VARIA

Water sorption and solubility of denture teeth acrylic resin reinforced with milled glass fiber

Özlem Gürbüz, Fatma Ünalan, Pinar Kursoglu

Istanbul, Turkey

Summary

Objectives. The objective of this study is to evaluate the water sorption and solubility of denture teeth acrylic resin reinforced with milled glass fiber

Methods. Test specimens were fabricated from silanized and unsilanized milled glass fiber reinforcement in four different concentrations (5, 10, 15 and 20% by weight) of denture teeth acrylic resin. Control specimens were unfilled acrylic resin. Water sorption and solubility were tested in accordance with International Standards Organization specification No. 1567 with the exception of the dimensions of the test specimens.

Kruskal Wallis test was used in the comparison of groups, post Hoc Dunn's multiple comparison test was utilized in the comparison of subgroups, and Chi square test was performed during the evaluation qualitative data.

Results. Denture teeth acrylic resin was reinforced with unsilanized and silanized milled glass fiber at four percentages (5, 10, 15, 20%) by weight as the test groups. Unreinforced acrylic resin was used as control group. The specimens were abraded on the two-body abrasion-testing device, which conforms to DIN 53516 standard.

Water sorption values were observed to differ significantly for the silanized and unsilanized milled glass fiber mixture varieties in all groups $(5,10,15,20 \ \%)$ (p < 0.05).

Water solubility values disclosed no significant difference among the groups (control, silanized, unsilanized) containing 10% and 20% of milled glass fiber (p > 0.05). In contrast, a significant difference was observed among the groups (control, silanized, unsilanized) containing 5% and 15% milled glass fiber (p < 0.05).

The 5-20% water sorption measurements of the specimens containing unsilanized milled glass fiber revealed a significant difference (p = 0.003).

Other measurements revealed no significant difference (p > 0.05).

Conclusions. Water sorption values increased with increased amount of milled glass fiber, but water solubility values were not affected. Silanization procedure decreased the water sorption and solubility of denture teeth acrylic resin.

Key words: denture teeth, acrylic resin, glass fiber, sorption, solubility.

Introduction

The solubility of materials in the mouth and the sorption of oral fluids by the material are important criteria in their selection [1]. The water sorption should be kept low for composites because excessive water sorption has a detrimental effect on the color stability and the wear resistance. Water sorption is believed to be a contributory factor to the eventual discoloration of the restorations and the hydrolytic degradation of teeth resin-filler interface. Thus it is desirable for both the water sorption and solubility fraction of polymers to be as smaller as possible [2].

Addition of filler to polymethylmethacrylate (PMMA) matrix may enhance the mechanical properties of the polymerized resin but there is a great concern about the stability of PMMA in aqueous environments [3]. This is mainly because the fillermatrix interface provides paths of facilitated diffusion similar to grain boundary diffusion [3,4]. Water sorption in composite materials is a diffusion-controlled process and occurs mainly in the resin matrix [5,6].

Glass fibers were shown to be most suitable for dental applications because of good cosmetic qualities and good bonding of glass fibers to the polymer matrix via silane coupling agents [7,8]. It has been reported that glass reinforcement significantly affects the water sorption and solubility of denture base resin [9,10]. Water can destroy the fiber-polymer matrix bond and plasticization of the polymer matrix by water molecules and these leads to strength reduction of glass fiber reinforced composites [11]. An aqueous environment such as the oral cavity can induce corrosion effects in the surface of glass fibers because of the water that diffuses through the polymer matrix [12]. This can lead to a reduction of the mechanical properties and changes in the composite structure [13].

The study in this paper is part of a continuing study of the reinforcement of a denture teeth acrylic resin with milled glass fiber (mgf). The aim of this study was to determine the water sorption and solubility of denture teeth acrylic resin reinforced with mgf at different weight concentrations.

Materials and Methods

Specimen preparation:

The specimen dimensions were 16 mm in diameter and 7 mm in thickness. Denture teeth acrylic resin (Rutinium Dental Manufacturing spa Ravigo, Italy) polymer/monomer ratio was 20g/10 ml by weight (wt) for all the specimens (unmodified or modified with fiber). The milled E glass fiber (1.2 μ m in diameter, 0.8mm in length and 2.7g/cm³ in density) was supplied from Cam Elyaf A. Þ, Çayýrova, Turkey. The percentages of milled glass fiber used were 5, 10, 15 and 20% by wt.

The silane coupling agent, A-174, 3methacryloxypropyl trimethoxy silane (3-MPS) was supplied from Union Carbide, UK.

