Determination of sulfur in soda-lime silicate glass by inductively coupled plasma atomic emission spectroscopy following separation using an alumina column

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We developed a simple, rapid, and accurate method to determine the quantity of sulfur in soda-lime silicate glass using inductively coupled plasma atomic emission spectroscopy (ICP-AES) and sulfur separation using an alumina column. This method requires a smaller number of samples and takes less time for the analysis than conventional methods. The samples of glass were decomposed using hydrofluoric acid, perchorlic acid, and an oxidizing agent. For the amber glass samples, the oxidizing agent used was potassium permanganate. The decomposed solution was diluted with perchorlic acid. The solution was passed through an alumina column to enable sulfur adsorption on the column. To desorb sulfur from the column, diluted ammonia was passed through it after rinsing it with diluted perchorlic acid. ICP-AES was used to determine the quantity of sulfur in the ammonia eluent. The method was validated using three certified glass samples. The sulfur quantities determined by this method were within the certified values. The relative standard deviation of the determined values was less than 2 %.

Key-words : Glass, Sulfur, Separation, Determination, Amber

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1. Introduction

Soda-lime silicate glass is the most widely used glass in the world and is the leader in the world’s glass market. The industrial production of glass involves batch preparation, melting, refining, forming, annealing, and post processes.¹) To obtain a high-quality glass product, the number of bubbles remaining in the final product should be reduced. Fining and refining are, therefore, important in the industrial production of glass. The fining agents reduce the bubbles in the glass. One of the commonly used fining agents in the production of soda-lime silicate glass is sodium sulfate (Na₂SO₄). The melting,²⁻⁵) and fining and refining⁶⁻¹⁸) reactions of soda-lime silicate glass were investigated. In oxidized melts, the main sulfate fining mechanism is thermal sulfate decomposition. It typically occurs between 1430 and 1480 °C¹⁷,¹⁸) as follows:

\[ \text{Na}_2\text{SO}_4 \rightarrow \text{Na}_2\text{O} + \text{SO}_2 + 1/2\text{O}_2 \]

In reduced melts, which contain cokes, CO, and organic contaminants, some sulfates react to produce sulfides as follows:

\[ 2\text{C} + \text{Na}_2\text{SO}_4 \rightarrow 2\text{CO}_2 + \text{Na}_2\text{S} \]
\[ 4\text{CO} + 2\text{Na}_2\text{SO}_4 \rightarrow 4\text{CO}_2 + 2\text{Na}_2\text{S} \]
\[ 4\text{C} + 2\text{Na}_2\text{SO}_4 \rightarrow 4\text{CO} + 2\text{Na}_2\text{S} \]

The sulfide formation typically occurs between 700 and 800 °C⁹,¹⁸) and the formed sulfides react with the remaining sulfates. Fining typically occurs between 1000 and 1350 °C as follows:

\[ \text{S}^{2-} + 3\text{SO}_4^{2-} \rightarrow 4\text{SO}_2 + 4\text{O}^{2-} \]

During annealing, which follows fining, fining gases, such as O₂ and SO₂, are chemically absorbed into the melt, and the process is called refining. Fining and refining reduce the number of bubbles in the final product.

Determining the quantity of sulfur in the final product is important to understand the refining and refining reactions. Sulfur is present in glass as sulfate and sulfide.¹⁹,²⁰) The total quantity of sulfur, consisting of sulfate and sulfide, also has to be determined, because sulfides can remain in a final product, such as amber glass.²¹⁻²⁴)

