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Dynamic and static elastic moduli of packable and flowable composite resins and their development after initial photo curing

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Summary *Objective*. The purpose of this study was to evaluate the dynamic (storage) shear modulus and the static shear modulus of elasticity of packable and flowable composite resins and to investigate their development after initial photo-curing.

Methods. Three pairs of a packable versus a flowable composite and a microfill composite resin were tested (Alert/Flow It, Filtek P60/Filtek Flow, Admira/Admira Flow, A 110). Cylindrical specimens (0.85 mm×18 mm) were made for each material. All specimens were conditioned and tested dry at 21 °C. The specimens were tested at 30 min, 24 h and 1 week after the end of photo curing.

Storage shear modulus and loss tangent were determined by conducting dynamic torsional loading in the frequency range from 1 to 150 Hz. Static shear modulus measurements were made by applying a constant load (below the proportional limit of the materials) for 10 s and recording the angular deformation of the specimens.

Data were analyzed by ANOVA and Duncan's Post hoc test ($\alpha=0.05$).

Results. Storage shear moduli (at 1 week measurement) ranged from 3.39 to 9.67 GPa, and loss tangents from 0.0735 to 0.0235; static shear moduli ranged between 2.66 and 9.80 GPa. High values of elastic moduli and low $\tan\delta$ values were obtained with packable composites, while low moduli values were obtained with flowable composites. Statistically significant ($\alpha=0.05$) differences were recorded between materials of the same category. Storage time, 24 h and 1 week after initial polymerization, resulted in significant increases in both moduli of elasticity. Dynamic shear storage moduli were highly correlated to the static ones ($r^2=0.92$; $P<0.05$).

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Significance. The results of the aging studies showed that the rigidity of these materials increases significantly even 1 week after the clinician turns off the curing unit.

Introduction

The elastic modulus of composite resin materials is an important property, which can yield useful data related to the materials behavior. It describes the relative stiffness or rigidity of a material and it is measured by the slope of the elastic region of the stress/strain diagram [1].

Elastic modulus plays an important role in the stress generated during shrinkage which accompanies the polymerization of resin composites. Stress is a critical parameter for the success or failure of the adhesive interface and may lead to marginal gap formation, marginal discoloration, post-operative sensitivity and secondary caries [2]. According to Hooke's law the force acting on the bonding interface equals the load induced deformation of the composite multiplied by its elastic modulus. The dry modulus at ambient temperature though is a simplification in that in the mouth, hydration and temperature will change and along with aging would affect the viscoelastic behavior of the composite material. Contraction stress build-up occurs since shrinkage is obstructed and the material is rigid enough to resist sufficient plastic flow to compensate for the original volume [3,4]. When restoring an adhesive cavity, the composite material is bonded to the walls of the rigid tooth structure and this is restrained from changing shape, except at the free surface, and further internal stresses will result. The magnitude of contraction stress was determined to depend, apart from the viscoelastic properties [3,5], on the configuration factor of the restoration (ratio of bonded to un-bonded composite surfaces) [3,6,7]. The elastic modulus of a composite must also be in close relationship with that of natural tooth structure. The tooth itself, however, is a composite of enamel and dentin, which elastically are totally different materials. From these materials the one with the lowest elastic modulus could be used as a standard, being dentin, with an elastic modulus of about 18 MPa [8].

The elastic modulus and other mechanical properties such as tensile strength, fracture toughness etc., are also important in determining the resistance to occlusal forces. Composites for cervical cavities must have a low modulus of elasticity to allow the composite to flex during tooth flexure. For occlusal, stress-bearing restorations, the modulus must be high to withstand occlusal forces and deformation [9,10]. The elastic modulus of resin based materials increases as the polymerization reaction proceeds [11]. As a result, the changes in the elastic modulus of resin based materials, during polymerization, is positively correlated to the degree of polymerization [12]. It is known that the polymerization reaction of light activated composites continues even after the end of light irradiation and the Degree of Conversion shows a gradual increase after light exposure [13].

Never before in restorative dentistry, have so many different materials been presented as alternatives for cavity restorations. Resin composites were first introduced as anterior restorative materials, but are being used more and more for posterior restorations. In the last few years, along with the development of resin modified glass-ionomer formulations and poly-acid modified resin composites (compomers), two new categories of resin composite have been developed: packable and flowable composites.

