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Changes in the physical properties and color stability of aesthetic restorative materials caused by various beverages

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This study investigates the effects of various beverages on the wettability, microhardness, and color stability of aesthetic dental restorative materials. A contact angle analyzer, Vickers hardness tester, and spectrophotometer were used to characterize the properties of the materials and a total of 225 specimens were prepared: 75 each for a resin composite, compomer, and giomer. Ingestion of energy drinks and cola caused the greatest deterioration in wettability and microhardness, and coffee caused the most significant color change. In addition, the change in the resin composite was lower than that of the other restorative materials. The extent of change in the restorative materials increased with duration and frequency of contact with the beverages, so a reduction in the frequency of ingestion of these beverages is recommended.

Keywords: Aesthetic restorative dentistry, Beverage, Color stability, Microhardness, Surface wettability

INTRODUCTION

Commonly used aesthetic restorative materials in dental clinics include resin composites, compomers, and giomers. The resin composite was created in 1962 by Bowen through the development of Bis-GMA[1)], and subsequent addition of an organic filler. Although resin composites possess good physical properties, their fluorine release ability is limited, which may lead to the development of secondary caries[2)].

Compomers (polyacid-modified resin composites) were originally developed by mixing a polyacid modified resin with a glass filler, which contained fluorine[3)]. However, compomers release less fluorine compared to glass ionomers and have a shorter release time and inferior physical properties compared to resin composites[4)].

Based on pre-reacted glass (PRG) technology, Shofu has developed "giomers", which are fluoride-releasing combination glass ionomer-polymer mixtures containing a PRG filler and a resin matrix[5)]. This material incorporates the fluorine glass properties of glass ionomers and the physical properties of resin composites, overcoming the disadvantages of both constituent materials[6,7)]. Furthermore, giomers have the ability to neutralize acid due to the compomer, preventing secondary caries[8)].

Aesthetic restorative materials require outstanding physical and mechanical properties, as well as color stability. According to Guler *et al.*, consuming coffee, tea, and wine as well as smoking causes extrinsic color changes in aesthetic restorative materials[9)]. According to Dietschi *et al.*, physiochemical stress induces disintegration of the surface of restorative materials and makes them more susceptible to color change[10)]. In addition, frequent ingestion of low pH beverages has been demonstrated to corrode teeth[11-13)]. As the consumption of acidic beverages has increased in conjunction with the demand for aesthetic restorative materials, there have been continuous developments in restorative materials. However, there is a lack of integrated short-term research regarding the influence of acidic beverages on various aesthetic restorative materials from a non-destructive perspective. According to the study by Badra *et al.*, after 7 days of immersion in a beverage, the restorative material was more stable in terms of microhardness than after 30 days[14)]. In addition, Dharrab studied energy drinks and their effect on the discoloration of resin composites after 1, 7, 30, and 60 days[15)]. The surface hardness of different restorative materials after long-term immersion in sports and energy drinks was measured by Erdemir *et al.*, who found that the surface hardness of the resin composite decreased significantly after 1 and 6 months of immersion[16)]. In addition, Tanthanuch *et al.* showed that the greatest change in microhardness occurred within 7 days of immersion[17)]. However, it was thought that the microhardness change within one week was insufficient, so the study was also conducted with a 5 day immersion period. Previous studies have found varying degrees of deterioration after one week, and we sought to directly observe changes occurring in the beverage and restorative materials in this short-term study.

This study examined the effect of water, cola, orange juice, coffee, and an energy drink on the wettability, surface hardness, and color of commonly used aesthetic restorative materials (resin composite, compomer, and giomer) as a function of exposure time. Furthermore, the effects of the immersion on the various restorative materials were compared. The null hypothesis of this study is that "the tested beverages will not affect the wettability, microhardness, or color of aesthetic restorations".

