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The effect of light rare earths on the chemical durability and weathering of Na_2O–CaO–SiO_2 glasses

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ABSTRACT

In this paper, the investigation on chemical durability and weathering of soda–lime–silicate glasses containing light rare earths (La, Ce and Nd) prepared with traditional melt-quenching methods was carried out by ICP-AES, optical microscopy, SEM and EDS. The weight losses of different glasses in different attacking media were measured, and the normalized releases were calculated. The results showed that the presence of La, Ce and Nd increased the water resistance of glasses and decreased the normalized releases of silicon. The addition of La and Nd improved the acid and alkaline resistance of glass; however, the effect of Ce was opposite. The results of weathering of glasses agree well with experimental data of chemical durability. Comparing with Nd-dopes glass, white plate crystals on the surfaces of undoped, La-dopes and Ce-doped glass were observed, moreover, isolated pits of 0.5–2.0 μm on the surface of Ce-doped glass appeared.

1. Introduction

Glass dissolution has been the subject of extensive research, because some glasses are used as confinement matrices for nuclear waste [1–4]. Ojovan and coworkers [5–8] have reported several papers on the alkali-borosilicate glasses (SiO_2–CaO–Na_2O system containing about 7.5 wt.% of B_2O_3) for immobilizing the radioactive and toxic waste, and they pointed out that this glass is currently being studied extensively with the aim of identifying the most important parameters determining its long-term performance as a host for nuclear waste. It is known that an ionic exchange takes place between mobile ions such as alkali ions and H^+, and that water molecules diffuse into the glass, react with Si–O–Si and Si–O–M, forming Si–OH silanol groups. Network hydrolysis reactions lead to the formation of a porous surface layer on the glass, resulting from the incongruent release of the glass constituents [1,4–11]. This layer is progressively enriched in heavy metals (transition metals, rare earths). This layer is supposed to be formed either by in situ recombination of insoluble elements or by precipitation of solubility limiting phase. It may become a diffusion barrier against corrosion. A preferential rare earth accumulation on the surface has been noted in natural glasses or in nuclear waste glasses, which is probably formed by an in situ recombination [12,13]. Rare earth glasses are known for their interesting mechanical properties, such as hardness, elastic moduli, thermal expansion coefficient, glass transition temperature, especially, high chemical durability, because rare earth cations have higher field strength than traditional network modifier cation, such as alkali or alkaline earth ions [14–16]. A high alkaline durability was noted in aluminosilicate glasses containing La_2O_3 and Y_2O_3. Investigations have been performed on the aqueous alteration of rare earth aluminosilicate glasses and a protective effect of rare earth has been noted, assimilated to metal passivation [1,3,12,13,17].

The weathering of float flat glass in soda–lime–silicate system glass is a worldwide issue [18]. Present measures taken to protect glass from weathering have some effects on glass in storage, but few effects on glass in use. As mentioned above, rare earth additives might improve the chemical durability and weatherability of flat glass. Therefore, the aim of this paper is to focus on the effect of light rare earth additives (La_2O_3, CeO_2 and Nd_2O_3) on chemical durability and weatherability of soda–lime–silicate flat glass.

2. Experimental

2.1. Synthesis of glass

Batch compositions (71.52SiO_2, 12.99Na_2O, 0.31K_2O, 8.87CaO, 0.58Al_2O_3, 5.73MgO, expressed in mol.%) were mixed with 1 mol.% of different light rare earths (La_2O_3, CeO_2, Nd_2O_3 respectively) and ground. The mixtures were fused in corundum crucibles by means of an electric furnace in the 1500–1580 °C temperature range for 2 h and 30 min. The melt was poured into a pre-heating stainless steel model to form, and then annealed at 520 °C for 1 h. Regular bulk glass samples were prepared for test of chemical stability and weathering. The glass samples were labeled FG, FGL, FGC

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and FGN, the FG indicates the soda–lime–silicate flat glass without doping rare earth, FGL, FGC, FGN indicate the glasses doped with La_2O_3, CeO_2, and Nd_2O_3 respectively.

