



Research article

Flake formation and composition in soda-lime-silica and borosilicate glasses

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ARTICLE INFO

Keywords:

Flake
Glass
Soda-lime-silica
Borosilicate
Corrosion

ABSTRACT

Glass is a food contact material that has been used for a long time in food packaging because it is chemically durable and stable. However, when used for a long time in an aqueous solution or under certain conditions in which alteration may occur, solid flakes may be formed. The phenomenon could be observed when the process of boiling water in a glass kettle is repeated. Transparent and shiny needle-shaped glass fragments appear floating in the water, which may cause complaints from consumers.

The purpose of this study is to investigate the conditions leading to the formation of flakes and to identify the components of the suspended flakes in glass container.

In this study we investigated the formation of flakes at different temperatures (70–100 °C), initial pH values (3–11) and varying the solution composition (with Na⁺, K⁺, Ca²⁺, Mg²⁺ concentrations from 0.2 to 40 mg/L). Two types of glass materials, soda-lime-silica glass and borosilicate glass (heat-resistance glass) were examined.

Results show that flakes were observed under the following conditions: 24 h at more than 90 °C, pH 8, and 20 mg/L Ca²⁺ for soda-lime-silica glass and more than 100 °C, pH 11 for borosilicate glass. The component of flakes was identified as a mixture of hydrates of magnesium, calcium, and aluminum silicate analyzed by X-ray fluorescence spectroscopy, inductively coupled plasma-optical emission spectroscopy, and X-ray diffraction.

1. Introduction

Glass is defined as an inorganic product that originating from the vitrification of liquid glass by cooling without crystallization [1]. Among the various types of glass, soda-lime-silica glass accounts for approximately 80% of the glass used in glass containers, sheet glass, and glassware. It contains silicon dioxide (SiO₂), alkali metals (e.g., Na and K), and alkaline earth metals (e.g., Ca and Mg). The leading constituent of soda-lime-silica glass is SiO₂ (70–75 wt%), followed by 12–16 wt% sodium oxide (Na₂O) and 10–15 wt% calcium oxide (CaO). Because Na₂O dissolves well in aqueous solutions, CaO is added to improve its insolubility. The borosilicate glass used in heat-resistant cooking includes SiO₂ (70–80 wt%) and boron oxide (B₂O₃, 7–13 wt%), which increases its chemical resistance [2].

Commonly, glass is known as a durable material that is chemically stable under normal environmental conditions, and food can be preserved for long periods in glass containers. As consumers continue to demand safer and healthier packaging, the annual global glass packaging market reached \$63.77 billion in 2019, and it is expected to increase by about 1.6 times by 2027 [3]. However, glass can be

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corroded under certain conditions, such as alkaline solutions, long-term storage, high temperature, and high humidity. One of the corrosive reactions in glass is called the ‘flaking phenomenon’ [4].

The flaking mechanism includes two steps [5]. The mechanism was shown in Fig. 1. (1) Ion exchange reaction [Fig. 1. (a)]. When water comes into contact with glass, alkali ions (particularly Na^+) in the glass diffuse into the water, and H^+ ions in water fill the vacant sites in the glass. This reaction is called incongruent dissolution. Due to ion exchange, the pH of the solution increases, and a Si-rich layer is formed on the glass surface. (2) Total dissolution in an alkaline solution [Fig. 1. (b)]. When the pH of water exceeds 9, the alkaline solution acts on the bridging oxygen connecting the SiO_4 tetrahedron, destroying the Si–O–Si bond and creating non-bridging oxygen. If this reaction continues, the network structure of glass completely collapses, and the silica-rich layer is delaminated, forming flakes [6–8].