Packable composites are characterized by a high filler load and a filler distribution that gives them a stiffer consistency compared to hybrid composites. They are recommended for stress bearing posterior restorations because they have improved handling properties, and can be applied using a technique similar to that used for amalgam. Furthermore, a bulk filling technique has been recommended suggesting high depth of cure and low polymerization shrinkage of packable composites [14]. Some mechanical properties of these materials have been reported showing a wide variety in the results obtained [15–19]. This is not surprising considering the differences in the filler loading and the filler types used. Also, remarkable differences in the resin phase exist since some products use the so called 'ormocer' technology which combines glass-like (inorganic) constituents with polymer (organic) constituents. Further research into stress/strain properties of these materials is thus required before one can truly speak of amalgam replacement materials.

Flowable composites are low viscosity resin composites which were created by retaining

the same small particle sizes of traditional hybrid composites, but reducing the filler content and allowing the increased resin to reduce the viscosity of the mixture. Flowable composites can be used as liners, fissure sealants and can restore tunnel preparations. The clinical applications of these materials have been critically reviewed by Bayne et al. [20] who examined their usefulness beyond flow, after a preliminary screening of physical properties. The authors expressed some concern regarding the inferior mechanical properties of the flowable composites when compared to traditional hybrid composites, and discouraged their use in high-stress applications for restorative dentistry. However, their increased flow capacity might provide more contraction stress relaxation and could probably reduce the frequency of marginal microleakage and possible debonding [10,21], when used as liners underneath packable composites.

The purpose of the present study was to determine the static and dynamic modulus of elasticity and the damping (tan δ) of packable and flowable composite resins, as well as their development after initial photocuring. An experimental method was used that is capable of measuring viscoelastic behavior in a variety of ways, including: creep, constant load rate, sub-resonant dynamic and resonant dynamic experiments in bending and torsion. In this study, the resonant dynamic method and constant load rate were used for the determination of the elastic moduli.

Materials and methods

Seven materials were investigated in this study: three pairs of a packable versus a flowable composite resin of the same manufacturer (Alert/-Flow It, Filtek P 60/Filtek Flow, Admira/Admira Flow) and a microfill composite resin (A110). They are all dimethacrylate based dental composites except for Admira and Admira Flow, which belong to the ormocer-based technology. As shown in Table 1 the filler loading varied from 67 to 40% vol.

Specimen preparation was the same as described in previous articles [22,23]. The uncured composites were injected into glass capillary tubes, resulting in finished specimens 0.85 mm in diameter and 18 mm in length. The resins were photopolymerized (Coltolux 4 light, Coltene Whaledent, Dentalvertriebs GmbH, Konstanz/Germany). The light was tested for light output (600 mW/cm^2) using the radiometer included in the Coltolux 4. The light was directed towards the side of the capillary tube using 40 s of exposure for each 5 mm length of the tube (tip diameter 7 mm). Thorough curing was achieved because of the small diameter of the specimens.

Table 1 Composite resins investigated.

Material-code	Composition	Manufacturer
Alert (AL) Packable	Matrix: functional dimethacrylates of ethoxylated bisphenol-A polycarbonate resins (EBPADMA-PCDMA), crushed glass fibers (CS2). Filler: Barium-boro-silicate glasses, combined with silanated colloidal silica (67% vol., 83.5% wt). Particle size: 0.8 μm. Photo initiator, amine accelerator, UV absorber, inorganic pigments	Jeneric/Pentron Inc. 53, North Plains, Industrial Road, Wallingford CT 06492
Flow-It (FT) Flowable	Ethoxylated Bis-GMA, TEGDMA. Filler: Barium Glass, Silica, TiO_2 (70% wt). Photoinitiator, accelerator, UV stabilizer, inorganic pigments	Jeneric/Pentron Inc., 53 North Plains, Industrial Road, Wallingford CT 06492
Filtec P60 (P0) Packable	Matrix: Bιs-GMA, UDMA, Bis-EMA resins. Filler: zirgonia/silica (61% vol., 83% wt). Particle size range of 0.01–3.5 μm. Initiators, inorganic pigments	3M Dental Products, St Paul, MN, USA
Filtek flow (FL) Flowable	Matrix: BisGMA and TEGDMA resins. Filler: zirconia/silica (47% vol.). Particle size 0.01 to 6.0 μm (average 15 μm)	3M Dental Products, St Paul, MN, USA
Admira (AD) Packable	Matrix: ormocers, Bis-GMA, Urethan dimethacrylate (UDMA), triethylene-dimethacrylate (TEDMA). Filler: barium–aluminium-boro-silicate glass (0.7 μm), silicone dioxide (0.04 μm) (77% wt, 60.2% vol.)	VOCO GmbH P.O.B. 767 27457, Cuxhaven/Germany
Admira flow (AF) Flowable	Matrix: ormocers, functional dimethacrylate groups. Filler: 63% wt	VOCO GmbH PO Box 767 27457, Cuxhaven/Germany
A110 (A0) Microfill	Matrix: BisGMA and TEGDMA resins. Filler: colloidal silica (40% vol.). Particle size: 0.01–0.09 μm (average 0.04 μm)	3M Dental Products, St Paul, MN, USA

