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Effect of Oral Environmental pH on the Dynamic Characterization of Bioactive Restorative Materials

(Kesan pH Persekitaran Oral terhadap Pencirian Dinamik Bahan Pemulihan Bioaktif)

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ABSTRACT

The objective of this study was to investigate the effects of oral environmental pH on the viscoelastic properties of bioactive restorative materials (BRMs) by using dynamic mechanical analysis. Stainless steel molds were used to fabricate 40 beam-shaped specimens (12 × 2 × 2 mm) for each material. The specimens were finished, measured, randomly divided into four groups (n = 10), and immersed in aqueous solutions of pH 3.0, 5.0, 6.8 and 10.0 at 37 °C for seven days. The specimens were then subjected to dynamic mechanical analysis with a 5 N load and frequency range of 0.1-10.0 Hz. Data were analyzed using one-way ANOVA/Dunnet T3's test ($\alpha = 0.05$). Mean elastic modulus spanned from 2.68 ± 0.17 to 6.49 ± 0.71 GPa, while viscous modulus ranged from 0.43 ± 0.03 to 0.62 ± 0.12 GPa. Loss tangent differed from 77.30 ± 4.90 to 164.50 ± 9.12. Significant differences among pH were discerned for (i) Elastic modulus: Cention N - pH 3.0, 5.0, 10.0 > 6.8; Activa Bioactive - pH 3.0, 6.8, 10.0 > 5.0, (ii) Viscous modulus: Cention N - pH 3.0, 5.0, 10.0 > 6.8, and (iii) Loss tangent: Activa Bioactive - pH 5.0 > 3.0, 6.8, 10.0. Significant differences in viscoelastic properties were noted among the BRMs with Activa Bioactive presenting the lowest elastic modulus for all pH. Immersion of all materials in pH 6.8 yielded the highest elastic modulus, except for Activa Bioactive. The effects of environmental pH on viscoelastic properties of BRMs are material-dependent.

Keywords: Bioactive; dynamic mechanical analysis; pH; viscoelastic

ABSTRAK

Objektif kajian ini adalah untuk mengenal pasti kesan pH persekitaran mulut pada sifat viskoelastik bahan pemulihan bioaktif (BRM) dengan menggunakan analisis mekanikal dinamik. Acuan keluli tahan karat digunakan untuk menghasilkan 40 spesimen ujian berukuran 12 × 2 × 2 mm bagi setiap bahan. Kesemua spesimen tersebut kemudiannya dirapikan, diukur dan dibahagikan secara rawak kepada empat kumpulan. Spesimen daripada setiap kumpulan (n =10) direndam di dalam larutan akueus yang mempunyai pH 3.0, 5.0, 6.8 dan 10.0 pada suhu 37 °C, selama tujuh hari. Spesimen kemudiannya tertakluk kepada analisis mekanikal dinamik dengan beban 5 N dan julat frekuensi di antara 0.1-10.0 Hz. Data dianalisis menggunakan ujian ANOVA/Dunnet T3 sehala ($\alpha = 0.05$). Purata modulus elastik mempunyai julat antara (2.68 ± 0.17 GPa) dan (6.49 ± 0.71 GPa), manakala purata modulus likat adalah antara (0.43 ± 0.03 GPa) dan (0.62 ± 0.12 GPa). Purata kehilangan tangen pula adalah antara (77.30 ± 4.9) dan (164.50 ± 9.12). Keputusan analisis dari segi pH adalah seperti berikut: (i) Modulus elastik: Cention N - pH 3.0, 5.0, 10.0 > 6.8; Activa Bioactive - pH 3.0, 6.8, 10.0 > 5.0, (ii) Modulus likat: Cention N - pH 3.0, 5.0, 10.0 > 6.8 dan (iii) Kehilangan tangen: Activa Bioactive – pH 5.0 > 3.0, 6.8, 10.0. Perbezaan ketara dari segi sifat viskoelastik antara pelbagai bahan telah dapat dikesan dan modulus elastik bagi bahan Activa Bioactive didapati paling rendah dalam semua pH rendaman. Semua bahan yang direndam di dalam pH 6.8 menghasilkan modulus elastik tertinggi, kecuali Activa Bioactive. Kesimpulannya, kesan pH persekitaran ke atas sifat viskoelastik BRM adalah bergantung kepada bahan-bahan ujian.

