Compositional optimization for molding of bioactive glasses in the SiO_2-Na_2O-CaO-P_2O_5 system

Otimização da composição para moldagem de vidros bioativos no sistema SiO2-Na2O-CaO-P2O5

DOI:10.34115/basrv5n4-007

Recebimento dos originais: 06/06/2021
Aceitação para publicação: 13/07/2021

Nícolas Lara
Master's student in Materials Science and Engineering. São Carlos School of Engineering – University of São Paulo (EESC-USP)
Avenida João Dagnone, 1100. 13563-120 - São Carlos, SP – Brazil
nicolaslara@usp.br

Maria Inês Basso Bernardi
Ph.D. in Materials Science and Engineering. São Carlos Institute of Physics – University of São Paulo (EESC-USP)
Avenida João Dagnone , 1100. 13563-120 - São Carlos, SP – Brazil

ABSTRACT
This paper shows the determination of the most energy efficient composition for molding of bioglasses in the SiO_2 -Na_2O-CaO-P_2O_5 system. Ten compositions were formulated in the range where the glass has a bioactivity index greater than 8 and the curves of viscosity as a function of temperature were drawn using the Vogel-Fulcher-Tammann (VFT) equation. Analyzing the curves, the composition 43SiO_2 -30CaO-21Na_2O-6P_2O_5 has the lowest viscosity over the entire working temperature range, requiring less heat to be molded and consequently consuming less energy in the ovens.

Keywords: bioactive glasses, Vogel-Fulcher-Tammann, vitreous conformation, biomaterials .

RESUMO
Este documento mostra a determinação da composição de maior eficiência energética para moldagem de biogás no sistema SiO2 -Na2O-CaO-P2O5. Dez composições foram formuladas na faixa onde o vidro tem um índice de bioatividade maior que 8 e as curvas de viscosidade em função da temperatura foram desenhadas usando a equação Vogel-Fulcher-Tammann (VFT). Analisando as curvas, a composição 43SiO2 -30CaO-21Na2O-6P2O5 tem a viscosidade mais baixa em toda a faixa de temperatura de trabalho, exigindo menos calor para ser moldado e conseqüentemente consumindo menos energia nos fornos.

Palavras-chave: vidros bioativos, Vogel-Fulcher-Tammann, conformação vítrea, biomateriais .

1 INTRODUCTION

Bioactive glasses and glass-ceramics have been developed to solve a wide range of medical problems, including bone repair, cancer therapy, soft tissue repair and dental restorations (KAUR, 2017).

To be molded into a product, a vitreous material must have its viscosity lowered by heating. The viscosity at a given temperature depends on the chemical composition of the material, so it is interesting to determine which composition achieves a lower viscosity at a lower temperature, so that the energy consumption of the ovens is as low as possible (VARSHNEYA and MAURO, 2019).

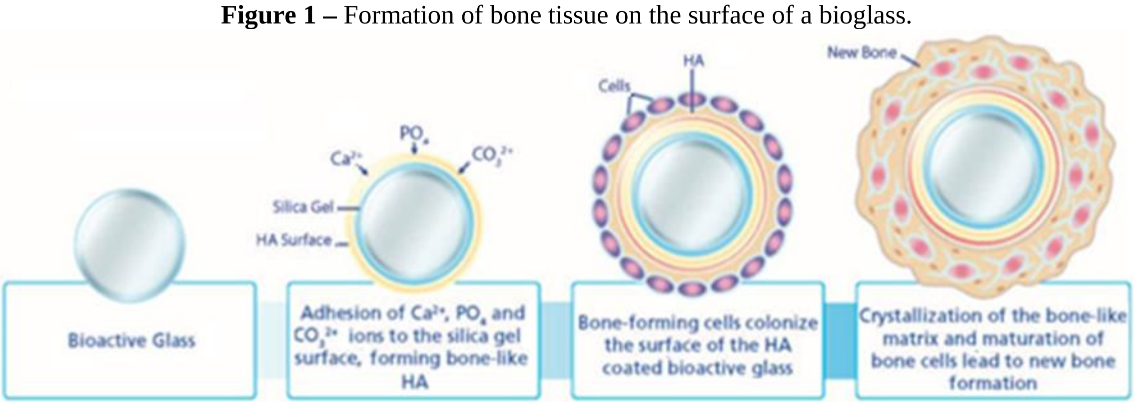
Bioglasses

In the early 1970s, hydroxyapatite, a natural ceramic and the main mineral constituent of bones, was the only synthetic material considered to be entirely compatible with the body. In the search for greater biocompatibility, Professor Larry L. Hench has developed a glass that can be mixed with other ingredients, such as calcium, to join fractured bones (KRISHNAN and LAKSHMI, 2013).

