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NASA TECHNICAL MEMORANDUM

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INCOMBUSTIBLE RESIN COMPOSITION

Toshio Akima



Translation of "Nannensei jushi soseibutsu;" Japanese Patent No. 51-57745 (1976), Applicant: Hitachi Kasei Kogyo K.K.; Date of Application, November 18, 1974; Date of Disclosure, May 20, 1976; pp.239-241

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1. Title of Invention

Incombustible resin composition.

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2. Scope of Patent Claim

Incombustible resin composition composed of aromatic compounds (however, at least one type of reaction ingredient is a halide) obtained through heat reaction of

a. combustible polymer material and

b. bisphenol A or halogenated bisphenol A and bisphenol A diglycidyl ether or halogenated bisphenol A diglycidyl ether.

3. Detailed Description of the Invention

This invention relates to incombustible resin compositions.

In recent years, there has been marked development of polymer materials, but various types of polymer materials are combustible, which is a lethal flaw for use in electrical parts, construction materials, clothing, bedding and general merchandise. Various organic halide compounds and organic phosphorous compound fire retardants have been developed in order to eliminate this flaw, and several of them have been perfected. For example, hexabromobenzene, the organic halogen compound with the highest halogen content, has outstanding incombustibility, but a flaw is its sublimation properties.

In addition, special groups of compounds among organic halide compounds have attracted concern because of toxicity to

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people, and conventional halide fire retardants have come under reexamination from the viewpoint of pollution in addition to their practical properties.

The inventors have completed this invention which eliminates these problems.

Specifically, this invention provides incombustible resin compositions composed of aromatic compounds (however, at least one type of reaction ingredient is a halide) obtained through heat reaction of

- a. combustible polymer material and
- b. bisphenol A or halogenated bisphenol A and bisphenol A diglycidyl ether or halogenated bisphenol A diglycidyl ether.

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The novel aromatic compound in this invention is an adduct of bifunctional phenols and bifunctional epoxy resins. Bifunctional phenols are substitution products of 1 to 8 halogens in bisphenol A, including bisphenol A and tetrabromobisphenol A, tetrachlorobisphenol A. Bifunctional epoxy resins are diglycidyl ethers of the aforementioned bifunctional phenols including bisphenol A diglycidyl ether and tetrabromobisphenol A diglycidyl ether. However, epoxy resins and/or phenols must contain halogen atoms.

The reaction between the aforementioned bifunctional phenols and bifunctional epoxy resins is implemented by heating in a conventional etherification reaction. Conventional catalysts are used, including lithium hydroxide.

The combustible polymer materials which are rendered incombustible through this invention include polyester resins, epoxy resins, polystyrene, polyethylene, polypropylene, ABS resin, phenol resin, polymethylmethacrylate, polyamide and nylon.

The novel aromatic compounds produced in this invention have no specific restriction regarding their molecular weight, but a molecular weight above 4,000 is desirable in relation to the melting point of the polymer material.

Blending polymer materials to a halogen content of 3 to 4%, specifically 5 to 15% in the case of bromine and 10 to 30% in the case of chlorine, would be effective. In addition, the fire retardant effect could be enhanced through the use of fire retardant promoters such as antimony trioxide.

Examples of synthesis of aromatic compounds are illustrated below.

Synthesis Example 1

489.6 g (0.9 moles) of tetrabromobisphenol A, 780 g (1.0 mole) (390 epoxy equivalents) of tetrabromobisphenol A diglycidyl ether and 0.24 g (0.01 moles) of basic catalytic lithium hydroxide were placed in a 1 liter three neck distillation flask and subjected to a heating reaction at 120°C. Since the viscosity of the reaction liquid increased after two hours of reaction, the temperature was gradually raised to 180°C, and the heating reaction was conducted for two hours. After the completion of the reaction, the solution was cooled, the solidified product was pulverized, and pale yellow powder was produced. (Bromine content of 51.5%). The softening point of the reaction product was 140 to 142°C (ring and ball method). The basic structure was confirmed through infrared absorption spectrum and NMR spectrum, and the molecular weight distribution was examined by gel paper chromatography (G.P.C.) and by high speed liquid chromatography to be 3,000 to 4,000.

Synthesis Example 2

A blend of 272 g (0.5 moles) of tetrabromobisphenol A, 925 g (0.25 moles) of tetrachlorobisphenol A, and 57.0 g (0.25 moles) of bisphenol A as the phenol with 780 g (1.0 moles) of tetrabromobisphenol A diglycidyl ether in epoxy resin was synthesized under the same conditions as in the aforementioned synthesis, producing a pale yellow resinous substance with a softening point of 130 to 135°C. Examination of the molecular weight distribution by GPC and high speed liquid chromatography revealed a virtually uniform composition with molecular weight of approximately 3,000.

Synthesis Example 3

0.24 g (0.01 mole) of catalytic lithium hydroxide was added to a blend of 335.2 g (0.8 moles) of tetrabromobisphenol A, 380 g (0.5 moles) of tetrabromobisphenol A diglycidyl ether, and 190 g (0.5 moles) (190 epoxy equivalents) of bisphenol A diglycidyl ether, and a heating reaction was implemented in accordance with synthesis example (1), producing pale yellow powder with a softening point of 132 to 133°C.

Actual examples of this invention are illustrated below.

Actual Example 1

25 weight parts of the compound produced in synthesis example 1, 10 weight parts of antimony trioxide, 20 weight parts of quartz glass, 80 weight parts of acidic anhydrous hardener HN-2200 (brand name of Hitachi Kasei Kogyo K.K.) and 3 weight parts of hardening promotor EMI24CN (made by Shikoku Kasei) were blended in 100 weight parts of commercial epoxy resin (Epiccoat 828, brand name of Shell Inc.), infused in a die, and set for 4 hours at 150°C.

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The resulting resin hardened material passed SE-O in UL-94 combustion tests.

Actual Example 2

30 weight parts of the adduct obtained in synthesis example 2 and 15 weight parts of antimony trioxide were thoroughly blended in 100 weight parts of commercial AAS-resin (V-6100 A, brand name of Hitachi Kasei Kogyo K.K.), and the product produced through conventional injection molding at 230°C passed SE-I in the UL-94 combustion tests.

Actual Example 3

15 weight parts of the compound produced in synthesis example 3 and 10 weight parts of antimony trioxide were blended in 100 weight parts of commercial ABS resin (V-3000, brand name of Hitachi Kasei Kogyo K.K.), and sample pieces produced through conventional molding passed SE-I in the UL-94 combustion tests.

The novel compound of this invention has outstanding thermal stability, and does not exhibit sublimation properties nor the odor of decomposition even at high temperatures (150 to 200°C) (differential thermal analysis 275°C, 5% weight reduction). In addition, its compatibility with polyolefins and other thermosetting resins is good. Since this compound is soluble in various organic solvents (aromatic hydrocarbons, ketones, esters), its advantages can be efficiently exhibited when applied to sheets, paper, fibers etc. which are to be coated to render them incombustible.

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