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MATERIALS AND METHODS

Research materials

Cola (Coca-Cola, Coca-Cola Co., Gyeonggi-do, Korea), orange juice (Delmonte premium orange juice100, Lottechilsung, Seoul, Korea), coffee (Cantata Americano, Lottechilsung), and an energy drink (Hot6, Lottechilsung) were selected as the test beverages. Mineral water (Jeju samdasoo, Jeju Province Development, Jeju, Korea) was used as a control. The aesthetic restorative materials used were Filtek Z250 (3M ESPE, St. Paul, MN, USA) as a resin composite, Dyract XP (Dentsply De Trey, Konstanz, Germany) as a compomer, and Beautifil II (Shofu, Kyoto, Japan) as a giomer (Table 1). All materials corresponded to the tooth shade A3.

Sample fabrication

For the fabrication of the resin composite, compomer, and giomer, disc-shaped acrylic molds 6 mm in diameter and 2 mm in height were prepared. Each material was placed in the mold, and an OHP film and glass plate were used to cover the outside of mold so that the formation of bubbles through pressurization could be suppressed and excess material removed. Following the manufacturer's instructions, LED light curing was conducted for 20 s for the resin composite, and 10 s for the compomer and giomer (on their front and back surfaces) using a LED light curing unit (Eliper Free Light 2, 3M ESPE; light intensity of 650 MW/cm^2). To form an even surface, the sample was polished with a polisher (CC261#2000, Deerfos, Seoul, Korea). The tests measured 5 samples per group, 75 samples per experiment, and a total of 225 samples.

Processing of samples

All samples were stored in an incubator (forced convection incubator, JISICO, Seoul, Korea) at 37°C during the experiment, and were submerged in the beverages for 3 h per day for 5 days. The wettability, surface hardness, and color stability of each aesthetic restorative material were measured before submersion in the beverages, and after 1 and 5 days of submersion. The carbonated beverages (cola and energy drink) were stirred for over 1 h, when before material submersion. The samples were stored in distilled water when not submerged and the beverages were replaced every day.

pH measurement of the beverages

To measure the pH of the experimental and control groups, 4 mL of each beverage was removed using a plastic centrifugal tube, and its pH was measured with a pH meter (Orion star series meter, Thermo Scientific, Waltham, MA, USA). Before the experiment, the pH meter was adjusted with standard solutions, and before each measurement distilled water was used to wash the electrode. The pH of each beverage was measured

Table 1 Components of the materials used in this study

Product	Type	Composition	Manufacturer
Filtek Z250	Resin composite	Bis-GMA, UDMA, Bis-EMA, TEDGMA, Zirconia/Silica filler	3M ESPE, St. Paul, MN, USA
Dyract XP	Compomer	UDMA, TCB, TMPTMA, TEGDMA, Strontium-alumino-sodium-fluoro-phosphor-silicate glass	Dentsply De Trey, Konstanz, Germany
Beautifill II	Giomer	Bis-GMA, TEGDMA, Fluoroboro-alumicosilicate glass	Shofu, Kyoto, Japan
Coca-cola	Cola	Purified water, High frustose corn syrup, White sugar, Carbonic-acid gas, Phosphoric acid, Caffeine	Coca-cola, Gyeonggi-do, Korea
Delmonte premuium orange juice 100	Orange juice	Purified water, Calcium Lactate, Vitamin C, Dl-a-tocopherylacetate, Maltodextrin, Silicon dioxide, Calcium pantothenate, Vitamin B6, Hydrochloride	Lottechilsung, Seoul, Korea
Cantata Americano	Coffee	Purified water, White sugar, Sodium bicarbonate, G-sodium ascorbate, Coffee solid content 0.672%	Lottechilsung
Hot6	Energy drink	Purified water, High fructose corn syrup, Carbon dioxide, Guarana extract, Taurine, Citric acid, Sodium citrate, Vitamin C, Siberian ginseng extract concentrate, Tea extraction powder, Inositol, Red ginseng concentrate	Lottechilsung
Jeju samdasoo	Water	Mineral	Jeju province development, Jeju, Korea

*Bis-GMA: bisphenol-A-glycidyl methacrylate, UDMA: urethane dimethacrylate, TEGDMA: triethyleneglycol dimethacrylate, Bis-EMA: ethoxylated bisphenol-A-dimethacrylate, TMPTMA: trimethylol propane trimethacrylate in triplicate, and the average pH value was calculated. All beverages were left at room temperature for 6 h without being opened, to measure all samples at the same temperature, and the carbonated beverages were measured after stirring with a magnetic stirrer (Stir PC-4022, Corning, NY, USA) for 6 h.

