Light Scattering and Gloss of an Experimental Quartz-filled Composite

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For samples of polymethylmethacrylate with and without quartz filler, the inverse of the contrast-gloss ratio is shown to be related to surface roughness and to the optical scattering coefficient. This finding adds to the importance of optical scattering, which has been widely studied because of its relation to color and translucency of materials. Furthermore, optical scattering by composite fillers is shown to be linearly related to the concentration of the filler material within the range of concentrations studied. Quartz fillers were incorporated at concentrations from 5 to 20 weight percent and were short fibers or granular powder, with the granular particles ranging in median equivalent spherical diameter from 15 to 3.3 μm. The efficiency of optical scattering for the granular quartz filler increased as the size of the filler decreased.

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Introduction.

The importance of surface gloss among the appearance properties of esthetic dental materials has been described by O'Brien *et al.* (1984). That investigation reported a significant relationship between the contrast-gloss ratio and the surface roughness of commercially available composite materials. However, no significant difference in contrast gloss was found among the materials studied of differing filler type and size. Since the contrast-gloss ratio is the ratio of light reflected specularly at a material surface to that diffusely reflected normal to the surface, the contrast gloss can be expected to be related to any optical scattering which occurs at the surface or within the body of an esthetic restorative material.

Optical scattering has been widely studied because of its important effects on the color and translucency of materials in general (Judd *et al.*, 1937; Kubelka, 1948), of paint films (Billmeyer and Abrams, 1973; Billmeyer and Phillips, 1973), and of dental materials (Judd *et al.*, 1937; Cook and McAree, 1985). Clewell (1941) has theorized that optical scattering is dependent upon the wavelength of the illumination for the case where the size of the scattering particle is approximately the wavelength of the illumination. This has been experimentally confirmed by Patrick (1951) for porcelain enamels in which crystals of titanium oxide have opacified the enamel by scattering. He found that optimum opacification occurs at a particle size of 0.4 times the wavelength of the scattered light.

However, when the size of the scattering particle is much greater than the wavelength of the illumination, Clewell's (1941) theory predicts that the scattering is inversely proportional to the size of the particle, and that wavelength has no effect. Filler particles utilized in some available composites have been shown by Ruyter and Øysæd (1982) to range from one to 50 μm. The uniform scattering coefficient for a dental composite material over the visible spectrum reported by Cook and McAree (1985) demonstrates that the effect of wavelength is of negligible concern with respect to scattering in these materials.

Commercial composite materials contain about 50 volume percent (75 weight percent) filler (Craig, 1985), and the filler can be expected to have a substantial effect on the optical properties of the composite. The similarity in optical phenomena involved in high-scattering and low-contrast gloss would imply a relationship between these two properties. The purposes of this investigation were to determine the effects of concentration of quartz filler of differing size and shape on the optical scattering and contrast gloss of a composite resin and to determine any relationship between the contrast gloss and the optical scattering and surface roughness of this material.

Materials and methods.

The composite specimens were composed of polymethylmethacrylate (PMMA) powder polymer and liquid monomer (Hygienic Perm Reline and Repair Resin, Shade Clear, Hygienic Corporation, Akron, OH 44310) to which quartz filler was added. The quartz filler was either short fibers or granular powder. The granular quartz (Quartz particles, 3M Corporation, Minneapolis, MN 55144) was incorporated in the resin as received, and also after automated grinding (Fisher Mortar Grinder model 155, Fisher Scientific, Pittsburgh, PA 15219) for 40 min, 3.3 hours, and 14 hours. The equivalent spherical particle size distribution was determined for each of the above four conditions of the granular quartz by a particle size analyzer (Sedigraph 5000D, Micromeritics Instrument Corp., Norcross, GA 30093). The fibrous quartz (Fibrous quartz, Astraquartz, Inc., New York, NY 11210) was hand-ground and -sieved to reduce the fiber length, and the size distribution was determined using photomicroscopy. The five fillers were incorporated at weight concentrations of 5, 10, 15, and 20 percent, with a sample replicate size of three. In addition, three specimens were formed of only PMMA. All specimens were formed in a mold of 25.4 mm diameter and 3.2 mm thickness. The specimens were finished by wet-sanding to 600-grit silicon carbide paper, by means of a standard metallographic polishing technique.

The scattering coefficient was determined at a wavelength of 550 nm for each specimen, with reflectance spectroscopy (Spectronic 20 spectrophotometer with reflectance attachment, Bausch and Lomb, Inc., Rochester, NY 14625) used as described by Woolsey *et al.* (1984). The arithmetic average roughness was determined (Surfanalyzer, Clevite Corp., Gauging and Control Div., El Monte, CA 91731) using a traverse speed of 0.5 mm/sec. A goniophotometer (Differential I, Science Spectrum, Santa Barbara, CA) was used to determine the contrast gloss, with incident tungsten light used at an angle of 30° to the specimen surface. The methods for roughness and gloss determination were as described by O'Brien *et al.* (1984).

