DENTAL RESTORATIVE MATERIAL

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to dental restorative materials, and more specifically to novel dental restorative materials, which are polymer composites containing an ultrafine inorganic filler in a high density. The term "dental restorative material" as used herein means a composite material for dental use, which is generally called "dental composite resin" and is used as a material for filling and restoring teeth, material for making inlays, artificial crowns, artificial teeth, abutment construction materials, or the like.

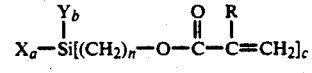
2. Description of Prior Art

Recently, in the field of dental care, compositions containing an inorganic filler and a monomer, for example composite resins for dental use, have come into use. It is known that the inorganic fillers used in composite resins for dental use will exert various desirable effects for dental care when the range of their particle sizes are properly selected. More concretely, while granular fillers having a size of not more than 20 μm had been used in the past, finer fillers having a particle size of not more than 0.1 μm have more frequently been used these days, as shown in Japanese patent Application Laid-open Nos. 107189/1979, 82303/1982, 101409/1984 and 148109/1986 and Japanese patent Kohyo Nos. 500150/1982 and 501090/1986. The use of such fine fillers is said to contribute to an increase of mechanical strength, an improvement in polishability (gloss development when polished) and abrasion resistance, and this technology therefore is becoming of high importance in the dental fields.

The inorganic fillers used in composite resins for dental use are generally subjected to preliminary surface treatment. The surface treatment improves the wettability at the filler-monomer interface, improves the dispersibility of the filler in the composition and makes it possible to increase the filler content. As a result, composite resin moldings obtained by polymerization of the monomer have improved mechanical strength owing to good adhesion at the filler-resin interface. Known in the art as surface treating agents for such purposes are silane coupling agents, typically γ-methacryloyloxypropyltrimethoxysilane.

On the other hand, for the purpose of increasing adhesiveness of the organic-inorganic interface, the use of surface-treating silane couling agents other than the above-mentioned γ-methacryloyloxypropyltrimethoxysilane, having a longer alkylene group as a spacer between the methacryloyl group and the silyl group have to some extent been attempted; for example:

(1) Japanese patent publication No. 20871/1969 discloses a composition incorporating an inorganic filler surface-treated with a compound represented by the general formula



wherein R is a hydrogen atom or an alkyl group, X is a hydrolyzable group, y is a hydrocarbyl group, n is an integer of 7 to about 20, a is an integer of 1 to 3, b is an integer of 0 to 2, c is an integer of 1 to 3, and a+b+c=4.

However, the above composition differs from the dental restorative material of the present invention in that it utilizes monofunctional monomers as the principal monomer and that it uses only a coarse inorganic filler having a particle size of at least 0.2 μm. Furthermore, the specification has no description anticipating its application to dental care field.

(2) J. Jang, H. Ishida and E. P. Plueddeman, Proc. 41st Annual Conference, Reinforced Plastics/Composite Institute, The Society of the Plastics Industry, Session 2-C (1986) describes 11-methacryloyloxyundecyltrimethoxysilane. The reference discloses a polymer composite comprising a filler surface-treated with the above silane compound and a thermoplastic polyester resin. It however does not suggest a (meth)acrylate monomer as a matrix resin. Besides, the filler is surface-treated in a way not effective for the afore-mentioned silane coupling agent, being different from the way of the present invention. And further, the paper contains no description anticipating the use in dental field.

(3) Nishiyama, The Journal of the Japanese Society for Dental Materials and Devices, 3 (2), 284 to 294 (1984) shows 10-methacryloyloxydecyltrimethoxysilane. However, in this paper, while a glass plate is surface-treated with the compound and the state of the treated surface is compared with that treated with conventional γ-methacryloyloxypropyltrimethoxysilane, the surface-treating effect for fine inorganic fillers is not studied. Furthermore, although the paper states, as a general comment, that 10-methacryloyloxydecyltrimethoxysilane can be used as a surface-treating agent for fillers, it contains no concrete description at all on how to use the compound as a surface-treating agent for dental composite resins or on the make-up of composite resins.

(4) Japanese patent Application laid-open No. 159214/1988 discloses a surface-treating process of a particulate silica-based oxide complex, which comprises preliminarily treating said complex with an aliphatic amine and then treating the same with a silane coupling agent. κ-methacryloyloxydecyltrimethoxysilane is used as an example of such coupling agents. The technical point of this process is, since it uses the particulate complex having a high acidity, pKa of −3.0 to 3.0 on the surface sites, to treat with a silane coupling agent the particulate complex after its neutralization with an amine. Accordingly, there is no thought to set up an object of incorporating a large amount of a fine filler having a size of 0.1 μm or less into a monomer and to find out a silane coupling agent suited therefor. The afore-mentioned fine fillers, particularly those having a size of 0.1 μm or less, can be dispersed in a monomer in a far smaller amount than coarser fillers since the fine fillers cause a markedly large viscosity increase. As a result, the advantage of incorporating an ultrafine filler having a size of 0.1 μm or less, such as improvement in mechanical strength, abrasion resistance and polishability, has not fully been realized.

There has therefore been desired a technique of incorporating an ultrafine filler in a high density into a monomer, i.e. a resin matrix. Also has been desired a technique for improving adhesion of filler-matrix resin interface, which governs the durability of the dental restorative materials.