SUPPORTING INFORMATION

A Chemical approach to optimizing bioactive glass dental composites

Savinda Aponso[1], Jyothir Ganesh Ummadi[1], Harry Davis[2], Jack Ferracane[2], Dipankar Koley[1,*]

[1]Department of Chemistry, Oregon State University, Corvallis, OR 97331, USA

[2]Department of Restorative Dentistry, Oregon Health & Science University, Portland, OR USA

*Corresponding Author: Dipankar.Koley@oregonstate.edu

Materials: The following materials were purchased: ETH 129 (a Ca^{2+} ionophore) and high molecular weight (~48000) polyvinyl chloride (PVC) from Sigma Aldrich (St. Louis, MO, USA); Trypticase peptone and Brain heart infusion (BHI) from BD Bioscience (Franklin Lakes, NJ, USA); protease peptone from Hardy Diagnostics (Santa Maria, CA, USA) ; carboxymethylcellulose sodium salt, aniline (99%), and 1-nitro-2-(n-octyloxy) benzene (NPOE) from Alfa Aesar (Haverhill, MA, USA); bis(2-ethylhexyl) sebacate and potassium tetrakis(4-chlorophenyl) borate from TCI America (Portland, OR, USA); a 25-µm Pt wire from Goodfellow (Coraopolis, PA, USA); tetramethyl silane from CIL; and sucrose from J.T. Baker. Vulcan carbon powder was a kind gift from the Cabot Corporation (Boston, MA, USA).

Artificial saliva (0.7 mM $CaCl_2$, 0.427 mM $MgCl_2$, 2.40 mM NaH_2PO_4, 2.39 mM Na_2HPO_4, 30 mM KCl, 1.19 mM $NaHCO_3$) and basal medium mucin (BMM) solution (10 g/L protease peptone, 5 g/L BBL trypticase peptone, 5 g/L yeast extract, 2.5 g/L KCl, 8 g/L carboxymethylcellulose) were freshly prepared before each experiment (Ionta et al. 2014). All solutions were prepared with 18 MΩ DI water (Elga Water Systems). *Streptococcus mutans* (UA159) was a kind donation from Jens Kreth (Oregon Health & Science University) (Ajdic et al. 2002).

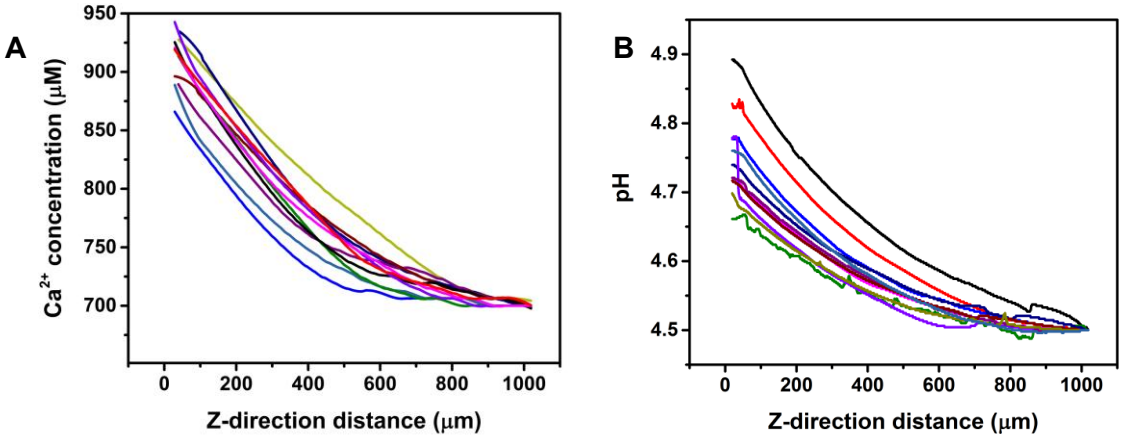
Preparation of bioactive glass

To formulate the bioactive glass, stoichiometric quantities of $Ca(OEtOMe)_2$ (calcium methoxide) $OP(OEt)_3$ (triethyl phosphate), and $Si(OEt)_4$ (tetraethyl orthosilicate) were combined under nitrogen gas to produce a homogeneous solution, after which it was incubated (37°C/100% humidity). A monolithic structure formed after about 7 d, to which deionized water was added to insure complete hydrolysis. After another 3 d, the H_2O and excess MeOEtOH were decanted and the gel was rinsed with 100% EtOH. The gel was then heated to 600 °C in a muffle furnace (Thermo Scientific, series 650–750) producing a transparent colorless glass in small pieces. Later a micronizer (Sturtevant, Hanover, MA, USA) was used to grind the BAG particles to

micrometer and sub-micrometer sizes within a grinding chamber using compressed air and particle-particle impact.

Range	Estimated avg. d_p (µm)	V_f	D_s (µm)
125-150	137.5	0.32	187.7
75-90	82.5	0.32	116.9
38-45	41.5	0.32	58.8
<38	30	0.32	42.5
<5	3	0.32	4.3

Appendix Table: Range of particle sizes from sieving (and micronizing), estimated average BAG particle size (d_p), particle volume fraction (V_f) incorporated into the resin, and estimated interparticle spacing (D_s) for the twelve different composite formulations.



Appendix Figure: Series of representative z-direction scans on a BAG-resin composite (<5 μm) in presence of artificial saliva at pH 4.5. (A) Ca^{2+} concentration scans and (B) pH scans. Ca^{2+} and pH microsensors were used for the respective scans.

Reference:

Ajdic D, McShan WM, McLaughlin RE, Savic G, Chang J, Carson MB, Primeaux C, Tian R, Kenton S, Jia H, Lin S, Qian Y, Li S, Zhu H, Najar F, Lai H, White J, Roe BA, Ferretti JJ. 2002. Genome sequence of Streptococcus mutans UA159, a cariogenic dental pathogen. *Proceedings of the National Academy of Sciences*. *99*(22):14434–14439.

Ionta FQ, Mendonça FL, de Oliveira GC, de Alencar CRB, Honório HM, Magalhães AC, Rios D. 2014. In vitro assessment of artificial saliva formulations on initial enamel erosion remineralization. *Journal of Dentistry*. *42*(2):175–179.