Several methods are already available to determine the quantity of sulfur in glass. These methods can be categorized into two types. One type decomposes the glass samples to determine the sulfur quantity in the decomposed solutions. An example of this type is the gravimetric
analysis of barium sulfate. The other type expels sulfur in the form of hydrogen sulfide and determines the sulfur quantity in the sample by trapping. These methods include distilled separation – methylene blue spectrophotometric method, Tin(II)-strong phosphoric acid decomposition - iodometric titration, Tin(II)-strong phosphoric acid decomposition – iodate titration, reduction-distillation - hydroxymercuribenzoate titration, hydrophosphorous acid – hydroiodic acid reduction, and sodium iodide – hypohosphorous acid reduction. However, these methods require technical skills and a large number of samples. Some methods even require special apparatuses to decompose glass samples and take a long time for the analysis. For example, the conventional gravimetric method requires a 3 g sample and at least 2 days for the analysis. The sodium iodide – hydroiodic acid – hypohosphorous acid reduction is unsafe because the samples have to be heated in a closed flask. Isotope dilution high-resolution inductively coupled plasma mass spectrometry (ICP-MS) equipped with a flow injection system is a simple and accurate method. It can determine even low levels of sulfur using only a small number of samples but requires high-resolution ICP-MS, and thus is still not used widely. X-ray fluorescence is used in the daily measurements at glass production sites. However, it requires standard materials with accurate quantities of sulfur that require calibration through other methods.

A simple, rapid, and accurate method is, therefore, required to determine the sulfur quantity in soda-lime silicate glass using a smaller number of glass samples. The use of commonly accepted equipment to decompose the glass samples and determine their sulfur quantity is preferable. Many elements in glass are determined using decomposition methods.

Separating the sulfur before determining its quantity is also preferable. The sulfur separation prevents interference by glass matrix elements and improves the sensitivity of the sulfur quantity determination. Chromatographic separation using an alumina column with the gravimetric analysis of barium sulfate was used to determine the sulfur quantity in iron and steel. This separation procedure was simple and rapid. However, a high blank value of sulfur (approximately 50 µg/g) was observed owing to the large alumina column and the large amount of agents used. We developed our method by improving the method already available to reduce the blank value of sulfur. In our method, the glass samples were decomposed using hydrofluoric acid, perchloric acid, and an oxidizing agent, and thereafter the sulfur in the glass matrix elements were separated using a small high-purity alumina column. The separation procedure, which is simple and speedy, can simultaneously treat up to 10 samples.

The method we propose determines the sulfur content of glass samples containing sulfide. An oxidizing agent has to be added to the glass samples in the decomposition procedure. In the absence of an oxidizing agent, sulfides evaporate in the form of hydrogen sulfide during the sample decomposition. Br₂ is a widely used oxidizing agent. However, to determine the sulfur content in glass using Br₂, additional apparatuses are required, and the decomposing reaction with cold water has to be slow. In this study, hydrogen peroxide and potassium permanganate were examined for their suitability as oxidizing agents that would reduce the decomposition time of the samples. Potassium permanganate is a typical oxidizing agent used to determine the arsenic content in glass. During the decomposition of the glass samples, potassium permanganate oxidizes trivalent arsenic to form pentavalent arsenic. Potassium permanganate prevents the evaporation of trivalent arsenic during the decomposition of the samples. We could confirm through this study that potassium permanganate is required to determine the total quantity of sulfur in amber glass to ensure that the determined value of sulfur is not lower than its certified value.

Inductively coupled plasma atomic emission spectroscopy (ICP-AES) was used to determine the sulfur quantity in the samples. ICP-AES, which is used widely, requires one hour for the process, which includes the time taken for equipment setting up and shutting down, standard solution measurements, and calculations performed to determine the quantity of sulfur in the samples.

2. Experimental procedure

2.1 Materials

Three certified reference soda-lime silicate glass samples were used in the experiment: the three glass types were R-1 (Japan Standard Sample Committee, Tokyo, Japan), SGT10 (Society of Glass Technology, Sheffield, United Kingdom), and SGT11 (Society of Glass Technology, Sheffield, United Kingdom). Table 1 lists the compositions of the three types of glass. R-1, SGT10, and SGT11 represent clear, amber, and green glass, respectively. Values in parentheses for SO₃ indicate 95% confidence intervals.

