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Flexible fiber-reinforced composites with improved interfacial adhesion by mussel-inspired polydopamine and poly(methyl methacrylate) coating

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ABSTRACT

To obtain a kind of light-curable fiber-reinforced composite for dental restoration, an excellent interfacial adhesion between the fiber and the acrylate resin matrix is quite essential. Herein, surface modification on glass fibers were carried out by coating them with poly(methyl methacrylate) (PMMA), polydopamine (PDA), or both. The PMMA or PDA coating was performed by soaking fibers in PMMA/acetone solution or dopamine aqueous solution. PDA/PMMA co-coated glass fibers were obtained by further soaking PDA-coated fibers in PMMA/acetone solution. These modified fibers were impregnated with bisphenol A glycidyl methacrylate (Bis-GMA)/triethylene glycol dimethacrylate (TEGDMA) (5:5, w/w) dental resin at a volume fraction of 75%, using unmodified fibers as reference. Light-cured specimens were submitted to evaluations including flexural properties, morphological observation, dynamic mechanical thermal analysis (DMTA) and pull-out test. In comparison with unmodified glass fibers, all the modified glass fibers showed enhancements in flexural strength and modulus of Bis-GMA/TEGDMA resin composites. Results of DMTA and pull-out tests confirmed that surface modification had significantly improved the interfacial adhesion between the glass fiber and the resin matrix. Particularly, the PDA/PMMA co-coated glass fibers displayed the most efficient reinforcement and the strongest interfacial adhesion due to the synergetic effects of PDA and PMMA. It indicated that co-coating method was a promising approach in modifying the interfacial compatibility between inorganic glass fiber and organic resin matrix.

1. Introduction

Most endodontically treated teeth require a post-and-core build-up for restoring the teeth to optimum health and function [1]. Prefabricated fiber posts are the most popularly used materials for these endodontical treatments due to their advantages in biocompatibility, mechanical properties and aesthetic features [2,3]. Many prefabricated fiber posts (e.g. RTD, 3 M and DMG) are now easily obtained worldwide [4,5]. However, each tooth in the arch exhibits anatomic characteristics such as root curvature, mesio-distal width and labio-lingual dimension. In other words, root anatomy dictates post selection for endodontical treatments. In the case of abnormal curved root canal, prefabricated fiber posts cannot fit into the canal well, and thus fail to achieve satisfactory endodontical treatment [6]. Flexible prepreg, i.e. pre-impregnated composite fibers, can be a good solution for this situation because of its chairside operability [7].

The bonding of a post to the tooth structure should improve the prognosis of the post-core restored tooth by increasing post retention and by reinforcing the tooth structure [8]. Prepregs composed of glass fibers and light-curable acrylate resin are preferred for dental restoration because they can form strong bonding to acrylate cements, which are so essentially required to fix posts in root canals firmly [9].

In the case of fiber-reinforced dental composites, a primary issue is the interfacial adhesion between the fiber and the resin matrix, especially in a moist environment [10,11]. Perfect adhesion is absolutely necessary to transfer load from the matrix to the fiber, i.e. the strong fiber carries the load, while the matrix distributes it and transfers it from one fiber to the other. Glass fibers are generally treated with silane coupling agent to enhance chemical bonds between the fiber and the resin matrix [12]. The Si group in the silane coupling agent is able to interact with inorganic surface of glass fiber, while another group in silane coupling agent is able to form covalent bonds with the organic resin matrix. Apparently, organic resin matrices with different chemical structures dictate silane coupling agent selection. The bonding between glass fiber and silane coupling agent via the formation of Si-O-Si is reportedly not so stable in aqueous environments [13]. Plasma treatment is also applied to improve fiber-matrix adhesion, in which newly activated components or surface roughness are introduced onto the fiber's surface.

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The adhesion between the fiber and the matrix was thus enhanced [14]. Due to its relatively high cost and low treating efficiency, however, plasma treatment is not popularly used in scaled-up production. For dental composites, an ideal modification method is low-cost, effective, nontoxic and causes no adverse effect on mechanical properties of glass fibers. In addition, it is welcomed if the modified fibers can remain acceptable interfacial adhesion when the composites are used in aqueous environments.

To achieve this goal, surface modification with mussel-inspired dopamine (DA) seems one of the simplest and most effective strategies in enhancing interfacial adhesion for various composites [15]. The amino acid dihydroxyphenylalanine (DOPA), has been identified to be largely responsible for the cohesive and adhesive strengths of mussel adhesive proteins in aqueous environments [16]. As an analogue of DOPA, DA resembles the strong adhesion ability of DOPA, and it is able to attach onto different substrates (e.g. metal, ceramic, polymer) by virtue of the strong anchoring force of its catechol functionality [17]. The DA coating process is easy and mild. Briefly, substrates are simply soaked in a DA aqueous solution at room temperature, and a polydopamine (PDA) layer on the substrate surface is readily formed via the oxidative polymerization of DA [18]. Various substrates including hydroxyapatite, carbon nanotube, glass, polytetraethylene (PTFE), polyester, silicon rubber, etc., have all been surface-coated with PDA in a similar manner [19–27]. The method would not ruin any structure of the original substrate, and it is extremely useful for biomedical applications because it does not require the time-consuming synthesis of complex linkers and the process is solvent-free and nontoxic.

