Use of the fiberglass reinforcement method in thermoplastic mouthguard materials to improve flexural properties for enhancement of functionality

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The purpose of this study was to evaluate the application of fiberglass reinforcement method in thermoplastic mouthguard materials to improve flexural properties and adhesive strength. Commonly used two types of commercial mouth guard materials (ethylene-vinyl acetate copolymer-based and polyolefin-based) were reinforced with glass fiber clothes by two-step hot press. Flexural strength and adhesive strength with each base material were examined via three-point bending test and delamination test, respectively. Ethylene-vinyl acetate copolymer-based fiberglass-reinforced material has significantly greater adhesive strength with base material and improvement of flexural properties compared with polyolefin-based material. These results suggest that flexural properties of both conventional commercial mouthguard materials were improved when the glass-fiber-reinforced method was applied to reinforce mouthguard materials, and more, ethylene-vinyl acetate copolymer was more desirable for the base material.

Keywords: Mouthguard, Fiberglass reinforcement method, Flexural properties, Enhancement of functionality

INTRODUCTION

The mouthguard helps prevent oral injuries and preserve the oral structures of athletes during contact and non-contact sports1-4. Among the three basic types: stock (ready-made), mouth-formed or boil-and-bite, and custom-made, the World Dental Federation (FDI: Fédération Dentaire Internationale) has recommended the use of custom-made mouthguards for players5. Custom-made mouthguards, besides preventing orofacial injuries, provide athletes with better comfort and wearability6 and can be processed for end-use applications7. Ethylene-vinyl acetate copolymer (EVA) is the most commonly used material for commercial and custom mouthguards due to its widespread availability and adequate physical and mechanical properties7. In Japan, polyolefin (PO) is another popular mouthguard material with satisfactory physical properties, and has both higher adhesive strength and lower water absorption than EVA8.

The thickness of a mouthguard has a vital role in absorbing the impact energy and dispersing it throughout the mouthguard material. According to Westerman et al., the force transmitted through the mouthguard material is inversely related to the thickness of the material up to 4 mm9. Therefore, the thicker the mouthguard, the better its ability to absorb impact energy. However, wearing an excessively thick mouthguard could be uncomfortable for the athletes during sports activity. The double-layered mouthguard has shown less thinning (34%) than the single-layered (38%) mouthguard during the fabrication process10. In addition, the double-layered mouthguard has more longitudinal dimensional stability than the single-layered mouthguard because of smaller residual stress accumulation during the forming process11. Enhancing the shock absorption ability of mouthguard material while minimizing its thickness has been a recent focus of research. To that end, the inclusion of air cells12, an intermediate layer of sorbothane between two layers of EVA13, a sponge insert14, the insertion of an acrylic layer with or without buffering space15,16, and the labial insertion of polyethylene terephthalate glycol-modified with buffering space17 showed higher shock absorption ability than the conventional laminated custom-made mouthguard. Reinforcing thermoplastic material with fiberglass is a frequently used and unique technique to increase the shock absorption ability of protective sports equipment such as face guards. Reinforcing face guard thermoplastic material with fiberglass already has proved very effective at improving shock absorption ability and decreasing the total thickness and weight of the material18,19. A first, this study evaluated the effect of fiberglass reinforcement of conventional mouthguard
materials (EVA and PO) on their flexural properties to develop a high functionality for mouthguards.

Adhesive strength, flexural properties, and the ability of the material to follow the curvature of the dental arch shape are factors of concern when athletes use such mouthguards over long periods of time. Takeda et al. described how EVA in a bilaminated mouthguard had enough adhesive strength for long term usage\(^{20}\). Suzuki et al. studied how PO mouthguard material exhibits satisfactory bonding, allowing good adhesion without any bonding agent\(^{8}\). In another study, Ihara et al. performed a delamination test on PO and EVA materials, which showed that both materials had good adhesive strength\(^{21}\). Shock absorption ability and flexural properties of fiberglass-reinforced thermoplastic material for face guards have already been evaluated\(^{18,19,22}\). However, adding different materials like fiberglass cloth to the thermoplastic material used for fabrication of mouthguards, then testing its flexural properties and adhesive strength, has not yet been investigated. Thus, we investigated the adhesive strength of fiberglass-reinforced materials laminated to each base of conventional mouthguard material (EVA and PO) and evaluated their functional efficacy, since it was assumed that mouthguards are created by laminating fiber-reinforced material with a base material.

