Flexural Strength of Surface-Treated Heat-Polymerized Acrylic Resin after Repair with Aluminum Oxide-Reinforced Autopolymerizing Acrylic Resin

Abstract

Background: A fracture of denture base *in situ* often occurs through a fatigue mechanism, which over a period of time leads to the formation of small cracks, resulting in fracture. **Aim and Objective:** To evaluate the flexural strength of repaired heat-polymerized acrylic resin, with different percentage of aluminum oxide (Al_2O_3) added to the repair resin and effect of two different surface treatments on the flexural strength of repaired heat-polymerized acrylic resin and also to evaluate quantification of filler particles using scanning electron microscopy. **Materials and Methodology:** Fifty specimens of heat-polymerized acrylic resin were prepared according to the American Dental Association specification no. 12 (65 mm × 10 mm × 2.5 mm). Al_2O_3 <50 nm particle size was silanized using metal alloy primer before incorporation in polymer. Two different percentages of Al_2O_3 nanoparticles, that is, 1% and 1.5% were added to autopolymerizing acrylic resin which was used as repairing material. **Results:** The study showed that repair resin incorporated with 1.5% Al_2O_3 in the group surface treated with silicon carbide paper improved the flexural strength of denture base resin. A proper filler distribution and deep penetration within the polymer matrix were observed by scanning electron microscope in the same group.

Keywords: *Aluminum oxide nanoparticles, flexural strength, polymethyl methacrylate, silane coupling agent*

Shraddha Hareshwar Tamore, K. S. Jyothi[1], Seshagiri Muttagi[2], Amit M. Gaikwad

Department of Prosthodontics and Crown and Bridge, MGM Dental College and Hospital, Navi Mumbai, Maharashtra, [1]Department of Prosthodontics and Crown and Bridge, Coorg Institute of Dental Sciences, Virajpet, Karnataka, [2]Department of Biochemistry, Royal Dental College, Palakkad, Kerala, India

Introduction

Dr. Walter Wright and Vernon brothers introduced polymethyl methacrylate (PMMA) in 1937.[1,2] The fracture of acrylic resin dentures is an unresolved problem in prosthodontics.[3,4] Low resistance to impact, flexural or fatigue, or poor fabrication technique can lead to denture fracture.[5] Several materials have been used to repair fractured acrylic resin dentures, including autopolymerized, heat-polymerized, visible light polymerized, and microwave-polymerized acrylic resins. The most popular one is the use of autopolymerizing resin.[6] The choice of denture base resin and repair material combination may also influence the final strength of denture repair.[7] Bur grinding, airborne-particle abrasion with 250-μm Al_2O_3 particles, carbon dioxide laser application, immersion in methyl methacrylate (MMA), and treatments with organic solvents are some of the techniques already been used.[8]

Adhesion between denture base and repair materials can be improved by first applying appropriate chemicals to the acrylic resin surfaces.[6] Previous studies investigated chemicals such as monomers, chloroform, methylene chloride, acetone, and ethyl acetate as wetting agents.[9] One of the common reinforcing methods is the use of metal wires embedded in the prosthesis. Carbon fibers are useful in strengthening PMMA. Glass fibers improve the mechanical properties, but their high cost limits their routine clinical use.[10] Previously, the addition of alumina into PMMA resulted in increased thermal conductivity and hardness. Silane coupling agent can be used for surface modification of filler which improves the surface bonding of filler and resin matrix.[11,12] Studies have shown that metal oxide nanoparticles have potential for improvement of resin-based dental materials. The results also showed that the antimicrobial properties of nanoparticles containing formulations were increased.[13]

Since only limited amount of data regarding the effect of metal oxide nanoparticles on repair resin with different surface treatments

Address for correspondence:
Dr. Amit M. Gaikwad, Assistant Professor, Department of Prosthodontics and Crown & Bridge, MGM Dental College and Hospital, Kamothe, Navi Mumbai, Maharashtra, India. E-mail: agaikwad1912@gmail.com

are available in literature, this study intends to see the flexural strength of surface-treated heat-polymerized acrylic resin after repair with Al_2O_3-reinforced autopolymerizing acrylic resin for a single repair surface design.

