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Synthesis and characterization of a novel *N*-vinylcaprolactam-containing acrylic acid terpolymer for applications in glass-ionomer dental cements

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Abstract

In this study a novel *N*-vinylcaprolactam (NVC)-containing copolymer of acrylic–itaconic acid was synthesized, characterized and incorporated into Fuji IX conventional glass-ionomer cement (GIC). Subsequently, the effects of incorporation of synthesized terpolymer on the mechanical properties of GIC were studied. The synthesized terpolymer was characterized using ^{1}H nuclear magnetic resonance, Fourier transform infrared and Raman spectroscopy. The viscosity and molecular weight of the terpolymer were also measured. The compressive strength (CS), diametral tensile strength (DTS) and biaxial flexural strength (BFS) of the modified GICs were evaluated after 24 h and 1 week of immersion in distilled water at 37 °C. The handling properties (working and setting times) of the resulting modified cements were also evaluated. One-way analysis of variance was used to study the statistical significance of the mechanical strengths and handling properties in comparison to the control group. The results showed that NVC-containing GIC samples exhibited significantly higher ($P < 0.05$) DTS (38.3 ± 10.9 MPa) and BFS (82.2 ± 12.8 MPa) in comparison to Fuji IX GIC (DTS $= 19.6 \pm 11.4$ MPa; BFS $= 41.3 \pm 10.5$ MPa). The experimental cement also showed higher but not statistically significant values for CS compared to the control material (CS for NVC-containing sample $= 303 \pm 32.8$ MPa; CS for Fuji XI $= 236 \pm 41.5$ MPa). Novel NVC-containing GIC has been developed in this study, with a 28% increase in CS. The presented GIC is capable of doubling the DTS and BFS in comparison to commercial Fuji IX GIC. The working properties of NVC-containing glass-ionomer formulations are comparable and are acceptable for water-based cements.
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1. Introduction

Glass-ionomer cements (GICs) have been successfully used as a dental restorative material since their invention by Wilson and Kent [1,2]. GICs contain ion-leachable fluoroaluminosilicate glass that can react with water-soluble acids such as polyacrylic acid (PAA) [3]. The cement is the product of an acid–base reaction between the silicate glass as the basic part and the polyacrylic homo- and copolymers. Similar to the powder of silicate glasses, glass-ionomer powder is finely ground ceramic, which is soluble in acids. The main components of powder are silica (SiO_2), alumina (Al_2O_3) and calcium fluoride (CaF_2) as flux, sodium fluoride (NaF) and cryolite (Na_3AlF_6) or aluminum phosphate ($AlPO_4$). Phosphate and fluoride are used in glass to modify the setting characteristics. The main structure of the glass is still the alumina and silica, which form the backbone and skeletal structure of the

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glass. The acid attacks and degrades the glass structure, releasing metal cations, which are subsequently chelated by the carboxylate groups and cross-link the polyacid chains [4].

GIC is well known for its unique properties such as direct adhesion to tooth structure and base metals [1–4], anticariogenic properties due to release of fluoride [5], minimal microleakage at the tooth–enamel interface due to low shrinkage, thermal compatibility with tooth–enamel and dentine due to the low coefficient of thermal expansion similar to that of tooth structure [6,7], biological compatibility, and low cytotoxicity [8,9]. However, they have disadvantages such as brittleness. Significant improvements have been made since the invention of GIC and further improvements are required in order to enhance the physical properties of these materials. Although stronger and more aesthetic materials with improved handling characteristics are now available, lack of strength and toughness are still major problems.