Some published studies have reported that the composition of glass, temperature, contact time, processing condition of glass, and pH of the solution influence the production of flakes [9–12]. In previous studies, the influencing factors were contact temperature (e.g., 45–121 °C, and 88 °C) [7,13], initial pH of water (e.g., pH 5–12, and pH 5.8–10) [14,15], and addition of ions (e.g., Ca^{2+} , Mg^{2+} , Na^+ , Si^{4+} , Al^{3+} , Fe^{2+} , Zn^{2+} , Mn^{2+} , and Ba^{2+}) [7,13–16]. Solutions are also one of the factors influencing flake formation. Deionized water, tap water and ethanol were used as solutions, and the ethanol solution had a lower Si elution than water. The higher the concentration of the ethanol solution, the lower the elution amount of the glass component [16]. Flakes are more easily generated in tap water than in deionized water because tap water contains many elements such as Ca and Mg. Water with high hardness, indicating the amount of divalent ions such as Ca^{2+} , and Mg^{2+} , quickly produces flakes [17]. However, to the best of our knowledge, no published studies have simultaneously attempted to identify various conditions that affect flake formation according to the glass type.

The aim of this study was to investigate the factors, such as initial pH (3–11), storage time (5–72 h), temperature (70–100 °C), and alkali ion concentration (with Na^+ , K^+ , Ca^{2+} , Mg^{2+} concentrations from 0.2 to 40 mg/L) that influence the formation of flake for both type of glass (soda-lime-silica and borosilicate glass) and identify their composition and structure using inductively coupled plasma-optical emission spectroscopy (ICP-OES), X-ray fluorescence spectroscopy (XRF), and X-ray diffraction (XRD). These results suggested basic research data on flakes by analyzing the various conditions for generating flakes and the structure of flakes depending on the glass type.

2. Materials and methods

2.1. Standards and reagents

All ICP standards, hydrochloric acid (HCl), potassium hydroxide (KOH), calcium chloride (CaCl_2), and magnesium sulfate (MgSO_4) were purchased from Sigma-Aldrich (St. Louis, MO). Deionized water was obtained using a Thermo Fisher Barnstead GenPure system (Thermo Electron LED GmbH, Germany).

2.2. Samples

To investigate the factors influencing flake formation, one soda-lime-silica glass and one borosilicate glass were selected as samples. Glass bottles commonly used in glassware, so they were selected as samples, and their capacity was 250 mL. The composition of the glass samples were given in Table 1. Both had SiO_2 contents of more than 70%. The soda-lime-silica glass sample contained higher alkali oxide content values, except for aluminum oxide (Al_2O_3) than those found in the borosilicate glass sample. However, the B_2O_3 content value was higher in the borosilicate glass sample than in the soda-lime-silica glass sample.

2.3. Investigation of factors affecting flake formation

Three factors influencing flake formation were investigated for soda-lime-silica glass and borosilicate glass samples: (1) Storage temperature: Glass samples containing water were examined at 70, 80, 90, and 100 °C in a drying oven. The pH of the water was adjusted to 9 by adding KOH. (2) pH of water: The pH of tap water (pH 7.5) was adjusted to pH 3, 5, 7, 8, 9, and 11 by adding HCl or KOH. The temperature was set to 90 °C. All pH was measured at room temperature. (3) Concentration of mineral ions in water: To

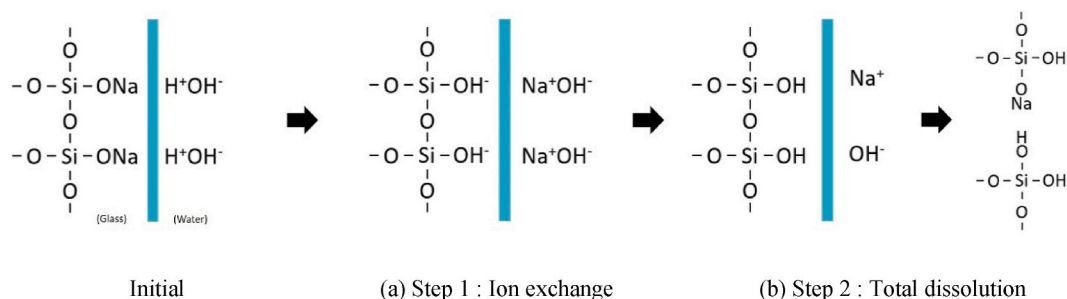


Fig. 1. The possible mechanism of flake formation.

