Fabrication of Shock Absorbing Photopolymer Composite Material for 3D Printing Sports Mouthguard

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Sports mouthguards (MGs) can reduce the risks of sports-related oral injuries. The aim of this study was to fabricate a composite photopolymer with shock-absorbing properties suitable for use in 3D-printed MGs. By using a commercial, flexible, rubber-like photopolymer as matrix and a commercial rigid simulated polypropylene photopolymer as a reinforcement material, five composites with different Shore A hardness levels were fabricated. Furthermore, four laminated materials were prepared to assess the improvement effects associated with adding a rigid outer layer. The five composites and four laminated materials were evaluated in terms of their shock absorbing capabilities via a steel ball drop impact test along with two types of conventional mouthguard materials. The rubber-like photopolymer composite material compounded with the rigid photopolymer with a Shore A hardness of 50 showed excellent shock absorbing capabilities that were compatible with conventional mouthguard materials, suggesting that this shock absorbing photopolymer composite is a candidate material for 3D-printed sports MGs. If the commercial flexible rubber-like photopolymer is to be applied alone without reinforcement, laminating a rigid photopolymer on the outer surface may be an effective means of improving the shock absorption capabilities of such a MG. We succeeded in fabricating a prototype of a double-layered mouthguard with these composite materials using a 3D digital dental workflow.

Key words: 3D printing, Mouthguard, Photopolymer, Composite material, Shock-absorbing capability

1. Introduction

A sports mouthguard (MG) offers protection by providing a resilient surface to absorb and disperse high-impact energy. MGs can reduce the risks of sports-related oral injuries to the teeth, alveolar bone, soft tissue, and temporomandibular joints, thereby minimizing the potential for concussions [1-11].

The first MG was originally made from gutta-percha in 1892 by Woolf Krause, who worked as a dental practitioner in London [12]. He fashioned such MGs for boxers by placing gutta-percha strips over the upper incisors and requesting them to bite down hard, thereby providing protection for the upper and lower jaws. The oldest MG in the United States of America was custom made from soft
rubber via the lost wax method in 1916 by a dentist in Chicago, Thomas A. Carlos [13,14].

The most popular materials for such custom-made MGs are ethylene-vinyl acetate (EVA) copolymers because of their valid shock absorption performance and ease of forming [15]. Additionally, olefin copolymer materials developed by Japanese manufacturers, such as an ethylene-alpha-olefin copolymer material and a polystyrene-polyolefin copolymer material, have been highly evaluated [16, 17].

Currently, research is being conducted to investigate the reinforcement effects of MGs that utilize titanium or glass fiber [18,19]. Studies have also been conducted to develop new MG materials with high functionalities [20,21]. Yoshida et al. have successfully developed a novel antibacterial mouthguard material manufactured using a silver-nanoparticle-embedded EVA copolymer masterbatch [21].

The modern standard method utilized to fabricate custom-made MGs is achieved via vacuum- or pressure-forming an elastic thermoplastic elastomer sheet over a dental stone model [22]. This thermoforming method is relatively simple compared to the old-fashioned lost wax method, but many working process are required between dental clinics and laboratories, including dental impressions, maxillo-mandibular registration, fabricating and trimming dental stone models, mounting these models on an articulator, thermoforming mouthguard materials, cutting, occlusal adjustments, and finally finishing and polishing such models.

The use of digital impressions reduces the number of clinical and laboratory steps, leading to fast and effective delivery of the final custom-made medical device [23-31]. The application of computer-aided designing and computer-aided manufacturing (CAD/CAM) technologies in dentistry is becoming increasingly popular. The CAD/CAM system offers an alternative to processing indirect dental restorations and fixed prostheses. Recently, these techniques have been expanded for application towards oral drug-delivery devices, mouthpiece-type orthodontic treatment devices, and MGs [32-35].

