Int. J. Odontostomat.,
16(2):311-320, 2022.

Effect of Color and Surface Roughness of Glass Ionomer Cements Submitted to Solutions Based on *Ilex paraguariensis*

Efecto del Color y Rugosidad Superficial de Cementos Ionómeros de Vidrio Sometidos a Soluciones en Base de Ilex Paraguariensi

Larissa Gabriela Ferreira Ojeda[1]; Tatiane Cristina Dotta[1]; Leonardo de Pádua Andrade Almeida[1]; Mayara Manfrin Arnez[1]; Raisa Castelo[1]; David Emanuel Ugarte[2]; Andréa Cândido dos Reis[1] & Alma Blasida Concepcion Elizaur Benitez Catirse[1]

OJEDA, L. G. F.; DOTTA, T. C.; ALMEIDA, L. P. A.; ARNEZ, M. M.; CASTELO, R.; UGARTE, D. E.; DOS REIS, A. C. & CATIRSE, A. B. C. E. B. Effect of color and surface roughness of glass ionomer cements submitted to solutions based on Ilex Paraguariensis. *Int. J. Odontostomat., 16(2)*:311-320, 2022.

ABSTRACT: The present study analyzed the color change (DE) and surface roughness (Ra) of glass ionomer cements (GICs) subjected to typical herbaceous South American beverage action - *Ilex paraguariensis*. 90 specimens (n = 10) were made: M1 - GIC Riva Light Cure® (SDI); M2 – GIC Riva Self Cure® (SDI); M3 - GIC Vitremer® (3M ESPE). After light curing/polymerization of GICs, specimens were divided for being submitted to solutions for one hour a day for 21 days: S1 – Yerba Mate Pajarito Traditional (Pajarito); S2 – Erva Mate Chimarrão (Barão de Cotegipe); S3 – Water (control). Four DE and Ra readings were taken: T0 – initial; T1 – 7 days; T2 – 14 days and T3 – 21 days, with a spectrophotometer and a roughness meter respectively. The obtained data were submitted to ANOVA statistical analysis and Bonferroni. S1 and S2 at T1 and T2 there was no statistical difference between the materials, and at T3 Materials M1=M2, M1=M3, and M2>M3. For S3 at T1, T2 and T3 there was no difference between the materials. Analyzing surface roughness, there was no statistical significance. Concluded that all materials changed color with greater influence of time, but without changing surface roughness.

KEY WORDS: Color, dental material, glass ionomer cements, *Ilex paraguariensis.*

INTRODUCTION

In order to preserve healthy dental structures, as well as the formation of reactionary tissues, restorative dentistry aims to employ appropriate materials and effective techniques to prevent recurrences or appearance of new cavities (Sidhu *et al*., 1997; Farrugia & Camilleri, 2015; Nicholson *et al*., 2020). Among these materials, the ones that stand out the most are glass ionomer cements (GIC) (Farrugia & Camilleri, 2015; Nicholson *et al*., 2020; Ana & Anggraeni, 2021).

Composed of inorganic glass particles dispersed in an insoluble hydrogel matrix (Francois *et al*., 2020; Klee *et al*., 2020; Nicholson *et al*., 2020), the GIC presents advantageous clinical characteristics such as: fluoride release (Forss & Seppa, 1990; Farrugia & Camilleri, 2015; Hafshejani *et al*., 2017; Francois *et al*., 2020; Klee *et al*., 2020; Nicholson *et al*., 2020; Ana & Anggraeni, 2021), adhesion to dental structures (Francois *et al*., 2020; Klee *et al*., 2020; Nicholson *et al*., 2020), coefficient of thermal expansion similar to the dental structure and biocompatibility (Frencken *et al*., 2006; Francois *et al*., 2020; Klee *et al*., 2020; Nicholson *et al*., 2020). And yet, in order to improve its properties, several materials were added in its composition (Farrugia & Camilleri, 2015; Francois *et al*., 2020; Klee *et al*., 2020; Nicholson *et al*., 2020; Ana & Anggraeni, 2021), as resinous components, as 2-hydroxyethyl methacrylate (HEMA), which resulted in resin-modified glass ionomer cements, presenting advantages such as greater strength and material stability (Sidhu *et al*., 1997; Francois *et al*., 2020; Klee *et al*., 2020; Nicholson *et al*., 2020; Ana & Anggraeni, 2021).

[1] Department of Dental Materials and Prosthodontics, Ribeirão Preto School of Dentistry, University of São Paulo, Ribeirão Preto, São Paulo, Brazil.
[2] Department of Rehabilitation, School of Dentistry, University National of Concepción, Concepción, Paraguay.

Received: 2021-03-04 Accepted: 2022-05-09

The color stability of restorative materials is very critical, due to intrinsic and extrinsic pigmenting agents (Prabhakar *et al*., 2013; Akay & Tanıs, 2017; Ozkanoglu & Akin, 2020). Certain daily actions can change the structure of the GIC, such as brushing, chewing and drinking beverages containing dye, which can alter the surface of the GIC, increasing their porosity and roughness, changing their staining (Prabhakar *et al*., 2013; Akay & Tanıs¸, 2017; Ozkanoglu & Akin, 2020).

Some of these pigments come from typical beverages such as Mate Tea, Chimarrão and Tereré, originating from Mate that have been widely consumed for hundreds of years as infusions, both of dried herbamate leaves and roasted mate leaves (Burris *et al*., 2012; Piovezan-Borges *et al*., 2016). Yerba mate (Ilex Paraguariensis St. Hilaire) is a tree species of the Aquifoliaceae family descending from the subtropical region of South America, used as a raw material for drinks and teas, especially in its traditional forms of Mate Tea and Chimarrão, which are consumed at temperatures between 60ºC to 70ºC (Burris *et al*., 2012; Cogoi *et al*., 2013; Piovezan-Borges *et al*., 2016; Fayad *et al*., 2020), and approximately 30 % of the population drinks more than 1 liter per day of these drinks (Piovezan-Borges *et al*., 2016). In addition to traditional drinks, yerba mate is also used in pharmaceutical preparations, being recommended as a stimulant, anti-inflammatory, anti-rheumatic, tonic and diuretic (Burris *et al*., 2012; Cogoi *et al*., 2013; Piovezan-Borges *et al*., 2016; Fayad *et al*., 2020).

