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EFFECT OF REINFORCEMENT OF HIGH-IMPACT ACRYLIC RESIN WITH ZIRCONIA ON SOME PHYSICAL AND MECHANICAL PROPERTIES

Efeito do reforço de resinas acrílicas de alto impacto com zircônia em algumas propriedades físicas e mecânicas

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Abstract

OBJECTIVES: To evaluate the effect of reinforcing high-impact acrylic resin (Metrocryl HI) with zirconia powder in two different concentrations on the transverse strength, impact strength, surface hardness, water sorption and solubility. **MATERIALS AND METHODS**: Fifteen specimens were prepared for each test, five from Metrocryl HI resin as control, five others were prepared from the 5% zirconia-modified Metrocryl HI resin, and another five specimens were prepared from the 15% zirconia-modified Metrocryl HI resin. Transverse strength was assessed with a 3-point bending test using a screw-driven testing machine. For the impact strength, a Charpy tester was used. Surface hardness testing was conducted using Vickers tester. The water sorption and solubility test was performed according to International Standards Organization specification No. 1567 for denture base polymers. Micro-structural examination of two specimens, one representing each ZrO_2 concentration group, was carried out using scanning electron microscopy. **RESULTS**: As determined by ANOVA and Bonferroni multiple comparison tests, the addition of zirconia resulted in a highly significant increase in transverse strength of high-impact acrylic resin ($P< 0.001$). This increase was proportional to the concentration of zirconia. No significant difference was detected in each of impact strength, surface hardness and water solubility ($P> 0.05$). Although a significant increase in water sorption was demonstrated ($P< 0.05$), still its mean value lies within the specification limits. **CONCLUSIONS**: Reinforcement of high-impact acrylic resin with zirconia powder increases its transverse strength significantly.

Keywords: High impact acrylic resins; Zircônia; Transversal strength.

Resumo

OBJETIVOS: *Avaliar o efeito do reforço de resina acrílica de alto impacto (Metrocryl HI) com pó de zircônia em duas diferentes concentrações, a forças transversais, de impacto, dureza de superfície, absorção de água e solubilidade.* ***MATERIAIS E MÉTODOS***: *15 espécimes foram preparados para cada teste, sendo cinco com resina Metrocryl HI como controle, cinco preparados com resina modificada em 5% e cinco preparados com resina modificada em 15%. As forças transversas foram determinadas com um teste de dobramento por máquina de parafuso. Para as forças de impacto, um testgador Charpy foi usado. A dureza de superfície foi verificada com teste de Vickers. A absorção de água foi testada de acordo com especificações n.o 1 567 da Organização Internacional de Standards para polímeros de bases de dentaduras. O exame microestrutural dos dois espécimes, um representando cada grupo de concentração de ZrO2, foi efetuado usando microscopia eletrônica.* ***RESULTADOS***: *Testes ANOVA e Bonferroni de comparação múltipla demonstraram que houve um aumento significante na resistência transversal das resinas de alto impacto (P<0.05). Este aumento foi proporcional na concentração de zircônia. Não houve diferenças significativas na resistência ao impacto, na dureza superficial e na solubilidade na água (P>0.05), permanecendo dentro dos limites das especificações.* ***CONCLUSÕES***: *O reforço de resinas de alto impacto com zircônia em pó aumenta significativamente a resistência transversal.*

Palavras-chave: *Resinas acrílicas de alta resistência; Zircônia; Resistência transversal.*

INTRODUCTION

Polymethyl methacrylate (PMMA) is one of the most widely used materials in prosthetic dentistry. Since it was introduced to dentistry, it has been successfully used in construction of denture bases. Although matching the appearance of the oral soft tissues, PMMA has poor strength characteristics and liability to fracture on accidental dropping (1). A study by Johnston et al. (2) showed that 68% of acrylic resin dentures break within a few years after fabrication. This occurs when the denture is accidentally dropped on a hard surface or fractures when subjected to high mastication forces (3, 4).

Approaches to strengthening acrylic resin polymer have included the incorporation of metal powder fillers. Silver, copper, and aluminum particles were added to PMMA. Although improving strength, (5) metal-reinforced dentures were found to be unesthetic. Modifications of chemical structure of acrylic resin by copolymerization with rubber have been attempted (6). The graft copolymers of rubber methacrylate are high-impact resins (7). Although the impact strength increased by such modifications, the transverse strength was found to be reduced (8).

