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Relationship between Chemical Durability, Structure and the Ionic-Covalent Character of Me-O-P Bond (Me = Cr, Fe), in the Vitreous Part of the System $60P_2O_5$-$2Cr_2O_3$-(38 – x) Na_2O-xFe_2O_3 (with 3 ≤ x ≤ 33 mol%)

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Abstract

The structure and chemical durability in the vitreous part of the system $60P_2O_5$-$2Cr_2O_3$-xFe_2O_3-(38 – x)Na_2O phosphate glasses (with 3 ≤ x ≤ 33 mol%) were investigated using various techniques such as IR spectroscopy, X-ray diffraction and Mössbauer spectroscopy. The presence of Cr_2O_3 and the increase of Fe_2O_3 at the expense of Na_2O in the glass network lead to a large number of covalent and rigid Fe-O-P and Cr-O-P bonds. The infrared and XRD spectra indicate a radical change of structure and show that the increase of the Fe_2O_3 content favors the depolymerization of the vitreous network towards pyrophosphate chains. The presence of Cr_2O_3 in the glass seems to favor the covalent Cr-O-P bonds linked to the most probable cyclic metaphosphate chains. However, when the Fe_2O_3 content increases (≥23 mol%), its impact on the glass network is stronger than that of Cr_2O_3. The infrared and XRD spectra indicate a radical change of structure and show that the increase in Fe_2O_3 content favors the depolymerization of the vitreous network to short pyrophosphate chains. The results of Mössbauer spectroscopy indicate the presence of both Fe (III) and Fe (II) ions which occupied more or less deformed octahedral sites. The growth of the glass transition temperature (Tg) with the increase of the iron oxide in the vitreous network, leads to an improvement of the glass rigidity. This explains the decrease of the ionic radius of the iron and the reinforcement of the interconnection of the chains of vitreous networks. The structure of sodium-chromium-iron phosphate glasses can be considered largely as pyrophosphate units linked to ferric and ferrous ions in octahedral or deformed octahedral coordination. The dissolution rate is 200 times lower than that of the silicate glasses.

Keywords

Chemical Durability, Phosphate Glasses, Chromium and Iron Oxides, IR, XRD, Nuclear and Chemical Wastes

1. Introduction

Phosphate-based glasses are particularly interesting in the scientific and technological fields because they generally have low melting temperatures as well as high coefficient of thermal expansion [1]-[7]. These properties make them good candidates for many applications. It has been found that the introduction of oxides, such as PbO, ZnO, Al_2O_3, Fe_2O_3 and Cr_2O_3, results in the formation of PO-Pb, P-O-Zn, Al-O-P, P-O-Fe and P-O-Cr bonds, leading to improvement of the chemical durability of phosphate glasses. Iron phosphate glasses generally have good chemical resistance [7]-[14]. The synergy of phosphate glasses with certain types of nuclear waste has indicated the possibility of a form of waste with a lower corrosion rate than borosilicate glass. It has been suggested that the chemical durability of sodium-chromium-iron phosphate glasses is attributed to the replacement of P-O-P, Na-O-P bonds by Fe-O-P and Cr-O-P bonds. The presence of Fe-O-P bonds at higher concentrations in the phosphate network makes the glass more resistant to chemical attackers [11] [12] [13] [14] [15]. The purpose of the present paper is to study the effect of the composition on structural modifications, chemical durability, and iono-covalent character of metal-oxygen-phosphorus bonds by varying the Fe_2O_3 ratio along with a series of phosphate glasses with the composition $60P_2O_5$-$2Cr_2O_3$-$(38 - x)Na_2O$-xFe_2O_3 (with $3 \le x \le 33$; mol%). It follows that the structural change causes a depolymerization to short pyrophosphate chains, and leads to a strengthening of bonds, better than that cyclic metaphosphate chains or isolated short orthophosphate chains, by the formation almost certainly aggregates of crystallites in perfect equilibrium with the vitreous bath. This results in high chemical durability [7] [11] [12] [15] [16] [17] [18] [19].