Green yerba mate is obtained by inactivating endogenous oxide reductase enzymes in the leaves, predominantly polyphenoloxidases and peroxidases (Burris *et al*., 2012). The presence of these enzymes, still active in the product, results in important changes in color, aroma and flavor, compromising the commercialization of the green product. The green coloring of yerba mate is the result of the presence of chlorophyll, where the concentration of the pigment determines the intensity of the green color (Morawicki *et al*., 1999; Burris *et al*., 2012; Piovezan-Borges *et al*., 2016). Chlorophyll can degrade in different ways due to the sensitivity of its pH, enzymes, oxygen, temperature and light, and the water in the food that defines the speed of reactions (Morawicki *et al*., 1999; Burris *et al*., 2012; Cogoi *et al*., 2013; Piovezan-Borges *et al*., 2016).

Considering the high consumption of these drinks in South America, and few citations of related works in the literature, this study seeks to assess the relationship between the color change and roughness of the restorative material Glass Ionomer Cement often used by dentists, when in contact with these typical drinks from the mate.

MATERIAL AND METHOD

Materials. Materials and solutions used throughout this study and their specifications are described Table I.

Experimental design. The study variables in this experiment are: ΔE (color change) and Ra (surface roughness). The variation factors are: (1) Material [3 levels: M1 – Glass Ionomer Cement Restorative Riva Light Cure® (SDI - Victoria, Australia); M2 – Glass Ionomer Cement Restorative Riva Self Cure® (SDI - Victoria, Australia); and M3 – Glass Ionomer Cement Restorative Vitremer® (3M ESPE - St. Paul, MN, EUA) (Table I)]; (2) Solution [3 levels: S1 – Yerba Mate Pajarito Traditional (Pajarito - Itapuá, Encarnación, Paraguay); S2 – Erva Mate Chimarrão (Barão de Cotegipe – Barão de Cotegipe, Rio Grande do Sul, Brazil); and S3 –Natural Mineral Water (control) Minalice Mining Company LTDA – São Paulo, Brazil) (Table I)]; and solution application time [4 levels: 24 h (T0), 7 (T1), 14 (T2) and 21 (T3) days after specimens were obtained].

Sample preparation. 90 specimens of Ionomer Glass Cement (Table I) were obtained (n=10) according to manufacturer's instructions, using a teflon matrix of 6 mm diameter and 2 mm depth on a glass plate and a polyester matrix.

The material was inserted in the matrix in a single increment with the aid of a resin spatula (Duflex, São Paulo, Brazil). A polyester matrix and a glass plate were put on top of the filled cavity. Additionally, a weight of 1 kg was put on top to guarantee the complete filling of the matrix and to produce the overflowing of any excess material. Next, the light activation was carried out on the specimens light-curable using a curing light VALO (Ultradent – São Paulo, Brazil), in accordance with instructions from the manufacturer.

After the polymerization, samples were taken out of the matrix and kept in artificial saliva for 24 hours, in the oven at 37±1 °C. Afterwards, each specimen was polished with Sof Lex (3M, São Paulo, Brazil) discs in decreasing grit sequence, wetting the specimen surface between applications to avoid overheating and the consequent alteration of the surface. One of the surfaces

Table I. Restoration Material and Solutions.

Material	Composition	Manufacturer
Glass Ionomer Cement Restorative Light Cure®	Powder: Aluminum silicate fluoride Liquid: Polyacrylic acid, tartaric acid, hydroxyethyl methacrylate, dimethacrylate, acidified monomer	SDI - Victoria, Australia
Glass Ionomer Cement Restorative Riva Self Cure®	Powder: Aluminum silicate fluoride, polyacrylic acid Liquid: Polyacrylic acid, tartaric acid	SDI - Victoria, Australia
Glass Ionomer Cement Restorative Vitremer®	Powder: Aluminosilicate fluoride glass, potassium persulfate, microencapsulated ascorbic acid, pigments Liquid: Aqueous solution with copolymers of polyalkenoic acid, photoinitiator with phonoquinone, water, HEMA	3M ESPE - St. Paul, NM, EUA
Yerba Mate Pajarito Traditional	Energetic value (26.36 kcal) (%RDD*) Protein (2.32 g) Glycids (3.67 g) Lipids (0 g) Dietary fiber (0 g) Thiamine (1.26 mg) Pyridoxine (B6) (0.83 mg) Magnesium (57.12 mg) Iron (2.19 mg)	Pajarito - Itapuá, Encarnación, Paraguay
Erva Mate Chimarrão	Resolution RDC nº 360, of December 23, 2003, approves Technical Regulation on Nutrition Labeling of Packaged Foods, making nutrition labeling mandatory. However, these Technical Regulations do not apply to coffee, yerba mate, tea and other herbs without adding other ingredients. This product is exempt from registration in accordance with RDC nº 23, 2000 and RDC nº 27, 2010.	Barão de Cotegipe - Barão de Cotegipe, Rio Grande do Sul, Brazil
Natural mineral water Minalice (control)	Strontium, Calcium, Magnesium, Potassio, Sodium, Vanadium, Sulfate, Carbonate, Biocarbonate, Fluoride, Chloride	Minalice Mining Company LTDA

Percentage of recommended daily dose (%RDD)

of each specimen was marked to serve as a positioning guide, to be used with the spectrophotometer and the rugosimeter. After polishing, color change readings were taken and surface roughness.

ΔE measurement. The color variations measurement were performed at initial (24 hours after test specimens were created), 7, 14 and 21 days.

ΔE was measured using an SP62S spectrophotometer and the QA-Master I software (X-Rite Incorporated - Neu-Isenburg, Germany). Each specimen was carefully manipulated using tweezers (Millennium, Golgran, São Paulo, Brazil), dried with a paper towel and kept in a device prepared to holdthe samples and take readings with white and opaque background.