Kata kunci: Analisis mekanikal dinamik; bioaktif; pH; sifat viskoelastik

Introduction

Resin-based composites are an integral part of the daily practice of dentistry. Despite their remarkable advancement, they still suffer a few setbacks, such as polymerization shrinkage, polymerization-induced stress, and microleakage (Ferracane 2005). Microleakage is a clinically imperceptible microscopic gap between cavity walls and the applied restorative material, thus, predisposing the tooth to secondary caries. As shown by a study, secondary caries is still the main reason why 72% of proximal restorations needed replacement (Nedeljkovic et al. 2020). Their finding corroborated multiple narratives and systematic reviews published over the last three decades (Demarco et al. 2012; Jokstad et al. 2001; Mjör et al. 1990). An attempt to overcome this negative phenomenon was observed by introducing contemporary bioactive restorative materials (BRMs) as a group of therapeutic biomaterials to resist acidic attacks and prevent secondary caries by buffering against the bacterial-induced decrease in environmental pH, promoting remineralization and encouraging precipitation of hydroxyapatite (Chan et al. 2018; Mayanagi et al. 2011).

Within the oral cavity, restorative materials are subjected to various mechanical and chemical challenges. Therefore, assessing physical properties under specific test conditions is necessary as this information is related to service longevity when these restorative materials are subjected clinically (Wang et al. 2003). Apart from mechanical stresses induced by mastication, restorative materials are also exposed to various chemical media, such as saliva, extrinsic/intrinsic acids and alkaline. Reportedly, acidic environments can compromise the structural integrity of restorative materials and contribute to the occurrence of secondary caries, while an alkaline environment accelerates material hydrolysis and infliction of surface microstructural damage of resin-based composites (Attin et al. 2014; Cilli et al. 2012).

Presently, most published literature about the physical properties of BRMs is conducted via static testing. BRMs are polymeric materials with viscoelastic behaviors. Hence, they are better assessed with a dynamic test, particularly dynamic mechanical analysis as it can determine both elastic and viscous properties (Vouvoudi & Sideridou 2012). While some *in-vitro* studies suggested that BRMs are comparable to resin-based composites, others reported inferior/variation of flexural properties (Alrahlah 2018; François et al. 2021; Sujith et al. 2020; Yap et al. 2021a). Given that BRMs may potentially prevent a tooth from spiraling down the restorative cycle, the prospects of BRMs are worth investigating. However, studies on the effect of oral environmental pH on the viscoelastic properties of BRMs are relatively unexplored. To better comprehend the viscoelastic properties of BRMs, this study aims to investigate to determine the effects of oral environmental pH on the viscoelastic properties of BRMs and compare the viscoelastic properties between different BRMs. The null hypotheses for this study are (a) viscoelastic properties of the various BRMs are not influenced by environmental pH, and (b) there is no difference in viscoelastic properties between the various BRMs.

Materials and Methods

Evaluated materials

The technical profiles of the materials examined are shown in Table 1. They involved a non-bioactive resin-based composite as control, Filtek Bulk Fill Posterior (FB) and four BRMs. Namely (i) giomer (Beautifil-Bulk Restorative (BB)), a group of restorative materials with fluoride ions leaching ability due to the incorporation of pre-reacted filler technology; (ii) bioactive composite (Activa Bioactive (AB)), a resin-modified glass ionomer cement containing patented bioactive shock-absorbing rubberized ionic-resin and bioactive glass; (iii) alkasite (Cention N (CN)), a relatively new group of bioactive materials with incorporation of alkaline fillers within a resin matrix of methacrylate and (iv) resin-reinforced glass ionomer restoratives (Riva Light Cure HV (RV)), a hybrid between glass ionomer cement and resin-based composites which leaches therapeutic ions.

Sample size calculation

Based on a previous study (Yap et al. 2021b) that investigated the viscoelastic properties of highly viscous glass ionomer cement after exposure to different chemical media, the effect size, d is determined from the formula, $\frac{M1 - M2}{S}$, where M_1-M_2 is the difference between group means; s is the standard deviation of either group. Hence, a minimum sample size of $n = 9$ specimens per group was calculated from the hypothesized effect size (d) of 0.4, Power (1- β) of 0.95, and a two-tailed significance level (α) of 0.05, df 19, critical f 1.64.