Table 1
Component of soda-lime-silica glass and borosilicate sample.

Component	Soda-lime-silica glass	Borosilicate glass
SiO ₂ (%)	71.9 ± 2.12 ^a	80.2 ± 0.42
Al ₂ O ₃ (%)	1.8 ± 0.73	2.6 ± 0.11
CaO (%)	9.0 ± 2.02	0.0 ± 0.01
MgO (%)	2.9 ± 0.11	<0.01
Na ₂ O (%)	14.1 ± 1.20	4.5 ± 0.20
B ₂ O ₃ (%)	<0.01	12.6 ± 0.14

^a Mean ± SD (n = 2).

examine the effect of mineral components of water in glass bottles water on flakes formation, Ca, Na, Mg, and K, those are the main minerals in water, were added to deionized water to be 0.2 mg/L and 20 mg/L to observe whether flakes were formed. They were stored at 90 °C for 72 h and pH was not adjusted. It was confirmed that Ca and Mg in water affect flake formation for soda-lime-silica glass. In order to determine the detailed concentration, Ca and Mg were added to deionized water up to 0.2, 1, 5, 10, 20, and 40 mg/L, respectively. The temperature and storage time were set in the same conditions. CaCl₂ and MgSO₄ were used as sources of Ca and Mg. The capacity of the sample is 250 mL and water (pH, temperature, calcium and magnesium concentrations are conditioned) was poured into the glass bottle in each experiment.

All experiments were carried out for up to 72 h under each condition in triplicate. After storage in each condition, the water was visually observed and the pH, Ca, Mg, Na, and Si content in water were measured with ICP-OES.

2.4. Instrumental analysis

2.4.1. XRF analysis

Flake samples were collected after passing through filter paper (Advantec 5B, 4 µm) and dried in a drying oven (NDO-520W) at 105 °C for 2 h. All flakes used in the experiment were obtained from soda-lime-silica glass because flake didn't occur enough in borosilicate glass. XRF analysis was performed using an XGT-7200 (Horiba, Kyoto, Japan) with a silicon drift detector to investigate the flake constituents. It has a 50 kV Rh X-ray tube with a diameter of 10 µm, a volume of 50 kV, and a current of 1 mA. The flake specimen, which was placed on the cell window, was transferred to a sample plate positioned in a vacuum chamber and analyzed five times for 300 s each.

2.4.2. ICP-OES analysis

The flakes and solution samples after storage were analyzed by ICP-OES. Hydrofluoric acid (5 mL), nitric acid (2 mL), and deionized water were added to the collected flake 0.2 g, and the latter was ignited twice using an electric muffle furnace (Jisico, Seoul, Korea) at 1000 °C. After cooling, the residues were completely dissolved in 10–15 mL of HCl and mass up to a 250 mL volumetric flask. ICP-OES analysis was performed using an Optima 8300 instrument with an S10 autosampler (PerkinElmer Waltham, MA). The operating conditions were as follows: flow gas, Ar; plasma gas flow, 10 L/min; auxiliary gas flow, 0.20 L/min; nebulizer gas flow, 0.65 L/min; power, 1350 W; flow rate, 1.5 mL/min. The wavelengths of Si, Mg, Ca, Na, and Al were 251.611, 285.213, 317.933, 589.592, and 396.153, respectively. The measurement of the elements was performed in triplicate.

Table 2
The flake formation according to the change of temperature and time.

