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Original Article

Effect of Novel Cycloaliphatic Comonomer on Surface Roughness and Surface Hardness of Heat-cure Denture Base Resin

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ABSTRACT

Background: Polymethyl methacrylate (PMMA) is a widely used resin in the field of prosthodontics for fabricating myriad orofacial prostheses. Albeit several advantages, it possesses certain lacunae concerning physicomechanical properties. **Purpose:** This *in vitro* research aimed to evaluate the surface roughness (SR) and hardness (SH) of heat-cured PMMA processed with a cycloaliphatic monomer, tricyclodecane dimethanol diacrylate (TCDDMDA), in methyl methacrylate at various concentrations. **Materials and Methods:** Groups have been divided into control (SRC and SHC) and experimental groups (SR10 and 20; SH10 and 20). Forty-five PMMA disc specimens were prepared. SR was assessed using a nanomechanical testing machine and the arithmetic roughness (Ra) was recorded. The same specimens were then subjected to Vicker's microhardness testing and Vicker's hardness number (VHN) was obtained. Data were compared using one-way analysis of variance (ANOVA) and *post hoc* Bonferroni tests (α=0.05). **Results:** The mean (standard deviation [SD]) of SRC, SR10, and 20 groups were 111.415 nm (0.789), 62.666 nm (0.482), and 41.004 nm (0.561), respectively. The mean (SD) VHN of SHC, SH10, and 20 groups were 21.003 (0.252), 23.975 (0.207), and 34.622 (0.079), respectively. **Conclusion:** The addition of TCDDMDA markedly decreased the SR and increased the SH of the experimental groups.

KEYWORDS: *Cycloaliphatic monomer, surface roughness, surface hardness, Vicker's hardness*

INTRODUCTION

Polymethyl methacrylate (PMMA) denture base resin has been extensively used in the field of prosthodontics. It possesses certain chief characteristics such as uncomplicated processing, chemical steadiness,

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longevity, affordability, lightweight, and color stability. Researches have shown a direct association between surface roughness (SR), the deposition of plaque, and attachment of *Candida albicans* on the denture surfaces.[1] Therefore, the surface properties of PMMA are of prime concern. This clinical scenario dictates the need for a featureless surface to deter the biofilm genesis on the denture surfaces. In the dental literature, clinically acceptable SR threshold level is ≤0.2 μm, below which no impact on plaque accumulation and retention could be expected on prosthetic and dental restorative materials.[2]

The surface properties of PMMA can also be affected by hardness. Hardness measurements are an inherent physical property of PMMA, exclusively important concerning resisting forces arising from in-service mechanical denture cleansing.[3] Measurements of PMMA hardness may be indicative of possible degradation of the polymeric matrix, which will result in reduced hardness, high perchance of fracture, and adversely affecting the durability of the denture base.[4] Residual monomer content in the polymerized resin has been found to adversely affect the surface hardness (SH).[5] This is because hardness assessments can be authenticated indirectly by evaluating the monomer to polymer conversion of heat- and auto-polymerizing resins.[6]

Despite favorable characteristics of PMMA, the intended surface and mechanical properties have not been fulfilled.[7] Hence, there arises a necessity for improvising these properties of PMMA. Numerous methods and techniques have been experimented to ameliorate its properties by modifying the polymer with the incorporation of fibers and/or fillers. However, the inherent compromised interfacial bonding between the organic matrix and inorganic fibers led to the advent of monomer (methyl methacrylate [MMA]) modifications. Modification of MMA was done with fluoro monomers, phosphate monomers, methacrylic acid monomer, itaconate monomers, nitromonomers, and nonspecific monomers or solutions.[8]

Modifying MMA with a cycloaliphatic monomer, tricyclodecane dimethanol diacrylate (TCDDMDA), significantly reduced the carbon–carbon double bond peak intensity (C = C) in infrared spectroscopy.[9] Therefore, it can be inferred that TCDDMDA invariably reduced the unpolymerized monomeric residues in the cured specimens. Also, modifying the proprietary monomer with TCDDMDA showed no cytotoxicity to the murine fibroblasts, which is attributed to reduced levels of residual monomer.[10] However, the effects of adding TCDDMDA comonomer on the surface characteristics of denture base resins are not been documented in the dental literature yet. Hence, this *in vitro* research aimed to evaluate the SR and SH of heat-cure denture base resin processed with TCDDMDA comonomer at 10% and 20% (v/v) concentrations. The null hypothesis was that the addition of TCDDMDA would not affect the SR and SH of the tested acrylic resin.

