

# The Effect of Temperature on Viscoelastic Properties of Glass Ionomer Cements and Compomers

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**Abstract:** The objective of this study was to determine the viscoelastic properties of different types of glass ionomer cements (GICs) and compomers under varying temperature conditions found in the mouth. The materials tested were a conventional GIC (Aqua Ionofil U), a resin modified GIC (Fuji II LC), a highly viscous GIC (Voco Ionofil Molar), and two polyacid modified composite resins/compomers (Glasiosite and Dyract Flow). Six groups of four specimens were prepared from each material. One group was stored dry for 24 h and was subsequently tested dry at 21°C. Each of the remaining five groups was stored for 24 h in distilled water at the temperatures 21, 30.5, 37, 43.5, and 50°C, respectively, and was subsequently tested at that temperature. Shear storage modulus and loss tangent were determined by conducting dynamic torsional loading. Static shear moduli were determined by applying a constant torque (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 test ( $\alpha = 0.05$ ). It was found that the viscoelastic properties varied significantly ( $p < 0.05$ ) across the different materials. The compomer Glasiosite, with the highest filler content, and the highly viscous GIC Voco Ionofil Molar exhibited the highest elastic moduli and lowest loss tangents. Viscoelastic properties varied also significantly ( $p < 0.05$ ) with temperature levels, but changes in the tested region were not indicative of a glass transition. Dynamic shear storage moduli were highly correlated to the static ones. Storage in water lowered the values of elastic moduli. © 2006 Wiley Periodicals, Inc. *J Biomed Mater Res Part B: Appl Biomater* 80B: 460–467, 2007

**Keywords:** polymer; viscoelastic; stress

## INTRODUCTION

Glass ionomer cements (GICs) are widely used in dentistry. Their advantages compared to more conventional dental materials, like composites, are good adhesion to tooth enamel and dentine,<sup>1,2</sup> good aesthetics<sup>3</sup> and long-term fluoride release.<sup>4,5</sup> However, these materials are limited in their applications because of low wear resistance, brittleness, and low strength.<sup>6–8</sup> Therefore, their use is generally restricted to specific indications such as small restorations in low stress bearing areas. Resin modified GICs and polyacid modified

composite resins (compomers) have been developed in an attempt to overcome the shortcomings in physical and mechanical properties of conventional glass-ionomers. These materials combine glass ionomer chemistry with resin composite technology, but have different setting mechanisms. The resin modified glass-ionomers are set by an acid–base reaction and free radical polymerization mechanisms.<sup>9,10</sup> The compomers are set by free radical polymerization, with only a limited acid–base reaction occurring later, as the material absorbs water from the oral environment.<sup>11,12</sup> Another recent modification of GICs is the highly viscous GICs, created to overcome the shortcomings of conventional GICs. These high powder/liquid ratio cements were designed as an alternative to amalgam for posterior preventive restorations.<sup>13</sup> In the mouth, restorative materials are subject to masticatory

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**TABLE I. Glass Ionomer Cements and Compomers Investigated**

Material-Code	Composition	Manufacturer
Fuji II LC (FL) Resin modified glassionomer	Powder: Calcium-alumino-fluoro-silicate glass (% m/m = 95–100). Mean particle size 4.5 $\mu\text{m}$ . Liquid: Polyacrylic acid (% m/m = 20–30), 2-hydroethyl methacrylate (HEMA) (% = 5–10), initiator (camphoroquinone).	G. C. Belgium N.V.
Voco ionofil molar (IM) Highly viscous glassionomer	Liquid: Polyacrylic acid, tartaric acid, water. Fillers: Calcium-alumino-fluorosilicate glass, 50 wt %, mean size 6 $\mu\text{m}$ .	Voco, Cuxhaven Germany
Aqua ionofil U (AI) conventional classionomer	Not available.	Voco, Cuxhaven Germany
Glasiosite (GL) compomer	UDMA, Bis-GMA mixture of diff. dimethacrylates, glass ceramics, silicates, initiators, additives 77.5 wt % average filler size 3 $\mu\text{m}$	Voco, Cuxhaven Germany
Dyract flow (DF) flowable compomer	Matrix: Phosphoric acid modified polymerizable monomers. Carboxylic acid modified macromonomers. Filler: Strontium-alumino-fluorosilicate glass 55.4 wt % Titanium dioxide Reactive diluent. Polymerization initiator, Stabilizer, Iron pigments.	DENTSPLY DeTrey GmbH De-Trey-str. 1 D-78467 Konstanz