Type of glass	Time (h)	Contact temperature							
		70 °C		80 °C		90 °C		100 °C	
Soda-lime-silica glass		Flaking	pH	Flaking	pH	Flaking	pH	Flaking	pH
	5	–	(9.2)	–	(9.2)	–	(9.4)	–	(9.3)
	10	–	(9.3)	–	(9.4)	–	(9.6)	O	(9.6)
	24	–	(9.4)	–	(9.5)	O	(9.5)	O	(9.5)
	48	–	(9.5)	–	(9.5)	O	(9.4)	O	(9.6)
	72	–	(9.5)	–	(9.5)	O	(9.6)	O	(9.8)
Borosilicate glasses									
	5	–	(8.7)	–	(9.0)	–	(9.2)	–	(9.2)
	10	–	(9.2)	–	(9.1)	–	(9.6)	–	(9.7)
	24	–	(9.1)	–	(9.2)	–	(9.5)	O	(9.5)
	48	–	(9.1)	–	(9.3)	–	(9.5)	O	(9.4)
	72	–	(9.0)	–	(9.3)	O	(9.5)	O	(9.4)

The flake was detected at higher 90 °C and over 24 h in soda-lime-silica glass and higher 100 °C and over 24 h in borosilicate glass.

3. Results and discussion

3.1. Factors affecting flake formation in glass

3.1.1. Storage temperature

As a result of storing water in a glass bottle at 70, 80, 90, and 100 °C, flakes were not generated below 80 °C (Table 2). Flake formation was observed at high contact temperatures in both glass samples. In soda-lime-silica glass, flakes formed after 24 h at 90 °C and 10 h at 100 °C. In borosilicate glass, flakes formed after 72 h at 90 °C and 24 h at 100 °C. These results are consistent with a previous study that has shown that temperature affects the reaction rate of flake formation [14].

The images of flake formation are shown in Fig. 2. It doesn't occur at the first [Fig. 2. (a)], but when it occurs a little, flakes are such as little particles [Fig. 2. (b)]. As time goes, it looks like a thin transparent sheet [Fig. 2. (c)], but breaks into fine flakes when shaken. After storage time, the content of Ca, Mg, Na, and Si in water were analyzed by ICP-OES and presented in Fig. 3. As for Ca and Mg, the occurrence of flakes was observed while the contents were decreased [Fig. 3. (a), (b)]. On the other hand, the generation of flakes was observed while increasing the content of Na and Si [Fig. 3. (c), (d)]. It is assumed that as flake formation progresses, Ca and Mg are consumed and used for flake formation. Na and Si appear to continuously elute into water without participating in flake formation. The results of this study show that the glass components Ca, Mg, Na, and Si are all released better at higher temperatures, but after a certain period of time, Ca and Mg form flakes and Na and Si continue to increase in solution. It means Ca and Mg are the elements limiting flake formation whereas Na and Si are not. The Si concentration increased dramatically from 1 mg/L to more than 40 or 70 mg/L when the temperature was 100 °C. The concentration of Mg rapidly decreased to 0 mg/L when flakes formed. Compared to borosilicate glass, flakes occurred at shorter time and lower contact temperatures in soda-lime-silica glass. It was surmised that soda-lime-silica glass has higher Ca and Mg contents, so that it is easier to form flakes because it is extracted into the solution through ion exchange.

3.1.2. Initial pH of the water

Table 3 shows flake formation depending on the pH and time. In soda-lime-silica glass, flake formation did not occur when the pH was less than 5, but it was produced after 72 h at pH 7, 24 h at pH 9, and 5 h at pH 11. In borosilicate glass, flakes were not generated below pH 7 but formed after 72 h at pH 9 and 10 h at pH 11. The two types of glass bottles exhibited similar characteristics. First, the higher the initial pH of the water, the earlier flakes formed. As mentioned above, total dissolution occurred effectively when the initial pH of the water was above 9. However, the pH of water did not continuously increase over time because OH^- was used to break Si–O–Si bonds [18]. Kim et al., reported the pH of the solution converged to pH 10 [14], but in our study, the pH of the solution increased over than 10. There is a limit to the ion exchange reaction, and the limit seems to vary depending on the composition of the glass. When the contact time elapses, the pH increases and equilibrates around pH 10, somewhat becoming lower. This is because in high pH solution, OH^- groups in the solution were reacted with Si–O bond and used to destroy the glass structure.