The strength of conventional GICs is influenced both by the type of glass powder used and the chemical composition of the polyacid applied in order to form the organic matrix [1–9]. Various studies have shown that very close attachment of carboxylic acid groups to the polymeric backbone of PAA resulted in a very rigid matrix, which causes steric hindrance. Subsequently not all the carboxylic acid groups will convert to carboxylate groups and very little polysalt bridge (Ca^{+2} dicarboxylate and Al^{+3} tricarboxylate complexes) formation takes place. In order to overcome this problem recent studies have incorporated different monomers such as *N*-vinylpyrrolidone (NVP) and amino acid derivatives as spacers between the carboxylic acid groups, rendering the polymeric backbone more flexible and allowing greater access for acid/base reaction (as the COOH groups then have greater freedom to react with Al and Ca ions); this results in improved filler–polymer interaction and more complete polysalt bridge formation. This approach was found to enhance the mechanical strength of GICs [10,11,35].

It was found that NVP-modified polyacids yielded dental GICs with enhanced mechanical strength [10,11]. Culbertson reported that acrylic acid–itaconic acid–NVP (AA–IA–NVP) polymers with different molar ratios have the ability to increase the mechanical properties of the GICs [10]. In another study Yamazaki et al. [13] showed that AA–IA–NVP polymer resulted in modified GICs with higher compressive strength (CS) and diametral tensile strength (DTS) than Fuji IX commercial GIC [10–13].

N-Vinylcaprolactam (NVC) is a chemical analogue of NVP. Applications of NVC in the area of biomedical materials, in stabilization of proteases and in controlled drug delivery and drug release have been published previously [14–16]. NVC, which has a ring that consists of seven carbon atoms (one carbon atom more than the NVP ring), can be used in the GIC polyacid structure. It is well known that the cycloheptane ring has different conformational forms, i.e. "chair", "bath" and "twisted bath" [17]. Therefore, it cannot exist in a flat conformation, and hence an NVC-containing GIC will exhibit increased steric hindrance, and consequently improved mechanical properties. The polymerization of NVC has not been studied as systematically as that of NVP. NVC can be polymerized by a radical mechanism with or without solvent [20]. NVC polymers have attracted much attention due to their thermosensitivity and biocompatibility. Due to the fact that hydrolysis of the amide group of NVC will not produce small amide compounds, NVC is suitable for biomedical applications [14–20].

It was envisaged that NVC molecules interspersed between the copolymers of itaconic and acrylic acid would act as a spacer and the degree of steric hindrance would decrease. In addition, the probability of ionic bond formation and subsequent polysalt bridge formation in the final set GIC would be increased significantly. Since there are more carboxylic acid groups available to make ionic bonds with Al^{+3} and Ca^{+2}, there will be more polysalt bridge formation and cross-linkings. Consequently the mechanical properties of the final set cement will be enhanced. Therefore, the presented GIC will be the material of choice for posterior teeth restoration and also as bone graft material in stress-bearing areas.

Therefore, the objective of this study was to synthesize the terpolymer of acrylic acid, itaconic acid and NVC using a free radical polymerization technique and to investigate the effect of incorporation of NVC-containing terpolymers into polyacid GICs on the mechanical and working properties of these materials.

2. Materials and methods

2.1. Materials

The glass powders and polyacid used in the experiments were commercially available Fuji IX (GC International, Tokyo, Japan), acrylic acid (AA), itaconic acid (IA), NVC, 2,2′-azobis (isobutyronitrile) (AIBN), isopropyl alcohol (2-propanol), methanol (CH_3OH) and anhydrous ethyl acetate ($CH_3COOC_2H_5$). All the reagents used in this study were of analytical grade and applied as received from Sigma–Aldrich Chemical Co.

2.2. Polymer synthesis

The experimental procedure employed in this study is a slight modification of methods reported previously [13,21–23]. The details are as follows. Initially 1 wt.% AIBN as the initiator of the polymerization reaction was dissolved in 75 ml of distilled water in a 250 ml three-necked flask. In the next step, 0.4 mol (27.43 ml) AA (density 1.05 g cm^{-3}), 0.05 mol (6.96 g) NVC (density 1.045 g cm^{-3}) and 0.05 mol (6.5 g) IA were measured and dissolved in 37.5 ml of distilled water in a beaker. A third solution was made up, consisting of 1 wt.% AIBN dissolved in 22.5 ml of distilled water in a beaker. The first solution was stirred with a mag-

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