This new vitreous material, when dissolving in a normal physiological environment, activates genes that control osteogenesis, producing bone with the same quality as the natural (XYNOS et al., 2000).

The surface of a bioglass implant, when subjected to an aqueous solution (body fluids), converts into a layer of silica gel rich in CaO and P_2O_5 that mineralizes to hydroxycarbonate in a matter of hours (Fig. 1). This gel layer is so similar to the hydroxyapatite matrix that the osteoblasts differentiate forming a bone layer (ANDERSSON et al., 1990; WALLACE et al. 1999).

Figure 1 – Formation of bone tissue on the surface of a bioglass.



Source: Velez (2016)

The original bioactive glass, called Bioglass 45S5, has the following mass composition: 45% SiO_2, 24.5% CaO, 24.5% Na_2O and 6% P_2O_5. It is composed of minerals that occur naturally in the body and the molecular proportions of calcium and phosphorus oxides are similar to those of bones (XYNOS et al., 2000).

Bioactivity

Professor Hench used the term "bioactivity" to describe the osteogenesis created by his glass (HENCH and WEST, 1996). The concept was then extended to other materials, being defined as those that provoke a specific biological response at the interface with the body, resulting in the formation of a bond with the living tissue (RATNER et al., 1996).

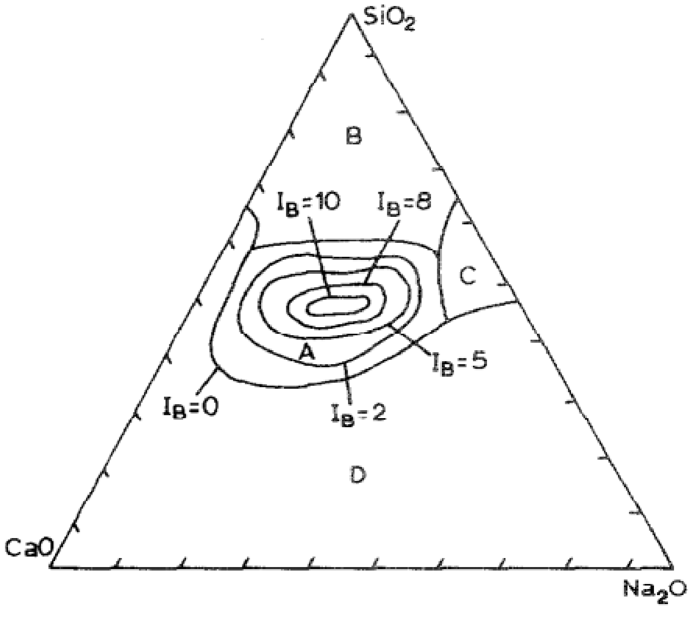
The Bioactivity Index (I_B) is measured by the time ($t_{0.5bb}$) it takes more than half of the interface to bind to the tissue:

$$I_B = \frac{100}{t_{0,5bb}} \tag{1}$$

any material with I_B greater than 8, such as the Bioglass 45S5, will bind to soft and hard tissue. Materials such as synthetic hydroxyapatite, with an I_B value smaller than 8 and greater than 0, will only bind to hard tissue (HENCH, 1990).

For bioglasses, I_B varies depending on the composition, allowing its use in various applications. Figure 2 shows how this index varies with the components percentages:

Figure 2 - Bioactivity index as a function of composition for bioglasses with 6%w of P_2O_5.