forces of varying magnitude and rate of changes, as well as fluctuating temperatures. Like resin composites, GICs and their resin modified branches consist of polymer matrix with inorganic fillers embedded. The polymer matrix is more susceptible to the above-mentioned changes than the filler and thus dominates the mechanical behavior of these materials. Polymers are characterized as viscoelastic materials, that is, the mechanical properties are strain rate dependent or equivalently sensitive to the rate of loading. Besides strain-rate sensitivity, the temperature dependence of the viscoelastic properties of polymers is also of paramount importance. Plastics and rubbers show extensive changes in properties with changing temperature.<sup>14</sup> However, despite the temperature dependence of polymers, their properties can be improved with the addition of inorganic fillers which increase their modulus of elasticity and even their  $T_g$  (glass-transition temperature) to some degree.<sup>15</sup> Early studies<sup>16,17</sup> have shown that, in general, fillers do not affect the shape of the mechanical spectra but do cause a shift in their position. For example, when no filler is present, the glass transition peak for polystyrene occurs at 95°C, but is raised to 107°C when 37% filler is added. The general behavior remains similar to that of the pure polymer.

During the consumption of hot food, the typical maximum tooth surface temperature is  $\sim 47^\circ\text{C}$ , and although temperatures ranging from 0 to 67°C have been reported<sup>18</sup> such extremes are considered unrealistic.<sup>19,20</sup> Such fluctuations in oral temperature imply corresponding fluctuations in the observed mechanical properties of composite resins.<sup>21–23</sup> However, most of the studies examine the mechanical properties of composites in the absence of any explicit statement about the temperature, at the prevailing room temperature (21–23°C), or at the nominal body temperature (37°C).<sup>24</sup> This leaves the question of the mechanical behavior of composite

materials under oral conditions unaddressed. Even published standards specifications have been ambivalent. For example, ISO 4049<sup>25</sup> had the material set at 37°C in distilled water, but the test was at 23°C, dry. The mechanical properties of these materials, under more realistic temperature conditions, need further investigation.<sup>26</sup>

The specific objectives of this study were to determine the dynamic and static modulus of elasticity and the damping ( $\tan \delta$ ) of three different types of GICs and two compomers, under varying conditions of temperature found in the mouth, in an attempt to better predict their clinical performance. Two hypotheses were statistically tested. The first was that the different types of glass ionomer materials have different viscoelastic properties and the second that temperature affects their viscoelastic properties.

## MATERIALS AND METHODS

The materials that were investigated in this study are a conventional GIC (Aqua Ionofil U, AI), a resin modified GIC (Fuji II LC, FL), a highly viscous GIC (Voco Ionofil Molar, IM), and two compomers (Glasiosite, GL and Dyract Flow, DF). Their specifications are listed in Table I. The preparation procedure of the specimens was the same as the one described in earlier articles.<sup>27–29</sup> Specifically, the materials were injected into glass capillary tubes, resulting in cured specimens 0.85 mm in diameter and 18 mm in length.

The fabrication of the specimens was completed according to manufacturers' instructions at room temperature ( $23 \pm 1^\circ\text{C}$ ). For AI and IM the liquid and powder were mixed to form a paste which was quickly injected into the glass capillary tubes and was left there for 1 h, so that the completion of cure could be ensured. In preparing the encapsulated FL,

the capsules were mixed in an amalgamator (Silamat: Viva-dent, Schaan, Liechtenstein) and then the mixture was quickly injected into the glass capillary tubes and photo-polymerized for the time recommended by the manufacturer. Finally, for the preparation of GL and DF, the materials were injected into the glass capillary tubes and then were also photo-polymerized for the time recommended by the manufacturer.

