International Journal of Research in Engineering, Science and Management
Volume 7, Issue 5, May 2024
https://www.ijresm.com | ISSN (Online): 2581-5792

Preparation and In-vitro Phase Characterization of BAG-S53P4 Glass-Ceramics using Microwave Energy Irradiation Approach

Oluwatuase Idowu Idowu[1], Olanireti Esther Isinkaye[2*], Oluwasola E. Akinwamide[3]
[1]*Chief Lecturer, Department of Glass and Ceramics, Federal Polytechnic, Ado-Ekiti, Nigeria*
[2]*Instructor, Department of Glass and Ceramics, Federal Polytechnic, Ado-Ekiti, Nigeria*
[3]*Lecturer, Department of Arts and Industrial Design, Federal Polytechnic, Ado-Ekiti, Nigeria*

***Abstract*: In this work, S53P4 bioglass-ceramics (BGCs) were prepared by melt-quenching followed by sintering approach using microwave assisted energy irradiation. The batch material composed of 53% SiO_2, 23% Na_2O, and 20% CaO and 4% P_2O_5 (all in weight percent) was mixed thoroughly and charged inside alumina crucible. The crucible was then place inside microwave kiln box and put inside microwave oven for melting. The oven was powered and timer set. The melt required 40 minutes for completed melting at a temperature of about 1300ºC when read with thermocouple. The melt was then quenched rapidly in water to produce bioglass (BG) frits. The obtained BG frits was later powdered using pulverizer and pelletized by addition of binder (1% PVA solution) followed by sintering at temperature 800 – 1050ºC to obtain BGCs. The samples were then immersed in biological fluids (Na-PBS and Tris-HCl) for 6 hours to determine their in-vitro bioactivity and hydroxyapatite (HAP) formation and afterward characterized. The hardness result showed significant improvement as temperature increase while the bulk density increase with decrease in porosity for the BGCs. All the samples showed good HAP formation in the biological fluids as indicated by the XRD after immersion in biological fluids. However, BG showed better HAP peaks compared with BGCs. The results showed sintering temperature have good impact on the BGCs tested properties while not impairing their in-vitro bioactivity.**

***Keywords*: S53P4 bioglass, in vitro bioactivity, phase analysis, melt-quenching, microwave.**

1. Introduction

As of today, several artificial biomaterials targeted for bone regeneration and repair have been developed. These artificial biomaterials include calcium orthophosphate, bioactive glasses and glass-ceramics, human demineralized bone matrix, variety of polymers and composites among others [1].

Among these biomaterials, bioglasses and glass-ceramics are materials of interest and well investigated considering their excellent biocompatibility and bone bonding ability [2]. Bioglasses are amorphous silicates-based materials which are biologically compatible, bond to bone and stimulate new bone growth whilst dissolving overtime. They possess the ability to restore diseased or damaged bone to its original state and function. Since its first discovery in the late 1969 by Hench et al. [3], several bioglasses have now been developed [4] among which S53P4 is prominent for its good bioactivity, osteointegration and antimicrobial properties [5].

BAG – S53P4 has been widely used for several clinical applications like chronic osteomyelitis therapy, filling of bone cavity and cranio-maxillofacial surgery among others [6], [7].

Despite their excellent bioactivity and biocompatibility, bioactive glasses are limited to very low load-bearing applications due to their poor mechanical properties resulting from their brittleness, and low fracture toughness [8]. As a result, bioactive glass-ceramics (BGCs) containing precipitation of various crystalline phases was developed with good combination of improved mechanical strength and bioactivity [9].

The two major methods that have been in practised to produce bioglasses are traditional melting approach using furnace [10] and sol-gel method [11].

Recently, the search for low-cost synthesis route ushers in the era of microwave assisted synthesis with advantage of achieving glass preparation in few minutes and also modify the reaction environment to obtain nano-phase powders [12].

In this regard, the present work aimed to develop S53P4 BGCs by microwave energy irradiation melt-quenching followed by sintering approach.

2. Materials and method

A. Materials

The starting materials used in this work are pure silica (SiO_2, 99% purity)), anhydrous sodium carbonate (Na_2CO3, 99.8% purity), calcium carbonate ($CaCO_3$, 99.8%), ammonium di-hydrogen orthophosphate ($NH_4H_2PO_4$, 99.9 % purity) as precursor for P_2O_5, and Polyvinyl Alcohol (1% PVA solution) as binder. These materials are of pure chemical grade and used in their as-received state.

