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Influence of a polymerizable eugenol derivative on the antibacterial activity and wettability of a resin composite for intracanal post cementation and core build-up restoration

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ABSTRACT

Objectives. Eugenol has been used in dentistry due to its ability to inhibit the growth of a range of microorganisms, including facultative anaerobes commonly isolated from infected root canals. The aim of this study was to evaluate the antibacterial activity of the experimental composites containing eugenyl methacrylate monomer (EgMA), a polymeric derivative of eugenol, against a range of oral bacteria, commonly associated with failure of coronal and endodontic restorations. *In vitro* composite behavior and wettability were also studied in conjunction with their antibacterial activity.

Methods. EgMA monomer (5 and 10% by weight) was added into BisGMA/TEGDMA resin based formulations with filler mixtures of hydroxyapatite (HA) and zirconium oxide ZrO_2. The antibacterial activity of the experimental composites against *Enterococcus faecalis*, *Streptococcus mutans* and *Propionibacterium acnes* were evaluated by direct contact test and compared with composite formulation without inclusion of EgMA. To clarify the antibacterial mode of action, agar diffusion test (ADT) was also performed. Water sorption, solubility, diffusion coefficient, contact angle and surface free energy as complementary clinically relevant properties were determined.

Results. Water sorption and wettability studies showed reduction of water uptake and surface free energy values with increasing content of EgMA monomer, resulting in significant increase in the hydrophobicity of the composites. No inhibition zones were detected in any of the composites tested against the three bacteria employed as expected, due to the absence of any leachable antibacterial agent. The covalently anchored EgMA monomer with the composite surface exhibited an effective bacteriostatic activity by reducing the number of CFUs of the three species of bacteria tested with no significant dependence on the concentration of EgMA at 5 and 10% by weight. The surface antibacterial activity *R* of the experimental composites were different against the three tested species with values in the range 2.7–6.1 following the order *E. faecalis* < *S. mutans* < *P. acnes*.

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Significance. The incorporation of EgMA monomer within polymerizable formulations provides a novel approach to yield intrinsically antibacterial resin composites for different dental applications.

1. Introduction

Endodontically treated teeth (ETT) are more susceptible to fracture than vital teeth due to the significant reduction of tooth tissue as a result of the endodontic and restorative treatment accompanied with changes in chemical composition of dentin due to loss of water and collagen [1]. Although it is well established that coronal coverage significantly improves the clinical success rate of endodontically treated posterior teeth, the choice of restoration depends on the amount of remaining tooth structure and functional requirements.

Composite resins have the advantage of bonding to residual coronal and root canal dentin, which may assist in strengthening the tooth [2,3] and offer an alternative technique for restoration of ETT [4]. Dual cure resin composites core materials with different viscosity are currently used within the canal for fiber posts cementation, to restore the structurally compromised ETT. They have superior mechanical properties than those of resin cements and result in lower stress, reducing the load transfer on the root dentin and surface of the post [2].

However, resin composites lack antibacterial properties and result in more plaque accumulation than other restorative materials [5]. In addition, any microleakage allows for new bacterial invasion, compounded by the fact that it is difficult to completely remove bacteria from the root canal system even after careful cleaning and shaping and the minimally invasive approach during restoration of teeth will possibly maintain more residual bacteria within the dentinal tubules [6].

There is a rising interest to endow dental restorative materials with sustained antibacterial activity to enhance long term performance [7], which is expected to lower the risk of reinfection [8] and secondary caries [9]. Different antibacterial agents such as chlorhexidine, fluoride, quaternary ammonium salts and metallic agents (silver, gold and zinc) have been incorporated in acrylic based composite formulation in order to achieve this goal [10]. However, most of these additives cause an adverse effect in terms of mechanical properties, discoloration of the material [11], toxicity and short-term antibacterial effectiveness [12].

Most antibacterial studies reported in literature evaluate the activity of different incorporated antibacterial agents against *Streptococcus mutans*, the main microbial etiological agent of dental caries and the leading cause of resin based composite failure [13], however other oral microorganisms such as, *Enterococcus faecalis*, *Candida albicans* and *Propionibacterium acnes* are also frequently associated with endodontic infections [14]. *E. faecalis*, in particular, is difficult to remove owing to its considerable virulence factors constituting a source of recurrent infection after conservative as well as surgical treatments [15]. *P. acnes* is an anaerobic Gram-positive bacterium responsible for a wide range of infections and inflammatory conditions [16]. Therefore, development of antibacterial restorative filling materials to be reliable for a variety of dental applications need a potent antimicrobial agent which acts against a wide range of oral microorganisms.

Eugenol (4-allyl-2-methoxyphenol) is a natural phenolic anti-oxidant essential oil that possesses antifungal activity [17] and inhibits the growth of several microorganisms including *Escherichia coli* [18] and facultative anaerobes commonly isolated from infected root canals [19]. This compound has been used in combination with zinc oxide in different dental applications such as temporary filling materials and root canal sealers. However, eugenol is not compatible with other methacrylate based restorative materials because of the presence of free eugenol, which interferes with the polymerization reaction of dental composite resins.

In contrast, eugenyl methacrylate (EgMA) an eugenol derivative [20] possess in its chemical structure a polymerizable methacrylic group (Fig. 1) that allows the monomer to participate in free radical polymerization reactions whilst maintaining the antibacterial activity of its natural precursor against different Gram-negative and Gram-positive bacterial species [21].

In our previous study, the experimental composites from Bis-GMA/TEGDMA, a commonly used dental resin system and EgMA were formulated with 65% by weight filler phase comprising of HA/ZrO_2 [22]. These composites were tailored to function as an antibacterial restorative material for intracanal

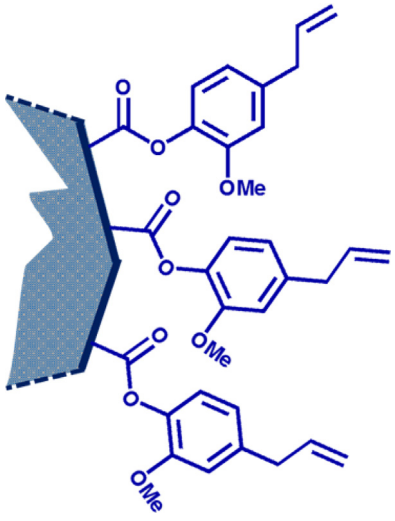

Fig. 1 – Proposed example of pendent eugenyl residues (eugenyl methacrylate antibacterial monomer) from the polymer network on the surface of the experimental composite.