The incorporation of zirconia in various dental materials has been studied and it was found to be biocompatible and that it improved mechanical properties (9-16). Also, the white color of zirconia powder is not expected to compromise the esthetic appearance. To date, no studies have examined the effects of incorporating zirconia in a high-impact denture base resin. Therefore, the aim of this study was to evaluate the effect of incorporating zirconia in two different concentrations on transverse strength, impact strength, surface hardness, water sorption and solubility of high-impact acrylic resin.

MATERIALS AND METHODS

A high-impact heat-cured acrylic resin (Metrocryl HI, Metrodent, Paddock, Huddersfield, UK), Zirconia powder (Zirconium oxide, Promochem GmbH Postfach, Wesel, Germany), and Zirconate coupling agent (KEN-REACT NZ 33, Kenrich petrochemicals Inc., Bagonne, NJ, USA) were used in this study.

Preparation of modified high-impact powder

A solution of 0.3 g of zirconate coupling agent in 100 ml of acetone was used to treat 30 g of zirconia powder (ZrO_2). The oxide powder was stirred in the coupling agent/acetone solution with a magnetic stirrer (ECM 5, Finemech Inc., CA, USA), for 60 minutes, after which acetone was completely evaporated using a rotary evaporator (Rotavapor R-200, Buchi Inc., Postfach, Switzerland) (9). Weighing of ZrO_2 powder was done using an electronic balance (Mettler Instrument AG, Greifensess, Switzerland). ZrO_2 powder was added in a concentration of 5% and 15% of the acrylic polymer powder weight. Thorough mixing of the oxide powder with the acrylic polymer powder was carried out using porcelain mortar and pestle, then, the monomer was added to the mixture (5).

Specimens' preparation

Fifteen specimens were prepared for each test according to the manufacturer's instructions. Five specimens were prepared from Metrocryl HI resin (HI) as control, five others were prepared from the 5% zirconia-modified Metrocryl HI resin (5%Zr-HI), and another five specimens were prepared from the 15% zirconia-modified Metrocryl HI resin (15%Zr-HI). After processing, resin specimens designed for strength testing were finished then stored in deionized water at room temperature for 2 weeks before testing.

Transverse strength testing

The transverse strength test (TS) was conducted according to International Standards Organization specification 1567 for denture base polymers (17). Rectangular-shaped specimens (65 x 10 x 2.5) were prepared. Testing was conducted under 3-point loading, with a crosshead speed of 5mm/min. using a screw-driven testing machine (Lloyd Lr 30K, Lloyd Instruments Ltd, Fareham, U.K.). Testing method was similar to that used by others (18, 19).

Impact strength testing

Rectangular-shaped specimens (60 x 7 x 4 mm) were prepared for impact strength testing (IS). Strength test method and specimens dimensions were similar to those used by Uzun et al (20). Using a notch cutter (Hounsfield notching machine, Tensometer Ltd., Croydon, U.K.), a 3.5 mm notch was prepared in each specimen. A Charpy-type impact tester (Hounsfield plastic impact machine, Tensometer Ltd.) was used to apply force to the specimens from the un-notched side. During testing those specimens that did not fracture at the first trial were excluded from the study.

Surface hardness testing

Rectangular-shaped specimens (30 x 10 x 2.5 mm) were prepared and measured for surface hardness (SH) using a digital Vickers microhardness tester (MXT70, Matsuzawa, Tokyo, Japan.). A 50 gf load was applied by the diamond pyramid for 5 seconds indentation time and VHN was obtained digitally.

Water sorption and solubility testing

Fifteen disc-shaped specimens (50 mm diameter, 0.5 mm thickness) were prepared according to International Standards Organization specification 1567 for denture base polymers (17). They were placed inside a desiccator (Wheaton, Millville, N.J.) at $37^oC \pm 1^oC$ for 23 hours, then, allowed to stand at ambient temperature for 1 hour. Each specimen was weighed by an electronic balance (Mettler Instrument AG, Greifensess, Switzerland), and the previously described cycle was repeated until the specimens reached constant mass. Specimens were immersed in distilled water at $37^oC \pm 1^oC$ for 7 days, then, they were removed from the water, weighed, and reconditioned to constant mass in the desiccator. Water sorption (WR) and solubility (WL) were calculated in mg/cm^2.