Measurement of wettability

The wettability of the aesthetic restorative materials was determined by contact angle measurement using a contact angle analyzer (Phoenix 300, Surface Electro Optics, Gyeonggi-do, Korea) and contact angle measurement software (Image XP version 5.9, Surface Electro Optics). The measurements were conducted before submerging the samples in the beverage, and after 1 and 5 days of submersion. During the measurements, a pipet was placed 1 cm above the sample, and a 20 μL droplet of distilled water was dropped onto the sample surface. The average value of the right and the left contact angles was determined after 3 s of contact.

Measurement of the surface hardness

The surface hardness of the aesthetic restorative materials was measured before submerging the samples in the beverages, and after 1 and 5 days of submersion. The samples were placed in a Vickers hardness tester (DMH-2, Matsuzawa Seiki, Tokyo, Japan), and a 200 g weight was applied for 10 s at different points of the samples and observed under 100× magnification. The major axis length of the pressed mark was measured at a magnification of 400× to determine the Vickers hardness (VHN). Measurements were performed at five different points of each sample (center, upper, lower, left, and right points) and the average value was calculated from these measurements.

Measurement of beverage color

Each beverage was placed in a transparent Petri dish 10 mm in diameter and 2 mm in height, which was then wrapped in Parafilm to prevent leakage. The color of the beverages was measured using a spectrophotometer (CM3500-d, Minolta, Tokyo, Japan). Before the measurement, calibration was conducted with a zero-calibration box and white calibration plate, and the $L^*a^*b^*$ values were measured using a CIE Lab system under a 10° field of view, and standard light source, D65. Herein, the CIE Lab system refers to the system regulated by the International Lighting Society, and the Lab values are defined as follows. The color of water was measured, followed by that of the beverages, confirming that the L of the water was 93.73±0.07, a was −0.35±0.05, and b was −0.16±0.10.

Measurement of color after submersion in beverages

A spectrophotometer was used to observe the change in color of the restorative materials after submersion in the beverages. The values were measured before submersion, and after 1 and 5 days of submersion. Three measurements were made at different locations of each sample to obtain an average value. According to the CIE color coordinate system, the Lab values and the values of the color change (ΔE) were measured. Herein, the value of the color change was calculated according to the following color difference formula[18,19]:

$$\Delta E^*=\sqrt{(\Delta L^*)^2+(\Delta a^*)^2+(\Delta b^*)^2}$$

Statistical analysis

The wettability, surface hardness, and color change of the aesthetic restorative materials before and after submersion in each beverage were statistically analyzed using the SPSS 18.0 (SPSS, Chicago, IL, USA) program. Statistical analysis of the wettability and surface hardness were performed using one-way ANOVA tests, and the statistical analysis of the color change assessed significance through independent t-tests. Post-analysis was conducted by performing Tukey's test, and the results were defined as significant when $p<0.05$.

RESULTS

Average pH

The four beverages of the experimental group except for the water-immersed control group were acidic (Table 2). The beverage with the lowest pH was cola, while water (control group) was neutral.

Comparison of wettability according to beverage type

To measure the wettability, a drop of beverage was dropped on each restorative material, and the contact angle was measured before and after submerging the samples in the beverages (Fig. 1). In all restorative materials, the contact angles were reduced by submersion in water (the control group), and the cola, orange juice, coffee, and energy drink of the experimental groups (Table 3). After 5 days of submersion, all samples showed significant differences in their respective contact angles ($p<0.05$).

A comparison of the contact angles before and after 5 days of submersion showed that the energy drink had the largest effect on the resin composite (49.00±4.23). For the compomer, cola exhibited the greatest effect (55.49±2.37), while coffee most significantly affected the giomer sample (59.02±8.11).