The analysis of the solution showed that the changes in Si and Mg concentration had a similar pattern to those above. The Na and Si concentration continued to increase [Fig. 4. (c), (d)], and the Ca and Mg concentration decreased sharply with flake formation [Fig. 4. (a), (b)]. We identified a relationship between flake formation, the initial pH of the solution, and consumption of Ca and Mg.

3.1.3. Inorganic elements composition in water

In order to check whether minerals contained in water affect flake formation, a certain concentration of Ca, Na, Mg, and K among minerals in water was added to deionized water and left at 90 °C for 72 h, and then the inside of glass bottle was visually observed. Among the major minerals in water, that is, Ca, Mg, Na, and K, only Ca and Mg were found to affect flakes formation for soda-lime-silica glass bottles. It was observed that flakes were formed after 24 h at a concentration of 20 mg/L for Ca, 48 h at a concentration of 0.2 mg/L for Mg, and 24 h at a concentration of 20 mg/L. It was confirmed once again that Ca and Mg in water affect flake formation

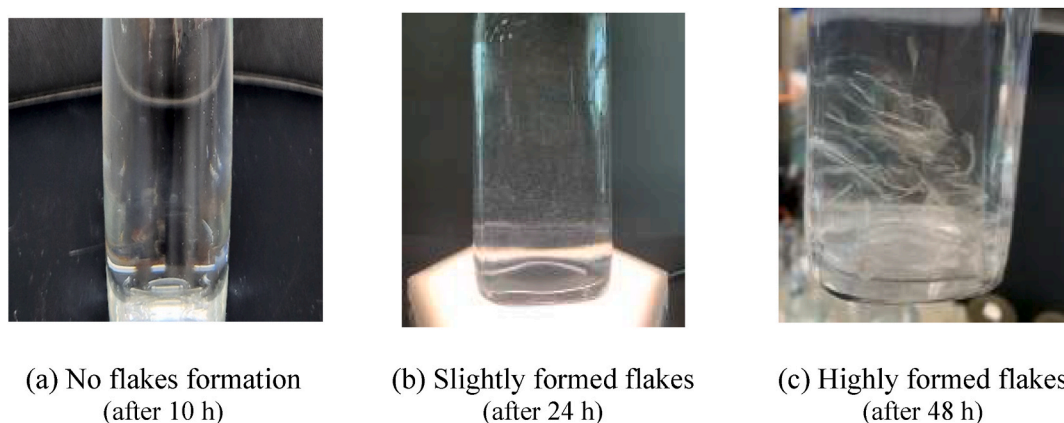


Fig. 2. The flake formation change in a glass at 90 °C in initial pH 9.

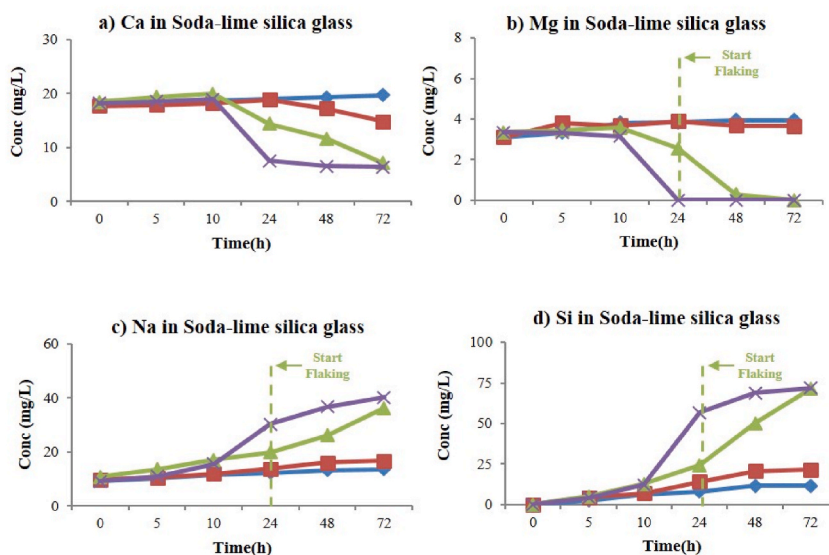


Fig. 3. Concentrations of Ca, Mg, Na, and Si in water for glass samples by contact temperature. As flake were formed, Ca and Mg decreased, and Na and Si increased: ■ 70 °C; ■ 80 °C; ■ 90 °C; ■ 100 °C.

