	114	116	118	110	111
	Yellowness index (YI)	7	11	13	15	17	21	14	23

Notes: 1) Production processes (a), (b) and (c) are as described herein (Examples 54-69 and Comparative Examples 39-42)

INDUSTRIAL APPLICABILITY

The resin composition of the present invention is free from problems, such as heat deterioration, accompanying a resin composition produced by a melt-extrusion method and, therefore, can be advantageously used for obtaining a shaped article which is markedly improved with respect to appearance, such as color, light stability and mechanical properties. Further, when the resin composition of the present invention is used as a masterbatch for preparing a molding material, wherein the masterbatch is added to a resin material which is the same as or different from the thermoplastic resin used in the resin composition, especially great advantages are brought about. The molding material obtained by adding the resin composition of the present invention as a masterbatch has advantages not only in that the molding material has improved melt moldability, but also in that, by virtue of the fact that the resin in the masterbatch has not experienced high-temperature conditions, a shaped article which is improved with respect to the above-mentioned properties can be easily produced from the molding material. Accordingly, the resin composition of the present invention can be advantageously used as a molding material for producing various shaped articles, for example, parts for house-

hold electric appliances, such as a VTR (video tape recorder), a distribution switchboard, a television set, an audio player, a condenser, a household plug socket, a cassette tape recorder, a videocassette, a
5 video disk player, an air conditioner, a humidifier and an electric machine for providing heated air; parts for office automation machines, such as a main frame (mechanical chassis) for a CD-ROM, a printer, a facsimile, a PPC (plain paper copier), a CRT (cathode-ray
10 tube), a word processor, a copying machine, an electronic register, an office computer system, a floppy disk drive, a keyboard, a typewriter, an ECRU (electronic cash resister), an electric calculator, a toner cartridge and a telephone; electric and electronic
15 parts, such as a connector, a coil bobbin, a switch, a relay, a relay socket, an LED (light-emitting diode), a variable condenser, an AC (alternating current) adapter, a FBT high-voltage bobbin, a FBT case, a FIT coil bobbin, a jack, a volume shaft and motor parts; and
20 parts for an automobile, such as an instrument panel, a radiator grille, a cluster, a speaker grille, a louver, a console box, a defroster garnish, an ornament, a fuse box, a relay case and a connector shift tape.

25



CLAIMS

1. A resin composition which is substantially the same as that produced by a method comprising the step
5 of mixing together:

(A) a thermoplastic resin, and

(B) at least one non-polymerizable liquid substance as an additive for said thermoplastic resin (A), said non-polymerizable liquid substance (B) being in a
10 liquid state at a temperature in the range of from 25 to 300 °C,

said mixing being conducted at a temperature in the range of from 25 to 300 °C, at which said at least one non-polymerizable liquid substance (B) maintains a
15 liquid state,

thereby causing said thermoplastic resin (A) to be dissolved in or swelled with said at least one non-polymerizable liquid substance (B).

20 2. The resin composition according to claim 1, wherein said at least one non-polymerizable liquid substance (B) is selected from the group consisting of a flame retardant, a plasticizer, a heat stabilizer and a light stabilizer.



3. The resin composition according to claim 1 or 2, wherein said thermoplastic resin (A) and said non-polymerizable liquid substance (B) are mixed together with (C) a solid substance as an additive for said thermoplastic resin (A), said solid substance (C) being a crystalline solid or an amorphous solid at a temperature of 300 °C or less.
4. The resin composition according to any one of claims 1 to 3, wherein said mixing is conducted at a temperature in the range of from 100 to 300 °C, at which said at least one non-polymerizable liquid substance (B) maintains a liquid state.
5. The resin composition according to any one of claims 1 to 4, wherein said mixing is conducted at a temperature in the range of from 150 to 250 °C, at which said at least one non-polymerizable liquid substance (B) maintains a liquid state.
6. The resin composition according to any one of claims 1 to 5, wherein the weight ratio of said thermoplastic resin (A) to said at least one non-polymerizable liquid substance (B) is from 10/90 to 90/10.



7. The resin composition according to claim 6, wherein the weight ratio of said thermoplastic resin (A) to said at least one non-polymerizable liquid substance (B) is from 30/70 to 70/30.

5

8. The resin composition according to any one of claims 1 to 7, wherein said thermoplastic resin (A) comprises 1 to 100 % by weight of a polyphenylene ether resin and 0 to 99 % by weight of a polystyrene resin, the total amount of said polyphenylene ether resin and said polystyrene resin being 100 % by weight.

10

9. The resin composition according to claim 8, wherein said thermoplastic resin (A) comprises 50 to 99 % by weight of a polyphenylene ether resin and 1 to 50 % by weight of a polystyrene resin, the total amount of said polyphenylene ether resin and said polystyrene resin being 100 % by weight.

15

10. The resin composition according to any one of claims 1 to 9, wherein said non-polymerizable liquid substance (B) is a flame retardant.

20

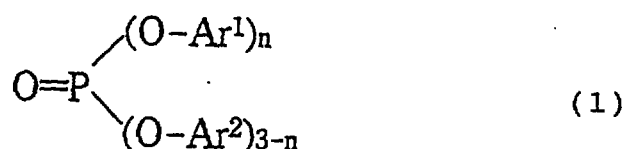
11. The resin composition according to claim 10, wherein said flame retardant comprises at least one

25

organic phosphorus compound.

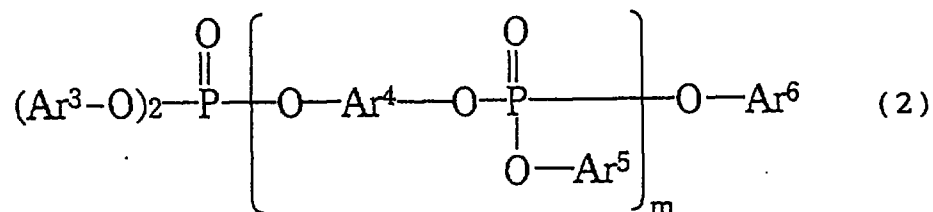
12. The resin composition according to claim 11,
wherein said organic phosphorus compound is at least
5 one aromatic phosphoric ester.

13. The resin composition according to claim 12,
wherein said aromatic phosphoric ester is a monomeric
aromatic phosphoric ester represented by the following
10 formula (1):



15 wherein each of Ar^1 and Ar^2 independently
represents a $\text{C}_6\text{-C}_{20}$ aromatic group which is
unsubstituted or substituted with at least
one $\text{C}_1\text{-C}_{10}$ hydrocarbon group; and each n is
identical to each other and represents an
20 integer of from 0 to 3.