Aim

The aim of this study is to check the flexural strength of repaired heat-cured acrylic resin after addition of aluminum oxide (Al_2O_3) to autopolymerizing repair resin.

Objectives

The purpose of this study was to investigate

1. The flexural strength of repaired heat-polymerized acrylic resin, with 1% and 1.5% of Al_2O_3 added to the repair resin
2. Effect of two different surface treatments, i.e., MMA and silicon carbide paper on the flexural strength of repaired heat-polymerized acrylic resin
3. Quantification of filler particles using scanning electron microscopy.

Materials and Methods

Fifty specimens of heat-polymerized acrylic resin were prepared according to the American Dental Association specification no. 12[11] (65 mm × 10 mm × 2.5 mm). The specimens were prepared using a metal trough of appropriate dimensions. The specimens were sectioned into halves using a disc to create a repair gap (3 mm × 10 mm × 2.5 mm). The sectioned specimens were placed in the metal trough, and a metal die (3 mm × 10 mm × 2.5 mm) was placed in the center of the repair gap for standardization as seen in Figure 1. The repair surface design for all specimens was butt joint. The fractured surfaces were treated with MMA for 180 s (20 samples) and with silicon carbide papers (20 samples) in ascending order of grit sizes, i.e., 100, 300, and 600 in order to get a uniform smooth surface. Control group had specimens without any filler particles and without any surface treatment (10 samples).

Al_2O_3 <50 nm particle size (Sigma-Aldrich) was silanized using metal alloy primer before incorporation in polymer [Figure 2]. Two different percentages of Al_2O_3 nanoparticles, that is, 1% and 1.5% were added to autopolymerizing acrylic resin which was used as repairing material. After silanization, the filler particles were incorporated in the monomer (1% and 1.5% by weight of repair resin). The monomer containing Al_2O_3 nanoparticles was mixed with polymer in the ratio of 1:3 by volume, and the fractured specimens were repaired [Figure 3].

The specimens were divided into three groups, that is, A, B, and C according to their filler particle percentage.
- Group A (control group) without any filler particle and with no surface treatment (*n* = 10) [Figure 4]
- Group B with 1% Al_2O_3 in repair resin material [Figure 5]
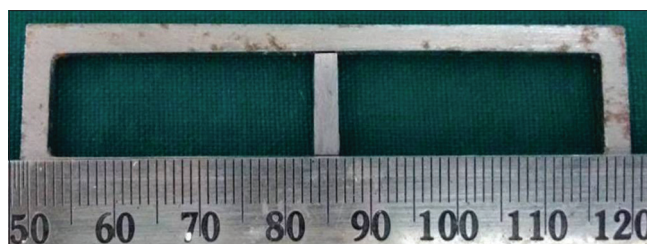 - Group B1 – Surface treated with MMA for 180 s (*n* = 10)

Figure 1: Metal die placed in the trough for maintaining the 3-mm repair gap

Figure 2: Mixture ready for silanization process

Figure 3: Repaired heat-cured specimen

- Group B2 – Surface treated with silicon carbide paper (*n* = 10)
- Group C with 1.5% Al_2O_3 in repair resin material [Figure 6]
  - Group C1 – Surface treated with MMA for 180 s (*n* = 10)
  - Group C2 – Surface treated with silicon carbide paper (*n* = 10).