Table 3

The flake formation according to the change of initial pH and time.

Type of glass	Time (h)	Initial pH of water									
		pH 3		pH 5		pH 7		pH 9		pH 11	
Soda-lime glass		Flaking	pH	Flaking	pH	Flaking	pH	Flaking	pH	Flaking	pH
	5	–	(3.2)	–	(6.1)	–	(8.0)	–	(9.4)	O	(10.7)
	10	–	(3.3)	–	(6.3)	–	(8.8)	–	(9.6)	O	(10.7)
	24	–	(3.5)	–	(6.4)	–	(9.2)	O	(9.5)	O	(10.6)
	48	–	(3.6)	–	(6.7)	–	(9.4)	O	(9.4)	O	(10.6)
	72	–	(4.2)	–	(7.3)	O	(9.8)	O	(9.6)	O	(10.6)
Borosilicate glasses											
	5	–	(3.2)	–	(5.7)	–	(8.4)	–	(9.2)	–	(10.6)
	10	–	(3.2)	–	(6.1)	–	(9.2)	–	(9.6)	O	(10.4)
	24	–	(3.2)	–	(6.6)	–	(9.4)	–	(9.5)	O	(10.0)
	48	–	(3.2)	–	(6.6)	–	(9.4)	–	(9.5)	O	(9.9)
	72	–	(3.2)	–	(6.6)	–	(9.3)	O	(9.5)	O	(9.8)

The flake was detected at pH 90 and 24 h in soda-lime-silica glass and at pH 11 and 24 h in borosilicate glass.

for soda-lime-silica glass. In order to determine the detailed concentration, Ca and Mg were added to deionized water up to 0.2, 1, 5, 10, 20, and 40 mg/L, respectively. Flakes were observed after 24 h when Ca was above 20 mg/L, or Mg ions were above 5 mg/L. In the case of borosilicate glass bottles, flakes were not observed up to 72 h, so it does not seem to be affected by minerals in water.

Based on these results, we conclude that flakes could be formed under specific conditions of storage temperature, initial pH, and Ca and Mg ion concentrations in water.

3.2. Flake composition

A few studies have demonstrated that the main constituent of flake was calcium silicate hydrate analyzed by energy dispersive spectroscopy (EDS) [7] and magnesium silicate hydrate analyzed by infrared spectroscopy (IR) [15]. In our study, XRF and ICP were employed to analyze flake structure and composition. As a result of XRF analysis of flakes obtained by forming flakes in a glass bottles, recovering and drying, Si, Mg, Al, and Ca were detected (Fig. 5). To confirm the content of the identified components, the flakes sample was completely melted, and analyzed by ICP-OES. When the content of each component converted into oxides the SiO₂ (47.1%) content was the highest, followed by those of MgO (18.7%), Al₂O₃ (7.3%), and CaO (5.0%), and the ignition loss was 20.3%. Thus it can be assumed that flakes are formed by the reaction of water and components that elute from the glass materials. As a result, similar to previous study [19], flakes are expected to be a mixture of hydrates of alkali ions. It means they are formed by dissolution of glass and secondary phase precipitation.