Scanning electron microscopic examination

Two specimens, one representing 5% Zr-HI and the other representing 15% Zr-HI, were sputter-coated with gold to a thickness of approximately 50 A^o in a vacuum evaporator (MED 010, Balzer Union, Balzers, Liechtenstein). SEM observation was conducted (SEMx1000) at 10 KV using (ISM-840A, JEOL, Tokyo, Japan).

Statistical analysis

Data entry and analyses were performed using SPSS statistical software program (SPSS Version 10 (SPSS, Inc., Chicago, IL, USA). The quantitative data were presented as means and standard deviations. One-way analysis of variance (ANOVA) at the 95% confidence level was used to examine variable effects. The test was considered significant when P<0.05 and highly significant when P<0.001. Bonferroni multiple comparison test was used to determine which group means were significantly different.

RESULTS

The mean values and standard deviations of the TS, IS, SH, WR and WL of the tested groups are shown in Table 1. Specimens reinforced with 15% ZrO_2 showed the highest transverse strength, followed by 5% Zr-HI specimens. Zirconia-free specimens exhibited the lowest TS values. Specimens that contained zirconia and Zirconia-free specimens showed close impact strength, surface hardness as well as water solubility values. Water sorption value of 15% ZrO_2 was higher than that of 5% ZrO_2 and that of zirconia-free specimens.

TABLE 1 - Means, standard deviation, ANOVA and Bonferroni comparison results of tests

	HI	5% Zr-HI	15%Zr- HI	ANOVA		Bonferroni multiple comparison		
	X ± SD	X ± SD	X ± SD	F	P	P_1	P_2	P_3
Transverse Strength (MPa)	79.1±1.148	101.8±1.55	139.3±13.9	70.18	0.00*	0.003*	0.000*	0.000*
Impact strength (J)	2.0 ± 0.13	1.85 ± 0.13	1.82 ± 0.08	2.102	0.203	0.499	.308	1.000
Surface Hardness (VHN)	16.6±1.627	15.9±1.018	17.6±1.866	1.454	0.272	1.000	1.000	0.346
Water sorption (mg/cm²)	0.55 ± 0.06	0.58 ± 0.02	0.66 ± 0.01	12.96	0.001*	0.446	0.001*	0.015*
Water solubility (mg/cm²)	0.01 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	2.600	0.115	1.000	0.379	0.147

± indicates the standard deviation value
* Significant P<0.05
P_1: HI & 5% Zr-HI
P_2: HI & 15% Zr-HI
P_3: 5% Zr-HI & 15% Zr-HI

From Table 1, one-way ANOVA revealed highly significant differences between the tested groups (P< 0.001) for transverse strength test. No significant difference was detected in each of impact strength, surface hardness and water solubility (P> 0.05). Although significant changes in water sorption values were demonstrated (P< 0.05), still their mean values lie within the specification limits. The results of Bonferroni multiple comparison test are displayed in Table 1 for TS, IS, SH, WR and WL. This test revealed significant differences between all the test groups in TS testing. The significant increase in water sorption was confirmed between 15% Zr-HI specimens and each of the other two groups. Figures 1, 2 and 3 represent SEM micro-structural examination of HI, 5% Zr-HI and 15% Zr-HI specimens respectively.

