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Effect of Immersion Duration in Coffee Beverage on Colour Stability of Polymethyl Methacrylate Denture Base Reinforced with 1% Chitosan Nanogel (Laboratory Study)

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ABSTRACT

Hot polymerized acrylic resin (RAPP) denture bases have several weaknesses, so RAPP requires reinforcing materials, one of which is chitosan. Extrinsic factors cause color changes in RAPP. The aim of this research was to determine the effect of soaking time for RAPP denture base material after adding 1% chitosan nanogel in coffee solution on color stability. Laboratory experimental research, using 16 cylindrical plates, diameter 15 x 2 mm (n = 8 from each group). There are 2 groups, the water-soaked group and the coffee drink group. Color stability measurements were carried out before and after soaking for 1, 2, 3, 4, and 7 days. The color stability of the samples was tested using a colorimeter and color changes were analyzed using the Kruskall-Wallis and Mann-Withney tests. There is an effect of soaking time for RAPP denture base material on color stability with a value of p = 0.0001. Soaking the RAPP denture base material after adding 1% chitosan nanogel in a coffee solution for color stability for 4 days is still clinically acceptable.

Keyword: Coffee; Chitosan Nanogel; Colour Stability; PMMA; Reinforcement

ABSTRAK

Basis gigi tiruan resin akrilik polimerisasi panas (RAPP) memiliki beberapa kelemahan, sehingga RAPP membutuhkan bahan penguat, salah satunya kitosan.. Kopi faktor ekstrinsik perubahan warna pada RAPP. Tujuan penelitian ini untuk mengetahui pengaruh lama perendaman bahan basis gigi tiruan RAPP pasca penambahan kitosan nanogel 1% dalam larutan kopi terhadap stabilitas warna. Penelitian eksperimental laboratoris, menggunakan 16 plat berbentuk silindris , diameter 15 x 2 mm (n = 8 dari setiap kelompok). Terdapat 2 kelompok, kelompok yang direndam air dan minuman kopi. Pengukuran stabilitas warna dilakukan sebelum dan sesudah perendaman selama 1, 2, 3, 4, dan 7 hari. Stabilitas warna sampel diuji dengan alat colorimeter dan perubahan warna di analisis menggunakan uji Kruskall-Wallis dan Mann-Withney. Ada pengaruh lama perendaman bahan basis gigi tiruan RAPP terhadap stabilitas warna dengan nilai p = 0,0001. Perendaman bahan basis gigi tiruan RAPP pascapenambahan kitosan nanogel 1% dalam larutan kopi terhadap stabilitas warna selama 4 hari masih dapat diterima secara klinis.

Kata kunci Kopi; Kitosan Nanogel**;** Stabilitas Warna; Polimetil Metakrilat; Bahan Penguat

1. Introduction

Polymethyl methacrylate (PMMA) is a dental material that is until now the most widely used in fabricating of denture bases because it has a number of advantages, including non-toxic, non-irritating, insoluble in oral fluids, having good aesthetics and easy to manipulate.[1] However, PMMA also has several disadvantages including porosity and the property of being inclined to absorb water or liquids, chemicals, both food and beverages and commonly fractures when dropped or impacted.[2] Based on the ISO 1567:1999 standard, the impact strength value of PMMA is 2 x 10^{-3} J/mm^2, while the transverse strength is 662 kg/cm^2.[3],[4] It is known that the standard hardness value of PMMA is 15-18 VHN,[1] while McCabe and Wallis (2008) said the optimal hardness value for PMMA is 20 VHN with its acceptable denture base, and the minimum hardness value is not below of 10 VHN (ADA, 1974).[5] Many studies have reported improvement of PMMA denture base material with the addition fiberglass, aluminum oxide, ultrahigh molecular weight polyethylene fiber, silica fiber and hydroxyapatite.