14. The resin composition according to claim 12,
wherein said aromatic phosphoric ester is an oligomeric
aromatic phosphoric ester represented by the following
25 formula (2):



5

wherein each of Ar^3 , Ar^4 , Ar^5 and Ar^6 independently represents a C_6 - C_{20} aromatic group which is unsubstituted or substituted with at least one C_1 - C_{10} hydrocarbon group; and

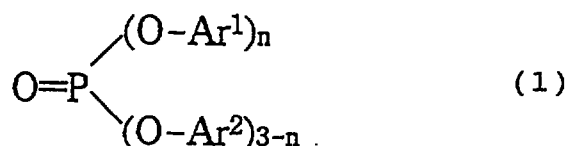
10

m is an integer of 1 or more.

15. The resin composition according to claim 12, wherein said at least one aromatic phosphoric ester is a mixture of:

15

a monomeric aromatic phosphoric ester represented by the following formula (1):

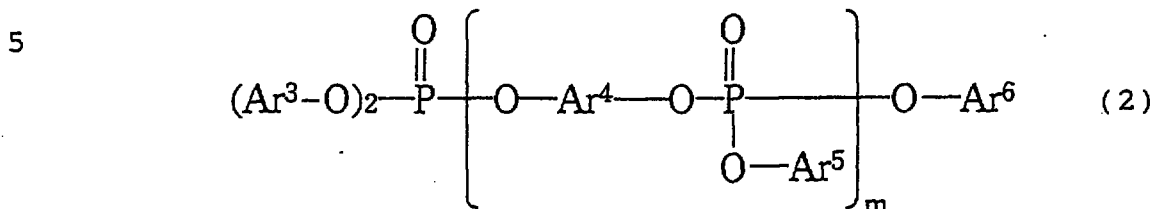


20

wherein each of Ar^1 and Ar^2 independently represents a C_6 - C_{20} aromatic group which is unsubstituted or substituted with at least one C_1 - C_{10} hydrocarbon group; and each n is identical to each other and represents an

25

integer of from 0 to 3, and
an oligomeric aromatic phosphoric ester represented by the following formula (2):



10 wherein each of Ar^3 , Ar^4 , Ar^5 and Ar^6 independently represents a C_6 - C_{20} aromatic group which is unsubstituted or substituted with at least one C_1 - C_{10} hydrocarbon group; and m is an integer of 1 or more.

15 16. The resin composition according to claim 15, wherein said mixture contains 1 to 99 % by weight of said monomeric aromatic phosphoric ester and 1 to 99 % by weight of said oligomeric aromatic phosphoric ester, the total amount of said monomeric aromatic phosphoric
20 ester and said oligomeric aromatic phosphoric ester being 100 % by weight.

17. The resin composition according to claim 16, wherein said mixture contains 1 to 50 % by weight of
25 said monomeric aromatic phosphoric ester and 50 to 99 %



by weight of said oligomeric aromatic phosphoric ester, the total amount of said monomeric aromatic phosphoric ester and said oligomeric aromatic phosphoric ester being 100 % by weight.

5

18. The resin composition according to any one of claims 3 to 17, wherein the amount of said solid substance (C) is from 0.000001 to 100 parts by weight, relative to 100 parts by weight of said thermoplastic resin (A).

10

19. The resin composition according to claim 18, wherein the amount of said solid substance (C) is from 0.1 to 10 parts by weight, relative to 100 parts by weight of said thermoplastic resin (A).

15

20. The resin composition according to any one of claims 3 to 19, wherein said solid substance (C) is at least one additive selected from the group consisting of an adhesion-preventive agent, an oil absorber, a heat stabilizer and a light stabilizer.

20

21. The resin composition according to any one of claims 1 to 20, wherein said thermoplastic resin (A) comprises an aromatic vinyl polymer and optionally at

25



least one compound selected from the group consisting of an aromatic vinyl monomer, an aromatic vinyl dimer and an aromatic vinyl trimer, the amount of said at least one compound being 1 % by weight or less, based on the weight of said resin composition.

22. The resin composition according to claim 1, wherein said mixing of said thermoplastic resin (A) and said non-polymerizable liquid substance (B) together is performed while heating at a temperature not exceeding 300 °C, and wherein, after said mixing, the resultant is cooled, followed by pelletization, whereby the resin composition is produced in the form of pellets.

23. The resin composition according to claim 1, wherein said mixing is performed in the presence of (D) a solvent, and, after said mixing, said solvent (D) is removed.

24. The resin composition according to claim 23, wherein the amount of said solvent (D) is 0.1 to 100 parts by weight, relative to 100 parts by weight of said thermoplastic resin (A).

25. The resin composition according to claim 23 or 24,



wherein said solvent (D) has a boiling point which is higher than the temperature at which said mixing is performed and which is not higher than 300 °C.

5 26. A method for producing an shaped article, which comprises subjecting to melt extrusion:

(i) a thermoplastic resin composition which is substantially the same as that produced by a method comprising the step of mixing together:

10 (A) a thermoplastic resin, and

(B) at least one non-polymerizable liquid substance as an additive for said thermoplastic resin (A), said non-polymerizable liquid substance (B) being in a liquid state at a temperature in the range of from 25 to 300 °C,

15

said mixing being conducted at a temperature in the range of from 25 to 300 °C, at which said at least one non-polymerizable liquid substance (B) maintains a liquid state,

20 thereby causing said thermoplastic resin (A) to be dissolved in or swelled with said at least one substance (B); and

(ii) a thermoplastic resin which is the same as or different from said thermoplastic resin (A).

25



27. The method according to claim 26, wherein said thermoplastic resin (ii) comprises at least one thermoplastic elastomer.

5 28. The method according to claim 26 or 27, wherein said resin composition (i) and said thermoplastic resin (ii) are subjected to melt extrusion together with (E) at least one additive selected from the group consisting of a flame retardant, an auxiliary agent for a
10 flame retardant, a light stabilizer, a mold release agent, a flowability-improving agent, a heat stabilizer, an anti-static agent, a coloring agent and a foaming agent, said at least one additive (E) being the same as or different from said non-polymerizable liquid
15 substance (B).

29. A shaped article produced by the method according to any one of claims 26 to 28.

20 30. A polyphenylene ether resin composition comprising:
(A') a polyphenylene ether resin comprising a plurality of polymer chains, each comprising recurring oxyphenylene units, said polyphenylene ether resin (A') containing hydroxyl groups in an amount of not more
25 than 0.5 mole, relative to 100 moles of said recurring



oxyphenylene units, and

(B') at least one additive for said polyphenylene ether resin (A').

