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VITRIFICATION OF IRON OXIDE RICH SLUDGE RESULTED FROM THE GROUNDWATER TREATMENT AS NEW GLASS CERAMIC MATERIALS

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ABSTRACT

This paper offers a new solution to vitrify the sludge resulted from washing the filters used for iron removal phase of the groundwater treatment process. The new glass ceramic materials, obtained after heat treatment at three different temperatures: 800, 900 and 1000 °C were characterized in terms of dimensional stability after firing, apparent density and porosity, hydrolytic stability and iron ions immobilization capacity. The effect of the calcined sludge amount upon the mentioned properties was analysed.

KEYWORDS: glass wastes, sludge waste, vitrification, glass ceramics

1. Introduction

The actual increase of industrial and agriculture wastes amounts primarily require an effective management of solid waste worldwide and is a major environmental concern. Due to insufficient space for land-filling and increasing cost for land disposal, it has become essential to recycle and reutilize these industrial waste [1]. Waste recycling provides alternative materials that facilitate the reduction in the depletion rate of natural materials [2] and also the space for land-filling [3].

Glass recycling become a very attractive concept in the last decades because it has been committed to developing technologies to comply with environmental policies [4]. In this context, it is necessary to create new recycling glass strategies because it is not biodegradable and remains stable for a long time [5]. The recycling of glass brings numerous benefits: reduction in raw material extraction [6], energy savings [6, 7], quality conservation [8], waste reduction [9] and decrease of the environmental contaminant [10].

The glass wastes vitrification is an energy consuming process that is economically viable only by manufacturing new glass-based products, such as glazes [11] glass ceramics [12], glass or glass-ceramic matrix composites [13, 14], glass fibres [15] glass foams [16, 17], and others. A new direction for glass wastes recycling is the vitrification of potential hazardous materials: municipal wastes [18-20], hospital wastes [21], radioactive wastes [22] etc.

The current work aims to offer a new alternative to use the sludge resulting from washing the filters used for iron removal in the groundwater treatment by vitrification together with packaging glass waste and kaolin as vitreous ceramic products able to retain iron ions in the glass-ceramic matrix.

2. Materials and methods

The bottle glass waste used have the oxide composition, determined using an RX fluorescence analyser type Niton XL 3, is presented in the Table 1.

The glass waste was grinded in a Pulverisette type laboratory mill using the material-balls-water ratio of 1:2:1, dried in an oven at 105 °C for 24 hours and then sieved, the granulometric fraction corresponding to passing through the 100 µm mesh sieve being retained.

The oxide composition of the used Bojidar kaolin, provided by the SC IPEC SA supplier is presented in the Table 2.

The sludge used comes from the process of washing the filters used for iron removal in the filtration process stage I for groundwater treatment plant SC Aquatim SA. It was dried and calcined at 750 °C for 6 hours. The main ions present in its composition, determined using RX fluorescence analyser type Niton XL 3, are indicate in the Table 3.

The glass ceramic samples were prepared based on the three mentioned precursors using the recipes detailed in the Table 4.

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Table 1. *The oxide composition of used glass waste*

Oxide	SiO_2	Na_2O	K_2O	CaO	MgO	Al_2O_3	Fe_2O_3
Amount (%)	74.42	12.90	0.19	11.27	0.46	0.75	0.01

Table 2. *The composition of Bojidar kaolin*

Oxide	SiO_2	Na_2O	K_2O	CaO	MgO	Al_2O_3	Fe_2O_3	TiO_2	P.C.
Amount (%)	49.29	0.14	0.87	0.56	0.44	35.18	0.78	0.43	12.31

Table 3. *The composition from the iron removal sludge*

Ion	Fe^{3+}	Mn^{2+}	Ca^{2+}
Amount (%)	83.00	10.03	5.41

Table 4. *Recipes for glass ceramic preparation*

Sample	Precursor amount (%)		
	Glass waste	Calcined sludge	Kaolin
1	60	0	40
2	50	10	40
3	40	20	40
4	30	30	40

The firing treatment was carried out in an electric furnace for 60 minutes at 800, 900 and 1000 °C respectively.

The samples dimensional deviations after firing were measured using an electronic calliper as the average value of three independent replicates.

The apparent density of the obtained foam glasses was measured using the liquid saturation method under vacuum with water as a working liquid.

The hydrolytic stability of the samples was determined according to ISO 719-1985. Two grams of grinded samples, having particles size less than 500 μm, were kept for 60 min in 50 mL deionized water at 98 °C. The volume of HCl needed for neutralization is recorded in order to express the equivalent Na_2O extracted.

The iron immobilization capacity was investigated by measuring the Fe^{3+} ions extraction using leaching tests performed according to the American Extraction Procedure Toxicity Test [23]. Deionized water was used as extraction medium at a constant temperature of 20 ± 2 °C, analysis being performed after 28 days using a Bruker Aurora ICP-MS.