The specimens were mounted, using a jig for centering, between a Plexiglas disc (0.5 mm thick) and a rod, by means of a self-cured composite (Concise, 3M Dental Products).

Each of the seven composite resins was tested at three different ages so that there were 21 experimental groups with four specimens each. All the materials were conditioned and tested dry at 21 °C. The composite resins were tested 30 min after the end of photo-curing, which is the shortest appropriate time for mounting the specimens, 24 h and 1 week (group A, group B and group C, respectively). The specimens of the groups B and C were stored dry at 21 °C, isolated from ambient light.

The apparatus used in this study is described by Lakes [24]. It has since been modified to enable the study of micro-samples of foams and composites and has also been used in studying other dental materials. [19,20] The apparatus is capable of torsion or bending tests upon cylindrical specimens, following static or dynamic methods. A permanent, high-intensity, samarium cobalt magnet was fixed to the specimen end, generating torque. The cylindrical magnet (19.06 mm in diameter and 6.35 mm thick) produced a torque of 2.85×10^{-3} Nm/ampere at the center of a Helmholtz coil. The torque on the specimen was controlled by the electric current in the coil. A thin mirror (8.2 mm in diameter and 0.635 mm thick) was cemented onto the magnet to reflect the spot of a low power helium neon laser beam on a calibrated chart at a distance of $D=749$ cm. The mirror rotation angle φ is given by $\varphi = X/2D$, where X is the displacement on the chart of the laser beam.

The weight of the magnet resulted in a constant small axial tensile stress on the specimen. There was no constraint on the specimen for either torsion or extension. In principle, a torsional load will generate an axial deformation in a specimen; however, this has a non-linear effect, which is negligible at the small torque levels used. Moreover, the method for measuring torsional angular displacement is totally insensitive to any axial deformation that may occur [25].

The distribution of shear strain, γ, in a circular cylinder in torsion is:

$$\text{Strain} \quad \gamma = \frac{r\phi}{L}.$$

In this equation r is the radial distance from the centerline and L is the length of the cylinder. The distribution of shear stress, σ, depends on the material properties of the specimen. If it is linearly elastic or linearly visco-elastic, the shear stress is given by:

$$\text{Stress} \quad \sigma = \frac{MR}{\pi R^4/2}$$

where R is the specimen radius. Interpretation of torsion results is straightforward when the stress is sufficiently small for the specimens to be linearly visco-elastic. At higher stress, caution is required, since only the outer layers of the specimen experience the peak stresses. Consequently, the intrinsic material non-linearity is underestimated in the results.

Static shear moduli (G) measurements

For the determination of the shear moduli of elasticity (G) of the tested materials, a constant torque was applied to the specimen for 10 s, the angular displacement was recorded (at 10 s) and then the torque was 'instantaneously' released. The shear modulus $G=\sigma/\gamma$ was calculated from the equation:

$$G = \frac{2ML}{\pi R^4 \phi}$$

The compliance J is the reciprocal of the shear modulus. The shear strain and the shear modulus G at 10 s reflect the instantaneous elastic response of the materials. Surface strain at 10 s for the specimens ranged from 1.4 to 2.8×10^{-4}. These strain levels are found to be well within the range for linear behavior of composite materials and well below the proportional limit, when measured in compression [26].

