

The Impact of Composite Structure on Its Elastic Response

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The non-destructive determination of Young's modulus of dental composites by means of the fundamental period was found to be reliable and accurate. Post-polymerization effects could clearly be detected. Exponential regression analysis showed a correlation coefficient of 0.92, after logarithmic transformation, with volumetric filler content.

The high accuracy and reliability of the measurements themselves are reflected in low standard deviations. The results are in excellent agreement with those of other investigations. Furthermore, the ease and speed of operation make this new procedure a powerful laboratory tool for material-testing and practical large-scale investigations.

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Introduction.

The need for testing materials in general and newly developed dental composites in particular before they are used in clinical practice is obvious. Up to the present, however, no foolproof laboratory test exists that can predict the clinical behavior of a material.

One quality of a material that could reasonably be assumed to be relevant to clinical behavior is its rigidity. Static, destructive test methods have been used to determine the rigidity and other mechanical properties of dental composites, but problems are encountered in assessment and comparison of the results, because of the elastic behavior of these composites under load (Nakayama *et al.*, 1974; Papadogianis *et al.*, 1984).

Young's modulus, a measure of rigidity, has several advantages over other parameters, since it is a function of the whole sample rather than of one weak part or of only the surface. Among the methods of determining Young's modulus non-destructively or dynamically are the ultrasonic pulse method and the torsion pendulum method (see Nakayama *et al.*, 1974; Greener *et al.*, 1979; and Whiting and Jacobsen, 1980b, for the former, and Finger, 1975, and Whiting and Jacobsen, 1980a, for the latter).

One problem with the ultrasonic method is the difficulty of measuring signal displacement in attenuated samples; another arises from changes in specimen geometry during water immersion. The torsion pendulum method is complicated by the problem of measuring low-level deformation.

A method of determining Young's modulus of solids using mechanical resonance frequencies has been perfected by Spinner and Tefft (1961). The theory of free vibrations of solids requires the use of the equations of vibratory motion and the accompanying stress or displacement boundary conditions. For the moment, only approximations can be obtained, although refinements in measuring techniques have made it possible to verify their validity. This dynamic resonance method has, therefore, become a standard technique for determining the elastic constants of solids (Schreiber *et al.*, 1973). The main drawback of this method is that the test equipment, and par-

ticularly the contact of the driver and the pick-up with the sample, interferes with the resonance frequency.

A new testing device (Grindo-Sonic®, Lemmens Elektronika, Haasrode, Belgium) has recently been developed to determine the fundamental period of a vibrating specimen without interfering with the oscillation. It was the aim of the present paper to see whether this method could be used on dental composites, and whether the Young's modulus so calculated from the fundamental period can make a useful contribution to the evaluation of composite restorative materials.

Materials and methods.

Table 1 shows the dental restorative composites included in this study. Ten rectangular samples of each product, 35 mm × 5 mm × 1.5 mm, were polymerized in a dismountable brass mold. These dimensions were selected on the recommendation of Spinner and Tefft (1961). All handling and testing were done at room temperature. The mixing and the placing of the self-cured composites were done within three minutes. The mold was then covered with a glass plate and held under firm finger pressure for five minutes. The samples were released from the mold ten minutes after mixing. Light-cured composites were inserted into the mold with minimal environmental light. A device with four light windows (Luxor® Activating Unit, ICI) was placed on a glass plate covering the composite, whereupon the sample was exposed for 60 seconds on the upper side through the glass plate and, upon removal, for an additional 60 seconds on the back side. All samples were finished with dry, 600-grit abrasive paper.

The actual test procedure was as follows: Flexural vibration was used to determine the fundamental period of a sample, because it is easier to excite flexural vibration than longitudinal vibration, especially in thin specimens. The sample was set in flexural transient vibration by a single shock excitation, thus creating standing waves in the sample. For flexural oscillation, there are two nodal points of the standing waves situated at about 0.224ℓ from the ends of the sample for the fundamental mode (Schreiber *et al.*, 1973).

