Current Trends in Natural Sciences
Vol. 12, Issue 23, pp. 285-290, 2023
https://doi.org/10.47068/ctns.2023.v12i23.034
Current Trends in Natural Sciences (on-line)
ISSN: 2284-953X
ISSN-L: 2284-9521
Current Trends in Natural Sciences (CD-Rom)
ISSN: 2284-9521
ISSN-L: 2284-9521

DETERMINING THE POSSIBLE PERSPECTIVE OF USING CATTLE BONE ASH AS OBTAINABLE RAW MATERIAL FOR BIOACTIVE GLASS

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Current Trends in Natural Sciences

Abstract
Biominerals and bioceramics are widely used in medicine in replacement as well as in the process of regeneration of diseased bone tissue. Bioactive glass is known as one of the effective materials for restoring bone tissue. Dozens of bioenamel materials of different composition and purpose are known at present. Rilligate, borate and other base biomines are known, the main characteristics of which are the presence of two types of oxides, namely P_2O_5 and CaO in the composition of such class of glasses. At the same time, one important thing is noted - the amorphism of biotin, in contrast to ceramics of bioactive crystalline nature, provides the possibility of adjusting the ratio of CaO/P_2O_5 (Ca/P) present in them within wide limits. It creates prospects for the purposeful regulation of the already mentioned properties and the growth of the fields of application.
Biomin materials that are practically usaed in medicine are obtained in compositions of religiate base, when they contain 4-9 wt. % P_2O_5 .
Non-technological compounds such as volatile P_2O_5 or H_3PO_4, hygroscopic or refractory salts of phosphoric acids are mainly used as phosphorus-containing raw materials for obtaining such glasses.
In order to expand the obtainable raw material base of bioactive enamel, a complex, containing CaO and P_2O_5 (total 90 wt. %) and presented in the form of bioactive hydroxyapatite, raw material - ash obtained from cattle bone (bone ash) was selected. 6-10 wt. % P_2O_5 containing glasses were obtained from the charges containing bone ash, quartz sand, chalk and calcined soda, and as well as crystallized materials by their further heat treatment. Their characteristic properties are: bending strength, 390-510 MPa (MPa). Chemical stability with weight losses of 1.1-1.6 wt. % and density 2660-2740 km/m3. By summarizing the research results it was concluded that it is possible to use of beef bone ash as a raw material containing P_2O_5 in the production of bioactive glass materials.

Keywords: bioglass, bone ash, crystallization, properties, glass synthesis.

1. INTRODUCTION

Realization of issues related to the human health recovery and humanization of his being is often directly related to the creation of purposeful materials that are biologically compatible with bone tissue (Williams, 2012). Dozens of materials of different compositions and functional purposes have been created at present: metals and alloys, inert and bioactive ceramics, bioglass, biopolymers, biocomposites, etc. that are used in traumatology and endoprosthetics in order to replace, restore and treat human bone tissue (Ray, 1992).

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In the given list two different types of materials (crystalline and amorphous) are distinguished: these are bioactive ceramics and bioglass. They have the ability to establish a direct active connection to the regenerating bone tissue with extensive involvement of the physiological environment. There is an opinion that introducing biomaterial into the physiological environment that surrounds the regenerating bone tissue leads to two parallel processes - the formation of new bone tissue on the bioactive material and the complete step-by-step regeneration of damaged bone tissue with newly formed bone. that is, in case of using biologically active material, there is mainly the initiation of new bone tissue formation processes (osteoconduction or osteoinduction). In order to conduct above mentioned processes, the use of materials with a certain composition as well as solubility in the physiological environment is required (Vallet-Regí, 2019; Kaur, 2013).
Bioceramics and bioglass used in medicine differ from one another mainly in three positions:
1. Chemical composition and aggregate state (crystalline and amorphous correspondingly);
2. Method for obtaining:
- Bioceramics are obtained by multi-step technology (Synthesis of compound (β-TCP, HAP) by "wet" method (i.e. in solutions)⟶cleaning⟶drying⟶high temperature drying⟶grinding⟶sieving);
- Bioglass is obtained (common option) by a relatively "simple" technology for obtaining industrial glass (material weighing⟶getting charge⟶high temperature synthesis (boiling)⟶smelt milling⟶grinding⟶ sieving);