The scattering coefficients were analyzed by analysis of covariance (Draper and Smith, 1966; Sokal and Rohlf, 1969, pp. 448–458). In this case, analysis of covariance is a method of determining the simple linear regression of scattering coeffi-

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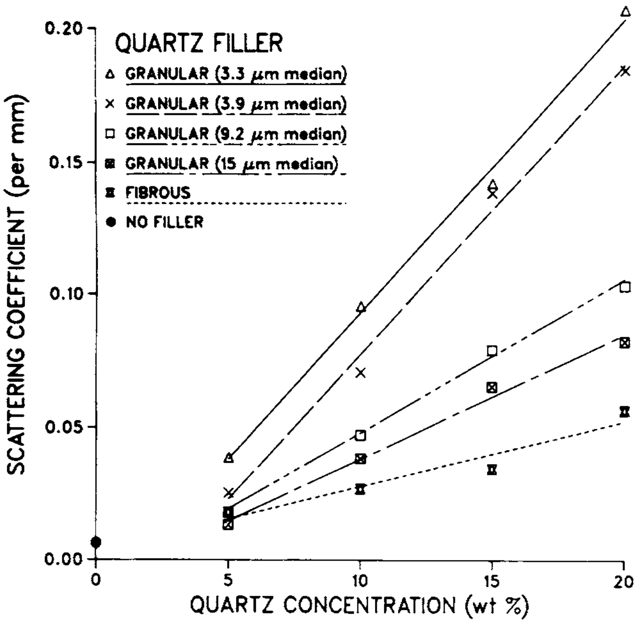

Fig. 1 — The mean scattering coefficients of the composites and of unfilled PMMA. The lines show the linear regression of the scattering coefficient to filler concentration for each filler type.

TABLE 1
STATISTICAL SUMMARY AND ANALYSIS OF OPTICAL SCATTERING COEFFICIENT (mm^{-1})

Regression Analyses by Filler

		Granular (Median Particle Size in μm)			
	Fibrous	15	9.2	3.9	3.3
Regression Constant (mm^{-1})	0.0032	−0.0086	−0.0090	−0.0313	−0.0165
Regression Coefficient (mm^{-1}/wt %)	0.0025	0.0047#	0.0057	0.0109	0.0110

Analysis of Covariance

Source of Variation	df	Mean Squares	F-ratio	Significance
Covariate	1	0.09011	190.58	<0.0005
Between Filler Means	4	0.01634	34.56	<0.0005
Overall Error	54	0.00047		
Overall Regression	1			
Equal Regressions	8	0.01092	152.75	<0.0005
Equal Slopes	4	0.00549	76.83	<0.0005
Error (Each Regression)	50	0.00007		
Total	59			

Underlines connect regression coefficients which are not significantly different at α = 0.05. All other regression coefficients are significantly different.

cients on filler concentration for each of the five quartz fillers, and then identifying any significant differences among the regression coefficients. The regression coefficient is the slope of the regression line and, for this study, is a measure of the increase in scattering due to an incremental addition of the filler. The roughness and contrast gloss were analyzed by two-way analyses of variance (Sokal and Rohlf, 1969, pp. 299–320). In addition, a relationship between the inverse of the contrast gloss and the roughness and scattering coefficient was obtained by means of multiple linear regression (Draper and Smith, 1966).

TABLE 2
STATISTICAL SUMMARY AND ANALYSIS OF ARITHMETIC AVERAGE ROUGHNESS (μm)

Sample Means (n = 3)
Quartz Filler

Concentration (%)	Fibrous	Granular (Median Particle Size in μm) 15	9.2	3.9	3.3
5	0.137	0.037	0.047	0.053	0.050
10	0.207	0.051	0.070	0.053	0.048
15	0.247	0.051	0.079	0.056	0.069
20	0.347	0.055	0.104	0.073	0.075

Analysis of Variance

Source of Variation	df	Mean Squares	F-ratio	Significance
Among Subgroups	19	0.0195	35.02	<0.00005
Among Quartz Fillers	4	0.0733	131.60	<0.00005
Among Concentrations	3	0.0116	20.84	<0.00005
Interaction	12	0.0004	6.37	<0.00005
Error	40	0.000056		
Total	59			

TABLE 3
MEANS AND ANALYSIS OF VARIANCE FOR CONTRAST GLOSS RATIO

Sample Means (n = 3)
Quartz Filler

Concentration (%)	Fibrous	Granular (Median Particle Size in μm) 15	9.2	3.9	3.3
5	240	12	151	136	105
10	161	81	79	80	70
15	108	66	68	50	53
20	81	76	41	41	28