2.2. Corrosion test

In order to evaluate the chemical stability and weatherability of the glasses doped with rare earth oxides, experiments on the chemical stability and weathering of glass samples were performed. Glass samples were firstly immersed in deionized water, acid (1 M HCl) and alkaline (mixture of 1 M NaOH and 1 M Na_2CO_3 with same volume) solution respectively at room temperature (about 25–30 °C), at fixed time intervals, they were remove from the solution, rinsed, dried at 100 °C, weighed, and then placed in the respective solutions again. The chemical durability of the glass samples was expressed by weight loss per surface unit in different solutions. In addition, the leachate in the deionized water was analyzed in order to detect the concentration of elements of glasses by ICP-AES. For weathering, the glass samples were submitted to accelerated weathering test under thermostat (75 °C) and humidistat (75% RH) on the basis of literature [9,18] for 13 days. Superficial decay of samples was directly observed by optical microscopy. Scanning electron microscopy equipped with an energy dispersive X-ray spectrometer was used to undertake a detailed study of the surface morphology and chemical composition.

3. Results

3.1. Leachate analyses

The regular FG, FGL, FGC, FGN samples were placed in Teflon containers with 50 ml deionized water, after 26 days test their leachates were characterized by the ICP-AES. Results of leachates analyses and the final weight losses are given in Table 1. The relative error introduced by diluting the leachate was below 10%. The [K], [Na], [Ca], [Mg], [Al], [Si] and [RE] concentrations in leachate are given in mg/l.

Normalized releases have been calculated from the relation [12,13,17] $N_i = [i]V/f_iS$, in 10^3 g/m^2, where $[i]$ is the i element concentration, f_i the weight fraction of the i element, V the leachate volume, S the glass surface area. Table 2 presents the ratios of the normalized releases.

3.2. Chemical durability

In Figs. 1 and 2, the weight losses (in μg/mm^2) for glasses FG, FGL, FGC and FGN in acid and alkaline media at room temperature are reported as a function of the time. The interaction between the leaching medium, the surface and network structure of glass was expressed by the weight loss as a function of time. In Table 3 the final weight losses of FG, FGL, FGC and FGN samples (after 26 days) contacted with 1 M HCl and mixture of 1 M NaOH and 1 M Na_2CO_3 with same volume solutions are summarized.

Table 2
Leaching elements ratios (ratios of the normalized releases).

Glasses	Na/Si	Ca/Si	Al/Si	Na/Al	Ca/Al	Na/Ca
FG	17.44	1.78	3.33	5.23	0.53	9.81
FGL	21.25	1.00	–	–	–	21.25
FGC	12.00	2.67	14.67	0.82	0.18	4.50
FGN	18.50	2.50	2.00	9.25	1.25	7.40

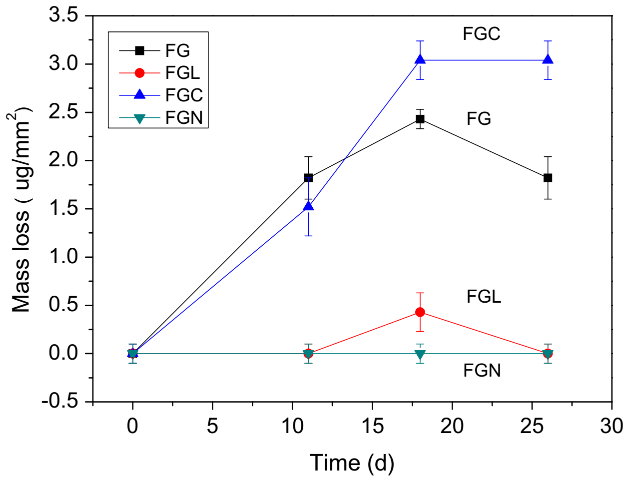


Fig. 1. Mass loss vs. time for different glass samples in 1 M HCl solution at room temperature.