B. Glass Melting

A 100 g batch material were composed comprising of $53SiO_2 – 23Na_2O – 20CaO – 4P_2O_5$ (wt. %) and accurately weighed by

*Corresponding author: olanireti12@gmail.com

electronic digital weighing balance (±0.01 accuracy).The weighed materials were thoroughly mixed to avoid batch inhomogeneity. The mixed batch was then poured inside alumina crucible prior to melting. The crucible containing the batch material was then enclosed inside a microwave kiln and placed inside microwave oven (Model: P90N30EP-ZK, 900W microwave power, 2.45GHz frequency) for melting. The complete melting of the batch was achieved at 40 minutes timer of the microwave oven corresponding to temperature of 1300°C measured by R-type thermocouple. The homogeneous bioglass melt was then quenched rapidly in water to avoid crystallization and obtained bioglass frits, then dried.

C. *Production of S53P4 BGCs by Microwave Sintering*

In the preparation of the S53P4 BGCs, the bioglass frits initially obtained were pulverized to obtain fine powder. The powdered frits are then compacted into cylindrical shaped pellets (50 x 10 mm) with addition of binder (1% PVA solution) and sintered using microwave energy irradiation in a microwave kiln enclosed inside microwave oven at temperature of 800, 900, 950 and 1050°C respectively. The sintered bioglass-ceramics were then left to cool to room temperature inside the microwave kiln.

D. *In vitro Bioactivity*

In this work, two different biological fluids namely: Tris buffer solution and sodium phosphate buffer solution (Na-PBS) were used respectively to assess the in vitro bioactivity of the prepared BAG-S53P4 and their glass-ceramics counterparts. 1M Tris buffer solution was prepared by adding Tris (hydroxymethyl) aminomethane/HCl to de-ionized water while Na-PBS was prepared by dissolving sodium chloride, monosodium phosphate and disodium phosphate into de-ionized water. The ion concentrations of the Tris buffer and Na-PBS as compared with blood plasma are shown in Table 1. The samples were then immersed in a clean plastic beaker containing the biological fluids (Tri-HCl and Na-PBS) and placed inside water bath at 37°C and pH 7.4 for 6 hours. After the period, respective sample was evacuated and dropped inside acetone for 5 seconds to stop the surface mineralization followed by drying in a desiccator. The samples were afterward analysed by X-ray diffraction to assess its bioactivity by assessment of CHA/HA (carbonate-hydroxyapatite) phase deposited.

E. *Characterization*

X-ray diffractometer (Rigaku miniflex 600, Japan) was used to investigate the crystalline phases before and after sintering. Also, to assess evolution of the hydroxyapatite layer respectively after in-vitro test. The bulk density and porosity of the samples were also evaluated following standard procedures while the hardness of the samples were investigated using Vicker's micro-hardness tester.

3. Results and Discussion

A. *Physical Properties*

Figure 1 represents the bulk density and the porosity of the S53P4 BG and BGCs respectively. In term of bulk density, the density of the S53P4 (control) is 2.6 g/cm^3 which is close to the theoretical density of bioglass (2.7 g/cm^3) in literature. Density of 1.49 g/cm^3 is obtained for S53P4/800 sintered at 800°C which might be due to porosity initiated as the binder is removed during sintering leaving behind pores. However, the density increase as the sintering temperature increase. It is also observed that porosity decrease as the sintering temperature increase for the glass-ceramics samples. The increase in density might be attributed to densification resulting from increase in vitreous portion by viscous flow at elevated temperature. The level of porosity also decrease across the S53P4 BGCs as the temperature increase which might be due to sealing of pores from the viscous flow. The precipitation of crystals in the glass enhanced the overall density of the S53P4 BGC samples especially for S53P4/1050 sintered at temperature of 1050°C which showed better density than other BGC samples. This similar trend has been reported by previous works [9], [13].