The null hypotheses of the present study were that glass fiber reinforcement method were not affected for adhesive strength of fiberglass-reinforced materials laminated to each base of conventional mouthguard material (EVA and PO) and flexural strength of fiberglass-reinforced materials.

**MATERIALS AND METHODS**

**Materials**

Two types of 3 mm-thick thermoplastic commercial mouthguard materials were chosen for the experiment: an EVA (Erkoflex\(^{®}\); Ev; Erkodent Erich Kopp, Pfalzgrafenweiler, Germany) and a PO (MG21TM; Po; CGK, Hiroshima, Japan) (Table 1). Ten sheets of plain-woven E-fiberglass cloth (M100X 104H; density: 100 g/m\(^2\), Unitika, Osaka, Japan) were used to reinforce each mouthguard thermoplastic material using the hot-press method (Fig. 1).

**Preparation of fiber-reinforced thermoplastics**

The two-step hot-press technique was used to impregnate fiberglass into the thermoplastic materials. Five sheets of the fiberglass were placed on the top and five sheets of the fiberglass were placed on the bottom of each sheet of conventional mouthguard material (Ev and Po). Ev and the sheets of fiberglass were pressed between two hot metal plates using a hot-press machine at 140\(^{\circ}\)C heater control setting (modified AH-1T, AS ONE, Osaka, Japan) (Fig. 2) with a compression load of 0.6 MPa while using the hot-press method (Fig. 2) with a compression load of 0.6 MPa while using the hot-press method (Fig. 2).

**Table 1** Physical and mechanical properties\(^{8}\)

<table>
<thead>
<tr>
<th>Material</th>
<th>Shore A hardness</th>
<th>Tensile strength (KPa)</th>
<th>Elongation (%)</th>
<th>Water absorption (%)</th>
<th>Tear strength (N/mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>EVA</td>
<td>81</td>
<td>1.8</td>
<td>800</td>
<td>0.012</td>
<td>48</td>
</tr>
<tr>
<td>PO</td>
<td>76</td>
<td>1.2</td>
<td>850</td>
<td>0.222</td>
<td>44</td>
</tr>
</tbody>
</table>

Ethylene-vinyl acetate copolymer (EVA) and polyolefin (PO)
a vacuum pump (MINIVAC PD-52; Yamato Scientific, Tokyo, Japan) to maintain a decompression environment for 20 min. To avoid poor impregnation of fiberglass into the mouthguard material, the process was repeated for the final compression, reducing the material thickness to 1.0 mm. The same procedure was followed to prepare Po with fiberglass (Po-Gf).

Three-point bending test
For the three-point bending test, hot-pressed 1.0 mm-thick sheets of Ev with fiberglass (Ev-Gf) and Po-Gf were cut by an ultrasonic cutter (Labo Sonic Cutter NE87; Nakanishi, Tochigi, Japan) into rectangular shapes 102 mm in length and 17 mm in width. Five specimens of each type were prepared and then ground smooth with waterproof SiC abrasive paper (#400 and #1000) to the proper dimension for the experiment (100×15 mm). A digital micrometer (Digimatic 293421-20, Minimum reading: 0.001 mm, Mitutoyo, Kanagawa, Japan) was used to measure the specimen’s dimensions. The three-point bending test was configured according to Japanese Industrial Standards (JIS) K7171-2008 and K7074-198823,24. It was therefore run with a 40 mm width support span and a crosshead speed of 1.0 mm/min using a universal test machine (EZ-LX, Shimadzu, Tokyo, Japan) (Fig. 3). The flexural strength and modulus data from the test were stored on a personal computer, and calculated using analysis software (TRAPEZIUM X ver. 1.4.0; Shimadzu). Five specimens of Ev-Gf and five specimens of Po-Gf were examined. All the tests were carried out in a dry environment in room temperature at 20–25°C under atmospheric pressure.