Comparing the reduction in the contact angles upon beverage treatment, the resin composite had the lowest contact-angle reduction rate of 28.91±16.41, and

Table 2 pH of the beverages

Beverage	pH
Cola	1.40 (0.01)
Orange juice	3.01 (0.01)
Coffee	4.45 (0.00)
Energy drink	2.18 (0.00)
Water (Control)	7.00 (0.00)

the reduction rate of the giomer was the highest at 48.80±17.31.

Surface hardness results as a function of beverage

The surface hardness of the restorative materials was measured by examining the microhardness with a Vickers hardness tester before submerging the samples in the beverages, and after 1 and 5 days of submersion (Fig. 2). All restorative materials showed significant differences in microhardness upon submersion (p<0.05).

For the resin composite and compomer, the energy

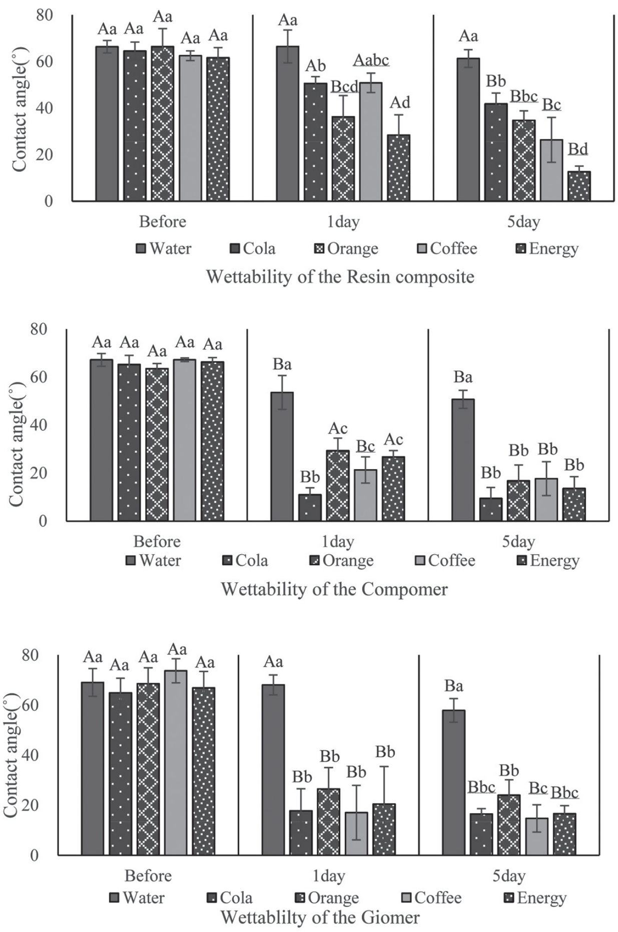

Fig. 1 Wettability of the aesthetic restorative materials.
A lowercase letter means a comparison to the same drink within each date. A capital letter means a comparison of each drink to the same date.