For the photo-polymerization of FL, GL and DF the Coltolux 4 light (Coltene Whaledent, Dentalvertriebs GmbH, Konstanz/Germany) was used. The light was tested for light output ( $600 \text{ mW/cm}^2$ ) using the radiometer included in the Coltolux 4. The light was directed towards the side of the capillary tube and polymerization was repeated for each 5 mm length of the tube (tip diameter 7 mm). Thorough curing was achieved because of the small diameter of the specimens.

Twenty-four specimens of each material were prepared and were randomly assigned to six groups of four. One group of each material was stored in a dry beaker at  $21^\circ\text{C}$  for 24 h after fabrication and was subsequently tested dry at  $21^\circ\text{C}$ . Each of the remaining five groups was stored for 24 h in distilled water at the temperatures of 21, 30.5, 37, 43.5, and  $50^\circ\text{C}$ , respectively, and was subsequently tested at that temperature. 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).

The device used in these experiments is described in a previous article,<sup>30</sup> and it has since been modified to enable study of microsamples of foams and composites. The modified device has also been used in studying other dental materials<sup>27-29</sup> and is capable of torsion or bending tests upon cylindrical specimens, following static or dynamic methods. In order to generate torque, a permanent high-intensity samarium cobalt magnet was fixed to the specimen end. The cylindrical magnet (19.06 mm in diameter and 6.35 mm thick) produced a torque of  $2.47 \times 10^{-3} \text{ Nm/A}$  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  $D = 749 \text{ cm}$ . The mirror rotation angle  $\phi$  is given by  $\phi = X/2D$ , 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 nonlinear 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.<sup>31</sup>

The distribution of shear strain,  $\gamma$ , in a circular cylinder in torsion is

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

where  $r$  is the radial distance from the centerline and  $L$  is the length of the cylinder. The distribution of shear stress,  $\sigma$ , depends on the material properties of the specimen. If it is linearly elastic or linearly visco-elastic, the shear stress is given by

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

where  $M$  is the torque of the magnet and  $R$  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 nonlinearity is underestimated in the results.

This experimental method is capable of measuring viscoelastic behavior in a variety of ways, including creep, constant load rate, subresonant dynamic, and resonant dynamic experiments in bending and torsion. The resonant dynamic method and constant stress in torsion were used in this study.

#### Measurements by Dynamic Vibration Shear Test

Viscoelastic parameters of the materials can be obtained following the dynamic mechanical analysis technique. When a sinusoidal stress is applied to a perfectly elastic solid the deformation occurs exactly in phase with it. A completely viscous material will respond with the deformation lagging  $90^\circ$  behind the applied stress. When a sinusoidal stress is applied to a viscoelastic material on the other hand, it will behave neither as a perfectly elastic nor as a perfectly viscous body and the resultant strain will lag behind the stress by some angle  $\delta$ , where  $\delta$  is  $<90^\circ$ . The magnitude of the loss angle is dependent upon the amount of internal motion occurring in the same frequency range as the imposed stress. The resulting signal produces a real (storage) or in phase modulus  $G_1$  (the real part of the complex modulus  $G^*$ ) and an imaginary (loss) or out of phase modulus  $G_2$  (the imaginary part of the complex modulus  $G^*$ ) which represent elastic and viscous behavior of the sample respectively, with the storage and loss depicting the energy paths. 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 ratio of loss modulus to storage modulus ( $G_2/G_1$ ) defines the loss tangent often referred to as internal damping or  $\tan \delta$ . The angle  $\delta$  is the phase angle between stress and strain sinusoids.  $\tan \delta$  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 150 Hz. The displacement or amplitude was measured on the chart for each frequency. Viscoelastic parameters were calculated from the resonance frequency,  $\nu_0$ , cor-

responding to the peak amplitude, and the resonance full width  $\Delta\nu$ , 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  $4 \times 10^{-7}$  kg m<sup>2</sup>. The loss modulus was calculated from  $G_2 = G_1 \tan \delta$ .

