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The Relationship Between Test Methodology and Elastic Behavior of Composites

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Comparisons were made of the Young's moduli obtained with tests that impose static, low-frequency, or high-frequency elastic deformations on dental composite systems.

The frequency of the imposed stress was reflected in the absolute value of Young's modulus. However, the values obtained at different test frequencies could be compared and understood by taking into account this frequency dependence. It was thus found that the composite structure largely determined the type of reaction to the imposed stress. The fundamental period test permitted the greatest differentiation in the elastic behavior of the investigated composites.

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Introduction.

In a previous article (Braem *et al.*, 1986), it was shown that the dynamic determination of Young's modulus can be used to distinguish several differently formulated dental composite materials and to study the sample condition non-destructively. That paper also pointed out the problem of comparing the results of dynamic and static testing. In the literature, the absolute values for Young's moduli of dynamic tests have been reported to be substantially higher than those obtained with static tests. This is believed to be inherent in the visco-elastic response of dental composites, which is a function of the applied strain rate in a particular test (Laeis, 1972; Nakayama *et al.*, 1974; Papadogianis *et al.*, 1984).

Very little is known of the comparability of the results obtained with these two test procedures. Therefore, the aim of the present paper was to investigate the comparability of results obtained at different test frequencies.

Materials and methods.

Sample preparation. — Table 1 lists the dental composites used in this study. Rectangular samples (35 × 5 × 1.5 mm) were prepared as described by Braem *et al.* (1986). Light-cured composites were cured for 60 sec on the top and for an additional 60 sec on the bottom (Luxor Activating Unit, ICI, Macclesfield, Great Britain). All the samples were finished with dry abrasive paper of 600 grit and were stored for 24 hours in air at room temperature before being tested at ambient temperature.

The determination of Young's modulus: The Fundamental Period Technique (FPT). — A complete description of this dynamic technique, with which Young's modulus is calculated on the basis of the duration of the fundamental period of a freely oscillating sample (Fig. 1), is given by Braem *et al.* (1986). From the period, the fundamental or resonance frequency could be calculated, which was found to range between 2000 and 4000 Hz for the composites. The results cited here are taken from that article. Ten samples of each composite were tested.

Dynamic Mechanical Thermal Analysis testing (DMTA). — Neither the thermal facilities nor the nitrogen gates of the DMTA apparatus (Polymer Laboratories Ltd., Loughborough, Great Britain; see Fig. 2) were used, since the tests were conducted at ambient temperature conditions.

This dynamic method requires a driven vibration to be applied to a clamped sample. If this sample is not an ideal elastic solid, and if internal molecular motion occurs in the same frequency range as the imposed vibration, it responds in a visco-elastic manner. Thus, the strain response lags behind the stress (Laeis, 1972; Finger, 1975; Wetton, 1984). Young's modulus can then be resolved into its elastic and viscous components, which are the storage or real (E′) and loss or imaginary (E″) components, respectively. They yield, by complex addition, the complex modulus E*. It is the real or storage component which is investigated in this paper. The imaginary or loss component, which is a measure of damping, will be the subject of later publications.

In the DMTA, the analyzer unit compares the applied stress and the corresponding strain signals. By using refined counting circuits, it resolves the strain into its storage and loss components. When the sample geometric constant is dialed into the instrument, log E′ is computed, and afterward is transformed into E′ (Wetton, 1984) for the present investigation. A single cantilever set-up was used (Fig. 2, inset). Minor modifications in the clamping device were made to compensate for the imperfect parallelism of the sample surface planes. The samples were tested at 0.1 Hz, 1 Hz, and 10 Hz. Three samples of each product were used.

The Static Test (STAT). — Knife-shaped edge supports were placed 11.6 mm from each other (Fig. 3). Between them, a small metal rod was positioned so that the sample could be loaded in a three-point bending apparatus (Dynstatgerät 5106, Zwick & Co., Einsingen, West Germany). A digital gauge was placed underneath the sample to record the bending in micrometers. In the case of proportionality, the measurement of the load required (**Δ**F) to bend the sample over an additional increment (**Δ**U) enabled Young's modulus to be calculated by means of Equation 1:

$$E = \frac{\ell^3 \, \Delta F}{4wh^3 \, \Delta U} \qquad (1)$$

where ℓ is the length between the supports, w the width of the sample, and h is the height.