Color measurements were obtained using the CIE L* a* b* color system. The ΔE*, that is, the total difference between two color stimuli was calculated using the following formula:

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

The CIE L* a* b* system uses three parameters to define color: luminosity, hue and saturation. The luminosity represents the degree of light and dark of the object represented by the value of L*, with L* = 100 for white and L* = 0 for black. The parameters of a* and b*, called chromatic scale (hue), represent red if + a* and green if –a*, yellow if + b* and blue if –b*. Saturation is the intensity of the hue and is given by the numerical value of a* and b*. The values of ΔL*, Δa*, Δb*, correspond to the difference of values of L*, a*, b*, respectively, in comparison to the first color reading (initial).

Ra measurement. Ra measurements were obtaind using a rugosimeter (SJ-201 P/M, Mitutoyo - Tokyo, Japan). Each specimen was dried with paper towel and positioned on the device using tweezers (Colgran - São Paulo, Brazil), taking three readings to register the mean value. Ra was performed at initial (24 hours after test specimens were created), 7 days, 14 days and 21 days.

Sample immersion. Samples were stored in artificial saliva, composed of methylparaben (2.0), sodium carboxymethylcellulose (10.0g), KCl (0.625g), MgCl 2.6H2O (0.059g) CaCl2-2H2O (0.166g), K2HPO4 (0.804g), KH2PO4 (0.326g) in 1000 mL of distilled water, in the oven at 37±1 °C during the entire experiment. The artificial saliva was changed daily and

only taken out to be exposed to the solutions and to take measurements at 24 hours, 7, 14 and 21 days. The solutions were prepared as follows: Mixing the respective Herbs with drinking water and heated to a temperature of 60ºC and the water for the control group for the control test was kept at room temperature. Then, the specimens were immersed in their respective solutions (Table I) for a period of 60 minutes, once a day during the proposed study time.

Statistical analysis. ΔE and Ra data were analyzed using the Shapiro-Wilk and ANOVA tests. The statistical software used to run these tests was IBM SPSS Statistics for Windows, version 21.0 (IBM Corp, Armong - New York, EUA).

RESULTS

Results of ΔE. The statistical analysis showed that the Solution Factor had a statistically significant effect on ΔE (p=0.004), where S1 had an average statistically similar to S2 and S3, while S2 had an average higher than S3 (Table II).

For the Time Factor on ΔE (p=0.001), was found that T3 had an average statistically higher than T1 and T2, while T1 had an average, statistically similar to T2 (Table II).

The Time x Material interaction was also significantly about ΔE (p=0.008), which in the sense of the columns, it appears that in the days of 07 days (T1), 14 days (T2) and 21 days (T3), respectively, there is no difference between the materials. Analyzing in the direction of the lines, it was observed that M1, M2 and M3, respectively, at 07 days presented averages similar to the averages of 14 days; at 21 days the average was higher than the averages of 07 and 14 days for M1 and M2, while M3 had a higher average at 21 days, when compared to the average of 7 days and was similar to the average of 14 days (Table III).

The Time x Solution interaction also influenced the ΔE (p=0.001), which analyzing the columns, it appears that in T1, S2 had a mean higher than S1 and S3, while the mean of S1 was statistically equal to S3; in T2, S3 had a mean lower than S1 and S2, and the means of S1 and S2 were statistically similar. And T3 there was no difference between the solutions. Analyzing in the direction of the lines, it was observed that S1 in T1 had a lower average thanT2 and T3, and in these times the averages were statistically equal. For the solutions S2 and S3, respectively, in T1 they presented averages statistically similar to the averages obtained in T2; as T3 presented higher averages, than in T1 and T2 (Table III).

In addition, the interaction Time x Material x Solution was statistically significant over ΔE (p=0.049), in which by checking the Solutions for each of the Materials, at different times, in the columns it appears that for S1 and S2, respectively, in T1 and T2 they had the same behavior, that is, there is no statistical difference between the materials studied; in T3, it is observed that M1 was statistically equal to M2 and M3, however M2 showed a higher average than M3, this for both solutions. As for S3, there is no statistical difference between the materials at different times. Analyzing the behavior of the Materials under the action of the Solutions in each of the Times, it appears that for M1, in T1, S1 presented an average lower than S2 and equal to S3, however the average for S2 was greater than S3; in T2, S1 had an average equal to S2 and S3, and S2 had a higher average than S3; in T3, there was no statistically significant difference between the Solutions. For M2, at T1 there was no difference between the Solutions; in T2, S1 had an average statisticallyequal to S2 and greater than S3, while S2 had an average equal to S3; and in T3, the mean of S1 was statistically equal to S2 and S3, and S2 had a highermean than S3. And for M3, there is no difference between the Solutions at different times. And analyzing the effect of the Time of use of the Solutions in the Materials, it appears that for M1 and M3, under the action of the solutions S1 and S2, respectively, there was no difference in the studied times, and the behavior determined by S3, were also similar, that is, in T1 the average was statistically equal in T2, and less than T3, and T2 the average was also less than in T3 (Table IV).

Table II. Color variation mean values of ΔE by Solution (S) and Time (T).

S1	S2	S3	T1	T2	T3
3.5(±1.9)	4.0(±2.2)	2.8(±1.9)	2.7(±1.8)	3.0(±1.9)	4.6(±1.9)

Table III. Color variation mean values of ΔE by interactions Time (T) x Material (M) and Time (T) x Solution (S).

	T1	T2	T3
M1	2.7(±1.8)	2.8(±1.7)	4.8(±1.9)
M2	2.4(±1.9)	2.8(±2.5)	5.0(±2.2)
M3	2.9(±1.6)	3.3(±1.6)	3.9(±1.5)
S1	2.7(±1.4)	3.5(±1.9)	4.2(±1.9)
S2	3.6(±1.9)	3.4(±2.3)	4.9(±2.2)
S3	1.7(±1.5)	1.9(±0.9)	4.6(±1.6)

Table IV. Color variation mean values of ΔE by interaction Time (T) x Material (M) x Solution (S).