Free vibration was obtained by placing the sample on triangular supports at these nodal points, where minimal displacement occurs (Fig. 1). The supported sample was then gently tapped once at its center by a small metal hammer activated by an electromagnet. The energy gain in the sample dissipates in oscillations. These vibrations are initially very complex and depend on the nature and energy of the stroke as well as on the characteristics of the tested material. However, such a free oscillating body tends to reach its fundamental vibration mode quickly, regardless of the initial complexity of its harmonics. This vibration was picked up by a microphone, so the experiment had to be conducted in an anechoic test chamber. Note that no part of the measuring equipment touched the sample. The signal was then fed into the special signal analyzer, the Grindo-Sonic®. The time of sampling was triggered by a drop in amplitude and was automatically delayed in order to allow the interfering overtones to die out. The

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TABLE 1
PRODUCTS, TYPE OF INITIATION (S IS SELF-CURED; L IS LIGHT-CURED), BATCH NUMBERS, AND MANUFACTURERS OF THE BRANDS INVESTIGATED

Product	S/L	Batch Number	Manufacturer
P-10	S	112983	3M Co, St. Paul, MN
P-30	L	Exp. Lot 5	
Concise	S	1994A + 1994B	
Silux U	L	041183 5502 U 4Y3	
Silar	S	8601A + 8601B	
Occlusin	L	Lot SP06 Mar 84	ICI Plc, Macclesfield, Great Britain
Estilux posterior XR1	L	061984 034	Kulzer & Co GmbH, Bad Hamburg, W. Germany
Estilux posterior Y	L	061984 182	
Estic microfill U	S	0684 045P + 131C	
Durafill U	L	061984 139	
Adaptic	S	053183 3A001	Johnson & Johnson, East Windsor, NJ
Miradapt	S	3D906 24051904	
Adaptic radiopaque	S	840514 CHB4135 + CHB4135/1	
Aurafill	L	L306159	
Answer	S	201804 21300	
Certain	L	021784P 3L1604	
Clearfil Posterior New Bond (=NB)	S	11127 PPU-2206 + CPU-2106	Keur & Sneltsjes Dental Mfg Co, The Netherlands
Clearfil	S	43005 BFXC-0204 + CFXC-0104	
Nimetic	S	0014 L157	Espe Dental Products, Lynbrook, NY
Nimetic-Dispers	S	L139 009 + 012	
Epolite 100	S	081131 CO21231 + EO81131	GC Dental Industrial Corporation, Japan
Microrest AP	S	230241	
Brilliant	S	150584-36	Coltène AG, Switzerland
Brilliant Lux U	L	D3 120684-20	
Biogloss	S	840522	De Trey AG, Zurich
Command Ultrafine	L	1 841286 BS U 30344	Kerr Mfg Co, Romulus, MI
Pedo Posterior	L	L28027/28	
Ful-fil Compules	L	041983 0224831	L.D. Caulk Co, Milford, DE
Prisma-fil Compules	L	041983 LYG 0306841	
Finesse	S	092183/6/ 83/12	
Amalux	L	40536	Pierre Roland, France
Scintilux 2	L	40412	
Heliomolar	L	050384	Vivadent, Schaan, Liechtenstein
Isomolar	S	B551183 + C701183	
Heliosit	L	22 1C-1D-2B 020584	
Isopast	S	22 B430484 + C370484	
Postifill	S	100483 U30826 + C30973	VMK, West Germany
Postilux	L	30932	
Barilux	L	100383	
Barifill	S	100483 U30974 + C31047	
Dentron Nano Lux 7	L	CH 40531 U	Dentron AG, St. Gallen, Switzerland

duration of eight cycles of the oscillation signal was then measured by comparison with an internal two-megahertz precision oscillator. The time of duration in microseconds of two periods was displayed and was called the Grindo-Sonic Reading (GSR). The fundamental frequency under flexure (f_F in Hz) can now be calculated from Equation 1 (Lemmens Elektronika):