Delamination test
A standard T-peel test was used as the delamination test20,21. For each test, two sheets of same base materials, one unaltered and one reinforced with fiberglass cloth (e.g., Ev-Gf and Ev, or Po-Gf and Po), were laminated together. First, an adhesive film sheet for separation (Molteno Separate Film, Molten, Hiroshima, Japan) was placed across the reinforced sheet material (Ev-Gf or Po-Gf) with its centerline at the center (50 mm position) of the reinforced sheet, to create an adhesive area at the center of the sheet 15 mm wide. Next, the base mouthguard material sheet (Ev or Po) was heated and placed on the reinforced sheet material so their edges aligned, then the materials were pressed in a vacuum at 0.012 MPa using a vacuum thermoforming machine (Vacuum Forming Machine, Keystone Industries, Cherry Hill, NJ, USA). The resulting laminated sheet was cut with a dumbbell-shaped cutter according to JIS K6251:2004 so that the adhesive area would be at the center of the isthmus, then was sectioned at the center to create two bottle-shaped specimens (Fig. 4). The final specimens with an adhesive area of 4.0×7.5 mm were then prepared for the delamination test (Fig. 4). Five specimens of Ev-Gf with Ev and five specimens of Po-Gf with Po were tested using the T-peel test (Fig. 4). Five specimens of Ev-Gf with Ev and five specimens of Po-Gf with Po were tested using the T-peel test to observe the adhesive strength. The size of each specimen was measured and confirmed by digital micrometer (Digimatic 293421-20) before the delamination test.

For each of the ten specimens, we proceeded with the T-peel delamination test by fixing the specimen to a universal test machine (1123, Instron, Canton, MI, USA) with a special jig to grip it firmly. During the delamination test, the crosshead speed was kept at 50 mm/min. Thus, the load on the adhesive bond rapidly increased when delamination started. The adhesive strength at the start of delamination was measured as the maximum load divided by the width of each specimen (approximately 4.0 mm). The displacement at the start of delamination, changes in the load and displacement before specimen fracture or break, and the displacement at fracture were recorded using material testing software (Series IX, Instron).

Statistical analysis
Flexural test results for Ev-Gf and Po-Gf samples, and
Values are shown as mean±standard deviation.
*There were significant difference between Ev-Gf vs. Po-Gf.

<table>
<thead>
<tr>
<th>Thickness (mm)</th>
<th>Flexural strength (Mpa)</th>
<th>Flexural modulus (Gpa)</th>
<th>Adhesive strength (N/mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ev-Gf</td>
<td>1.237±0.033</td>
<td>29.7±1.6</td>
<td>4.85±0.47</td>
</tr>
<tr>
<td>Po-Gf</td>
<td>1.226±0.123</td>
<td>14.8±0.6</td>
<td>3.17±0.49</td>
</tr>
</tbody>
</table>

DISCUSSION

There is a wide range of materials suitable for making mouthguards. EVA material is commonly used for making commercial, ready-made, or custom-made mouthguards because of its widespread availability as well as good mechanical and physical properties. PO is an emerging mouthguard material that has gained wide usage, exhibiting better shock absorption capability with lower water absorption and higher adhesive strength, comparable to EVA. For these reasons, EVA and PO materials were selected for this research. Functional and mechanical properties are important factors in selecting such materials for mouthguards, particularly when mouthguards are used for longer periods of time inside the oral cavity. Impregnating the base material like Ev or Po with another reinforcing material could be a useful way to change the effect of such properties.

As proved in the study of Kismet, changing mechanical properties by using reinforced hydrolyzed polyester-epoxy system electrostatic powder in a polyolefin base comparatively improved the physical bonding mechanism. That study clearly showed that the physical bonding mechanism and bending strength were enhanced; however, the study used gamma-radiated specimens. Other studies stated that reinforcing thermoplastic denture base resin by impregnating it with fiberglass is a useful method to improve overall mechanical properties, yielding higher elastic modulus and improved flexural properties with satisfactory reinforcing effects. Studies by both Abe et al. and Wada et al. proved that in the case of sports protective equipment made using thermoplastic core materials for faceguards, reinforcing with fiberglass results in improved flexural properties and better shock absorption capabilities. Using such a procedure also might lead to a decrease in the total thickness and weight of the faceguard to lower values than for one that is commercially or conventionally custom-made. Thus, this research work was structured to investigate reinforcing mouthguard material with fiberglass to observe the mechanical and functional behavior of its adhesive strength and flexural properties.