	T1			T2			T3		
	S1	S2	S3	S1	S2	S3	S1	S2	S3
M1	2.5(±1.1)	4.3(±2.0)	1.3(±0.9)	3.2(±0.9)	3.7(±2.1)	1.5(±0.9)	4.0(±2.1)	4.9(±1.8)	5.4(±1.5)
M2	2.6(±1.7)	3.0(±2.1)	1.5(±1.6)	3.7(±2.4)	3.0(±3.3)	1.6(±0.8)	5.4(±1.7)	5.8(±2.9)	3.7(±1.3)
M3	2.7(±1.5)	3.6(±1.3)	2.3(±1.7)	3.6(±2.3)	3.5(±1.3)	2.7(±0.7)	3.4(±1.5)	3.8(±1.2)	4.7(±1.5)

Results of ΔL

The analysis of variance showed that the Solution Factor had a statistically significant effect on ΔL (p=<0.001), which S3 had a statistically higher average than S1 and S2. While S1 presented an average statistically equal to S2. We then observed that the specimens immersed in water (control) (S3) showed a lighter color when compared to the other solutions, when analyzed separately. Negative ΔL means for solutions S1 and S2 signal that there is a loss of luminosity, that is, there is a darkening of the material (Table V).

The Time Factor also proved to be significant on ΔL (p=<0.001), where the specimens presented higher averages in T3, and lower averages in T1. It is then observed that at 07 days (T1) the specimens showed a darker color compared to the other times, and over time, they tend to become lighter (Table V).

The interaction Time x Material was also significant on ΔL (p=0.029), in which, observing the columns, it appears that in T1, M1 had an average lower than M2. This, on the other hand, presented an average statistically equal to M3, and M1 statistically equal to M3. However, in T2 and T3, there was no statistical difference between the materials. Analyzing in the direction of the lines, it was observed that the M1 in T1 presented a lower average than in T2 and T3, and the average in T2 was lower than in T3. And it turns out that for M2 and M3, the averages at T1 are statistically equal to the averages at T2, and the averages at T3 are statistically higher than those at T1 and T2, for both materials, respectively. For ΔL, this interaction demonstrates that the first study days 7 days and 14 days showed a lower average value of luminosity than in the 21 days. Perhaps it is due to the degree of polymerization of the materials (Table VI).

The Time x Solution interaction also influenced the ΔL (p=0.004), in which analyzing the columns, it appears that in T1, T2 and T3, S1 showed an average statistically equal to S2. S3, on the other hand, had a statistically higher average than S1 and S2. In the sense of the lines, it is observed that S1, in T1, presented a statistically similar average than in T2. In T3, the average was higher than in T1 and T2. Regarding S2 and S3, in T1 the means were statistically lower than in T2 and these were lower than in T3, respectively. When analyzing the parameter ΔL in this interaction, it appears that at all times, the solutions S1 and S2 presented lower mean values than S3, which demonstrates that there is a loss of luminosity at all times. It was also found that there is no difference in effect between the S1 and S2 solutions (Table VI).

Results of Δa. The analysis of variance showed that the Material Factor had a statistically significant effect on Δa (p=0.005), in which M2 had significantly higher values than M3, while M1 had an average statistically equal to M2 and M3 (Table VII).

The Factor Solution also had an influence on Δa (p=<0.001), in which S2 showed significantly lower values than S1 and S3, while S1 is statistically equal to S3. We then observe that the values represent that the Erva Chimarrão (S2) determines a trend towards green compared to the other solution (Table VII).

The Time Factor had significance in the Δa (p=<0.001), where in T1 the mean value was statistically lower than in T2 and T3. While in T2 the

Table V. Color variation mean values of ΔL by Solution (S) and Time (T).

S1	S2	S3	T1	T2	T3
-0.7(±2.1)	-0.7(±1.7)	1.1(±1.3)	-1.8(±1.7)	-1.3(±2.2)	2.8(±2.2)

Table VI. Color variation mean values of ΔL by interactions Time (T) x Material (M) and Time (T) x Solution (S).

	T1	T2	T3
M1	-2.2(±1.8)	-1.3(±2.1)	3.0(±2.3)
M2	-1.1(±1.4)	-0.7(±2.0)	2.8(±2.1)
M3	-2.0(±1.7)	-1.8(±2.3)	2.5(±2.2)
S1	-1.9(±1.5)	-2.1(±2.2)	2.0(±2.7)
S2	-2.6(±1.7)	-1.9(±2.0)	2.5(±1.7)
S3	-0.8(±1.3)	0.3(±1.3)	3.9(±1.5)

average was statistically equal to the average in T3. This means that in 07 days (T1) the specimens showed a greener color compared to other times, and over time, they tend to red (Table VII).

The Time x Material interaction was also significant over Δa (p=<0.001), since analyzing the columns, it appears that in T1 and T3, respectively, M1 had means statistically lower than M2, and M2 had a higher average than M3. M1 presented an average statistically similar to M3 in the three periods. At Time T2 there was no difference between Materials. In the analysis in the direction of the lines, it is observed that the M1 in T1 presented a lower average than in T2 and a similar average than in T3; and T2 the average was higher than in T3. It is also verified that the M2 in T1 and T2 presented similar averages, and in T3 the average was higher than in T1 and T2. For material M3, it was found that in T1 it presented a lower mean than in T2, where the means obtained in T3 were similar in T1 and T2, respectively (Table VIII).

And the Time x Solution interaction was also significant on Δa (p=<0.001), in which analyzing the columns, it appears that at T1, S1 had an average greater than S2, and equal to S3. S2 had a mean lower than S3. In T2, S1 presented an average similar to S2 and S3; and S2 showed a statistically lower average than S3. At 21 days (T3), there was no difference between the solutions. In the analysis in the direction of the lines, it is observed that S1 and S2, respectively, in T1 presented means statistically lower than in T2 and T3; at times T2 and T3 they presented similar averages. The S3 solution did not determine a significant difference in times T1, T2 and T3 (Table VIII).

Results of Δb. The analysis of variance showed that the Material Factor had a statistically significant effect on Δb (p=0.0014), in which M3 presented a mean lower than M1 the materials M1 and M2. While M1 was statistically equal to M2 (Table IX).

The Factor Time also had a significant effect on Δb (p=<0.001in which in T1 the average was the same as in T2 and less than in T3. While in T3 the means were statistically higher than the means obtained in T1 and T2 (Table IX).