2.2 Apparatus

A 100 mL polytetrafluoroethylene (PTFE) dish was used as the decomposition container. An alumina column,

| Composition (wt%) |
|-------------------|
|                  |
| R-1 (Clear)       |
| SGT10 (Amber)     |
| SGT11 (Green)     |
| SiO₂ 72.2         |
| Al₂O₃ 1.75        |
| Na₂O 13.8         |
| K₂O 0.84          |
| MgO 4.01          |
| CaO 6.72          |
| BaO —             |
| Fe₂O₃ 0.08        |
| TiO₂ 0.03         |
| Cr₂O₃ —           |
| SO₃ 0.20          |
|                  |
|                  |
|                  |
|                  |
|                  |

Table 1. Certified compositions of the samples
InterSep AL-N Al₂O₃ 500 mg/2mL (GL Sciences Inc., Tokyo, Japan), placed on a vacuum filtration kit (GL Sciences Inc., Tokyo, Japan) was used to separate the sulfur in the samples. An inductively coupled plasma atomic emission spectrometer, SP5520 (Hitachi High-Tech Corporation, Tokyo, Japan) was used to determine the sulfur quantity and other glass matrix elements in the glass samples. An agate mortar and pestle were used to grind the samples. A 50 mL volumetric flask and a 100 mL volumetric flask were used.

2.3 Agents and chemicals

Hydrofluoric acid (50 wt%), perchloric acid (60 wt%), ammonia solution (25 wt%) of atomic absorption spectrometry grade (Kanto Chemical Co., Inc., Tokyo, Japan), hydrogen peroxide (30 wt%) of special grade (Kanto Chemical Co., Inc., Tokyo, Japan), and 0.1 mol/L of the potassium permanganate solution of reagent grade (Kanto Chemical Co., Inc., Tokyo, Japan) were used. The standard solutions used for the ICP-AES analysis were sulfate ion standard solution (1 g SO₄²⁻/L) of ion chromatography grade (Kanto Chemical Co., Inc., Tokyo, Japan), aluminum standard solution (1 g Al/L), sodium standard solution (1 g Na/L), potassium standard solution (1 g K/L), magnesium standard solution (1 g Mg/L), calcium standard solution (1 g Ca/L), iron standard solution (1 g Fe/L), titanium standard solution (1 g Ti/L), and chromium standard solution (1 g Cr/L) of atomic absorption spectrometry grade (Kanto Chemical Co., Inc., Tokyo, Japan). Deionized water was used throughout the experiments.

2.4 Procedures

2.4.1 Decomposition procedure of R-1 and SGT11 samples

Glass samples were dried in an oven for 2 h at 110 °C after they were ground with a mortar and pestle. Each ground sample (0.1 to 0.3 g in weight) was accurately weighed in a PTFE dish and moistened with 1 mL of water. Some samples contained reduced sulfur, and 1 mL of 30 wt% hydrogen peroxide was added to the dish to facilitate the strong oxidization of reduced sulfur present in SGT10 samples, which were made of amber glass. Without strong oxidization, reduced sulfur is converted into hydrogen sulfide and evaporated during the decomposition of the samples. Manganese dioxide precipitated after the decomposition of the samples because of the reduction of potassium permanganate. The precipitated manganese dioxide was dissolved by adding 0.5 mL of 3 wt% hydrogen peroxide, which served as a reducing agent. Figure 1 demonstrates the decomposition procedure of samples.

2.4.2 Decomposition procedure of SGT10 samples

Decomposition procedure of SGT10 samples was similar to that of R-1/SGT11 samples except for the use of an oxidizing agent. Instead of hydrogen peroxide, potassium permanganate (2.5 mL of 0.1 mol/L) was added to the dish to facilitate the strong oxidization of reduced sulfur present in SGT10 samples, which were made of amber glass. Without strong oxidization, reduced sulfur is converted into hydrogen sulfide and evaporated during the decomposition of the samples. Manganese dioxide precipitated after the decomposition of the samples because of the reduction of potassium permanganate. The precipitated manganese dioxide was dissolved by adding 0.5 mL of 3 wt% hydrogen peroxide, which served as a reducing agent.