Materials and Methods

Cycloaliphatic monomer, TCDDMDA (Sigma-Aldrich, Germany; CAS Number 42594-17-2) and heat-polymerizing denture base resin (DPI Heat Cure, Dental Products of India, Mumbai, India) were used. The control group comprises PMMA specimens with 100% MMA. Experimental groups comprise PMMA specimens prepared by adding TCDDMDA in MMA at 10% and 20% (v/v) concentrations. Table 1 describes the grouping of specimens for SR and SH.

Preparation of specimens

Forty-five PMMA disc specimens (20 × 3 mm^2) were prepared out of dies, laser-cut from commercially available acrylic sheets. To obtain mold space, dies were impressed in the putty impression (Photosil Soft putty, DPI-Dental Products of India, Mumbai, India) in the brass dental flask. The polymer and the monomer were mixed (3:1 ratio) and packed into the mold space in dough stage at a packing pressure of 3500 psi in the mechanical press (Sirio Dental Srl, Meldola FC, Italy) for 10 min. This was followed by heat-curing at 74°C for 8 h followed by terminal boiling treatment at 100°C for 1 h in an acrylizer (Unident Instruments, New Delhi, India). After half-an-hour bench cooling and deflasking, the processed specimens were retrieved. Then the specimens were finished using silica-carbide (SiC) emery

Table 1: Grouping of specimens

No. of specimens (N = 45)	Surface roughness (SR)	Surface hardness (SH)
I. Control groups: PMMA specimens with 100% MMA		
N = 15	Group-SRC	Group-SHC
II. Experimental groups: PMMA specimens with 10% and 20% TCDDMDA		
N = 15	Group-SR10	Group-SH10
N = 15	Group-SR20	Group-SH20

PMMA = polymethyl methacrylate, MMA = methyl methacrylate

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sheets of grit 400, 600, 800, 1000, 1200, and finally 1500. The specimens were eventually polished using pumice. All the specimens were prepared by a single investigator. The final thickness of the specimens was reassured with the aid of a digital Vernier caliper (Mitutoyo 500-197-20, Mitutoyo South Asia Pvt. Ltd., Bangalore, India). All polymerized specimens were immersed in distilled water at 37ºC for 48 ± 2h before testing for the monomeric residues to leach out.

Surface roughness assessment

SR was assessed using the nanomechanical testing machine (Hysitron; TI 700 Ubi 1, Florida, USA) [Figure 1]. It is equipped with a movable camera (10× zoom) to focus the area of interest on the specimen. An area of 10 × 10 μm^2 was scanned at a 1 Hz scan rate and 2 μN set point using scanning probe microscopy (SPM). Inbuilt TriboScan software, Bruker India Scientific Pvt. Ltd., Bangalore, India (version 8.1.1) with TriboView suite is used for image analysis and to create three-dimensional (3D) plots. Each specimen was scanned for five measurements at different random sites to get arithmetic mean or average roughness (R_a) values in nanometer (nm).[11]



Figure 1: Nanomechanical testing machine

Vicker's hardness test

For SH, Vicker's hardness test was experimented with an HMV-G21 device (Micro Hardness Tester, Shimadzu, Singapore) [Figure 2] with a 490.3 mN (50 g) load and 15 s penetration. The specimens were placed on the X–Y specimen stage for focusing and indentation. This device is equipped with an automatic lens (40×) switching function that switches the lens to suit the size of the indentation. For each specimen, five indentations were done at five random points and averaged. Standardized automatic length measurement function (HMV-G test software) using a CCD camera built in a novel G frame is used to measure the length of the diagonals with no risk of human error. The resultant conversion hardness of the specimens was obtained as Vicker's hardness number (VHN). For both SR and SH, the test machine operators were blinded from the type and composition of the specimen.

Statistical analysis

Statistical analysis was performed using Statistical Package for the Social Sciences (SPSS) software program, SPSS Inc., Chicago, IL, USA, version 20.0. Preliminary results of the Shapiro–Wilks test indicated the data were normally distributed ($P > 0.05$). Descriptive statistics, including mean, standard deviation (SD), standard error, maximum, and minimum were calculated. Concerning inferential statistics, the level of significance between the three groups for R_a and VHN was tested with one-way analysis of variance (ANOVA). To compare the groups, the *post hoc* Bonferroni test (α=0.05) was performed. A value of $P < 0.05$ was considered for statistical significance.