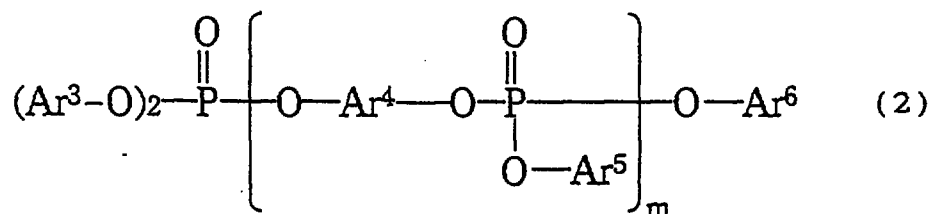
- 5 31. The resin composition according to claim 30, wherein said at least one additive (B') is selected from the group consisting of a flame retardant, a plasticizer, a heat stabilizer and a light stabilizer.
- 10 32. The resin composition according to any one of claims 30 and 31, wherein the weight ratio of said polyphenylene ether resin (A') to said at least one additive (B') is from 10/90 to 90/10.
- 15 33. The resin composition according to any one of claims 30 to 32, wherein said at least one additive (B') is a flame retardant.
- 20 34. The resin composition according to claim 33, wherein said flame retardant comprises at least one organic phosphorus compound.
- 25 35. The resin composition according to claim 34, wherein said organic phosphorus compound is at least one aromatic phosphoric ester.

36. The resin composition according to claim 35, wherein said aromatic phosphoric ester is a monomeric aromatic phosphoric ester represented by the following formula (1):



wherein each of Ar^1 and Ar^2 independently represents a C_6 - C_{20} aromatic group which is unsubstituted or substituted with at least one C_1 - C_{10} hydrocarbon group; and each n is identical to each other and represents an integer of from 0 to 3.

37. The resin composition according to claim 35, wherein said aromatic phosphoric ester is an oligomeric aromatic phosphoric ester represented by the following formula (2):



wherein each of Ar^3 , Ar^4 , Ar^5 and Ar^6 inde-

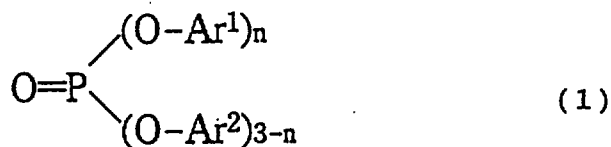


pendently represents a C₆-C₂₀ aromatic group which is unsubstituted or substituted with at least one C₁-C₁₀ hydrocarbon group; and m is an integer of 1 or more.

5

38. The resin composition according to claim 35, wherein said at least one aromatic phosphoric ester is a mixture of:

a monomeric aromatic phosphoric ester represented by the following formula (1):



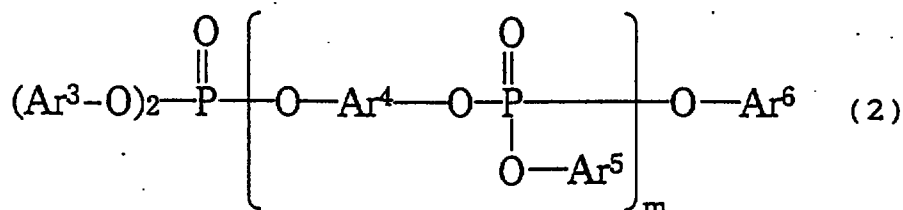
15

wherein each of Ar¹ and Ar² independently represents a C₆-C₂₀ aromatic group which is unsubstituted or substituted with at least one C₁-C₁₀ hydrocarbon group; and each n is identical to each other and represents an integer of from 0 to 3, and

20

an oligomeric aromatic phosphoric ester represented by the following formula (2):

25





wherein each of Ar^3 , Ar^4 , Ar^5 and Ar^6 independently represents a $\text{C}_6\text{-C}_{20}$ aromatic group which is unsubstituted or substituted with at least one $\text{C}_1\text{-C}_{10}$ hydrocarbon group; and
5 m is an integer of 1 or more.

39. The resin composition according to claim 38,
wherein said mixture contains 1 to 99 % by weight of
10 said monomeric aromatic phosphoric ester and 1 to 99 %
by weight of said oligomeric aromatic phosphoric ester,
the total amount of said monomeric aromatic phosphoric
ester and said oligomeric aromatic phosphoric ester
being 100 % by weight.

15 40. The resin composition according to claim 39,
wherein said mixture contains 1 to 50 % by weight of
said monomeric aromatic phosphoric ester and 50 to 99 %
by weight of said oligomeric aromatic phosphoric ester,
20 the total amount of said monomeric aromatic phosphoric
ester and said oligomeric aromatic phosphoric ester
being 100 % by weight.

41. A shaped article produced from the resin composition of any one of claims 1 to 25, which is a film, a
25



sheet or a foamed article.

42. A shaped article produced from the resin composition of any one of claims 1 to 25, which is a part for
5 an office automation machine, a part for an electric
appliance, a part for an electronic appliance or a part
for an automobile.

43. Use of the resin composition of any one of claims
10 1 to 25 as a material for injection molding.

44. The shaped article according to claim 29, which is
a film, a sheet or a foamed article.

15 45. The shaped article according to claim 29, which is
a part for an office automation machine, a part for an
electric appliance, a part for an electronic appliance
or a part for an automobile.

20 46. The shaped article according to claim 29, which is
a pellet for use as a material for injection molding.

47. A shaped article produced from the resin composition of any one of claims 30 to 40, which is a film, a
25 sheet or a foamed article.



48. A shaped article produced from a resin composition
of any one of claims 30 to 40, which is a part for an
office automation machine, a part for an electric
5 appliance, a part for an electronic appliance or a part
for an automobile.

49. Use of the resin composition of any one of claims
30 to 40 as a material for injection molding.

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ABSTRACT

Disclosed is a resin composition which is substantially the same as that produced by a method comprising the step of mixing together (A) a thermoplastic resin and (B) at least one non-polymerizable liquid substance as an additive for the thermoplastic resin (A), wherein the non-polymerizable liquid (B) is in a liquid state at a temperature in the range of from 25 to 300 °C and wherein the mixing is conducted at a temperature in the range of from 25 to 300 °C, at which the non-polymerizable liquid (B) maintains a liquid state, thereby causing the thermoplastic resin (A) to be dissolved in or swelled with the non-polymerizable liquid (B). By molding the resin composition of the present invention, a shaped article having excellent appearance, excellent light stability and excellent mechanical properties can be obtained. Therefore, the resin composition of the present invention can be advantageously used as a molding material for producing various shaped articles, such as parts for office automation machines, parts for household electric appliances, and electric and electronic parts. Further, the molding material obtained by adding to a resin material the resin composition of the present invention as a masterbatch is especially advantageous not only in that the molding material has improved melt moldability, but also in that, by virtue of the fact that the resin in the masterbatch has not experienced high-temperature conditions, a shaped article which is improved with respect to the above-mentioned properties can be easily produced from the molding material.

DECLARATION

I, Maho KASEKI, c/o the Inoue & Associates of 3rd Floor,
Akasaka Habitation Building, 3-5, Akasaka 1-chome, Minato-ku,
Tokyo, Japan do solemnly and sincerely declare that I am con-
versant with the Japanese and English languages and that I have
executed with the best of my ability this translation into Eng-
lish of the drawings of the international application No.
PCT/JP97/03719, and believe that the translation is true and
correct.