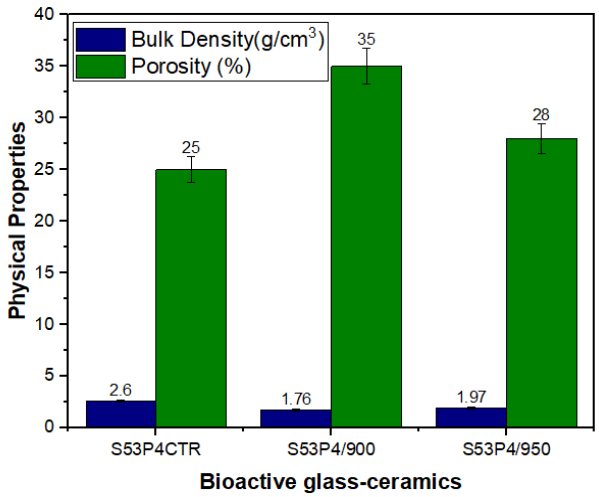

Fig. 1. Density and Porosity

B. *Mechanical Property (Hardness)*

Figure 4 shows the result of the Vickers hardness (HV) compared with its micro-hardness (GPa) value. The result showed that hardness increases with increase in sintering temperature for the S53P4 BGCs. This improvement in hardness observed for S53P4 BGCs might be attributed to increase in densification and decline in porosity of the samples resulting increased viscous flow at elevated temperature. The improvement of hardness for S53P4 BGCs over the S53P4 bioglass (Control) might be due to precipitation of crystals from the glass during sintering. Similar results have been reported by previous works [9], [14], [15].

Table 1
Ion concentration (mM/ Litre) of Na-PBS, Tris-Hcl and human blood plasma

Ion	Na^+	K^+	Mg^+	Ca^+	HCO_3^-	HPO_4^{2-}	SO_4^{2-}	Cl^-	$H_2PO_4^-$
Na-PBS	156.2	-	-	-	-	24.9	-	100.9	5.5
TRIS	-	-	-	-	-	-	-	45	-
Human Blood Plasma	140.0	5.0	1.5	2.5	27.0	1.0	0.5	103.0	-

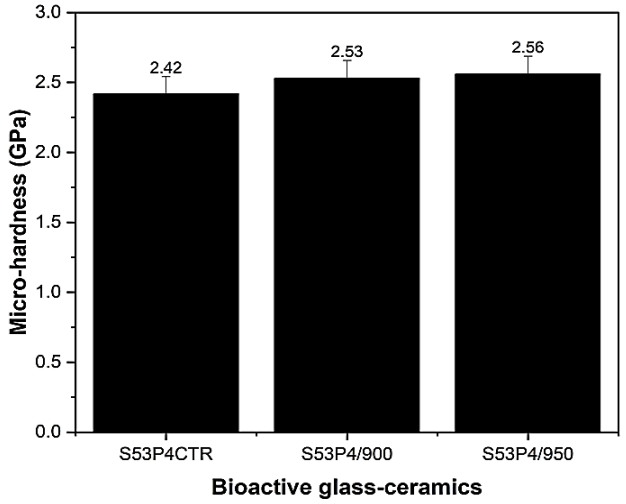

Fig. 2. Micro-hardness

C. Phase Characterization (Before Immersion in Biological Fluids)

Figure 3 shows the superimposed diffractogram of the S53P4 BG and the bioglass-ceramic (BGCs) counterparts sintered at 900 and 950ºC respectively. In Figure 3, it is observed that the S53P4 BG showed characteristic amorphous phase with a broad band peaks which affirmed that a bioglass was obtained from the melt-quenching process. However, a low intensity peak at θ = 27º characteristic of Combeite ($Na_2Ca_2Si_3O_9$) is obtained as previously reported [19]. In Figures 8 and 9 which represent the S53P4 BG sintered at 900 and 900ºC respectively, it is observed that sample sintered at 900ºC (S53P4/900) fully crystallized with major peak of Combeite ($Na_2Ca_2Si_3O_9$) observed. However, for S53P4/950 sintered at 950ºC, a broad band peaks between 15 – 32º characteristics of amorphous phase can be seen to occur with several crystalline peaks identified with Combeite while peaks of wollastonite ($CaSiO_3$) are also seen.