Results

Table 2 presents the mean (SD) of the control and experimental groups for both R_a and VHN. One-way ANOVA showed the level of significant differences among the groups ($P = 0.000$). Bonferroni multiple comparison tests [Table 3] showed a statistically significant difference between the groups of both R_a and VHN ($P = 0.000$). Regarding the R_a, the specimens of the experimental groups (SR10 and SR20) showed smoother surfaces than the control group SRC. SR20 possessed the smoothest surface of all. Figures 3–5 illustrate the 3D topographies of SRC, SR10 and SR20

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specimens. Concerning SH, experimental groups had harder surfaces than the control. Among the groups, SH20 showed the hardest surface.

Discussion

Modifying the MMA monomer of heat-cure denture base acrylic resins is not uncommon. Various studies had executed monomer modifications in an effort to improvise the physicomechanical properties of denture base acrylic resins.[8] TCDDMDA is a recently identified, novel crosslinking, di-functional, dual-reactive, cycloaliphatic acrylic monomer. Ajay *et al.*[9] showed copolymerization of TCDDMDA with PMMA and it is noncytotoxic to the mouse fibroblast.[10] However, there are no previous studies about the effect of this novel monomer on SR and SH. In this study, the addition of TCDDMDA with MMA decreased SR and increased SH of denture base resin. Hence, the null hypothesis was rejected.

In this research, the SR of the experimental groups (0.041–0.062 µm) was very well below the SR threshold level of 0.2 µm. Therefore, the perchance of microbial adherence and colonization is negligible on the denture surface. This, in turn, will dampen mucosal inflammation and ease the denture cleaning by the geriatric population whose manual dexterity might have compromised. The exact reason for the smooth surface of the experimental groups is unknown. Perhaps TCDDMDA comonomer influenced the differences elicited in SR. Similar studies on SR with monomer modifications have been performed. Fluoralkyl methacrylate in MMA reduced

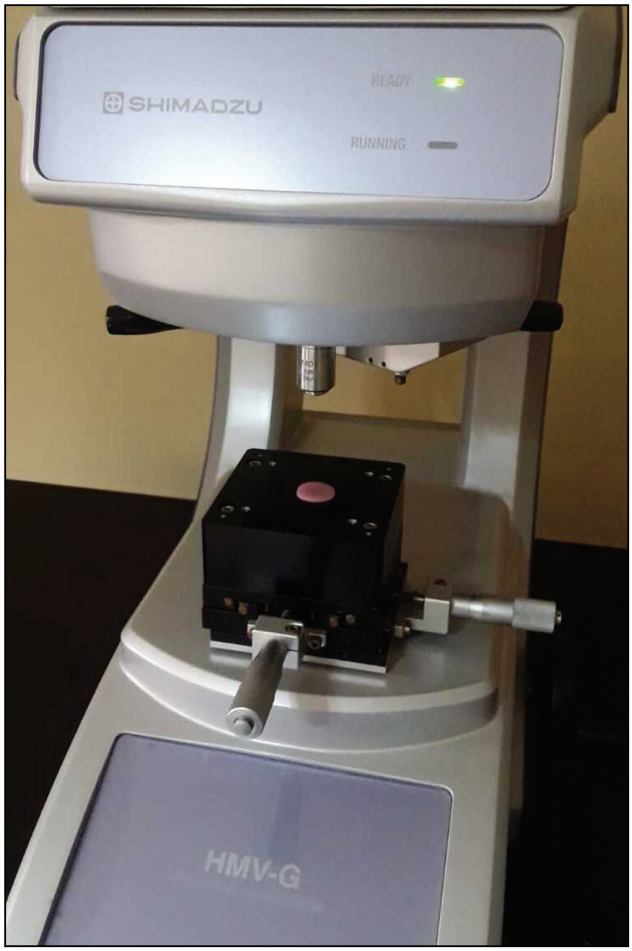


Figure 2: HMV-G21 microhardness tester

Table 2: One-way analysis of variance

I. Surface average roughness (R_a)			
Groups	**Mean (SD)**	***F* Value**	***P* Value**
SRC	111.415 (0.789)	49994.948	0.000
SR10	62.666 (0.482)		
SR20	41.004 (0.561)		
II. Vicker's hardness number (VHN)			
Groups	**Mean (SD)**	***F* Value**	***P* Value**
SHC	21.003 (0.252)	20416.384	0.000
SH10	23.975 (0.207)		
SH20	34.622 (0.079)		