Analysis of Variance

Source of Variation	df	Mean Squares	F-ratio	Significance
Among Subgroups	19	7616.2	14.32	<0.00005
Among Quartz Fillers	4	12635.5	23.75	<0.00005
Among Concentrations	3	26431.7	49.69	<0.00005
Interaction	12	1239.3	2.33	<0.025
Error	40	531.9		
Total	59			

TABLE 4
ANALYSIS OF VARIANCE FOR REGRESSION OF INVERSE OF CONTRAST GLOSS RATIO ON AVERAGE ROUGHNESS AND SCATTERING COEFFICIENT

Source of Variation	df	Mean Squares	F-ratio	Significance
Regression	2	0.000516	62.23	<0.00005
Error	18	0.0000083		
Total	20			

Results.

The median equivalent spherical diameters of the granular quartz were found to be 15, 9.2, 3.9, and 3.3 μm, respectively, for the four conditions of as-received samples, ground for 40 min, 3.3 hours, and 14 hours. The median fiber length was found to be 60 μm for fibers of uniform diameter (10 μm). The mean scattering coefficients of the composites and of unfilled PMMA are shown in Fig. 1, which also displays the regression lines for the fillers studied. The regression coefficients and constants and the analysis of covariance of the scattering coefficients are given in Table 1, with non-signifi-

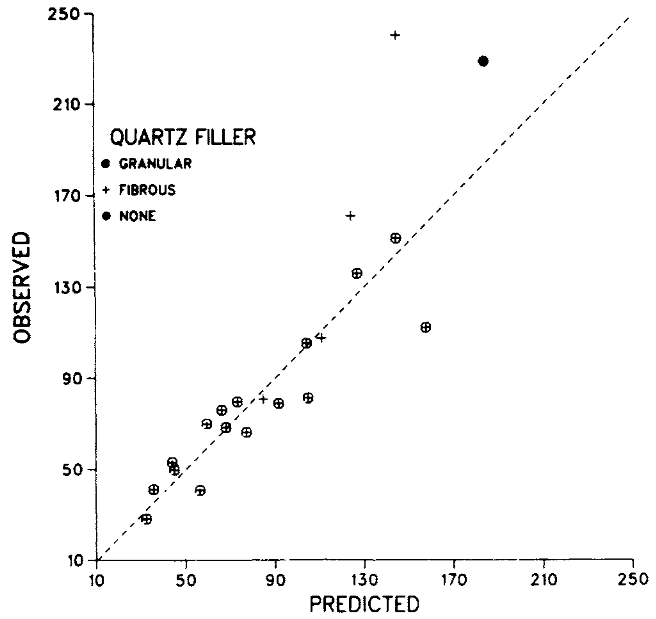

Fig. 2 — The agreement between the observed contrast gloss ratio and that predicted by linear regression to scattering coefficient and roughness.

cantly different regression coefficients marked by an underline. The means and the analyses of variance of the roughness and gloss parameters are given in Tables 2 and 3, respectively, for the filled samples. The unfilled PMMA sample had a mean contrast gloss of 229 and a mean roughness of 0.07 μm.

The regression of the inverse of mean surface gloss to the means of the scattering coefficient and roughness yielded:

$$\frac{1}{G} = 0.0047 + 0.13\ S - 0.00012\ R$$

where G is the contrast gloss ratio, R is the roughness in μm, and S is the scattering coefficient in mm^{-1}. The analysis of variance is shown in Table 4, and the agreement of the predicted and observed sample means is diagrammed in Fig. 2. For this diagram, there are 18 degrees of freedom, since the three constants of the regression equation were determined from 21 sample means.

Discussion.

The scattering coefficient has been known for its importance in determining the color and translucency of materials and is now shown to be related to surface gloss for translucent composite materials. The relationship of the inverse of surface gloss to surface roughness and scattering coefficient is due to the increased diffuse reflection of a rougher surface, or of a more-highly-scattering material, or of both.

The second major finding of this investigation is the linear relationship between the optical scattering coefficient and the filler concentration, at least within the limits of the range of concentrations studied. The difficulty in studying the scattering coefficient of opaque materials has already been established (Woolsey *et al.*, 1984). At high filler concentration, the scattering coefficient can be expected to reach a plateau value, as supported by the absence of significant differences among the contrast glosses of commercially available composite materials (O'Brien *et al.*, 1984). The linear relationship of the scattering coefficient of quartz-filled PMMA to the concentration of the quartz filler is useful not only in predicting this important optical property of composite materials at the filler concentrations studied in this investigation, but also in determining the efficiency with which various fillers scatter light within the composite material. The greater efficiency for scattering by the smaller particles studied supports the theory of Clewell (1941) and the conclusions of Patrick (1951) and Ruyter and Øysæd (1982) with regard to particle size for optimum scattering.

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