Assessment of the Effect of Acid and Base Cycling on Mechanical Properties of Various Esthetic Restorative Materials

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

Original Research Article

ABSTRACT

Objective: The purpose of this study is the evaluation of the effect of pH cycling, including both acidic and alkaline environments, on the mechanical properties of tooth-colored restorative materials.
Methods and Materials: 20 rectangular bar specimens of one bulk-fill restorative composite, two conventional nanohybrid restorative composites, and one restorative resin-modified glass ionomer were produced according to ISO 4049. Half of the materials were stored in an acid and base cycling defined as two-day storage in acidic (pH =4) and alkaline (pH=8) solutions. The rest of the materials were incubated in distilled water as a control group. The storage lasted for 48 days.

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Finally, flexural strength, elastic modulus, and microhardness of the specimens in each group determined. Data analyzed with Kruskal-Wallis, Dunn, MANOVA, Tukey HSD and T-test.

Results: The pH cycling model had a significant influence on all mechanical properties of the bulk-fill restorative composite and resin-modified glass ionomer than those stored in water ($P < 0.05$). One of the conventional nanohybrid restorative composites showed a significant reduction in elastic modulus and microhardness while the other one showed a significant reduction only in flexural strength.

Conclusion: pH cycling negatively affects the mechanical properties of resin composites, and the materials' composition is an important factor in the degradation of the resin-based materials examined.

1. INTRODUCTION

Tooth-colored restorative materials are primarily used restorative materials in dental practice [1], and improving their clinical performance is a focal point of new research. In this regard, one important aspect is increasing hydrolytic stability. Chemical hydrolysis of tooth-colored restorative materials occurs in water, which hydrolyzes the condensation type bonds [2]. Two types of categories influence the hydrolysis rate. The first category includes factors that are related to the environment like acids [3], bases [4], enzymes [5] and etcetera. The second category relates to the structural factors of the tooth-colored materials like the type of the resin matrix [6], filler content [7], filler type [8], and filler size [9].

Since evaluating all these factors' simultaneous effects is difficult in the laboratory, many researchers have to reproduce a restricted form and work on models. Some researchers use a acid and base cycling that usually contains acidic pH since most of the beverages have pH below 4 [10], and regular intake of fermentable carbohydrates leads to acidic pH as low as 3.9 in the dental biofilm [11]. There is conflicting evidence whether the effect of hydrogen ion on hydrolysis rate is primarily from the matrix, the filler particles, or the interfacial-coupling agent. According to the study which is done by Soderholm et al., hydrogen ions possibly accelerate ion exchange at the filler surface. Tooth-colored resin restorations, when are stored in water, leach Alkaline-earth elements such as Strontium and Barium. Leaching of these positive ions leaves the remaining Si-O-Si structures negatively charged, and the charge imbalance could delay the release of more ions on the surface. However, positive ions such as H^+ in the environment could help neutralize the negative charges and facilitate further leaching of glass modifiers [12,13].

Evaluation of the effect of an alkaline environment on tooth-colored restorative materials is not sufficient. However, it has been proposed that leaching of glass modifiers results in OH ion production at the resin-filler interface. Charles proposed that this ion hydrolyzes the silanol bonds and makes the reaction autocatalytic [13]. It is reported that the pH of this alkaline environment for barium-containing glasses at the filler interface is 9.3 [4].

Therefore, OH and H ions theoretically have different action mechanisms on dental resin materials. This phenomenon is a complex process that may lead the composite polymer matrix to collapse, causing several problems such as filler-polymer matrix debonding, release of residual monomers, and wear and erosion caused by food, chewing which affecting dental resin restorations' mechanical properties [12-14].

However, most studies have not evaluated the effect of different pH mediums on dental resin materials' mechanical properties. Most studies had been confined to surface roughness, microhardness, water sorption, and solubility [4,14-16]. In this way, the present research aims to evaluate the interaction between resin-based restorative materials, and acid and base cycles on their mechanical properties. The null hypothesis tested were no differences in the flexural strength, elastic modulus, and microhardness of recent tooth-colored restorative materials after storage in different pH media over time.

2. METHODS AND MATERIALS

2.1 Preparation of Samples

Three resin-composite materials, including one bulk-fill and two conventional materials, and one resin-modified glass ionomer are tested. Product specifications are presented in Table 1.