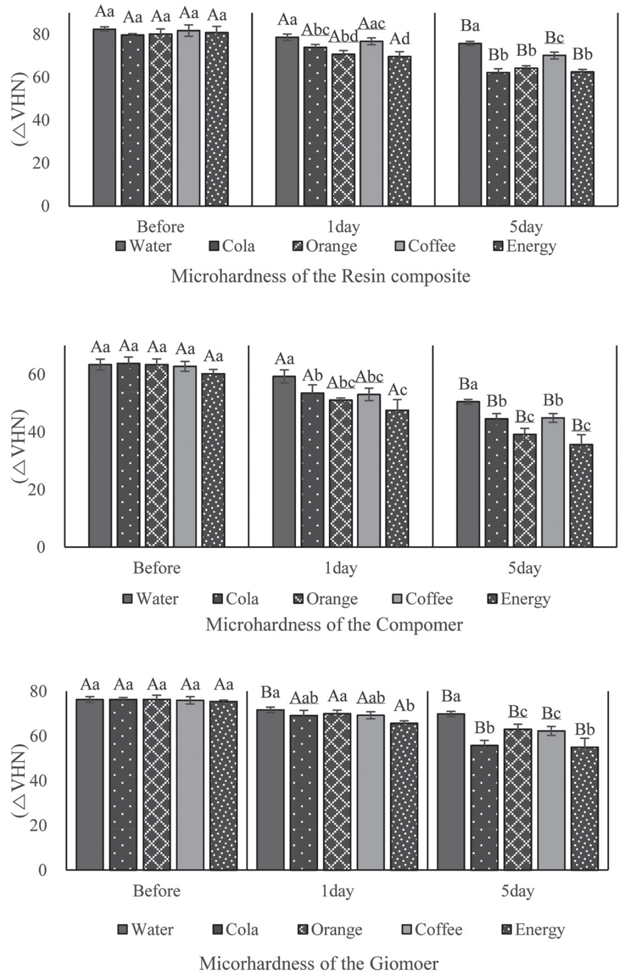

Fig. 2 Microhardness of the aesthetic restorative materials.
A lowercase letter means a comparison to the same drink within each date. A capital letter means a comparison of each drink to the same date.

Table 3 Comparison of the ΔContact angle among the beverages

Group	ΔContact angle (Before –5 day)		
	Resin composite	Compomer	Giomer
Cola	22.68 (6.95)	55.49 (2.37)	48.71 (8.63)
Orange	31.65 (9.39)	46.77 (6.53)	44.53 (4.23)
Coffee	36.13 (10.70)	49.53 (7.36)	59.02 (8.11)
Energy drink	49.00 (4.23)	52.73 (5.56)	50.30 (6.56)
Water (Control)	5.10 (3.40)	16.53 (3.79)	12.04 (7.19)

Table 4 Comparison of the microhardness reduction rates among the beverages

Group	ΔVHN (Before –5 day)		
	Resin composite	Compomer	Giomer
Cola	17.60 (1.37)	19.23 (2.79)	20.63 (1.62)
Orange	15.90 (1.58)	24.28 (3.86)	13.40 (1.94)
Coffee	11.60 (3.41)	17.90 (2.15)	13.73 (2.11)
Energy drink	18.33 (3.84)	24.60 (3.66)	20.45 (3.54)
Water (Control)	6.68 (1.89)	12.93 (1.65)	6.40 (1.36)

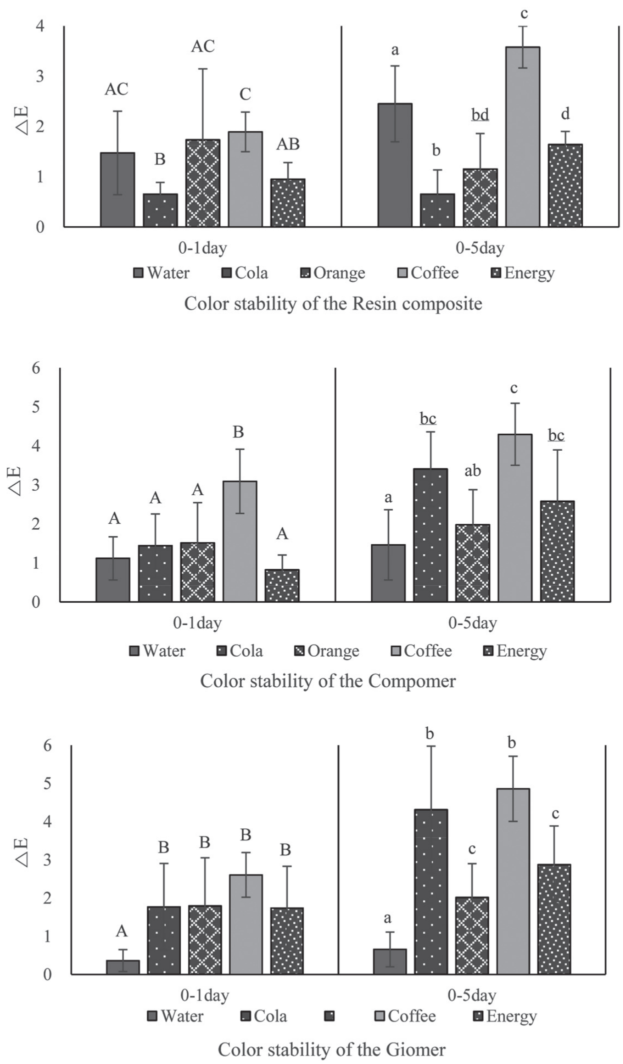

Fig. 3 Color stability of the aesthetic restorative materials.
The uppercase letter indicates a statistically significant difference for the color change rate between day 0 and 1. The lowercase letter indicates statistical significance of the color change rate between day 0 and 5.