Although many 3D printing materials exist, rubber-like photopolymers are considered to have the greatest potential for development into new mouthguard materials. However, the hardnesses of rubber-like photopolymers are lower than those of conventional MG materials. Therefore, the hardness values of rubber-like photopolymers may be adjusted via combination with rigid materials.

Additionally, the synergetic effects that provide a rigid-soft laminated structure should be investigated. Considering the typical structure of protective gear utilized in sports, such as helmets, it is well known that the inner layer of this equipment should be soft to provide cushioning while the outer shell should be rigid to provide protection [36].

The aim of this study was to fabricate a composite photopolymer with shock-absorbing properties suitable for 3D printing MGs. We fabricated the candidate materials by compounding and/or laminating different types of 3D printing photopolymers. Next, we investigated their shock absorbing capabilities through the steel ball drop impact test and analyzed the effective structural patterns among the results. We executed a 3D digital workflow of a custom-made MG using a photopolymer composite material and reviewed the quality of the resulting prototype.

2. Experimental

2.1. Fabrication of photopolymer composite materials

FDA-approved flexible rubber-like (Agilus30; Stratasys Ltd., Rehovot, Israel) and rigid simulated polypropylene (Rigur; Stratasys Ltd., Rehovot, Israel) photopolymers were utilized (Table 1). Five types of Agilus30/Rigur photopolymer composites with Shore A hardness values of 40, 50, 60, 70, and 95 (denoted as Compo40, Compo50, Compo60, Compo70, and Compo95, respectively) were fabricated in this study.

Table 1. Mechanical properties of rubber-like and rigid simulated polypropylene photopolymers used in this study [37].
Rigur was utilized as a reinforcement material that was forming the three-dimensional grid structure compounded with the matrix composed of Agilus30. The liquid droplets of the two photopolymers were sprayed according to the composition ratio required to achieve the desired hardness in a similar format to the Polyjet 3D printer (Objet500 Connex3: Stratasys Ltd., Rehovot, Israel) with the software (GrabCAD Print: Stratasys Ltd., Rehovot, Israel) and cured with successive ultraviolet light via cooperation with a digital printing manufacturer (Altech Co., Ltd., Tokyo, Japan).

To assess the improvement effects obtained by adding a rigid outer layer, four kinds of laminated materials were also manufactured to laminate the Rigur with 0.5- and 1-mm thicknesses over Compo50 (R0.5 Compo50, R1.0 Compo50) and Agilus30 (R0.5 Ag30, R1.0 Ag30) by using the same Polyjet 3D printer with the software and cured with ultraviolet light.

2.2. Steel ball drop impact test

The drop test was performed using a modified IM-201 impact testing machine (Tester Sangyo Co., Saitama, Japan) in the laboratory at room temperatures between 20 to 25 °C. Many different impact tests exist, including the Charpy test, Izod test, tensile test, and drop test. The steel ball drop test was selected in this study because it is a popular method that yields results that correspond closely to the actual impact strength of the examined material [38].

![Fig. 1. Experimental setting of free-fall drop impact test.](image)

The impact force was generated by a free-falling stainless-steel ball (weight: 32.6 g, diameter: 20 mm) dropped from a 60-cm height onto the test specimen, which was placed on a steel platform that was supported by a load cell sensor system. Three dynamic compression load cells with rated capacities of 1 kN (LMA-A-1KN; Kyowa Electronic Instruments Co., Tokyo, Japan) were located between the steel platform and base plate of the impact testing machine, which were located 120° apart from one another (Fig. 1).

The control load was set at 660 N based on data obtained for the fracture loads of the upper teeth and jaws of humans. Previous studies have shown that the fracture load of the upper incisor of a human is 607 to 894 N [39,40]. The minimum impact tolerance and average fracture loads of a human maxilla are reported to be approximately 660 and 1,100 N, respectively [41].