The Time x Material interaction was significant over Δb (p=0.017), where analyzing the columns, it shows that in T1 and T3, respectively, there is no statistical difference between the Materials; in T2, M1 and M2 showed statistically similar averages, and M3 had lower averages, when compared to the means of materials M1 and M2, respectively. Analyzing in the direction of the lines, it is observed that the M1 and M3 in T1 and T2 presented statistically similar means, while M2 in T1 presented values of means lower than in T2. In T3 the averages were higher than the averages obtained in T1 and T2, for the 3materials, respectively (Table X).

And the Time x Solution interaction was also significant over Δb (p=<0.001where analyzing the columns, it appears that in T1 and T2 there was no difference between the solutions; in T3, S1 showed an average statistically similar to S2 and S3, while S2 determined a mean higher than S3. The analysis in the direction of the lines, it is observed that S1, S2 and S3, in T1 presented average statistically similar to the averages obtained in T2; and in T3 the averages were higher than in T1 and T2 (Table X).

Results of Ra. After obtaining the data, the analysis of variance showed no statistical significance ($p \leq 0.05$) of surface roughness (Ra) for none of the factors alone and for the interactions. Having no statistical significance, it was only placed as complementary information.

Table VII. Color variation mean values of Δa by Material (M), Solution (S) and Time (T).

M1	M2	M3	S1	S2	S3	T1	T2	T3
-0.3(±0.6)	-0.0(±0.7)	-0.5(±0.5)	-0.1(±0.5)	-0.6(±0.8)	-0.1(±0.4)	-0.4(±0.6)	-0.2(±0.6)	-0.2(±0.7)

Table VIII. Color variation mean values of Δa by interactions Time (T) x Material (M) and Time (T) x Solution (S).

	T1	T2	T3
M1	-0.5(±0.6)	-0.1(±0.6)	-0.3(±0.5)
M2	-0.2(±0.7)	-0.1(±0.7)	0.3(±0.7)
M3	-0.5(±0.5)	-0.3(±0.5)	-0.5(±0.6)
S1	-0.3(±0.4)	-0.1(±0.5)	-0.0(±0.6)
S2	-0.9(±0.7)	-0.4(±0.8)	-0.4(±0.9)
S3	-0.1(±0.4)	0.0(±0.3)	-0.2(±0.7)

DISCUSSION

Esthetics is a determining factor for the satisfaction of patients and professionals in dental treatment (Martin *et al*., 2016), but restorative materials are subject to several factors capable of altering their properties, such as staining and color change, mainly

Table IX. Color variation mean values of ∆b by Material (M) and Time (T).

M1	M2	M3	T1	T2	T3
1.1(±1.9)	1.1(±2.7)	-0.3(±2.2)	-0.2(±1.9)	-0.1(±2.4)	2.3(±2.5)

Table X. Color variation mean values of ∆b by interactions Time (T) x Material (M) and Time (T) x Solution (S).

	T1	T2	T3
M1	0.2(±1.4)	0.4(±2.0)	2.7(±2.1)
M2	-0.1(±2.3)	0.6(±2.9)	2.8(±2.9)
M3	-0.8(±1.7)	-1.3(±1.7)	1.3(±2.1)
S1	-0.3(±1.6)	-0.5(±2.5)	2.0(±2.5)
S2	-0.1(±2.4)	0.3(±2.9)	3.2(±2.9)
S3	-0.2(±1.6)	-0.1(±1.7)	1.6(±1.8)

due to ingestion of food and drinks, leading to esthetic restorative failure (Akay & Tanıs¸, 2017; Hamid *et al*., 2018). However, the superficial staining of some materials does not necessarily affect their mechanical properties or even the durability of the restoration, so it is necessary that the professional has knowledge of the properties of the materials (Akay & Tanıs¸, 2017; Hamid *et al*., 2018) There is a very traditional custom in the South of Brazil, in Paraguay, Uruguay and Argentina, which is the consumption of mate and chimarrão based on *Ilex paraguariensis*, which has the characteristic green color, which could affect the color of these esthetic materials (Burris *et al*., 2012; Cogoi *et al*., 2013; Piovezan-Borges *et al*., 2016).

The color change can be seen with the naked eye and can also be measured by specific equipment. Three different ranges are used to distinguish changes in color values from esthetic restorations: DE*<DE*3,3, easily observed, that is, it is clinically unacceptable (Vichi *et al*., 2004; Rüttermann *et al*., 2010; Freitas *et al*., 2012; Ceci *et al*., 2017).

The S1 (Yerba Mate Pajarito) solution showed a statistically similar average to S2 (Erva Mate Chimarrão) and S3 (Water-control), while S2 had a higher average than S3. According to the averages it can be said that the color change promoted by the two herbs (S1 and S2), from the *Ilex paraguariensis* plant have chlorophyll in its composition (Morawicki *et al*., 1999; Piovezan-Borges *et al*., 2016), change the color above the clinically acceptable reference values. These herbs contain chlorophyll which is the most important pigment present in plants, responsible for determining the intensity of the green color (Yu *et al*., 2019). This can be considered a factor for the staining and color change of the samples, as noted. It is known that several pigments present in the composition of beverages and foods are capable of being deposited on the surface layer of restorations (Adusumilli *et al*., 2016; Akay & Tanıs¸, 2017; Al-Samadani, 2017; Ozkanoglu & Akin, 2020). Yerba Mate Chimarrão (S2) is characterized by a more intense green color, which may justify a greater change than the control. Negative ∆L means can justify the color change for solutions S1 and S2, which signal that there is a loss of luminosity, that is, there is a darkening of the material.

And yet, according to Hse et at. (1999), although CIVs have anticariogenic properties, they do not have color stability due to the material's polyacid content and can be explained by the degradation of metal polyacrylate salts. According to Chhabra et at. (2014), the discoloration of the GIC may be due to adsorption or absorption of stains that can be influenced by the porosity of the glass particles, dehydration after setting and drying and micro-cracks that allow the restoration to be stained and discolored.

For the Time Factor, the results showed that at 7 (T1) and 14 days (T2) there was no difference and the values can still be considered clinically acceptable. At 21 days (T3), the change was greater than at 7 and 14 days, with a value exceeding the limit of 3.3, which means that it is not clinically acceptable. This shows that the longer the immersion time in the solutions used, the greater the color change observed in the samples. Previous studies have already shown that the immersion time in coloring solutions directly affects the intensity of the color change of the restorative materials (Bezgin etal., 2015; Savas *et al*., 2019).