Dynamic experiments-storage shear moduli (G_1) measurements

In a dynamic experiment, when the viscoelastic behavior is linear, both stress and strain vary sinusoidally, but strain lags behind the stress. The storage modulus G_1 (the real part of the complex modulus G^*) is in phase with the strain, whereas, the loss modulus G_2 (the imaginary part of the complex modulus G^*) is 90° out of phase with the strain and is related to dissipation of energy. In most cases of stiff solids, G_2 is small compared to G_1. The complex shear modulus G^* is, therefore, approximately equal to G_1 and is sometimes loosely referred to as modulus G. The loss tangent ($\tan\delta$) is a measure of damping and is defined as the ratio of the imaginary part to the real part of the complex modulus G^*. The angle δ is the phase angle between stress and strain sinusoids. Tan δ is proportional to

the energy loss per cycle within the framework of linear viscoelasticity.

In this study, steady-state dynamic torsional vibration was applied to the specimens by driven frequencies that ranged from 1 to 180 Hz. The displacement or amplitude was measured on the chart for each frequency. Viscoelastic parameters were calculated from the resonance frequency, ν_0, corresponding to the peak amplitude, and the resonance full width Δ_ν, that is the difference between the two frequencies at which the amplitude is half of the maximum.

The loss tangent is obtained from the relation

$$\tan\delta = \left(\frac{1}{\sqrt{3}}\right)\frac{\Delta\nu}{\nu_0}$$

The storage shear modulus G_1 was calculated from the relation

$$\nu_0 = \left(\frac{1}{2\pi}\right)\sqrt{\frac{G_1\pi r^4}{2LI}}$$

where r is the specimen radius, L is its length and I is the moment of inertia of the magnet that was measured to be 4×10^{-7} kg m^2. The loss modulus was calculated from $G_2 = G_1 \tan\delta$.

Statistical analysis

The mean values of both moduli of elasticity and loss tangents of the materials were analyzed by means of one-way analysis of variance (ANOVA) and Duncan's post hoc test at $P<0.05$ level. This was performed separately for each of the different ages. Linear regression analysis was performed to study the relationship between static shear moduli (G) and storage shear moduli (G_1) at the three different ages. A two-way ANOVA (independent variables: materials and age) was used to assess the effect of age.

Results

The mean values of the storage shear moduli (G_1), loss $\tan\delta$ and static shear moduli (G) of the examined materials are given in Table 2. Storage shear moduli (at 1 week measurements) ranged from 3.39 (AF) to 9.67 GPa (P0), loss tangents from 0.07 (FT) to 0.02 (P0) and static shear moduli from 2.66 (AF) to 9.80 GPa (AL). One-way ANOVA analysis and Duncan's test showed that the elastic moduli of packable composites were significantly higher ($P<0.05$) than those of flowable composites. The microfill composite A0 had significantly lower

Table 2 Viscoelastic parameters of composite resins investigated.

Material filler	Age	Loss tangent (tan δ)		Storage mod. (G_1)		Loss mod. (G_2) (Gpa)	Static shear mod. (G)	
		Mean	(std error)	GPa	(std error)		Gpa	(std error)
AL	30 min	0.0569	0.002	7.73	0.018	0.44	5.90	0.016
67% vol.	24 h	0.0292	0.001	9.54[a]	0.019	0.28	9.74	0.011
83.5% wt	1 week	0.0281	0.001	9.54[a]	0.013	0.27	9.8	0.011
P0	30 min	0.059	0.003	7.5	0.018	0.45	5.17	0.014
61% vol.	24 h	0.0275	0.001	9.38	0.018	0.26	8.93	0.012
83% wt	1 week	0.0235	0.001	9.67	0.015	0.23	9.18	0.011
AD	30 min	0.079	0.001	5.55	0.013	0.44	3.75	0.011
60.2% vol.	24 h	0.046	0.003	6.78	0.012	0.31	5.93	0.013
78% wt.	1 week	0.0377	0.001	7.16	0.017	0.27	6.69	0.018
A0	30 min	0.0626	0.004	3.69	0.019	0.23	2.80	0.012
40% vol.	24 h	0.0413	0.001	4.32	0.013	0.18	3.95	0.015
	1 week	0.0351	0.003	4.48	0.015	0.16	4.21	0.012
FL	30 min	0.0717	0.003	3.45	0.018	0.25	2.00	0.011
47% vol.	24 h	0.0599	0.001	4.19	0.012	0.25	3.48	0.013
	1 week	0.0467	0.001	4.64	0.019	0.22	4.21	0.011
FT	30 min	0.1206	0.002	2.7	0.012	0.33	1.51	0.011
70% wt	24 h	0.0785	0.001	3.51	0.015	0.28	2.51	0.016
	1 week	0.0735	0.001	3.63	0.018	0.27	2.72	0.018
AF	30 min	0.1012	0.003	2.66	0.015	0.27	1.67	0.016
63% wt	24 h	0.0712	0.002	3.39[b]	0.013	0.24	2.57	0.012
	1 week	0.0698	0.001	3.39[b]	0.017	0.23	2.66	0.011

$N=4$ specimens per group. Homogeneous statistical groups (alpha = 0.05) are shown by superscript letters.