2.4.3 Separation procedure of sulfur in the samples using an alumina column

For washing, 20 mL of 0.7 mol/L NH₄OH and 10 mL of water were passed through an alumina column at a flow rate of 20 mL/min using a vacuum kit. For conditioning, 10 mL of 1.8 mol/L HClO₄ was passed through the column. The decomposed sample solution was passed through the column at a flow rate of 2 mL/min to enable the column to absorb the sulfur in the solution. To remove the glass matrix elements in the solution, 20 mL of 1.8 mol/L HClO₄ and 20 mL of water were passed through the column at a flow rate of 20 mL/min. To desorb sulfur from the column, 10 mL of 0.7 mol/L NH₄OH was passed through the column at a flow rate of 2 mL/min. The eluent was collected in a 50 mL volumetric flask and diluted with 0.7 mol/L NH₄OH until its volume was 50 mL. The obtained solution is called the measurement sample solution. The quantity of sulfur in the solution was determined as explained in section 2.4.5.

2.4.4 Recovery of the sulfur in the standard solution

The recovery of sulfur was made using the procedure explained in section 2.4.3. A standard solution of sulfur diluted with perchloric acid was introduced in place of the decomposed sample solution (section 2.4.3). The standard solution contained 5 mL of HClO₄ and 10 mL of HF were also added to the dish. The dish was placed on a hot plate and heated until perchloric acid fumes were observed. After the dish had cooled, 5 mL of HF was added to the dish. The dish was heated again until perchloric acid fumes were observed. Silicon, which is the main element in glass, was volatilized as silicon fluoride during decomposition. After the dish had cooled, the decomposed solution in the dish was transferred to a 50 mL beaker. The solution was thereafter diluted to approximately 25 mL using 1.8 mol/L HClO₄ to form a decomposed sample solution. This decomposed sample solution contained sulfur and other elements that were in the glass.

2.4.2 Decomposition procedure of SGT10 samples

Decomposition procedure of SGT10 samples was similar to that of R-1/SGT11 samples except for the use of an oxidizing agent. Instead of hydrogen peroxide, potassium permanganate (2.5 mL of 0.1 mol/L) was added to the dish to facilitate the strong oxidization of reduced sulfur present in SGT10 samples, which were made of amber glass. Without strong oxidization, reduced sulfur is converted into hydrogen sulfide and evaporated during the decomposition of the samples. Manganese dioxide precipitated after the decomposition of the samples because of the reduction of potassium permanganate. The precipitated manganese dioxide was dissolved by adding 0.5 mL of 3 wt% hydrogen peroxide, which served as a reducing agent. Figure 1 demonstrates the decomposition procedure of samples.
solutions used were 0.1 to 4.5 mol/L perchloric acid containing 100 µg of sulfur, and 1.8 mol/L perchloric acid containing 5 to 1000 µg of sulfur. After completing the procedure explained in section 2.4.3, the sulfur quantity in the measurement sample solution was determined as explained in section 2.4.5. The recovery of sulfur is defined as the ratio of the quantity of sulfur determined to the quantity of sulfur introduced. The repeatability of the sulfur recovery was checked using 1.8 mol/L perchloric acid containing 100 µg of sulfur. The experiment was repeated six times using different alumina columns. It was repeated 10 times using the same alumina column to determine whether a column could be used several times.

2.4.5 Determination of the sulfur and glass matrix element quantities present in the measurement sample solution

The quantities of sulfur and glass matrix elements present in the measurement sample solution were determined using ICP-AES. The quantities of glass matrix elements were determined using a 0.3 g SGT11 sample. The removal ratio of each glass matrix element was calculated by subtracting the ratio of the quantity of each element determined to its theoretical quantity. The theoretical quantities of glass matrix elements in the 0.3 g SGT11 sample were calculated using the certified composition listed in Table 1. Table 2 lists the instrumental conditions of ICP-AES. The sulfur quantity in a glass sample was calculated as sulfur trioxide.

2.4.6 Blank test procedure

A blank test was conducted throughout the entire procedure consisting of the decomposition procedure, separation procedure, and determination procedure explained in sections 2.4.1, 2.4.3, and 2.4.5, respectively. The test was repeated 16 times to calculate the mean blank value, standard deviation, detection limit, and quantification limit of sulfur in the samples. The lower limit of detection is defined as the mean of the blank plus three standard deviations. The lower limit of quantification is defined as the mean of the blank plus 10 standard deviations.