Specimen preparation and immersion protocol

Stainless steel molds were used to fabricate 40 beam-shaped specimens for each material according to the

TABLE 1. Technical profiles and manufacturers of the evaluated materials

Material (abbreviation)	Manufacturer	Type and curing method	Resin	Filler	Filler content % (Weight/ Volume)
Filtek bulk-fill (FB) [As control]	3M ESPE, St Paul, MN, USA	Bulk-fill composite (light cured)	ERGP-DMA DD-DMA UDMA Aromatic-UDMA	Nano scaled zirconia, silica, Ytterbium trifluoride	76.5/58.4
Cention N bulk fill (CN)	Ivoclar, Vivadent Inc., NY, USA	Alkasite (self-curing powder/liquid with optional additional light-curing)	UDMA DCP Aromatic aliphatic-UDMA PEG-400 DMA	Br-Al-Si glass filler, Ytterbium trifluoride, Isofiller (copolymer), Calcium barium Aluminium fluorosilicate glass and calcium fluorosilicate glass	78.4/NA
Beautifil-bulk restorative (BB)	Shofu, Kyoto, Japan	Giomer (light cured)	Bis-GMA UDMA Bis-MPEPP TEGDMA	S-PRG filler based on fluoro-alumino-silicate glass and nano filler	87/74.5
Activa Bioactive restorative (AB)	Pulpdent, Watertown, MA, USA	Bioactive composite (ACTIVA enhanced -RMGIC) (dual-cured/light-cured)	Blend of UDMA and other methacrylates with modified polyacrylic acid	Silica, amorphous and sodium fluoride	55.4/NA
Riva light-cure HVGIC (RV)	SDI Limited, Bayswater, Australia	Encapsulated resin reinforced high viscosity glass ionomer cement(light-cured)	Compartment 1: Polyacrylic acid, Tartaric acid, HEMA, dimethacrylate-cross-linker Acid monomer	Compartment 2 : Fluoro-alumino-silicate glass	95.0/NA

Bis-GMA= Bisphenol-A glycidyl methacrylate, DD-DMA= 1,12-dodecanediol dimethacrylate, UDMA = Urethane dimethacrylate, TEGDMA= Triethylene glycol dimethacrylate, PEGDMA= Polyethylene glycol dimethacrylate, DCP= Tricyclodecan-dimethanol dimethacrylate, Aromatic aliphatic-UDMA= Tetramethyl-xylylendiurethane dimethacrylate, PEG-400 DMA= Polyethylene glycol 400 dimethacrylate, Bis-MPEPP= 2,2-Bis (4-methacryloxypolyethoxyphenyl) propane, HEMA= 2-hydroxyethyl methacrylate

mini flexural test specifications (12 × 2 × 2 mm) as it may be more clinically relevant (Yap et al. 2020). The specimens were manipulated according to manufacturers' instructions where applicable and/or placed into the molds in a single increment. The material-filled molds were compacted between two mylar strips with glass slides to remove excess material. A light-emitting diode (LED) curing light (Bluephase N, Ivoclar Vivadent) with a wavelength of 385-515 nm, power of 1,200 mW/cm^2 (high power), and an exit window of 8 mm was used for light polymerization. FB and BB were light polymerized for 10 s each in two overlapping irradiations from the top and bottom surfaces, and similarly, RV was polymerized for 20 s on each surface. The light-curing unit was recharged after every ten specimens, and a radiometer (Bluephase Meter II, Ivoclar Vivadent) was used to validate the consistency of its performance. CN was mixed according to manufacturers' instructions and allowed to set for five minutes. As for AB, the material was mixed using an auto-mix syringe and was dispensed directly into the molds. It was allowed to self-cure for 20 s before being light polymerized for 20 s.