Specimens were stored in distilled water at 37°C for 7 days to simulate use in oral environment. Flexural strength of the specimens was determined using three-point bending test in a universal testing machine. A load was applied in the center of the specimens (center of the repair area) at the crosshead speed of 5 mm/min. The specimens were loaded until the first sound of a crack was detected. The flexural
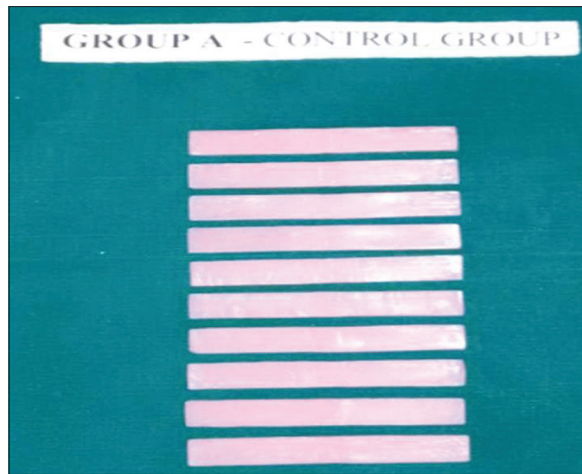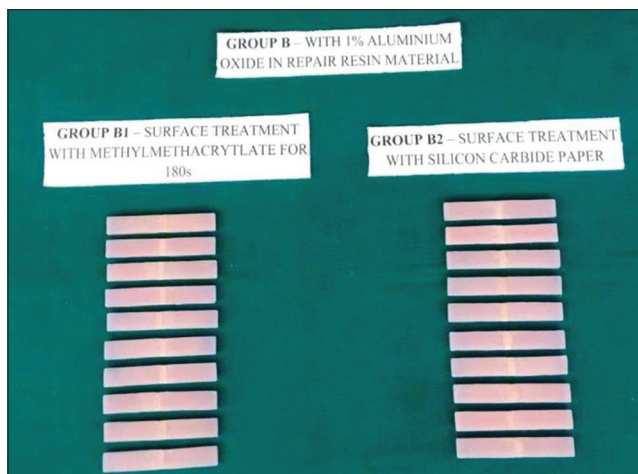
Figure 4: Control group

Figure 5: Specimens with 1% aluminum oxide in repair resin material with two different surface treatments
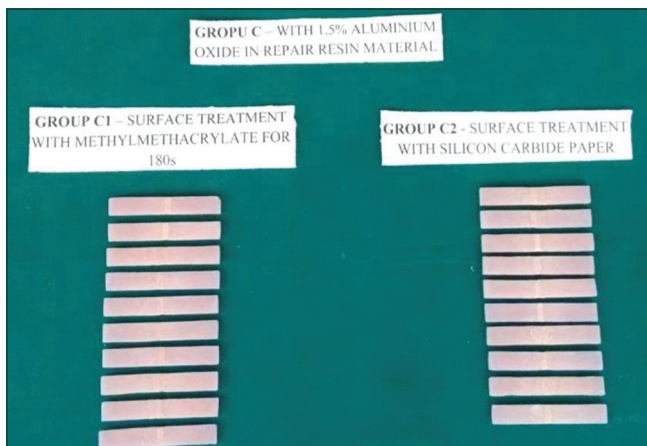

Figure 6: Specimens with 1.5% aluminum oxide in repair resin material with two different surface treatments

strength values of each specimen were calculated with the following formula:

$S = 3WL/2bd^2$.

Where, S – Flexural strength (in Megapascals)

W – Fracture load (in Newtons)

L – Distance between the supports (50 mm)

B – Specimen width (10 mm)

D – Specimen thickness (2.5 mm).

To evaluate the effects of the surface treatments and repair resins on the surface of the denture base resin, one specimen from each group was selected after three-point bending test. The fractured surface of specimens was subjected to scanning electron microscope (SEM). These selected specimens were gold sputtered and examined under a field emission SEM at 10.0 kV.

Results

The results show that there is a significant difference in the mean flexural strength between the test groups and the control and between all the test groups. For the surface treatment with MMA, when the mean flexural strength of Groups B1 and C1 was compared, the results were highly significant ($P < 0.05$) [Figure 7]. Repair resin material with 1% Al_2O_3 showed better results. However, when mean flexural strength of Groups B2 and C2 was compared, the results were not significant [Figure 8]. To satisfy the second objective, effect of the surface treatments was compared within 1% group and within 1.5% group. For the 1% Al_2O_3 group, the results were not significant. A highly significant value was seen when flexural strength of 1.5% Al_2O_3 group with two different surface treatments was compared. Repair resin incorporated with 1.5% Al_2O_3 and the group surface treated with silicon carbide paper showed better results. An SEM study was done to quantify the distribution of filler particles to satisfy the third objective. Group B1 showed voids in between the filler particles, whereas for Group B2, a number of dense agglomerates of filler particles were seen [Figure 9]. Group C1 showed superficially placed nanoparticles. In Group C2, a deep and uniform distribution of filler particles throughout the repair resin material was seen [Figure 10].