Maho Kaseki
Maho KASEKI

February 29, 2000
(date)

FIG.1

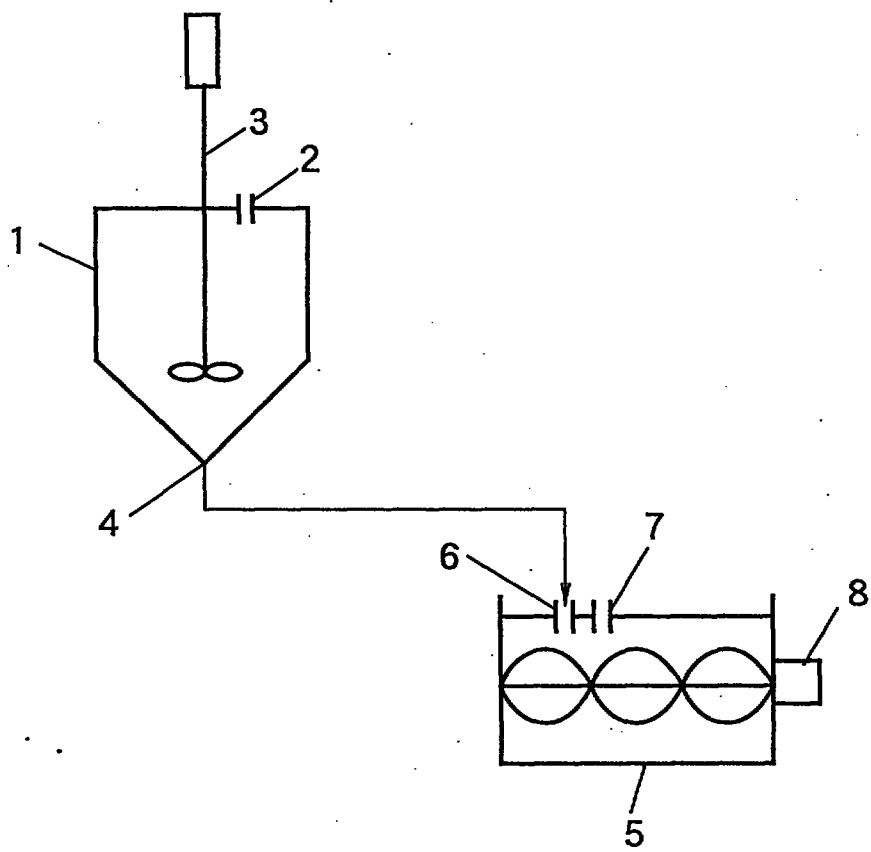


FIG.2

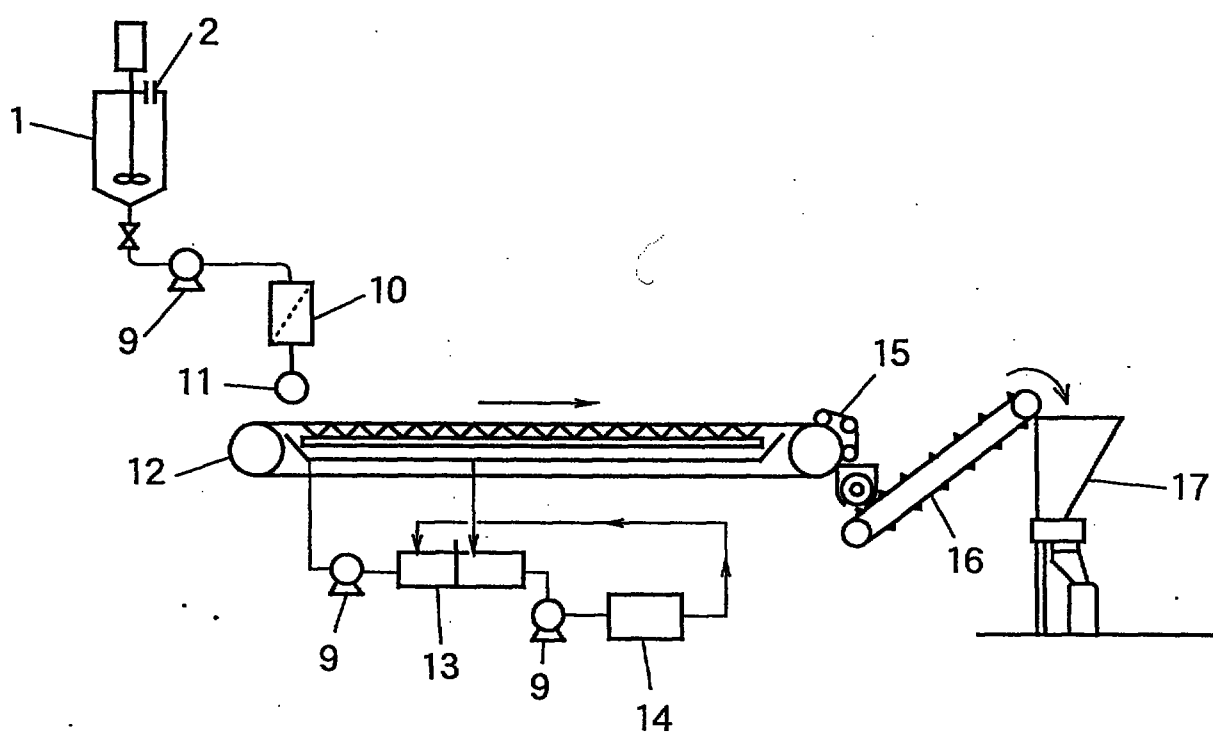


FIG.3 (a)

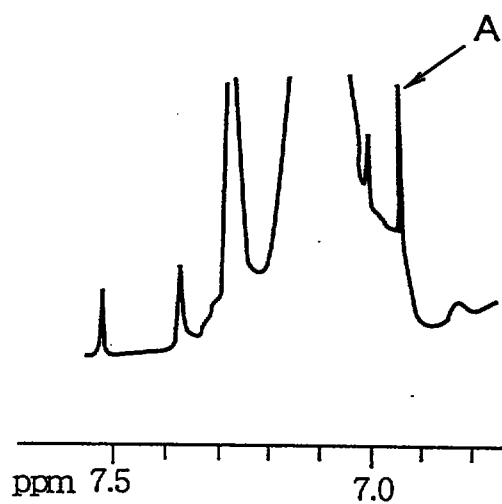


FIG.3 (b)

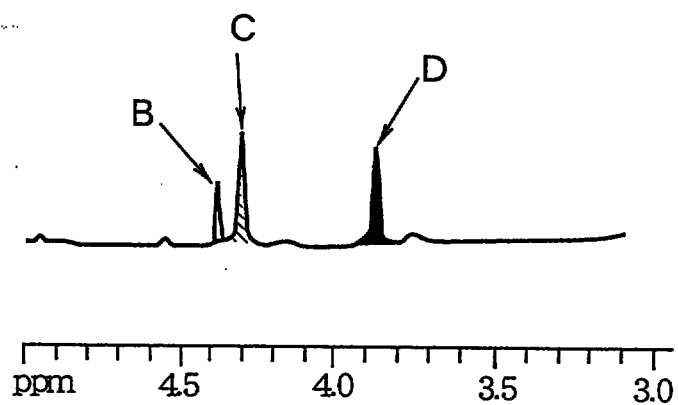


FIG.4 (a)