After light polymerization/self-curing, the materials were left undisturbed in their molds for five minutes. Later, specimens were removed from their molds and finished with fine contouring/polishing discs (Sof-Lex, 3M ESPE, St Paul, MN, USA). A digital caliper (ABS Digimatic, Mitutoyo, Kawasaki, Japan) was used to verify the dimensions of the finished specimens and the parallelism between their opposite surfaces. The specimens (n=10 each) were randomly divided into four groups of 10 and subsequently stored for 7 days at 37 °C in the following immersion mediums, reflected in Table 2. Artificial Saliva Gal Fovet (SAGF) medium was the media of choice to prepare the artificial saliva used in this study because its pH of 6.8 resembles the intra-oral saliva when excreted from salivary canals. Moreover, the major constituents of this artificial saliva represent most of the composition found in human saliva (Gal et al. 2001). All immersion mediums were freshly prepared for each set of tests and verified with a digital pH meter (pH 2700, Eutech, Singapore).

TABLE 2. Immersion groups with their respective pH values and composition

Immersion group	pH values	Composition	Titratable acidity mean volume of 0.1M NaOH
1	3.0	Artificial saliva titrated with 0.02 M hydrochloric acid	1.20 ± 0.2 mL
2	5.0	Artificial saliva titrated with 0.02 M lactic acid	0.90 ± 0.1 mL
3	6.8	Artificial saliva	-
4	10.0	Artificial saliva titrated with 0.1 M sodium hydroxide	-

TITRATABLE ACIDITY

Besides pH measurements, titratable acidity (TA) was also determined in triplicated immersion mediums using the same pH meter by gradually adding 0.1 M of sodium hydroxide into 80 mL of respective immersion mediums. The mediums were stirred manually until a stable pH measurement was obtained after each addition of sodium hydroxide until the titration endpoint of pH 8.2 was achieved (Friedrich 2001). TA is then calculated by multiplying a correction factor of 0.96, obtained by standardizing 0.1M sodium hydroxide solution with Potassium Hydrogen Pthalate (KHP) (Syed & Chadwick 2009). Efforts of standardization of respective pH values throughout the entire immersion period included verification with digital pH on a daily working day basis and application of self-sealing semi-transparent film on the top opening of immersion containers.

DYNAMIC TESTING

Specimens were subjected to dynamic mechanical testing (DMA RSA-G2, TA Instruments, New Castle, DE, USA) in a 3-point bending mode at 37 °C and a frequency of 0.1 to 10 Hz after the 7 days immersion period. The range of frequency and temperature simulates the average human masticatory rate and intra-oral temperature (Po et al. 2011). The distance between two supports was set at 10 mm, with an axial load of 5 N. Parameters such as elastic modulus, viscous modulus and loss tangent were determined by using the following Equation (1-3):

Elastic modulus

$$E' = (\sigma^o/\varepsilon^o)\cos\delta = (f_o/b\mathrm{k})\cos\delta \quad (1)$$

Viscous modulus

$$E'' = (\sigma^o/\varepsilon^o)\sin\delta = (f_o/b\mathrm{k})\sin\delta \quad (2)$$

Loss tangent

$$\mathrm{Tan}\,\delta = E''/E' \quad (3)$$

where σ° is the maximum stress at the peak of the sine wave; ε° is the strain at the maximum stress; f_o is the force applied at the peak of the sine wave; b is the sample geometry term and, k is the sample displacement at the peak.

The sample geometry for a three-point bending bar was calculated as follows (Equation 4):

$$4B_s H_s^{\,3} / L_s^3 \quad (4)$$

where B is the width in millimeters; H is the height in millimeters; L is the distance between support in millimeters; and s denotes the specimen.

STATISTICAL ANALYSIS

Data analyses were performed with SPSS statistical program (Version 26.0, IBM Corp., New York, USA). Shapiro-Wilk test was employed to determine the normality of data, and all were found to be normally distributed ($P > 0.05$). Hence, parametric analyses were performed at a significance level of $P = 0.05$. Inter-material and inter-medium differences were first assessed with Levene's test for homogeneity of variances before proceeding with one-way ANOVA and Dunnett's T3 post hoc test.

RESULTS

The titratable acidity mean volume of 0.1 M sodium hydroxide used to achieve the titration endpoint is reflected in Table 2. Mean elastic modulus, viscous modulus and loss tangent for various BRMs are shown in Tables 3 to 5 and Figures 1, 2, and 3 after immersion in environmental pH of 3.0, 5.0, 6.8, and 10.0.