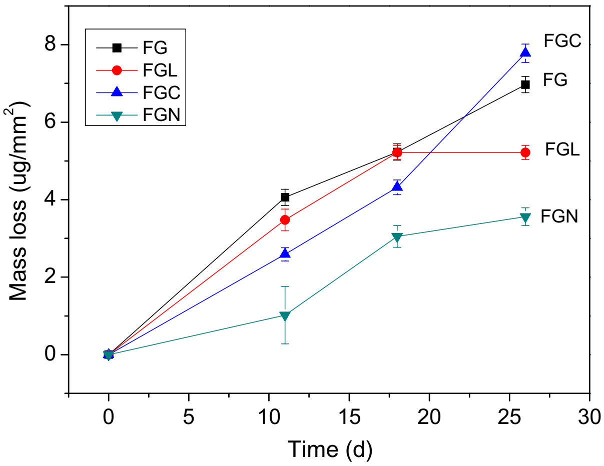


Fig. 2. Mass loss vs. time for different glass samples in alkaline solution at room temperature.

3.3. Weathering

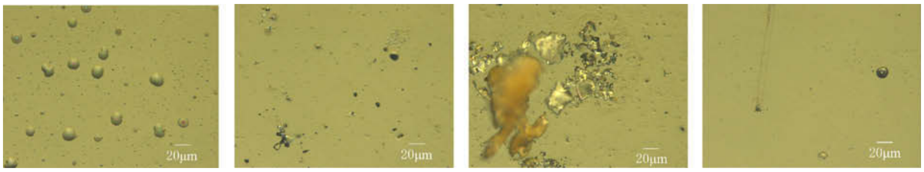
Before the accelerated weathering test, the regular glass samples were polished with PowerPro 4000 apparatus. Fig. 3 presents

Table 1
Results analyses of leachates performed in deionized water at room temperature for 26 days.

Glasses	Weight loss (μg/mm^2)	[K] (mg/l)	[Mg] (mg/l)	[Na]		[Ca]		[Al]		[Si]		[RE]
				mg/l	g/m^2	mg/l	g/m^2	mg/l	g/m^2	mg/l	g/m^2	mg/l
FG	1.24	0.36	0.21	17.00	1.57	1.03	0.16	0.17	0.30	3.31	0.09	<0.05
FGL	0.182	0.16	0.23	21.60	0.85	0.59	0.04	<0.05	–	3.12	0.04	<0.05
FGC	0.121	0.18	0.32	7.95	0.36	1.03	0.08	0.51	0.44	2.52	0.03	<0.05
FGN	0.523	0.15	0.21	8.00	0.37	0.63	0.05	0.05	0.04	1.10	0.02	<0.05

Table 3
Final weight losses of glasses after 26 days in HCl and NaOH + Na_2CO_3 solutions at room temperature.

Glasses	FG (μg/mm^2)	FGL (μg/mm^2)	FGC (μg/mm^2)	FGN (μg/mm^2)
HCl	1.82	0.00	3.04	0.00
NaOH + Na_2CO_3	6.97	5.22	7.78	3.56



(FG) (FGL) (FGC) (FGN)

Fig. 3. Optical microscopy images of glass samples after weathering.

the optical microscopy images of glasses surface subjected to weathering at 75 °C and 75% RH for 13 days. Fig. 4 shows the SEM images of glass samples weathered under same conditions. EDS microanalyses performed on the zone outside and inside the product weathered are summarized in Table 4.

Table 4
EDS microanalyses carried out on the glasses submitted to the weathering (wt.%).

Oxides	FG0	FG1	FGL0	FGL1	FGC0	FGC1	FGN0	FGN1
SiO_2	50.43	35.69	49.15	41.32	47.47	43.87	46.43	51.00
Na_2O	7.13	7.44	7.41	14.67	6.87	8.30	6.77	7.10
K_2O	1.04	0.74	–	–	1.41	1.61	0.82	1.01
CaO	9.84	7.26	9.28	7.31	8.71	7.41	7.92	9.11
MgO	1.88	9.46	2.42	1.65	2.60	1.89	2.06	2.70
Al_2O_3	3.54	3.81	3.31	2.68	4.08	11.07	4.36	5.14
RE_2O_3	–	–	5.55	4.24	4.24	–	5.21	4.29