Discussion

Combination of tensile and compressive strengths is the flexural strength which is relevant to the masticatory loading characteristics of a denture base in the clinical situation.[9,14] Important factors which lead to fracture are low resistance to impact, flexural or fatigue, or poor fabrication technique.[5,14] Any repairing procedure should be quick, dimensionally stable, cost-effective and should have adequate strength and color.[8] Al_2O_3, also referred as alumina, possesses strong ionic interatomic bonding with most stable hexagonal alpha phase at elevated temperature. By addition of alumina, there was proper distribution and bonding of the filler within the matrix which improved the flexural strength of the polymer tested.[15] Porosity weakens acrylic resin prosthesis and it has been reported that introduction of nanosized metal oxide pigments reduces

porosity and yields a better dispersion.[13,16] Thus, in this study, nanosized Al_2O_3 particles were incorporated in repair resin material to evaluate the flexural strength of repaired material.

To achieve good bonding between filler and resin matrix, modification of filler surface is necessary which results in the improved strength of materials.[11] Silane coupling agent improves surface bonding with resin matrix. Tri-methacryloxypropyltrimethoxysilane (MPS) modifies the surface of silica and glass filler blended with resin matrix.[11,17,18] MPS was selected based on its solubility parameters for MMA and PMMA bonding. In a previous study, Al_2O_3 powder was incorporated from 5% to 20% by weight into conventional heat-polymerized denture base resin which resulted in an increase in both flexural strength and thermal diffusivity.[15] However, in this study, 1% and 1.5% concentration of nano-Al_2O_3 was added in repair resin.

Alkurt *et al.*[8] established that the transverse strength of the repaired specimen can be increased by immersing them in MMA for 180 s. Exposure to monomer softened the PMMA and formed pits in the bond surface which helps the repair material to diffuse into the bond surface and develops adhesion. Therefore, MMA was taken as one of the methods for surface treatment. Other methods used were manual grinding with silicon carbide paper as it was said that it modified the surface morphology and enhances mechanical retention.[6]

A repair gap of 3 mm was used in this study as it decreased the degree of polymerization contraction and also reduced any color difference between the denture base and repair material.[8] Ayaz and Durkan[10] observed that mechanical strength of the specimens reached optimum strength after immersing them in water for 1 day–1 week. Hence, the specimens were stored in distilled water at 37ºC for 7 days.

Statistical significant difference was found when the control group was compared with Al_2O_3 added group. In both the percentage groups, that is, 1% and 1.5%, better flexural strength value was seen in Group C2. This could be explained by the fact that the surface morphologic changes could have enhanced the mechanical retention between the fractured surface and repaired acrylic resin as seen in previous study done by Alkurt *et al.*[8] The same results can be explained by the SEM image which shows a uniform distribution and deep penetration of nano-Al_2O_3 particles in repair resin as compared to superficially placed agglomerates of nanoparticles seen in Group B2 [Figures 11 and 12]. When the mean flexural strength was compared between Groups B1 and C1, higher values were seen for Group B1 which could be explained by stating that an increase in filler fraction does not necessarily lead to an increase in strength, because higher filler fractions create more defects that weaken the materials. Low filler fractions used might have caused higher strength values
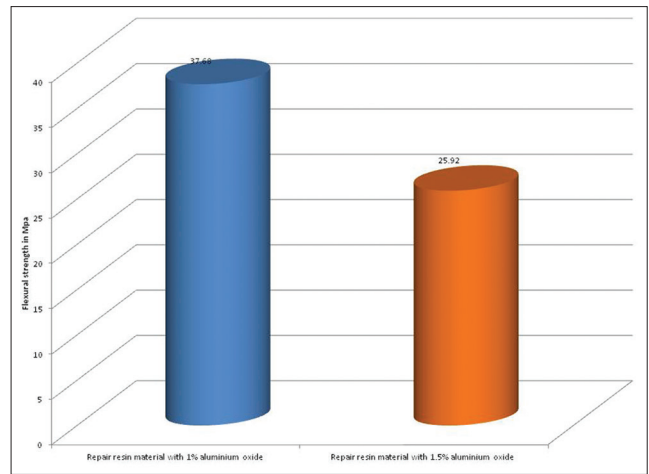