TABLE 3. Mean elastic modulus values (GPa) of the various BRMs (standard deviations in parentheses)

Materials [x] \ Environmental pH[y]	pH 3.0	pH 5.0	pH 6.8	pH 10.0
FB	5.83^{Aa} (0.66)	6.01^{Aab} (0.96)	6.16^{Aa} (0.60)	6.11^{Aa} (0.86)
BB	5.97^{Aa} (0.81)	6.04^{Aa} (0.79)	6.49^{Aa} (0.71)	6.38^{Aa} (0.75)
CN	4.22^{B} (0.58)	4.51^{Ab} (0.59)	5.32^{Aa} (0.48)	4.11^{B} (0.64)
AB	2.85^{AB} (0.39)	3.49^{A} (0.27)	2.95^{AB} (0.51)	2.68^{B} (0.17)
RV	6.03^{Aa} (0.69)	6.08^{Aa} (0.65)	6.14^{Aa} (0.98)	6.11^{Aa} (0.56)

x = Values with the same lowercase letters in the same column are not significantly different between materials, y =Values with the same uppercase letters in the same row are not significantly different between environmental pH

TABLE 4. Mean viscous modulus values (GPa) of the various BRMs (standard deviations in parentheses)

Materials [x] \ Environmental pH[y]	pH 3.0	pH 5.0	pH 6.8	pH 10.0
FB	0.53^{Aa} (0.07)	0.60^{Aa} (0.10)	0.62^{Aa} (0.12)	0.61^{Ab} (0.09)
BB	0.54^{Aa} (0.08)	0.56^{Aab} (0.12)	0.54^{Aa} (0.07)	0.54^{Ab} (0.06)
CN	0.49^{ABa} (0.07)	0.43^{Bb} (0.06)	0.50^{Aa} (0.05)	0.45^{ABa} (0.08)
AB	0.46^{Aa} (0.07)	0.46^{Aab} (0.04)	0.46^{Aa} (0.08)	0.43^{Aa} (0.03)
RV	0.45^{Aa} (0.06)	0.47^{Aab} (0.08)	0.49^{Aa} (0.08)	0.53^{Ab} (0.05)

x = Values with the same lowercase letters in the same column are not significantly different between materials, y =Values with the same uppercase letters in the same row are not significantly different between environmental pH

TABLE 5. Mean loss tangent values ($\times 10^{-3}$) of the various BRMs (standard deviations in parentheses)

Materials [x] \ Environmental pH [y]	pH 3.0	pH 5.0	pH 6.8	pH 10.0
FB	93.00^{Aa} (8.06)	101.80^{Aa} (8.61)	101.60^{Ab} (16.11)	103.00^{Aab} (14.46)
BB	93.10^{Aa} (6.79)	94.20^{Aa} (13.33)	85.10^{Aa} (5.57)	85.90^{Aa} (6.19)
CN	116.90^{B} (5.38)	95.50^{Aa} (3.81)	107.40^{ABb} (7.60)	111.60^{Bb} (7.04)
AB	162.70^{A} (5.40)	132.30 (4.35)	159.60^{A} (10.34)	164.50^{A} (9.12)
RV	77.30^{B} (4.90)	78.30^{AB} (7.09)	81.40^{ABa} (6.20)	88.30^{Aa} (4.85)

x = Values with the same lowercase letters in the same column are not significantly different between materials, y =Values with the same uppercase letters in the same row are not significantly different between environmental pH