In the aspect of using PDA modifications to improve interfacial adhesion in composites, some achievements were reported in recent years. To improve the dispersibility and interfacial interaction of nanofillers in polymer nanocomposites, a layer of PDA was constructed onto the surface of clay. It was found able to benefit not only the dispersion of clay in epoxy resin matrix but also to enhance the effective interfacial stress transfer [27]. PDA coating was also used to coat aramid fibers. In comparison with silane coupling modifications, the adhesion between the PDA-modified fibers and rubber matrix was remarkably improved [28]. The surface free energy of both carbon and glass fibers was found to increase after PDA coating; therefore, the fibers displayed quite good wettability to epoxy resin [29].

With these approaches, in this study, PDA coating on glass fibers was proposed to achieve good infiltration with light-curable acrylate resin, and thus their interfacial adhesion could be improved. In view of the structure similarity and excellent compatibility between poly(methyl methacrylate) (PMMA) and acrylate resins [30–32], PMMA coating or PDA/PMMA co-coating on glass fibers was also investigated. As illustrated in Fig. 1, prepregs consisting of glass fibers and bisphenol A glycidyl methacrylate (Bis-GMA) dental resin were prepared, light-cured and submitted to characterizations including flexural properties, dynamic mechanical thermal analysis (DMTA) and pull-out test. In comparison with unmodified glass fibers, the null hypothesis of the present study was that the aforementioned modifications on glass fibers could not significantly improve the interfacial adhesion between the fiber and the resin matrix.

2. Experimental

2.1. Materials

The glass fibers used in this study were SE8400LS from Owens Corning Co., Ltd. (USA). The fibers were cleaned in distilled water under ultrasonication for 10 min before they were used in the following modifications. Dental resins as Bis-GMA, triethylene glycol dimethacrylate (TEGDMA), 2-(dimethylamino) ethyl methacrylate (DMAEMA), camphorquinone (CQ) and PMMA (M_w = 35,000) were purchased from Sigma-Aldrich (USA). Dopamine hydrochloride (DA · HCl) was also purchased from Sigma-Aldrich and used directly. Tris(hydroxymethyl aminomethane) (Tris) was purchased from Alfa Aesar Company (USA). Other chemicals used in the study were from Beijing Chemical Plant (China).

2.2. Coating modifications on glass fibers

2.2.1. PDA coating process

A certain amount of DA · HCl was dissolved in distilled water to get a transparent solution with concentrations of 1.0, 2.0 or 4.0 mg/mL, and the solution pH was adjusted to 8.5 by adding Tris. Pre-cleaned glass fibers were then immersed into the solution at room temperature for different times (2, 10 or 24 h). After the reaction, glass fibers were

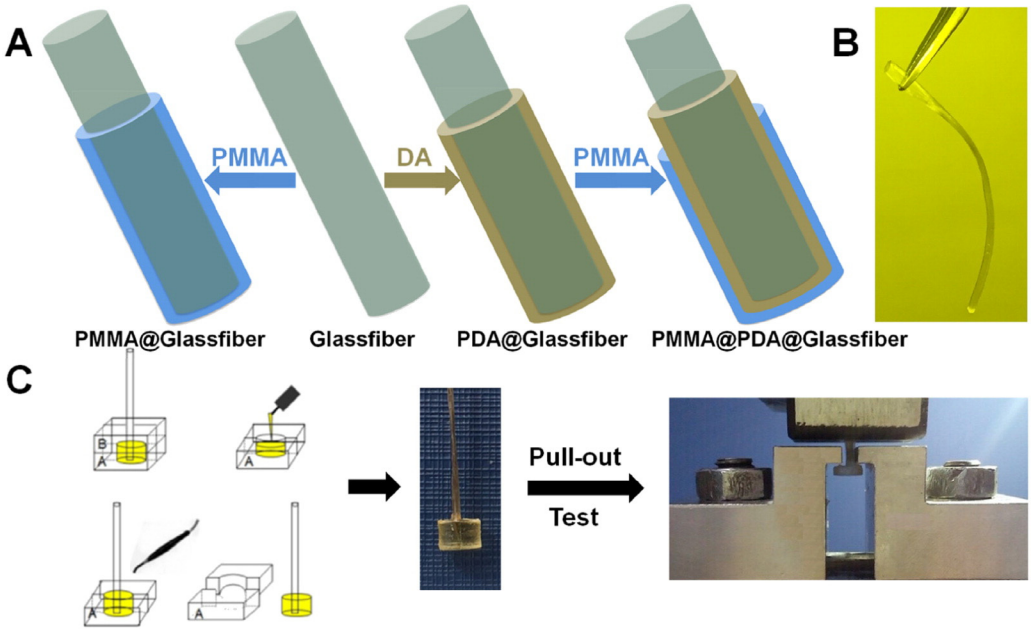

Fig. 1. Schematic description of glass fibers with different surface modifications (A), the prepared prepreg (B) and the following composite samples for pull-out test (C).