Influence of different glass fiber reinforcements on denture base polymer strength (Fiber reinforcements of dental polymer)

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ABSTRACT

Aim Assessment of flexural strength values of dental base polymers reinforced with different glass fibers (“dental” and “industrial” origin) after performed artificial ageing procedures.

Methods Three hundred specimens (dimensions 18 x 10 x 3 mm) were produced of denture base polymers reinforced with different glass fibers. The “short beam” testing method was used to determine the flexural strength of the specimens after polymerization, immersion in water of temperature 37°C for 28 days, and thermocycling.

Results Flexural strength of the polymer specimens was 91.76-122.75 MPa, while specimens reinforced with glass fibers demonstrated rise of flexural strength values (103.10-163.88 MPa), no matter which type (“dental” or “industrial”) of fibers was used. Microscopic examination revealed partial bonding between the glass fibers and polymer material.

Conclusion Both of the investigated glass fibers had similar strengthening effect, but due to better investment/benefit ratio “industrial” glass fibers could be recommended for dental laboratory use. Prolonged polymerization of denture base materials should be proposed because it had direct impact on the improvement of flexural strength values.

Key words: dental materials, polymers, glass fibers, dental prosthesis, mechanical phenomena
INTRODUCTION

Life span of the world population has been considerably prolonged in the last century because of the huge progress of preventive and curative medicine. Therefore, the number of elderly people (60 and more years old) is increasing. There is also evidence of progressive loss of teeth connected with the age of patients, and the increased loss of teeth is dominant in the population between 50 and 60 years of age (1). Total loss of teeth is especially frequent in patients living in non-developed countries due to improper dental care and low health education of the population. Dental prostheses are at the second place in frequency of different appliances that are used by elderly population, just next to the glasses (1). Today dental prostheses are predominantly made of methyl metacrylate polymer which has proven to be the most reliable denture base material. Despite many advantages, methyl metacrylate is prone to fracturing. Therefore, fractures of the dentures are very frequent, almost equal to the number of newly made dentures (2). These dentures produced from methyl metacrylate may be strengthened by incorporation of various reinforcements into the denture base polymer, because such reinforcements enhance flexural strength and resistance to impact (3). Physical and mechanical properties of acrylic dentures can be enhanced by integration of different fibers with different fiber architectures into the denture base polymer (4-9). In order to improve flexural and impact strength of denture base polymer, graphite (7-11), glass (5,6,12-16), and organic fibers, such as, aramide (15,17,18) and polyethylene fibers with high molecular weight (9,19-27), are used.

Today the most acceptable fibers for dental polymer reinforcements are glass fibers, because of their good aesthetics (6,12-14,28,29) and good bonding with polymers via silane coupling agents (30-32). Also, they can easily be adapted to the desired shape and length (33) which is then suitable for incorporation into denture base polymer material. Different procedures of pre-impregnation of glass fiber reinforcements with silane coupling agents are used to establish chemical bond with denture base polymer material. Glass fiber reinforcements produced by dental manufacturers, which are present on market, are already pre-impregnated with silane coupling agents, but they are rather expensive. Common industrial E-glass fibers are usually not pre-impregnated during manufacturing. They are rather cheap, and it is possible to pre-impregnate them with silane coupling agents in dental laboratory.

The main goal of this study was the assessment of flexural strength of dental base polymers reinforced with silane pre-impregnated glass fibers of different architecture produced by dental products manufacturers. These results should be compared with the results of flexural strength of dental base polymers reinforced with “industrial” E-glass fibers after they are pre-impregnated in dental laboratory using silane coupling agent. Obtained results could give the preference of which glass fiber reinforcement to use, with regard to the investment/benefit ratio.