$$f_F = \frac{2 \times 10^6}{\text{GSR}} \quad (1)$$

Before the elastic properties can be calculated from this frequency, certain characteristics of the tested bar need to be known: the surface area of the cross-section (A), the moment of gyration of the sample (I), the radius of gyration (i), and the density of the material (ρ). The latter two are determined by Equations 2 and 3, respectively:

$$i = \frac{I}{A} \quad (2)$$

where $I = wh^3/12$, $A = wh$, w is the width of the sample, and h is the height (or thickness), and

$$\rho = \frac{m}{LA} \quad (3)$$

where ρ is the density, m the weight of the sample, L the length, and A the cross-sectional area. Once these constants are known, the dynamic Young's modulus under flexure (E_{df}) is given by Equation 4, according to the Norm of the Belgian Concrete Industry (NBN B15-230):

$$E_{df} = 4 \times 10^{-6} \frac{\pi^2}{4.73^4} \frac{L^4}{i^2} f_F^2 \rho C \quad (4)$$

where C is a correction factor that depends on the radius of gyration i and Poisson's Ratio ν and is given by Equation 5

TABLE 2
DYNAMIC YOUNG'S MODULUS UNDER FLEXURE (IN MPa), AND VOLUME PERCENT OF INORGANIC FILLER OF DENTAL COMPOSITES (LITERATURE VALUES)

Product	After 24 Hours in Air (Mean ± SD)	After 3 Months in Air (Mean ± SD)	Volume Percent of Inorganic Filler
P-10	25117 ± 429	26826 ± 377	69.1
Occlusin	23774 ± 225	24886 ± 235	69
P-30	23385 ± 223	24886 ± 234	69.6
Concise	22531 ± 305	23590 ± 298	56.8
Estilux posterior XR1	21805 ± 238	23266 ± 223	66.2
Adaptic	21412 ± 230	22936 ± 195	56.3
Clearfil Posterior NB	21073 ± 475	22219 ± 183	
Clearfil	20373 ± 282	21141 ± 417	55.2
Miradapt	20320 ± 196	21346 ± 230	60.8
Adaptic radiopaque	19619 ± 365	21001 ± 285	55
Nimetic	19550 ± 552	20908 ± 549	58.9
Epolite 100	18206 ± 498	19314 ± 382	53
Aurafill	17985 ± 537	20210 ± 418	
Estilux posterior Y	17408 ± 476	19027 ± 575	58.1
Brilliant	16586 ± 276	17602 ± 263	53.9
Biogloss	15190 ± 385	16454 ± 213	51.9
Command Ultrafine	14803 ± 168	16409 ± 114	49.9
Brilliant Lux U	14451 ± 176	15119 ± 253	49.9
Pedo Posterior	13849 ± 571	16329 ± 550	
Ful-fil Compules	13842 ± 208	15007 ± 244	52.8
Amalux	13372 ± 221	14106 ± 242	
Prisma-fil Compules	13362 ± 210	14818 ± 185	51
Scintilux 2	11360 ± 304	11958 ± 285	40.5
Heliomolar	10612 ± 240	10980 ± 172	49.1
Nimetic-Dispers	10147 ± 175	11031 ± 181	40.5
Answer	9932 ± 275	10681 ± 300	
Isomolar	9619 ± 307	9996 ± 314	45.3
Silux U	9382 ± 155	9829 ± 136	36.3
Silar	9075 ± 157	9222 ± 173	35
Certain	3770 ± 123	9456 ± 187	
Microrest AP	8679 ± 250	9071 ± 137	17.1
Postifill	8471 ± 165	8831 ± 145	
Postilux	7707 ± 136	8496 ± 116	
Barilux	7594 ± 234	8406 ± 182	
Estic microfill U	6473 ± 58	6810 ± 43	36
Finesse	6437 ± 157	6900 ± 134	15.8
Heliosit	6401 ± 142	6683 ± 94	24.3
Barifill	6272 ± 150	6816 ± 150	
Durafill U	6085 ± 88	6497 ± 85	34.8
Dentron Nano Lux 7	5860 ± 242	6994 ± 87	
Isopast	5436 ± 268	5596 ± 275	20.6