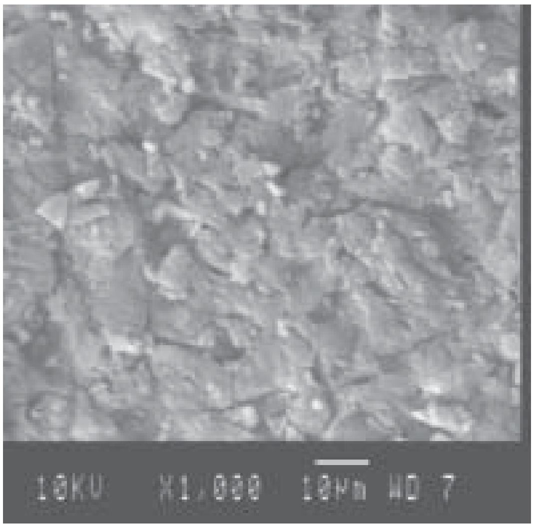

FIGURE 1 - SEM micro-structural image (x 1000) of HI

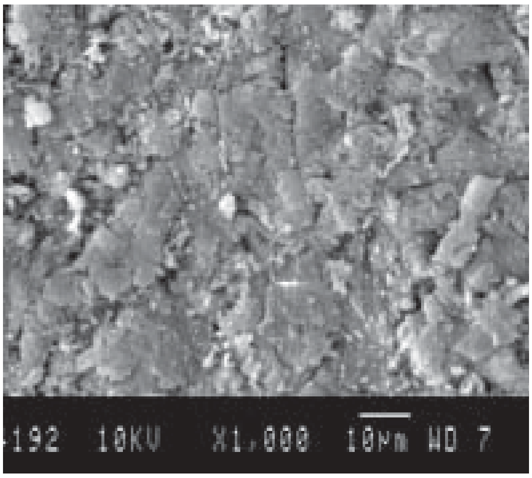

FIGURE 2 - SEM micro-structural image (x 1000) of 5% Zr-HI

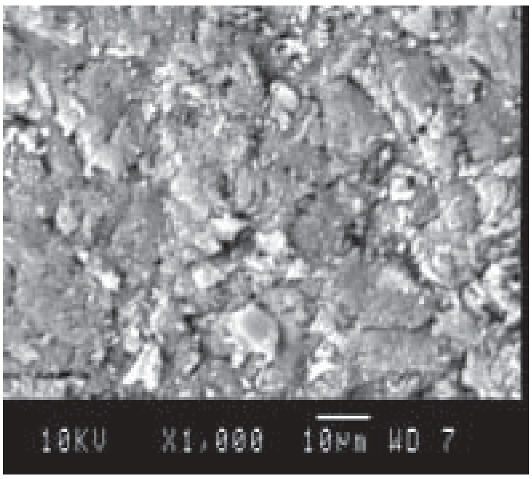

FIGURE 3 - SEM micro-structural image (x 1000) of 15% Zr-HI

DISCUSSION

Denture base resins with high impact strength should also withstand high loads of mastication. This study evaluated the effects of reinforcement with zirconia in two concentrations on the transverse strength, impact strength, surface hardness, water sorption and solubility of a high-impact acrylic resin, (Metrocryl HI). Zirconium oxide white colored powder that is not expected to adversely affect the esthetic appearance of the denture base has been selected for this study as it was stated by others (14-16) to be biocompatible, and able to improve fracture resistance of dental materials.

In this study, zirconia powder was used in a concentration of 5% and 15% after being pretreated with zirconate coupling agent to obtain a chemical bond between zirconium oxide and acrylic resin powder. The transverse strengths of the test groups were measured using 3-point bending test. The impact strength was also measured using Charpy tester to determine whether adding zirconia had an effect on the impact strength of the high-impact resin or not. Also, surface hardness was measured using a digital Vickers micro hardness tester. Water sorption and solubility were measured to detect changes in the properties of the high-impact resin after being reinforced with zirconia.

The results of this study showed that TS of 5% Zr-HI increased by 29 % compared to zirconia-free Metrocryl HI, meanwhile TS of 15% Zr-HI increased by 76 %. This increase in transverse strength can be explained on the basis of transformation toughening. When sufficient stress develops and a crack begins to propagate, a transformation of ZrO_2 from the metastable tetragonal crystal phase to the stable monoclinic phase occurs which depletes the energy of crack propagation. Also, in this process, expansion of ZrO_2 crystals occurs and places the crack under a state of compressive stress and crack propagation is arrested (21). Both mechanisms improve fracture resistance of denture base under applied mastication loads.

The results of this study are in agreement with the findings reported by others (10-13) who concluded that reinforcement of ceramics, dental restorative resins as well as acrylic resin with zirconia could exhibit improvement in their mechanical properties.

The results of this study also revealed that the impact strength, hardness as well as water solubility of high-impact acrylic resin did not change significantly after being reinforced with zirconia powder for any of the concentrations used.

Many studies have been conducted on water sorption of denture base resins, and it has been concluded that increased water sorption may cause a decrease in mechanical properties (22). The results of this study showed significant increases in water sorption of 15% Zr-HI when compared to zirconia-free high-impact resin (P< 0.05), but still its mean value lies within the specification limits (17). These results are in agreement with those of others (23) who found that addition of reinforcing particles generally increased water sorption by acrylic resin.

CONCLUSIONS

Within the limits of this study, the following conclusions were drawn:

1. The transverse strength of Metrocryl HI high-impact denture base resin can be increased significantly by a factor of 29% and 76% when reinforced with zirconia in a concentration of 5% and 15% respectively.
2. The impact strength, surface hardness as well as water solubility of the zirconia-reinforced high-impact resin were not significantly different from that of zirconia-free high-impact resin.
3. Although the water sorption values of zirconia-reinforced high-impact resin were significantly different from that of zirconia-free high-impact resin, still their values lie within the specifications limits.