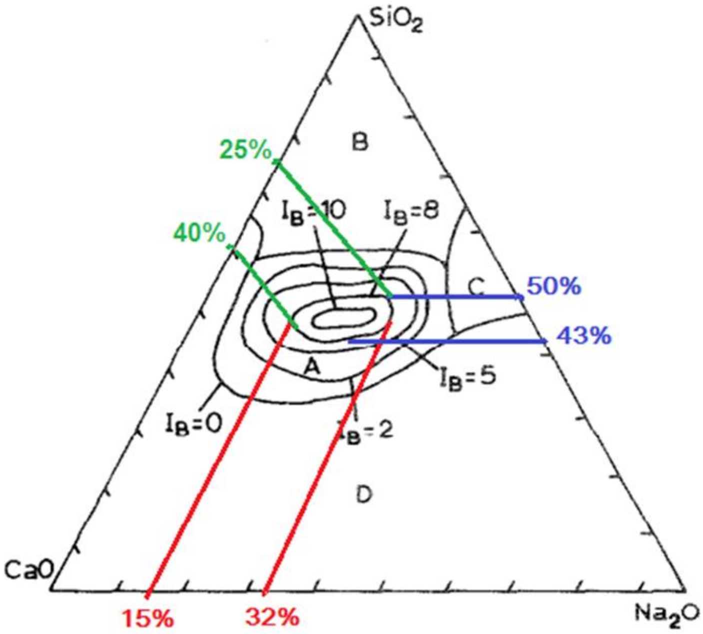
Source: Hench (1990)

In Figure 2, region A is bioactive; in region B there is no bonding with living tissue because the reaction is very slow; in region C binding does not occur because the reaction is too fast; and in region D, bioactivity can occur, but the composition does not favor the formation of glasses (RAWLINGS, 1992).

2 METHODOLOGY

Composition intervals where the bioactivity index (I_B) is at least 8 were selected, as shown in Figure 3:

Figure 3 – Determination of composition ranges where $I_B \geq 8$



Source: adapted from Hench (1990)

The composition ranges determined by Figure 3 are shown in Table 1:

Tabela 1 – Compositions where $I_B \geq 8$.

Component	Mass percentage (%)
SiO_2	43 – 50
CaO	25 – 40
Na_2O	15 – 32
P_2O_5	6

Source: based on Hench (1990)

Ten compositions were elaborated from the determined intervals and their viscosity curves were plotted as a function of temperature, using the Vogel-Fulcher-Tammann (VFT) equation (VOGEL, 1921):

$$log\ \eta(T) = A + \frac{B}{T - T_0} \qquad (2)$$

where A, B and T_0 are constants depending on the chemical composition of the material.

With the curves of each glass, the composition that requires less energy to be softened and, therefore, the most advantageous for manufacturing, was determined.

3 RESULTS AND DISCUSSIONS

The formulated materials are shown in Table 2 and the coefficients A, B and T_0 of the VFT equation were determined using the global statistical modeling of Fluegel (2007):