drink exhibited the highest reduction rate (18.33±3.84), while for the giomer, submersion in cola resulted in the highest reduction rate (20.63±1.62), and the detailed results are listed in Table 4. Furthermore, comparison of the extent of surface hardness reduction revealed that the resin composite had the lowest reduction rate (14.02±5.04), while the compomer had the highest reduction rate (19.79±5.17).

Beverage-induced color changes

The color change measurements were performed using a spectrophotometer, and the values were measured before submersion, and after 1 and 5 days of submersion (Fig. 3, Table 5). The ΔE values (before and after 5 days of submersion) were 3.58±0.40, 4.30±0.80, and 4.85±0.85 in coffee for the resin composite, compomer, and giomer, respectively (Table 6). For the resin composite and giomer after the 5th day, the color changes between the water of the control group and the cola, orange juice, coffee, and energy drink of the experimental group were significant. On the other hand, for the compomer, the color change after the 5th day showed no significant color changes due to the orange juice (p>0.05), while the other beverages exhibited significant changes when compared with the color of the control (p<0.05).

A comparison of the reduction rate of color change as a function of the beverage treatment between the restorative materials showed that the resin composite had the smallest reduction rate (1.88±1.18), and giomer had the highest reduction rate (2.88±1.83).

DISCUSSION

El-Sharkawy *et al.* suggested that microhybrid resin composites have the lowest degree of water absorption, whereas giomers and compomers have higher degrees of water absorption. In addition, compomers contain methacrylates which increase water absorption compared to the resin composite. The results of this study also showed that the wettability is better for the compomer than the resin composite[20]. McCabe and Rusby showed that the giomer has significantly greater water absorption than the compomer[21]. These results conflict with those of the present study, in which the wettability of the compomer is not greater than the giomer.

Table 5 Color of the beverages

Group	*L*	*a*	*b*
Cola	44.24 (0.03)	23.97 (0.04)	62.13 (0.07)
Orange	30.30 (0.14)	14.18 (0.01)	50.75 (0.20)
Coffee	42.40 (1.10)	26.95 (0.77)	68.18 (1.75)
Energy	82.70 (0.16)	0.98 (0.05)	37.56 (0.05)
Water (Control)	93.73 (0.07)	−0.35 (0.05)	−0.16 (0.10)

Table 6 Comparison of the ΔE among the beverages

Group	ΔE (Before –5 day)		
	Resin composite	Compomer	Giomer
Cola	0.65 (0.47)	3.40 (0.96)	3.76 (1.88)
Orange	1.15 (0.69)	1.98 (0.90)	2.02 (0.88)
Coffee	3.58 (0.40)	4.30 (0.80)	4.85 (0.85)
Energy	1.64 (0.26)	3.05 (1.71)	2.87 (1.02)
Water (Control)	2.45 (0.73)	1.82 (1.34)	0.65 (0.46)

Table 7 Comparison of the ΔE of the restorative materials immersed in mineral water and distilled water

Group	Resin composite	Compomer	Giomer
Mineral water	2.45 (0.76)	1.47 (0.90)	0.65 (0.46)
Distilled water (Control)	0.55 (0.05)	0.52 (0.06)	0.66 (0.34)