2. Experimental Procedure

The synthesis of phosphate glasses with the composition $60P_2O_5$-$2Cr_2O_3$-$(38 - x)Na_2O$-xFe_2O_3 (with $3 \le x \le 33$; mol%) was carried out by directly melting the mixtures with suitable proportions of $(NH_4)H_2PO_4$, Na_2CO_3,

Cr_2O_3 and Fe_2O_3. The reagents were finely ground and then introduced into a porcelain crucible. They were initially heated at 300˚C for 1 h and then held at 500˚C for 1 h to complete their decomposition. The reaction mixture was then heated at 1060˚C - 1100˚C for 20 min. The homogeneous liquid obtained was poured into an aluminum plate, which had been preheated to 200˚C to avoid thermal shock. Pellets of ≈1 cm in diameter and 2 - 3 mm thick were obtained. The dissolution rate (D_R) of these glasses was evaluated based on the weight loss of the sample. The samples were immersed in Pyrex beakers containing 100 ml of distilled water and heated to 90˚C. The surface of the sample was constantly immersed in distilled water for 30 consecutive days. The IR spectra of the phosphate glasses studied were determined in the zone between 400 and 1600 cm^{-1} with a resolution of 2 cm^{-1}. The samples were finely ground and mixed with KBr. Infrared spectroscopic analysis of the materials was performed using a Vertex Spectrometer 70 and recorded on a DTGS (triglycine deuterium sulfate) detector. The vitreous state was evident by its brilliance and transparency, and confirmed by XRD. The glasses S_1 and S_4 were annealed at 500˚C and 650˚C, respectively, for 48 hours. Structural determination was performed using XRD which allowed us to follow the structural evolution using a Pro MPD Panalytical X'Pert diffractometer. Differential scanning calorimetry was performed using DSC 121 type SETARAM, at a heating rate of 10˚C/min in a flowing Argon atmosphere (30 cm^3/mn) with alumina crucibles. ^{57}Fe Mössbauer spectra were recorded at room temperature in transmission geometry using a conventional constant-acceleration spectrometer with a 57Co(Rh) source. The Mössbauer samples consisted of powdered glass, containing 5 mg Fe per cm^2. A α-Fe foil was used as a calibration of the spectrometer, and the values of the isomer shift (δ) of the measured samples are reported relative to that of α-Fe measured at 300 K. The Mössbauer spectra were fitted by means of the NORMOS program, assuming the superposition of two quadrupolar doublets with Lorentzian line.

3. Chemical Durability

As presented in **Table 1** and **Figure 1**, the dissolution rate (D_R), defined as the weight loss of glass (expressed in $g \cdot cm^{-2} \cdot min^{-1}$), showed an impressive decrease which was dependent on the Fe_2O_3 content of samples immersed in 100 ml of distilled water at 90˚C for 30 days [16] [20]. **Figure 1** shows a gradual decrease

Table 1. Compositions, calculated O/P ratio, D_R and transition temperature (Tg) of the glasses series $60P_2O_5$-$2Cr_2O_3$-xFe_2O_3-(38 − x) Na_2O with (3 ≤ x ≤ 33; mol%).

Echantillon de verre	Oxyde de mélanges de départ mol%				Rapport [O/P]	D_R ($g/cm^2/min$)	Tg (˚C) (±2)	T_f (˚C)
	Fe_2O_3	Na_2O	Cr_2O_3	P_2O_5				
S_1	3	35	2	60	2.92	$(1.34 \pm 0.50) \times 10^{-5}$	315	1060
S_2	13	25	2	60	3.08	$(2.32 \pm 0.50) \times 10^{-8}$	471	
S_3	23	15	2	60	3.25	$(2.32 \pm 0.50) \times 10^{-10}$	560	
S_4	33	5	2	60	3.42	$(2.35 \pm 0.50) \times 10^{-10}$	500	

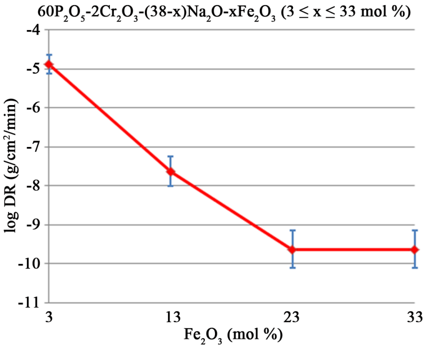

Figure 1. Dissolution rates (D_R) of the glass series $60P_2O_5$-$2Cr_2O_3$-$(38 - x)Na_2O$-xFe_2O_3 versus Fe_2O_3 (mol%).

in the dissolution rate of the glasses from 1.34×10^{-5} to 2.32×10^{-10} $g \cdot cm^{-2} \cdot min^{-1}$ when the Fe_2O_3 content was increased from 3 to 23 mol% to the detriment of Na_2O. Beyond 23 to 33 mol% of Fe_2O_3, the dissolution rate becomes practically constant.