0 Analyses were spotted on the zones outside the product weathered, 1 inside the product.

4. Discussions

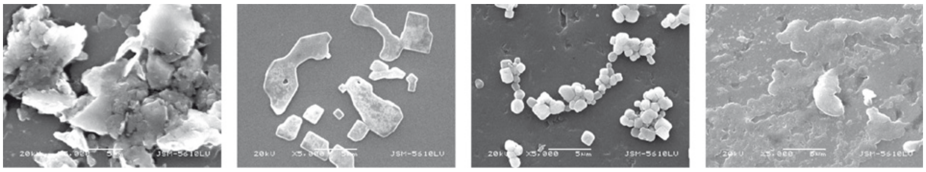
4.1. Leachate analyses

In the case of glasses without doping rare earths, the final weight loss and silicon concentration (Table 1) are 1.24 μg/mm^2 and 3.31 mg/l respectively. When the rare earth oxides (La_2O_3, CeO_2 and Nd_2O_3) are introduced into the glasses, the final weight losses and silicon concentration are about 0.121–0.523 μg/mm^2 and 1.10–3.12 mg/l. The incorporation of rare earth oxides decreases the final weight loss and silicon release, namely, they improve the water resistance of glass overall.

According to the final weight loss, water resistance is seen to increase in the order: FGC > FGL > FGN > FG. The cation field strength (CFS) is very important for the chemical durability of glass, so the order should be as follows based on the CFS theory: FGN > FGC > FGL > FG, because of their CFS (Z/r^2, Z is the valence of the corresponding element and r is its radius) [19] order: $Nd^{3+} > Ce^{3+} > La^{3+}$. However, the results of experiment are not like that. This abnormal result may be related to the compactness of glass structure, thereby the mass densities were measured by Archimedes method, densities of FG, FGL, FGC and FGN are 2.467, 2.521, 2.529 and 2.514 g/cm^2, therefore the poor water resistance of FGN comparing with FGC and FGL can ascribe to the looser glass structure.

The different constituents are usually extracted from the glass at different rates resulting in the formation of a surface layer which has a composition different from that of the corresponding bulk glass. The thickness of the layer increases with time. In the case of more durable glasses such surface layer decreases the dissolution rates of glass constituents and can eventually stop leaching of glass by attacking solution [20]. In order to investigate the constituents release, comparing the Na/Si, Ca/Si, Al/Si ratios (Table 2) for FG, FGL, FGC and FGN samples, the decreasing order for FG glass as follows: Na/Si > Al/Si > Ca/Si, it indicates that the order of preferential releasing element: Na > Al > Ca > Si; for FGL, FGC and FGN glasses, decreasing orders respectively as follows: Na/Si > Ca/Si > Al/Si (Al concentration in leaching solution of FGL is low the detectable limit of 0.05 mg/l, therefore the ratios of Al/Si should be less 1), Al/Si > Ca/Si > Na/Si and Na/Si > Ca/Si > Al/Si respectively, thereby the preferential releasing elements: Na > Ca > Al > Si, Al > Na > Ca > Si and Na > Ca > Al > Si respectively. The presence of La and Nd decreases the Al release, on the contrary, the introduction of Ce increases the Al release, even more than Na (Na/Al = 0.82).

Properties changes must be related to glass structure changes. Structural investigation on rare earth aluminosilicate glasses, carried out by MAS-NMR, Raman and FTIR [14–16], have shown that for rare earth aluminosilicate glasses, the increasing content of the rare earth ions causes a wide distribution of structural units Q_n to occur, (Q refers to a silica tetrahedron with n bridging oxygen atoms). At the same time, the aluminum ions are in four, five and sixfold coordination. Conversely, for cerium aluminosilicate glasses, where the cerium ions are present as Ce^{3+} and Ce^{4+}, the



(FG) (FGL) (FGC) (FGN)

Fig. 4. SEM images of glass samples after weathering.

coordination sites of aluminum ion are Al(6) and Al(4), and the larger fractions of aluminum ion are in Al(6) coordination. Thereby, the change of Al(4)–Al(6) causes Al of FGC glass releases easily due to the changes of glass network connection caused by introducing the ceria.

The compared releases of Si and Al (ratios of Al/Si for all glasses samples) reflect an incongruent leaching process, which shows that the silicon is retained in the alteration layer compared to aluminum, to form a protective silicon-rich layer on the glass surface.

