Preparation of Light-Cured 3-Arm Star-shape Poly(acrylic acid)-based Glass-ionomer Cements via Aqueous Atom Transfer Radical Polymerization

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Introduction:

Resin modified glass ionomer cement (RMGIC) has been used as dental restoratives for decades [1]. Typical RMGIC cement is formulated from two-component system: a calcium fluoroaluminosilicate glass powder and a polyacid solution with polymerizable units [2]. Although RMGIC has numerous advantages, such as good biocompatibility, anti-cariogenicity, and direct adherence to tooth structure, it is still considered inferior to resin composites in mechanical strength.

One of the approaches to improve RMGIC's mechanical properties is to increase the molecular weight (MW) of polyacids. It is known that high MW versions of currently used linear polyacids are very likely to yield a viscous solution, thus making the cement mixing problematic.

Recently, Xie et al found that star-shape polyacid synthesized via atom transfer radical polymerization (ATRP) in organic solvent is able to form solutions with reasonable viscosities, even with a higher MW. The formed RMGIC showed improved mechanical properties as compared to commercial products [3].

In this study, we propose to utilize ATRP reaction in aqueous medium to synthesize star-shape polyacids for improved mechanical strengths. The objective of this study was to synthesize 3-arm poly(acrylic acid) via aqueous ATRP, graft the polyacid with pendent polymerizable groups, formulate it with commercial Fuji II LC glass powder and evaluate the mechanical properties of the formed cements.

Materials and Methods:

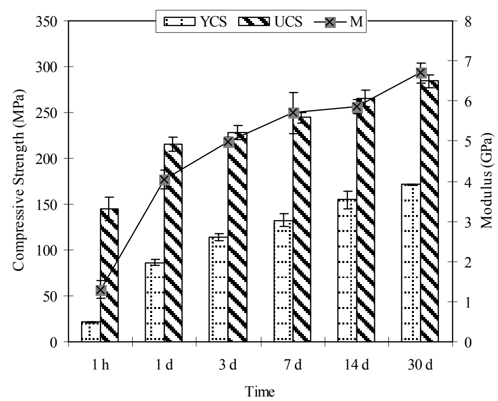
The 3-arm water soluble initiator was synthesized via similar procedures described in previous paper [3]. Generally, tris(hydroxymethyl)aminomethane (THMAM) was linked with trimellitic anhydride (TMA) by amide formation, followed by esterification with 2-bromopropionyl bromide (BPB). The 3-arm polyacid was synthesized in a methanol/water mixture through ATRP reaction in the presence of the synthesized 3-arm initiator and sodium acrylate in methanol-water mixtures [4]. 2-isocyanatoethyl methacrylate (IEM) was then grafted onto the polyacid at 50% molar ratio of carboxyl groups [3].

The cement was formulated by mixing Fuji II LC glass powder with a solution of IEM-grafted polyacid, co-monomer and water at a weight ratio of 50/25/25. Photo initiation system was used to cure the cement. The cement was fabricated into Φ4mm×8mm cylinders for compressive strength (CS); Φ4mm×2mm plates for diametral tensile strength (DTS) and 3mm×3mm×25mm bars for flexural strength (FS) tests. Specimens were

conditioned in distilled water at 37 °C prior to testing. Testing was conducted on a screw-driven mechanical testing machine with a crosshead speed of 1 mm/min. The FS test was performed in three-point bending, with a span of 20 mm between supports. The sample sizes were n = 6-8 for each test. One-way ANOVA with the *post hoc* Tukey-Kramer multiple range test was used to determine significant differences of strengths at a level of $\alpha = 0.05$.

Results

We have successfully synthesized the 3-arm polyacids via aqueous ATRP and used them to formulate the LCGICs. The effects of MW, comonomer and P/L ratio were significant. The cement containing polyacid with a MW of 4274 (Daltons) showed the most desirable mechanical and working properties. The cement containing methacryloyl beta-alanine (comonomer) showed a higher mechanical strength than the one with HEMA. Higher P/L ratio gives higher CS but also makes cement mixing relatively difficult. A sustained CS increase was observed in the course of aging in water over 1 month, with the values (MPa) of 284.1 for ultimate CS, 171.7 for yield CS, and 6700 for modulus. The experimental cement was significantly higher in mechanical strength than Fuji II LC cement.



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References:

1. Wilson AD. Int J Prosthodont 1990;3:425-429
2. McCabe JF. Biomaterials. 1998; 19:521-527
3. Xie D, Park JG, Zhao J. Dent Mater. 2006 Mar 14
4. Wang XS, Lascelles SF, Jackson RA, Armes SP. Chem Commun. 1999; 18:1817-181