SD = standard deviation

Table 3: *Post hoc* Bonferroni test

I. Surface average roughness (R_a)			
Group	**Compared group**	**Mean difference**	**Sig.**
SRC	SR10	48.749*	0.000
	SR20	70.411*	0.000
SR10	SR20	21.662*	0.000
II. Vicker's hardness number (VHN)			
Group	**Compared group**	**Mean difference**	**Sig.**
SHC	SH10	–2.972*	0.000
	SH20	–13.619*	0.000
SH10	SH20	–10.647*	0.000

*The mean difference is significant at the 0.05 level

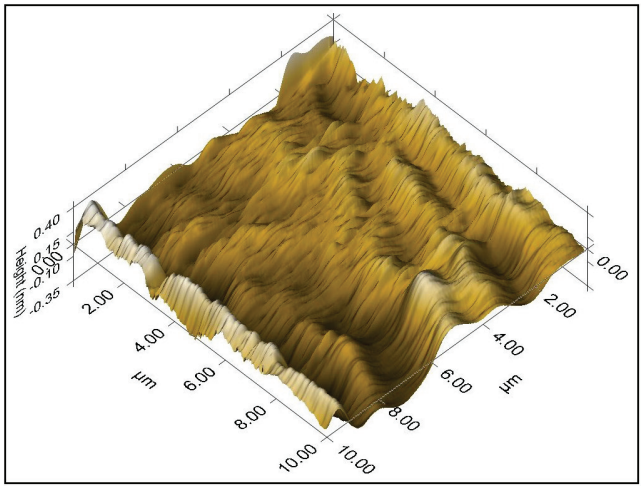


Figure 3: 3D topography of SRC group

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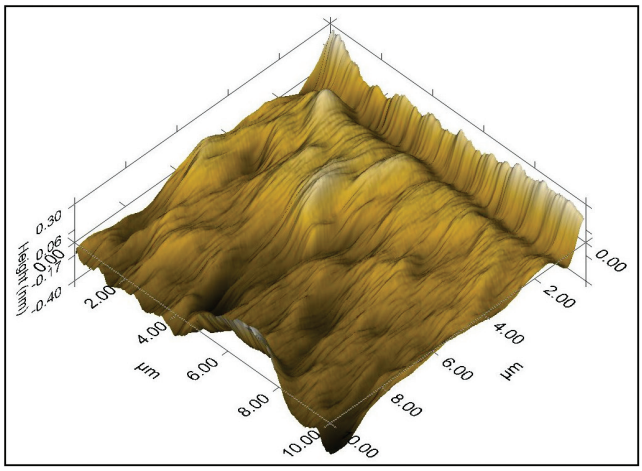


Figure 4: 3D topography of SR10 group

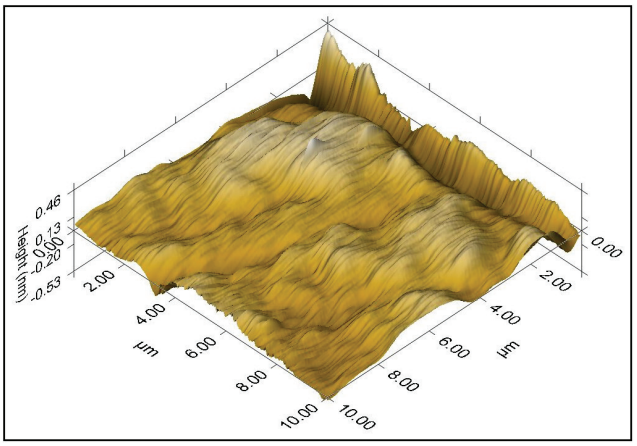


Figure 5: 3D topography of SR20 group

the SR of PMMA specimens when compared to the control, which was not so significant.[12] The addition of methacryloyloxy undecyl pyridinium bromide to MMA did not significantly alter the SR when compared to control.[13] *N*-acetyl cysteine (NAC) in MMA resulted in increased SR with increasing the NAC concentration and it was concluded that adding NAC beyond the concentration of 0.3% (0.86 µm) had negative influences on the SR.[14] However, this is greater than the SR threshold value. Higher SR values were found for the control group and they were reduced as the methacrylic acid concentration in MMA increased in the experimental groups.[15] None of the studies have pointed out the exact reason for the SR modulation in PMMA after monomer modifications which mandate future investigations in this field.