3. Results and discussion

3.1 Study of the separation procedure using an alumina column

Figure 2 displays the recovery of sulfur using various perchloric acid concentrations. The recovery was almost 100% for 0.1 to 4.5 mol/L of perchloric acid. A wide range of perchloric acid concentrations displayed a good recovery. We chose 1.8 mol/L of perchloric acid to dilute the decomposed solution. Figure 3 displays the recovery of sulfur from 5 to 1000 µg. The recovery was between 95 and 102%. Sulfur in the range of 5 to 30 µg showed a relatively variable recovery. The reason for this variable recovery can be attributed to procedural or measurement errors caused by the small quantity of sulfur present in the samples. The average recovery, which was repeated six times using different alumina columns, was 99.1%. The standard deviation was 0.25%. The sulfur quantity in the glass samples has to be between 100 and 1000 µg for the sulfur recovery to exceed 98%. The average recovery, which was repeated ten times using the same alumina column, was 98.4% and the standard deviation was 0.33%. Thus, an alumina column can be used more than once.

3.2 Removal of glass matrix elements using an alumina column

Table 3 shows the removal ratios of glass matrix elements when a 0.3 g SGT10 sample was used. The removal ratios were over 99.9%, except for aluminum. Aluminum has to be an eluent from the alumina column because it was detected even in the blank test. Glass matrix elements were removed by the separation procedure using the alumina column and sulfur was successfully separated from the glass matrix elements.

Table 2. Instrumental conditions of ICP-AES

| Instrumental Condition | Condition       |
|------------------------|-----------------|
| RF power               | 1.2 kW          |
| Plasma gas flow        | 15.0 L/min      |
| Auxiliary gas flow     | 1.5 L/min       |
| Nebulizer gas pressure | 0.75 MPa        |
| Emission line          |                 |
| S: 180.669 nm          |                 |
| Al: 167.019 nm         |                 |
| Na: 589.592 nm         |                 |
| K: 766.491 nm          |                 |
| Mg: 279.553 nm         |                 |
| Ca: 396.847 nm         |                 |
| Fe: 259.94 nm          |                 |
| Ti: 334.941 nm         |                 |
| Cr: 205.56 nm          |                 |

Fig. 2. Recovery of 100 µg of sulfur using various perchloric acid concentrations and a separation procedure using an alumina column.

Fig. 3. Recovery of 5 to 1000 µg of sulfur with 1.8 mol/L perchloric acid and a separation procedure using an alumina column.
Table 3. Quantities of glass matrix elements present in the measurement solution, using a 0.3 g of SGT11 sample

| Matrix element quantity in the glass sample (µg) | Determined value (µg) | Removal ratio (%) |
|-------------------------------------------------|----------------------|-----------------|
| Al                                              | 2900                 | 150             | 95               |
| Na                                              | 30000                | <0.1            | >99.9            |
| K                                               | 1700                 | <0.1            | >99.9            |
| Mg                                              | 3900                 | <0.1            | >99.9            |
| Ca                                              | 22000                | <0.1            | >99.9            |
| Fe                                              | 720                  | <0.1            | >99.9            |
| Ti                                              | 120                  | <0.1            | >99.9            |
| Cr                                              | 420                  | <0.1            | >99.9            |

3.3 Determination of the sulfur quantities in R-1 and SGT11 samples

The sulfur quantities in R-1 and SGT11 samples were determined using the procedures explained in sections 2.4.1, 2.4.3 and 2.4.5. Table 4 lists the sulfur quantity determined in the form of oxides. The quantities determined were within the certified values. The relative standard deviations were all within 2%. The developed procedures shown in sections 2.4.1, 2.4.3 and 2.4.5 were validated to determine the sulfur quantities in R-1 and SGT11 samples accurately.

It took 4–5 h to complete the entire procedure; 2–3 h to decompose a glass sample, 1 h to separate sulfur in a decomposed sample solution using an alumina column, and 1 h to determine the quantity of sulfur in the measurement sample solution using ICP-AES. The procedure can use up to 10 samples simultaneously. The separation procedure using an alumina column is simple. Even any newcomer can easily learn and use the procedure.

