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Eugenol functionalized poly(acrylic acid) derivatives in the formation of glass-ionomer cements

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ABSTRACT

Eugenol possesses analgesic and anti-inflammatory properties with the ability to relieve pain in irritated or diseased tooth pulp, thus, incorporating polymers with eugenol moieties in dental cements is attractive. An acrylic derivative of eugenyl methacrylate (EgMA) was copolymerized with acrylic acid (AA) using a radical initiator, to yield a water soluble copolymer of acrylic acid and eugenyl methacrylate {p(AA-co-EgMA)}, which was then applied in the formulation of glass-ionomer cements for potential application as dental cements. Three concentrations of the p(AA-co-EgMA) copolymer in water were studied by, 30 wt%, 40 wt% and 50 wt%, and used with different powder:liquid ratios to formulate the glass-ionomer cements. The setting kinetics showed that both the concentration of the copolymer and the powder:liquid ratio influenced the working and setting times. Thus, selected formulations were used for further characterization of their mechanical properties, water uptake and fluoride release, to optimize the cement formulation. The experimental glass-ionomer cements exhibited physical and mechanical properties in compliance to ISO standard requirements with the benefit of the initial pH being greater than the commercial formulation used as the standard cement.

Furthermore, the presence of the eugenyl moieties bound to the polymer matrix was advantageous with respect to moisture sensitivity and anti-bacterial properties.

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1. Introduction

Eugenol (4-allyl-2-methoxyphenol) has been used in dental materials since 19th century. It is currently used in combination with zinc oxide (ZOE) as temporary pulp capping agents and as filling materials in root canals, where eugenol functions as an obturation agent producing a soothing effect on the pulp. These cements produce some adverse effects in vivo probably due to the release of unreacted eugenol, which, in some concentrations, can produce tissue irritation and induce inflammatory reactions over the oral mucous mem-

brane. The presence of free eugenol in the cements can also cause a detrimental effect on the physical and mechanical properties of the overlying permanent dental composite resins that cure mainly by free radical polymerization [1–3]. However, literature reports indicate that there are numerous advantages of eugenol such as pharmacological properties, analgesic and anti-inflammatory properties with the ability to relieve pain in irritated or diseased tooth pulp [2] antimicrobial and anti-aggregating function [3], antipyretic activity [4] and anti-anaphylactic properties preventing mast cell degranulation [5]. In addition, eugenol can prevent lipidic peroxidation

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in the initial steps due to the presence of the phenolic group, which can scavenge free radicals [6].

Combining the properties of eugenol molecules with glass-ionomer cements is an interesting concept, especially as a chemically bound entity, namely a methacrylate derivative, which is expected to imbibe anti-inflammatory effects and alter moisture sensitivity of the cement. Eugenyl methacrylate (EgMA) monomer [7] is a suitable molecule for introducing the eugenol activity in the glass ionomer formulations via copolymerization with acrylic acid (AA). Glass ionomer cements (GICs) are self-hardening cements that are formed by the reaction of an ion leachable calcium fluoro aluminosilicate glass powder and polyalkenoic acid. The setting involves neutralization of the acid groups available from the water-soluble polymer and the base, which is a calcium fluoro aluminosilicate glass powder. The setting reaction occurs between the two phases, thus, it is heterogeneous and is sensitive to the particle size of the glass, the concentration and the nature of the poly acid [8]. Conventional glass-ionomers suffer from some disadvantages such as a short working time, and are generally brittle in nature. Glass ionomers cements are also sensitive to moisture in the early stages of placement and can lose matrix-forming ions in presence of excessive moisture or desiccation in case of patients who breathe through the mouth. This work reports the synthesis of a new copolymer derived from acrylic acid and eugenol methacrylate, which can be potentially used in glass ionomer cement formulations with improved properties in comparison with the conventional cements.