3. Bioceramics and bioglass differ in terms of chemical stability, i.e. they differ in solubility in physiological environmentt: Both β-TCP (β-$Ca_3(PO_4)_2$) and HAP ($Ca_{10}(PO_4)_6(OH)_2$) bioceramic materials are characterized by unchanged solubility in the physiological environment. The only way to change their kinetics is by adjusting the grain size and creating polymer-bioceramic composites. at the same time, in the case of bioglass, apart from the two mentioned approaches (adjustment by granulometry, inclusion in composites), there is another important way that is varying the solubility of bioglass on account of changing its chemical composition.
Despite the current practical achievements using of bioceramics and bioglass in medicine, the work of obtaining new compositions with their specific properties as well as finding a new food base, is still an actual issue today - goal is to obtain a biomaterial with improved and required properties that will be affordable at the same time. The implementation of above mentioned was the main goal of our research, that was implemented in the direction of obtaining biomins at the new grocery base was carried out in the direction of obtaining bioglasses.

2. MATERIALS AND METHODS

We selected three preconditions for the experimental research.
The first precondition was the base composition of the research bioglasses. Bioactive glasses of generally known composition were obtained in the Na_2O-CaO-SiO_2-P_2O_5 system, including the "classic" 45S5 glass Na_2O (24.5%)+CaO (24.5%)+SiO_2 (45%) and P_2O_5 (6%). This means that the main components are: Na_2O, CaO and SiO_2, that is, the initial source is the three-component system Na_2O-CaO-SiO_2 as well as the glasses obtained in it, however real results are achieved by adding 4-9% P_2O_5 to them, i.e. 6% on average, that is in case of L. Hench's 45S5 glass. The content of all four oxide compounds included in the composition glasses has been selected for research, and this is the first precondition (wt.% 40-50 (R_2O+RO); 45-60 SiO_2 and 5-10 P_2O_5 (Nuha Al-Harbi, 2021; Hench, 2014).

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The second precondition is the selection of raw materials which are characterized by economic and technological expediency. In this regard, we have based on the experience in getting silicate products, namely, the so-called "Royal" bone porcelain. About 40% of P_2O_5 and 53% of CaO are presented in the composition of bone ash although not free, but mainly with distinguished by bioactivity HAp and TCP (more than 90% in total). It is also significant that bone ash contains more or less essential microelements (Mg^{2+}, Fe^{2+}, Mn^{2+}, etc.) as well, that increase its value. The chemical structure of the new raw material bone ash and other raw materials used to obtain bioglass are presented in Table 1.

Table 1. Chemical composition of obtainable materials for new composition bioglass

#	Name of construction material	Content of oxides, mass.%									sum
		P_2O_5	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	LOI	
1	bone ash	40,3	0,4	0,3	0,1	53,2	2,8	-	-	3,3	100
2	calcium carbonate ($CaCO_3$ "pure")	-	-	-	-	55,8	-	-	-	44,2	100
3	calcined soda (technical)	-	-	-	-	-	-	-	58,4	41,6	100
4	quartz sand (Novosiol)	-	99,30	0,13	0,08	-	-	0,05	0,02	0,45	100

The third precondition was the expected bioactivity of the glasses/bioglasses obtained on the new raw material base and its assessment.There is normative documentation standard and the main standard P UCD-13175-3-2015, which regulates the certification properties of two bioactive ceramic materials (with HAp and TCP) in bioceramic material: granulometry, number and type of crystalline phases in the material, porosity and type of pores, material density, material solubility at 37±10C in buffer (pH=7.3±0.1) solution for 24, 48 and 72 hours. Considering the listed amorphousness, four test properties were acquired, these are: density; solubility/weight loss in saline; water absorption/porosity (Kaur, 2019; Standart P UCO 13175-3-2015).

3. RESULTS AND DISCUSSION

An experimental study was carried out to determine the possibility of obtaining bioactive glass in the Na_2O-CaO-SiO_2-P_2O_5 system on the basis of a new raw material such as bone ash.

At the initial stage of the research 12 base glasses of different compositions were selected (the chemical composition of the glasses is given in Table 2 and the material composition of the corresponding four-component glasses in Table 3).