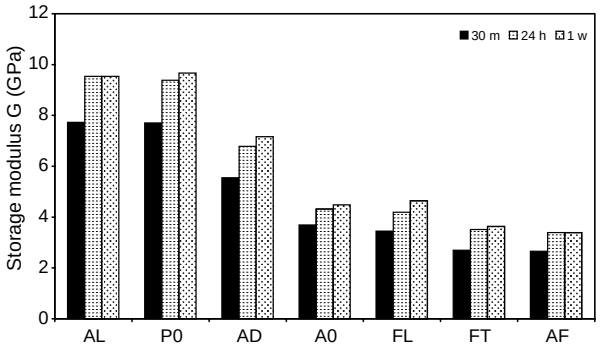

Figure 1 Dynamic elastic modulus development in composite resins after initial photocuring.

($P<0.05$) moduli than the packable composites, but significantly higher ($P<0.05$) than the flowable composites.

The storage time influenced both static and dynamic storage shear moduli of elasticity of the tested materials. Statistically significant ($P<0.05$) increases were observed in the values of dynamic and static moduli of elasticity for 24 h and 1 week aged specimens as compared to the values for 30 min aged specimens (Figs. 1 and 2). For the storage shear moduli (G_1) these increases ranged between 17 and 30% for 24 h and from 21 to 34% at 1 week as compared to the values for 30 min aged specimens. The increases observed in the values of the static shear moduli (G) varied between 41 and 74% for 24 h specimens, whereas, for 1 week aged specimens the increases ranged from 50 to 111% as compared to the values for 30 min aged specimens (Table 3). Storage time also resulted in a statistically significant ($P<0.05$) decline on damping (tan δ) of all the materials tested (Fig. 3).

Lower values were recorded for all the materials tested by the static method, except for the 24 h and 1 week aged specimens of AL, where the static shear moduli were higher than the storage shear moduli. The differences recorded between the dynamic and static moduli were higher in 30 min aged specimens for all the materials and ranged from 32 to 79% (Table 4).

Table 3 Dynamic and static moduli increase by aging (%).

Material	Dynamic modulus		Static modulus	
	30 min →24 h	30 min →1 week	30 min →24 h	30 min →1 week
AL	23.41%	23.41%	65.08%	66.10%
FO	21.80%	25.58%	72.72%	77.56%
AD	22.16%	29.00%	58.13%	78.40%
AO	17.07%	21.40%	41.00%	50.35%
FL	21.44%	34.49%	74.00%	110.5%
FT	30.00%	34.00%	66.22%	80.13%
AF	27.44%	27.44%	53.89%	59.28%

Linear regression analysis revealed significant correlation between the dynamic and static moduli ($r^2=0.92$; $P<0.05$) for all ages tested (Figs. 4–6).

Considerable differences were found between materials of the same category in both static and dynamic moduli of elasticity. Among the packable composites tested, AD showed significantly lower ($P<0.05$) values in both static and dynamic moduli than AL and P0. The storage shear moduli of AL were significantly higher ($P<0.05$) than those of P0 for 30 min and 24 h aged specimens, while in 1 week aged specimens P0 exhibited significantly higher ($P<0.05$) storage shear moduli than AL. As for the static shear moduli, AL showed significantly higher values ($P<0.05$) than P0 for all ages tested. Statistically significant differences between the flowable composites were also found in both dynamic and static shear moduli. FL had significantly higher values ($P<0.05$) than FT and AF in both dynamic and static moduli. The storage shear moduli of FT were significantly higher ($P<0.05$) than those of AF. An alteration was observed in the static shear moduli between these two materials.