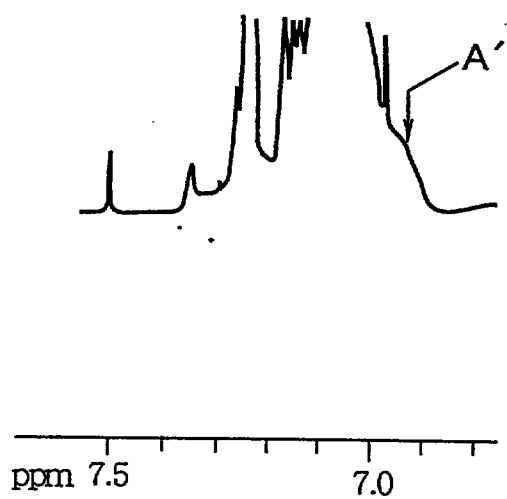


FIG.4 (b)

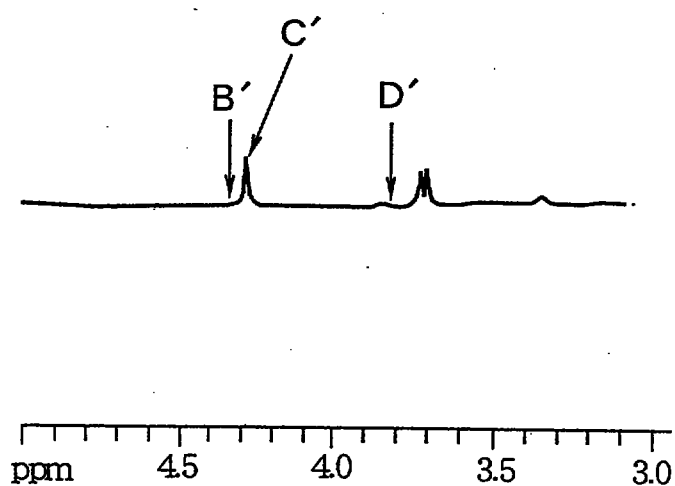


FIG.5

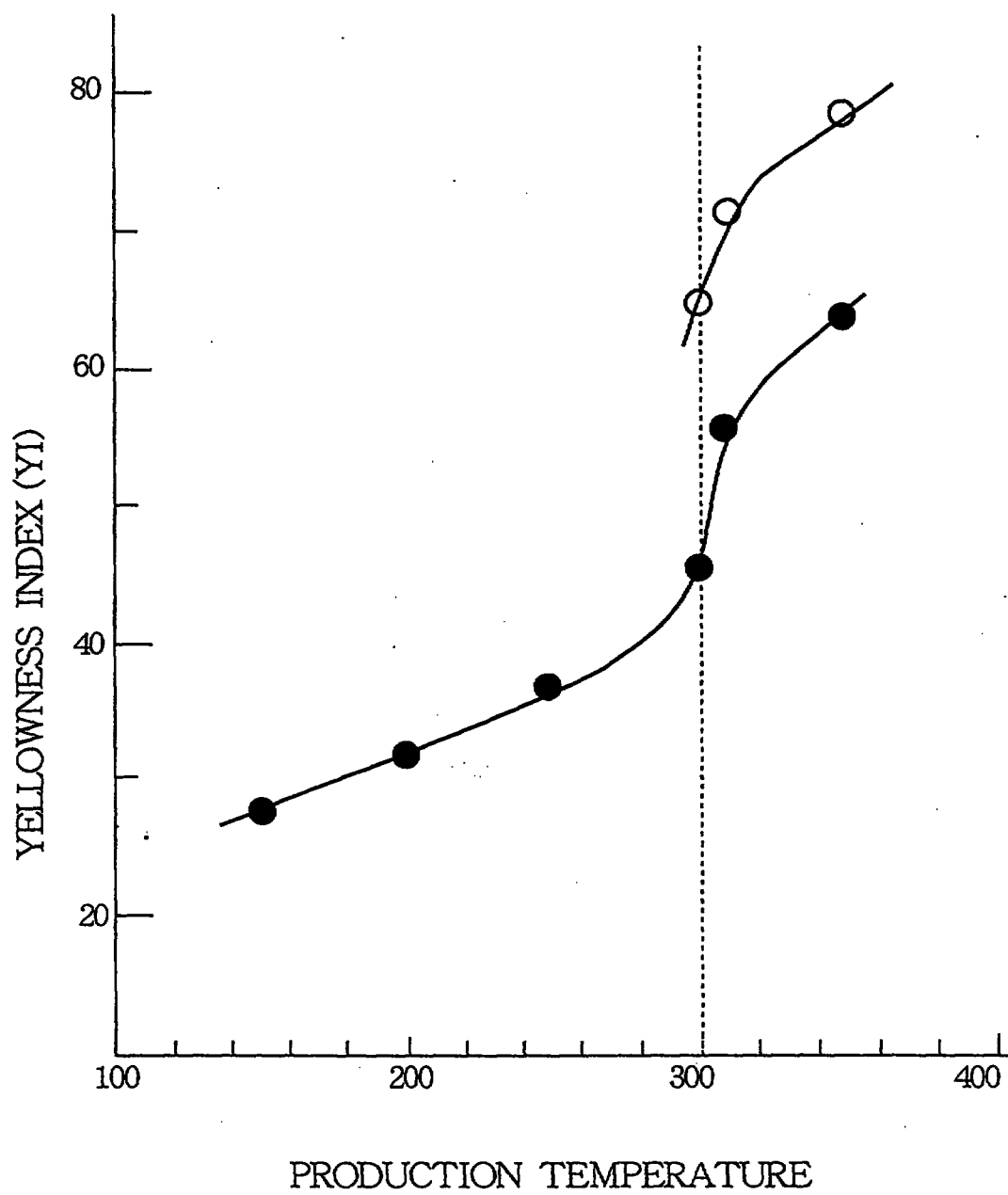