One of many natural resources that are being studied to be utilized as PMMA reinforcing materials is chitosan. Chitosan as a biopolymer or natural polymer in the form of a white amorphous solid that does not dissolve in alkalis and mineral acids. Chitosan is specific in shape (powder, gel, and flakes) and contains an amino group (NH_2) in its carbon chain.[6] Based on its viscosity, chitosan can be grouped into low-molecular chitosan, medium-molecular chitosan, and high-molecular chitosan. Rumengan I, et al (2018) article, the molecular weight of chitosan can affect its antibacterial properties. Chitosan with a lower molecular weight will produce better antibacterial properties than high-molecular chitosan.[7] Kurniasari, et al (2017) concluded that the development and advancement of technology, now chitosan is modified into nanoparticle size. Nanoparticles are particles within the size range from 1-1000 nm containing macromolecular material. The use of chitosan on a nanometer scale can increase the surface area which will increase the effectiveness in binding to other chemical groups.[8]

Colour stability is one of the important characteristics of the denture base because the colour of denture base must be maintained to preserve its aesthetic value. Discolourization occurs due to external factors and internal factors. External factors are causes that come from outside, for example drinks such as tea, coffee, chocolate and from denture cleaning agents, while internal factors come from chemical changes of the material itself, imperfect polymerization, and the addition of a substance to the composition of acrylic resins.[9]

Adiana ID., et al (2016) the addition of high-molecular chitosan nanogel with concentrations of 0.25%, 0.50%, 0.75%, 1%, and 1.5% each as much as 2 ml to PMMA with a ratio of *powder* and *liquid* as much as 23 g : 10 ml obtained results that the best transversal strength, impact strength, and colour stability were found in the addition of 1% chitosan nanogel with a result of 79.003 MPa, 7.91 ± 0.41 x 10^{-3} J/mm^2, and 0.076 cm^{-1}.[10] Ismiyati T (2017) reported that there is an influence of chitosan mixture with acrylic resin denture base material on its hardness using a concentration of 0.5%, and 1% with two different acid materials, acetic acid as material A and acrylic acid acetone solvent as material B obtained the highest average hardness in both material A (19.72 VHN) occurring at a concentration of 1%, and material B (15.14 VHN) occurred at a concentration of 0.5%.[5]

Most Indonesians drink coffee as part of their habit. Coffee is a type of beverage made by processing and extracting coffee plant beans. *The International Coffee Organization* (2018) Indonesia's domestic coffee consumption has almost quadrupled since 1990, the equivalent of 4.8 million 60-kilogram bags of coffee in 2019/2020.[11] The habit of consuming coffee knows no time, it can be done in the morning, afternoon, or evening. On average, each person can consumption 3-4 cups of coffee in a day.[12] Tarigan EB, et al (2015) conducted an analysis of the level of consumer preference for arabica blended coffee with robusta. From the results of the hedonic test, the results obtained showed that respondents preferred coffee with a robusta formula over arabica with a ratio of 3:1.[13] Kasuma N, et al (2015) assume that it takes about 15 minutes for an individual to drink coffee. Immersion of PMMA in coffee beverage for 1 week (7 days) is equivalent to (7 days x 24 hours x 60 minutes) divided by 15 minutes equals 672 days, this means equal to 2 years of use. He concluded that there was an increase in the absorbance value, which means that there was a change in colour stability in the PMMA of immersion in the coffee beverage for 1, 3, 5, and 7 days. It showed that immersion duration process caused more noticeable change in colour stability of the acrylic resin.[14]

This study was aims to determine the effect of immersion of PMMA denture base material in coffee beverage for 1 day (equal to the use of dentures for 3.2 months), 2 days (the same as the use of dentures for 6.4 months), 3 days (the same as the use of dentures for 9.6 months), 4 days (the same as the use of dentures for 12 month), and 7 days (same as the use of dentures for (22 months) after 1% chitosan nanogel addition to colour stability.