Concerning the La, Ce and Nd releases, their concentration were too low to detect with ICP-AES (test limit of 0.05 mg/l). Therefore, they must be lower than silicon and aluminum releases, indicating an incongruent dissolution process. Research on the dissolution of oxide glasses containing rare earths used for simulating nuclear waste have been carried out by some researchers.

Leturcq et al. [17] have investigated the initial and long-term dissolution rates of aluminosilicate glasses enriched with Ti, Zr and Nd in detail. They supposed that the decrease in dissolution rates could be related to the protective effects of alteration films on the glass surface. These effects differed according to the glass composition, and notably with regard to the nature and concentrations of the network modifiers. SiAlON glass containing 53% Nd_2O_3 instantaneously developed a passivating layer.

In the glass dissolution study of Menard et al. [21], an important retention of rare earth in the alteration layers was observed, over 98.5% of the lanthanides released from the glass were retained in the alteration products on the glass surface, probably by coprecipitated with a siliceous gel. They outline the fact that lanthanides released by glass dissolution preferentially form colloids.

Yttrium and lanthanum releases in the dissolution of lanthanide aluminosilicate glasses have been studied by Bois et al. [12]. It was concluded that La and Y were retained in the alteration layer compared with Si. Y and La releases were influenced by pH of the leachant, their releases could be controlled by the solubility of a solid phase such as a hydroxide or hydroxycarbamide which are very pH dependent. Besides the pCO_2 is also an important factor for lanthanides releases.

In our present studies, silicon normalized releases of glasses containing lanthanum, cerium and neodymium are 0.02–0.04 g/m^2, which are lower than 0.09 g/m^2 of glass without rare earth (Table 1). The low silicon release could be explained by the presence of lanthanum, cerium and neodymium which probably increases the compactness and connection of the glass structure and then reduces the dissolution rate of silica.

4.2. Chemical durability

The weight losses of glass samples in 1 M HCl solution are shown in Fig. 1. It can be seen that the weight losses of FGL and FGN in the HCl solution are considerably lower than that of FG, indicating introducing of La and Nd enhances the acid resistance of glasses. For the FGC glass, the weight loss is similar to that of FG glass at first, however, the value is larger than that of FG after 11 days. In the view of CFS theory, addition of rare earths should increase the glass network structure and connection due to their stronger CFS. However, as stated above, the introducing of cerium causes the fourfold coordinated Al to change into sixfold coordination. It is known that fourfold coordinated Al is the network former, which enhances the glass network structure and then improve the chemical durability through connecting with $[SiO_4]$ tetrahedral. However, the sixfold coordinated Al is a network modifier, which decreases the glass network structure. Thereby, the presence of cerium decreases the acid resistance of glass.

Fig. 2 presents the corrosion behavior of glasses samples in alkaline solution (1 M NaOH + 1 M Na_2CO_3). Up to 18 days, the resistance against alkaline attacking is seen to increase in the order: FGN > FGC > FGL > FG, as expected. However, after 18 days, the increasing orders as follows: FGN > FGL > FG > FGC. It also can be explained by the changing between Al(6) and Al(4).

In explaining the experimental data two possible mechanisms, i.e. ion-exchange leaching and matrix dissolution, were considered for the release of cations from the glass structure in different aqueous solutions [22]. The leaching mechanism involves the replacement of cations (mainly alkali metal ions) with a hydrogen-bearing ion (such as H^+ or H_3O^+) from the solution leading to the formation of a leached layer. Therefore, in neutral and acid media, the initial stage of the reactions is the diffusion of alkali ions through the glass network and across the leached layer into the solution. The diffusion rate of alkali ions gradually slows down as the leached layer becomes thicker and this behavior is represented by line of FGC in Fig. 1. The mass loss of FGC glass undergoes a rapid ascending and reaches a plateau value of 3.04 $\mu g/mm^2$ (Table 3). The matrix dissolution reaction implies the dissolution of the Si–O network by which the glass dissolves directly into the solution either after leaching or without leaching. This type of chemical reaction is characteristic of alkaline media and leads to a higher mass loss as a function of time, dissolution rate can not slow down gradually with time because there is no protective layer. It is highlighted by Fig. 2.

