

was filtered to remove residual crystalline p-hydroxy benzoic acid. The benzene was stripped out with a vacuum and heating on a steam bath. The product was washed three times using a total of 500 ml. of pentane. The pentane was removed by evaporation and the product was thinned with toluene and mixed with activated carbon and a filtering aid and filtered to remove residual crystals of p-hydroxy-benzoic acid. The solvents were removed by the use of a vacuum, moderate heating and stirring, yielding a clear, yellow, slightly viscous liquid having a refractive index of $n_D^{24}=1.507$. The yield of 115 gm. was approximately 40% of the theoretical yield. When an amine accelerator such as N,N-di-methyl-sym-m-xylylidine or N,N-di-methyl-toluidine was added to this liquid product and its mixture subsequently combined with particulate fillers containing benzoyl peroxide, lauroyl peroxide, or both, the mixture hardened at room temperature within 0.5 to 60 minutes, depending upon the concentrations of the amine accelerator, the benzoyl peroxide and the nature and surface treatment of the particulate filler. For example, when the product contained 0.58% of N,N-dimethyl-sym-m-xylylidine and was then mixed with zinc oxide powder containing 1% benzoyl peroxide, the mixture hardened in approximately 1 minute at room temperature. However, when to the product additional stabilizers (inhibitors) were added so as to give a total of 0.58% N,N-dimethyl-sym-m-xylylidine, 0.23% butylated hydroxy toluene and 0.19% di-t-butylsulfide, this formulation, when mixed with zinc oxide powder containing 1% benzoyl peroxide had a hardening time of about 6 minutes.

When used as a coupling agent between dentin and a composite restorative material, the adhesive bond strength was approximately twice that of a control as shown in Table 2.

The structure of the instant compound may be compared closely with that of methyl-p-hydroxybenzoate used as a preservative for galenicals, etc., and butyl-p-hydroxybenzoate, which is used as a preservative for cosmetic emulsions. It is theorized for provisional utility that the present compound 2-methacryloxyethyl-p-hydroxybenzoate may have commercial possibilities as an antiseptic, bacteriostat, and germicide.

EXAMPLE 3

Method of preparation of 2-methacryloxyethyl gallate (III)

Gallic acid monohydrate (110 gm.; 0.585 mol, 2-hydroxyethyl methacrylate (296 ml.), and p-toluene sulfonic acid monohydrate (practical grade, 96%; 9.9 gm.) were stirred together and heated gradually to a maximum temperature of 150° C. until the theoretical amount of water of condensation was removed by distillation. The toluene-soluble portion of the product was washed repeatedly with water and with pentane. The filtered product was stripped of solvents using a vacuum and slight warming.

The resulting product was a viscous (but pourable), clear amber liquid with refractive index of $n_D^{24}=1.517$. Various methods were attempted to crystallize the product without success. As is indicated in Table 2, a 10% acetone solution of this product significantly increased bonding between a composite resin material and surfaces of dentin, compared to controls in which the acetone solvent only was applied to the dentin.

The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows:

1. A composite dental filling material comprising a metal oxide particulate material and a polymeric adhesive material prepared by the polymerization of a condensation product selected from 2-methacryloxyethyl vanillate, 2-methacryloxyethyl-p-hydroxybenzoate, and 2-methacryloxyethyl gallate in the presence of an amine-peroxide free-radical polymerization initiator.

2. The filling material of claim 1 wherein the reaction product utilized is 2-methacryloxyethyl vanillate.

3. The filling material of claim 1 wherein the polymerization initiator is a tertiary amine-peroxide.

4. The method of treating human teeth for restorative dentistry which consisting of applying as a cavity primer a solution of a condensation product selected from 2-methacryloxyethyl vanillate, 2-methacryloxyethyl-p-hydroxybenzoate, and 2-methacryloxyethyl gallate.

5. The method of claim 4 wherein the reaction product is 2-methacryloxyethyl vanillate.

6. The method of treating human teeth for restorative dentistry which comprises applying as a cavity liner a solution of a reaction product selected from 2-methacryloxyethyl vanillate, 2-methacryloxyethyl-p-hydroxybenzoate, and 2-methacryloxyethyl gallate, zinc oxide and an amine peroxide free radical polymerization initiator.

7. The method of claim 6 wherein the reaction product is 2-methacryloxyethyl vanillate and the free radical polymerization initiator comprises N,N-dimethyl-sym-m-xylylidine and benzoyl peroxide.

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