Figure 7: Mean flexural strength of specimens with 1% and 1.5% aluminum oxide in repair resin for the group surface treated with methyl methacrylate for 180 s
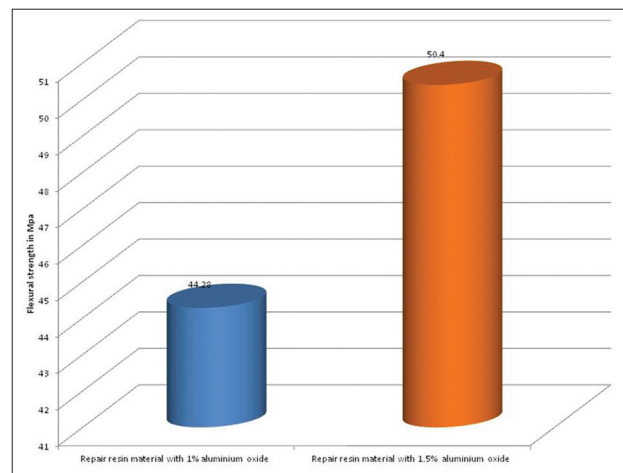

Figure 8: Mean flexural strength of specimens with 1% and 1.5% aluminum oxide in repair resin for the group surface treated with silicon carbide paper
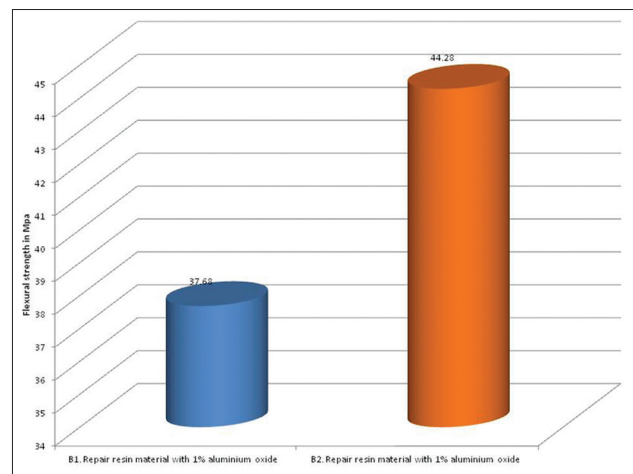

Figure 9: Mean flexural strength of Group B – repair resin incorporated with 1% Al_2O_3, in two surface treatment groups

obtained in the test groups.[19] On the other hand, when the mean flexural strength was compared between Groups

B2 and C2, higher values were seen for Group C2, which showed that surface treatment played an important role in bonding of Al_2O_3 nanoparticle-incorporated repair resin to heat-polymerized PMMA resin [Figures 13 and 12].

The material treated with MMA (Groups B1 and C1) may form microscopic pores which caused penetration of Al_2O_3 particles [Figures 14 and 15]. As this Al_2O_3 is a positively charged molecule which might form ionic bond between the molecules (i.e. PMMA powder and Al_2O_3 particles), and thus gave extra flexural strength to the material in comparison to the control. When MMA-treated groups were compared with silicon carbide paper-treated group, MMA group showed less flexural strength and this could be due to the fact that monomer is not a powerful solvent for PMMA and would therefore not remove the debris efficiently. However, when the material is treated with silicon carbide paper, there may not be a formation of pores, and moreover, ionic interaction may be less. Therefore,