It is also observed that in the times of 7 and 14 days of immersion in solutions S1, S2 and S3, respectively, there was no difference in behavior for any of the materials studied, however it is important to consider the values of Delta E obtained, which are noticeable. At 21 days of immersion in solutions S1 and S2, respectively, M2 showed greater color change than M3 and the same behavior as M1. Evidence that the immersion time is relevant. This may be justified when considering that M2 is a chemical reaction glass ionomer cement, to the degree of polymerization of materials or maturation of conventional GIC, which can take up to weeks (Hamid *et al*., 2018). The control solution showed no difference at different times and the values were within the clinically acceptable range. Studies have evaluated the effect of water on the color change of esthetic restorative materials (Bagheri *et al*.,

2005; Bezgin *et al*., 2015; Savas *et al*., 2019; Kamath & Nasim, 2020). As in this study, they demonstrated statistically significant differences between the initial and final values of color after immersion.

The values of ΔL, for the interaction Time x Material demonstrate that the first study times 7 days and 14 days, presented a lower average value of luminosity than in the time of 21 days, which can support the color change. Color changes after 24 hours of immersion can be attributed to the post-irradiation polymerization reaction that extends up to 24 hours whereas long-term changes in immersion can be attributed to the hygroscopic absorption of water in the material (Bagheri *et al*., 2005; Bezgin *et al*., 2015; Savas *et al*., 2019; Kamath & Nasim, 2020). Other factors such as the composition of the material can also affect its color and clinical appearance, due to its degradation over time (Al-Samadani, 2017; Hamid *et al*., 2018).

It was found that M2 and M3, had the same behavior, that is, both had the same tendency for luminosity at 7 and 14 days, and at 21 days the luminosity had greater value. This demonstrates the greater sensitivity of these materials, in the early times, probably due to having in their composition, resinous components, which are more susceptible to staining (Savas *et al*., 2019).

When analyzing the parameter ΔL for the interaction Time x Solution, it appears that at all times, the solutions S1 and S2 presented lower mean values than S3, which demonstrates that there is less light at all times. It was also found that there is no difference in behavior between the S1 and S2 solutions, in the three times. Therefore, the relevance of this interaction explains the color change.

The trend of Δa of M1 and M3 (photoactivated), at 7 days and 21 days of immersion was more towards the greenish color than M2 (trapped by chemical reaction). This is probably due to the composition of the materials, with M1 and M3 containing resinous components (Savas *et al*., 2019).

Regarding the effect of the solutions used at different times, the results show that at 7 days the solution S2 had a tendency towards very intense green, compared to the other solutions, and at 14 days it was equal to S1 and greener than the solution control, which makes sense, due to the color of the solution under study, characterized by the strong greenish color (Morawicki *et al*., 1999; Piovezan-Borges *et al*., 2016; Yu *et al*., 2019). The fact that S2 determines a greener color at 7 days, in comparison to other solutions, can be explained by the type of packaging of products studied, since, at Yerba mate Pajarito, it has Kraft paper packaging and has high gas permeability. and water vapor (Burris *et al*., 2012; Fayad *et al*., 2020). According to reports from the yerba mate processing industries, the product does not have a shelf life longer than 45 days, which causes a decrease in the concentration of chlorophyll, in which the main deterioration factors of dehydrated vegetables are the oxidation of lipids, non-enzymatic browning reactions, moisture adsorption, oxidation reactions of vitamins and oxidation of pigments such as chlorophylls and carotenoids (Burris *et al*., 2012). In addition, adsorption or moisture gain leads to sensory changes, microbial growth, favors chlorophyll degradation and promotes enzymatic browning (Piovezan-Borges *et al*., 2016). At Erva Chimarrão (Barão de Cotegipe), it has vacuum filling by Bioriented Metallized Polypropylene, where the metallization of flexible films aims toimprove the barrier properties of gases, water vapor and light of the substrates to which it is applied, in addition to giving a metallic appearance and shine, whose packaging has properties that improve the water vapor and light barrier (Valduga *et al*., 2005).

Therefore, comparing the materials used, the packaging of the products is different and can influence the conservation of the product. The packaging used for the Erva Barão de Cotegipe is more resistant to external events that lower the levels of chlorophyll found in mate herbs, probably allowing the preservation of a higher concentration of chlorophyll levels in the final product (Yu *et al*., 2019).

There were no different conditions in industrial processing and packaging that could explain the difference in the concentration levels of chlorophyll or another component, which would change the color of the samples made in the laboratory.

The Δb for M3 at 7 and 14 days tended towards blue and at 21 days there was a tendency towards yellow, for all materials studied (M1, M2 and M3), as well as for all solutions (S1, S2 and S3).

In this study, a complementary surface roughness test was performed, which demonstrated that none of the factors under study had a significant effect on this variable. However, the literature was controversial, since studies have shown that the action of extrinsic factors, such as food and beverage intake,

can promote higher roughness values in conventional glass ionomer cements (Sari *et al*., 2018; Guler *et al*., 2021) and modified by resin (Savas *et al*., 2019; Ong & Yoo, 2021). This variation can occur because the surface roughness of the CIV depends on several factors, such as a) size and shape of the glass particles; b) adhesion between the particles and the matrix; c) resistance inherent to the constituents of cements, and d) reaction of setting of each material (Savas *et al*., 2019; Ozkanoglu & Akin, 2020).

Taking into account the behavior of glass ionomer cements against the ingestion of food and dye drinks and their direct action on the behavior of dental materials (Choi *et al*., 2019), studies with other experimental conditions, such as brushing or polishing restorations, are important to clarify the susceptibility of these and other dental materials, in relation to use of *Ilex paraguariensis* mate herb, for application and guidance of patients on the best way to preserve your restorations.

