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The influence of particle size and fluorine content of aluminosilicate glass on the glass ionomer cement properties

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ABSTRACT

Objective. Glass ionomer cements (GIC) are clinically accepted dental restorative materials mainly due to their direct chemical adhesion to both enamel and dentin and their ability to release fluoride. However, their mechanical properties are inferior compared to those of amalgam and composite. The aim of this study is to investigate if combinations of nano- and macrogranular glass with different compositions in a glass ionomer cement can improve the mechanical and physical properties.

Methods. Glasses with the composition 4.5 SiO_2–3 Al_2O_3–1.5 P_2O_5–(5 – x) CaO – x CaF_2 (x = 0 and x = 2) were prepared. Of each type of glass, particles with a median size of about 0.73 μm and 6.02 μm were made.

Results. The results show that the setting time of GIC decreases when macrogranular glass particles are replaced by nanogranular glass particles, whereas the compressive strength and Young's modulus, measured after 24 h setting, increase. The effects are more pronounced when the nanogranular glass particles contain fluoride. After thermocycling, compressive strength decreases for nearly all formulations, the effect being most pronounced for cements containing nanogranular glass particles. Hence, the strength of the GIC seems mainly determined by the macrogranular glass particles.

Cumulative F^--release decreases when the macrogranular glass particles with fluoride are replaced by nanogranular glass particles with(out) fluoride.

Significance. The present study thus shows that replacing macro- by nanogranular glass particles with different compositions can lead to cements with approximately the same physical properties (e.g. setting time, consistency), but with different physicochemical (e.g. F^--release, water-uptake) and initial mechanical properties. On the long term, the mechanical properties are mainly determined by the macrogranular glass particles.

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1. Introduction

Glass ionomer cements (GIC), also known as polyalkenoate cements, were discovered in the late 70s and are known for their good esthetical properties, their anti-cariogenic potential and their biocompatibility. Moreover, they have the unique ability to bind to wet tooth tissue. This prevents microleakage and increases the durability of a restoration. However, GIC are highly sensitive to moisture during initial hardening and have poor mechanical properties compared to other restorative materials, such as resin-modified GIC and composites [1,2]. For these reasons they are predominantly used as luting and base cements and as filling materials in low load-bearing dental restorations.

Conventional GIC set by an acid-base reaction between a polyalkenoic acid and an aluminosilicate glass powder [3]. The aluminosilicate glass powder normally contains Al, Si, and Ca. The ratio of these components determines whether the glass will form a cement. Si^{4+} and Al^{3+} are important compounds as they form a network of tetrahedrae with oxygen bridges [3–5]. In this network, Al^{3+} induces negative charges. These charges are compensated by Ca^{2+}, which can also make the glass more basic by its network modifying capability [3,4]. When mixed with polyalkenoic acid, the glass degrades, so that Ca^{2+} and Al^{3+}-ions are set free. These ions form complexes with the acid, so that a firm gel is formed. In a later stage, the silica ions eluted from the glass condensate and form a silica gel matrix [3,6].

The chemistry and formulation of the basic glass and the polyalkenoic acid both affect the setting reaction and the properties of the GIC. An increase of the molecular weight of the polyalkenoic acid results in improved mechanical properties, but reduces the handling properties [7]. Lyophilized polyalkenoic acid was introduced, as an aqueous solution is unstable and becomes viscous in time [1]. Photopolymerizable resin modified glass ionomer cements (RM-GIC) were developed by adding hydroxyethylmethacrylate (HEMA) and photo-initiators to a modified polyalkenoic acid. These cements are stronger and have better handling characteristics, but are still prone to water-uptake [2,8].

Over the years, many researchers have focused on the optimization of the composition of the glass to improve the physical and mechanical properties of the cement [5,6,9,10]. Griffin and Hill have incorporated fluoride and phosphate in the calcium aluminosilicate glass and investigated the effect on the setting and mechanical properties of the GIC [11,12]. F^- was originally added to the glass mixture as a flux to decrease the melting temperature [10]. It was found that fluoride disrupts the glass-network by forming Al–F–Ca(*n*) and F–Ca(*n*) species. In glasses with high fluoride content, additional Si–F–Ca(*n*) species are formed. The incorporation of calcium into these species results in a reduction of the number of available cations to charge balance non bridging oxygens [13]. This makes the glass more reactive and in this way it improves the mechanical properties of the GIC [4,9,10,14]. Furthermore, as the polyalkenoic acid attacks the glass during setting, F^- leaches from the glass and a reservoir of F^- is formed within the matrix, which leads to a long-term release of F^- [15]. F^- has been shown to have an anticariogenic effect due to the inhibition of the formation of bacterial plaques and due to the formation of fluorapatite, which is more resistant to acid dissolution than hydroxyapatite [16]. Phosphor, on the other hand, can integrate in the tetrahedral network, which might create extra places for the acid to degrade the glass. However, phosphor also balances the charge deficit caused by aluminum and leads to a less reactive glass. During setting, cations can also bind to PO_4^{3-}, so that less cations are available to react with the acid groups. This consequently leads to weaker GIC with longer setting times [5].

Apart from the chemical composition of the glass and the polyalkenoic acid, the contact area between these components also controls the setting and mechanical properties of GICs. So, powder/liquid (P/L) ratio as well as particle size distribution are determining parameters for the setting rate and mechanical strengths of the set cement. Increasing the P/L ratio increases the setting rate and mechanical strength of GICs [17]. Smaller particles reduce the setting time [12], and improve wear resistance, surface hardness and compressive strength [18,19]. However, viscosity increases, which impedes the workability [11]. This is a result of the higher surface area that is available for the acid to react with the glass [12]. On the other hand, a combination of small particles with the original glass particles of the same composition results in GIC with better mechanical and handling properties [11,19].

Although several studies have demonstrated that the properties of GICs can be optimized by changing the composition or the particle size and distribution of the glass, the combined effect of such changes has not yet been explored.

The purpose of this study is two-fold. First, the effect of the particle size of the glass on the physical and mechanical properties of the GIC is evaluated. Secondly, as the properties of a GIC are also determined by the composition of the glass, the effect of combining nano- and macrogranular particles of glasses with different compositions on the physical and mechanical properties of the GIC is investigated. The hypothesis is that mixing nano- and macrogranular particles of glasses with different compositions can result in GIC with optimal physicochemical and mechanical properties.

2. Materials and methods

For all solutions, demineralized water was used (Milli-Q system, Millipore, Bedford, MA, USA).

2.1. Synthesis of the glass powder

Glass with the composition 4.5 SiO_2–3 Al_2O_3–1.5 P_2O_5–$(5-x)$ $CaO-x$ CaF_2 was prepared, as described by Griffin and Hill, by mixing appropriate amounts of SiO_2 (Merck 7536), Al_2O_3 (Merck 1095), $CaCO_3$ (Merck 2066), P_2O_5 (Fluka 79612) and CaF_2 (Mallinckrodt (AR) 4168) [6,10]. A total batch size of 100 g was made. Glass without ($x=0$) and with ($x=2$) fluoride was synthesized.

The powder mixture of glass without fluoride and glass with fluoride was heated respectively at 1475 °C and 1450 °C in alumina crucibles in a furnace (HTF17, Carbolite, Hope valley, UK). After homogenization, the glass melt was poured in demineralized water to produce a glass frit [6,10].