(NBN B15-230). In the present experiment, C ranged from 1.0123 to 1.0148. It has been found that this equation yields the most representative results (Van Riet, 1984) and is easy to program:

$$C = \frac{1}{2} + \frac{4.73^2}{2} \times \frac{i^2}{L^2} [1 + \frac{6}{5} \times 2(1+\nu)] + \sqrt{\frac{1}{4} + \frac{4.73^2}{2} \times \frac{i^2}{L^2} [1 + \frac{6}{5} \times 2(1+\nu)] + \frac{4.73^4}{4} \times \frac{i^4}{L^4} [1 - \frac{6}{5} \times 2(1+\nu)]^2} \quad (5)$$

Poisson's Ratio is a function of the type of composite material used and varies between 0.25 and 0.35. A value of 0.30 was chosen since it was found that a variation of this value by ±0.05 resulted in changes in Young's moduli considerably smaller than the standard deviation.

All samples were stored for 24 hours in air, after which 30 measurements were made on each of them. The samples were then stored for three months in air and measured again to evaluate post-polymerization.

Results.

Table 2 gives the results of all the measurements for each brand. The products are ranked following their baseline values, ranging from 5436 MPa for Isopast® to 25,117 MPa for P-10®.

The rise in Young's modulus upon storage may be quite substantial, depending on the product. In the case of Dentron Nano Lux 7®, the result after three months in air was 19.6% higher than the baseline value. Silar® showed the lowest increase, with 1.6%. Fig. 2 presents the Young's moduli of only the posterior composites within the group of materials investigated. The Young's moduli of the posterior hybrid systems are about five times higher than that of the microfilled material Isopast®.

Discussion.

This non-destructive test to determine the fundamental period has advantages over those methods that act on the sample by a periodic series of impulses having a frequency equal or

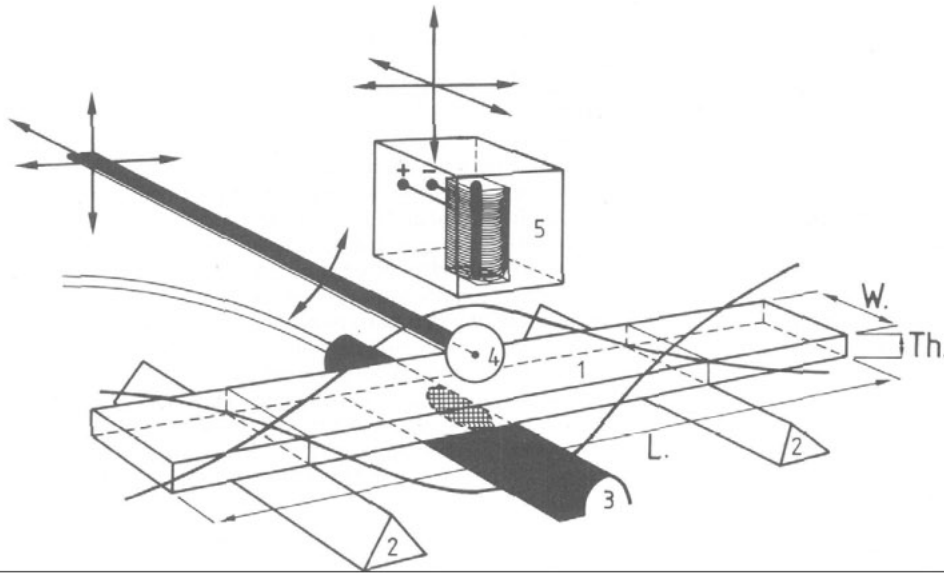


Fig. 1 — Schematic presentation of the rectangular sample (1) resting on two triangular supports (2). The microphone is immediately beneath the sample (3); the metal hammer (4) with the electromagnet (5) is above the sample.