CONCLUSION

According to the methodology used and within its limitations, it is concluded that:

- GIC Riva Self Cure® (M2) showed greater color change than CIV Vitremer® (M3) for the Yerba Mate Pajarito (S1) and Erva Mate Chimarrão (S2) Solutions in 21 days (T3);
- GIC Riva Light Cure® (M1) and CIV Riva Self Cure® (M2) showed greater color change at 21 days;
- Vitremer® (M3) showed a greater color change at 21 days than at 7 days;
- None of the Solutions had a statistical difference between the Materials in the Times of 7 (T1) and 14 days (T2);
- The solution Erva Mate Chimarrão (S2) determined a greater color change than the control (S3);
- The time factor has an influence on the color change, being greater at 21 days;
- The factors: Material, Solution and Time had no significant effect on surface roughness.

ACKONOWLEDGMENTS

Dr. Ana Paula Macedo, Laboratory II Specialist (Department of Dental Materials and Prosthesis, School of Dentistry, Ribeirão Preto – University of São Paulo).

OJEDA, L. G. F.; DOTTA, T. C.; ALMEIDA, L. P. A.; ARNEZ, M. M.; CASTELO, R.; UGARTE, D. E.; DOS REIS, A. C. & CATIRSE, A. B. C. E. B. Efecto del color y rugosidad superficial de cementos ionómeros de vidrio sometidos a soluciones en base de *Ilex paraguariensis*. *Int. J. Odontostomat., 16(2)*:311-320, 2022.

RESUMEN: En el presente estudio se analizó el cambio de color (DE) y la rugosidad superficial (Ra) de los cementos ionómero de vidrio (CIV) sometidos a la acción típica de las bebidas herbáceas sudamericanas - *Ilex paraguariensis*. Se realizaron 90 especímenes (n = 10): M1 - GIC Riva Light Cure® (SDI); M2 – GIC Riva Self Cure® (SDI); M3 - GIC Vitremer® (3M ESPE). Después de la fotopolimerización/ polimerización de los CIV, los especímenes se dividieron y fueron sometidos a soluciones durante una hora al día durante 21 días: S1 - Yerba Mate Pajarito Tradicional (Pajarito); S2 – Erva Mate Chimarrão (Barão de Cotegipe); S3 – Agua (control). Se tomaron cuatro lecturas de DE y Ra: T0 – inicial; T1 – 7 días; T2 – 14 días y T3 – 21 días, con espectrofotómetro y rugosímetro respectivamente. Los datos obtenidos fueron sometidos a análisis estadístico ANOVA y Bonferroni. S1 y S2 en T1 y T2 no hubo diferencia estadística entre los materiales y en T3 Materiales M1=M2, M1=M3 y M2>M3. Para S3 en T1, T2 y T3 no hubo diferencia entre los materiales. Al analizar la rugosidad de la superficie, no hubo significación estadística. En conclusion, todos los materiales cambiaron de color con mayor influencia del tiempo, pero sin cambiar la rugosidad de la superficie.

PALABRAS CLAVE: color, material dental, cementos de ionómero de vidrio, *Ilex paraguariensis***.**

REFERENCES

Adusumilli, H.; Avula, J. S.; Kakarla, P.; Bandi, S.; Mallela, G. M. & Vallabhaneni, K. Color stability of esthetic restorative materials used in pediatric dentistry: An *in vitro* study. *J. Ind. Soc. Pedod. Prev. Dent., 34(3)*:233-7, 2016.

Akay, C. & Tanıs¸, M. Ç. Evaluation of color changes of dental luting materials in food colorant solutions. *Int. J. Artif. Organs, 40(9)*:503-9, 2017.

Al-Samadani, K. H. Glass ionomer restorative materials response to its color stability with effect of energy beverages. *World J. Dent., 8(4)*:255-61, 2017.

Ana, I. D. & Anggraeni, R. Development of bioactive resin modified glass ionomer cement for dental biomedical applications. *Heliyon, 7(1)*:e05944, 2021.

Bagheri, R.; Burrow, M. F. & Tyas, M. Influence of food-simulating solutions and surface finish on susceptibility to staining of aesthetic restorative materials. *J. Dent., 33(5)*:389-98, 2005.

Bezgin, T.; Özer, L.; Tulga, Öz. F. & Özkan, P. Effect of toothbrushing on color changes of esthetic restorative materials. *J. Esthet. Restor. Dent., 27*:1-9, 2015.

Burris, K. P.; Harte, F. M.; Davidson, P. M.; Stewart Jr., C. N. & Zivanovic, S. Composition and bioactive properties of yerba mate (Ilex paraguariensis A. Sr.-Hil.): a review. Chil. *J. Agric. Res., 72(2)*:268-74, 2012.

Ceci, M.; Viola, M.; Rattalino, D.; Beltrami, R.; Colombo, M. & Poggio, C. Discoloration of different esthetic restorative materials: A spectrophotometric evaluation. *Eur. J. Dent., 11(2)*:149-56, 2017.

Chhabra, C.; Tavargeri, A.; Anegundi, R.; Patil, S. & Chhabra, K. G. Color stability of esthetic restorative material after topical fluoride application. *J. Res. Dent., 2(6)*:496-504, 2014.

Choi, J.; Lee, M. J.; Oh, S. H. & Kim, K. M. Changes in the physical properties and color stability of aesthetic restorative materials caused by various beverages. *Dent. Mater. J., 38(1)*:33-40, 2019.

Cogoi, L.; Giacomino, M. S.; Pellegrino, N.; Anesini, C. & Filip, R. Nutritional and phytochemical study of *Ilex paraguariensis* fruits. *J. Chem., 2013*:750623, 2013.

Farrugia, C. & Camilleri, J. Antimicrobial properties of conventional restorative filling materials and advances in antimicrobial properties of composite resins and glass ionomer cements-A literature review. *Dent. Mater., 31(4)*:89-99, 2015.

Fayad, E.; El-Sawalhi, S.; Azizi, L.; Beyrouthy, M. & Abdel-Massih, R. M. Yerba Mate (Ilex paraguariensis) a potential food antibacterial agent and combination assays with different classes of antibiotics. *LWT, 125*:109267, 2020.

Forss, H. & Seppa, L. Prevention of enamel demineralization adjacent to glass ionomer filling materials. *Eur. J. Oral Sci., 98(2)*:173-8, 1990.