4. X-Ray Diffraction and Differential Scanning Calorimetry Measurements

X-ray diffraction confirmed the vitreous nature of all samples studied [11]. The values of glasses transition temperature (Tg) increase from S_1 to S_4, when the iron oxide increases in glass network involving an improvement glass rigidity (see **Table 2**). The heat treatment of the S_1 and S_2 glasses at 540˚C and 630˚C, respectively, for 48 hours, gives the XRD patterns shown in **Figure 2**. In **Figure 2(a)**, major metaphosphate phases occurred in the sample S_1, whereas the spectrum for the sample S_4, shown in **Figure 2(b)**, indicates the total dominance of pyrophosphate groups [18].

5. Infrared Spectra

The IR spectra of sodium-chromium-iron-phosphate glasses were obtained in a frequency range between 399 and 1600 cm^{-1} using a Fourier-transform spectrometer. The IR spectra of these glasses are shown in **Figure 3**. The bands at 474 - 530 cm^{-1} are assigned to skeletal deformation δske (P-O-P) [21]. The band at 759 - 777 cm^{-1} is assigned to the symmetrical vibration mode ν_{sym} (POP) [12], while the band at 883 - 945 cm^{-1} is attributed to the asymmetric vibration νasym (PO-P) [11] [19] [22]. The band observed at 1074 - 1099 cm^{-1} is assigned to stretching ν_{sym} (PO_3)/ν_{sym} (PO_2), and the region 1268 - 1278 cm^{-1} was attributed to the stretching of ν_{asym} (PO_3) [11] [12]. It should be noted that with increasing values of x, the intensity of characteristic metaphosphate bands tended to decrease when the Fe_2O_3 content was increased to the detriment of Na_2O. This phenomenon appears clearly when the Fe_2O_3 content exceeds 23 mol%, when we observed the disappearance of the vibration band at 1268 - 1288 cm^{-1} assigned to ν_{asym} (PO_3) and attributed to the metaphosphate groups, which became a simple shoulder, while the vibration of the ν_{sym} (PO_3)/ν_{sym} (PO_2) band, located at 1099 -

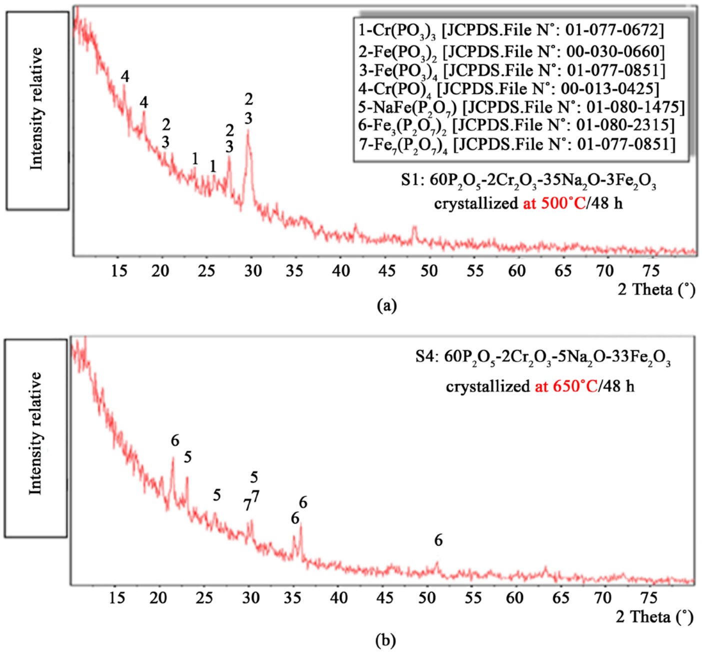

Figure 2. X-ray diffraction spectra for the samples S1 and S2 partially crystallized, respectively, at 500˚C and 650˚C for 48 h.