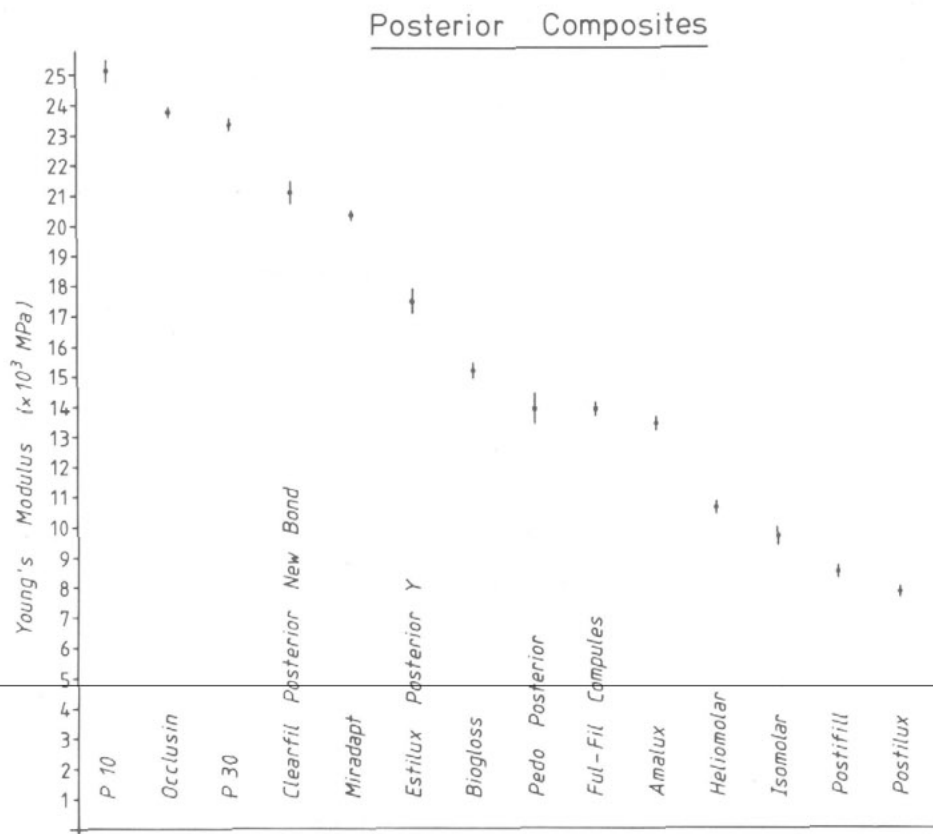


Fig. 2 — The Young's moduli of composites marketed as posterior materials. The vertical bars represent the standard deviations.

nearly equal to one of the natural frequencies of oscillation of the system. First, all mechanical influences induced by the contact of a driver, pick-up, or accelerometer are avoided, and, second, the direct measurement of the fundamental period that corresponds to the fundamental frequency (*i.e.*, the lowest resonance frequency of a sample) eliminates the need to scan the whole frequency range in search of the fundamental frequency

of the sample, as must be done with the driven mechanical resonance frequency methods (Spinner and Tefft, 1961).

The strain amplitude imposed on the samples in this method is extremely low, even relative to the strain applied in a low-strain, static, non-destructive test. Second, the strain rate is very high, which could account for the high values found with this method as compared with those found using low frequen-

cies or static tests, about 30% higher on the average (Whiting and Jacobsen, 1980a). Indeed, the higher the strain rate, the higher the Young's modulus of a visco-elastic material, since the most rigid component of the composite material is then enhanced (Nakayama *et al.*, 1974; Papadogianis *et al.*, 1984).

Finger (1974) also demonstrated an increase in Young's modulus as a function of storage time at room temperature. This phenomenon is called post-polymerization. It is suggested that free methacrylate groups continue to polymerize after the initial curing of the composites (Ruyter and Svendsen, 1978).