Results.

The results are shown in Table 2. The values of the DMTA and the STAT results were close together, but substantially lower than those obtained with the FPT procedure. The values for the STAT test (O Hz) were the lowest, except for two composites. With the DMTA method, the imposition of a higher

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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	
Silux U	L	041183 5502 U 4Y3	
Silar	S	8601A + 8601B	
Occlusin	L	Lot SPO6 Mar 84	ICI PLC, Macclesfield, Great Britain
Estilux posterior Y	L	061984 182	Kulzer & Co. GmbH, Bad Homburg, W. Germany
Durafill U	L	061984 139	
Adaptic	S	053183 3A001	Johnson & Johnson, East Windsor, NJ
Miradapt	S	3D906 24051904	
Answer	S	201804 21300	
Brilliant	S	150584-36	Coltène AG, Altstätten, Switzerland
Brilliant Lux U	L	D3 120684-20	
Biogloss	S	840522	De Trey AG, Zürich, Switzerland
Heliomolar	L	050384	Vivadent, Schaan, Liechtenstein
Isomolar	S	B551183 + C701183	

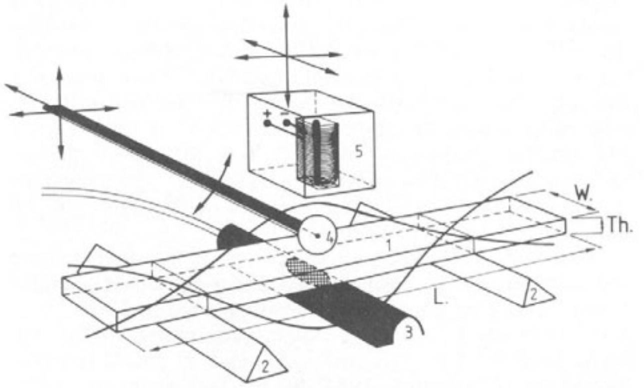

Fig. 1 — Experimental set-up for the determination of the fundamental period (FPT). The rectangular sample (L = length, W = width, Th = thickness) (1) rests on two triangular supports (2). The microphone (3) is immediately beneath the sample; the metal hammer (4) with the electromagnet (5) is above it.

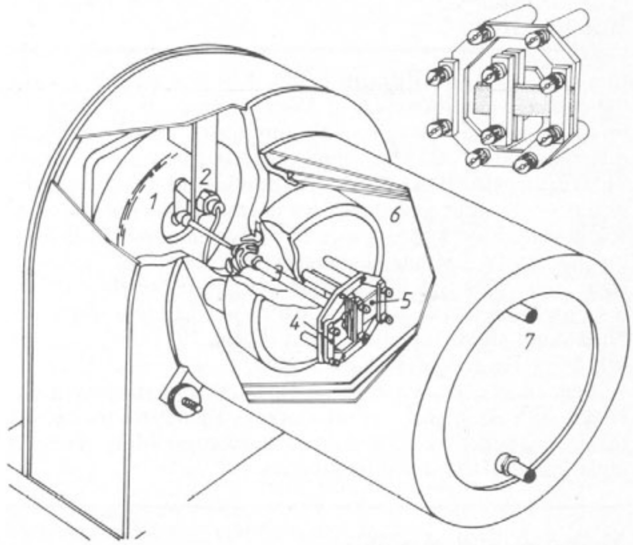

Fig. 2 — Mechanical head of the DMTA showing the essential features of sample mounting (inset), vibrating system, and transducer: (1) vibrator; (2) displacement transducer; (3) drive shaft; (4) clamps; (5) sample; (6) temperature enclosure; and (7) liquid nitrogen gates (not used in the present investigation). Inset: single cantilever clamping of the composite samples.

test frequency seemed to result in an increase in Young's modulus, although it was not statistically significant. This phenomenon could be observed in all tested composites. The values obtained with the FPT procedure (resonance frequencies) were the highest.