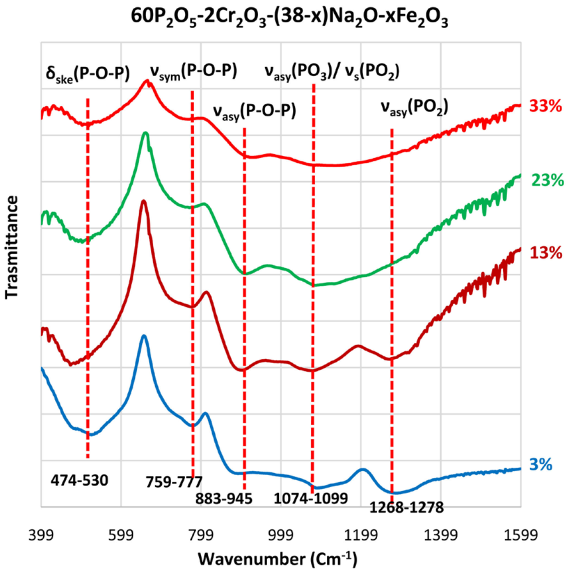

Figure 3. IR spectra of the series $60P_2O_5$-$2Cr_2O_3$-$(38 - x)Na_2O$-xFe_2O_3; x = 3, 13, 23 and 33 mol%.

Table 2. Glass compositions expressed in terms of quaternary systems.

Glass samples	Chemical composition	Composition of glasses in the ternary diagram
S1	$35Na_2O{\cdot}3Fe_2O_3{\cdot}2Cr_2O_3{\cdot}60P_2O_5$	$0.05(Cr_2O_3\text{-}P_2O_5){\cdot}0.875(Na_2O\ P_2O_5){\cdot}0.075(Fe_2O_3\text{-}P_2O_5)$
S2	$25Na_2O{\cdot}13Fe_2O_3{\cdot}2Cr_2O_3{\cdot}55P_2O_5$	$0.05(Cr_2O_3\text{-}P_2O_5){\cdot}0.625(Na_2O\text{-}P_2O_5){\cdot}0.325(Fe_2O_3\text{-}P_2O_5)$
S3	$15Na_2O{\cdot}23Fe_2O_3{\cdot}2Cr_2O_3{\cdot}55P_2O_5$	$0.05(Cr_2O_3\text{-}P_2O_5){\cdot}0.375(Na_2O\text{-}P_2O_5){\cdot}0.575(Fe_2O_3\text{-}P_2O_5)$
S4	$5Na_2O{\cdot}33Fe_2O_3{\cdot}2Cr_2O_3{\cdot}55P_2O_5$	$0.05(Cr_2O_3\text{-}P_2O_5){\cdot}0.125(Na_2O\text{-}P_2O_5){\cdot}0.825(Fe_2O_3\text{-}P_2O_5)$

1070 cm^{-1} and attributed to the majority pyrophosphate groups, became wider [9] [11] [23] [24]. So, as we can be seen clearly on the ternary diagram (**Figure 4**), the localization of S1, S2, S3, and S4 indicates the depolymerised structure from metaphosphate (S1) to predominant pyrophosphates groups (S4).

57Fe Mössbauer spectrometry and iron redox phenomenon: The 300 K Mössbauer spectra of sodium chromium iron phosphate glasses of composition (38 − x)Na_2O-xFe_2O_3-2Cr_2O_3-60P_2O_5 (with $8 \le x \le 38$; mol%) are illustrated in **Figure 5**. They exhibit a central prevailing quadrupolar doublet with broadened lines and a small line at higher velocities. The first part is well described by means of at least four quadrupolar doublets, which can be unambiguously attributed to high-spin-state Fe^{3+} in octahedral sites. A quadrupolar splitting distribution can also be successfully used, which is consistent with a disordered local structure, as expected in a glassy system [12]. The second part of the Mössbauer spectra can be associated with a minor quadrupolar component, which is well described by two doublets, unambiguously ascribed to the presence of Fe^{2+}. The refined values of the mean Mössbauer hyperfine parameters, such as isomer shift (δ), quadrupolar splitting (ΔE_Q) and fraction of Fe^{2+} are listed in **Table 3**. The values of the quadrupolar splittings suggest Fe^{2+} and Fe^{3+} with distorted octahedral coordination [12] [25]. This description allows us to determine the iron redox ratio, $Fe^{2+}/(Fe^{2+} + Fe^{3+})$, from the corresponding spectral area of each component, assuming the same value of the f-Lamb Mössbauer factor. It can be concluded that the increasing Fe_2O_3 content favours a reduction of Fe^{3+} to Fe^{2+} when melting in air. It seems, then, that the significant factor affecting the redox state of the iron in sodium chromium-iron phosphate glasses is the presence of ammonium phosphate, $(NH_4)H_2PO_4$, in a batch composition, which acts as a source of P_2O_5. As a matter of fact, reducing conditions were created during the melting process, which favours the reduction of Fe^{3+} to Fe^{2+} [12]. However, this