FIG.6

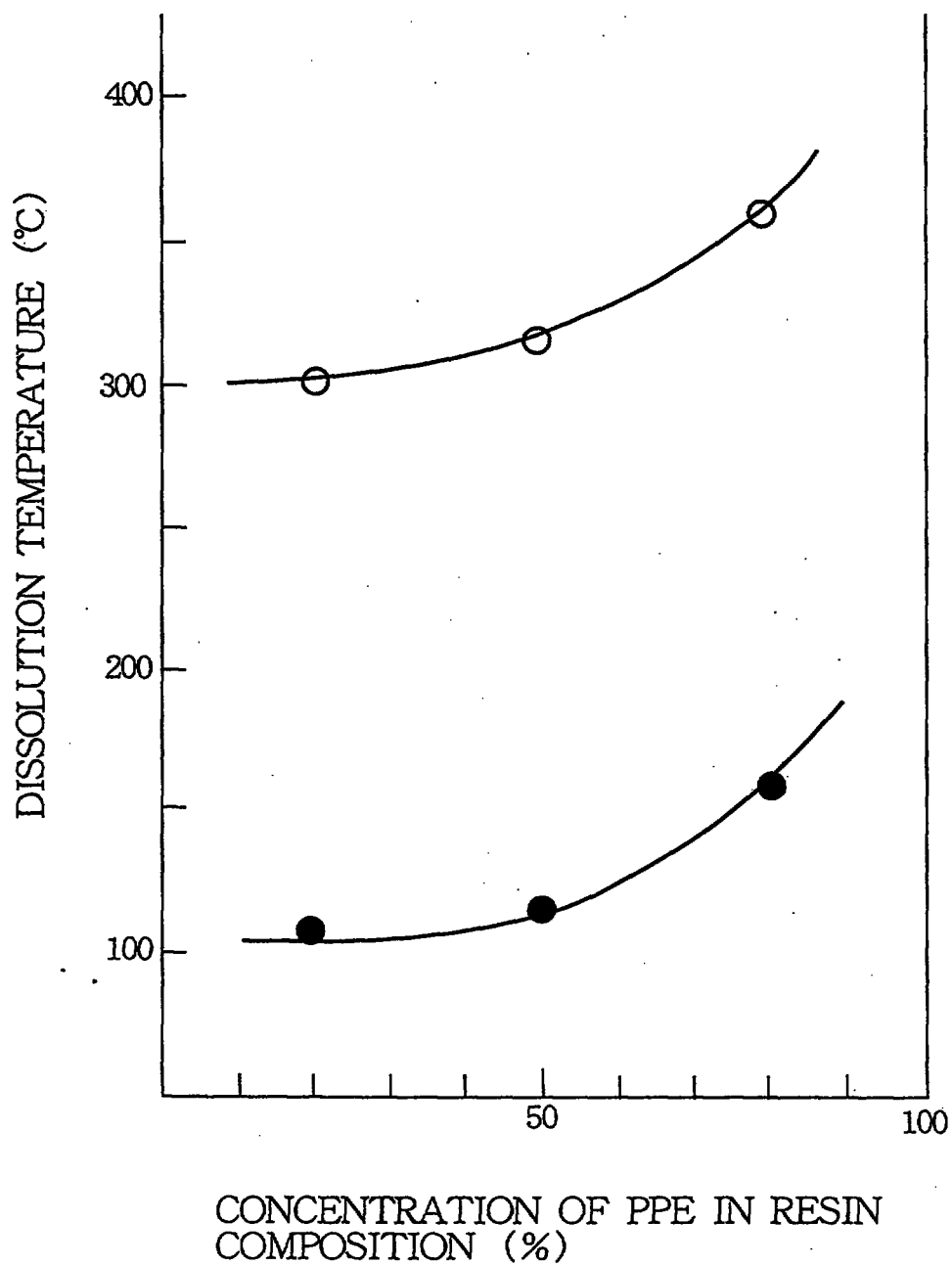


FIG.7

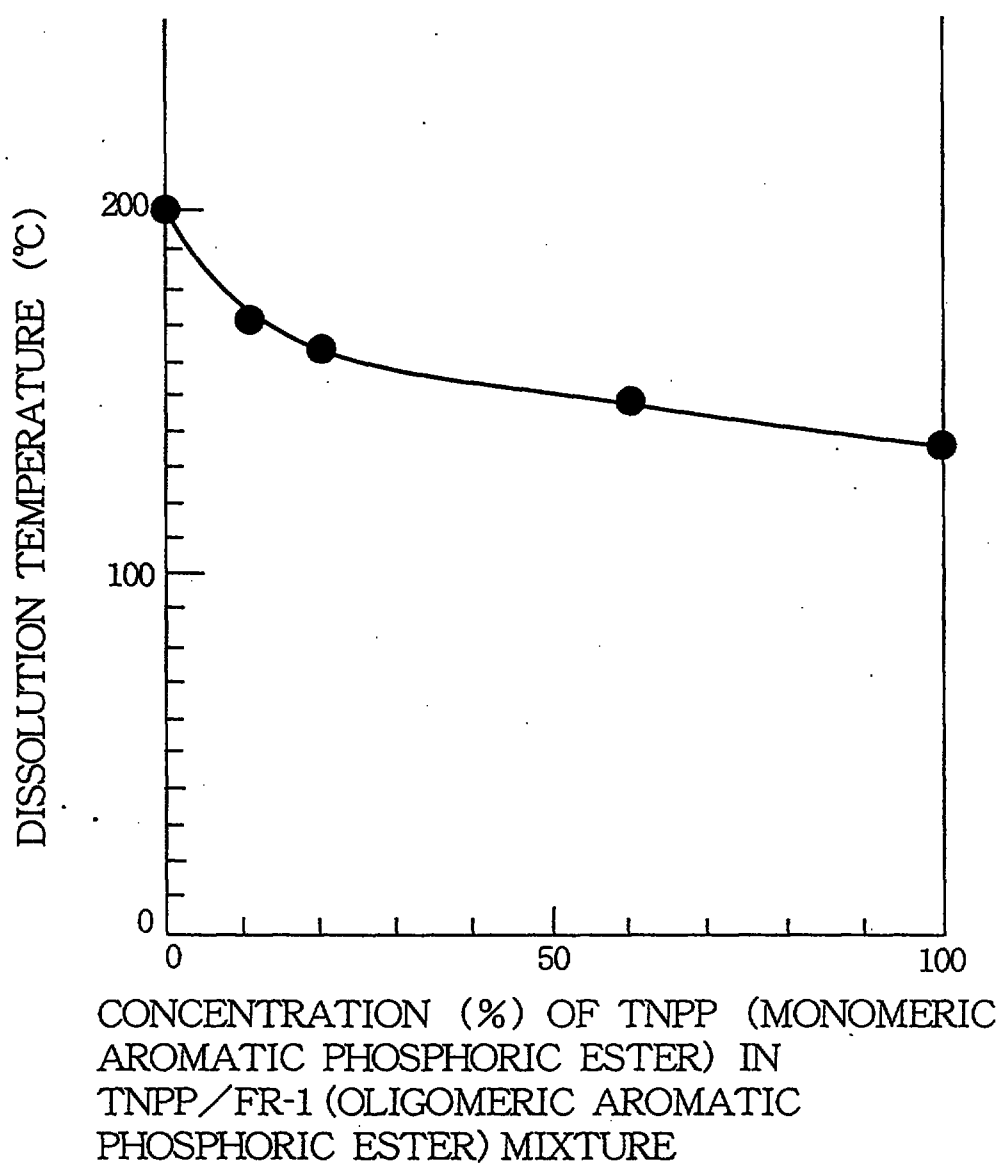


FIG.8