Further studies are recommended to evaluate the effect of reinforcement with zirconia on mechanical properties of other high-impact acrylic resins systems, stored in water for longer periods of time that may affect the mechanical properties (23).

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REFERENCES

1. Gutteridge DL. The effect of including ultra-high modulus polyethylene fiber on the impact strength of acrylic resin. Br Dent J. 1988;164(6):177-80.
2. Johnston EP, Nicholls JI, Smith DE. Flexural fatigue of 10 commonly used denture base resins. J Prosthet Dent. 1981;48(5):478-83.
3. Kim SH, Watts DC. The effect of reinforcement with woven E-glass fibers on the impact strength of complete dentures fabricated with high-impact acrylic resin. J Prosthet Dent. 2004;91(3):274-80.
4. Manley TR, Bowman AJ, Cook M. Denture bases reinforced with carbon fibers. Br Dent J. 1979;146(1):25-5.
5. Sehajpal SB, Sood VK. Effect of metal fillers on some physical properties of acrylic resin. J Prosthet Dent. 1989;61(6):746-51.
6. Matsukawa S, Hayakawa T, Nemoto K. Development of high-toughness resin for dental application. Dent Mater. 1994;10 (6):343-6.
7. Stafford GD, Bates JF, Huggett R, Handley RW. A review of the properties of some denture base polymers. J Dent. 1980;8 (4):292-306.
8. Jagger DC, Harrison A, Jandt KD. Review: the reinforcement of dentures. J Oral Rehabil. 1999;26(3):185-94.
9. Yoshida K, Greener EH. Effects of coupling agents on mechanical properties of metal oxide polymethacrylate composites. J Dent. 1994;22(1):57-62.
10. Giodano R. A comparison of all-ceramic restorative systems: Part 2. Gen Dent. 2000;48(1):43-8.
11. Furman B, Rawls HR, Wellinghoff S, Dixon H, Lankford J, Nicolella D. Metal-oxide nanoparticles for the reinforcement of dental restorative resins. Crit Rev Biomed Eng. 2000;28(3, 4):439-43.

12. Zuccari AG, Oshida Y, Moore BK. Reinforcement of acrylic resins for provisional fixed restorations. Part I: Mechanical properties. Biomed Mater Eng. 1997;7(5):327-43.

13. Tinschert J, Natt G, Mautsch W, Augthun M, Spiekerman H. Fracture resistance of lithium disilicate-, alumina-, and zirconia-based three unit fixed partial dentures: a laboratory study. Int J Prosthodont. 2001;14(3):231-8.

14. Ichikawa Y, Akagawa Y, Nikai H, Tsuru H. Tissue compatibility and stability of a new zirconia ceramic in vivo. J Prosthet Dent. 1992;68(2):322-6.

15. Minamizato T. Slip-cast zirconia dental roots with tunnels drilled by laser process. resin. J Prosthet Dent. 1990;63(6):677-84.

16. William D, Callister JR. Materials science and engineering: an introduction. 4th ed. New York: John Wiley & Sons, Inc.; 1997. p. 532-3.

17. International Organization for Standardization. Specification 1567: denture base polymers. 2nd ed. Switzerland: ISO; 1988.

18. Dixon DL, Breeding LC. The transverse strengths of three denture base resins reinforced with polyethylene fibers. J Prosthet Dent. 1992;67(3):417-9.

19. Williamson DL, Boyer DB, Aquilino SA, Leary JM. Effect of polyethylene fiber reinforcement on the strength of denture base resins polymerized by microwave energy. J Prosthet Dent. 1994;72(6):635-8.

20. Uzun G, Hersek N, Tincer T. The effect of five woven fiber reinforcements on the impact and transverse strength of a denture base resin. J Prosthet Dent. 1999;81(5):616-20.

21. Anusavice KJ. Phillips' science of dental materials. 11th ed. Philadelphia, PA: Saunders; 2003. p. 703-4.

22. Hargreaves AS. Equilibrium water uptake and denture base resin behavior. J Dent. 1978;6(4):342-52.

23. Zuccari AG, Oshida Y, Miyazaki M, Fukuishi K, Onose H, Moore BK. Reinforcement of acrylic resins for provisional fixed restorations. Part II: changes in mechanical properties as a function of time and physical properties. Biomed Mater Eng. 1997;7 (5):345-55.

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