2. Methods and Materials

This laboratory experimental study obtained ethical clearance from the Ethical Committee of the Faculty of Medicine, Universitas Sumatera Utara, with decision letter 971/KEPK/USU/2022. This experimental study uses a posttest-only control group design. The purpose of this experiment was to determine the effect of immersing PMMA denture base material in robusta coffee (*Coffee canephora)* beverage with a 1% concentration of chitosan nanogel for an extended period, and how this affects the material's colour stability. The specimens was a PMMA with 1% chitosan nanogel with thickness of 15 x 2 mm (ADA No. 27 of 1993).[15]

There are 2 groups, immersed in water and coffee beverage, each group consisting of 8 specimens. Chitosan powder as much as 3 g dissolved in a beaker glass with 300 ml of acetic acid 1% then enter the magnetic bar for assisting the stirrer process at 400 rpm for 24 hours and sonication for 2 hours. Sodium Tripolypospat 1% solution (NaTPP) of 60 ml was added to form a chitosan gel, chitosan solution returned in the stirrer 24 hours and sonication for 3 hours to obtain nano-size. The nanoparticle size was tested with a Particle Size Analyzer (PSA). Chitosan nanogel was added to PMMA in the ratio of 11.5 g polymer: 5 ml monomer : 1.25 ml 1% chitosan nanogel. Chitosan nanogel is stirred first with PMMA monomer for 5 minutes, then mixed with polymer and stirred until homogeneous. Then reach dough stage phase is inserted into the mold. The pressured was applied of 1000 psi followed by 2000 psi. The specimens were cured by gradually raising from room temperature to 70ºC over a period of 90 min, before it was further cured over a duration of 30 min at 100ºC. The processed specimens then polished with a rotary grinder to the intended size. Coffee beverage is made with coffee grounds and water ratio of 1:10. The water and coffee beverage are grouped into 8 containers of 5 ml each.[16] The specimens measured with a colorimeter, and taken before immersion, and after 1, 2, 3, 4, and 7 days. The CIELAB coordinates of each sample were determine with use of a reflexive colorimeter at a wavelength of 360-750 nm under the D65 light source. The measurements were made by one operator. Then color change of the samples was determine using the formula below:

$$\Delta E\,(L,a,b) = [\,(\Delta L)^2 + (\Delta a)^2 + (\Delta b)^2]^{1/2}$$
$$\Delta L = Lb - Li$$
$$\Delta a = ab - ai$$
$$\Delta b = bb - bi$$

Before measurements were taken, the specimens were removed from their coffee beverage, washed with distilled water, dried using tissue paper, and transferred into the desiccator for 1x24 hours. The data were calculated and analyzed with the Kruskall-wallis test to see the long period of immersion of the PMMA denture base material after the addition of 1% chitosan nanogel in coffee beverage against colour stability to see significant differences between groups, the Mann-Whitney test was conducted.[17]

3. Results

The colour stability value of the PMMA with 1% chitosan nanogel of immersion in water and coffee beverage for 1, 2, 3, 4, and 7 days in this study was obtained by measuring results from each specimen using a colorimeter and obtained the result in the form of a ΔE value from each specimen tested. In this study, the specimens without immersion in coffee beverage was a specimens that was immersed in water as a control group. In this study, C1, C2, C3, C4, C5, and W1, W2, W3, W4, W5 are PMMA immersion on coffee beverage and water in 1, 2, 3, 4, and 7 days.

Table 1. Value and mean (± SD) of Colour Stability on PMMA with 1% Chitosan Nanogel Immersion in Water and Coffee Beverage for 1, 2, 3, 4, and 7 days