The mechanical properties of dental composite systems are known to depend to a large degree on the concentration and particle size of the filler. It has been found that Young's modulus is the mechanical quantity most highly correlated with volumetric filler content (Draughn, 1981; Boyer *et al.*, 1982). This correlation has been quantified before by Boyer *et al.* (1982). They investigated the Young's modulus vs. the volumetric filler content of 10 composites and found a correlation coefficient of 0.91. Comparison of the Young's moduli of 30 products, as determined above, with volumetric filler percentages compiled from the literature showed an exponential regression between Young's modulus and the volume percent of inorganic filler (Fig. 3). The correlation coefficient calculated from the logarithmic transformation is 0.92, which illustrates the accuracy and reliability of this new method.

Although Young's modulus is supposed to be independent of the particle size, it should be dependent on the maximum particle packing fraction, which is defined as the ratio of true particle volume to the apparent volume occupied by the particles (Draughn, 1981). This ratio is determined by the particle shape and size distribution. This could explain why the ranking of the Young's moduli values does not perfectly match that of

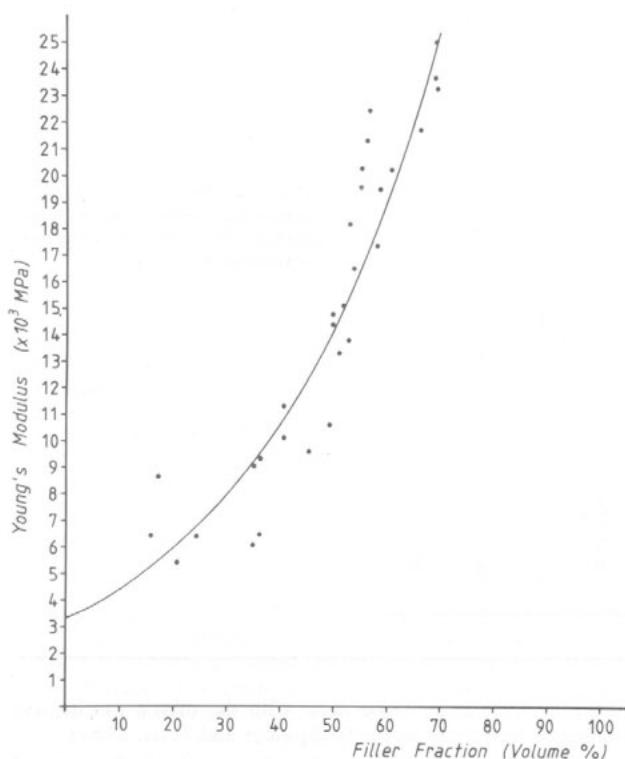


Fig. 3 — Young's modulus as a function of the volume fraction of inorganic filler. The values for the volume percentages were compiled from the literature.

the volume percent of inorganic filler, as is shown in Table 2. Furthermore, the use of this ratio may well lead to the engineering of composites that retain close particle-to-particle distances, this type having been designated as resin-bonded ceramics (RBC-type) by Cross *et al.* (1983). They result in high Young's moduli, since more energy is dissipated in the ceramic filler and less in the resin binder (Cross *et al.*, 1983). However, there is a significant risk with high filler fractions. If de-bonding occurs in such filler-rich composites, mechanical properties such as tensile strength and compressive strength will deteriorate more rapidly than otherwise, since the particles are close to, or even in contact with, each other. Since there is no bond between them, the strength will be drastically diminished (Söderholm, 1985). This could explain why many of the posterior composites are in the Young's modulus range of the micro-filled composites (Fig. 2). Although, clinically, these low-filler composites seem to be sufficiently wear-resistant in stress-bearing areas (Lambrechts *et al.*, 1984), the percentage of collapsed fillings rises dramatically after four years of clinical service (Lambrechts *et al.*, 1985).

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