any difference that was found between the two percentage groups for silicon carbide paper treatment is only because of the increased concentration of Al_2O_3, and there may be chances that greater number of Al_2O_3 molecules comes in close contact with PMMA material which may also help in formation of ionic bond between molecules. Hence, this group, that is, 1.5% treated with silicon carbide paper showed highly significant results. Silicon carbide paper treatment causes only physical removal of material to give microporosities without damaging structural matrix like MMA. Probably, this is the reason why silicon carbide paper group showed better flexural strength compared with MMA. When two surface treatments were compared within 1.5% group, it was found that MMA treatment decreased the flexural strength. This was probably because higher percentage of unreacted filler particles remained on the surface, thereby preventing proper bonding with the repair resin. Similar results were seen when surface treatments were compared within 1% group. Adhesive fractures were the most common type of failure to occur in the present
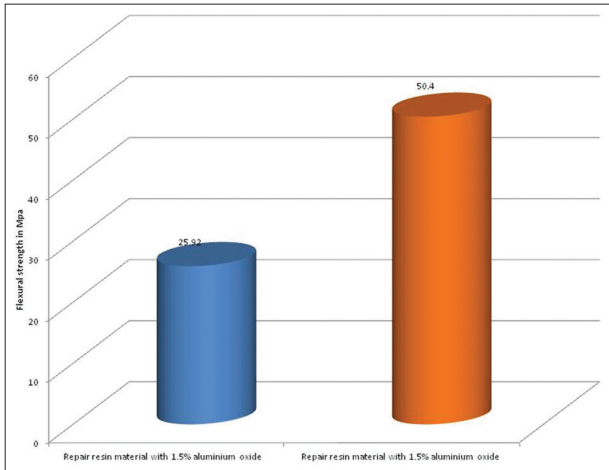

Figure 10: Mean flexural strength of Group C – repair resin incorporated with 1.5% Al_2O_3, in two surface treatment groups
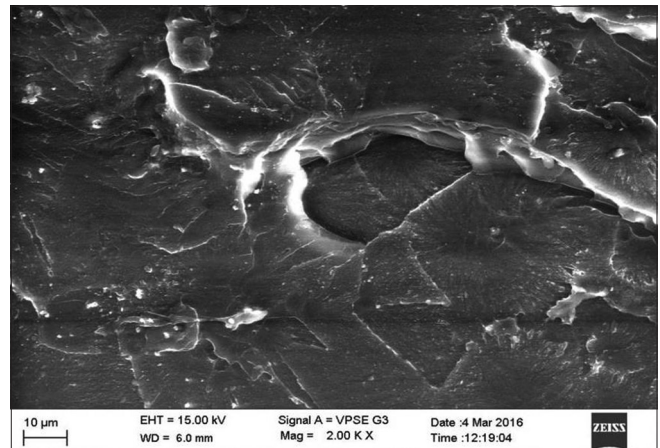

Figure 11: Scanning electron microscope image of sectioned specimens of control group
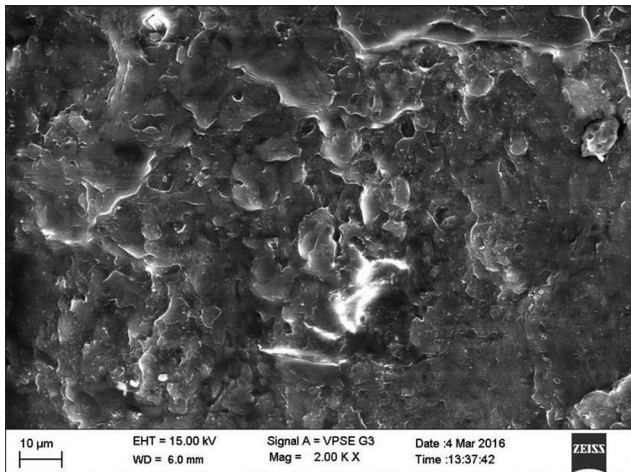