Francois, P.; Fouquet, V.; Attal, J. P. & Dursun, E. Commercially available fluoride-releasing restorative materials: A review and a proposal for classification. *Materials, 13(10)*:2313, 2020.

Freitas, D. B.; Godoi, A.; Trauth, K. G. S.; Colucci, V. & Catirse, A. B. C. E. B. Study of the color alteration of esthetic restorative materials influenced by chlorhexidine associated with sweet red wine. *Int. J. Clin. Dent., 5(2)*:113-20, 2012.

Frencken, J. E.; Taifour, D. & van't Hof, M. A. Survival of ART and amalgam restorations in permanent teeth of children after 6.3 years. *J. Dent. Res., 85(7)*:622-6, 2006.

Guler, E. B. G.; Bayrak, G. D.; Unsal, M. & Selvi Kuvvetli, S. Effect of pediatric multivitamin syrups and effervescent tablets on the surface microhardness and roughness of restorative materials: An *in vitro* study. *J. Dent. Sci., 16(1)*:311-7, 2021.

Hafshejani, T. M.; Zamanian, A.; Venugopal, J. R.; Rezvani, Z.; Sefat, F.; Saeb, M. R.; Vahabi, H.; Zarrintaj, P. & Mozafari, M. Antibacterial glass-ionomer cement restorative materials: A critical review on the current status of extended release formulations. *J. Control Release, 262*:317-28, 2017.

Hamid, D. M. A.; Mahmoud, G. M.; El-Sharkawy, F. M. & Abou Auf, E. A. Effect of surface protection, staining beverages and aging on the color stability and hardness of recently introduced uncoated glass ionomer restorative material. *Fut. Dent. J., 4(2)*:288-96, 2018.

Hse, K. M. Y.; Leung, S. K. & Wei, S. H. Y. Resin-ionomer restorative materials for children: A review. *Aust. Dent. J., 44(1)*:1-11, 1999.

Kamath, K. A. & Nasim, I. Effect of hard water on properties of glass ionomer cement - An *in vitro* study. *Int. J. Pharm. Res., 12(2)*:100-6, 2020.

Klee, J. E.; Renn, C. & Elsner, O. Development of novel polymer technology for a new class of restorative dental materials. *J. Adhes. Dent., 22(1)*:35-45, 2020.

Martin, J.; Rivas, V.; Vildósola, P.; Moncada, L.; Oliveira Junior, O. B.; Saad, J. R.; Fernandez, E. & Moncada, G. Personality style in patients looking for tooth bleaching and its correlation with treatment satisfaction. *Braz. Dent. J., 27(1)*:60-5, 2016.

Morawicki, R. O.; Schmalko, M. E. & Känzig, R. G. Chlorophyll stability in Yerba Maté leaves in controlled atmospheres. *Braz. Arch. Biol. Technol., 42(1)*:1-5, 1999.

Nicholson, J. W.; Sidhu, S. K. & Czarnecka, B. Enhancing the mechanical properties of glass-ionomer dental cements: A review. *Materials (Basel), 13(11)*:2510, 2020.

Ong, S. & Yoo, S. Surface roughness and chemical composition changes of resin-modified glass ionomer immersed in 0.2% sodium fluoride solution. *J. Dent. Sci., 16(1)*:389-96, 2021.

Ozkanoglu, S. & Akin, E. G. G. Evaluation of the effect of variuos beverages on the color stability and microhardness of restorative materials. *Niger. J. Clin. Pract., 23(3)*:322-8, 2020.

Piovezan-Borges, A. C.; Valério-Júnior, C.; Gonçalves, I. L.; Mielniczki-Pereira, A. A. & Valduga, A. T. Antioxidant potential of yerba mate (*Ilex paraguariensis* St. Hil.) extracts in Saccharomyces cerevisae deficient in oxidant defense genes. *Braz. J. Biol., 76(2)*:539-44, 2016.

Prabhakar, A.; Pattanshetti, K. & Sugandhan, S. A comparative study of color stability and fluoride release from glass ionomer cements combined with chlorhexidine. *Int. J. Clin. Pediatr. Dent., 6(1)*:26-9, 2013.

Rüttermann, S.; Suyoun, K.; Raab, W. H. & Janda, R. Effect of exposure time on the color stability of resin-based restorative materials when polymerized with quartz-tungsten halogen and LED light. *Clin. Oral Invest., 14(5)*:599-605, 2010.

Sari, P. B.; Herda, E. & Damiyanti, M. Variables affecting conventional glass ionomer cement surface roughness after exposure to Coca-Cola®. *J. Phys. Conf. Series, 1073*:1-6, 2018.

Savas, S.; Colgecen, O.; Yasa, B. & Kucukyilmaz, E. Color stability, roughness, and water sorption/solubility of glass ionomer-Based restorative materials. *Niger. J. Clin. Pract., 22(6)*:824-32, 2019.

Sidhu, S. K.; Sherriff, M. & Watson, T. F. In vivo changes in roughness of resin-modified glass ionomer materials. *Dent. Mater., 13(3)*:208-13, 1997.

Valduga, E.: Javornik, G.; Sordi, M. & de Freitas Rezende, D. Evaluation of the quality of mate conditioned in different packaging materials. *Braz. J. Food. Technol., 8(2)*:99-105, 2005.

Vichi, A.; Ferrari, M. & Davidson, C. L. Color and opacity variations in three different resin-based composite products after water aging. *Dent. Mater., 20(6)*:530-4, 2004.

Yu, X.; Hu, S.; He, C.; Zhou, J.; Qu, F.; Ai, Z.; Chen, Y. & Ni, D. Chlorophyll metabolism in postharvest tea (*Camellia sinensis* L.) leaves: variations in color values, chlorophyll derivatives, and gene expression levels under different withering treatments. *J. Agric. Food Chem., 67*:10624-36, 2019.

Corresponding author:
Andréa Cândido dos Reis
Department of Dental Materials and Prosthesis
School of Dentistry of Ribeirão Preto
University of São Paulo
Av. do Café, s/n, 14040-904
Ribeirão Preto – SP
BRAZIL

E-mail: andreare73@yahoo.com.br / andreare@forp.usp.br

ORCID: https://orcid.org/0000-0002-2307-1720