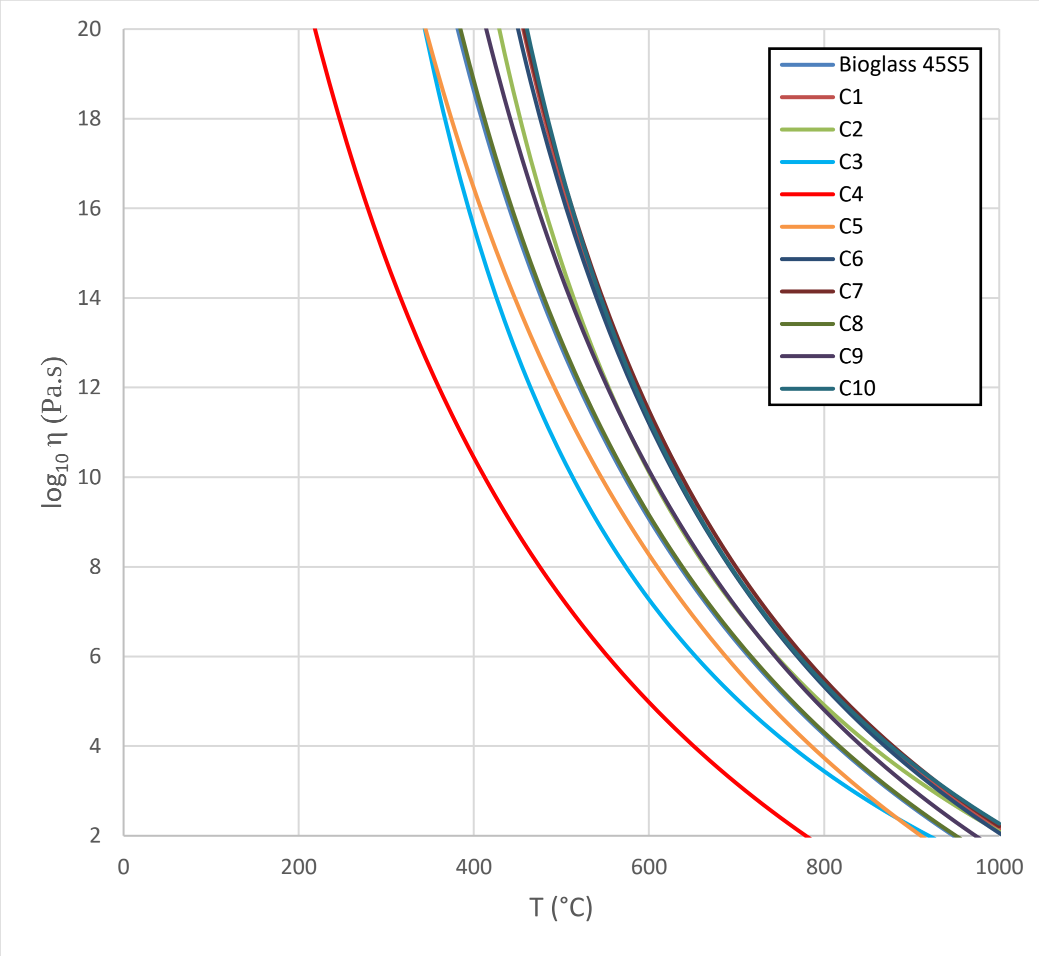
Table 2 - Compositions and coefficients of the VFT equation for bioglasses with $I_B \geq 8$ and 6%w of P_2O_5

Composition code	Mass percentage				VFT equation coefficients		
	SiO_2	CaO	Na_2O	P_2O_5	A	B	T_0
Bioglass 45S5	45	24,5	24,5	6	-10,329	11764	-6,38
C1	45	34	15	6	-8,5282	9300	130,28
C2	50	25	19	6	-7,4158	8314	125,75
C3	50	29	15	6	-6,9382	7699	178,25
C4	43	30	21	6	-11,171	12785	-11,60
C5	43	25	26	6	-12,101	14229	-78,4
C6	43	36	15	6	-9,3798	10301	99,6
C7	45	32	17	6	-9,0385	9965	94,3
C8	45	25	24	6	-10,279	11686	-1,2
C9	43	32	19	6	-10,639	12022	21,7
C10	47	32	15	6	-7,8100	8529	153,9

Source: based on Krishnam and Lakshmi (2013) and Hench (1990)

With the coefficients in hand, the viscosity curves as a function of temperature were plotted:

Figure 4 - Viscosity as a function of temperature



Source: based on Fluegel (2007)

Figure 4 shows that the glass of composition C4 ($43SiO_2$-$30CaO$-$21Na_2O$-$6P_2O_5$) has the lowest viscosity throughout the entire working range, requiring less energy to be softened and molded.

4 FINAL CONSIDERATIONS

The selected composition is different from commercial bioglasses, having lower viscosity, which facilitates its molding and reduces the energy consumption of the ovens. The results obtained open a range of research on the $43SiO_2$-$30CaO$-$21Na_2O$-$6P_2O_5$ composition: *in vitro* cytotoxicity tests, biocompatibility studies among many other properties must be evaluated, such as tenacity, chemical stability and machinability, so that the material can be implemented in the biomedical market.

REFERENCES

ANDERSSON, H.; KARLSSON, K. H.; KANGASNIEMI, K. **Calcium phosphate formation at the surface of bioactive glass in vivo**. Journal of Non-Crystalline Solids, vol. 119, n. 3, p. 290-296, May 1990. http://dx.doi.org/10.1016/0022-3093(90)90301-2.

CONSTANTIAN, M.B. **Use of Ear Cartilage in Orbital Floor Reconstruction.** Plastic and Reconstructive Surgery, v. 69, n. 6, p. 951-955, Jun. 1982. Ovid Technologies (Wolters Kluwer Health). http://dx.doi.org/10.1097/00006534-198206000-00006.

