DENTAL ADHESIVE COMPOSITION

This application is a continuation of application Ser. No. 07/911,512, filed on Jul. 10, 1992, now abandoned.

BACKGROUND OF THE INVENTION

This invention relates to a dental adhesive composition for bonding acrylic resin to teeth and, more specifically, to a dental adhesive composition for restorative teeth and for bonding dental composite material (composite resin) to enamel and dentin. A bonding force with respect to dentin is particularly needed for restorative teeth.

With a conventional adhesive for bonding restorative composite resin to an enamel, a bonding strength higher than 100 kg/cm^2 that meets clinical requirements well enough is obtained between an enamel and the restorative composite resin. This is true even when an enamel is treated by acid etching using phosphoric or citric acid, then washed with water and dried, and finally coated with a primer which is composed of a methacrylic ester monomer and a curing agent and shows no adhesion to the dentin. However, as only poor adhesion to a dentin is obtained by using the primer having no adhesion to the dentin, various dentin treatment solutions or primers considered to be adhesive have been proposed. For instance, Japanese Patent Publication No. 55-30768 describes a phosphoric ester compound as being adhesive to the dentin, but it fails to give the aforesaid high bonding strength, as measured by the present inventors.

Japanese Patent Laid-Open No. 54-12338 discloses a functional monomer 4-methacryloxyethyl trimellitic anhydride (hereinafter 4META for short), and the "Journal of the Japan Society for Dental Apparatus and Materials", 23(61), pp. 29–32 (1982) teaches that when a dentin is treated with an aqueous solution of 10% citric acid and 3% ferric chloride and then restored with a restorative filler (4META-containing methyl methacrylate/tri-n-butyl borane/polymethyl methacrylate), a bonding strength of 12–18 Mpa is obtained. When measured by the present inventors, however, such a high bonding strength could not be obtained. Furthermore, when using a chemical polymerization type restorative composite resin made up of a redox initiating system of a tertiary amine-benzoyl peroxide, a problem will arise in connection with the reaction between the tertiary amine and 4META.

In view of the present state of the art where no clinically efficacious adhesive is obtained yet, as mentioned above, we have made an effort to achieve an adhesive which is not only improved in terms of its adhesion to a dentin but serves to reinforce a dentin as well, and so have accomplished the present invention.

SUMMARY OF THE INVENTION

As a result of strenuous studies made to provide a solution to the state-of-the-art problem mentioned above, we have now found that a dental adhesive composition containing a compound having a carbodiimide in its molecule has much higher bonding strength to a dentin than ever before. This would appear to be so because the protein component of a dentin is chemically combined with a methacrylic ester monomer having a hydroxyl or amino group due to their dehydration reaction which takes place under the catalytic action of the carbodiimide. In addition, the carbodiimide serves to crosslink dentinal protein, so that composite resin can be firmly bonded to a dentin by the dental adhesive composition containing a compound having a carbodiimide in its molecule.

DETAILED EXPLANATION OF THE INVENTION

In the ensuing description, this invention will be explained at great length.

The dental adhesive composition according to this invention is characterized by containing a compound having a carbodiimide in its molecule. Mentioned for this compound, for instance, cyclohexylcarbodiimide, 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide and 1-cyclohexyl-3-(2-morpholinoethyl)carbodiimide. These compounds should be used in an amount lying in the range of preferably 0.1 to 20.0% and more preferably 0.5 to 10.0% with respect to the total weight of the composition.

Any morphological limitation is not imposed on this dental adhesive composition, provided that it contains a compound having a carbodiimide in its molecule. For instance, it may be available in the form of a primer composition (which does not cure by itself) diluted with a solvent, a photo-polymerizable type composition, a chemically polymerizable type composition which is divided into two or more parts and a composite resin composition in which the monomer contains that compound. Preferable for this invention is a dental adhesive composition made up of:

a) a compound having a carbodiimide in its molecule,

b) a methacrylate or acrylate having at least one unsaturated double bond, and

c) a polymerization initiator.

For the methacrylate or acrylate b) which has at least one unsaturated double bond, use may be specifically made of methyl methacrylate, ethyl methacrylate, isopropyl methacrylate, hydroxyethyl methacrylate, n-butyl methacrylate, isobutyl methacrylate, hydroxypropyl methacrylate, tetrahydrofurfuryl methacrylate, glycidyl methacrylate, 2-methoxyethyl methacrylate, 2-ethylhexyl methacrylate, 2,2-bis(methacryloxyphenyl)propane, 2,2-bis[4-(2-hydroxy-3-methacryloxypropoxy)phenyl]propane, 2,2-bis(4-methacryloxydiethoxyphenyl)propane, 2,2-bis(4-methacryloxypropoxyphenyl)propane, ethylene glycol dimethacrylate, diethylene glycol dimethacrylate, triethylene glycol dimethacrylate, butylene glycol dimethacrylate, neopentyl glycol dimethacrylate, 1,3-butanediol methacrylate, 1,4-butanediol dimethacrylate, 1,6-hexanediol methacrylate, trimethylolpropane trimethacrylate, trimethylolethane trimethacrylate, pentaerythritol trimethacrylate, trimethylolmethane trimethacrylate and pentaerythritol tetramethacrylate and their acrylates as well as methacrylates and acrylates containing an urethane bond in their molecules. Note that particular preference is given to di-2-methacryloxyethyl-2,2,4-trimethylhexamethylene dicarbamate or its acrylate as well as compounds having the following structural formula: