08 Nov 2013

Relating PH and Ion Release from Ga2O3-Na 2O-CaO-ZnO-SiO2 Bioactive Glasses

T. J. Keenan

A. W. Wren

A. Coughlan

Mark R. Towler
Missouri University of Science and Technology, mtowler@mst.edu

et. al. For a complete list of authors, see https://scholarsmine.mst.edu/che_bioeng_facwork/1139

Recommended Citation

T. J. Keenan et al., "Relating PH and Ion Release from Ga2O3-Na 2O-CaO-ZnO-SiO2 Bioactive Glasses," *Proceedings of the IEEE Annual Northeast Bioengineering Conference, NEBEC*, pp. 98 - 99, article no. 6574376, Institute of Electrical and Electronics Engineers, Nov 2013.
The definitive version is available at https://doi.org/10.1109/NEBEC.2013.133

Relating pH and Ion Release from Ga_2O_3-Na_2O-CaO-ZnO-SiO_2 Bioactive Glasses

T.J. Keenan, A.W. Wren, A. Coughlan, M.R. Towler, M.M. Hall
Kazuo Inamori School of Engineering
Alfred University
Alfred, NY 14802
e-mail: tjk2@alfred.edu

***Abstract*—Three glasses were designed for this study, including one Ga-free glass (*Control*), and two Ga-containing glasses (*TGa-1, TGa-2*). In the Ga-containing glasses, Ga_2O_3 is included at the expense of ZnO. This study focuses on the relation between pH and ion concentration present in solution in which these bioactive glasses have been submerged for periods of 1, 7, and 14 days.**

Keywords-bioactive; glass; gallium; pH

I. Introduction

Bioactive glasses can contain ions that are analogous to those found in bones and tissue fluids, and upon implantation, these therapeutic ions leach from the glass, resulting in the formation of a chemical bond between the material and the surrounding tissue. This study focuses on the relation between the pH and the ion concentration present in aqueous solutions in which there has been submerged, a series of bioactive glasses that has had its zinc (Zn) content incrementally replaced by gallium (Ga). Ga is included in these glasses because it has exhibited anti-bacterial properties [1], and anti-cancer abilities [2]. This study was conducted to evaluate the ability of these glasses to release ions upon submersion in aqueous solution, as well as to evaluate the pH of the solutions. It is important for these glasses to possess the ability to release ions without causing significant changes in pH in the local environment as this can affect bioactivity, prohibit further ion release, and potentially damage host cells [3].

II. Experimental Methods

A. Synthesis

Three glasses were designed for this study including one Ga-free glass (*Control*), and two Ga-containing glasses (*TGa-1, TGa-2*), in which Ga_2O_3 is included at the expense of ZnO. Glass compositions can be found in (Table I).

TABLE I

Glass Compositions (mol fraction)

	Control	*TGa-1*	*TGa-2*
SiO_2	0.42	0.42	0.42
Ga_2O_3	0.00	0.08	0.16
ZnO	0.40	0.32	0.24
Na_2O	0.10	0.10	0.10
CaO	0.08	0.08	0.08

B. Sample Preparation

To obtain glass frit samples, the powdered mixes of analytical grade reagents (Sigma-Aldrich, Dublin, Ireland) were fired (1500°C, 1h) in platinum crucibles and shock quenched into water. The resulting frits were dried, ground, and sieved to obtain glass powders with a maximum particle size of 90 μm.

C. Surface Area Determination, Ion Release, and pH Measurements

Gas adsorption analysis was utilized along with the Brunauer-Emmett-Teller (BET) method to determine the surface area of each powder, in order to ensure equivalent surface areas for each sample being submerged in aqueous solution. Each powder sample (n=3) was then submerged in ultra-pure water and placed in an incubator for 1, 7, or 14 days. Upon completion of their respective incubation times, ion release studies were performed using Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES) on a Perkin-Elmer 5300UV (Perkin Elmer, Ma, USA). pH measurements were then conducted on these solutions.

III. Results

Gas adsorption analysis was used in combination with the BET method to determine the surface area of each glass powder.

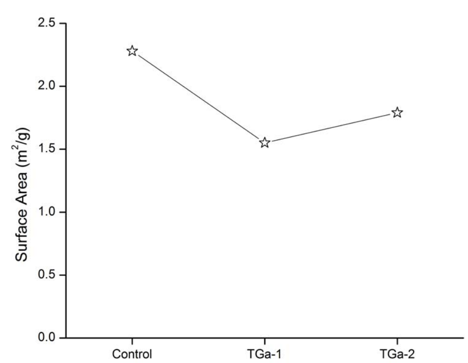

Figure 1. Surface area determination for each glass.

The surface areas of the Control, TGa-1, and TGa-2 glass powders were calculated to be 2.28, 1.55, and 1.79 (m^2/g), respectively.

978-0-7695-4964-4/13 $26.00 © 2013 IEEE
DOI 10.1109/NEBEC.2013.133

Ion release concentrations were calculated after 1, 7, and 14 days. Average release concentrations for each ion are displayed.