For a multilayered laminated impregnate fabrication (base material and sheets of reinforcement material) using a one-step hot-press technique under certain conditions, interfacial adhesive strength usually significantly decreases as the thickness or number of the sheets increases because poorly impregnated areas remain due to the material's significant impregnation resistance gradient in the direction of its thickness. Because impregnation resistance gradient (base material and reinforcement material) is also influenced by the temperature gradient and thermal conductivity, even slightly inappropriate the temperature, pressure, and holding time would lead to displace and spread fibers from their initial position in the base material. Therefore, it is not appropriate to fabricate multilayered laminates using a one-step hot-press technique because of difficulties both in impregnation, and in establishing correct fiber position of each layer homogenously with fine orientation. In contrast, the two-step hot-press technique for multilayered thermoplastic proved useful.
for the matrix base to hold the fibers in a desired position homogeneously without exposing the fibers on the surface of finished specimens\textsuperscript{18,29}. In the process of adding reinforcing fiberglass cloth to mouthguard materials in our study, impregnation was followed by a two-step hot-press technique to avoid poor impregnation of fiberglass cloth. Temperature was continuously maintained by a hot-press heater regulator, thermal conductivity was maintained by an induction motor, and the vacuum machine was used to prevent insertion of any bubbles during impregnation with fiberglass. There are many types of fiberglass cloth that can be used for reinforcement in thermoplastic materials. For example, continuous unidirectional S-fiberglass, continuous unidirectional E-fiberglass, and short-rod fiberglass have shown good results in several studies\textsuperscript{18,27,30). In those studies, it was found that continuous unidirectional E-fiberglass showed the most effective and satisfactory adhesive strength with higher flexural strength and flexural modulus values. This is possibly attributable to the fact that the unidirectional E-fiberglass has better density, composition, thickness, and mesh size compared to other types of fiberglass. The selected glass cloth was epoxysilane-treated, plain weave, thickness 0.12 mm, density 100 g/m², and weaving density (threads/25 mm): 19 (vertical)-19 (horizontal) according to the relevant Japanese Industrial Standards (JIS R 3413)\textsuperscript{31), tensile strength is estimated to be 392 (N/25 mm) or more in both the vertical and horizontal directions. Plain weave is the simplest type of weave, made by alternately crossing one warp thread and one weft thread. The warp and weft yarns appear evenly on both sides of the fabric, and since there are many points where the yarns intersect. It is considered strong and durable against friction. During final preparation of the specimens for the delamination test and three-point bending test, specimens were re-shaped to the required dimensions using an ultrasonic cutter, and then the edges were refined with waterproof abrasive paper.

The fiberglass reinforcement method was effective for improving the properties of conventional mouthguard materials. The bending strength of Ev-Gf was ten times higher and Po-Gf was seven times higher than that of each base material, which was reported by Abe et al.\textsuperscript{22). In the same study, the authors also reported the flexural strength of conventional thermoplastic materials used for face guards (3 types: 21.6 MPa, 31.5 MPa, 44.8 MPa) and that of denture base resin (101.4 MPa), which shows that the flexural strength of Ev-Gf was closer to that of face guard material. Based on the flexural strength of poly ethylene terephthalate-glycol used in the previous study\textsuperscript{15,16}, it is required 30 MPa or higher as flexural strength\textsuperscript{22).}