The experiments were carried out for Agilus30, Rigur, Agilus30/Rigur photopolymer composites, the laminated materials, and two conventional MG materials (an EVA material: Erkoflex; Erkodent Erich Kopp GmbH, Pfalzgrafenweiler, Germany; and an Ethylene-alpha-olefin copolymer material: MG21; CGK Co., Hiroshima, Japan) (Table 2). The diameters of all samples were 50 mm. Each specimen was tested five times. A control test was performed in the same manner in the absence of a specimen.

![Table 2. Materials and thickness of the test samples.](image)

2.3. Data recording and analysis

The impact force applied to the test specimen was measured by three load cells and recorded in a personal computer through an amplifier (EDX-100A; Kyowa Electronic Instruments Co., Tokyo, Japan) at a sampling rate of 50 kHz. The sum of the impact loads rapidly increased immediately after the steel ball collided with the specimen, attained a peak value, and then decreased. The first peak intensity of the sum of the impact load was regarded as the
maximum impact force (IF\textsubscript{max}). The time required to reach IF\textsubscript{max} (IF\textsubscript{max}-t) and the impulse from the onset of the impact force up to 1.0 ms (\int F dt) were also calculated.

The data were statistically analyzed using one-way ANOVA and Tukey’s HSD tests via JMP14 (SAS Institute Inc., Cary, Nc, USA). The levels of significance were set at \( \alpha = 0.05 \).

![Fig. 2. Computer-assisted designing of the double-layered MG. Upper left: a dental model, upper right: intraoral digital data, middle left: 3D image of inner layer, middle right: 3D image of outer layer, lowers: 3D images of double-layered MG.](image)

2.4. Prototyping of a sports mouthguard with a photopolymer composite material

To manufacture a prototype MG with the fabricated photopolymer composite material, digital information of the dentition and gum were constructed from a dental model (D18FE-500A-QF; Nissin Dental Product Inc., Kyoto, Japan) using an intraoral scanner (CEREC Omnicam; Dentsply Sirona, North Carolina, U.S.A.).

The intraoral data was output as STL (stereolithography) file and the data for double-layered MG with 3-mm thicknesses were designed utilizing the maxillary dentition data using CAD software (Sirona Splint; Dentsply Sirona, North Carolina, U.S.A.) (Fig. 2). The design data of the inner layer and outer layer were output as STL files, and both data of this MG were integrated with CAD software (Materialise Magics; Materialise, Leuven, Belgium). A prototype of a MG was then fabricated using a PolyJet 3D printer (Objet500 Connex3: Stratasys Ltd., Rehovot, Israel) via cooperation with a digital printing manufacturer (Stratasys Japan Co., Ltd, Tokyo, Japan).

The outline shape of the MG was evaluated via ocular inspection. Next, the dimensional accuracy was assessed digitally in terms of the fit of the prototype MG via an optical hand-held 3D scanner (Artec Spider, Artec Group Inc, Luxembourg) and a 3D analysis software (Artec Studio 12 Professional, Artec Group Inc, Luxembourg). The dental artificial model and the inner and outer surfaces of the MG were scanned with the 3D scanner. The digital data were saved as STL files. To assess the fit of the MG, the STL file of the MG was superimposed with the reference STL file of the model on the 3D analysis software and the mean root mean square (RMS) values of the gap space between the model and MG were calculated.

3. Results and discussion

3.1. Photopolymer composite materials

The cross-sectional observations of the compounded and the laminated photopolymer composite materials fabricated in this study are shown in Fig. 3. No inadequacies such as air bubble contamination or gaps between the interfacial surfaces of the hard and soft materials were observed, and the specified thicknesses were maintained.