Generally, the ranking of the results from the STAT test and the FPT was comparable. However, this similarity was not found for the DMTA results of the microfilled types of composites.

Discussion.

This investigation was carried out at room temperature to eliminate additional thermal effects such as post-curing and temperature weakening.

From this investigation, it seems that the interpretation of the test results of static *versus* dynamic tests is a function mainly of the applied test frequency. The results in Table 2 show that Young's modulus is a function of the frequencies used in the various methods. Indeed, the measurements from the STAT test, the DMTA tests, and the FPT method yield values of Young's moduli at 0 Hz, 0.1 Hz, 1 Hz, 10 Hz, and the fundamental frequency of the investigated composites (from 2000 to 4000 Hz). An additional investigation with the FPT procedure showed that by individually changing the thickness of the sample, we could obtain a uniform resonance frequency of 4000 Hz for each tested composite (Table 2). Those results did not differ significantly from the previous ones, which emphasized that Young's modulus is constant within that frequency range. Therefore, in the following discussion the frequency range between 2000 Hz and 4000 Hz will be referred to as one resonance frequency.

In order to investigate the frequency influence, we made regression analyses of Young's modulus with respect to the volumetric filler fractions for each of the five frequencies investigated here (Söderholm, 1976; Braem, 1985). In a previous paper (Braem *et al.*, 1986), it was shown that these regression curves are of the form:

$$E = E_r e^{bx} \quad (2)$$

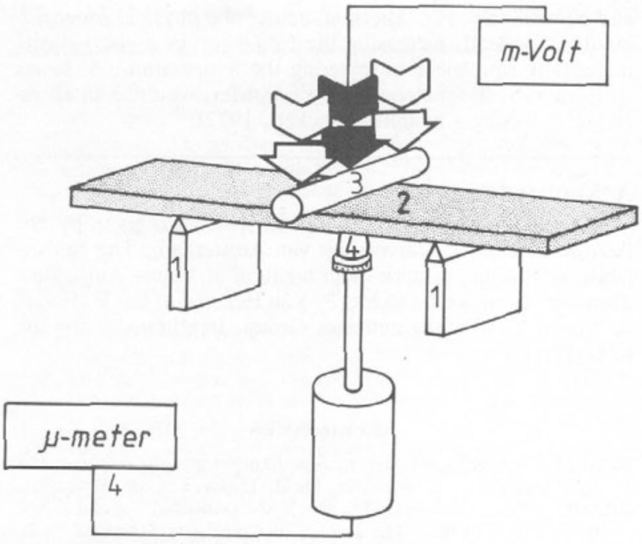

Fig. 3 — Schematic presentation of the static test (STAT) used: (1) supports; (2) rectangular sample; (3) metal rod transmitting the load; and (4) digital gauge and micrometer recording the bending of the composite sample.

where E is Young's modulus of the composite, E_r is Young's modulus of the matrix, b is the coefficient of the exponent, and x is the volumetric filler fraction. This equation is valid for the values of x included in Table 2. It gives only an extrapolation of Young's modulus for higher filler loads, since one must take into account that at such filler loads there is insufficient matrix material to bind the filler particles (Söderholm, 1985). One must also take into account that this phenomenological model assumes a perfect coupling of both phases.

In Fig. 4, the regression curve for each of the five frequencies investigated is shown on a semi-log scale. From this Fig., it seems that the curves at different frequencies approach the value of Young's modulus for silica, which is about 70,000 MPa (Van Vlack, 1975), extrapolated to 100% filler concentration. But, for the unfilled resin, the methods used yielded different results, in such a way that the slope (b) decreased with increasing frequencies. However, this impression could not be demonstrated statistically, probably because of the relatively small number (15) of composites investigated in the STAT and the DMTA methods, as compared with the 57 products in the FPT procedure. This resulted in larger standard deviations for the coefficient b than that obtained with the FPT test (Van Doren *et al.*, 1986). However, the same authors showed that by assuming comparable numbers and deviations for both the DMTA and STAT tests, they found statistically significant differences among the slopes of the five curves in Fig. 4. This would lead to the conclusion that the slopes of the curves decrease with increasing frequencies.