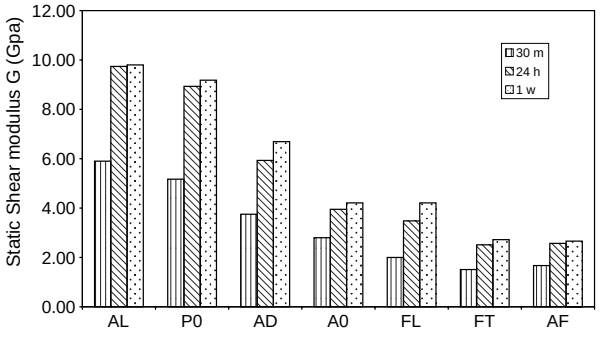

Figure 2 Static elastic modulus development in composite resins after initial photocuring.

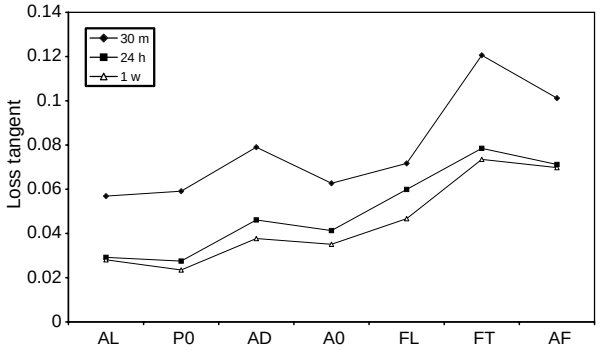

Figure 3 Loss tangent (damping) of composites decreased with aging.

Table 4 Static shear moduli were lower than dynamic storage shear moduli in all ages tested (% relationship).

Material	30 min	24 h	1 w
AL	31.00%	2.00%[a]	2.65%[a]
FO	48.93%	5.035%	5.33%
AD	48.00%	14.33%	7.02%
AO	31.78%	9.36%	6.41%
FL	72.50%	20.40%	10.21%
FT	78.80%	39.84%	33.45%
AF	59.28%	31.90%	27.44%

[a] Static modulus > than dynamic.

While AF showed significantly higher values ($P < 0.05$) than FT in the specimens aged 30 min and 24 h, in 1 week aged specimens the static shear moduli of FT were significantly higher ($P < 0.05$) than those of AF.

Discussion

It is desirable that the elastic modulus of a restorative material matches that of the tissue it replaces. There are no consistent values in the literature for enamel and dentin elastic moduli. According to Craig and Peyton [27] human dentin's Young's modulus is 18 GPa. Watts [28] reported a modulus of elasticity for dentin of 13 GPa measured in compression at body temperature. In a more recent study, Xu et al. [29] measured the elastic modulus of human enamel and dentin by an indentation method; they obtained a mean value of 19 GPa for dentin. Young's modulus of enamel exhibited a significant dependence on tooth orientation; for the occlusal section it was 94 GPa, while for the axial section it was 80 GPa.

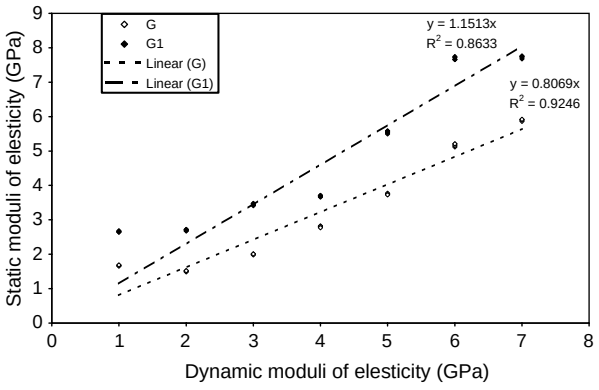

Figure 4 Correlation between static and dynamic moduli of elasticity at 30 min (linear regression).

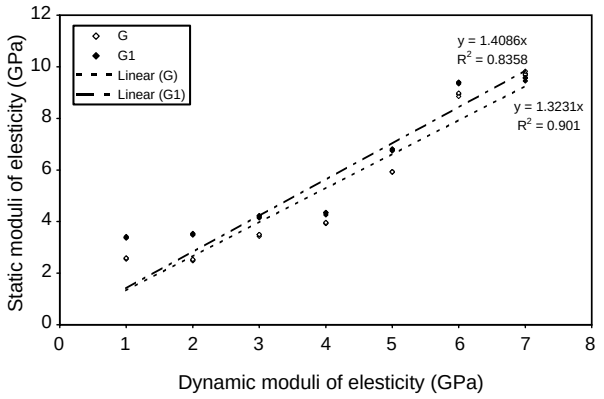

Figure 5 Correlation between static and dynamic moduli of elasticity at 24 h (linear regression).