In these viscoelastic materials, the storage shear modulus  $G_1$  is for brevity referred to as an elastic modulus in the following.

### Static Shear Moduli ( $G$ ) Measurements

For the determination of the static 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

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

where  $M$  is the torque of the magnet,  $L$  is the specimen length, and  $R$  its radius.

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  $1.4 \times 10^{-4}$  to  $2.8 \times 10^{-4}$ . 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.<sup>32</sup>

### Statistical Analysis

The mean values of storage shear moduli, loss tangents and static shear moduli of elasticity 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 procedure was followed separately for each of the different temperature. In order to study the relationship between static shear moduli ( $G$ ) and storage shear moduli ( $G_1$ ) at the six different levels of temperature, linear regression analysis was performed. A

two-way ANOVA (independent variables: materials and temperature) was used to assess the effect of temperature.

## RESULTS

The mean values of viscoelastic parameters of GICs and compomers measured in this study are given in Table II. It was found that there were statistically significant differences ( $p < 0.05$ ) in both the dynamic and static shear moduli of the materials tested. The compomer Glasiosite showed the higher elastic moduli values under most of the tested conditions and was followed by the highly viscous GIC IM. However, among the specimens that were tested dry at 21°C, the resin modified GIC FL exhibited significantly higher ( $p < 0.05$ ) dynamic and static elastic moduli values than the rest of the materials. At the temperatures of 43.5 and 50°C the storage shear moduli of IM were significantly higher ( $p < 0.05$ ) than those of all other materials tested (Table II, Figure 1). The conventional GIC AI had the lowest elastic moduli values.

Loss tangent  $\delta$  (damping) values were found to have statistically significant differences ( $p < 0.05$ ) among the materials tested (Table II, Figure 2). The highly viscous GIC IM showed less damping than all the other materials except for the dry stored specimens at 21°C. In the latter case the GL and FL showed the same  $\tan \delta$  values, significantly lower than those of the rest of the materials (Figure 2).

The elastic moduli decreased substantially for all the examined materials with the rise of the temperature. This temperature dependence was even greater for the flowable compomer DF and resin modified GIC FL. When tested in wet conditions, these materials showed the highest decrease in storage shear modulus ( $G_1$ ) between the temperatures of 21 and 50°C, 49% for DF and 45.9% for FL (Table III). For the static shear moduli ( $G$ ) the decrease was even higher (Table III). Loss tangent on the other hand, exhibited a statistically significant ( $p < 0.05$ ) increase between 21 and 50°C for all the specimen groups.

When tested dry at 21°C all specimen groups showed statistically significant ( $p < 0.05$ ) higher elastic moduli and lower  $\tan \delta$  values than the corresponding specimen groups tested in water. This result was more evident for AI and for FL. Between dry and wet conditions at 21°C, AI and FL showed a 44.68% and 22.12% decrease in the storage shear modulus respectively (Table IV).

Lower elastic moduli values were recorded for all the specimens tested by the static method. Linear regression analysis revealed significant correlation between the dynamic and static shear moduli for all the test conditions (Figures 3 and 4).

## DISCUSSION

The finding that the mechanical properties of GICs and compomers depend on the condition of test temperature was not unexpected and is relevant to their performance and survival

TABLE II. Viscoelastic Parameters of Glass Ionomer Cements and Compomers Investigated

