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Optimization of strontium aluminate-based mechanoluminescence materials for occlusal examination of artificial tooth

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ABSTRACT

This work presents a novel approach for evaluating the occlusal examination of artificial tooth based on the mechanoluminescence (ML) materials. The rare earth doped strontium aluminate ($SrAl_2O_4$: Eu^{2+}, Dy^{3+}; SAOED) was chosen as the ML material, which was further composited with the commercial denture base resin (DBR) to determine its feasibility for the mechanics analysis of artificial tooth occlusion. To eliminate negative factors for occlusal analysis, SAOED was first optimized to exhibit a rapid decay of afterglow and enhanced ML intensity. The luminescent characterizations of the SAOED/DBR composites suggest DBR is a desirable elastic-supporter for nondestructive ML generation. Furthermore, the introduction of SAOED improved the mechanical performance of DBR, and its biocompatibility was maintained at the same time. These results suggest the feasibility of the idea to detect the mechanics in occlusal examination of artificial tooth based on ML. The bright and sensitive ML from the constructed standard artificial tooth models could guide clinicians to purposefully adjust the occlusal surface until a balanced occlusion established.

1. Introduction

Dentition defect is a common oral-maxillofacial disease, which will further arouse a series of complications (e.g., temporomandibular joint disorder, decreased masticatory efficiency, alveolar bone atrophy, etc.) if there is no artificial tooth applied [1, 2]. In addition to the properties of artificial tooth itself, the occlusal-articulation of the filled tooth is crucial adjective in clinic [3, 4]. The inappropriate occlusion of the artificial tooth would also result in dysfunction of the oral-maxillofacial system and temporomandibular joints. An accurate identification of the occlusal high points and interference points of artificial tooth is prerequisite for the establishment of a correct occlusion in clinic [5]. At present, articulating paper is widely employed as a diagnostic tool for occlusal analysis of dental practice [5, 6]. It is extensively accepted that occlusal contact point and occlusal load could be exhibited by the mark size and spot of articulating paper [7]. Generally, the occlusal area with greater contact pressure represents markings with darker color. Although articulating paper is able to record occlusion information, it suffers the problems of low sensitivity and inaccuracy which have received increasing attention [8]. In some recent researches [9, 10], they even showed that the relationship between the color of contact area of articulating paper and contact force was reversed compared with the usually recognized one. Therefore, it is highly required to develop a more reliable approach to detect mechanics of artificial tooth during occlusal test.

Mechanoluminescence (ML) is a type of luminescence produced by mechanical stimuli, e.g., grinding, impact, rub, press and stretching [11]. The phenomenon of ML has been known for centuries. However, it is only in recent years that ML has attracted much attention [12–14]. This is because most of the previously observed ML belongs to destructive luminescence, showing limited application prospect. Since the non-destructive elastico-mechanoluminescence was realized by Xu et al. in 1999 [15], much more researches about ML have been carried out, and a series of advanced applications based on ML have been explored, e.g., stress sensor [16], flexible handwriting device [17], and wind-driven displaying device [18]. Since ML materials could be driven by various mechanical stimuli and the oral environment is filled with mechanics and friction, the combination of ML materials and artificial tooth is supposed to be an effective approach to respond to the mechanics information for occlusal analysis.

Inspired by the above consideration, we applied ML materials for articulating test in this work. The rare earth doped strontium aluminate ($SrAl_2O_4$: Eu^{2+}, Dy^{3+}; SAOED) was employed as the ML component because of its bright and sensitive ML, as well as the excellent chemical

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and physical stability [16, 19]. In addition, compared with the sulfide-based ML materials [20, 21], SAOED is environmentally friendly and non-toxic, which is much more suitable for oral occlusion application. It should be noted that SAOED is also a typical long-lasting phosphor which could emit light for hundreds of hours after removing the irradiation source [22]. Such afterglow behavior goes against the occlusal analysis. To eliminate the interference from afterglow, the concentration and trap depths in structure were regulated first via adjusting the chemical composition of SAOED. The optimized SAOED showed rarely afterglow after removing the irradiation source just for 20 s, while its ML intensity was greatly enhanced. The optimized SAOED was then composited with the widely used denture base resin (DBR). The investigation results suggest that the introduction of SAOED in DBR could not only emit intense ML, but also exhibit good biocompatibility and enhanced mechanical and tribological performance. The construction of oral standard models based on the SAOED/DBR composite further demonstrates the feasibility of ML materials for occlusal analysis.

2. Experimental details

2.1. Synthesis of SAOED

$Sr_{1-x-y}Al_2O_4$: xEu^{2+}, yDy^{3+} (x = 0.01, 0.02, 0.03, 0.04, y = 0, 0.01, 0.02, 0.03) was synthesized via a solid-state reaction. For each sample of $Sr_{1-x-y}Al_2O_4$: xEu^{2+}, yDy^{3+}, the total amount was set to 0.05 mol. First, stoichiometric $SrCO_3$ (99%), Al_2O_3 (99%), Eu_2O_3 (99.99%), and Dy_2O_3 (99.99%) were mixed and thoroughly ground in an agate mortar with the assistance of ethanol. Then, the mixture was transferred to an alumina crucible and sintered in a tube furnace (GSL-1600X, Hefei Ke Jing Material Technology Co., Ltd.) at 1300 °C for 4 h under reducing atmosphere (90% N_2/10% H_2). After cooling to room temperature, the reacted sample were collected and ground for later use.