An optimal restoration should mimic the structural, mechanical and physical characteristics of dentin and enamel, which could only be achieved if different restorative materials were combined.

It is clear that the values obtained with the tested materials are far from those of enamel. However, only approximate comparisons can be made with other research reports. Factors such as the choice of mechanical models (bending, shear, compression, and tension), and the testing conditions (strain rate, curing time, temperature etc.) have considerable effect on the ultimate elastic modulus value of viscoelastic resin composites. For a valid comparison between elastic modulus values, the factors described earlier must be equivalent. Any difference between these factors will make comparisons between elastic moduli not viable. If a mean Poisson's ratio [30] of 0.3 is selected for the examined materials, then only AD shows a static shear modulus, in 24 h specimens, approaching the Young's modulus of dentin. The rest of the packable composites showed higher values, whereas the flowable composites had lower values. Wide

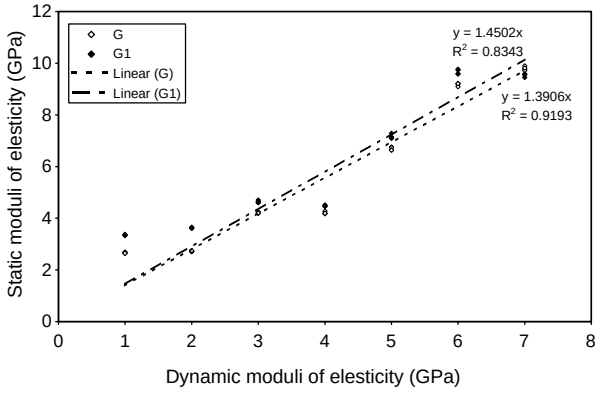

Figure 6 Correlation between static and dynamic moduli of elasticity at 1 week (linear regression).

variations were observed in both moduli of elasticity for all the materials tested.

All the tested materials exhibited statistically significant ($P<0.05$) increases in both elastic moduli 24 h and 1 week after photo curing. The dynamic elastic modulus of only two materials (AL and AF) was stable after 24 h of storage (Table 2). Previous studies [31] have shown that 30 min after light-curing only about 50–60% of the final modulus of the composites was developed and only about 60% of their flexural strength. In the present study, water storage of the specimens was avoided in order to exclude the effect of water and investigate only the effect of post-irradiation polymerization on the elastic moduli. As already mentioned in the Section 1, the elastic modulus of resin-based materials is positively correlated to the degree of polymerization. The increases in the elastic moduli found in this study can be attributed to post-irradiation polymerization.

Kildal and Ruyter [32] reported that the degree of conversion of proprietary resin-based inlay materials increased by approximately 13% after 24 h of storage when cured with a hand-held curing unit, while when they were initially polymerized to a greater degree with a light-curing oven, showed no post-irradiation polymerization. In a more recent study [33], it was found that composites with a greater degree of conversion showed less post-irradiation polymerization. Therefore, there seems to be an inverse relationship between the amount of post-irradiation polymerization and the degree of conversion in as-cured composites. A possible explanation for this inverse relationship is based on the diffusion rate of radicals. It has been reported that high viscosities of composite pastes induced slow diffusion of radicals [34]. This possibly explains the lower increases in elastic moduli of packable composites, as compared to the flowable ones, found in this study.

The amount of post-irradiation polymerization in dental composites depends on the reaction that takes place during light exposure. In the case of a rapid reaction, most parts of the conversion process end immediately after light exposure, leading to reduced post-irradiation polymerization. On the other hand, in the case of a slow advance of the reaction there is increased post-irradiation polymerization. The lower increases in the moduli values exhibited by the microfill composite A0 may be suggesting a more rapid completion of cure with this filler system, possibly due to less light scattering and more efficient activation. It has been previously reported [12] that the overall conversion of an experimental microfill composite was higher than that of a hybrid 1 h after light activation.