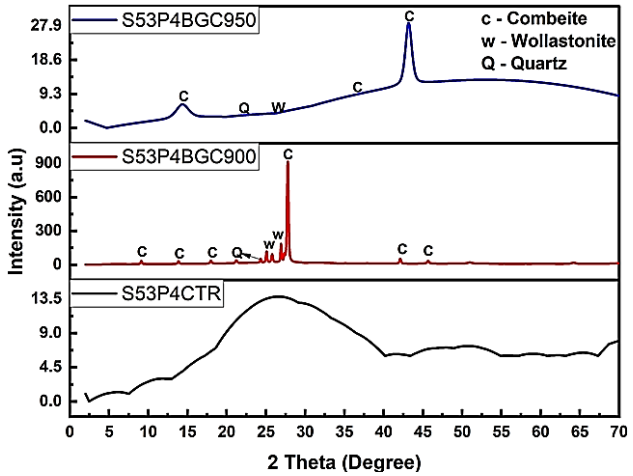

Fig. 3. Superimposed XRD pattern of bioglass and bioglass-ceramics samples

D. Degradability (Weight Loss) in Biological Fluids

Figure 4 represents the result of the weight loss of the bioglass and glass-ceramic samples after immersion in Na-PBS and Tris-HCl respectively for 6 h. It is observed from the results that S53P4 BG showed high weight loss in both Na-PBS and Tris-HCl which might attributed to high release of ions such as Ca, Si, P and Na from the glass into the solutions [16]. For glass-ceramics version, the degradability (weight loss) is not as high compared to S53P4 BG. The glass-ceramic samples showed minimal weight loss in Na-PBS while their weight loss in Tris-HCl is higher with the exception of S53P4/900 though not pronounced. The small changes in weight loss observed for in Na-PBS might be due to an equal amount of elements dissolving from the glass-ceramics and HAP (hydroxyapatite) forming on the surface [17]. Previous work also reported similar trend in Tris-HCl [17]. Resistance to degradation can be improved through crystallization to obtain glass-ceramics as observed in this work and has previously reported [18].

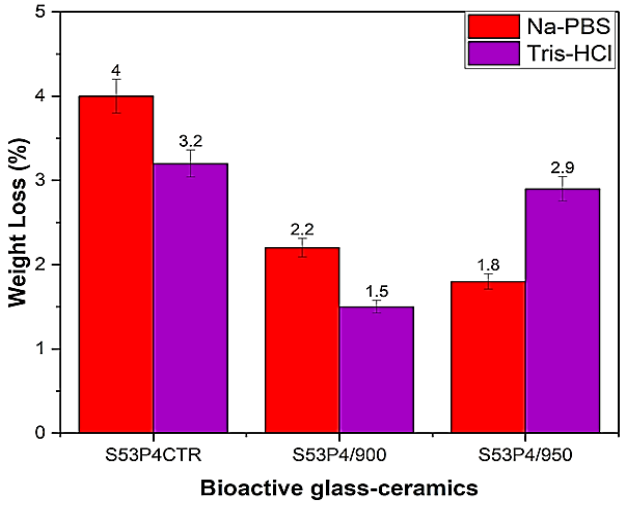

Fig. 4. Weight loss of the samples in biological fluids

E. pH of the Immersion Solutions

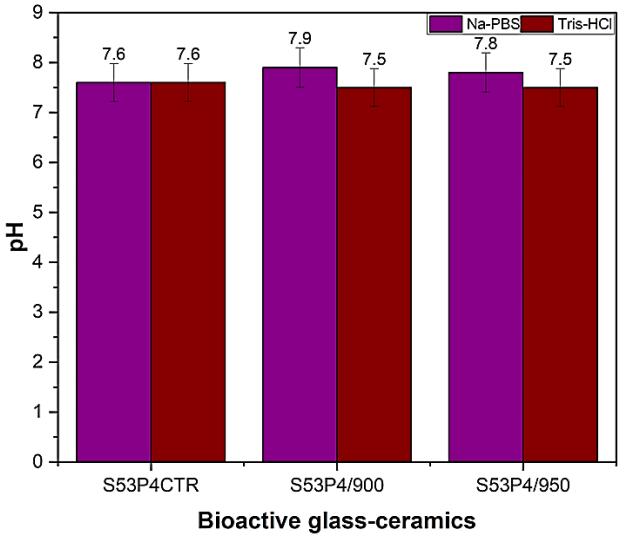

Fig. 5. pH of the biological fluids after immersion of samples

The result of the pH of the Na-PBS and Tris-HCl buffered solutions after immersion of the bioglass and glass-ceramics version for 6 h is shown in Figure 5. It can be observed that the pH of the Na-PBS increase throughout the immersion period, although not significant (7.6 – 7.9). However, in Tris-HCl, a constant pH within 7.6 – 7.5 was maintained throughout the

immersion period. The increase in pH indicates higher reactivity of the glass as reported by Varila et al. [17]. However, since the solutions have differing buffer capacities, the order of reactivity cannot be entirely concluded from pH measurements [17].

