DENTAL MATERIALS XXX (2018) XXX–XXX

Available online at www.sciencedirect.com

ScienceDirect

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Novel magnetic nanoparticle-containing adhesive with greater dentin bond strength and antibacterial and remineralizing capabilities

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ARTICLE INFO

Article history:
Received 17 February 2018
Received in revised form
22 May 2018
Accepted 1 June 2018
Available online xxx

Keywords:
Magnetic nanoparticles
Magnetic force

ABSTRACT

Objectives. A nanoparticle-doped adhesive that can be controlled with magnetic forces was recently developed to deliver drugs to the pulp and improve adhesive penetration into dentin. However, it did not have bactericidal and remineralization abilities. The objectives of this study were to: (1) develop a magnetic nanoparticle-containing adhesive with dimethylaminohexadecyl methacrylate (DMAHDM), amorphous calcium phosphate nanoparticles (NACP) and magnetic nanoparticles (MNP); and (2) investigate the effects on dentin bond strength, calcium (Ca) and phosphate (P) ion release and anti-biofilm properties.

Methods. MNP, DMAHDM and NACP were mixed into Scotchbond SBMP at 2%, 5% and 20% by mass, respectively. Two types of magnetic nanoparticles were used: acrylate-functionalized iron nanoparticles (AINPs); and iron oxide nanoparticles (IONPs). Each type was added into

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https://doi.org/10.1016/j.dental.2018.06.001

Please cite this article in press as: Li Y, et al. Novel magnetic nanoparticle-containing adhesive with greater dentin bond strength and antibacterial and remineralizing capabilities. Dent Mater (2018), https://doi.org/10.1016/j.dental.2018.06.001

Dental adhesive
Dentin bond strength
Anti-biofilm
Remineralization

the resin at 1% by mass. Dentin bonding was performed with a magnetic force application for 3 min, provided by a commercial cube-shaped magnet. Dentin shear bond strengths were measured. *Streptococcus mutans* biofilms were grown on resins, and metabolic activity, lactic acid and colony-forming units (CFU) were determined. Ca and P ion concentrations in, and pH of biofilm culture medium were measured.

Results. Magnetic nanoparticle-containing adhesive using magnetic force increased the dentin shear bond strength by 59% over SBMP Control ($p < 0.05$). Adding DMAHDM and NACP did not adversely affect the dentin bond strength ($p > 0.05$). The adhesive with MNP + DMAHDM + NACP reduced the *S. mutans* biofilm CFU by 4 logs. For the adhesive with NACP, the biofilm medium became a Ca and P ion reservoir. The biofilm culture medium of the magnetic nanoparticle-containing adhesive with NACP had a safe pH of 6.9, while the biofilm medium of commercial adhesive had a cariogenic pH of 4.5.

Significance. Magnetic nanoparticle-containing adhesive with DMAHDM and NACP under a magnetic force yielded much greater dentin bond strength than commercial control. The novel adhesive reduced biofilm CFU by 4 logs and increased the biofilm pH from a cariogenic pH 4.5–6.9, and therefore is promising to enhance the resin–tooth bond, strengthen tooth structures, and suppress secondary caries at the restoration margins.

1. Introduction

Dental adhesives and resin composites are the first choice for tooth defect restorations due to their excellent esthetics and direct-filling capabilities [1–4]. Unfortunately, composite restorations have a relatively higher failure rate with an average replacement time of only 5.7 years [5,6]. Secondary caries at the tooth-restoration interface has been suggested in previous studies as one of the primary reasons for restoration failures [7,8]. Despite great improvements in dental adhesives, the dentin–resin interface is still the weakest area of the composite restorations due to dentinal bond degradation [9–13]. Microleakage and gap formation in this weakened interface provide an effective pathway for invasion of oral plaque biofilms and the development of secondary caries around the tooth-restoration margins. Therefore, improving the bond durability and preventing bacterial invasion are pivotal issues for inhibiting secondary caries and increasing the restoration longevity [11,14,15].

Dentin bonding relies on a micromechanical interlocking mechanism that involves the infiltration and subsequent entanglement of adhesive resin into the dentin collagen matrix to form the hybrid layer [16]. A major concern with contemporary adhesives is their limited ability to infiltrate into the collagen fibril network of the demineralized dentin exposed by acid-etching or self-etch processes [14,15]. Incomplete infiltration of resin into the demineralized dentin and hydrolysis of the polymerized resin result in the exposure of the denuded dentin collagen matrix along the dentin–resin interface. Acids and enzymes produced by bacteria, as well as activated host-derived proteases, further deteriorate this defective bonded interface, thereby compromising the longevity of the resin–dentin bond [17,18]. Several methods were developed to facilitate the optimal infiltration of resin into the collagen matrix of the demineralized dentin, including the use of hydrophilic resin monomers, catalysts, various solvents, and the ethanol wet-bonding technique [19–23]. The use of hydrophilic monomers such as 2-hydroxyethyl methacrylate (HEMA) can improve the resin wettability, promote the re-expansion of the dried dentin collagen, help displace water in the bonded interface, and facilitate the subsequent resin infiltration [24]. However, adhesives rich with hydrophilic monomers exhibit greater water sorption from the host dentin and are consequently susceptible to hydrolytic degradation [25–27]. This shortcoming was described by many studies showing increased interfacial nanoleakage and lower long-term bonding effectiveness [28,29]. In addition, adhesive resin polymerization in such a moist environment is a challenge. The compromise in the degree of conversion of adhesive monomers may allow water permeation along the bonded interface, which in turn would result in the formation of a porous hybrid structure with reduced sealing ability [30]. Furthermore, incomplete polymerization of adhesive monomers may also reduce the mechanical properties, which may shorten the durability of the adhesive restoration [31,32]. The ethanol wet-bonding technique is based on the theory that ethanol dehydration renders the acid-etched dentin less hydrophilic and maintains the dehydrated collagen matrix in an extended state to facilitate the relatively hydrophobic monomers to infiltrate by water replacement from the interfibrillar and intrafibrillar spaces [20]. Although better stability of the resin–dentin bonds has been created, ethanol wet-bonding is highly technique sensitive and difficult for clinical applications.

To solve the limited resin penetration problem, designing a novel adhesive to be forced to infiltrate and penetrate the interfibrillar spaces in the acid-etched dentin would be highly desirable. Unlike self-diffusion which is a passive process, magnetic forces can potentially enhance the adhesive penetration. Under the guidance of an external magnetic force, magnetic nanoparticles (MNP) can transport more drugs to a target than either diffusion or iontophoresis [33,34]. However,

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