ELLIS, E.; TAN, Y. **Assessment of internal orbital reconstructions for pure blowout fractures: cranial bone grafts versus titanium mesh**. Journal of Oral and Maxillofacial Surgery, vol. 61, n. 4, p. 442-453, Apr. 2003. http://dx.doi.org/10.1053/joms.2003.50085 .

FLUEGEL, A. **Glass viscosity calculation based on a global statistical modeling approach.** Glass Technology, v. 48, n. 1, p. 13-30, March. 2007.

HENCH, L. L. **The compositional dependence of bioactive glasses and glass-ceramics**. Paper presented at the 7th CIMTEC World Ceramic Congress, Montecatini Terme, Italy, 1990.

HENCH, L. L.; WEST, J. K. **Biological Applications of Bioactive Glasses**, Life Chemistry Reports, Vol. 13, 1996, pp. 187-241.

KAUR, G. **Bioactive Glasses,** Springer International Publishing. 2017. http://dx.doi.org/10.1007/978-3-319-45716-1.

KLEIN, M.; GLATZER, C. **Individual CAD/CAM Fabricated Glass-Bioceramic Implants in Reconstructive Surgery of the Bony Orbital Floor**. Plastic and Reconstructive Surgery, v. 117, no. 2, p. 565-570, Feb. 2006. Ovid Technologies. http://dx.doi.org/10.1097/01.prs.0000200770.83864.bc .

KRISHNAN, V.; LAKSHMI, T. **Bioglass : a novel biocompatible innovation** . Journal of Advanced Pharmaceutical Technology & Research, vol. 4, n. 2, p. 78-79, 2013. http://dx.doi.org/10.4103/2231-4040.111523 .

MEYER, D.; WLADIS, E. **Orbital Fracture**. Available at: https://www.eyefacialplasticsurgery.com/orbital-fracture-blow-out-medial-wall-fracture-trauma.html . Accessed on: Nov. 20, 2020.

RATNER, B. D.; Hoffman A. S.; Schoen, F. J.; LEMMONS, J. E. **An Introduction to Materials in Medicine**. Biomaterials Science; 1996. p. 484.

RAWLINGS, R. D. **Bioactive glasses and glass-ceramics** . Clinical Materials, v. 14, n. 2, p. 155-179, Jan. 1993. Elsevier BV. http://dx.doi.org/10.1016/0267-6605(93)90038-9.

STOOR, P.; MESIMÄKI, K.; LINDQVIST, C.; KONTIO, R. **The use of anatomically drop-shaped bioactive glass S53P4 implants in the reconstruction of orbital floor fractures—A prospective long-term follow-up study**. Journal of Cranio-Maxillofacial

Surgery, vol. 43, n. 6, p. 969-975, Jul. 2015. http://dx.doi.org/10.1016/j.jcms.2015.03.029 .

VARSHNEYA, A. K.; MAURO, J. C. **Fundamentals of Inorganic Glasses**. 3. ed. Amsterdam: Elsevier, 2019.

VELEZ, J. **Ceramic Biomaterials**. Available at: https://openwetware.org/wiki/Ceramic_ Biomaterials ,_ by_Jon_Velez . Accessed on: Nov. 23 2020

VOGEL, H. **Das Temperaturebhaengigkeitsgesetz der Viskositaet von Fluessigkeiten**. Physikalische Zeitschrift n. 22, p. 645, 1921.

WALLACE, K. E.; HILL, R. G.; PEMBROKE, J. T.; BROWN, C. J.; HATTON, P. V. **Influence of Sodium Oxide Content on Bioactive Glass Composite**. Journal of Materials Science: Materials in Medicine, vol. 10, n. 12, p. 697-701, 1999. Springer Science and Business Media LLC. http://dx.doi.org/10.1023/a:1008910718446 .

XYNOS, I. D; HUKKANEN, M. V. J.; BATTEN, J. J.; BUTTERY, L. D.; HENCH, L. L.; POLAK, J. M. **Bioglass ®45S5 Stimulates Enhances Bone Osteoblast Formation and Turnover in vitro: implications and applications for bone tissue engineering**. Calcified Tissue International, v. 67, no. 4, p. 321-329, Oct. 2000. Springer Science and