Various factors such as grits of SiC emery sheets, type of polishing, and polishing agents, time, and efficiency of manual polishing influence the SR directly. Previous studies have shown mechanical polishing yielding smoother surfaces than the chemical polishing techniques.[16-18] Hence, in this study, the mechanical polishing technique was executed. From the dental literature, diamond stylus profilometers (DSPs) and atomic force microscopy (AFM) are the two commonly used methods to analyze SR of denture base resins. For hard materials SR measurements with traditional DSP are adequate. Polymers, lacquers and pure metals have smooth surfaces. For SR measurements on such smooth surfaces, DSP cannot be used because they tend to scratch the specimen's surface and the R_a measured will be meaningless. On the contrary, with AFM, the interaction force between the tip of the probe and the specimen is very small with high spatial resolution.[19] In this research, a new triaxial TriboScanner was used to analyze the SR of the specimens. The interaction force between the Berkovich probing tip and the specimen surface is negligible. Nevertheless, for bacterial colonization, R_a at the nano-metric scale becomes the prime concern. Hence, SR evaluation by TriboScanner is justified.

Concerning SH, experimental groups (SH10 and SH20) possessed higher VHN values than the control group. This can be attributed to residual monomer content which adversely affects SH.[20] The plasticizing effect of residual monomer reduces the polymer inter-chain forces. Hence, indentation appears readily under hardness test loads.[21] Ajay *et al.*[9] reported that the addition of TCDDMDA in MMA decreased the uncured residual monomer in the polymerized specimens. This was confirmed by the disappearance of the carbon–carbon double bond (C = C) peak from the infra-red spectra. The tricyclodecane (TCD) tri-ring central group of TCDDMDA offers a steric hindrance effect. Therefore, it slows down the polymerization rate and facilitates the conversion of monomer into polymer. The decrease in VHN values of control group (SHC) specimens can be ascribed to the progressive polymerization reaction, release of monomer, and monomer's bonding with liberated active oxygen radicals.[22] An increase in VHN for experimental groups can also be attributed to the cross-linking nature of TCDDMDA. This is a difunctional and dual-reactive monomer. Pavlinec *et al.*[23] reported that crosslinking PMMA with polyfunctional monomers increases the SH. Kawaguchi *et al.*[24] concluded that the addition of methacrylated dendrimer cross-linker in the MMA lead to increase SH of the specimens. Porwal *et al.*[22] reported that less decrease in hardness of high impact resins is because of high cross-linking of polymer.

This research is a triple-blinded study. The author, the operator and the statistician were blinded by concealing the concentration of TCDDMDA substituted in

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MMA to avoid expectation bias that might creep into the result. This is the only study with TCDDMDA concerning SR and SH. The denture base acrylic resin is subjected to thermal and cyclic masticatory load stresses in the oral environment that need to be focused in the future experiments. Nonetheless, the effects of various denture cleansers and mechanical denture cleansing directly affect the SR and SH. Hence, future investigations are necessary to evaluate the effect of TCDDMDA more than 20% v/v concentration considering the aforementioned factors.

Conclusion

The conclusions of this study were as follows:

(1) Addition of TCDDMDA in MMA decreased the SR and increased the SH of PMMA.
(2) TCDDMDA at 20% (v/v) concentration in MMA had the smoothest and hardest surface.

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Nil.

Conflicts of interest

There are no conflicts of interest.

