Preparation of A PQAS-containing Dental GIC Cement for Improved Antibacterial Function

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Introduction

Secondary caries that often occurs at the interface between the restoration and the cavity preparation is mainly caused by demineralization of tooth structure due to invasion of plaque bacteria (acidproducing bacteria) such as Streptococcus mutans (S. mutans) in the presence of fermentable carbohydrates [1]. Although glass-ionomer cements (GICs) are found to be the most cariostatic and somehow antibacterial due to release of fluoride [2], annual clinical surveys found that secondary caries was still the main reason for GIC failure [3], indicating that the fluoride-release from GICs is not potent enough to inhibit bacterial growth or combat bacterial destruction. Although numerous efforts have been made on improving antibacterial activities of dental restoratives, most of them have been focused on release or slow-release of various incorporated low molecular weight (MW) antibacterial agents such as antibiotics, zinc ions, silver ions, iodine and chlorhexidine (CHX). However, release or slow-release can lead or has led to reduction of mechanical properties of the restoratives over time, short-term effectiveness, and possible toxicity to surrounding tissues if the dose or release is not properly controlled [4]. Polymers containing quaternary ammonium (QAS) or phosphonium salt (QPS) groups have been studied extensively as an important antimicrobial material and used for a variety of applications due to their potent antimicrobial activities [4].

The objective of this study was to synthesize a new poly(acrylic acid-co-itaconic acid) with pendent quaternary ammonium salt (PQAS) and explore the effects of this PQAS on mechanical strength and antibacterial activity of commercial Fuji II LC and recently developed experimental high-strength cements.

Materials and Methods

The synthesis of the poly(acrylic acid-co-itaconic acid) (PAAIA) copolymer followed the procedures described elsewhere [5]. The synthesis of the antibacterial QAS and grafting it onto the PAAIA to form the PQAS, where the grafting ratio of QAS = 50% by mole, followed the similar published protocols [4,5]. The synthesis of the light-curable star-shape poly(acrylic acid) for formulation of the experimental high-strength cement has been described in our previous publication [6]. The experimental cement (EXPGIC) formulation and sample preparation followed the published protocol [6].

Fuji II LC was used as control and sample mixing was conducted per manufacturer's instruction. Fuji II LC glass powder was either used alone or mixed

with the synthesized PQAS to formulate the cements. Specimens were exposed to blue light for 2 min, followed by conditioning in 100% humidity for 15 min and then in distilled water at 37 °C prior to testing.

The CS test was performed on a screw-driven mechanical tester. The sample sizes were n = 6-8 for each formulation. The antibacterial test was conducted following the published procedures [7]. S. mutans (oral bacterial strain) was used for evaluation of antibacterial activity of GICs. Triple replica was used to obtain a mean value for each material. One-way analysis of variance (ANOVA) with the post hoc Tukey-Kramer multiplerange test was used to determine significant differences of both CS and antibacterial tests among the materials in each group. A level of $\alpha=0.05$ was used for statistical significance.

Results

With PQAS addition, the studied cements showed a reduction in CS with 25-95% for Fuji II LC and 13-78% for the experimental cement and a reduction in S. mutans viability with 40-79% for Fuji II LC and 40-91% for the experimental cement. The experimental cement showed less CS reduction and higher antibacterial activity as compared to Fuji II LC. The long-term aging study indicates that the cements are long-lasting antibacterial with no PQAS leaching. It appears that the experimental cement is a clinically attractive dental restorative that can be potentially used for long-lasting restorations due to its high mechanical strength and long-lasting antibacterial function.

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