Figure 12: Scanning electron microscope image of sectioned specimens incorporated with 1.5% aluminum oxide for the group surface treated with silicon carbide paper
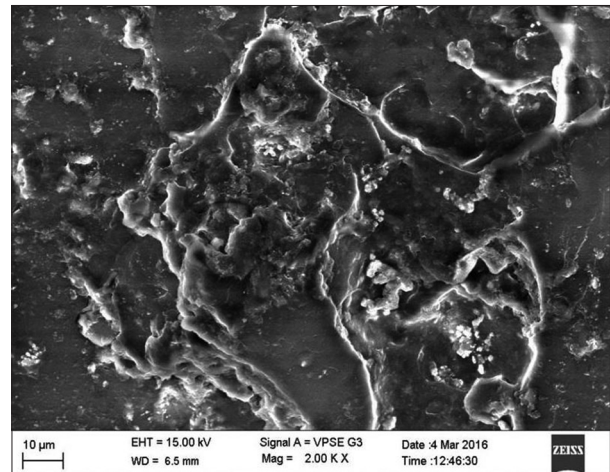

Figure 13: Scanning electron microscope image of sectioned specimens incorporated with 1% aluminum oxide for the group surface treated with silicon carbide paper
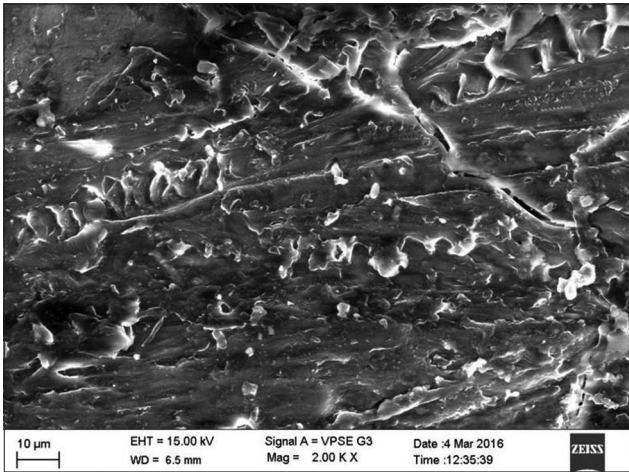
Figure 14: Scanning electron microscope image of sectioned specimens incorporated with 1% aluminum oxide for the group surface treated with methyl methacrylate for 180 s
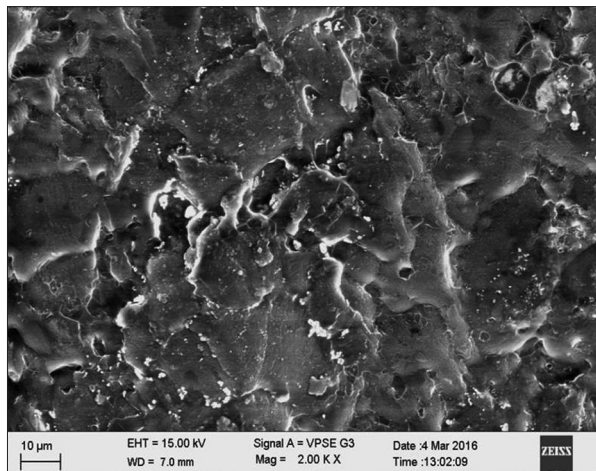

Figure 15: Scanning electron microscope image of sectioned specimens incorporated with 1.5% aluminum oxide for the group surface treated with methyl methacrylate for 180 s

study, indicating that the overall poor bond strengths were achieved between repair material incorporated with Al_2O_3 particles and PMMA denture base polymer.

The limitations of the study include the absence of artificial aging with thermal cycling and the use of rectangular specimens instead of more complex denture shapes. *In vitro* studies are limited in their ability to predict the success of a material or technique in a clinical situation. Further *in vitro* studies and clinical research are necessary to investigate the effects of different surface treatments and incorporation of various metal oxide nanoparticles on the physical and mechanical properties of repaired acrylic resin.

Conclusion

Within the limitations of the study, it could be concluded that:

- Repaired heat-polymerized acrylic resin incorporated with 1.5% Al_2O_3 in the group surface treated with

silicone carbide paper showed the highest flexural strength
- Silicon carbide paper surface treatment showed increased flexural strength as compared to MMA
- A proper filler distribution and deep penetration within the polymer matrix were seen in specimens incorporated with 1.5% Al_2O_3 and in the group surface treated with silicone carbide paper as seen by SEM.