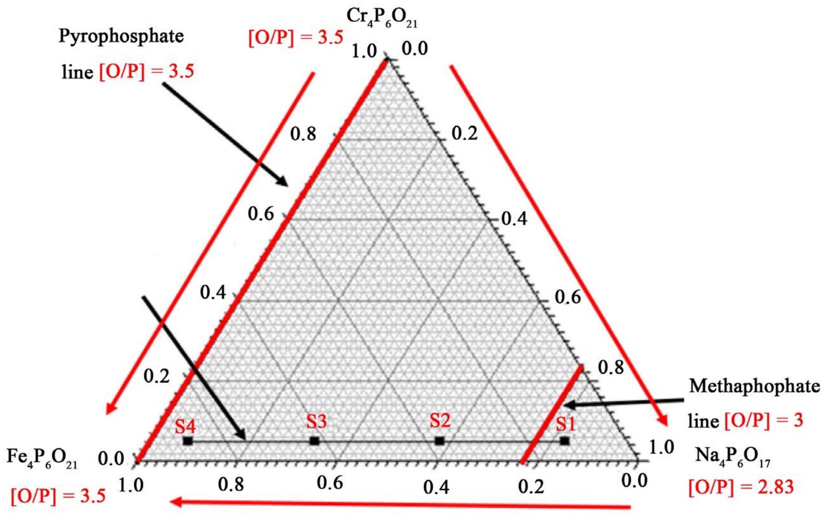

Figure 4. Localization of the investigated glasses compositions in the ternary diagram of ($Cr_2O_3 \cdot P_2O_5$)-($Na_2O \cdot P_2O_5$)-($Fe_2O_3 \cdot P_2O_5$). **Table 1** gives the corresponding compositions in the quaternary system Na_2O-Fe_2O_3-Cr_2O_3-P_2O_5.

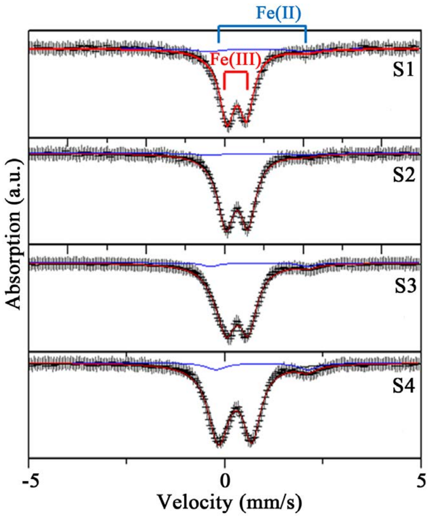

Figure 5. Room temperature Mössbauer spectra of the series of $60P_2O_5$-$2Cr_2O_3$-xFe_2O_3-$(38 - x)Na_2O$ glasses.

Table 3. Mössbauer characteristics of the series of $60P_2O_5$-$2Cr_2O_3$-xFe_2O_3-$(38 - x)$ Na_2O glasses.

Echantillon	Site-1-Fe^{3+}				Site-2-Fe^{2+}			
	A (%)	ΔEQ (mm/s)	Γ (mm/s)	δ (mm/s)	A (%)	ΔEQ (mm/s)	Γ (mm/s)	δ (mm/s)
S1	**92.5**	0.50	0.44	**0.42**	**7.5**	2.48	0.75	**0.93**
S2	**95.3**	0.56	0.50	**0.42**	**4.7**	2.42	0.75	**0.93**
S3	**94.6**	0.56	0.63	**0.43**	**5.4**	2.46	0.5	**0.95**
S4	**90**	0.844	0.64	**0.38**	**10**	2.34	0.65	**1.06**

reduction does not exceed 10%. Some authors have suggested that the $Fe^{2+}/(Fe^{3+} + Fe^{2+})$ ratio is related more to the melting temperature than to the glass composition [12] [26]. The obtained values lie in the same range of about $[Fe^{2+}]/[Fe^{2+}] + [Fe^{3+}] = 5\% - 10\%$.