Table 1. Product specifications of materials evaluated

Materials	Abbreviation	Manufacturer	Shade	Filler Weight (%)	Filler type	Filler size (μ)	Matrix
Tetric N-Ceram Bulk-Fill (nanohybrid composite resin)	TNB	Ivoclarvivadent	IVA	75-77	barium glass, prepolymer filler	0.04to 3	Bis-GMA, UDMA
Grandio (Universal nanohybrid composite resin)	GR	Voco	A2	87	silica glass	0.02 to 1	TEGDMA, Bis-GMA
Clearfil APX (universal, nanohybrid composite resin)	CL	Kuraray	A2	86	Silanated barium glass, Silanated silica, Silanated colloidal silica	0.02 to 17	TEGDMA, Bis-GMA
Fuji II LC GC (Resin modified glass ionomer cement)	RMGI	GC America	A2	Distilled water, polyacrylic acid, 2-hydroxyethylmetacrylate, urethane dimethacrylate, camphorquinone, fluoroaluminosilicate filler			

A rigid transparent acrylic barwas prepared in 2×2×25 mm dimensions according to ISO 4049:2019 by water cooling three-axis CNC milling machine. A mold was then made by pressing the plastic specimen into the silicone impression putty (Speedex, Coltene).

Twenty rectangular bar specimens of each tooth-colored restorative material were prepared at room temperature (23±2°C). The mold was slightly overfilled with uncured material in a single increment. A polyester strip with a glass slab was placed over the mold to obtain a flat surface and standardize the curing distance by placing the light tip in contact with the glass. As the specimen length was 25 mm, and the light exit diameter was 5 mm, each specimen was polymerized in five overlapping sections for 20 seconds per section in a pulse curing mode. The calibrated light-curing unit (CL37, Jerry, China) had an irradiance of 1000 mW/cm2, and wavelength ranges were about 420–480 nm. After curing, each specimen was gently removed from the mold, and specimens with any cracks or voids were replaced. The excess minor materials were removed carefully by a sharp blade at the periphery, and no polishing was performed. Final specimens were pre-stored in distilled water at room temperature for seven days.

2.2 Acid and base Cycling

The specimens were then randomly divided into experimental and control groups (n= 10). The experimental group was stored in a pH cycling, and the control group was immersed in distilled water. The pH cycling was defined as two-day storage in acidic solution (pH=4) and two-day storage in alkaline solution (pH=8). After every two days, solutions were refreshed, and samples in the experimental group were rinsed with distilled water before entering the next solution. The pH solutions were prepared based on the citrate-phosphate buffer (McIlvaine's Buffer Solution) described by Ortengren et al. [15]. Specimens were kept in sealed glass containers in an incubator at 37 °C for 48 days.

2.3 Mechanical Properties Evaluations

After storage, a universal testing machine (SANTAM STM-20) in three-point bending mode

was used to measure the flexural strength (FS - MPa) and flexural modulus (FM - GPa) of all samples until fracture occurred. The crosshead speed was 1 mm/min, and the test span was 15 mm. Flexural strength (σ) and elastic modulus (E) were calculated as follows:

$$\sigma = 3PL/2b\text{h}^2$$

Where P is the peak load from the load–displacement curve in N, L is the span in mm, b is the specimen width in mm, and h is the specimen height in mm.

$$E = PL^3/4bh^3d$$

Where P is the maximum load in N, L is the span in mm, b is the specimen width in mm, h is the specimen height in mm, and d is the deflection corresponding to the load P in mm.

Microhardness measurements were performed on the samples' top surface by a diamond pyramid indenter using a Vickers microhardness tester (MH3, Koopa, Iran). It was applied 0.98N load for 10s on the upper surface, and the average value of the two indentation diagonals was measured using a microscope. Two measurements were takenand averaged. All measurements were carried out at a constant room temperature.

2.4 Statistical Analysis

Statistical analyses were performed using SPSS Statistics 21.0 and Microsoft Excel 2016. Assuming four types of materials and two storage mediums (distilled water and pH cycling), mechanical properties data were analyzed for eight groups. The normality of data distribution was verified by Shapiro-Wilk. Experimental groups' data were not homoscedastic. Therefore, Kruskal-Wallis was applied to determine the mean values of mechanical properties, and Dunn's test was applied for pairwise comparison of materials' mechanical data. Control groups' data were homoscedastic. Therefore, MANOVA was used to determine the mean values of mechanical properties, and Tukey HSD was used for pairwise comparison of materials' mechanical data. In all analyses, the level of significance was justified at $p < 0.05$.