The physical explanation of this finding is related to the nature of the constituent phases. The silica responds purely elastically, which implies a negligible frequency dependence of Young's modulus. However, the matrix phase exhibits a much more pronounced frequency dependence because of its visco-elastic nature.

Thus, for the unfilled resins, it can be expected that the visco-elastic component will be much more pronounced in the test result. Indeed, the lower the frequency imposed on the sample (a low strain rate), the lower will be Young's modulus of a visco-elastic material, since the response of the most viscous component is enhanced under such conditions (Nakayama *et al.*, 1974; Papadogianis *et al.*, 1984). Therefore, low frequency tests will explicitly deform such composites, so that their results will show a greater deviation from tests which reflect purely elastic deformation.

In highly filled composites, the filler phase predominates over the visco-elastic matrix phase, and one may expect smaller differences between the results obtained at different frequencies than when the low-filled composites are considered. This is indeed the case, as can be derived from Table 2. Two composites (Occlusin and P-30), however, show rather deviating results when tested with the STAT procedure. A possible explanation could be that the substantial bending applied in this test causes the particles to come into closer interaction in these highly filled composites, thereby giving a higher Young's modulus. It must also be noticed that this test is believed to be the least sensitive testing procedure of the three methods investigated.

The interpretation of the behavior of the phases of highly filled composites at different frequencies is also in good agreement with the less-pronounced visco-elastic behavior of these materials as reported by Papadogianis *et al.* (1984, 1985). Further evidence can be found in Table 3, which gives the results of ultrasonic evaluation at 5 MHz (Whiting and Jacob-

TABLE 2

STATIC AND DYNAMIC YOUNG'S MODULUS UNDER FLEXURE (IN MPa), AND VOLUME PERCENT OF INORGANIC FILLER (VFC) OF DENTAL COMPOSITES (BRAEM, 1985)

Product	VFC %	STAT (MPa) O Hz Mean ± SD	DMTA (MPa) 0.1 Hz Mean ± SD	DMTA (MPa) 1 Hz Mean ± SD	DMTA (MPa) 10 Hz Mean ± SD	FPT (MPa) 2 kHz - 4 kHz Mean ± SD	FPT (MPa) 4 kHz
P-10	69.1	13,997 ± 848	14,125 ± 326	14,791 ± 410	15,488 ± 418	25,117 ± 429	25,706
Occlusin	69.0	16,532 ± 503	12,886 ± 218	13,808 ± 211	14,458 ± 180	23,774 ± 225	23,605
P-30	69.6	16,396 ± 427	13,647 ± 112	14,458 ± 130	15,140 ± 112	23,385 ± 223	24,110
Adaptic	58.4	12,697 ± 336	12,883 ± 537	13,490 ± 676	14,125 ± 568	21,412 ± 230	20,170
Miradapt	63.2	12,666 ± 729	12,023 ± 168	12,589 ± 121	13,183 ± 120	20,320 ± 196	19,281
Estilux posterior	58.1	9856 ± 492	9705 ± 148	10,000 ± 132	11,482 ± 143	17,408 ± 476	18,057
Brilliant	53.9	9951 ± 645	10,233 ± 403	10,715 ± 437	11,220 ± 461	16,586 ± 276	15,498
Biogloss	51.9	9017 ± 604	9205 ± 208	9795 ± 187	10,233 ± 197	15,190 ± 385	14,483
Brilliant Lux	49.8	8544 ± 244	6824 ± 109	7719 ± 56	8541 ± 77	14,451 ± 176	14,322
Heliomolar	49.1	6415 ± 284	5445 ± 229	6061 ± 276	6631 ± 301	10,612 ± 240	10,641
Answer	39.7	5718 ± 323	3119 ± 182	4436 ± 193	4775 ± 208	9932 ± 275	10,281
Isomolar	45.3	3912 ± 339	4677 ± 114	5333 ± 129	5984 ± 126	9619 ± 307	10,120
Silux	36.3	5912 ± 247	5140 ± 177	5534 ± 214	5916 ± 242	9382 ± 155	9066
Silar	35.4	5565 ± 228	5848 ± 43	6194 ± 40	6501 ± 49	9075 ± 167	8447
Durafill	37.5	2877 ± 196	2825 ± 162	3289 ± 119	3837 ± 113	6085 ± 88	6664