MATERIALS AND METHODS

Quadratic specimens with smooth surfaces and dimensions of 18 x 10 x 3 mm were made of Meliodent Heat Cure and Meliodent Rapid (Heraeus Kulzer, Hanau, Germany) polymer. Some of the specimens made from aforementioned polymers were reinforced with glass unidirectional fibers and nets produced exclusively for application in dentistry (StickTech Ltd., Turku, Finland), and some with industrial E-glass unidirectional fibers and nets (Kelteks, Duga Resa, Croatia). All the specimens were split across ten different groups with thirty specimens each (n=300). Special metal cuvette was constructed in order to produce uniform specimens. It consisted of two thick polished metal parts on the sides and two thin metal parts in the middle. The middle metal parts had ten quadratic perforations of the size of a specimen (18 x 10 mm). One thin metal part was 1 mm thick and the other thin metal part was 2 mm thick. In that way the thickness of the specimens and the proper glass fiber alignment were obtained. So, the fiber reinforcements were placed 1 mm from one side and 2 mm from the other side of a specimen. Metal parts of the cuvette were smeared twice with Ivo-
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clar Separating Fluid (Ivoclar Vivadent, Schaan, Liechtenstein). Pre-impregnated glass fibers for dental use (StickTech Ltd., Turku, Finland), unidirectional (Stick™) or net shaped (Stick™ Net) were impregnated with Meliodent Heat Cure polymer mixture of a syrup consistency (with polymer/monomer weight ratio 10:8) for eight minutes. During impregnation time of the glass fibers Meliodent Heat Cure polymer was prepared according to the manufacturer’s instructions and placed in both cuvette halves (consisting of one thick side part + one thin middle part). Just impregnated glass fibers, unidirectional (Stick™) or net shaped (Stick™ Net) were taken from polymer syrup and placed on one cuvette half. During that alignment procedure unidirectional glass fibers were laid along the specimens, in order to be orthogonal to the force to be applied. The net shaped glass fibers were placed at an angle of 45° to the length of the specimen. The cuvette halves were then closed together and put in a hydraulic press (Zlatarne, Celje, Slovenia) under the 200 bars pressure. After pressing, the cuvette was moved to a manual bench vice and put in a polymerizing apparatus (Type 5518, KaVo EWL, Biberach, Germany) where polymerization was performed according to the manufacturer’s instructions. For the specimens produced from Meliodent Rapid auto polymerizing material a similar procedure was performed. The only difference was shorter impregnation time of glass fibers (two minutes instead of eight) due to the auto polymerizing character of the material.

Production of the specimens reinforced with industrial E-glass fibers was different only in the performance of pre-impregnation of the glass fibers in dental laboratory.

Firstly, industrial non-impregnated fibers were weighted in weighing-machine Mettler H 311 (Mettler Instr. AG, Zurich, Switzerland) with the accuracy of 0.1 mg, to match the weight of dental glass fibers used in equivalent specimens. Then, industrial E-glass fibers were cleaned with 1.6 mol sulphuric acid for 30 sec., rinsed in distilled water, and air dried at room temperature for 24 hours. After that they were dipped into 98% γ-metacryloxypropyl-trimethoxysilane (Sigma-Aldrich Co., St. Louis, MO, USA), placed on a clean sheet of paper, put in a dental sterilizer (ISO 400, Aesculap, Tuttingen, Germany) and heated at temperature 100°C for 2 hours. Afterwards, these so pre-impregnated glass fibers were impregnated with adequate polymer syrup (Meliodent Heat Cure or Meliodent Rapid) and the specimens were produced in the manner of the aforementioned procedures.

After polymerization of the denture base materials and cooling, the cuvettes were opened and the specimens were detached. Thin polymer excess on all the specimens was removed with a carbide bur (Ivomill, Ivoclar Vivadent), and the specimen margins were finished using sandpaper (Sianor 7/0B, Frauenfeld, Switzerland). The specimens were checked with calipers (Dentarium 042-751, Dentarium, Ispringen, Germany) for the accuracy of dimensions, and the maximum allowed deviation was 0.05 mm.

All the specimens were tested in the universal testing machine (Amsler, Schafhausen, Switzerland) using the short beam method (Figure 1) (34), and the moving speed of the blade was set to 1.5 mm/min in order to determine the samples’ flexural strength. All ten groups of specimens were subdivided into three subgroups of ten specimens each which were tested after polymerization of the specimens, immersion of the specimens in distilled water with temperature at 37°C (thermostat Btuj, Poznan, Poland) for 28 days, and after thermocycling of the specimens according to Hansson’s method (35). Specimens were placed in a testing holder, in a position where the fiber reinforcements were closer to the testing holder’s posts (1 mm away from the posts and 2 mm from the blade). The force causing breakdown of the

![Figure 1. Scheme of the specimen loading](d, thickness of the specimen, F, applied force)
specimens was recorded and the flexural strength was calculated according to the formula:

$$\sigma_{\text{max}} = \frac{F_{\text{max}} \cdot l}{4} \cdot \frac{6}{b \cdot h^2} \left(\frac{N}{\text{mm}^2} = \text{MPa}\right)$$

\(F_{\text{max}}\) – measured force of the loader (N); \(l\) – distance between posts (here 15 mm); \(b\) – width of the specimen (here 10 mm); \(h\) – height of the specimen (here 3 mm).

Obtained numerical results were analyzed with SPSS statistical package (SPSS Inc., Chicago, USA). Statistical analysis was performed using descriptive statistics, tests of between-subjects effects, Scheffe’s test, and Student’s t-test.