3. RESULTS

Table 2 shows the mean flexural strength, elastic modulus, and microhardness of each group after 48 days storage in water or pH cycling. Grandio showed a significant reduction in flexural strength, while Clearfil APX showed a significant reduction in elastic modulus and microhardness. All mechanical properties of Tetric N-Ceram Bulk-Fill and RMGI reduced significantly during pH cycling than those stored in distilled water. Additionally, all mechanical properties of TNB were more susceptible to pH cycling than conventional resin composites.

Table 2. Mean values and standard deviation for flexural strength, elastic modulus, and microhardness in experimental and control groups

groups	Flexural strength (MPa)	Elastic modulus (GPa)	Microhardness (VH)
		Mean ($\pm$SD)	
TNB	51.72(15.54)[C]	31.77(5.41)[C]	2041.12(1315.50)[C]
TNB (control)	94.73(11.12)[c]	50.89(3.61)[c]	4042.94(1212.61)[c]
GR	114.34(8.67)[A]	111.26(8.42)[A]	6522.71(1124.39)[A]
GR (control)	132.79(18.34)[a]	107.64(8.36)[A]	6407.75(1225.67)[A]
CL	86.31(17.52)[B]	52.8(4.11)[B]	2831.38(628.43)[B]
CL (control)	87.34(19.63)[B]	79.73(3.16)[b]	4175.02(757.88)[b]
RMGI	13.5(13.34)[D]	14.07(2.85)[D]	1151.71(328.23)[D]
RMGI (control)	56.99(14.31)[d]	33.48(2.81)[d]	2766.68(660.93)[d]

According to Kruskal-Wallis and MANOVA tests, same superscript upper case letters indicate no statistical significance among values for each used material. SD (standard deviation), TNB (Tetric N-Ceram Bulk-Fill), GR (Grandio), CL (Clearfill APX ES-2), RMGI (Resin modified glass ionomer)

4. DISCUSSION

As shown by the present findings, all mechanical properties of TNB were more susceptible to pH cycling than conventional composites. TNB has lower filler content and contains barium glass and pre-polymerized fillers. Pre-polymerized fillers may facilitate water and ion transportation inside dental resin materials. Water sorption results in resin matrix swelling, but the swelling of the pre-polymerized fillers is retained. Since the bonding between the pre-polymerized fillers and the matrix may not be optimized, gaps will open, acting as pathways and letting the interaction between internal fillers and the surrounding medium [17]. There is also evidence that water sorption of dental resin restorations is mainly associated with the polymeric matrix, and tooth-colored resin materials with lower filler content absorb more water than the opposite [6]. It was reported that higher filler content could improve composites' resistance against acidic mediums. However, a linear relationship could not be observed and, filler types, filler distribution, and surface treatment of fillers were also important factors [7]. Another important consideration is that the TNB resin matrix is more hydrophobic than Grandio and Clearfill APX resin matrixes. In previous study conducted a comparison between all these factors. They compared water sorption of Tetric Bulk-Fill and Grandio in distilled water for up to a year. It was found the general water sorption of Tetric Bulk-Fill was statistically greater than Grandio water sorption, which could be attributed to its lower filler content [6] . Their results concur with those of Alrahlah et al. who conducted an almost identical study [18]. The flexural strength of GR and elastic modulus and microhardness of CL reduced significantly in the pH-cycling model than those of the distilled water. Therefore, it can be proposed that CL had been more susceptible to pH challenge than GR. CL and GR have nearly identical structures except in the filler type, in which GR contains silica glasses purely, while CL contains silica glasses plus Barium glass modifiers. There is evidence that glass modifiers often reduce filler particles' hydrolytic stability [8,13]. They accelerate siloxane bond hydrolysis. Alkaline-earth elements incorporated in fillers leach from the filler particles, react with water, and make the surroundings alkaline. When a particular hydroxyl ion concentration is reached, these hydroxyl ions could start attacking the silica network and make the siloxane bond hydrolysis automatic [13]. However, more studies showed that the storage medium's effect is more pronounced