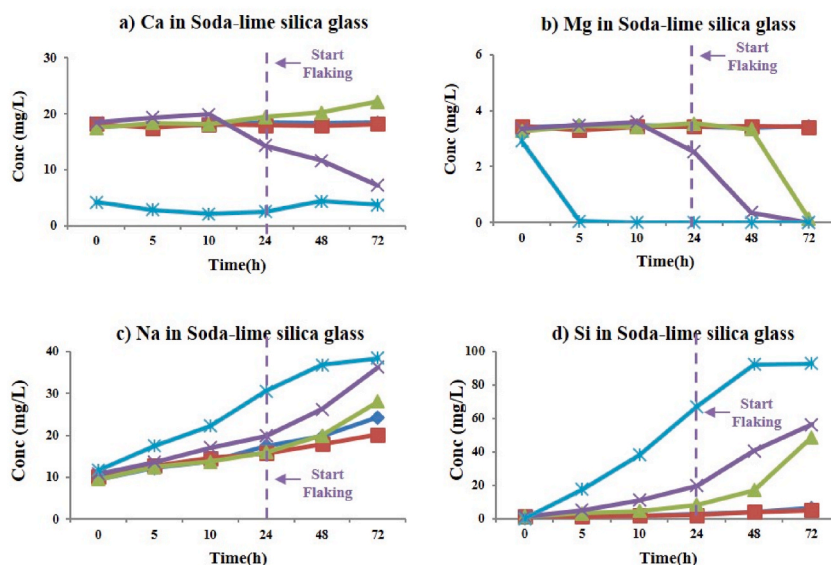


Fig. 4. Concentrations of Ca, Mg, Na, and Si in water for glass samples by initial pH of the water. As flake were formed, Ca and Mg decreased, and Na and Si increased: ■ pH 3; ■ pH 5; ■ pH 7; ■ pH 9; ■ pH 11.

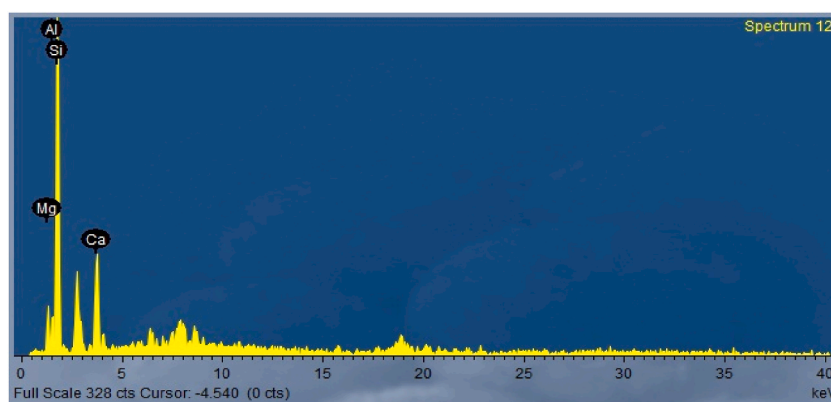


Fig. 5. Mg, Al, Si, and Ca were detected as results of XRF analysis.

4. Conclusions

This study investigated several factors that affect the formation of flake using soda-lime-silica and borosilicate glass bottles. The factors were storage temperature (from 70 °C to 100 °C), initial pH (from pH 3 to pH 11), and concentration of Ca and Mg (from 0.2 mg/L to 20 mg/L). In the soda-lime-silica glass bottle, flakes were generated at more than 90 °C, pH 9, and 20 mg/L Ca for 24 h. In the borosilicate glass bottle, flake formation occurred at more than 100 °C, pH 11 based on 24 h, and any minerals did not affect flake formation. However, when these factors were combined, flakes were formed. As flakes formation proceeds, the Na and Si concentration in the water continued to increase over time. The concentration of Ca and Mg in water decreased rapidly as flakes were produced, so that Ca and Mg might have been consumed during flake formation. In particular, this study identified the components and structure of glass flakes. Following analysis by XRF and ICP, flakes were assumed to be a mixture of hydrates of magnesium, calcium, and aluminum silicate.

Author contribution statement

Jung Eun LEE: Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Eunbee KIM, Joung Boon HWANG: Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

Jae Chun CHOI, Jong Kwon LEE: Conceived and designed the experiments.

Data availability statement

Data will be made available on request.

Additional information

No additional information is available for this paper.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported by the Korea Ministry of Food and Drug Safety [grant number 20161MFD017].

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