Materials	Test Temp. (°C)	Loss Tangent (tan $\delta$ )		Storage Mod. $ G_1 $ (GPa)		Loss Mod $ G_2 $ (GPa)	Static Shear Modulus $ G $ (GPa)	
		Mean	Std error	Mean	Std error		Mean	Std error
Glasiosite (GL)	21 <sup>a</sup>	0.0295 <sup>b</sup>	0.001	5.48	0.025	0.162	5.46	0.020
	21	0.0375	0.033	5.35	0.021	0.2	0.13	0.018
	30.5	0.0522	0.001	4.75	0.020	0.24	4.62	0.012
	37	0.0665	0.001	4.15	0.018	0.27	4.02	0.013
	43.5	0.0811	0.002	3.8 <sup>c</sup>	0.015	0.3	3.66	3.15
	50	0.0971	0.003	3.4	0.014	0.33	2.9	0.020
Voco Ionofil Molar (IM)	21 <sup>a</sup>	0.0332	0.002	4.88	0.013	0.162	4.75	0.018
	21	0.0362	0.002	4.52	0.019	0.163	4.47 <sup>d</sup>	0.021
	30.5	0.0383	0.001	4.03	0.014	0.154	3.99	0.011
	37	0.0425	0.002	3.92	0.012	0.166	3.87	0.014
	43.5	0.0443	0.002	3.83 <sup>c</sup>	0.017	0.169	3.79	0.015
	50	0.0477	0.003	3.72	0.013	0.177	3.68	0.012
Fuji ULC (FL)	21 <sup>a</sup>	0.0295 <sup>b</sup>	0.003	5.83	0.021	0.1	5.75	0.018
	21	0.0641	0.002	4.54	0.019	0.29	4.48 <sup>d</sup>	0.021
	30.5	0.0963	0.002	3.63	0.023	0.349	3.35	0.013
	37	0.1104	0.001	3.22	0.018	0.35	2.57	0.011
	43.5	0.1232	0.001	2.79	0.014	0.344	2.2	0.015
	50	0.1368	0.001	2.47	0.015	0.33	1.84	0.011
Dyract Flow (DF)	21 <sup>a</sup>	0.0533	0.001	3.23	0.023	0.17	3.15	0.016
	21	0.063	0.001	3.02	0.019	0.19	2.98	0.019
	30.5	0.0809	0.001	2.45	0.018	0.198	2.42	0.019
	37	0.0947	0.003	2.09	0.013	0.198	1.99	0.018
	43.5	0.109	0.002	1.77	0.018	0.193	1.6	0.015
	50	0.1243	0.002	1.54	0.016	0.191	1.36	0.033
Aqua Ionofil U (AI)	21 <sup>a</sup>	0.0482	0.001	1.88	0.012	0.092	1.85	0.017
	21	0.0702	0.001	1.04	0.021	0.0733	1.02	0.011
	30.5	0.0772	0.003	0.9	0.026	0.069	0.87	0.015
	37	0.0826	0.001	0.88	0.024	0.0732	0.85	0.018
	43.5	0.0857	0.002	0.87	0.012	0.075	0.82	0.015
	50	0.0909	0.001	0.85	0.015	0.077	0.8	0.018

Note: N, four specimens per group.

<sup>a</sup> Stored dry for 24 h at 21°C, then tested dry at 21°C.

<sup>b,c,d</sup> Homogeneous statistical groups ( $\alpha = 0.05$ ) are shown by superscript letters.

in the oral environment. All the materials tested in this study showed significant decreases in storage shear moduli and static shear moduli and increases in loss tangents with in-

creasing temperature conditions. These findings are consistent with the temperature dependence of these parameters. The effect of temperature is due to the relaxation in polymers

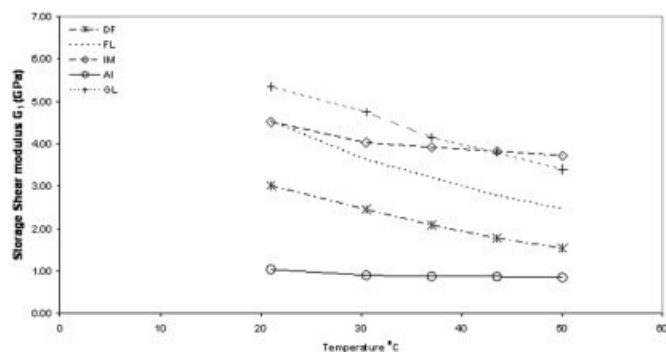


Figure 1. Storage modulus of Glassionomer cements and compomers decreased with rise in temperature (wet specimens).