2.2. Synthesis of SAOED/epoxy resin composites

To achieve non-destructive elastico-ML for characterization and optimization, the as-synthesized SAOED were composited with epoxy resin. SAOED and epoxy resin were mixed with a weight ratio of 1:7. The mixture was mechanically stirred in a beaker, and then poured into a customized spherical mold with size of 25 mm in diameter and 20 mm in thickness. Finally, the mixture was solidified under 60 °C for 2 h in an oven and the SAOED/epoxy resin composite was obtained.

2.3. Synthesis of SAOED/DBR composites

SAOED/DBR composites were prepared for evaluating the feasibility of ML materials for mechanics detection of artificial tooth. SAOED and the commercial DBR powder (composed of 99% polymethyl methacrylate powder and 1% benzoyl peroxide) were first mixed with a certain weight ratio in an agate mortar. After thoroughly ground with an appropriate amount of ethanol, the mixture powder was transferred to a beaker, and methyl methacrylate monomer was added with half weight of the powder. In order to prepare samples with size of 25 mm in diameter and 20 mm in thickness for ML tests, the mixture was transferred to a customized mold and cured under room temperature for 1 h. The composites prepared with the SAOED to DBR weight ratios of 1:8, 1:7, 1:6 and 1:5 were named to S1-D8, S1-D7, S1-D6 and S1-D5, respectively. The artificial tooth models were fabricated by employing a denture mold, and the other procedures were same with those of SAOED/DBR composites.

2.4. MTT assay

The SAOED/DBR composites with size of 10 × 10 × 2 mm were sterilized under ultraviolet (UV) radiation for overnight and used for cell cultures. L929 Fibroblasts (a clonal mouse fibroblasts cell line, ATCC, Rockville, MD, USA) were employed to evaluate the biocompatibility and proliferation behaviors of the prepared SAOED/DBR composites. The fibroblasts L929 cells were cultured at a density of 10^4 cells per sample in Dulbecco's modified Eagle's medium (DMEM, Gibco, USA) supplemented with fetal bovine serum (FBS, 10%, Australia Origin, Gibco, USA), Penicillin-Streptomycin (100 UI mL^{-1}) and D-glucose (4.5 g L^{-1}) under 5% aseptic CO_2 atmosphere and the temperature of 37 °C. The media was refreshed every two days until the cells reached confluence. Then, the composites were placed into a 24-well plate and seeded with 10^4 cell/mL concentration on each well. At a given time (1, 2 and 4 days), 100 μL of 3-(4,5-dimethyl-2-thiazolyl)-2,5-diphenyl-2-H-tetrazolium bromide (MTT, St. Louis, MO, USA) solution was injected into each well. The cells were cultured for another 4 h. The lower blue formazan reaction product was dissolved by adding 750 μL of dimethylsulfoxide. Then, the mixed solution was transferred into a 96-well plate, and its absorbance was measured by a microplate reader (Bio-Rad iMark) for three times for calculating the average absorbance value. For comparison, the absorbance of the cells only in the culture medium was selected as control. The proliferation performance of the as-prepared samples was obtained by comparing the absorbance of experiment group and control group.

2.5. Characterizations

The crystal structure of SAOED was studied by X-Ray Diffraction (XRD, D/max-2400, Rigaku, Cu-Kα). The absorption spectra were collected by a UV–visible (UV–vis) spectrophotometer (PE lamda 950, USA) using $BaSO_4$ as a reference. The photoluminescence (PL) spectra and decay curves were measured by a fluorescence spectrophotometer (Omni-λ300i, Zolix, China). The ML performance were tested using a ML test system built by Lanzhou Institute of Chemical Physics that consists of electronic universal testing machine (WDT-5, Tian Shui Hong Shan Testing Machine New Technology Development Co., Ltd), rotary tribometer (MS-T3001, Lanzhou Huahui Instrument Technology Co., Ltd.), and photomultiplier tubes and data collection system (DCS103, Zolix, China).

The tribological properties of the SAOED/DBR composites was evaluated using a CSM reciprocating tribometer at ambient conditions (RH = 30 ± 5%). A GCr15 bearing steel ball with a diameter of 6 mm was used as the counterpart. The reciprocating frequency and stroke were 0.5 Hz and 5 mm, respectively. The normal force was selected as 10 N. The coefficient of friction and sliding passes were recorded automatically.

The wear rates of composites after the tests were calculated as follows:

$$W = \frac{V}{L \times D}$$

where W, V, L and D represent wear rate, wear volume, applied load and sliding distance, respectively. The cross-sectional area of the thickness of the samples was measured by Alpha-Step D-100 profilemeter (KLA-Tencor, San Francisco, CA). The hardness of composite is presented with Shore A durometer. Three replicate tests were carried out for each specimen.

3. Results and discussion

The crystal structure of monoclinic $SrAl_2O_4$ with space group of $P2_1$ is illustrated in Fig. 1a. In the unit cell, there are three independent cation sites for Eu^{2+} and Dy^{3+} doping, i.e., two nine-coordinated Sr^{2+} sites and one four-coordinated Al^{3+} site. Because of the radius difference among Eu^{2+} (0.117 nm), Dy^{3+} (0.091 nm), Sr^{2+} (0.118 nm) and Al^{3+} (0.053 nm), it is suggested that Eu^{2+} and Dy^{3+} ions should substitute the sites of Sr^{2+} ions in SAOED [23, 24]. The crystal phase of the as-prepared SAOED powder was determined by XRD, as shown in Fig. 1b. It is observed that the XRD patterns of SAOED is identical with