F. Phase Characterization (After Immersion in Biological Fluids)

Figure 6 shows the superimposed XRD pattern of the samples after immersion in Na-PBS while Figure 7 represents the superimposed XRD spectra of the samples immersed in 1M Tris-HCl both for 6 h respectively. It is observed for the samples immersed in both Na-PBS and Tris-HCl that new peaks identified to be HAP (Hydroxyapatite) are obtained which indicates the apatite forming ability of the samples in biological fluids and ultimately their compatibility if use in living tissue. However, samples immersed in Na-PBS showed better intensify peaks of HAP than that observed for Tris-HCl, which indicates that hydroxyapatite formation of the samples is more pronounced in Na-PBS probably due to the presence of phosphate in the solution [20].

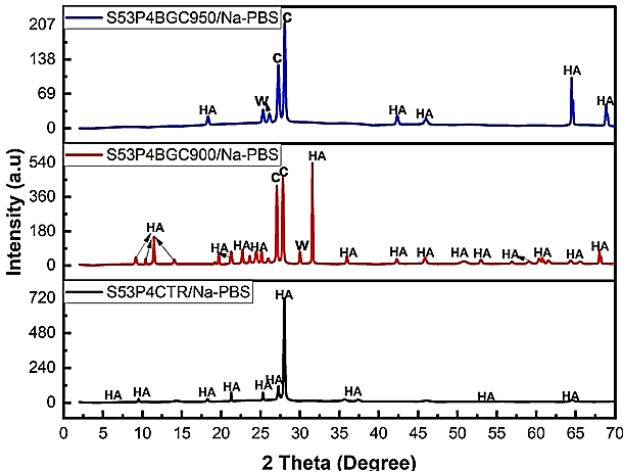

Fig. 6. Superimposed XRD pattern of samples immersed in Na-PBS solution

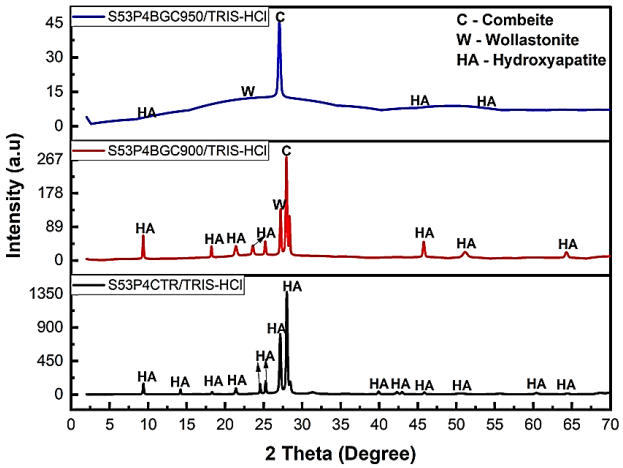

Fig. 7. Superimposed XRD pattern of samples immersed in Tris-HCl solution

4. Conclusion

The present work has successfully developed S53P4 bioglass using melt-quench approach by microwave assisted irradiation and subsequent sintering at different temperatures using microwave energy irradiation. The following conclusions were drawn from this work:

- Microwave assisted energy irradiation provided easy route to melt bioglass within short possible time and less energy consumption.
- The bulk density of the sintered samples increase while the percent porosity decrease as sintering temperature which is attributed formation of vitreous phase arising from viscous flow during sintering.
- The hardness of the samples improved noticeably as sintering temperature increase compared with control sample.
- Weight loss of the sintered samples is considerably lower compared with S53P4 bioglass (control) in biological fluids.
- Hydroxyapatite formation on the samples in Na-PBS is more pronounced compared to Tris-HCl. However, the formation of HAP is well pronounced in non-sintered bioglass compared to sintered bioglass-ceramics.

References

[1] LeGeros, R.Z. Properties of osteoconductive biomaterials: calcium phosphate. Clinical Orthopaedics and Related Research, 395 (2002) 81 – 98.

[2] Palakurthy, S., Venugopal Reddy, K., Pattel, S. and Abdul Azeem, P. A cost effective SiO2– CaO–Na2O bio glass derived from bio waste resources for biomedical applications. Progress in Biomaterials, 9 (2020) 239 – 248.