6. Discussion

In this study, we prepared glass series with different percentages of phosphate oxides (Na_2O, Fe_2O_3, Cr_2O_3 and P_2O_5) as the basic constituent. In this series we substituted the Na_2O oxide with Fe_2O_3 oxide while keeping the percentage of P_2O_5 and Cr_2O_3 oxides constant. Both the presence of Cr_2O_3 and the substitution of Na_2O with Fe_2O_3 in the glass network led to considerable improvement in chemical durability. The presence of Cr_2O_3 oxide in the glass seems to favor covalent Cr-O-P bonds linked to the most probably cyclic metaphosphate chains [19] [27]. However, the infrared spectra and XRD indicate, both, a radical change in the structure and show that increasing Fe_2O_3 content from 3 to 33 mol% lead to the increase of O/P ratio and favors the depolymerization of the vitreous network by creating predominant short pyrophosphate chains [12] [28]. This depolymerization becomes more obvious when the Fe_2O_3 content reaches 23 mol%, and its impact on the glass network becomes stronger than that of Cr_2O_3, which leads to a strengthening of the chemical bonds by the covalent bond formation Fe-O-P at detriment of P-O-P and Na-O-P bonds, easily hydrated, and possibly Cr-O-P. On the other hand, the increase in the glass transition (Tg) when Fe_2O_3 increases in the phosphate network confirms an improvement in the glass rigidity [29]. This phenomenon explains the decrease of the ionic radius of the iron due, both, to the coordination bonds, translated by the hybridization phenomenon, and to the distorted sites occupied by the iron, which become smaller in the phosphate glass compared to the crystalline sites [30] [31] [32]. Therefore, the electron density becomes very important and leads to a reinforcement of the bonds between the irons and non-bridge oxygen atoms and thus to a reinforcement of the interconnection of the chains of vitreous networks [29] [30]. The results of the Mössbauer spectroscopy indicate a partial reduction of the Fe III iron ions in the state of the Fe II iron ions during the melting. The hyperfine parameters show that these occupied purely octahedral sites more or less deformed. The presence of FeII and FeIII enhances the covalent Fe-O-P band. The increase in the number of covalent Fe-OP bonds favors the formation of short pyrophosphate chains and leads to a strengthening of the bonds, better than those of cyclic metaphosphate or isolated orthophoqphatic chains, by the probably formation of aggregates of crystallites in perfect equilibrium with the vitreous bath [16] [23] [25] [27]. It follows that the structure of sodium-chromium-iron phosphate glass can be considered largely as pyrophosphate units linked to ferric and ferrous ions in octahedral or octahedral coordination [12]. The dissolution rate obtained was 200 times lower than that of the silicate glasses.

7. Conclusion

The structure and chemical durability of $60P_2O_5$-$2Cr_2O_3$-xFe_2O_3-$(38 - x)Na_2O$ phosphate glasses (with $3 \leq x \leq 33$; mol%) were investigated using various techniques such as IR, XRD and Mössbauer spectroscopy. Increased Fe_2O_3 content in the glass composition leads to stronger bondings in the vitreous network. Both, the presence of FeII and FeIII reinforces the covalent Fe-O-P band and makes the vitreous network exceptionally rigid. The IR and DRX spectra indicate the formation of short pyrophosphate chains, which provides important chemical durability. These studied glasses have better chemical durability than silicate glasses and can be used as an alternative form in the vitrification of nuclear waste.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

[1] Sanford, L.M. and Tick, P.A. (1982) US Patent 4.314.031.

[2] Shih, P.Y., Yung, S.W. and Chin, T.S. (1998) Thermal and Corrosion Behavior of P_2O_5-Na_2O-CuO Glasses. *Journal of Non-Crystalline Solids*, **224**, 143-152. https://doi.org/10.1016/S0022-3093(97)00460-2

[3] Shih, P.Y., Yung, S.W., Chen, C.Y., Liu, H.S. and Chin, T.S. (1997) The Effect of SnO and $PbCl_2$ on Properties of Stannous Chlorophosphate Glasses. *Materials Chemistry and Physics*, **50**, 63-69. https://doi.org/10.1016/S0254-0584(97)80185-X

[4] Wei, T.Y., Hu, Y. and Hwa, L.G. (2001) Structure and Elastic Properties of Low-Temperature Sealing Phosphate Glasses. *Journal of Non-Crystalline Solids*, **288**, 140-147. https://doi.org/10.1016/S0022-3093(01)00612-3

[5] Niida, H., Takahashi, M., Uchino, T. and Yoko, T. (2002) Preparation and Structure of Organic-Inorganic Hybrid Precursors for New Type Low-Melting Glasses. *Journal of Non-Crystalline Solids*, **306**, 292-299. https://doi.org/10.1016/S0022-3093(02)01188-2

[6] Liu, H.S., Shih, P.Y. and Chin, T.S. (1996) Thermal, Chemical and Structural Characteristics of Erbium-Doped Sodium Phosphate Glasses. *Physics and Chemistry of Glasses*, **37**, 227.