The functional and mechanical properties of mouthguards should be evaluated by testing adhesive strength, using the tensile test and tear test, which evaluate the durability of a laminated mouthguard material. Several test methods such as T-peel, 180° peel, 90° peel, or floating roller have been useful for examining such behavior. Delamination testing using a T-peel test is one of the most effective methods to investigate adhesive strength\textsuperscript{5,20}, and has been adopted by most standards bodies (e.g. ISO 11339, ASTM D 1876)\textsuperscript{31). Therefore, the T-peel test was conducted in this study to evaluate adhesive strength. During the procedure, delamination started when the load was rapidly increased. It was important to observe the initial load at the start of delamination, which was more important than the final load, which resulted in interfacial fracture both for Ev-Gf and for Po-Gf in our study. According to Ihara et al., the adhesive strength of EVA at the start of the delamination was significantly greater than that of PO in a dry environment\textsuperscript{21). In that study, the fracture pattern observed during the delamination test showed that EVA had interfacial fractures and PO had cohesive fractures, suggesting that EVA adhesive strength can be improved by changing the laminating condition or pattern. Our study results support the results reported by Ihara et al.\textsuperscript{21). Therefore, specimens in our research contained reinforcing fiberglass in Ev and Po, where testing results showed Ev-Gf (1.90 N/mm) had significant higher adhesive strength than did Po-Gf (0.33 N/mm). The same types of material as EVA and PO used in this study are both conventionally used as materials for so-called hot-melt adhesives, which exhibit adhesive functions when heated to melt and then cooled to solidify. EVA is known to have higher viscosity than PO\textsuperscript{32). Therefore, it was thought that the properties were improved by the EVA penetrating more into the fiber cloth used in this study. The adhesive strength of the bond between Ev-Gf and Ev in our study was approximately one-third of the adhesive strength between Ev-Gf and EVA when bonded to each other at 120°C or higher. On the other hand, another study showed that the adhesive strength between EVA and EVA when bonded to each other at about 100°C\textsuperscript{33} was almost the same as the adhesive strength between Ev-Gf and EVA in our study. A sagging distance of 15 mm below the clamp was considered the most suitable configuration for the forming process when the center of the softened sheet sagged as for the mouthguard vacuum forming procedure\textsuperscript{34}. The test specimens for the delamination test were prepared according to the procedures in their report because the method was successful\textsuperscript{34). Their report inferred that the temperature of the adhesive surface was approximately 110°C just before forming. However, some studies showed that higher adhesive strengths could be obtained using solvents and by adjusting the heating conditions\textsuperscript{20,35). This could be attributed to the fact that although the preparation of the specimens was different, their results were comparable. The present study was conducted under normal atmospheric pressure in dry conditions without using any resin modifier or any processing agent. The bond strength is considered to be clinically more than 4 N/mm\textsuperscript{35). The bond strength might be improved by examining the surface treatment method of the glass fiber. From the results of the present study, it was also thought that molding in a way that the material is sandwiched by the base material is preferable from the biochemical safety point of view to compensate
for the lack of adhesive strength. It showed the possibility of using this fiber-reinforced material in this study as like poly ethylene terephthalate-glycol in the report from Takeda et al.\textsuperscript{15,16}. It is assumed that the glass fibers will not come into contact with the actual oral tissues (teeth, mucosa, etc.). In the future, it needs to evaluate the biocompatibility of the material in a strict sense.

Additional functional and mechanical properties of mouthguards could be evaluated by determining flexural strength and flexural modulus using a three-point bending test. The basic methodological approach in the present study used the three-point bending strength and flexural modulus to compare values and evaluate differences between Ev-Gf and Po-Gf. Moreover, it is appropriate to evaluate the mechanical and functional properties of such experimental reinforced thermoplastic material using a bending test to observe the deformity of the specimens. In addition, these properties determine the degree to which deformation of new reinforced thermoplastic material is minimized during occlusion. Therefore, such materials used for mouthguards must possess sufficient flexural strength and an appropriate elastic modulus. Our research found that the flexural strength of Ev-Gf (29.7 MPa) was significantly higher than that of Po-Gf (14.8 MPa). Perpendicular force should be applied to the specimen’s cross-section in a three-point bending test, and the filler material should be dispersed throughout the sample materials, which was accomplished in the present study experiment.\textsuperscript{20} Thus, the energy generated by the force must either break the filler particles or be dissipated throughout the specimen, showing more resiliency. Therefore, in the present study, it was more difficult to break the reinforced Ev-Gf sample, demonstrating a more resilient structure.

One limitation of the present study is related to the testing of dry specimens only. Further studies are needed in the future under different environmental conditions. The peel durability of the adhesive surface between the specimen, showing more resiliency. Therefore, in the present study, it was more difficult to break the reinforced Ev-Gf sample, demonstrating a more resilient structure.

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CONCLUSION

This pioneer study on reinforced fiberglass mouthguard materials compared the functional and mechanical properties of the two most commonly used materials to determine the gold standard for reinforced thermoplastic mouthguard materials. The fiberglass-reinforced Ev-Gf has the better functional properties for fabricating mouthguards, and these properties will help to reduce the thickness of the mouthguard for single-layer, laminated, or hard insertion type objects. When the fiberglass reinforcement method was applied for advancement of mouthguard materials, EVA proved to be the more desirable base material.

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CONFLICT OF INTEREST

The authors declare no conflicts of interest.

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