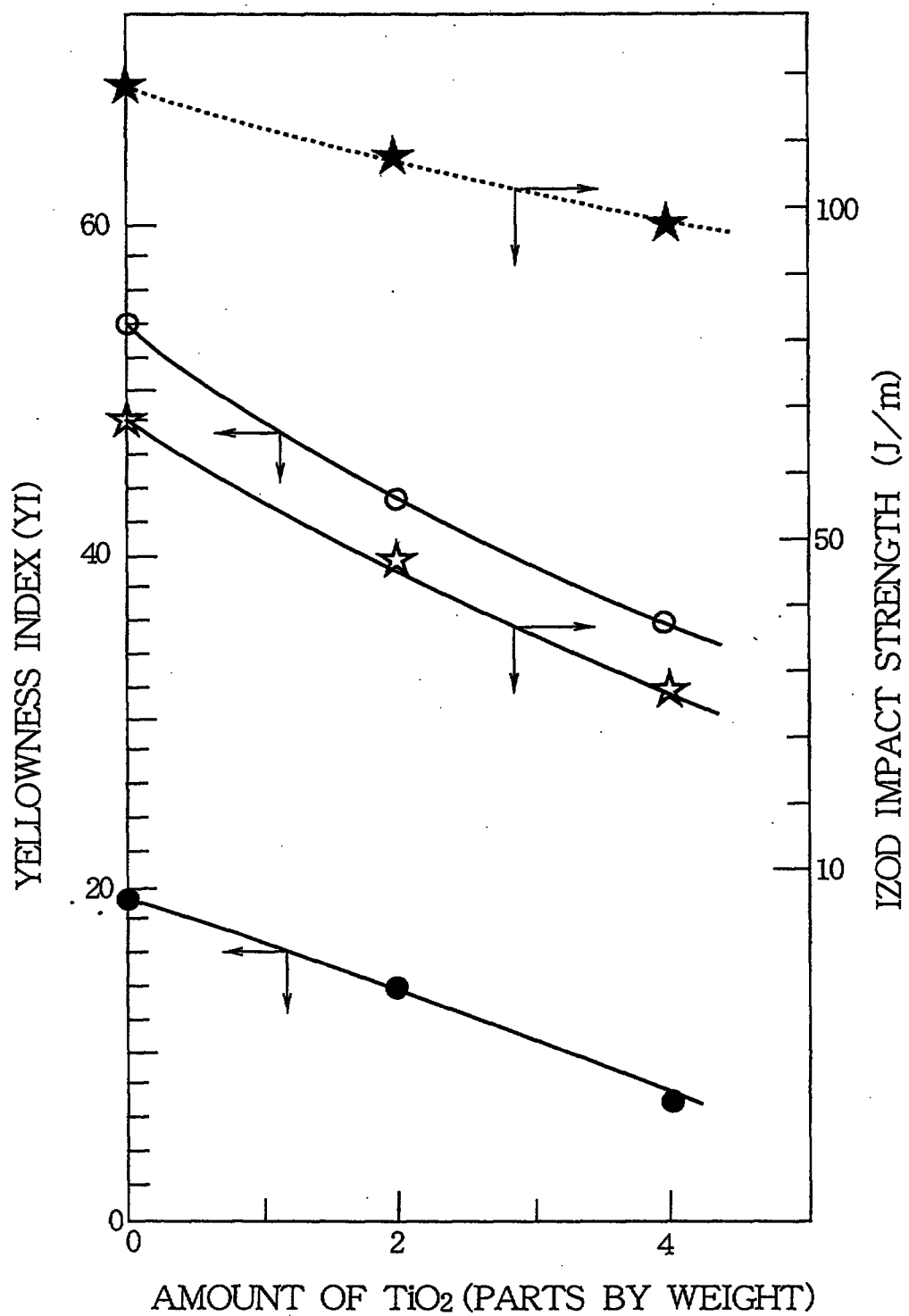


FIG.9 (a)

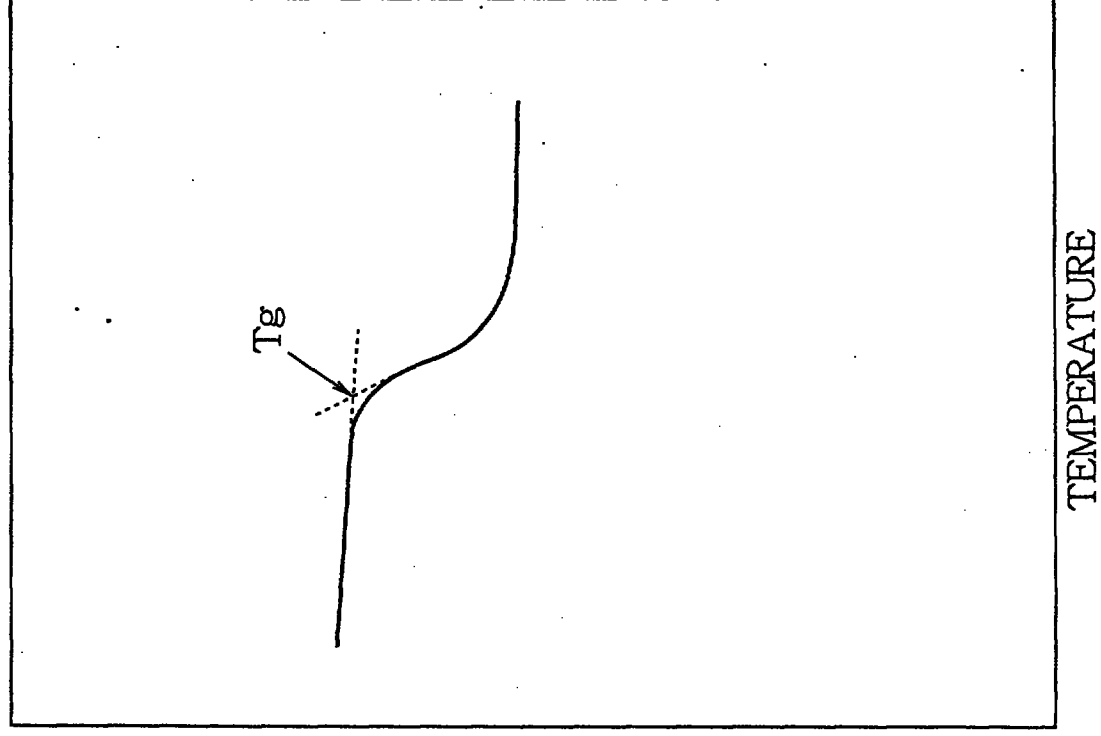
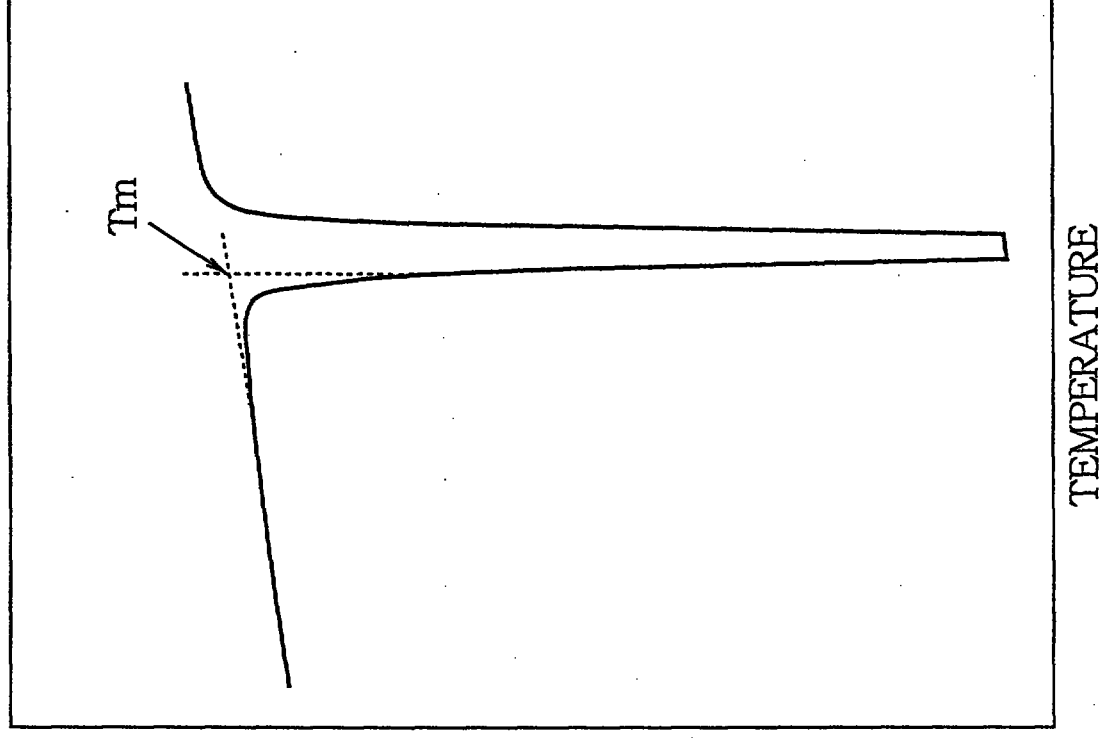