Awliya *et al.* claimed that there is no significant difference in the microhardness of resin-based composite materials before and after immersion in coffee[22]. Badra *et al.* revealed that the microhardness of materials immersed in coffee and Coca-Cola remained stable up to 7 days, but showed a decrease after 30 days[14]. However, Saba *et al.* showed that immersion in beverages resulted in decreased microhardness and color change in CAD/CAM hybrid compared to feldspathic ceramic blocks[23]. Tanthanuch *et al.* also showed a decrease in microhardness in restorative materials after being immersed in beverages[17], and the results presented herein agree with those previously reported. The microhardness values of the restorative materials significantly decreased after 5 days of immersion in the experimental beverage groups compared to the initial value, and Coca-Cola most significantly affected the giomer microhardness. As the polymer material absorbs water, the coupling agent causes the loss of chemical bonds and hydrolysis occurs between the resin matrix and filler particles, affecting the microhardness[20].

The resin composite color change before and after 5 days of submersion in water was 2.45±0.73, and coffee induced the greatest color change value of 3.58±0.40. This figure was significantly higher compared to the color changes induced by water, which were 1.82±1.34 and 0.65±0.46 for the compomer and giomer, respectively. In the study conducted by Tekçe *et al.*, the resin composite exhibited a larger color change induced by water compared to the compomer[24]. On the other hand, Ertan *et al.* measured the color changes of various resin composite products exposed to beverages and showed that water resulted in a small change compared to other beverages[25]. This is likely because the components and proportions differ according to the type of resin composite; they may exhibit different properties, and experimental conditions of the studies differed[25,26]. In addition, in previous studies the control group was water, not distilled water, so ions and other substances in the water may induce additional chemical reactions. To explore the influence of species present in water, an additional experiment was conducted using the same sample fabrication process, submersion, and circulation. When submerged in distilled water, the ΔE values between before and 5 days after were 0.87±0.29, 0.52±0.06, and 0.66±0.34 for the resin composite, compomer, and giomer, respectively. With the exception of the giomer, these values are lower than those observed when immersed in mineral water (Table 7). In addition, for all restorative materials, the color change in coffee was the most evident. To determine whether the color of the beverage itself induced the color change, an additional experiment that measured the color of the beverages was performed. When submerged in coffee, the L(46.63), a(−0.68), and b(3.45) values of the samples changed to values similar to those of coffee (L(42.4), a(26.95), and b(68.18)). However, other beverages showed different tendencies, and although the color of the drink itself may influence the aesthetic restorative material, this hypothesis may not be valid for all beverages.

This study sought to determine the changes in aesthetic restorative materials induced by immersion in various beverages. To maintain the submersion conditions for all beverages and simulate the environment of the mouth, the beverages were stored at 37°C, however, the remineralization environment of saliva could not be recreated[27]. In this study, not all components of the beverages and restorative materials were analyzed, and therefore interpretations of the mechanisms could not consider physiochemical reactions. Therefore, future experiments exploring the mechanisms of the physiochemical reactions between the specific components of the beverages should be performed.

CONCLUSIONS

In this study, the changes in wettability, surface hardness, and color of aesthetic restorative materials induced by various beverages were analyzed before and after submersion in the beverages. The analysis of the results can be summarized as follows.

1. As the amount of beverage consumed and contact time with the aesthetic restorative material increased, its wettability increased significantly (p<0.05), surface hardness decreased significantly (p<0.05), and although there were color changes, they were not significant for all beverages. The change in the wettability for the resin composite submerged in the energy drink was greatest (p<0.05). For the compomer, cola induced the greatest change, and coffee had the most significant effect on the giomer (p<0.05).
2. A comparison of surface hardness results showed that the energy drink induced the most significant reduction in the hardness of the resin composite and compomer, while for the giomer, cola had the highest reduction rate (p<0.05).
3. With regard to the color change, as in the other restorative materials, the largest change was observed upon immersion in coffee (p<0.05).

Therefore, the null hypothesis that "the tested beverages will not affect the wettability, microhardness, or color of aesthetic restorations" can be rejected. It is recommended that the contact time with the beverages tested herein in the experimental groups and the frequency of their consumption be reduced. In addition, comparison of the restorative materials indicated that the changes in the resin composite were the smallest. This is likely due to its superior physical properties, and it is recommended that physical properties be considered when selecting aesthetic restorative materials.

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