[3] Hench, L.L., Splinter, R.J., Allen, W.C. and Greenlee T.K. Bonding mechanism at the interface of ceramic prosthetic materials. J. of Biomed. Mater. Res., 5 (1971) 117 – 141.

[4] Vallet-Regí, M. and Ruiz-Hernández, E. Bioceramics: from bone regeneration to cancer nanomedicine. Adv. Mater., 23 (2011) 5177–5218.

[5] Nandi, S.K., Mahato, A., Kundu, B., Mukherjee, P. Doped bioactive glass materials in bone regeneration, advanced techniques in bone regeneration. IntechOpen, (2016) 275 – 328.

[6] Lindfors, N.C., Heikkila, J.T., Koski, I., Mattila, K., Aho, H.J. Bioactive glass and autogenous bone as bone graft substitutes in benign bone tumors. J. Biomed. Mater. Res. B: Appl. Mater., 90 (2009) 131 – 136.

[7] Stoor, P., Pulkkinen, J., Grenman, R. Bioactive glass S53P4 in the filling of cavities in the mastoid cell area in surgery for chronic otitis media. Annotol Rhinol Laryngol, 119 (2010) 377 – 382.

[8] Kokubo, T. Bio-ceramics and their clinical applications. Wood head Publishing Limited, England, (2008).

[9] Leenakul, W., Tunkasiri, T., Tongsiri, N., Pengpat, K. and Ruangsuriya, J. Effect of sintering temperature variations on fabrication of 45S5 bioactive glass-ceramics using rice husk as a source for silica. Mater. Sci. Eng. C., 61 (2016) 695–704.

[10] Franks, K., Abrahams, I., Georgiou, G. and Knowles, J.C. Investigation of thermal parameters and crystallization in a ternary CaO-Na2O-P2O5-based glass system. Biomaterials, 22 (2001) 497–501.

[11] Ramila, A. and Vallet-Regi, M. (2001). Static and Dynamic in-vitro Study of a Sol–gel Glass Bioactivity, Biomaterials, 22, 2301–2306.

[12] Sarkar, S.K. and Lee, B.T. Synthesis of bioactive glass by microwave energy irradiation and its in-vitro biocompatibility. Bioceramics Development and Applications, 1 (2011) 1 – 3.

[13] Filho, O., Torre, G., Hench, L. Effect of crystallization on apatite-layer formation of bioactive glass 45S5. J. Biomed. Mater. Res., 30 (1996) 509–514.

[14] Fujikura, K., Karpukhina, N., Kasuga, T., Brauer, D.S., Hill, R.G., Law, R.V. Influence of strontium – substitution on structure and crystallization of bioglass 45S5. J. Mater. Chem., 22 (2012) 7395.

[15] Vyas, V.K., Kumar, A.S., Prasad, S., Ershad, M., Singh, S.R. and Pyare, R. Preparation and characterization of cobalt oxide doped 45s5 bioactive glass- ceramics. Innovation in Corrosion & Mater. Sci., 5 (2015) 86 – 92.

[16] Chen, J., Zeng, L., Chen, X., Liao, T., Zheng, J. Preparation and characterization of bioactive glass tablets and evaluation of bioactivity and cytotoxicity in vitro. Bioactive Materials, 3 (2018) 315 – 321.

[17] Varila, L., Fagerlund, S., Lehtonen, T., Tuominem, J., Hupa, L. Surface reactions of bioactive glasses in buffered solutions, J. of the Eur. Ceram. Soc., 32 (2012) 2757 – 2763.

[18] Plewinski, M., Schickle, K., Lindner, M., Kirsten, A., Weber, M., Fischer, H. The effect of crystallization of bioactive bioglass 45S5 on apatite formation and degradation. Dent Mater., 2013.

[19] Swea, T.T., Shariffa, K.A., Mohamada, H., Ishikawab, K., Hayashib, K., Abu Bakar, M.H. Behavioural response of cells and bacteria on single and multiple doped Sr and Ag S53P4 sol-gel bioglass. Ceram. Int., 46 (2020) 17881–17890

[20] Fagerlund, S., Hupa, L., Hupa, M. Comparison of reactions of bioactive glasses in different aqueous solutions. Ceram Trans., 218 (2010) 101–113.