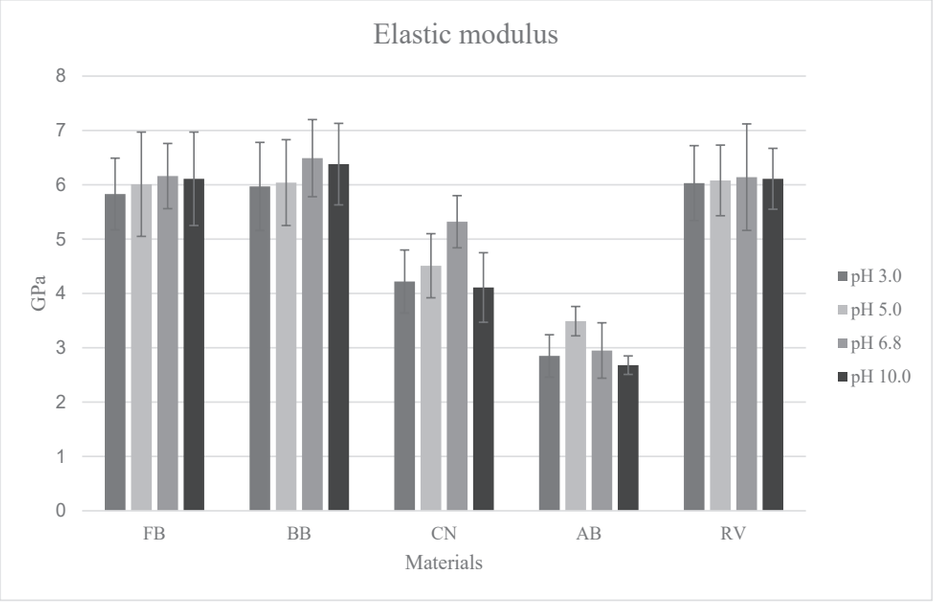

FIGURE 1. Mean elastic modulus (GPa) of different materials at when conditioned in all four mediums

ELASTIC MODULUS

Mean elastic modulus varied from 2.68 ± 0.17 to 6.49 ± 0.71 GPa. AB in pH 10.0 presented the lowest mean value, while BB in pH 6.8 presented the highest. When comparing the various immersion mediums for individual BRMs (Table 3), pH 3.0 and 10.0 generally yielded the lowest elastic modulus of all BRMs, whereas immersion in pH 6.8 resulted in the highest elastic modulus, except in AB. Within AB, the immersion in pH 5.0 had yielded the best elastic modulus compared to other environmental pH's. When comparing the mean elastic modulus of different BRMs in the same immersion mediums, AB

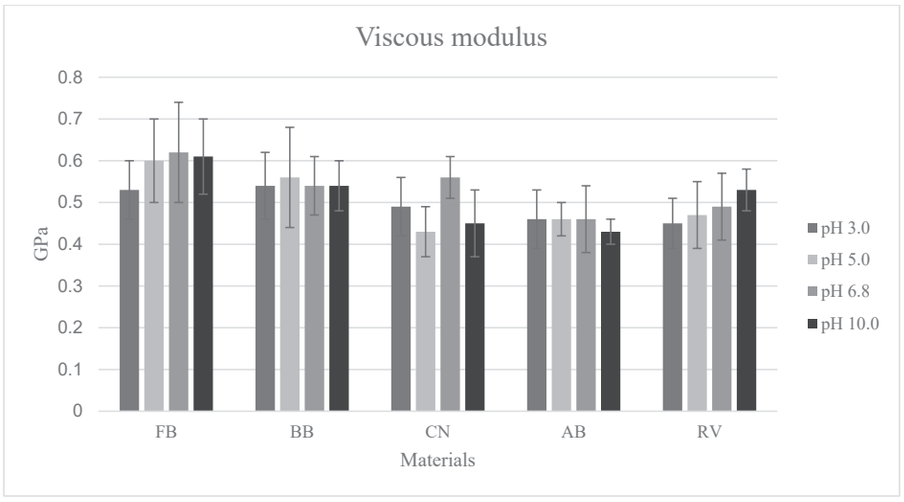

FIGURE 2. Mean viscous modulus (GPa) of different materials at when conditioned in all four mediums

