DENTAL MATERIALS XXX (2018) XXX–XXX

Available online at www.sciencedirect.com

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Fluoride containing bioactive glass composite for orthodontic adhesives — Apatite formation properties

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ARTICLE INFO

Article history:
Received 10 November 2017
Received in revised form
14 March 2018
Accepted 27 April 2018
Available online xxx

Keywords:
Bioactive glass
Fluoride
Apatite
White spot lesions
Orthodontic adhesives

ABSTRACT

Objectives. Dental materials that can form apatite offer the potential to not only prevent demineralisation but enhance remineralisation of the enamel. The objective of this study was to investigate the ability of a novel BAG-resin adhesive to form apatite in 3 immersion media.

Methods. A novel fluoride containing BAG-resin adhesive described previously, with 80% by weight filler load, was used to fabricate 90 disks. Each disk was immersed in 10 ml of either tris buffer (TB), or artificial saliva at pH = 7 (AS7) or pH = 4 (AS4). At ten time points (from 6 h to 6 months), three disks were taken from each of the solutions and investigated by ATR-FTIR, XRD and SEM.

Results. The BAG-resin formed apatite on the disk surface, which increased with time, especially in AS4 and AS7. The apatite crystals formed in AS7 were highly oreintated and the oreintation increased with time.

Significance. This novel BAG-resin adhesive differs from the currently used adhesives by promting apatite formation, particularly under acidic conditions. Thus, applied in the clinical situation to bond orthodontic brackets, it may discourage the frequent occurrence of white spot lesion formation around the brackets.

1. Introduction

Bioactive glasses (BAGs) were first used in dentistry for the treatment of bony defects [1]. This material was further developed as a commercial dentifrice to treat teeth with dentine hypersensitivity and enamel demineralisation [2]. The reduction of dentine hypersensitivity occurs by occluding exposed dentinal tubules with apatite formation using a calcium-sodium-phospho-silicate glass. Remineralisation of small enamel defects caused by acid erosion or caries is enhanced through the release of calcium and phosphate ions [3,4]. More recent research has focused on incorporating BAGs in dental composites and adhesives [5] to prevent and/or

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https://doi.org/10.1016/j.dental.2018.04.009

treat demineralisation of teeth through beneficial long term ion release and their acid neutralising characteristics [6,7]. Depending upon the glass composition, restorative materials containing BAG can release locally, and more continually, a range of therapeutic ions such as Ca^{2+}, Sr^{2+}, PO_4^{3-} and F^- at the site of demand, rather than less specifically through toothpaste or remineralising gel.

The mechanism of enamel demineralisation and remineralisation has been extensively studied over several decades. Hydroxyapatite crystals in enamel are in quasi dynamic equilibrium with the aqueous phases of saliva and plaque fluids [8]. The amount of dissolution of enamel is directly related to both the pH and the concentration of calcium and phosphate ions in the solution [2,9]. On the other hand the rate of apatite formation of the BAG has been shown to increase dramatically upon increasing the phosphate content, whilst maintaining the network connectivity (NC) at a low value [10,11]. Another factor that increases the rate of apatite formation is the addition of a small amount of fluoride, whilst again maintaining the NC [11–14]. The composition of the immersion solution can also affect the apatite formation process depending on the presence or absence of ions necessary for apatite crystal formation and on the pH of the solution. In our previous study [15], we manufactured and tested a novel fluoride containing BAG resin adhesive for its potential on ion release and acid neutralising effect. It was found that F^-, Ca^{+2}, PO_4^{-3} were released substantially over a six-month period and the release was higher and faster in acidic media at pH4 compared to pH7. The BAG resin was also shown to have a long term neutralising effect [15].

Very few studies exist evaluating the ability of the BAG adhesives or composites to form apatite, particularly with regards to the effect of the immersion media on apatite formation. The aim of the present study was to investigate the potential of the previously reported BAG resin adhesive [15] to form apatite in three different solutions: artificial saliva at pH4 (AS4), and pH7 (AS7), and tris buffer (TB) at pH7.35, simulating the extreme range of conditions present in the mouth. The characterisation of the apatite would also be investigated.