The denture teeth acrylic resin was polymerized at 175 C under pressure of 160 Barr for 3 minutes (Elimko 2200 Hidrocontrol Machine, Ankara, Turkey) and then cooled with water under a pressure of 160 Barr for 3 minutes. After demolding, the specimens were removed and finished to remove excess material by honing with fine emery paper. All specimens were prepared with the same procedure in the same mold and carried out at room temperature (23 1C).

The nine groups of specimens (totally 90 specimens) were divided as follows: 10 specimens of unfilled acrylic resin used as a control group (C) and the others test groups – Acrylic resin modified with 5, 10, 15, 20% by wt unsilanized mgf added to polymer (n = 10, totally 40 specimens) and Acrylic resin modified with 5, 10, 15, 20% by wt silanized mgf added to polymer (n = 10, totally 40 specimens).

Water sorption and solubility testing:

Water sorption and solubility were tested according to ISO specification 1567 [14], with the exception of the dimensions of the test specimens. Control and test specimens were measured with fine digital micrometer (Mitutoya Digimatic Caliper 500154/CD15C, England) and their volumes (V) (including fibers) were determined for calculation of water sorption and solubility values. After the specimens were dried for 23 hours at 37°C in desiccators, the specimens were weighed (m₁). All specimens were stored in distilled water in a thermostatically controlled water bath (Elektromag-M96K Water Bath, Turkey) at 37° C for a week, then weighed (m₂). Then the specimens were again dried for 23 hours at 37°C in desiccators, after that being weighed (m₃). All specimens were weighed until the mass was constant to an accuracy of 0.0002 mg with an analytic balance (Metler H20, nearest 0.001 mg Switzerland). After completing the storage periods, the water sorption (W_{sp}) and solubility (W_{SI}) values (mg/mm³) were measured and calculated as described below:

$$W_{sp}$$
: $(m_2 - m_1)/V$
 W_{sl} : $(m_1 - m_3)/V$

Statistical analysis:

Statistical calculations were performed with GraphPad Prisma V.3 program for Windows. Besides standard descriptive statistical calculations (mean and standard deviation), Kruskal Wallis test was used in the comparison of groups, post Hoc Dunn's multiple comparison test was utilized in the comparison of subgroups, and Chi square test was performed during the evaluation qualitative data. Statistical significance level was established at p < 0.05.

Results

Water sorption values were observed to differ significantly for the silanized and unsilanized milled glass fiber mixture varieties in the 5, 10, 15, 20% groups (*Table 1, 2*).

| | | Control | Unsilanized Milled Glass (mgf) | Silanized Milled Glass (mgf) | р |
|---------------------|-----|-------------------|--------------------------------------|------------------------------------|--------|
| | 5% | | 2.372 ± 0.436 | 2.372 ± 0.091 | 0.0008 |
| Water sorption | 10% | | 2.680 ± 0.166 | 2.425 ± 0.081 | 0.0003 |
| ····· | 15% | | 2.978 ± 0.256 | 2.449 ± 0.144 | 0.0002 |
| | 20% | 4.037 ± 0.451 | 3.042 ± 0.470 | 2.296 ± 0.160 | 0.0003 |
| | р | | 0.003 | 0.066 | |
| XX /-4 | 5% | | 4.428 ± 1.362 | 4.236 ± 0.155 | 0.013 |
| Water solubility | 10% | | 4.395 ± 1.515 | 4.140 ± 0.133 | 0.089 |
| · | 15% | | 4.346 ± 0.125 | 4.046 ± 0.206 | 0.003 |
| | 20% | 4.021 ± 0.067 | 4.042 ± 0.487 | 4.109 ± 0.742 | 0.79 |
| | Р | | 0.12 | 0.17 | |

Table 1. Means, standard deviations and statistical comparisons for water sorption and solubility values (mg/mm³) of all groups

The weight changes recorded in the 5-20% of unsilanized milled glass mixtures displayed a statistically significant difference in water sorption. Both 15% and 20% unsilanized mgf containing groups displayed increased water sorption compared to the 5% unsilanized mgf group, p value being respectively: (p < 0.05) and (p < 0.01) (*Table 3*). Other measurements revealed no significant difference (p > 0.05).