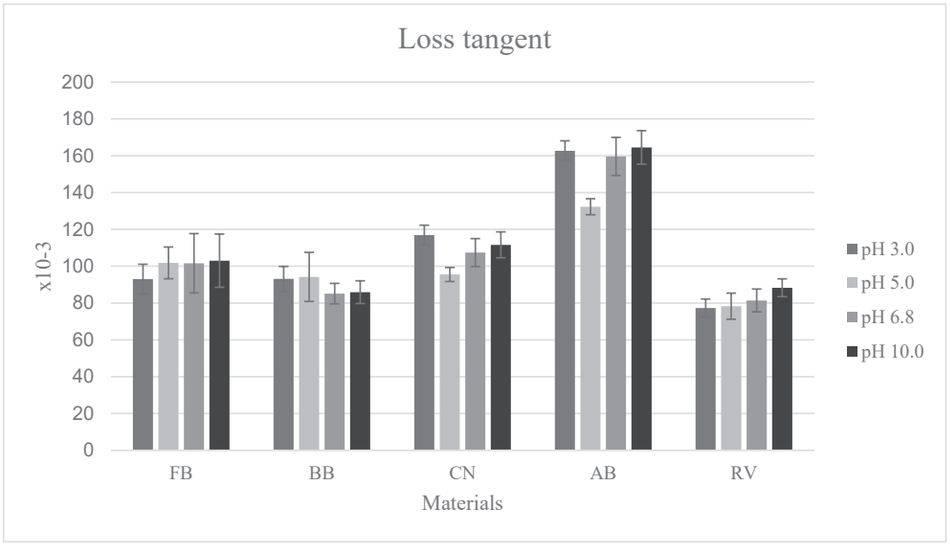

FIGURE 3. Mean loss tangent ($\times 10^{-3}$) of different materials at when conditioned in all four mediums

performed significantly the poorest in all mediums, followed by CN, while FB, BB, and RV were comparable.

VISCOUS MODULUS

Mean viscous modulus ranged from 0.43 ± 0.03 to 0.62 ± 0.12 GPa. AB in pH 10.0 and CN in pH 5.0 exhibited the lowest viscous modulus, while FB in pH 6.8 presented the highest. In terms of impacts of different environmental pH's within similar BRMs (Table 4), mean viscous modulus across all pH's seemed to be comparable, except CN. When exposed to pH 6.8, it was significantly greater than the others. When the mean viscous modulus of different BRMs in the same immersion mediums are compared, RV in pH 3.0, CN in pH 5.0, AB in pH 6.8 and 10 exhibited the lowest values, while FB appeared to be greater than other BRMs except in pH 3.0. Overall, the mean viscous modulus of all materials was comparable, except CN in pH 5.0 and AB, CN in pH 10.0, where they were significantly lower than the others.

LOSS TANGENT

Mean loss tangent differed from 77.30 ± 4.90 to 164.50 ± 9.12. The lowest loss tangent was observed with RV in pH 3.0, while AB was highest in pH 10.0. Immersion mediums of different environmental pH's within the same BRMs showed comparable results among all (Table 5), except CN, AB and RV. As for comparison of different BRMs in the same immersion mediums, AB was significantly greater than the others in all immersion mediums, while RV recorded significant lower mean loss tangent values in pH 3.0 and 5.0.

DISCUSSION

This study determined the effects of environmental pH's on the viscoelastic properties of BRMs using dynamic mechanical analysis. Given that the viscoelastic properties of BRMs were affected by their chemical environments and differences were seen between inter-mediums and inter-materials across the 7 days of immersion, the initial null hypotheses were duly rejected.

The period of artificial aging of 7 days was chosen to allow post-irradiation hardening of the resin composite and stabilization of the acid-base reaction of glass ionomer cement (Marghalani 2010; Yap et al. 2005). Resistance against acidic/alkaline attacks of BRMs was investigated by titration of hydrochloric acid to mimic regurgitation of gastric acid (pH 3.0) and lactic acid to mimic the acidic environment induced by cariogenic bacteria (pH 5.0) into artificial saliva. Whereas titration of sodium hydroxide (pH 10.0) was to simulate ingestion of alkaline beverages, such as mineral water and green tea (Attin et al. 2012; Fuss et al. 2017; Moon et al. 2015). Both pH and TA are indicators of the erosive potential of acidic solutions. However, TA may be a better predictor for erosive potential (Valinoti et al. 2008). Nevertheless, both pH and TA were found to be proportional. Therefore, both are viable indicators to predict the erosive potential of acidic aqueous solutions.

Elastic modulus represents stiffness and rigidity, and viscous modulus represents a material's ability to flow and stress dissipation. Loss tangent indicates the relative degree of energy dissipation of material and the ratio of viscous modulus to elastic modulus (Ong et al. 2018). Ideally, restorative materials should possess high elastic and low viscous modulus to resist great functional stress under cyclic masticatory force, especially at high stress-bearing areas, such as posterior regions. On a side note, materials with high viscous modulus may be advantageous in delaying material failure via dissipation of accumulated functional stress in the form of heat energy. However, the mean viscous modulus values across all materials observed in this study were relatively small to be of any significance. In the aspect of loss tangent, a lower value would indicate a more 'elastic' behavior, thus enabling a faster return to its original state after stress removal. A higher value would indicate a more 'viscous' behavior, enabling a higher release amount of energy in heat form (Mesquita et al. 2006).