In order to reveal the quality of bonding between glass fibers and denture base polymer, glass fiber reinforced specimens were randomly chosen and sealed in Durofix material (Struers, Rodovre, Denmark). Subsequently they were ground, and polished according to the routine procedure (36) to obtain a smooth surface suitable for microscopic examination. It was performed with the use of a light microscope Olympus BH2-UMA (Olympus Optical, Tokyo, Japan) and the characteristic images were photographed with Olympus C-5050 Ultra Zoom camera (Olympus Optical, Tokyo, Japan).

**RESULTS**

During testing specimens made of Meliodent Heat Cure and auto polymerizing Meliodent Rapid polymer (control groups) demonstrated the lowest flexural strength, whereas, the specimens reinforced with both types of fibers showed higher flexural strength values (Figure 2, Table 1). Scheffe’s test applied across ten investigated groups of specimens revealed a statistically significant difference (\(P<0.05\)) between some of these groups, as shown in Table 1. The test between subject’s effects revealed that the type of polymer (heat cure or auto polymerizing), type of glass fibers (unidirectional or net) and ageing procedures (immersion in distilled water at temperature 37 °C for 28 days, thermocycling) had a significant influence (\(P<0.05\)) on the achieved flexural strength values, whereas, the origin of glass fibers (“dental”-Stick

Table 1. Descriptive statistics and Scheffe test between the groups of specimens

<table>
<thead>
<tr>
<th>Groups*</th>
<th>N</th>
<th>Mean (MPa)</th>
<th>Standard Deviation</th>
<th>Standard Error</th>
<th>95% Confidence Interval for Mean</th>
<th>Min.</th>
<th>Max.</th>
<th>Scheffe test between groups - Statistically significant difference</th>
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<tr>
<td>1</td>
<td>30</td>
<td>140,900</td>
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<td>5,9762</td>
<td>128,6771 153,1229</td>
<td>86,251</td>
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<td>130,633</td>
<td>12,1853</td>
<td>32,7338</td>
<td>126,0833 135,1834</td>
<td>110,00</td>
<td>148,75</td>
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<tr>
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<td>30,00</td>
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<td>122,50</td>
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<tr>
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<td>4,05921</td>
<td>149,2813 165,8854</td>
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<td>30,00</td>
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<tr>
<td>6</td>
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<td>313,31182</td>
<td>1,80894</td>
<td>131,6551 138,7749</td>
<td>28,75</td>
<td>236,25</td>
<td>1  2  3  4  5  6  7  8  9  10</td>
</tr>
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</table>

*1. Meliodent Rapid polymer+Kelteks fibers (net); 2. Meliodent Rapid polymer+Kelteks fibers (unidirectional); 3. Meliodent Rapid polymer+Stick fibers (net); 4. Meliodent Rapid polymer+Stick fibers (unidirectional); 5. Meliodent Rapid polymer (control); 6. Meliodent Heat Cure polymer (control); 7. Meliodent Heat Cure polymer+Kelteks fibers (net); 8. Meliodent Heat Cure polymer+Kelteks fibers (unidirectional); 9. Meliodent Heat Cure polymer+Stick fibers (net); 10. Meliodent Heat Cure polymer+Stick fibers (unidirectional); N, number of specimens; Mean, arithmetic mean of the recorded flexural strength values in megapascals.
or “industrial”-Kelteks) had no statistically significant influence. That was also confirmed by Student’s t-test ($t=1.367$). Auto polymerizing polymer material was weaker than heat-cure polymerizing dental base material ($P<0.05$), and unidirectional glass fibers positioned perpendicularly across the applied force, on the specimen, strengthen polymer material more successfully than net shaped glass fibers positioned at an angle of $45^\circ$ ($P<0.05$). Thermocycled specimens had the highest flexural strength ($P<0.05$), whereas, there was no statistically significant difference ($P>0.05$) between specimens tested after polymerization and after immersion in distilled water for 28 days.

After a short beam test was performed, the specimens reinforced with both types of fibers revealed mostly adhesive type of breakdown between fiber reinforcements and polymer matrix resulting in pullouts of fibers from the matrix. Microscope image analysis showed the existence of voids between glass fibers and polymer matrix and partial bonding between glass fibers and polymer material (Figure 3).