[7] Fang, X., Ray, C.S., Marasinghe, A. and Day, D.E. (2001) Iron Redox Equilibrium, Structure and Properties of Iron Phosphate Glasses. *Journal of Non-Crystalline Solids*, **283**, 162-172. https://doi.org/10.1016/S0022-3093(01)00416-1

[8] Ouchetto, M. (1993) Caractérisation et approche structural de la région vitreuse du système ternaire Li_2O-CdO-P_2O_5. Diplôme D'étude de 3ème Cycle Sciences Physiques, Faculty of Sciences Rabat, University Mohammed V, Morocco.

[9] Sales, B.C. and Boatner, L. (1986) Physical and Chemical Characteristics of Lead-Iron Phosphate Nuclear Waste Glasses. *Journal of Non-Crystalline Solids*, **79**, 83-116. https://doi.org/10.1016/0022-3093(86)90040-2

[10] Sales, B.C. and Boatner, L.A. (1984) Lead-Iron Phosphate Glass: A Stable Storage Medium for High-Level Nuclear Waste. *Sciences*, **226**, 45-48. https://doi.org/10.1126/science.226.4670.45

[11] Aqdim, S., Sayouty, E.H. and Elouadi, B. (2008) Structural and Durability Investigation of the Vitreous Part of the System (35−z)Na_2O-zFe_2O_3-5Al_2O_3-60P_2O_5. *Eurasian Chemico-Technological Journal*, **10**, 9-17.

[12] Aqdim, S., Elouadi, B. and Grenech, J.M. (2012) Chemical Durability and Structural Approach of the Glass Series (40−y)Na_2O-yFe_2O_3-5Al_2O_3-55P_2O_5—by IR, X-Ray Diffraction and Mössbauer Spectroscopy. *Material Sciences and Engineering*, **27**, Article ID: 012003. https://doi.org/10.1088/1757-899X/27/1/012003

[13] Reis, S.T., Karabulut, M. and Day, D.E. (2002) Structural Features of Lead Iron Phosphate Glasses. *Journal of Non-Crystalline Solids*, **304**, 188-194. https://doi.org/10.1016/S0022-3093(02)01021-9

[14] Yu, X., Day, D.E., Long, G.J. and Brow, R.K. (1997) Properties and Structure of Sodium-Iron Phosphate Glasses. *Journal of Non-Crystalline Solids*, **215**, 21-31. https://doi.org/10.1016/S0022-3093(97)00022-7

[15] Rais, S.T. and Martinelli, J.R. (1999) Cs Immobilization by Sintered Lead Iron Phosphate Glasses. *Journal of Non-Crystalline Solids*, **247**, 241-247. https://doi.org/10.1016/S0022-3093(99)00078-2

[16] Aqdim, S. and Albizane, A. (2015) Structural Feature and Chemical Durability of Sodium Aluminium Iron Phosphate Glasses. *Journal of Environmental Science, Computer Science and Engineering & Technology*, **4**, 509-521.

[17] Makhkhas, Y., Aqdim, S. and Sayouty, E.H. (2013) Study of Sodium-Chromium-

Iron-Phosphate Glass by XRD, IR, Chemical Durability and SEM. *Journal of Materials Science and Chemical Engineering*, **1**, 1-6.
https://doi.org/10.4236/msce.2013.13001

[18] Santic, A., Mogus-Milankovic, A., Furic, K., Bermanec, V., Kimd, W. and Daye, D.E. (2007) Structural Properties of Cr_2O_3-Fe_2O_3-P_2O_5 Glasses, Part I. *Journal of Non-Crystalline Solids*, **353**, 1070-1077.
https://doi.org/10.1016/j.jnoncrysol.2006.12.104