FIG.9 (b)

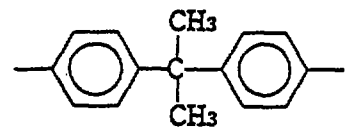


REPLACEMENT SHEET

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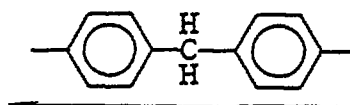
98 Substitute sheet -
6 January 1999

wherein each Ar^1 represents



, and

each Ar^2 represents



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The above-mentioned silicone resin used as a flame retardant auxiliary is one which has a three-dimensional network structure which is obtained by combining the structural units SiO_2 , $\text{RSiO}_{3/2}$, RSiO and $\text{RSiO}_{1/2}$. In these structural units, each R independently represents an alkyl group, such as a methyl group, an ethyl group or a propyl group, an aromatic group, such as a phenyl group or a benzyl group, or a vinyl group-containing substituent group obtained by introducing a vinyl group to the above-mentioned alkyl group or the above-mentioned aromatic group. Especially, a silicone resin, in which R is the above-mentioned vinyl group-containing substituent group, obtained by introducing a vinyl group to the above-mentioned alkyl group or the above-mentioned aromatic group, is preferred.

The above-mentioned silicone resin can be obtained by hydrolyzing organohalosilanes corresponding to the above-mentioned structural units and subsequently polymerizing the hydrolyzed products.

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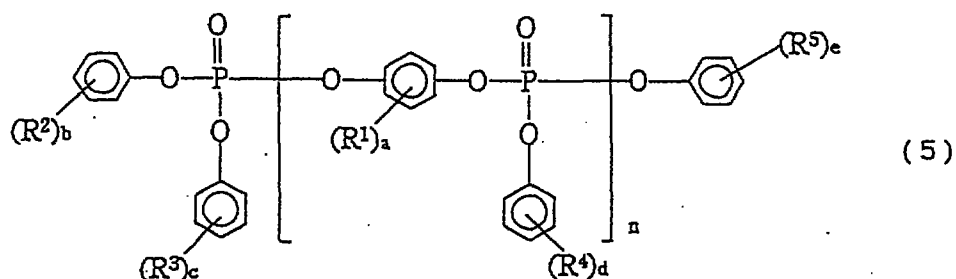
A silicone oil used as a flame retardant auxiliary

28 OCT 1999

73 Substitute sheet - 27/10/99

preferred are compounds of formula (2) in which Ar^4 is a phenyl group or a bivalent group derived from a diarylalkane, from the viewpoint of achieving high resistance to hydrolysis and thermal stability. Specific examples of such compounds of formula (2) include bisphenol A bis(diphenylphosphate) and bisphenol A bis(dicresylphosphate).

Further preferred examples of compounds of formula ~~(2)~~² include an oligomeric aromatic phosphoric ester represented by the following formula (5):



wherein each of a, b, c, d and e independently represents an integer of from 0 to 3; each of R^1 to R^5 independently represents a C_1 - C_{10} hydrocarbon group; and n represents an integer of from 1 to 3.

With respect to the oligomeric aromatic phosphoric esters represented by formula (5), it is preferred that a is 0; each of b, c, d and e is 2; and the substitution positions of each of the di-substituted aromatic rings are 2- and 6-positions. Such a compound of

REPLACEMENT SHEET

28 OCT 1999

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hold electric appliances, such as a VTR (video tape recorder), a distribution switchboard, a television set, an audio player, a condenser, a household plug socket, a cassette tape recorder, a videocassette, a video disk player, an air conditioner, a humidifier and an electric machine for providing heated air; parts for office automation machines, such as a main frame (mechanical chassis) for a CD-ROM, a printer, a facsimile, a PPC (plain paper copier), a CRT (cathode-ray tube), a word processor, a copying machine, an electronic register, an office computer system, a floppy disk drive, a keyboard, a typewriter, an ~~ECRU~~ register (electronic cash ~~resister~~), an electric calculator, a toner cartridge and a telephone; electric and electronic parts, such as a connector, a coil bobbin, a switch, a relay, a relay socket, an LED (light-emitting diode), a variable condenser, an AC (alternating current) adapter, a FBT high-voltage bobbin, a FBT case, a FIT coil bobbin, a jack, a volume shaft and motor parts; and parts for an automobile, such as an instrument panel, a radiator grille, a cluster, a speaker grille, a louver, a console box, a defroster garnish, an ornament, a fuse box, a relay case and a connector shift tape.