In water solubility values, a significant difference was observed between the silanized and the unsilanized groups containing 5% of mgf (p = 0.013).

| | (Water sorption) | | | | (Water solubility) | |
|-------------------------------------|------------------|-----------|-----------|----------|--------------------|----------|
| Dunn's Multiple Comparison Te st | %5 | %10 | %15 | %20 | % 5 | %15 |
| control / unsilanized mgf | P < 0.05 | P > 0.05 | P > 0.05 | P > 0.05 | P > 0.05 | P < 0.05 |
| control / silanized mgf | P < 0.001 | P < 0.001 | P < 0.001 | P <0.001 | P > 0.05 | P > 0.05 |
| unsilanized / silanized mgf | P > 0.05 | P > 0.05 | P > 0.05 | P > 0.05 | P < 0.05 | P < 0.05 |

Table 2. The comparisons of subgroups

mgf: milled glass fibe

In the 15% mgf group, water solubility difference between groups was significant (p = 0.003) (*Table 1, 2*).

No significant difference of water solubility was observed between the 5-20% measurements of the specimens containing silanized and unsilanized mgf (p > 0.05).

Discussion

It has been reported that glass reinforcement significantly affects the water sorption and solubility of denture base resin [9]. Ladizesky et al [10] studied the water sorption of denture base acrylic resin reinforced with woven polyethylen fiber, adding up to 4-6% of the volume. The high fiber content reduced the water sorption by 25%. A study of the water sorption of continuous unidirectional glass fiber-reinforced (11% by wt) specimens demonstrated a decrease in water sorption values [9]. Çal et al [15] reported that the water sorption of denture base polymers is lower when the specimens are reinforced with glass fibers in continuous and woven form. The present study, in accord with the referred papers, revealed that denture teeth acrylic resin with milled glass fiber incorporation held lower water sorption values than did acrylic resin without glass. Water sorption occurs mainly as a direct absorption by the resin. The glass filler will not absorb into bulk of the material but can adsorb water onto its surface. Thus, the amount of water sorption is dependent on the resin content of the composite and the quality of the bond between the resin and the filler [2].

Well impregnated composites have theoretically lower water sorption than poorly impregnated composites in which the fibers are not completely embedded with the resin, which resulted in void in the structure of the polymerized composite that could increase water sorption [9,16-19]. The glass fillers reduced the quantity of water absorbable material and the glass filler didn't absorb water and thus the water sorption of glass fiber composite should be less compared to that of the matrix polymer [9].

| Dunn's Multinle Composison Test | Water sorption Unsilanized | |
|---------------------------------|-------------------------------|--|
| Dunn's Multiple Comparison Test | | |
| %5 / %10 | P > 0.05 | |
| %5/%15 | P < 0.01 | |
| % 5 / % 20 | P < 0.05 | |
| %10 / %15 | P > 0.05 | |
| %10/%20 | P > 0.05 | |
| %15/%20 | P > 0.05 | |

Table 3. Comparisons of concentrations

Chow et al [20] reported in the paper are part of a continuing study of the reinforcement of acrylic denture base resins with highly drawn linear polyethylene fibers. Water sorption is significantly reduced by incorporation of these fibers, even though the water diffusion processes as such remain broadly unaffected. A relative high water sorption value for a composite may indicate a number of possibilities. The resin may contain air voids, introduced during mixing or placement, and another possibility is that hydrolytic breakdown of the bond between the filler and the resin has occurred, allowing adsorption onto the surface of the filler particles [2]. Glass fibers may cause irregular voids in the composite structure and voids absorb water by means of capillary forces and thus may be responsible for variations in water sorption with increasing percentages [2].

Glass fiber reinforcement affected the solubility values of the PMMA specimens [9]. Fiber inclusion and post curing of polymer matrix reduced water sorption and solubility [18]. Results of this study show that the addition of unsilanized or silanized glass

References

1. Craig RG, O'Brien WJ, Powers JM: Properties of Materials In: Dental Materials 6th ed. St. Louis: Mosby, 1996; p. 14.

2. Noort van R: Chemical Properties In: Introduction to Dental Materials. 2ⁿd ed. London: Mosby, 2002; p. 62.

3. Kalachandra S, Turner DT: Water sorption of plasticized denture acrylic lining materials. *Dental Materials*, 1989; **5**: 161-164.

4. Philliphs RW: Denture base resins: Technical considerations, miscellaneous resins, and techniques, In Eugene W. Skinner's Science of Dental Materials, 9 th ed Philadelphia: PA Saunders Co; 1991, pp. 177-213.

5. Braden M, Causton EE and Clarke RL: Diffusion of water in composite filling materials. *Journal of Dental Research*, 1976; **55**: 730-732.