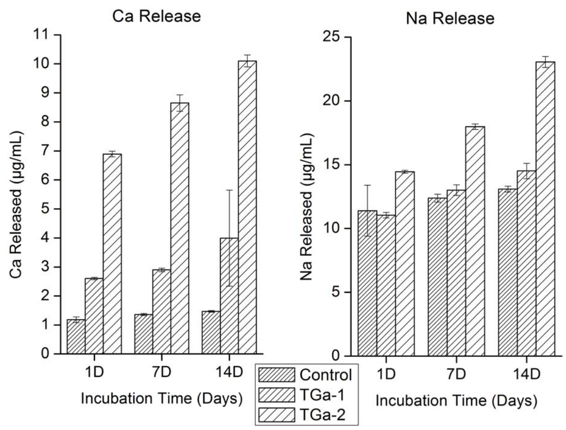

Figure 2. Ca and Na release from all three glasses over 14 day study.

This figure shows that the highest Ca release was observed in the TGa-2 glass, releasing an average total of 6.9, 8.7, and 10.1(µg/mL) after 1, 7, and 14 days respectively. It also shows that the highest Na release was also observed in the TGa-2 glass, releasing an average total of 14.4, 18.0, and 23.0 (µg/mL) after 1, 7, and 14 days, respectively.

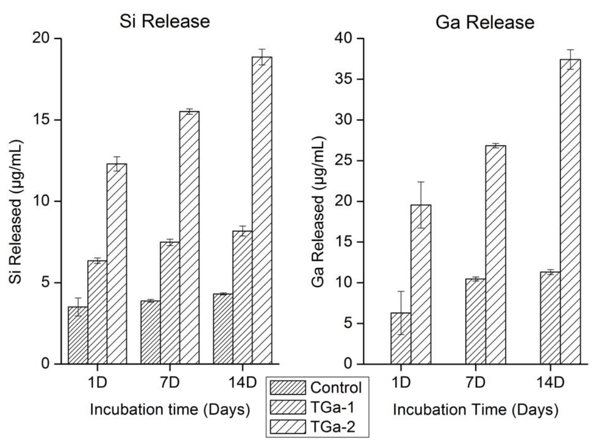

Figure 3. Si and Ga release from all three glasses over 14 day study.

This figure shows that the highest Si release was observed in the TGa-2 glass, releasing an average total of 12.3, 15.5, and 18.9 (µg/mL) after 1, 7, and 14 days, respectively. It also shows that the highest Ga release was also observed in the TGa-2 glass, releasing an average total of 19.5, 26.8, and 37.4 (µg/mL) after 1, 7, and 14 days, respectively.

The ion release studies performed on these glasses produced several observable trends. The first trend observed was an increase in total ion release as submersion time increased, although no Zn was released from any of the samples. The second trend was that the majority of the release of each ion occurred in the first 24 hours of the samples being submerged in water. Another trend observed was as the Ga-content of the glasses increased, so did the Ga-release into solution. The final trend observed was that the total amount of each ion released after the full 14 day period was highest in the TGa-2 samples, then second highest in the TGa-1 samples, and lowest in the Control samples. These last two trends were unexpected as prior studies had suggested that Ga was acting as a glass network former in this series, which should translate to increased Ga-content producing lower concentrations of ion release.

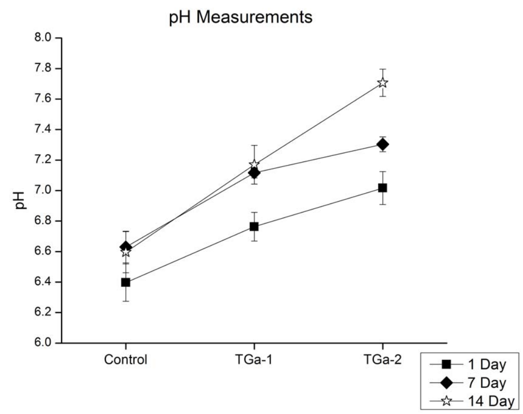

Figure 4. pH measurements of solutions over 14 day study.

The pH measurements performed on the aqueous solutions in which these glasses had been submerged exhibited an increase in the pH of the solution as submersion time increased. This was expected as longer submersion time should translate to more cations leaching from the glasses, resulting in a higher pH. It was also observed that as Ga content increased, the pH of the solution increased, with the solution in which TGa-2 was submerged increasing from an average pH of 7.02, to 7.30, to 7.71 after 1, 7, and 14 days, respectively. This correlates with the Ga ion release studies which exhibited that increased Ga-content results in increased Ga-release.

IV. Conclusion

From this study it can be concluded that other than Zn, all of the other cations present in these glasses leach out upon submersion in an aqueous environment, and that longer submersion time will correlate to larger concentrations of ions being released. This study also suggests that longer submersion times translate to higher levels of ion release and therefore, higher pH levels. The pH of these solutions remain near neutral, suggesting the dissolution products of these glasses will not negatively affect the host cells upon implantation.

References

[1] O. Olakanmi, B.E. Britigan, and L.S. Schlesinger, "Gallium Disrupts Iron Metabolism of Mycobacteria Residing Within Human Macrophages", *Infection and Immunity*, vol. 68, pp. 5619-5627, 2000.

[2] M.M. Hart and R.H. Adamson, "Antitumor Activity and Toxicity of Salts of Inorganic Group IIIa Metals: Aluminum, Gallium, Indium, and Thallium", *Proceedings of the National Academy of Sciences of the United States of America*, vol. 68, pp. 1623-1626, 1971.

[3] T.R. Arnett and D.W. Dempster, "Effect of pH on Bone Resorption by Rat Osteoclasts *in Vitro*", *Endocrinology*, vol. 119, pp. 119-124, 1986.