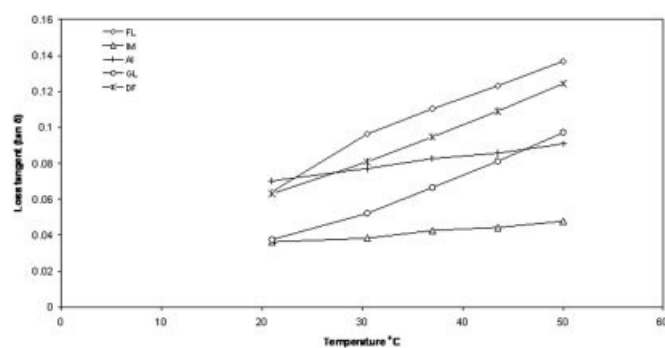


Figure 2. Loss tangent of Glassionomer cements and compomers increased with rise in temperature (wet specimens).

**TABLE III. Dynamic and Static Moduli Decrease with Rise in Temperature (%)**

Material	Dynamic Modulus (%)		Static Modulus (%)	
	Wet 21°C→50°C		Wet 21°C→50°C	
GL	36.44		43.46	
IM	17.69		17.67	
FL	45.59		59.02	
DF	49		54.36	
AI	18		21.56	

is by activated processes, being diffusive in nature. This accounts for the observed decline in elastic moduli with increasing temperatures at a constant torque level, as has been reported before.<sup>21,28,29,33</sup>

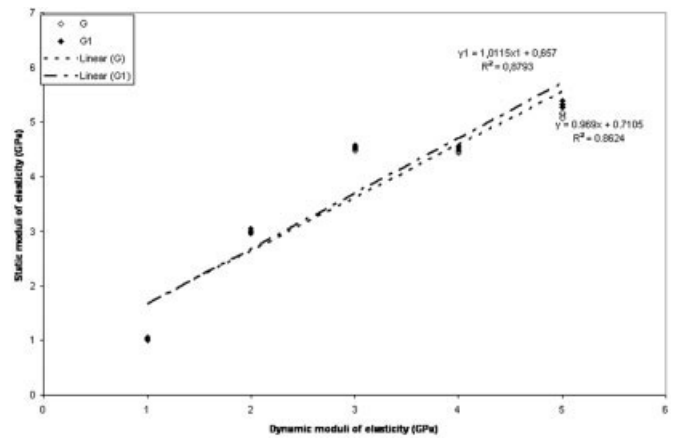
However, the viscoelastic behavior of all the materials tested in this study was stable within the temperature range of 21–50°C, that is, no large shifts indicative of the glass transition were evident. The data recorded on the elastic moduli were of the order of 10<sup>9</sup> N/m<sup>2</sup> (GPa) (Table II). The fact that the elastic moduli remained within this order of magnitude indicates that no major transitions were observed. A shift from the glass-like state to the rubber-like state (above the glass transition level) would subsequently have been followed by a decrease in the value of the elastic modulus, from a few 10<sup>9</sup> N/m<sup>2</sup> to the order of 10<sup>8</sup> N/m<sup>2</sup> or even 10<sup>7</sup> N/m.<sup>14,34</sup> The drop in the moduli values observed among the different temperature levels may be characteristic of minor or secondary transitions. This implies that these materials should maintain their properties in the temperature range found in the mouth. Temperature changes from 37°C are expected to be small and should lead to only modest changes in modulus.

Static shear modulus of the materials was highly correlated to the dynamic storage shear modulus at all the tested conditions as shown in Figures 3 and 4, even though the values of the static shear modulus were lower than those obtained by the dynamic method. The higher values obtained by the dynamic method is attributed to the high frequency rate applied on the specimens during testing.<sup>34</sup> It can also be noticed that the variations of the storage shear moduli, as affected by temperature rise, are smaller compared to those of the static ones (Table III).