Immersion in pH 3.0 and pH 10.0 yielded the least elastic modulus of all tested materials. Hence, it shows that the effects of an alkali medium are equally destructive as a highly acidic environment. As the catalytic reaction rates are acidity and alkaline dependent, magnitudes of pH can be positively correlated to the severity of mechanical degradation by the infliction of hydrolysis of the resin matrix (Prakki et al. 2004). Meanwhile, immersion in artificial saliva (pH 6.8) had produced the best elastic modulus for all materials, except AB. According to manufacturers, AB contains patented bioactive shock-absorbing rubberized ionic-resin and bioactive glass. These components enable acid neutralization and apatite formation by releasing/recharging high amounts of calcium, phosphate and fluoride ions when facing an acidic challenge. Based on this study, AB performed better in the mimicked cariogenic environment of pH 5.0 than other immersions of different environment pH. The latter could be attributed to the ionization process between protonated hydrogens in acidic solution and the release of therapeutic ions to form a mineral apatite-like complex on the surface layer (Wang & Yap 2009). The formation resin-hydroxyapatite complex may have fortified AB and thus yielded a significantly higher elastic modulus value in an acidic media. However, this finding should not be hastily concluded as there was no scanning electron micrograph (SEM) investigation to confirm the existence of such a mineral complex.

In terms of elastic modulus, AB performed significantly the poorest, followed by CN. Based on the positive correlation between elastic modulus and percentage of filler by weight, this may reasonably explain why the least filled AB had the lowest elastic modulus, while the more heavily filled RV and BB had higher elastic modulus (Mesquita et al. 2006). Between CN and FB, FB had a higher elastic modulus, although CN is comparatively heavier in filler weight. This compelling finding concurred with the works of Yap et al. (2020), in which the better performance of FB may be accredited to its stability of material formulation and resin modification. Viscous modulus of FB, BB, and RV in pH 10.0 were significantly greater than CN and AB.

Prolonged exposure to alkaline media has been reported to cause crack formation between filler particles and resin matrix, partial dissolution and exfoliation due to excess hydroxyl ions (Bagheri et al. 2007). This phenomenon may first increase the inter-filler particle space and then promote inter-filler particle slippage and subsequently increase material flowability and dissipation of heat energy (Boparai & Singh 2018). Generally, the mean viscous modulus of FB and BB was higher than the others in all immersion mediums. Due to continuous immersion, fluid uptake may have induced unreacted components to leach from the resin matrix (Sideridou & Karabela 2011). It can be implied that FB and BB were more prone to filler leachability regardless of chemical environments. As for loss tangent, RV was lower than the others, implying they were showing a more elastic behavior. Conversely, FB, BB, CN and AB were exhibiting a more viscous behavior.

Like any other *in-vitro* study, this work had its limitations. In the present study, all BRMs were continuously immersed for 7 days. The subjection of materials in artificial saliva seems plausible, but constant exposure to a high acidic and alkaline environment may not be clinically realistic. However, extensive exposure to an acidogenic environment can occur in patients with gastroesophageal reflux disease, bulimia nervosa and restorative materials with plaque retentive margins (Aframian et al. 2010; Chenicheri et al. 2017). The duration of the immersion period can also be prolonged to signify the aging resistance of evaluated restorative materials. Other than mechanical evaluation, aging resistance also correlates to the prediction of clinical longevity of restorative materials (Yilmaz & Sadeler 2018). Lastly, the beneficial effect of mineral complex formation on the elastic modulus of BRMs also warrants further investigations.

Conclusion

Within the limitation of the study, the following conclusion could be made. The viscoelastic properties of BRMs were found to be material and immersion medium dependent. AB presented the lowest elastic modulus across all immersion mediums of different environmental pHs. Overall, all materials tend to yield better elastic modulus when immersed in pH 6.8, except AB. When exposed to cariogenic environments, the elastic modulus of AB is found to be improved.

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