[19] Beloued, N. and Aqdim, S. (2016) Correlation between Chemical Durability Behavior and Structural Approach of the Vitreous Part of the System $55P_2O_5$-$2Cr_2O_3$-(43−x)Na_2O-xPbO. *Advances in Materials Physics and Chemistry*, **6**, 149-156.
https://doi.org/10.4236/ampc.2016.66016

[20] Reis, S.T., Karabulut, M. and Day, D.E. (2001) Chemical Durability and Structure of Zinc-Iron Phosphate Glasses. *Journal of Non-Crystalline Solids*, **292**, 150-157.
https://doi.org/10.1016/S0022-3093(01)00880-8

[21] Videau, J.J. and Le Flem, G. (2009) Les verres phosphatés de la spécificité de l'atome de phosphore à la formation, la structure et la durabilité chimique de phosphates vitreux. 3eme Cycle, Université de Bordeaux, 66.

[22] Vast, P. and Semmoud, A. (1994) Comportement Thermique de Difluorodi-Oxophosphate Ferreux. *Journal of Thermal Analysis*, **41**, 1489-1493.
https://doi.org/10.1007/BF02549945

[23] Moss, R.M., Abou Neel, E.A., Pickup, D.M., Twyman, H.L., Martin, R.A., Henson, M.D., Barney, E.R., Hannon, A.C., Knowles, J.C. and Newport, R.J. (2010) The Effect of Zinc and Titanium on the Structure of Calcium-Sodium Phosphate Based Glass. *Journal of Non-Crystalline Solids*, **356**, 1319-1324.
https://doi.org/10.1016/j.jnoncrysol.2010.03.006

[24] Santic, A., Skoko, Z., Gajovic, A., Reis, S.T., Day, D.E. and Mogus-Milankovic, A. (2011) Physical Properties of Iron Phosphate Glasses Containing Cr_2O_3. *Journal of Non-Crystalline Solids*, **57**, 3578-3584.
https://doi.org/10.1016/j.jnoncrysol.2011.07.011

[25] Nelson, N.B. and Exarhos, G.J. (1979) Vibrational Spectroscopy of Cation-Site Interactions in Phosphate Glasses. *The Journal of Chemical Physics*, **71**, 2739.
https://doi.org/10.1063/1.438679

[26] Kim, C.W., Ray, C.S., Day, D.E., Zhu, D., Ghombert, D., Alloy, A., Mogus Milan-kovic, A. and Karabulut, M. (2003) Chemically Durable Iron Phosphate Glasses for Vitrifying Sodium Bearing Waste (SBW) Using Conventional and Cold Crucible Induction Melting (CCIM) Techniques. *Journal of Nuclear Materials*, **322**, 152-164.
https://doi.org/10.1016/S0022-3115(03)00325-8

[27] Makhlouk, R., Beloued, N. and Aqdim, S. (2018) Study of Chromium-Lead-Phosphate Glasses by XRD, IR, Density and Chemical Durability. *Advances in Materials Physics and Chemistry*, **8**, 269-280.
https://doi.org/10.4236/ampc.2018.86018

[28] Aqdim, S. and Ouchetto, M. (2013) Elaboration and Structural Investigation of Iron (III) Phosphate Glasses. *Advances in Materials Physics and Chemistry*, **3**, 332-339.
https://doi.org/10.4236/ampc.2013.38046

[29] Shaharuddin, S.I.S., Ahmed, I., Furniss, D., Parsons, A.J. and Rudd, C.D. (2015) The Glass Transition Temperature of Iron and Titanium Containing Phosphate Glasses. *Advanced Materials Research*, **1115**, 178-181.
https://doi.org/10.4028/www.scientific.net/AMR.1115.178

[30] El Hadrami, A. (2003) Etude de condition de confinement du plomb et du cadmium

dans des verres à base de metaphosphate de Zinc. Université Cadi Ayyad, Marrackech, Maroc, Université Bourdeaux 1.

[31] Lenglet, M. (2004) Iono-Covalent Character of the Metal Oxygen Bonds in Oxides: A Comparison of Experimental and Theoretical Data. *Active and Passive Electronic Components*, **27**, 1-60. https://doi.org/10.1080/0882751031000116142

[32] Calas, G., Levitz, P., Petiau, J., Bondot, P. and Loupias, G. (1980) Etude de l'ordre local autour du fer dans des verres silicatés naturels et synthétiques à l'aide de la spectrométrie d'absorption X. *Revue de Physique Appliquee*, **15**, 1161-1167. https://doi.org/10.1051/rphysap:019800015060116100