6. Braden M and Clarke RL: Water characteristics of dental microfine composite filling materials.

fiber to the denture teeth acrylic resin either does not affect or decrease water solubility.

The solubility behavior of composite resin materials will be affected by the type of filler used, the treatment of the filler (i.e. silane treatment) [21,22] as well as air voids within the composite resin material, often leading to a decrease of mass of the material [23]. This study revealed a decrease in water sorption values and generally unaffected solubility with silanization.

Conclusions

The strengthening of denture teeth PMMA with milled glass fiber resulted in a composite structure with reduced water sorption and generally unaltered solubility.

Acknowledgements

We thank Prof. Selim Küsefoðlu and Prof. Nihan Nugay from Boðaziçi University, Department of Chemistry, for their support.

Proprietary Materials. *Biomaterials*, 1984; 5:369-372.

7. Vallittu PK: The effect of glass fiber reinforcement on the fracture resistance of a provisional fixed partial denture. *Journal of Prosthetic Dentistry*, 1998; **79**: 125-130.

8. Friskopp J, Blomlöf L: Intermediate fiberglass splints. *Journal of Prosthetic Dentistry*, 1984; **51**: 334-337.

9. Miettinen VM, Valittu PK: Water sorption and solubility of glass fiber- reinforced denture polymethyl methacrylate resin. *Journal of Prosthetic Dentistry*, 1997; **77**: 531-534.

10. Ladizesky NH, Chow TW, Cheng YY: Denture base reinforcement using woven polyethylene fiber. *International Journal of Prosthodontics*, 1994; **7**: 307-314.

11. Lassila LVJ, Nohrström T, Valittu PK: The influence of short-term water storage on the flexural properties of unidirectional glass fiber-reinforced composites. *Biomaterials*, 2002; **23**: 2221-2229.

12. Ehreinstein GW, Schmiemann A, Bledzki A,

Spaude R: *Corrosion phenomena in glass fiber-reinforced thermosetting resins. In Cheremisinoff.* NP editor. Handbook of Ceramics and Composites, New York: Marcel Decker; 1990; pp 231-268. (Reference 13).

13. Vallittu PK: Effect of 180-week water storage on the flexural properties of E glass and silica fiber acrylic resin composite. *International Journal of Prosthodontics*, 2000; **13**: 334-339.

14. International Organization for Standardization. *Specification 1567: Dentistry-Denture Base Polymers.* 2nd ed, Geneva: Switzerland; International Standards Organization, 1988.

15. Çal NE, Hersek N, ^aahin E: Water sorption and dimensional changes of denture base polymer reinforced with glass fibers continuous unidirectional and woven form. *International Journal of Prosthodontics*, 2000; **13**: 487-493.

16. Vallittu PK: The effect of void space and polymerization time on transverse strength of acrylic-glass fiber composite. *Journal of Oral Rehabilitation*, 1995; **22**: 257-561.

17. Valittu PK: Impregnation of glass fibers with poltmethylmethacrylate using a powder-coating

method. Applied Composite Materials, 1995; 2: 518.

18. Miettinen VM, Narva KK, Valittu PK: Water sorption, solubility and effect of post curing of glass fiber reinforced polymers. *Biomaterials*, 1999; **20**: 1187-1199.

19. Valittu PK: Acrylic resin-fiber composite-Part II: The effect of polymerization shrinkage of polymethylmetacrylate applied to fiber roving on the transverse strength. *Journal of Prosthetic Dentistry*, 1994; **71**: 613-617.

20. Chow TW, Cheng YY, Ladizesky NH: Polyethylene fiber reinforced poly(methylmethacrylate) water sorption and dimensional changes during immersion. *Journal of Dentistry*, 1993; **21**: 367-372.

21. Söderholm KJ: Degradation of glass filler in experimental composites. *Journal of Dental Research*, 1981; **60**: 1867-1875.

22. Söderholm KJ, Zigan M, Ragan M, Fischlschweiger W, Bergman M: Hydrolytic degradation of dental composites. *Journal of Dental Research*, 1984; **63**: 1248-1254.

23. Oysaed H and Ruyter IF: Water sorption and fiber characteristics of composites for use in posterior teeth. *Journal of Dental Research*, 1986; **65**: 1315-1318.

Correspondence to: Dr. Fatma Unalan, Associate Professor, DDS, PhD - Istanbul University, Faculty of Dentistry, Department of Prosthodontics. 34390 Çapa-Istanbul, Turkey. E-mail: fat-maunalan@yahoo.com, pinarkurs@hotmail.com, pinarkurs@hotmail.com