2. Materials and methods

The detailed composition of the novel BAG resin adhesive was described in the previous study [15]. The BAG was composed of 35.25% SiO_2, 6% Na_2O, 43% CaO, 5.75% P_2O_5, and 10% CaF_2 and was prepared via the melt quench technique. The resin was composed of 42.25% BisEMA, 55% TEGDMA, 0.25% DMAEM, 0.5% camphorquinone and 2% 4-Meta.

2.1. Preparation of the BAG-resin disks

Ninety disks were prepared using Teflon moulds measuring 10 mm in diameter and 1.2 mm in thickness. The BAG-resin:weight ratio was 80:20%. The disks were divided into three groups (n = 30) to be immersed individually in three types of solutions. Each disk was immersed in a 15 ml polypropylene centrifuge tube (Fisher Scientific UK Ltd, Leicestershire, UK) containing 10 ml of the solution.

2.2. Preparation of the immersion media

2.2.1. Tris buffer (TB)

The preparation of TB solution was by dissolving 15.09 g tris-(hydroxymethyl)aminomethane (Sigma-Aldrich) in 800 ml deionized water, adding 44.2 ml 1 M hydrochloric acid (Sigma-Aldrich) and overnight heating to 37 °C. The pH was adjusted to 7.3 the next day, using 1 M hydrochloric acid and a pH meter (Oakton Instruments, Nijkerk, the Netherlands). Deionized water was then added to make up a total volume of 2.0 l. The solution was kept at 37 °C [11].

2.2.2. Artificial saliva

Artificial saliva (demineralising and remineralising buffers) were prepared according to Ten Cate et al. [16]:

1. Demineralising buffer (AS4): 2.0 l AS4 was prepared by dissolving 0.4411 g of $CaCl_2{\cdot}2H_2O$, 0.245 g KH_2PO_4 (all Sigma-Aldrich) in 800 ml of deionized water, and adding 5.72 ml acetic acid (Sigma-Aldrich). The pH was adjusted to 4, by adding 0.5 M KOH (Sigma-Aldrich), and deionized water was then added to make up a total volume of 2.0. The solution was stored in a fridge at 2 °C.
2. Remineralising buffer (AS7): 2.0 l AS7 was prepared by dissolving 0.4411 g $CaCl_2{\cdot}2H_2O$, 0.245 g KH_2PO_4, 9.532 Hepes and 19.386 g KCL (all Sigma-Aldrich) in 800 ml of deionized water. The pH was adjusted to 7, by adding 0.5 M KOH (Sigma-Aldrich) and deionized water was then added to make up a total volume of 2.0 l. The solution was stored in a fridge at 2 °C.

2.3. Investigations for apatite formation

For the characterisation, the samples were investigated by the following techniques

(1) Attenuated total reflection-Fourier transform infrared spectroscopy (ATR-FTIR) using a Spectrum GX (Perkin-Elmer, Waltham, MA, USA) — The disk was pressed against the ATIR-FTIR lens to attain maximum signal intensity. Data collected from 1800 to 500 cm^{-1} in absorbance mode.

(2) X-ray diffraction (XRD) using an X'pert Pro Diffractometer (Panalytical, Netherlands) with Cu-Kα alpha radiation. The samples were analysed using a 2θ range of 3–70°, with a step size of 0.03 and a step time of 200 s.

(3) Scanning electron microscope (SEM) — The immersed disk was halved and one half was imbedded in a cold cure acrylic resin in a Teflon mould of 10.2 mm diameter and 5 mm depth. The embedded disk was polished at the fracture surface using a sequence of silicon carbide grinding papers (P300, P1000 and P4000 respectively) in a Kent 4 Automatic Lapping and Polishing Unit (Kemet International Ltd, Maidestone UK). The samples were attached to SEM stubs and carbon coated to minimize charging and improve the imaging resolution.

2.3.1. Bioactivity of BAG — 24 h study

In order to investigate the bioactivity and its potential of the BAG, an initial 24 h study was carried out. A sample of the BAG powder (75 mg) was immersed in 50 ml of TB in a 250 ml polypropylene container and one BAG-resin disk was immersed in 10 ml of TB in a 15 ml polypropylene centrifuge

Please cite this article in press as: Al-eesa NA, et al. Fluoride containing bioactive glass composite for orthodontic adhesives — Apatite formation properties. Dent Mater (2018), https://doi.org/10.1016/j.dental.2018.04.009