![Fig. 3. Microscope observations of the photopolymer composite materials (×100, VHX-1000: KEYENCE Co., Osaka, Japan). Upper: Compo50, lower left: R0.5Compo50, lower right: R1.0Compo50.](image)

PolyJet technology, which was used in this study, is a common material jetting technology in a layered or additive approach with thermoplastic and photopolymers. By utilizing this technology, we may easily produce composite materials with three or more layers in a varied sandwich structure (for example, Agilus-Rigur-Agilus and Rigur-Agilus-
Rigur-Agilus). The effects of such multi-layered structures may be investigated via this method. In near future, the biocompatibility of the photopolymers must be assessed via in vitro tests and animal models to ensure patient safety.

3.2. Shock absorbing capability

The IFmax data of the shock absorbing capability and the results of the statistical analyses are shown in Table 3. Agilus30 by itself demonstrated a greater IFmax than the conventional MG materials, which confirmed its poor shock absorbing capability (p < 0.05).

Table 3. The IFmax data obtained from steel ball drop impact test performed in this study. Units: N, S.D.: standard deviation, Rate: the ratio of sample load with the control, statistical*: same letter were not significantly different (p<0.05).

| Material         | IFmax (N) | S.D. | Rate (%) | Statistical* |
|------------------|-----------|------|----------|--------------|
| Control          | 666.2     | 16.5 | 100.0    |              |
| Agilus30         | 487.2     | 15.9 | 73.1     | D            |
| Compo40          | 381.2     | 6.4  | 57.2     | I, J         |
| Compo50          | 367.9     | 5.7  | 55.2     | J, K         |
| Compo60          | 374.0     | 1.2  | 56.1     | I, J, K      |
| Compo70          | 406.4     | 6.2  | 61.0     | G            |
| Compo95          | 513.5     | 4.7  | 77.1     | C            |
| R-0.5mm          | 591.6     | 9.2  | 88.8     | B            |
| R-1mm            | 652.7     | 11.9 | 98.0     | A            |
| R0.5Ag30         | 401.4     | 5.3  | 60.3     | G            |
| R1.0Ag30         | 434.2     | 8.4  | 65.2     | E, F         |
| R0.5Compo50      | 393.9     | 4.4  | 59.1     | G, H         |
| R1.0Compo50      | 430.0     | 5.4  | 64.5     | E, F         |
| ERK-1mm          | 446.2     | 13.8 | 67.0     | E            |
| ERK-2mm          | 423.7     | 4.0  | 63.6     | F            |
| ERK-3mm          | 376.3     | 4.6  | 56.5     | I, J         |
| ERK-4mm          | 358.9     | 3.6  | 53.9     | K            |
| MG21-2mm         | 425.1     | 2.6  | 63.8     | F            |
| MG21-3mm         | 387.6     | 4.5  | 58.2     | H, I         |
| MG21-4mm         | 372.8     | 2.9  | 56.0     | I, J, K      |

The IFmax of the Compo series was significantly smaller than that of Agilus30 (p < 0.05). There was no significant difference between the IFmax data of the Compo series and the conventional MG materials for the same 3-mm thickness (p = 0.323). The Compo 40, Compo50, and Compo60 samples demonstrated the maximum level of shock absorbing capability. They were equivalent to the conventional MG material with a 3- to 4-mm thickness.

The IFmax data of the photopolymer composite materials was approximated by a quadratic curve, as shown in Fig. 4. The Compo50 performed the best among the Compo series, suggesting that the optimal hardness for the shock absorbing photopolymer composite for 3D-printed MGs differed by approximately 20 to 30 in terms of Shore A hardness compared to the conventional MG materials.

The IFmax data of the R0.5Compo50 and R1.0Compo50 samples were significantly greater than that of Compo50 (p < 0.05), while those of the R0.5Ag30 and R1.0Ag30 samples were significantly smaller than that of Agilus30 (p < 0.05). Likewise, the fFdt data of the R0.5Compo50 and R1.0Compo50 samples were greater than that of Compo50 (Fig. 5). Accordingly, the synergetic effects of these materials in combination with the composition and lamination reinforcement were not apparent. Instead, the lamination process resulted in the deterioration of the shock absorbing capability of the resulting samples.