3. Results and Discussion

3.1. Dimensional deviations after the firing process

The glass ceramic shrinkage after the heat treatment is due to the structural changes that occur at high temperature. The volume contractions of the samples during combustion are shown Figure 1 for the three firing temperatures used for synthesis. The lowest shrinkage was recorded for the samples obtained at 800 °C, ranging between 1.21-1.46 % while the synthesis at 1000 °C leads to higher dimensional deviations, ranged between 12.31-19.83 %. As the firing temperature increases, the vitreous phase melts and becomes more fluid, occupying the pores in the glass ceramic mass, thus generating a higher shrinkage of the samples. The substitution of the glass waste with calcined sludge leads to a decrease in contractions during firing, less important at a temperature of 800 °C but more pronounced at 1000 °C.

3.2. Apparent densities and apparent porosities of the glass ceramics

The apparent density and the apparent porosity of the investigated glass ceramic compositions are illustrated in Figures 2 and 3. The apparent density values range between 1.35-1.71 g/cm^3 for the samples treated at 800 °C and 1.63-1.88 g/cm^3 for those fired at 1000 °C. The apparent porosities for the glass ceramics sintered at 800 °C are between 35.09-56.78 % and between 25.61-43.33% when heat treated at 1000 °C.

For all the studied samples, increasing the sintering temperature generates larger amount of melted glass that fills the pores, leading to a higher apparent density and a smaller apparent porosity. Replacing the vitreous precursor with calcined sludge waste produces a lower amount of melt able to fill the pores and therefore higher apparent porosities and lower apparent densities were recorded for the three used thermal treatment temperatures.

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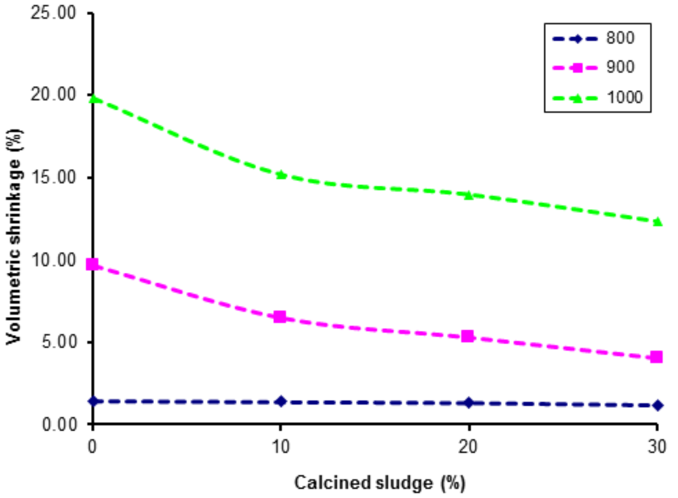

Fig. 1. *Evolution of volume contractions with calcined sludge amount for the studied samples*

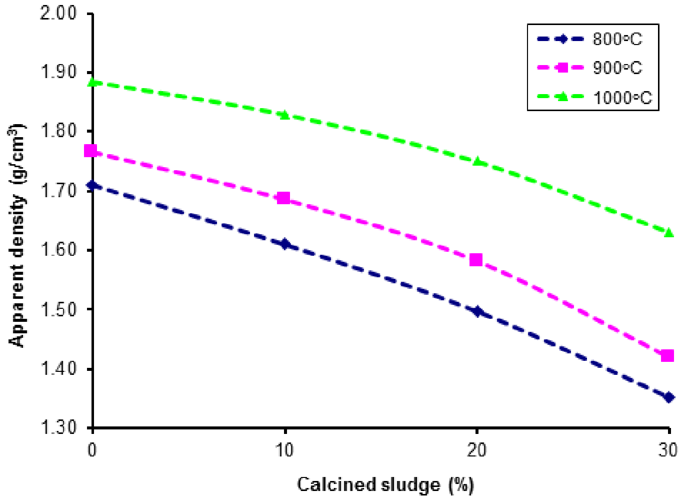

Fig. 2. *Evolution of apparent density with calcined sludge amount for the studied samples*

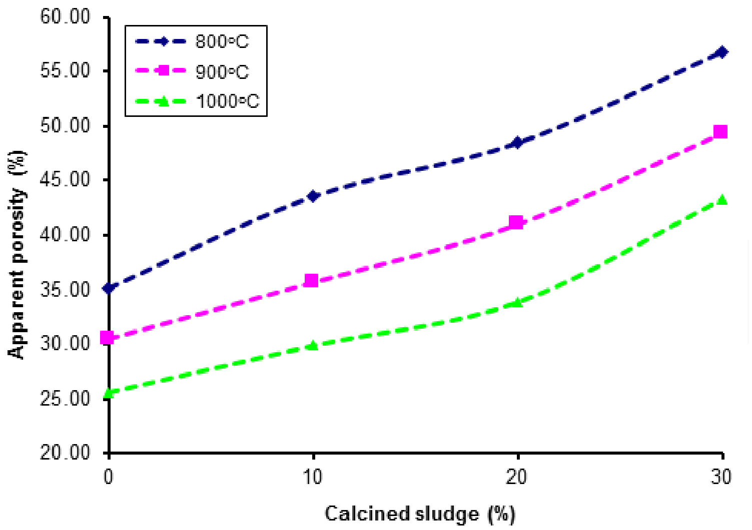

Fig. 3. *Evolution of apparent porosity with calcined sludge amount for the studied samples*

3.3. Hydrolytic stability of the studied glass ceramics

The synthesized glass ceramics stability in water was studied according to ISO 719-1985, the obtained results being summarized in Table 5.