Financial support and sponsorship

Conflicts of interest

There are no conflicts of interest.

References

1. Arora N, Jain V, Chawla A, Mathur VP. Effect of addition of sapphire (aluminium oxide) or silver fillers on the flexural strength thermal diffusivity and water sorption of heat polymerized acrylic resins. IJOPRD 2011;1:21-7.
2. 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.
3. Polyzois GL, Andreopoulos AG, Lagouvardos PE. Acrylic resin denture repair with adhesive resin and metal wires: Effects on strength parameters. J Prosthet Dent 1996;75:381-7.
4. Ajaj-Alkordy NM, Alsaadi MH. Elastic modulus and flexural strength comparisons of high-impact and traditional denture base acrylic resins. Saudi Dent J 2014;26:15-8.
5. Hamza T, Wee AG, Alapati S, Schricker SR. The fracture toughness of denture base material reinforced with different concentrations of POSS. J Macromol Sci A Pure Appl Chem 2004;A41:897-906.
6. Sarac YS, Sarac D, Kulunk T, Kulunk S. The effect of chemical surface treatments of different denture base resins on the shear bond strength of denture repair. J Prosthet Dent 2005;94:259-66.
7. Seó RS, Neppelenbroek KH, Filho JN. Factors affecting the strength of denture repairs. J Prosthodont 2007;16:302-10.
8. Alkurt M, Yeşil Duymuş Z, Gundogdu M. Effect of repair resin type and surface treatment on the repair strength of heat-polymerized denture base resin. J Prosthet Dent 2014;111:71-8.
9. Bural C, Bayraktar G, Aydin I, Yusufoğlu I, Uyumaz N, Hanzade M, *et al.* Flexural properties of repaired heat-polymerising acrylic resin after wetting with monomer and acetone. Gerodontology 2010;27:217-23.
10. Aydogan Ayaz E, Durkan R. Influence of acrylamide monomer addition to the acrylic denture-base resins on mechanical and physical properties. Int J Oral Sci 2013;5:229-35.
11. Chaijareenont P, Takahashi H, Nishiyama N, Arksornnukit M. Effect of different amounts of 3-methacryloxypropyltrimethoxysilane on the flexural properties and wear resistance of alumina reinforced PMMA. Dent Mater J 2012;31:623-8.
12. Kamble VD, Parkhedkar RD, Mowade TK. The effect of different fiber reinforcements on flexural strength of provisional restorative resins: An *in vitro* study. J Adv Prosthodont 2012;4:1-6.
13. Acosta-Torres LS, Lopez-Marin LM, Nunez-Anita RE, Hernandez-Padron G, Castano VM. Biocompatible metal-oxide

nanoparticles: Nanotechnology improvement of conventional prosthetic acrylic resins. J Nanomater 2011;2011:941561.

14. Diaz-Arnold AM, Vargas MA, Shaull KL, Laffoon JE, Qian F. Flexural and fatigue strengths of denture base resin. J Prosthet Dent 2008;100:47-51.

15. Ellakwa AE, Morsy MA, El-Sheikh AM. Effect of aluminum oxide addition on the flexural strength and thermal diffusivity of heat-polymerized acrylic resin. J Prosthodont 2008;17:439-44.

16. Ahmed MA, Ebrahim MI. Effect of zirconium oxide Nano-fillers addition on the flexural strength, fracture toughness, and hardness of heat-polymerized acrylic resin. WJNSE 2014;4:50-7.

17. Kanie T, Arikawa H, Fujii K, Ban S. Flexural properties of denture base polymers reinforced with a glass cloth-urethane polymer composite. Dent Mater 2004;20:709-16.

18. Kanie T, Fujii K, Arikawa H, Inoue K. Flexural properties and impact strength of denture base polymer reinforced with woven glass fibers. Dent Mater 2000;16:150-8.

19. Asar NV, Albayrak H, Korkmaz T, Turkyilmaz I. Influence of various metal oxides on mechanical and physical properties of heat-cured polymethyl methacrylate denture base resins. J Adv Prosthodont 2013;5:241-7.