As a result of the base glass synthesis at 1350±50^{0}C temperature, three compositions were identified, namely, C-1, C-2 and C-3 glasses (presented in Table 4), which are distinguished by their technology as they provide homogeneous melts. The correlations of individual oxides included in the corresponding compositions are given in the same table. Another result is that in the technologically distinguished C-1, C-2 and C-3 compositions the content of P_2O_5 is 6-10%, what it is provided by adding bone ash in the amount of 13-28% in the mentioned case. It was also

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established that change of glass properties and expected structural transformations, if they exist, should be directly related to the SiO_2/P_2O_5 relation because their value clearly varies from 4.5 to 7.8 (Table 4).

Table 2. Chemical composition of the studied base glasses

oxide	glass index and oxide content (mass.%)											
	B-1	B-2	B-3	B-4	B-5	B-6	B-7	B-8	B-9	B-10	B-11	B-12
Na_2O	20	20	25	25	20	20	25	25	15	15	15	15
CaO	20	20	25	25	25	25	20	20	20	20	25	20
SiO_2	50	55	40	45	50	45	45	50	55	60	50	60
P_2O_5	10	5	10	5	5	10	10	5	10	5	10	5

Table 3. Material composition of base glass charge

glass (charge) index	The number of components of the charge (weght/100 weght glass)				
	Bone ash	Quartz sand	Chalk (incorporated CaO, %)	Calcined soda	The weight of the charge 100 weght on the glass
B-1	22.4	50.5	10.4(5.8)	35.5	118.3
B-2	12.2	55.6	29.2(12.9)	35.5	126.0
B-3	22.4	56.4	19.4(10.8)	35.5	133.2
B-4	12.4	45.5	32.2(17.9)	43.8	133.7
B-5	12.2	50.5	32.2(17.9)	35.0	129.9
B-6	22.4	45.5	19.4(10.8)	35.0	122.3
B-7	22.4	45.5	10.4(5.8)	43.8	122.1
B-8	12.2	50.5	23.2(12.9)	43.8	129.7
B-9	22.4	55.6	10.4(5.8)	26.3	114.7
B-10	12.2	60.6	23.2(12.9)	26.3	122.3
B-11	22.4	55.5	19.4(10.8)	26.3	103.6
B-12	12.2	60.6	32.2(17.9)	26.3	131.3

Table 4. Studied glasses of optimal composition of Na_2O-CaO-SiO_2-P_2O_5 and the ratio of contained in them oxides

Composition	Content of oxides in glasses, mass. %				Ratio of oxides in glasses			
	Na_2O	CaO	SiO_2	P_2O_5	%(R_2O+RO)/ %(SiO_2 +P_2O_5)	%CaO/ % SiO_2	%CaO/ % P_2O_5	% SiO_2/ \ %P_2O_5
C-1	25	22	47	6	0.89	0.49	3.7	7.8
C-2	23	24	45	8	0.89	0.53	3.0	5.6
C-3	20	25	45	10	0.82	0.56	2.5	4.5

One of the important parts of the conducted research was study of the crystallization ability of the glasses of optimal composition (C-1, C-2 and C-3), which was carried out in the temperature range of 600-1000^{0}C. The lead of the temperature was 50^{0}C, and the delay at individual temperatures was 2 hours. It was necessary to determine the temperature interval for the crystallization of C-series glasses, considering that the final cycle of biomaterial obtaining is the implant making from powders (shaping) and caking (fixing the shape). It is expected that the amorphous nature of the

