METHOD AND COMPOSITION FOR PROMOTING IMPROVED ADHESION TO SUBSTRATES

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Background of the Invention

1. Field of the Invention

The present invention is directed to a composition for and methods of improving adhesion of conventional adhesives to substrates. More particularly, the present invention is directed to methods and compositions for the improvement of adhesive bonding of acrylic resins to substrates found in industrial, natural and dental environments, such as those involved in dental restorations and for protective sealants.

2. Description of Related Art

The ability to form strong, durable bonds between restorative or sealant materials and skeletal tissue, such as bone and tooth structure, has a number of important advantages. Effective adhesives for such purposes, particularly dental adhesives, provide a means of bonding that does not require the removal of healthy dental tissue, e.g., by eliminating the need for mechanical undercuts for retention of restorations. In addition, the use of dental adhesives can eliminate microleakage at the restorative-tooth interface, which, if unresolved, can lead to gap formation, margin discoloration, sensitivity, and the potential formation of secondary caries. In addition, dental adhesives improve the stress distribution at the tooth-restoration interface and can reinforce decay-weakened tooth structures. Adhesives are also capable of enhancing the bonding of protective sealant materials to tooth structure, especially to dental tissue that is not amenable to acid etching, a method which has been used in some situations heretofore but which has certain attendant detrimental effects. A wider application of conservative and preventive dental practices thus depends on the availability of effective dental adhesive systems.

A major shortcoming of polymer-based restoratives and sealants, as well as other types of materials, is their inherent inability to adhere to enamel and, particularly, dentin. Adequate adhesion to enamel, in most situations, can be achieved by acid etch techniques that employ aqueous solutions of inorganic or organic acids, such as phosphoric, nitric, citric, maleic acids, etc. These techniques enable dental resins to infiltrate the microporous surface of the etched enamel and form interlocking polymeric tags. Forming bonds of suitable strength to dentin, which is a vital, more complex, heterogeneous substrate than enamel, has only recently been achieved. As currently practiced, bonding to dentin involves a number of steps, usually including a dentin conditioner or etchant, that alters the surface of the dentin. This etchant has typically been an acidic solution similar to, but usually more dilute than, those employed with enamel. The cleansed dentin surface is then conditioned further with a surface-active compound referred to as a "primer", commonly in solution, and finally treated with an adhesion-promoting monomer (also commonly used in solution) that can diffuse and polymerize in the conditioned dentin to form a resin-impregnated interface or hybrid layer.

As indicated above, currently used dentin bonding systems generally include 3 basic components: (1) a dentin conditioner, (2) a dentin primer, and (3) a resin-based bonding agent or polymer-forming monomer(s). Dentin conditioners are generally aqueous solutions of inorganic or organic acidic agents, such as phosphoric, nitric, citric, maleic acids, etc. Some conditioners employ chelating compounds, based on ethylenediaminetetraacetic acid (EDTA), which are also applied to dentin in the form of aqueous solutions. The function of the dentin conditioner is to remove or alter what is known as the "smear layer", a coating of debris that forms on the dentin surface as a result of the cutting and grinding processes of cavity preparation. Application of the dentin conditioner is regarded as a critical step in establishing effective bonding to dentin as this makes it possible for the other components of the dentin adhesive system to wet, infiltrate or penetrate, and conform to the heterogeneous, vital dentinal substrate.

Primers are surface-active compounds that exhibit both an affinity for dentin and adhesive resin systems and participate in the polymerization process, thereby promoting adhesion between the primarily hydrophilic dentin and the predominantly hydrophobic polymeric adhesives or monomers from which they are formed. Primers are applied to dentin in solution form, such as acetone, ethanol, monomeric solutions, water and various mixed solvent systems. A widely used primer is N-phenylglycine (NPG), which, in addition to its surface-active properties, also functions as a co-initiator or activator during interfacial polymerization.

The bonding resins or monomers used to form the bonding adhesives are generally simple acrylic monomers such as methyl methacrylate, 2,2-bis[p-(2'-hydroxy-3'-methacryloxypropoxy)phenyl]propane (Bis-GMA), triethylene glycol dimethacrylate, 2hydroxyethyl methacrylate, etc., as well as various monomeric combinations (monomer systems). Especially effective monomeric bonding agents are those that have surface-active functional groups, e.g., carboxylic acid, carboxylic acid anhydride, phosphate, sulfonate, sulfinate, aldehyde, isocyanate, hydroxyl, amide, etc. A particularly effective surface-active monomer is the reaction product of 2-hydroxyethyl methacrylate and pyromellitic acid dianhydride (PMDM). In addition to the above components, dentin adhesive systems incorporate chemical, photochemical and dual-curing free-radical initiators. Bonding systems which have proven fairly useful in recent years include a number described by Rafael L. Bowen in several of his U.S. Pat. Nos. : 4,514,527; 4,521,550; 4,588,756; 4,659,751 and 5,270,351. These patents disclose a bonding procedure for achieving adhesion to tooth structures and other substrates that involve the use of an acid, such as nitric acid, for removing the smear layer, the use of an N-aryl-α-amino acid such as N-phenylglycine to prime the surface and the application of a surface-active monomer such as PMDM.

Some of these bonding systems are characterized by the ability to undergo a self-initiated interfacial polymerization and are especially effective for bonding to dentin. For example, the sequential application of a dilute aqueous nitric acid solution as the etchant, N-phenylglycine (NPG) in acetone as the primer, and an acetone solution of a carboxylic acid-containing monomer, PMDM, has been shown to yield strong composite-to-dentin bonding. In this system, effective interfacial free-radical polymerization is initiated by the interaction of PMDM with the infused NPG. A number of other N-aryl-α-amino acids have been used as primers in place of NPG, and a variety of carboxylic acid monomers have been substituted for PMDM in this bonding system in attempts to further enhance the efficiency of this type of bonding system. The procedures described, however, require the use of separate etchant and primer steps to prepare and predispose the dentin surface for bonding to resin-based dental materials.

Although current dentin adhesive systems such as the aforementioned, show considerable improvement over their