Specimen	Colour Stability (ΔE) PMMA + 1% Chitosan Nanogel									
	W1	**C1**	**W2**	**C2**	**W3**	**C3**	**W4**	**C4**	**W5**	**C5**
1	0.8**	2.53**	1.39	2.63	2.15	2.82	2.78*	3.19	3.43	6.27
2	0.66	1.85	1.49	2.47	2.51	3.05	2.97	3.33	3.62	6.47
3	0.78	2.02	1.62**	2.7**	2.92	3.23**	3.2	3.38**	3.48	6.28
4	0.61	1.76	1.35	2.20	2.35	2.93	2.87	3.25	3.39*	6.25
5	0.72	1.52*	1.47	1.51*	2.76	2.91	3.16	3.02*	3.54	6.16
6	0.59*	1.91	1.07*	2.26	3.19**	2.81*	3.26**	3.35	3.48	6.77**
7	0.61	1.61	1.57	1.76	2.04*	3.13	2.91	3.31	3.79**	6.29
8	0.77	2.22	1.32	1.9	2.52	2.93	2.89	3.26	3.47	6.12*
± SD	0.7 ± 0.09	1.92 ± 0.33	1.41 ± 0.17	2.18 ± 0.43	2.56 ± 0.39	2.98 ± 0.15	3.00 ± 0.18	3.26 ± 0.11	3.52 ± 0.13	6.33 ± 0.21

Description: * smallest value
** greatest value

Based on result the Kruskall-wallis statistical test obtained significance value p = 0.0001 ($p < 0.05$). This suggests that there is a significant effect of prolonged immersion of PMMA denture base material after 1% chitosan nanogel addition in coffee beverage on colour stability (Table 2).

Table 2. Effect of Immersion Duration in Coffee beverage on Colour Stability of PMMA Denture Base Reinforced with 1% chitosan Nanogel

Group	Colour Stability (ΔE)			*p*
	Day	**n**	**±SD**	
Coffee	1	8	1.92 ± 0.33	0.0001*
	2	8	2.18 ± 0.43	
	3	8	2.98 ± 0.15	
	4	8	3.26 ± 0.11	
	7	8	6.33 ± 0.21	

Description: *significant (p<0.05)

Table 3. Different Effect of Immersion Duration in Coffee beverage on Colour Stability of PMMA Denture Base Reinforced with 1% chitosan Nanogel

Group	Duration		*p*
Coffee	C1	C2	0.27
		C3	0.0001*
		C4	
		C5	
	C2	C3	
		C4	
		C5	
	C3	C4	0.003*
		C5	0.0001*
	C4	C5	

Description: *significant (p<0.05)

The *Kruskall-wallis* and Mann-withney statistical test was used to determine which treatment group was affected. The results of the *Mann-withney* statistical test showed that in the immersion group in the coffee beverage of 1 day and 2 days of immersion there was no difference with the value of p = 0.027 ($p < 0.05$) (Table 3).

4. Discussions

The colour stability values were found to vary in the set of specimens in the water and coffee groups (Table 1). The results of this study are in accordance with the research by Savitri RPA, et al (2022) that the colour change value in in vitro studies is clinically acceptable if $\Delta E \leq 3.70$, while in in vivo studies the colour change is acceptable if $\Delta E \leq 6.80$. The results of the study in all groups were still acceptable at 4 days of immersion in the assumed coffee beverage for 1 year of use.[18] The composition, material properties, chemical structure, and grinding from PMMA's fabrication process can influence colour changes in PMMA's denture base.[19] Several factors can cause variations in colour stability values that cannot be controlled and can affect the PMMA polymerization process, including because the mixing process between polymers and monomers of PMMA denture base materials is not carried out simultaneously for all specimens and manual stirring techniques that cause the speed cannot be controlled perfectly. The difference values explained above could also happen due to can its microporosity difference. These microporosity difference could occur due to the from the improper stirring process between polymer and monomer components, also it could happen due to the evaporation of unreacted monomers and low polymer molecular weight, its also known as porosity. This difference in values can also occur because when the polishing process of each specimens uses a *rotary grinder* which is done by hand or manually, causing differences in the surface and thickness of the specimens due to uneven pressure.[5]