Static shear modulus was highly correlated to the dynamic storage shear modulus at all ages tested as shown in Figs. 4–6, although all of the materials exhibited lower static than dynamic modulus of elasticity. The high values obtained by the dynamic method are due to the high frequency rate applied on the specimens during testing [35]. When looking at the shear moduli as a function of time, it can be noticed that the variations of the dynamic storage shear moduli are smaller compared to those of the static ones.

In the present study, three pairs of a packable versus a flowable composite resin (each pair produced by the same manufacturer) (Table 1) were tested. Supposing that those materials have a similar chemical composition, the main difference would be their filler load that is reduced approximately 22–25%. Among packable composites higher values of both moduli of elasticity were obtained by AL and P0. They are both highly filled (67 and 61% vol., respectively), dimethacrylate based composites. The higher values obtained by AL could be attributed to the higher filler load and to the content of large-size irregular glass fibers [36]. The third packable composite investigated AD, exhibited significantly lower elastic moduli than the other two packable composites (AL and P0). Besides its lower filler content, AD has a different matrix composition. It uses a so-called 'ormocer' (Organically Modified Ceramic) technology with a matrix consisting of ceramic polysiloxane (silicon–oxygen-chains). The alcoxysilyl groups of the silane allow the formation of an inorganic Si–O–Si network by hydrolysis and polycondensation reactions, while the (meth) acrylate groups are available for thermally or photochemically induced organic polymerization [37].

As opposed to the high values obtained with packable composites, low moduli were recorded with flowable composites in the present study. Their moduli values ranged from 52 to 65% less than those of the packable composite resins, produced respectively, by the same manufacturer. This is consistent with previous findings [38]. The lower moduli of the flowable compared to those of the highly viscous composites is indirect evidence that flow ability is achieved mainly by increasing the proportion of monomer in the formulation of the composite paste. The observed moduli of FT are lower than those of FL, although FT has higher filler content. This could be explained in terms of a relatively lower degree of conversion. An alternative explanation could be that FT contains resin matrix with inherently higher flexibility, thus

introducing a confounding factor that interacts with filler loading.

The microfill composite resin A0 exhibited higher values for both moduli than the flowables FL and FT; in spite its lower filler content. This is possibly due to a higher degree of conversion as mentioned earlier. However, since A0 was the only microfill composite included in this study, it may be premature to conclude that the filler technology used for microfill composites produces materials with higher rigidity than that of the flowables.

Creep of composites is related to mechanical damping by a Fourier integral; if damping is relatively small, the loss tangent is proportional to the slope of the creep curve $J(t)$ on a log log scale: $\tan\delta \approx (\pi/2)\mathrm{d}\ln J(t)/\mathrm{d}\ln t$. For example, in a power-law creep, $J(t)=At^{n}$ he loss angle $\delta = n\pi/2$. For $\tan\delta = 0.02$ (AL and P0 at 24 h and 1 week), the creep would be 4.3% per decade, while for $\tan\delta = 0.07$ representative for FT and AF the creep would be 10.8% per decade (factor 10 in time). From the above it can be speculated that composite resins with low loss tangents and high moduli of elasticity, like packables, might show better clinical performance concerning deformation behavior. Knowledge of the deformation behavior of restorative materials is important in predicting the functional behavior in the mouth especially when these materials are designated for stress bearing areas.

To summarize, these laboratory results cannot be directly extrapolated to the clinical situation. Temperature rise and humidity in the oral environment are factors that would affect the viscoelasticity of composite resins. However, they are evidence of the rigidity of the materials investigated and the development of this rigidity after the clinician turns off the curing unit. Among the restorative resins, those marketed as packable and flowable tend to be at the opposite ends of the spectrum of elastic moduli values. The flowables are less rigid than the packables and the development of their elastic moduli (rigidity) after curing is not exactly the same as compared to the corresponding packable of the same manufacturer. It has been previously reported that flowables shrink more than the composites with higher viscosity [39]. It is also known that composite resins with high volumetric shrinkage develop high shrinkage stress. Hence, the effect of flowable composites or the combination of flowable and packable on interfacial stress build-up in the restoration cannot be easily predicted. Future clinical studies are needed to predict the behavior of these materials in the oral environment.

Conclusions

The experiments conducted in the present study, revealed a wide range of both dynamic and static moduli of elasticity among the different categories of resin based materials.

Post-irradiation time resulted in significant increases in both dynamic and static moduli of elasticity of the tested materials.

High correlations were observed between dynamic and static moduli of elasticity, and dynamic moduli were found to be higher than static.

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