Table 5. *Hydrolytic stability class of the investigated glass ceramics*

Firing temp. (ºC)	Calcined sludge (%)	Equivalent Na_2O extracted (mg)	Stability class
800	0	68.75	HGB3
	10	63.5	HGB3
	20	58.15	HGB2
	30	53.25	HGB2
900	0	65.5	HGB3
	10	56.05	HGB2
	20	50.7	HGB2
	30	48.45	HGB2
1000	0	63.25	HGB3
	10	52.5	HGB2
	20	45.2	HGB2
	30	31.75	HGB2

The alkaline oxide is mainly extracted from the vitreous phase; therefore, the glass ceramics hydrolytic stability increases with the increase of the calcined sludge amount. The higher firing temperature leads to a better integration of the vitreous phase in the glass ceramic matrix, being less exposed toward the water dissolution.

3.4. Iron ions immobilization in the glass ceramics matrix

Iron ions dissolution from the studied glass ceramics after 28 days in deionized water are illustrated in Figure 4.

The amounts of Fe^{3+} extracted after 28 days, for all the samples studied, were extremely low, ranging between 0-0.0023%. The increase in the amount of calcined sludge due to the corresponding decrease in the amount of waste glass leads to an increase of the Fe^{3+} leached, due to the fact that the sludge is the main vector carrying iron ions in the studied masses.

The increase in the firing temperature generates a decrease of iron leached due to an increase of the encapsulation of the calcined sludge waste, associated with a better melting of the glass particles.

This confirms the very good iron immobilization capacity in the studied glass-ceramic matrices.

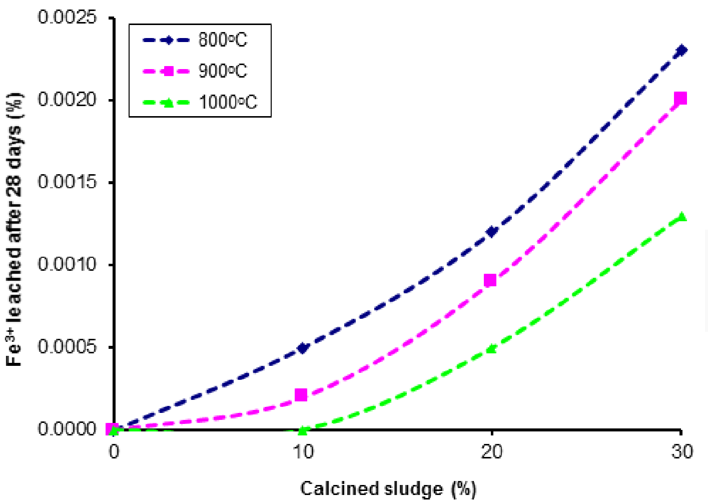

Fig. 4. *Evolution of iron ions leached after 28 days with calcined sludge amount for the studied samples*

4. Conclusions

Glass ceramic masses were synthesized starting from bottle glass wastes and a calcined sludge waste rich in Fe_2O_3 derived from the washing process of the filters used for iron removal in the groundwater treatment together with kaolin as main ceramic precursor.

The dimensional stability of the obtained samples after the firing process range between 1.21-1.46% at 800 ºC and 12.31-19.83% at 1000 ºC respectively. The substitution of the glass waste with calcined sludge leads to an increase of the dimensional stability after the heat treatment.

The apparent density of the synthesized samples range between 1.35-1.88 g/cm^3 while the apparent porosity values are between 25.91-56.78%, depending of the firing temperature. Using higher amounts of calcined sludge waste generates a lower quantity of melt to fill the pores and leading to higher apparent porosities and lower apparent densities.

The hydrolytic stability of the glass ceramics samples, studied according with ISO 719-1985, qualifies them as HGB2-HGB3 materials, having good water resistance. The samples hydrolytic stability increases with the increase of the calcined sludge amount.

The obtained glass ceramic matrix immobilized iron ions very well, the lixiviation values after 28 days range between 0-0.0023% of the total iron content. The increase in the amount of calcined sludge leads to an increase of the Fe^{3+} leached, the sludge being the main vector carrying iron ions in the investigated materials.

These results confirm the viability of the proposed solution for immobilizing iron rich calcined sludge in glass ceramic matrix using bottle glass wastes and kaolin as precursor materials, with multiple economic advantages.

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