2. Materials and methods

2.1. Materials

Acrylic acid was purchased from Sigma–Aldrich and purified by distillation under reduced pressure. The monomer EgMA was synthesized as reported previously [7]. 2,2′ Azobisisobutyronitrile (AIBN) (Merck) was recrystallized from methanol (mp 104 °C). The solvents ethanol (Scharlau) and acetone (SDS) were purified by standard procedures. Fuji IX was purchased from Kent Dental, UK.

2.2. Synthesis and characterization of the copolymer

A mixture of acrylic acid and EgMA with a AA:EgMA feed molar ratio of 80:20 was deoxygenated at room temperature for 15 min. The mixture was copolymerized at 60 °C in ethanol ([M] = 1 mol/l), using AIBN (1 wt% with respect to monomers) as a radical initiator, for 24 h in order to obtain soluble products. Once the reaction time was over, the solvent was removed by flash distillation under reduced pressure and the copolymer was washed with acetone to remove residual monomers. Subsequently, the copolymer was dissolved in a minimum amount of water and lyophilized.

The copolymer was characterized by NMR spectroscopy (Varian XLR-300 spectrometer) and ATR-FTIR spectroscopy (PerkinElmer Spectrum One). The molar fraction of each monomeric unit in the copolymer was determined from the ^1H NMR spectrum by using Eq. (1), where $f(M_1)$ is the molar

fraction of M_1 unit (acrylic acid), I_{M_1} the integration of the signal assigned to the carboxylic proton of the acrylic acid and I_{M_2} is the integration of the signals assigned to aryl and allyl protons of the M_2 unit (EgMA).

$$f(M_1) = \frac{I_{M_1}/N^0H^+}{I_{M_1}/N^0H^+ + I_{M_2}/N^0H^+} \quad (1)$$

The molecular weight of the copolymer was determined by size exclusion chromatography (SEC). A solution was prepared by dissolving an appropriate amount of the polymer in a $\text{CH}_3\text{CN}/\text{KNO}_3$ buffer solution (0.1 M), to give a concentration of 10 mg/ml, stirring it during 2 h to dissolve completely. After that, the solution was then filtered through a 0.45 μm PTFE membrane prior to the measurement. The equipment used for the determination was a GPC Waters 1515 Isocratic HPCL Pump, with a precolumn and two columns Waters Ultrahydrogel™ 500 and 250 maintained at 30 °C using a thermostat, and connected to a refractive index detector, Waters 2414. The solvent used was $\text{CH}_3\text{CN}/\text{KNO}_3$ buffer solution (0.1 M). The flow rate was 0.5 ml/min. Equipment was previously calibrated with monodisperse poly(ethylene oxide) (PEO) samples (Waters).

2.3. Cement preparation

New GICs were prepared using different concentrations of aqueous solutions of the synthesized copolymer as a liquid phase. Solutions with 30% (LP30), 40% (LP40) and 50% by weight (LP50) of the p(AA-co-EgMA) copolymer were prepared in distilled water and allowed to dissolve completely at room temperature. The powder component of the commercial GIC, Fuji IX, was used as the basic glass in all cement formulations. Different powder:liquid ratios were employed and are shown in Table 1. The experimental GICs were prepared by hand mixing of the corresponding liquid phase with the GC Fuji IX powder. The commercial GC Fuji IX GP cement was used as a control formulation. The cement specimens were allowed to set for 24 h and then stored in distilled water for 24 h prior to mechanical testing, fluoride release, water uptake and pH surface measurements. Specimens were also aged for 6 weeks in distilled water at 37 °C for mechanical testing.

Table 1 – Experimental glass ionomer cements formulated in this work

Liquid phase	Solid phase	Solid:liquid ratio
GC Fuji IX GP liquid	GC Fuji IX GP Powder	3:1 ^{a,b}
LP30	GC Fuji IX GP Powder	2:1 ^a
		3:1 ^a
		4:1
LP40	GC Fuji IX GP Powder	2:1
		3:1 ^{a,b}
		2:1 ^{a,b}
LP50	GC Fuji IX GP Powder	3:1
		5:2

^a Used for mechanical properties testing.

^b Used for water uptake and fluoride release.

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