As shown in Table 3, Figs. 1 and 2, the introduction of La and Nd improves the acid and alkaline resistance of glass, Ce decreases the properties.

4.3. Weathering

The weathering mechanism of glass is divided into four steps on the basis of literature sources [11,18,23,24]. At the early stages of glass weathering, water is absorbed by the glass surface with silicon-oxygen unsaturated bonds; secondly, ion-exchange reaction between alkali and alkaline earth ions in glass and H^+ or H_3O^+ of absorption water on the glass surface: $-Si-O-R + H_2O = -Si-OH + -ROH$ or $-Si-O-R + 2H_2O = -Si-O-H_3O + ROH$. The weathering process is not like that of the dissolution of glass in the aqueous solution, the hydroxyl ions produced by ion-exchange reaction can not be diluted, therefore a more serious hydrolysis reaction is caused: $-Si-O-Si- + OH^- = -Si-OH + -Si-O^-$. The final step of weathering is the formation of weathering products by reaction between ROH created by ion-exchange reaction and CO_2 in atmosphere.

Firstly, the whole glass surface was observed by optical microscopy, as shown in Fig. 3. It can be seen that after 13 days of weathering, the FGC glass samples doped with ceria weathered the most serious, and even large tracts of weathered film occurred, while the degree of weathering of the FGL doped with lanthanum oxide and the FGN doped with neodymium oxide glass samples were significantly less than that of the FG without rare earth oxides glass and the FGC glass, there is only a small number of spots and stripes. At the same time we can see that the weatherability of the FGN glass was the strongest.

After 13 days weathering at 75 °C and 75% RH, the surface of glass FG showed very serious deterioration (Fig. 4), a large number of white plate crystals and aggregates produced by weathering were observed. Glass FGL and FGC, smaller white plate crystals and aggregates also appeared, in addition, there was a evident different phenomenon occurred on the surface of FGC glass, isolated pits of 0.5–2.0 μm appeared. For glass FGN, morphology of surface was significant different others, a layer seems to form on the surface.

The EDS microanalyses (Table 4) performed on the zones outside the crystals of glass surface showed a decrease in the Na_2O, MgO, Al_2O_3 and SiO_2 content, in comparison with these components concentration in the original glass, and a relative increase of the CaO content. The decrease of Al_2O_3 and SiO_2 may be due to the fact that their concentration on the surface reduced in the glass formation and annealing process because of their large surface tension. Decrease of Na_2O and MgO can attribute to the higher release tendency [12]. The EDS microanalyses corresponding to the zones of crystals of glass surface indicated that a considerably increasing in the Na_2O content, comprising with the Na_2O concentration outside the crystals, and a decreasing of the CaO, SiO_2 (except for FGN) and RE_2O_3 content was detected. These observations suggest that the white plate crystal on the surfaces of glass FG, FGL and FGC may be Na_2CO_3 or $NaHCO_3$ produced by weathering [18], for glass FGN, it may be calcium silicate or compounds containing Na. Detailed information needs to further test.

5. Conclusions

The results of this study highlight the possibility of improving the chemical durability and weathering resistance of float flat glasses by introducing the suitable rare earth oxides into the glassy products. If only according to the final weight losses, adding light rare earths (La_2O_3, CeO_2 and Nd_2O_3) improves water resistance of float flat glasses, they increase in the order: FGC > FGL > FGN > FG. The incorporation of lanthanum and neodymium decreases the normalized releases of silicon and aluminum, and also cerium reduces the silicon normalized release, However its presence increases the aluminum normalized release because of changing between fourfold coordinated and sixfold coordinated aluminum. Corrosion resistance of float flat glasses doped with lanthanum and neodymium in 1 M HCl and 1 M NaOH + 1 M Na_2CO_3 solutions is enhanced, conversely doping with cerium decreases these properties due to changes of glass structure. Weathering tests suggest that the white plate crystal on the surfaces of glass FG, FGL and FGC may be Na_2CO_3 or $NaHCO_3$, for glass FGN, it may be calcium silicate.

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