Dynamic tests, such as dynamic mechanical analysis, are particularly well suited for viscoelastic materials, since they

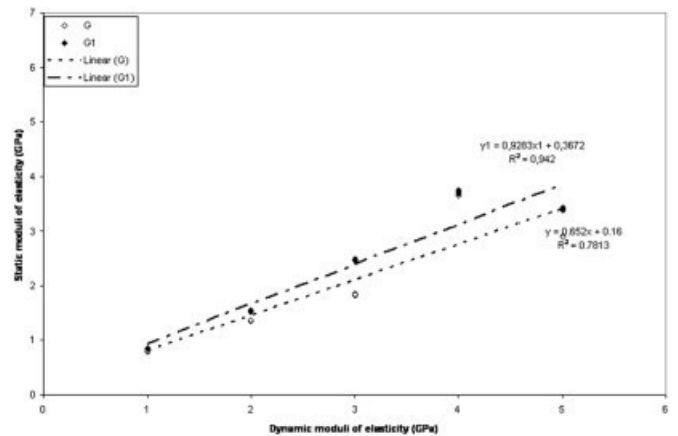
**TABLE IV. Storage Shear Moduli Decrease in the Wet Stored Specimens When Compared with the Dry Stored Specimens at 21°C (%)**

Material	Dry 21°C → Wet 21°C (%)
GL	2.37
IM	7.37
FL	22.12
DF	6.5
AI	44.68



**Figure 3.** Correlation between static and dynamic modulus of elasticity at specimens tested wet at 21°C (linear regression).

can determine both the elastic and viscous responses of the sample. Besides, since dental filling materials are clinically subjected to masticatory stresses, dynamic tests better mimic the cyclic masticatory loading than static tests under load. The tested materials showed low loss tangent values that ranged from 0.0295 to 0.0533 in dry specimens at 21°C and from 0.0477 to 0.1368 at 50°C. The observed loss tangent values were relatively small, typical of polymers in the glassy region, and much lower than values (near 1) typical of the glass–rubber transition. By contrast, metals including stainless steel and gold typically exhibit loss tangent below 0.001. An increase in loss tangent with temperature is typical in polymers below the glass transition temperature ( $T_g$ ). In the present study, even though the increases in the loss tangent were generally small, they were more noticeable in the resin containing materials FL, GL, and DF as compared to the highly viscous GIC IM. The latter showed significantly lower loss tangent values than the rest of the tested materials under all tested conditions (Table II, Figure 2). The modest increase of loss tangent with temperature is also representative of the glassy region. The results indicate the glass transition is not



**Figure 4.** Correlation between static and dynamic modulus of elasticity at specimens tested wet at 50 C (linear regression).

approached at temperatures likely to be encountered in the mouth. Therefore, excessive creep or viscoelastic flow under the quasi-static component of time-varying masticatory forces is not likely to be a problem in these materials.

The elastic modulus of all the materials tested was significantly lower when tested wet at 21°C as compared to the group of specimens tested dry at the same temperature. When water absorption occurs its plasticizing effect lowers the value of the elastic modulus. The effect of water was more apparent to the materials AI and FL (Table IV). This was no surprise and agrees with previous investigation findings.<sup>35,36</sup> GICs are hydrophilic and absorb water that continues to take a mediating role in the acid–base reaction, a process important for the development of the material's properties. Besides, on immersion, apart from dissolution effects, the high osmotic potential of the matrix leads to a large absorption of water. Therefore, the decline in elastic modulus of AI is the predicted natural consequence. The other material that showed a remarkable drop in elastic modulus, after immersion in water, was the resin modified GIC FL. The dry stored specimens at 21°C of FL had the highest values in both elastic moduli among all the other materials tested under these conditions. However, these values dropped in the specimens tested wet throughout the whole test temperature range, including the case of the 21°C and they were significantly lower than the values of GL and IM but higher than those of DF and AI. This water sorption effect might be attributed to the resin component of FL. Previous investigators have showed that restorative resin modified glass ionomer materials can absorb water up to 7% by mass and that the amount of water uptake depends on their poly (HEMA) content.<sup>37,38</sup> Other investigators also attributed the lower hardness of FL, as compared to a highly viscous GIC, to the water sorption by the resin component in FL.<sup>36</sup> The Young's modulus ( $E$ ) of FL, reported by the above investigators, was found to be proportional to the shear modulus of FL determined in the present study, under the same conditions (37°C).