than the effect of filler type [19]. Continuity of the explained mechanism is dependent on the ion exchange with the environment. Leaching of Alkaline-earth elements forms negative charges within the silicone network. Such negative charges could delay further leaching of these ions retained inside the network until a charge balance is re-established. Such a charge balance could occur if positive ions diffuse from the environment to the filler surface and neutralize the negative charges. Thus by neutralizing the filler particle charges, Alkaline-earth elements otherwise retained within the filler particles would be more readily available for further release [12]. The present study used pH cycling, containing both acidic and alkaline environments. Based on the above explanations, it can be assumed that hydrogen ions in an acidic medium accelerate the leaching of barium from CL since they penetrate the spaces previously occupied by barium and maintain the charge balance inside the silica. However, theoretically, it seems that in an acidic medium, the autolytic degradation of CL and GR glass fillers by hydroxyl ions stops. Contrariwise, in an alkaline medium, the charge imbalance restricts the leaching of barium from CL. A million times as many hydroxyl ions as would be produced from barium glasses are present in the solution, and the autocatalysis of CL and GR siloxane bonds by hydroxyl ions intensifies. Therefore, surface fillers of tooth-colored resin materials become improperly bonded gradually, and as a result of such a debonding, the mechanical properties will decrease substantially. A previous research evaluated the effect of filler type and filler size on the flexural strength of composites and showed the flexural strength of barium-containing resin decreased after water immersion more than that of the quartz-containing resin as the particle size decreased [8]. Several studies report similar findings regarding the lowering effect of glass modifiers on resin-based restorative materials' different physical properties [4,15,20]. Resin modified glass ionomer had the lowest mechanical data in both experimental and control groups than composite resins. Their mechanical properties reduced significantly in the acid and base cycling than in distilled water. These findings could be explained by differences in the physical and chemistry of resin-modified glass ionomers and resin composites. Resin-modified glass ionomer consist of aluminum fluorosilicate glasses and modified polyacrylate networks cross-linked by poly-HEMA. It is proposed that the coherence between the polyacrylate acid network and the poly-HEMA may be insufficient,

and gaps will form as a result of phase separation [21]. Besides, the poly-HEMA is a water-based resin system that contains a high proportion of hydrophilic functional groups and takes up a lot of water [22]. Therefore, water and H+ and OH- ions can more easily diffuse into the matrix and provoke ionic species' dissolution from the glass particles. Consequently, there is a gradual reduction in the mechanical properties of RMGI. Moreau et al. compared flexural strength and elastic modulus changes of Fuji II LC and Heliomolar in lactic (pH=4) or acetic (pH=5.5) acids after 84 days. Results showed that immersion decreased mechanical properties, significantly for Fuji II LC compared to Heliomolar' [23]. Changes in the RMGI microhardness coincide with other studies evaluating RMGI microhardness changes in acidic mediums [24-26]. However, Gomec et al. evaluated surface microhardness of different tooth-colored restorative materials in different acidic solutions and showed contrary results. The microhardness of composite reduced in all acidic media; however, the microhardness of RMGI increased in orthophosphoric acid. Authors have attributed the higher hardness values to the deposition of insoluble salts like calcium phosphate and aluminum phosphate with high ionic bonding and low solubility on the surface of RMGI [27]. However, in the present study, citrate-phosphate buffer solution was used, and citric acid has large complexing ability with metal actions that may inhibit insoluble salts' deposition on RMGI. Nicholson et al. evaluated gain in the mass of different restorative materials in different acidic solutions and showed that citric acid was an especially aggressive medium towards these materials, particularly for glass-ionomer cement [28]. Regarding the effect of alkaline medium on glass ionomers, two studies were evaluated, and the results suggested that glass ionomers are more prone to alkaline medium than acidic medium. Yanikoğlu et al. studied the solubility of different dental cement in different pH mediums for a month. The solubility of glass polyalkenoateionomer (Voco, Germany) in alkaline medium (pH=9) was higher than that of acidic medium (pH=3) [29]. Bagheri et al. evaluated surface degradation of composites and compomers stored in different pH mediums and found that degradation of all materials in all chemical media, but to a significantly greater depth of degradation after exposure to NaOH media [20]. Limitations of this study were non-inclusion of other essential factors such as saliva, temperature and loading on the chemical hydrolysis of resin-based restorative materials to create a more clinically relevant testing environment. Therefore, future clinical trial research accompanies other aging factors should consider the evaluation of newly introduced restorative materials. Further in vitro studies should be carried out to address more tests to predict the clinical performance of various restorative materials over time.

5. CONCLUSION

In light of the presented results, it can be concluded that pH cycling negatively affects the mechanical properties of resinous restorative materials. However, the extent of the effect depends on the materials' composition. Filler loading and incorporation of glass modifiers and pre-polymerized fillers are some structural factors of materials that could have been important impact in mechanical properties.

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CONFLICT OF INTEREST

The authors of this article certify that they have no proprietary, financial, or other personal interest of any nature or kind in any product, service, and/or company that is presented in this article.

CONSENT

It is not applicable.

ETHICAL APPROVAL

It is not applicable.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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