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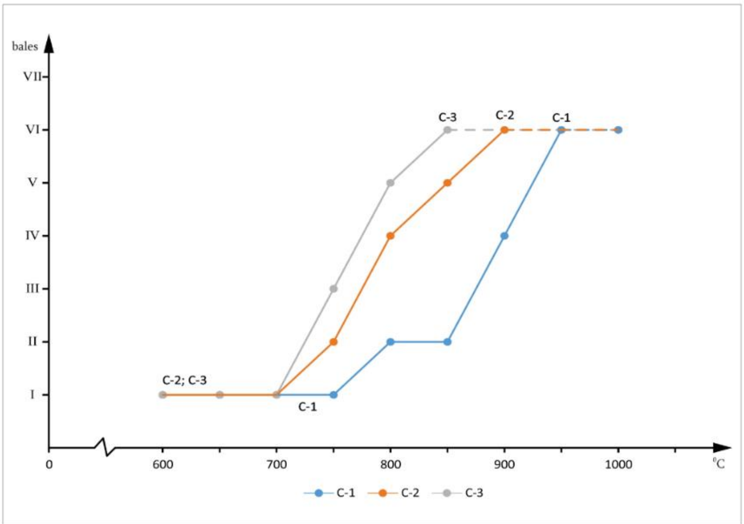
bioglass will be transformed into a bioglasscrystalline material and the properties of the glass will change (Hench, 1998; Suchanek, 1998).

The glasses synthesized of C-series were evaluated on the ability of crystallization and it was determined that according to the crystallization levels, all three glasses can be written down in the following order: C-3 >C-2>C-1. Accordingly, from the graphics given in Figure 2, it can be seen that the C-1 glass starts and finishes crystallization at higher temperature than the C-2 and C-3 glasses (Fig.1).



degree	level of crystallization
I	is not
II	Membrane
III	Surface layer up to 1 mm
IV	The same up to 3 mm
V	up to 50% in volume
VI	up to 70% in volume
VII	up to 90% in volume

Figure 1. Crystallization ability of the studied glasses

Selected test characteristic properties were determined both for C-1, C-2 and C-3 synthesized glasses and for crystallized ones respectively D-1, D-2 and D-3 glass materials obtained by their 2 hour heat treatment at 800^0C. The glass material properties of both series are presented in Table 5. They are completely dependent on the structure and nature of the material with the amorphous-crystalline state.

Table 5. Characteristic properties of the studied and crystallized glasses

#	property	dimension	Composition index and property value					
			C-1	C-2	C-3	D-1	D-2	D-3
1	Compression strength (P)$^{(1)}$	MPa	510	480	470	390	410	480
2	Chemical stability in weight loss (Δg,%)$^{(2)}$	%	1.6	1.5	1.3	1.2	1.2	1.1
3	density (d, g//cm^3)$^{(3)}$	g/cm^3	2.66	2.67	2.69	2.72	2.73	2.74
4	water absorption (W, %)$^{(4)}$	%	0.05	0.06	0.05	1.30	1.25	1.20

Note: (1) - cube-shaped (10×10×10 mm) massive sample;
(2) – $0.1 \le \delta_{avg} \le 0.5$ mm fractionation grains. Reagent-physiological solution (37 ± 1^0C, delay 6 hours)
(3) - massive dowels (mass 3-5 g);
(4) – $0.5 \le \delta_{avg} \le 2.0$ mm fractionation grains.

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4. CONCLUSIONS

Using bone ash in order to determine the properties of glass materials, among them crystalglass. Several important results were obtained by the conducted experimental studies, the main essence of which is as follows:

- It was established that in Na_2O-CaO-SiO_2-P_2O_5 composition it is possible to obtain glasses containing 6-10 mass % P_2O_5, when in the composition of P_2O_5 (phosphoric anhydride) glass the so-called bone ash, the raw materials with a complex composition obtained by processing cattle bone containing CaO and P_2O_5 are used as an input raw materials
- It was determined that using bone ash on a new raw material base, with its content of up to 30% by mass, it is possible to obtain bioglasses corresponding to "classical" bioactive glass qualities, the synthesis of which takes place at 1350-1400^{0}C;
- Conducted experiments have showed that it is possible to vary the properties of the obtained biomaterials and in this way obtain targeted bioglasses with a set of required properties by changing the composition (on account of of the crystallization process) and in this way ensuring the targeted obtainment of biominerals having a set of required properties;
- The conducted studies have confirmed the possibility of obtaining bioactive glass materials using cattle bone ash, a complex raw material, which will make it possible to expand the raw material base for receiving biomaterials and in the future it will lead to the improvement of technical and economic indicators of their reception.

5. ACKNOWLEDGEMENTS

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Williams D.F. (2012). Jhc. Williams Dictionary of Biomaterials. Liverpool University Pross. DOI: https//doi.org/10.5949/upo9781846314438

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