The study obtained significant findings in the effect of immersion duration of PMMA denture base material after the addition of 1% chitosan nanogel in coffee solution on colour stability (Table 3). In this study, the 1% chitosan nanogel used had a characteristic yellowish-white colour, so that chitosan itself had given colour to PMMA before immersion with coffee beverage, the colour of the PMMA denture base material with the addition of 1% chitosan nanogel looks paler compared to what is not added 1% chitosan nanogel. In accordance with Sugita's research (2009) reported that chitosan has a high absorption capacity ability and chitosan is often used as an effort to remove dyes in textile waste so that there is a possibility that with a high level of chitosan absorption there is an absorption of tannins from coffee into PMMA denture base materials that can cause discolourization.[20]

The obtained results were not consistent with the statement that chitosan possesses high chemical reactivity due to the presence of other compounds, such as the amine group (NH2), which allows it to bind water effectively.[11] Ismayati T (2017) mixing PMMA and chitosan with various concentrations of hardness with material A in the form of chitosan with acetic acid solvent and B material chitosan acrylic acid acetone solvent. The average hardness value and standard deviation in group A were 21.84 ± 0.84 VHN, while in group B it was 15.14 ± 0.55 VHN. It is obtained that material A is greater in hardness than material B due to material A, EDGMA (ethylene glycol dimethacrylate) contained in monomers is insoluble in acetic acid, so that in the presence of EDGMA (ethylene glycol dimethacrylate) it produces material A has a higher hardness compared to material B.[5]

One of the contents of coffee compounds is tannins containing polyphenols which are acids known to interfere with the hydrolysis reaction between phenols and polymethyl methacrylate esters on the acrylic resin plate so that the polymer chain bond is disturbed and can increase porosity on the inside of the acrylic resin. This will affect the discolourization due to an increase in the absorption of tannins. The content of tannins has a polar structure that causes physical chemical bonds that occur stronger and commonly oxidized oxygen, so it will commonly result in discolourization. The advantage of robusta coffee used in this study is that it has a deeper colour compared to the type of arabica coffee can be one of the factors supporting colour changes.[21],[22]

The results of the *Mann-Whitney* statistical test (Table 3) showed that the coffee beverage group at 1 day and 2 days of immersion there was no difference in influence with the value of p = 0.027 ($p > 0.05$). In the other group there was a significant colour stability difference in the effect on the base material of PMMA

dentures after the addition of 1% chitosan nanogel in coffee beverage on with a value of p = 0.0001 ($p < 0.05$). Based on the colour change value obtained, it is known that the longer the immersion, the greater the colour change value. According to the research conducted by Said, et al (2015) conclude that the discolourization of artificial tooth acrylic plates is caused by 2 factors, internal and external, which external can be caused by coffee, tea, nicotine and mouthwash. In denture users who often consume coffee for a long period of time, the coffee beverage will enter and be absorbed through the poreus of the cavity on the acrylic plate. It reports that the duration of immersion affects the colour change value in the specimens, the higher the day it is, the higher the colour stability value obtained and when there is a colour change.[21]

The high absorption of coffee stains into the PMMA, which may be supported by chitosan properties that have high absorption, is one of the factors that can cause discolourization due to the immersion duration.[19] Coffee that contains phenol compounds in the form of tannins which are colour-bearing and colour binders the longer they are consumed, the more they will form stains, affecting pigmentation or colours that can cause brown colour.[21] Acrylic resin material has one of the properties, absorbing water slowly over a certain period, the diffusion process occurs continuously so that when in prolonged contact with acrylic resin it can penetrate the material and damage the polymer chain bond, resulting in a decrease in physical and mechanical properties, one of which is discolourization.[21]

5. Conclusions

In conclusion, the colour stability of the denture base material of PMMA after the addition of 1% chitosan nanogel in coffee beverage during 4 days is still clinically acceptable. There is an effect of addition chitosan nanogel in PMMA, which can impact strength and transverse strength but can negative impact on color stability. The discoloration of acrylic resin is not always permanent can be reduced after cleaning with toothbrush. Denture users it is recommended to diligently clean with toothbrush and come to the dentist for polishing to keep the colour of denture remains stable.

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7. Conflicts of Interest

The authors declare that there are no conflicts of interest to disclose concerning this study.

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