References

1. Radford DR, Challacombe SJ, Walter JD. Denture plaque and adherence of *Candida albicans* to denture-base materials *in vivo* and *in vitro*. Crit Rev Oral Biol Med 1999;10:99-116.
2. Bollen CM, Lambrechts P, Quirynen M. Comparison of surface roughness of oral hard materials to the threshold surface roughness for bacterial plaque retention: a review of the literature. Dent Mater 1997;13:258-69.
3. Pinto Lde R, Acosta EJ, Távora FF, da Silva PM, Porto VC. Effect of repeated cycles of chemical disinfection on the roughness and hardness of hard reline acrylic resins. Gerodontology 2010;27:147-53.
4. Asad T, Watkinson AC, Huggett R. The effects of various disinfectant solutions on the surface hardness of an acrylic resin denture base material. Int J Prosthodont 1993;6:9-12.
5. Frangou MJ, Polyzois GL. Effect of microwave polymerization on indentation creep, recovery and hardness of acrylic denture base materials. Eur J Prosthod Restorative Dent 1993;1:111-5.
6. Lee SY, Lai YL, Hsu TS. Influence of polymerization conditions on monomer elution and microhardness of autopolymerized polymethyl methacrylate resin. Eur J Oral Sci 2002;110: 179-83.
7. Abdulrazzaq Naji S, Behroozibakhsh M, Jafarzadeh Kashi TS, Eslami H, Masaeli R, Mahgoli H, *et al*. Effects of incorporation of 2.5 and 5 wt% tio2 nanotubes on fracture toughness, flexural strength, and microhardness of denture base poly methyl methacrylate (PMMA). J Adv Prosthodont 2018;10:113-21.
8. Ajay R, Suma K, Ali SA. Monomer modifications of denture base acrylic resin: a systematic review and meta-analysis. J Pharm Bioallied Sci 2019;11:112-25.
9. Ajay R, Suma K, JayaKrishnaKumar S, Rajkumar G, Kumar SA, Geethakumari R. Chemical characterization of denture base resin with a novel cycloaliphatic monomer. J Contemp Dent Pract 2019;20:940-6.
10. Ranganathan A, Karthigeyan S, Murugesan SV, Kandasamy B, Veeramalai D, Natesan D. Evaluation of *in vitro* cytotoxicity of heat-cure denture base resin processed with a dual-reactive cycloaliphatic monomer. J Contemp Dent Pract 2019;20:1279-85.
11. Zafar MS, Ahmed N. The effects of acid etching time on surface mechanical properties of dental hard tissues. Dent Mater J 2015;34:315-20.
12. Cunha TR, Regis RR, Bonatti MR, de Souza RF. Influence of incorporation of fluoroalkyl methacrylates on roughness and flexural strength of a denture base acrylic resin. J Appl Oral Sci 2009;17:103-7.
13. Regis RR, Zanini AP, Della Vecchia MP, Silva-Lovato CH, Oliveira Paranhos HF, de Souza RF. Physical properties of an acrylic resin after incorporation of an antimicrobial monomer. J Prosthodont 2011;20:372-9.
14. Jiao Y, Ma S, Li J, Shan L, Yang Y, Li M, *et al*. The influences of N-acetyl cysteine (NAC) on the cytotoxicity and mechanical properties of poly-methylmethacrylate (PMMA)-based dental resin. PeerJ 2015;3:e868.
15. Azevedo AM, Regis RR, Chaves CAL, Souza RF, Fernandes RM. Physical properties of a denture base acrylic resins after incorporation of anionic charges. Rev Odontol Bras Central 2010;19:290-94.
16. Al-Rifaiy MQ. The effect of mechanical and chemical polishing techniques on the surface roughness of denture base acrylic resins. Saudi Dent J 2010;22:13-7.
17. Al-Kheraif AA. The effect of mechanical and chemical polishing techniques on the surface roughness of heat-polymerized and visible light-polymerized acrylic denture base resins. Saudi Dent J 2014;26:56-62.
18. Rahal JS, Mesquita MF, Henriques GE, Nóbilo MA. Surface roughness of acrylic resins submitted to mechanical and chemical polishing. J Oral Rehabil 2004;31:1075-9.
19. Cortes-Sandoval G, Martinez-Castanon GA, Patino-Martin N, Martinez-Rodriguez PR, Loyola-Rodriguez JP. Surface roughness and hardness evaluation of some base metal alloys and denture base acrylics used for oral rehabilitation. Mater Lett 2015;144:100-05.
20. Arab J, Newton JP, Lloyd CH. The effect of an elevated level of residual monomer on the whitening of a denture base and its physical properties. J Dent 1989;17:189-94.
21. Ayaz EA, Durkan R, Koroglu A, Bagis B. Comparative effect of different polymerization techniques on residual monomer and hardness properties of PMMA-based denture resins. J Appl Biomater Funct Mater 2014;12:228-33.
22. Porwal A, Khandelwal M, Punia V, Sharma V. Effect of denture cleansers on color stability, surface roughness, and hardness of different denture base resins. J Indian Prosthodont Soc 2017;17:61-7.
23. Pavlinec J, Lazar M, Janigova I. Influence of crosslinking on surface hardness of poly(methyl methacrylate). J Macromol Sci A – Pure Appl Chem 1997;34:81-90.
24. Kawaguchi T, Lassila LV, Vallittu PK, Takahashi Y. Mechanical properties of denture base resin cross-linked with methacrylated dendrimer. Dent Mater 2011;27:755-61.