

TABLE 4-continued

	Example 21	Example 22	Comparative Example 18	Comparative Example 19	Comparative Example 20
Brinell hardness	92	81	66	68	73

(1) 11-methacryloyloxyundecyltrimethoxysilane

(2) γ -methacryloyloxypropyltrimethoxysilane

EXAMPLE 23

A mixture containing 50 parts by weight of an ultrafine γ -alumina having an average particle size of 0.02 μm (Aluminum Oxide C®, available from Nippon Aerosil Co.), 10 parts by weight of 11-methacryloyloxyundecyltrimethoxysilane and 500 ml of toluene was refluxed for 2 hours under heating with vigorous stirring and then allowed to cool. Toluene was first distilled off in vacuo, and the residual mixture was dried in vacuo for 12 hours and then heated at 90° C. for 2 hours in vacuo to fully distill off toluene, to give a surface-treated ultrafine alumina filler. Next, an amount of lanthanum glass powder (GM 3168®, available from Shott) was pulverized and sieved to give a powder having an average particle size of 2.8 μm and a particle size range of 0.1 to 10 μm . Surface treatment was conducted on 100 parts by weight of the thus obtained powder with 2 parts by weight of 11-methacryloyloxyundecyltrimethoxysilane in the same manner as for the borosilicate glass in Example 21.

A monomer composition was prepared by mixing together 30 parts by weight of D-2.6E, 35 parts by weight of U-4TH, 35 parts by weight of DD and 0.5 part by weight of 2,4,6-trimethylbenzoyldiphenylphosphine oxide.

Using these fillers and monomer composition thus obtained, a polymerizable composition shown in Table 5 was prepared and evaluated in the same manner as in Example 21. The results are shown in Table 5.

COMPARATIVE EXAMPLE 21

Surface treatment was conducted on 50 parts by weight of the γ -alumina powder of Example 23 with 10 parts by weight of γ -methacryloyloxypropyltrimethoxysilane. This filler was then used to obtain a polymerizable composition of the same filler/monomer ratio as that in Example 23. However, all of the filler could not be kneaded into the monomer composition because of increase in viscosity.

COMPARATIVE EXAMPLE 22

A polymerizable composition with components and the filler/monomer ratio shown in Table 5 was prepared using the surface-treated alumina filler obtained in Comparative Example 21 and evaluated in the same manner as in Example 23. The results are also shown in Table 5.

TABLE 5

	Example 23	Comparative Example 22
<u>Monomer</u>		
Composition	D-2.6E/ U-4TH/DD = 30/35/35	D-2.6E/ U-4TH/DD = 30/35/35
Parts by weight	100	100
<u>Ultrafine filler</u>		
Surface treating agent	11-MUS	γ -MPS
Parts by weight	200	150
<u>Glass filler</u>		
Surface treating agent	11-MUS	11-MUS

TABLE 5-continued

	Example 23	Comparative Example 22
Parts by weight	800	350
Paste consistency (mm)	20.5	20.6
Compressive strength (kg/cm ²)	5920	4820
Flexural strength (kg/cm ²)	1680	1480
Brinell hardness	92	66

EXAMPLE 24

Test specimens for flexural strength were prepared by using the polymerizable composition obtained in Example 21 and by the same curing method as used in Example 21. The specimens were, for the purpose of accelerating deterioration, kept immersed in water at 70° C., and measured for flexural strength after one day, 10 days, 30 days and 100 days of immersion. The results are shown in Table 6.

COMPARATIVE EXAMPLE 23

A monomer composition was prepared by mixing and dissolving 65 parts by weight of Bis-GMA, 35 parts by weight of 3G and 0.5 part by weight of 2,4,6-trimethylbenzoyldiphenylphosphine oxide.

A polymerizable composition was prepared by kneading 100 parts by weight of the monomer composition obtained above, 150 parts by weight of the OX-50 surface-treated with γ -MPS and 300 parts by weight of borosilicate glass surface-treated with γ -MPS, the latter two having been obtained in Comparative Example 18. The composition was evaluated in the same manner as in Example 24. The results are shown in Table 6.

TABLE 6

	Flexural strength (kg/cm ²)				
	Initial	immersed in water at 70° C. for:			
		1 day	10 days	30 days	100 days
Example 24	1,770	1,680	1,670	1,640	1,620
Comparative Example 23	1,450	1,330	1,280	1,150	1,020

It is clear from Table 6 that the restorative material of the present invention maintains its high strength over a long period of time in water at 70° C., and is thus excellent in water resistance.

In the Examples above, the definitions and measuring methods of various characteristics are as follows.

(i) Average particle size and particle size range

For the ultrafine powders with a particle size of 0.1 μm or less, the particle size was determined based on transmission electron photomicrograph or by conversion from the specific surface area determined by BET method. For the fillers with a particle size exceeding 0.1 μm , a Horiba model CAPA 500 particle size autoanalyzer was used. The measurement was made with the centrifugal and gravitational sedimentation and light transmission technique.

(ii) Paste consistency