**DISCUSSION**

The prime and most frequent fracture of the upper denture occurs in the medial line and the fracture mechanism and the influence of the masticatory load which is applied onto the dentures are very similar to the three point bending test – short beam test (37,38). During chewing denture base material is submitted to the flexural deformation due to applied masticatory load. Therefore, tests of the flexural strength are very appropriate for testing polymer dental materials (23, 37-41), because simulation of the masticatory load is very close to the real conditions in the mouth. Also, the influence of material fatigue on mechanical properties is decisive (42). For that reason different procedures of artificial aging, such as, underwater storage in thermally controlled conditions and cyclic temperature changes (thermocycling) are used in denture materials studies. They are used to determine the longevity of the obtained mechanical properties of dental materials in the demanding environment of the oral cavity. For that purpose different authors use different time periods of underwater storage, but the important influence of water on the flexural strength occurs during the first four weeks of immersion causing decrease of the flexural strength values. Since longer period of immersion does not perform a statistically significant decrease of flexural strength (43,44) 28 days immersion in water at 37 °C was used in this study.

Immersion in water enables molecules of water to penetrate into the areas between the polymer chains, remain there, and act like wedges between these chains. Water entry in polymer material during immersion is primarily caused by diffusion, and partly by the polarity of the polymer chains that is caused by unsaturated molecules and unbalanced intermolecular forces. Penetration of water molecules may also cause softening of the denture base, as absorbed water can act as a poly(methyl methacrylate) (PMMA) plasticizer. Water absorption diminishes the mechanical properties of the material, resulting in lower flexural strength and lower modulus of elasticity (45). Also, the ageing effect of thermocycling procedure which simulate ingestion of cold and hot food/beverages can have a significant influence on mechanical properties of polymer materials (46,47).

After the ageing procedures were performed no statistically significant difference ($P>0.05$) was found between specimens tested just after polymerization and after immersion in distilled water for 28 days. On the contrary, the flexural
strength values remained the same or were even slightly increased in some of the specimens subgroups (Figure 2). Since water absorption that occurred during the immersion of specimens in distilled water had no negative influence on the flexural strength, it could be stated that water absorption caused relaxation of the stress in polymer material that occurred during polymerization shrinkage. That could be the explanation for the increase of flexural strength values for the tested polymer materials (15,48).

As mentioned, thermocycling procedure can have a significant impact on lowering the mechanical properties of polymer materials (46,47). On the contrary, in this investigation thermocycling procedure increased the flexural strength values. It appears as if the increase in temperature during thermocycling procedure resulted in the effect of prolonged polymerization, which could in turn result in the decrease of the residual monomer volume thus enhancing the mechanical properties of the polymer (increase of the flexural strength). Therefore, prolonged polymerization of the polymer material should be proposed discarding the manufacturer’s instructions on the relatively short duration of polymerization, because, in the authors’ opinion, it had a direct impact on the improvement of mechanical properties.

Glass fibers, no matter if they were of dental or industrial origin, considerably strengthened the polymer material, because testing load was shared between polymer material and the fiber reinforcements. Because of similar strengthening effect, but due to relatively high costs of the “dental” glass fiber reinforcements, low cost “industrial” glass fibers treated with self made pre-impregnation could be recommended for dental laboratories.

Auto polymerizing polymer material was significantly weaker than heat-cure polymerizing dental base material (P<0.05) regardless of the details of fiber reinforcements. That fact could be attributed to the type of polymerization procedure resulting in higher residual monomer volume at auto polymerizing material that is directly influencing its mechanical properties.

As expected, unidirectional glass fibers strengthen polymer material more successfully than net shaped glass fibers, because fibers were positioned perpendicularly across the applied force (opposite to the net shaped glass fibers positioned at an angle of 45°), thus giving better strengthening effect. Therefore they are more appropriate to be used, but in cases when the direction of the braking force is known.

After short beam test was performed specimens reinforced with both, “dental” and “industrial”, fibers revealed mostly adhesive type of breakdown between fiber reinforcements and polymer material resulting in pullouts of fibers from the polymer. That arouses suspicion about inadequate bonding between glass fibers and polymer material, but there was no difference between specimens reinforced with expensive fibers produced by dental manufacturers (already pre-impregnated with silane coupling agents) and specimens reinforced with cheap “industrial” E-glass fibers treated with described self made pre-impregnation.

Microscope image analysis of specimens reinforced with both types of glass fibers (“dental” and “industrial”) showed existence of voids between glass fibers and polymer revealing partial bonding between glass fibers and polymer material (Figure 3) which is, in our opinion, the result of established dental laboratory routine production, without idealization of specimens’ production conditions. It is the intent of the study to generate reproducible results comparable to the general laboratory practice. But even such “imperfect” reinforcements are good enough to significantly increase the flexural strength of the investigated polymer materials.

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