Conversely, the fFdt data of R0.5Ag30 and R1.0Ag30 were slightly improved in the latter section from 0.6 ms to 1.0 ms. Therefore, it was considered that maintaining the hardness difference between materials may be important to adequately realize the lamination reinforcement effect. However, it was unclear what effects such lamination will provide when another type of reinforcement material is used, such like carbon- and/or glass-fiber [42], which should be examined in future studies.

These results suggest that Compo50 is a leading material candidate for use in 3D-printed sports MGs. Additionally, it was found that if a commercial rubber-like material such as Agilus30 is applied without 3D-forming composition reinforcement, it is better to laminate rigid material onto the outer surface of such a structure to improve the shock absorption performance of the material.

The IFmax-t data obtained from the steel ball drop impact test with 3-mm thickness samples
performed in this study are shown in Fig. 6. The IFmax-t of both conventional MG materials were nearly identical (approximately 0.5 ms). The IFmax-t data of the Compo series and the lamination series were significantly shorter than those of the conventional MG materials ($p < 0.05$). The data collected for Compo50 was the closest among all examined materials to that of the conventional MG materials.

Fig. 5. The impulse data obtained from steel ball drop impact test performed in this study. Left: Compo50 series, right: Agilus30 series.

Fig. 6. The IFmax-t data obtained from steel ball drop impact test performed in this study. Units: ms, error bars: standard deviation, values with same superscript letter were not significantly different ($p < 0.05$).

3.3. 3D-printed prototype of a sports mouthguard

The 3D-printed prototype of a MG was successfully manufactured with a desired shape in approximately 2 hours (Fig. 7). The reproducibility of the desired thickness was high. The mean RMS values of the gap space between the model and prototype MG obtained by digital analysis were 0.225 mm. These mean RMS values of the gap space were much smaller than the values obtained when fabricating a MG using the conventional thermoforming technique, which demonstrated that the adaptation to the dentition and gingiva on the dental model was satisfactory [47]. Well-fitted custom-made MGs have been shown to be more effective in suppressing the vibration of impacted teeth [48].

Digital dentistry has the potential to produce a MG in a single day [33,34]. When fabricating MGs with a conventional vacuum or pressure forming machine, uniform thicknesses could not be achieved and may vary depending on the anatomic features of the dentition model [33]. Once the digital data for an MG is created and stored as an STL file, there is no need to restart the procedure if the anatomic features of the dentitions, gingiva, and jaws do not change, such that MGs with uniform thicknesses...
may be duplicated many times using a 3D printer.

There are two methods utilized to 3D print multi-layered MGs [49]: (1) the simultaneous printing method, wherein the inner and outer layers are printed simultaneously; and (2) the engaging method, wherein the inner and outer layers are printed separately. The former method, which was used in this study, provides more accurate manufacturing compared to the latter. However, a slight gap is sometimes formed between the interfacial surfaces of the inner and outer layers in either method when conventional commercial dental CAD software is used, as these programs do not possess the functionality to support the use of this method. This issue may be solved in conventional dental CAD software by setting the base point and supplementing the interlaminar data in industrial CAD software when fabricating a multi-layered structure with multiple materials. Therefore, it is necessary to create an exclusive software program for the 3D manufacturing of multi-layered MGs.

4. Conclusion

The composite materials for the fabrication of MGs were prepared by forming various patterns of a rigid simulated polypropylene photopolymer and a soft rubber-like photopolymer, and the shock absorbing properties of these materials were investigated. The rubber-like photopolymer composite material compounded with the rigid photopolymer with a Shore A hardness of 50 demonstrated excellent shock absorbing capabilities that are compatible with conventional MG copolymer materials. If the soft rubber-like photopolymer is to be applied alone without reinforcement, the shock absorption characteristics of the MG may be improved via lamination of the rigid photopolymer on the outer surface of the MG.

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