DENTAL FILLING MATERIAL

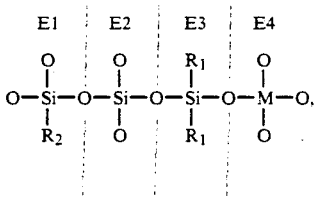
The present invention relates to a novel dental filling material which contains a specific filler.

Dental restorative materials based on polymerizable compounds, so-called composites, contain obligatorily a mineral filler in addition to one or more polymerizable monomers, especially (meth)acrylic acid esters, activators, optionally polymerization catalysts and other components.

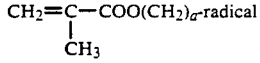
Depending on the type and amount, this filler determines the physical properties of the filling made using the composite. The greater the filler content and the larger the particle size, the better it is for the physical properties but usually the worse for the polishability of the filling.

A need therefore arose to develop dental filling materials which not only have good physical properties but can be also satisfactorily polished.

This object is achieved in that dental filling materials based on otherwise customary components, especially at least one polymerizable (methy)acrylic acid ester, contain 20-90 percent by weight, calculated on the total composition, of a compound consisting of the structural element E2 and at least one of the structural elements E1 and/or E3 and/or E4 of the general formula



where R_1 denotes a methyl, ethyl, n-propyl, isopropyl or an unsubstituted or CH_3—C_3H_7-substituted phenyl radical, R_2 denotes a CH_2=CH—, CH_2=CHCOO(CH-2)— or



or R_1, n denotes 0, 1, 2 or 3, and M denotes titanium or zirconium.

By using these organically modified silica compounds as fillers not only is outstanding polishability of the composites achieved, but also the physical properties, especially the mechanical strength and abrasion resistance of the fillings, are very substantially improved.

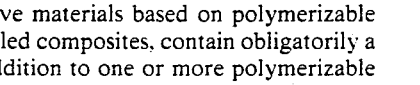
The structural unit E2 of the above general formula is present in combination with at least one of the structural units E1, E3 or E4, in which case the preferred molar ratio of E2 to the other structural elements is between 50:1 and 10:1, preferably between 30:1 and 20:1, particularly about 25:1.

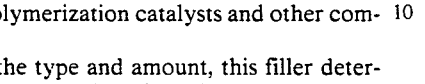
If the compound consists of more than two structural elements, the ratio of E2 to E1and E3 likewise is preferably between 50:1:1 and 10:1:1, particularly 30:1:1 and 20:1:1; the same is of course also true with regard to the combination E2/E1/E4 or E2/E3/E4.

If all the structural elements of the general formula are present together, then the molar ratio of the structural elements E2:E1:E3:E4 is between 50:1:1:1 and 10:1:1:1, preferably between 30:1:1:1 and 20:1:1:1, particularly about 25:1:1:1.

Examples of suitable compounds are the following:

E2:E1 = 50:1





or

E2:E3 = 25:1
R_1 = CH_3, from which follows:
$(CH_3)_2$—$SiO_{2/2}$.25SiO_2
or
E2:E3:E4 = 25:1:1
R_1 = CH_3
M = Zr, from which follows:
$(CH_3)_2SiO_2$.ZrO_2.25SiO_2

These inorganic-organic polymers are basically known from the prior art and are designated inter alia as "ORMOCER" or "ORMOSIL".

They are described, for example, in the 1987 activities report of the Fraunhofer-Institut für Silikatforschung, Würzburg, pp. 48-74, and in a survey published in "Bild der Wissenschaft" No. 11/1987, p. 29.

These polymers are prepared by the sol-gel process in the presence of an acid or basic catalyst in alcoholic or aqueous alcoholic solution at about 25° to about 300° C. by reacting a tetraalkoxysilane, for example tetraethoxysilane, with (meth)acryloxypropyltrimethoxysilane and, if desired, a tetraalkoxyzirconium or tetraalkoxytitanium and, if desired, dialkyldialkoxysilane.

It is necessary to keep the amount of hydroxysilane groups as low as possible, which can be achieved by adjusting the corresponding pH.

The halides of the corresponding silanes may be also expediently employed as starting products. The resultant reaction product is separated from the reaction solution, dried at about 100° C. to about 500° C. and ground.

Should the amount of SiOH groups be undesirably high, these can be completely removed by a basic postcondensation or by additional silylation using, for example, (meth)acryloxypropyltrimethoxysilane.

The preparation of the ORMOCERs may be generally expressed as follows: reaction of an alkoxysilane $Si(OR_1)_4$ with an alkoxysilane R_2—$Si(OR_1)_3$ and/or an alkoxysilane $(R_1)_2$—$Si(OR_1)_2$ and/or a metal ester $M(OR_1)_4$, R_1, R_2 and M having the meaning defined above.

The surface of the ORMOCERs used according to the invention as fillers is between about 10 and about 50 m^2/g, particularly 20-30 m^2/g.

The ORMOCER fillers used according to the invention may be the only filler comprised in the dental filling materials, but it appears expedient to combine these with other fillers known per se.

The total filler content in the dental filling materials according to the invention is between about 55 and not more than 90 percent by weight based on the total composition, preferably between about 65 and about 85 percent by weight.

Suitable fillers to be used in combination with the ORMOCERs and known per se, are preferably silylated