Storage moduli of GICs and compomers were found to be lower than those of restorative composite resins reported in earlier studies using the same method.<sup>21,29</sup> These values are at the same levels as those of resin cements with lower filler loading than restorative composite resins.<sup>28</sup> Among other properties, elastic moduli of the materials are highly correlated with filler loading, filler properties, filler's size, and matrix composition. The materials that exhibited the highest moduli and the least damping were the compomer Glasiosite and the GIC Voco Ionofil Molar. IM is a highly viscous new generation GIC with a 50% by weight filler loading and particle size of 6  $\mu\text{m}$ . As mentioned before, highly viscous GICs' rely on improved chemistry and high powder/liquid ratio.<sup>13</sup> It has been previously reported that the mechanical properties of GICs are improved with increasing powder/liquid ratio up to a certain critical level.<sup>39</sup> When a mean Poisson's ratio of 0.3 is selected,<sup>40</sup> the values of storage shear modulus ( $G_1$ ) for IM obtained in the present study at 37°C is proportional to Young's modulus ( $E$ ), measured at 37°C, by Yap et al.<sup>36</sup> for F IX GP Fast (highly viscous GIC).

The elastic moduli of the compomer GL were in general higher than those of IM and similar to the composite resin Aelitefil with similar filler loading and particle size, measured by the same technique.<sup>22</sup> It has been previously reported that the viscoelastic behavior of compomers is similar to that of resin composites.<sup>41</sup> In the current study, the effect from rise in temperature was more marked to the specimens of GL at 43.5 and 50°C. The elastic moduli of GL were significantly lower than those of IM in the above temperature range, probably due to the presence of resin in the composition of GL. On the other hand, the higher stiffness of IM at elevated temperatures may be accounted for by the lack of resin in its composition. It seems that GICs named "highly viscous" may be viscous during the preparation phase, but viscosity does not result in a higher viscoelasticity of the polymerized solid. Yap et al.<sup>36</sup> also found the Young's modulus ( $E$ ) of Fuji IX (highly viscous GIC) to be higher than the composite-based materials investigated, including an ormocer, a composite, and a compomer. They inferred that the inclusion of glass ionomer phases increases the stiffness of the materials.

The flowable compomer DF exhibited lower moduli than the conventional compomer GL. The presence of less filler volume and the low viscosity of DF could explain the lower values obtained here. Sabbagh et al.<sup>41</sup> also reported that flowable composites and flowable compomer Dyract had nearly 20–30% lower values for both dynamic and static elastic moduli ( $E$ ) than universal composites.

The viscoelastic properties have the following relevance to the use of dental materials. First, they disclose the presence or absence of glass transition in the temperature range of clinical use. Second, if creep (related to loss tangent) is excessive, the restorations will suffer indentation. The differences in the viscoelastic properties between the materials evaluated do not necessarily mean that one material is better than the other, but that each material would better fit in a certain clinical application. Material selection for clinical applications does not depend on viscoelastic properties alone. Other physical and mechanical properties like strength, wear, resistance to environmental degradation, and aesthetics should also be considered.

## CONCLUSIONS

The research hypotheses of the current study were confirmed. Viscoelastic properties of GICs and compomers, measured by a torsion resonance and a static method in torsion, varied across the different materials and test conditions. The compomer GL, which has the highest filler content, and the highly viscous GIC, showed the highest elastic moduli and the lowest loss tangents. Viscoelastic properties varied with temperature levels (21, 30.5, 37, 43.5, and 50°C), but changes in this region were not indicative of a glass transition. Storage in water had a plasticizing effect on materials. This effect was more evident with conventional and resin modified GIC, probably due to increased water sorption.

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