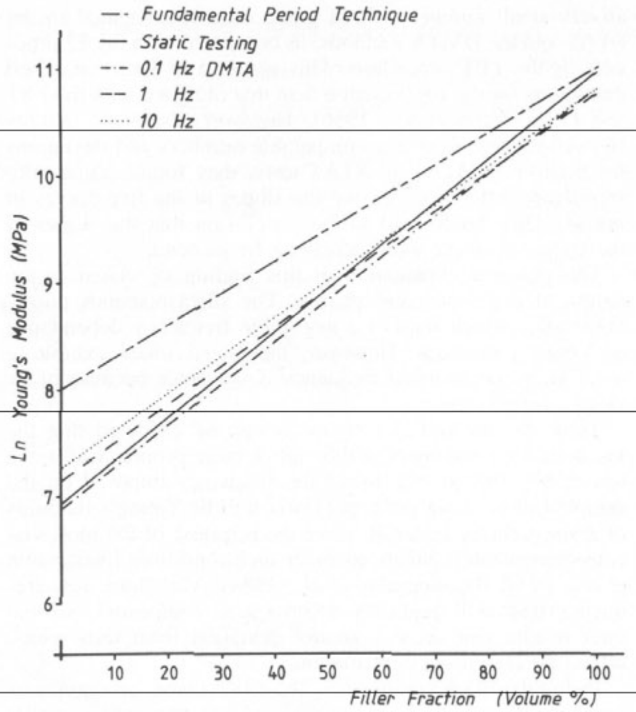

Fig. 4 — Regression analysis for the three frequencies investigated with the DMTA, the static test (STAT), and the fundamental period test (FPT), given on a semi-log scale.

TABLE 3
YOUNG'S MODULI OBTAINED *VIA* ULTRASONIC EVALUATION AND STATIC TESTS AT AMBIENT TEMPERATURE (LITERATURE RESULTS), COMPARED WITH THE FPT RESULTS

Product	Static Testing (MPa) Mean ± SD	FPT 2-4 kHz (MPa) Mean ± SD	Ultrasonic Testing (MPa) Mean ± SD
Concise	11,721 ± 1721	22,531 ± 305	24,550 ± 420
Adaptic	12,800	21,412 ± 230	24,500 ± 410
Isopast		5436 ± 268	7410 ± 50

son, 1980) and other static determinations of Young's modulus under comparable conditions, as reported in the literature (Dennisson and Craig, 1972; Draughn, 1981). The ultrasonic method yields Young's moduli that are slightly higher, as compared with the FPT results for the more highly filled composites such as Adaptic: 21,412 MPa for the FPT *versus* 24,500 MPa for the ultrasonic test. This effect is again much more pronounced for the low-filled composites such as Isopast: 5436 MPa for the FPT (Braem *et al.*, 1986) *versus* 7410 MPa for the ultrasonic evaluation. Also, the result for the static determination of Adaptic is in good agreement with the present findings.

Such a frequency dependence of the Young's modulus can also be understood by interpretation of the physical processes involved. Indeed, increasing the frequency in a visco-elastic material is equivalent to lowering the temperature. At lower temperatures, the material becomes stiffer, resulting in an increase in Young's modulus (Lockett, 1972).

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