3.4 Determination of the sulfur quantity in SGT10 samples

Amber soda-lime silicate glass contains reduced sulfur, which during decomposition of a sample is converted into hydrogen sulfide and evaporated. Hydrogen peroxide and potassium permanganate were compared to determine the oxidizing agent to be used. A procedure was also implemented without an oxidizing agent. Table 5 lists the results of determined quantities of sulfur in SGT10 samples as oxides. With no oxidizing agent, the sulfur quantity was below the lower quantification limit. With 1 mL of 30 wt % hydrogen peroxide, the determined sulfur quantity was lower than the certified value. With 2.5 mL of 0.1 mol/L potassium permanganate, the determined sulfur quantity was within the certified value.

Without oxidizing agents, most of the sulfur in the samples is evaporated as H₂S because reduced sulfur is present in glass in the form of sulfide, as mentioned in XANES studies. Hydrogen peroxide was not sufficient to oxidize the sulfide in SGT10 samples during their decomposition. Hydrogen peroxide allowed the evaporation of some of the reduced sulfur as H₂S. Potassium permanganate properly oxidized the sulfide in the SGT10 samples during their decomposition and succeeded in preventing the sulfur from evaporating during the decomposition of the samples. The process took almost the same time as indicated in section 3.3. Precipitated manganese dioxide requires approximately 10 min to dissolve. Unlike the method that uses Br₂ as an oxidizing agent, the method using potassium permanganate as the oxidizing agent does not require any additional apparatus or a decomposition procedure.

Table 5. Quantities of SO₃ present in SGT10 (amber) samples with and without the oxidizing agents

| Oxidizing agent | SO₃ (wt%) |
|-----------------|----------|
| Without an oxidizing agent | 30% H₂O₂ | 0.1 mol/L KMnO₄ |
| N = 1           | <0.002   | 0.037             | 0.053             |
| N = 2           | <0.002   | 0.039             | 0.054             |
| Avg.            | <0.002   | 0.038             | 0.053             |

3.5 Results of the blank test

The mean blank value of sulfur was 0.49 µg. The standard deviation was 0.25 µg. The limits of detection and quantification were 1.3 and 3.0 µg, respectively. The limit of quantification is low enough to determine the sulfur quantity in soda-lime silicate glass because soda-lime silicate glass contains over 100 µg of sulfur.

The mean blank value of sulfur, 0.49 µg, obtained in this study is much lower than the value obtained in a previous study that used a large alumina column and the large amount of agents. The higher blank value obtained in this previous study would have been due to the small quantities of sulfur contained in the large alumina column and the agents. In our study, we reduced the blank value of sulfur with a decomposition procedure that used hydrofluoric acid and perchloric acid, and a separation procedure that used a small and high-purity alumina column. However, the mean blank value of sulfur obtained in the study, 0.49 µg, is 10 times higher than the value obtained in the study done by Makishima, who purified the agents for decomposition and used two charcoal filters in a flow injection system to reduce sulfur contamination. These processes are not required in the method we used because our limit of quantification of sulfur is low enough to determine the quantity of sulfur in soda-lime silicate glass as mentioned above.
4. Conclusion

A simple, rapid, and accurate method to determine the sulfur quantity in soda-lime silicate glass was investigated. Glass samples were decomposed using hydrofluoric acid, perchloric acid, and an oxidizing agent. Potassium permanganate was used as the oxidizing agent with glass samples containing reduced sulfur, such as those made of amber glass. The decomposed solution was diluted using perchloric acid. The sulfur in the decomposed solution was adsorbed on the alumina column and was separated from the glass matrix elements by rinsing with diluted perchloric acid. The sulfur was desorbed from the column by passing diluted ammonia through the column. ICP-AES was used to determine the sulfur quantity in the ammonia eluent. The method requires approximately 0.1 to 0.3 g glass samples depending on the sulfur quantity in the glass. The separation procedure with the alumina column is simple and rapid: it takes only 1 h, and it can simultaneously treat up to 10 samples. The method was validated using three certified glass samples. The sulfur values determined are within the certified values. The relative standard deviations of the sulfur values were less than 2%. This method does not require any special apparatus to decompose a glass sample.

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