

Determination of Sodium and Aluminum in the Glass Sample by Neutron Activation Technique

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Abstract- Elemental analysis of sodium and aluminum in glass sample with reference to IAEA standard has been determined simultaneously by using neutron activation and off-line gamma ray spectrometric technique. The amount of sodium and aluminum was also calculated from the composition of the glass sample quoted by the supplier and compared with the experimental value in the present work. It was found that the experimental value for sodium is in good agreement with the theoretical values. However, in the case of aluminum, the experimental value is around seven times higher than the theoretical value. Reason for this discrepancy has been investigated.

Keywords- Neutron Activation Analysis; off-Line Gamma Ray Spectrometry; Glass Sample; Sodium and Aluminum Estimation

I. INTRODUCTION

Different types of glasses are available based on their uses. The types of glasses are based on the percentage of SiO₂ and oxides of other elements like Na, K, Mg, Ca, Li, B and so on. As for example borosilicate glass has the composition of 60% SiO₂, 20% B₂O₃, 5% Na₂O, 5% Li₂O, 2% K₂O. Pyrex glass has the compositions of 80.6% SiO₂, 13% B₂O₃, 4% Na₂O, 2.3% Al₂O₃ and 0.1% other trace element oxide. Similarly, lead glass has the composition of 62.9% SiO₂, 13.6% B₂O₃, 2.6% Al₂O₃, 10.3% MgO, 2.1% ZrO₂ and 8.5% PbO. Different constituents of glasses of different types are important from the point of their applications in various fields. As for example Pyrex glass is used in chemical environment in chemistry and biochemistry laboratory, lead glass in radioactive fields and float glass for Resistive Plate Chambers (RPCs) to be deployed in 50 kilo ton Indian Neutrino Observatory (INO) detector (Bhuyan, M. et al., 2012). Thus it is important to estimate the different elements in a glass sample in quantitative way. Various destructive methods such as gravimetric method (Hillebrand, W.F. et al. 1929, Wells, R.C. et al. 1934) flame spectrometry (Broderick, E J. et al. 1951, Williams, J.P., et al. 1954, Roy, N. 1956), and x-ray microanalysis (Roe, M. et al., 2006, Newbury, Dale. E. 2002) are used to estimate the composition of glass. In gravimetric method, glass has to be dissolved first and chemical separation of different element

is usually done. Then the element of interest has to be taken into a particular chemical composition for gravimetric analysis (Hillebrand, W.F. et al. 1929, Wells, R.C. et al. 1934). In the case of flame spectrometry (Broderick, E J. et al. 1951, Williams, J.P., et al. 1954, Roy, N. 1956) also, glass has to be dissolved and the element of interest has to be separated. Then flame spectro-photometry method has to be used for the analysis of various alkali earth metals. The above two methods are destructive and thus are time consuming. In x-ray micro analysis method (Roe, M. et al., 2006, Newbury, Dale E., 2002), the estimation of different element of glass to be estimated has to be compared with a standard for its estimation. On the other hand, non-destructive method such as neutron activation analysis has been also used by various authors (Corless, J. T. 1966, De Soete, D., 1972, Penev et.al., 1985) for the estimation of different elements of glass samples such as Si, Na, K, Al, Mg and Ca etc. Similarly, other non-destructive technique such as photon activation analysis (Kanda, Y. et al., 1980) was also used for the estimation of multi-elements in glass sample. The neutron and photon activation technique is not only non-destructive but also less time-consuming. Among these two techniques, neutron activation analysis is used by Penev et. al., 1985 for the estimation of various elements in samples other than glass like rock and pottery. In the present work, we have determined sodium and aluminum in the float glass sample using neutron activation and off-line gamma ray spectrometric technique.

II. EXPERIMENTAL METHOD

About 49.42 mg of glass sample and 44.96 mg of IAEA standard samples were separately sealed in alkathene bags. They were additionally sealed together in another alkathene bag and mounted into a irradiation capsule. The sealed sample was irradiated for one minute at a neutron flux of $5 \times 10^{13} \text{ ncm}^{-2}\text{s}^{-1}$ using pneumatic carrier facility of the heavy water moderated natural uranium fueled reactor DHRUVA (Reddy, A.V.R. et al., 2007). Separate irradiations were done for different samples. After irradiation the samples were open and the alkathene covers were cut open to take out the

samples. Then the irradiated glass sample and IAEA standard were mounted separately in two different perspex plates without alkathene covers and taken for gamma-ray spectrometry. Thus, the chemical composition and weight of alkathene covers are not relevant for the gamma ray spectrometry. The direct gamma-ray counting of the irradiated samples was done using energy and efficiency calibrated 120 cm³ HPGe detector coupled to a PC based 4096 channel analyzer. The resolution of the detector system was 2.0 keV at 1332.0 keV of ⁶⁰Co. The gamma-ray counting was done in live time mode and the dead time of the counting was kept less than 10% by placing the sample at a suitable distance of 5 cm away from the detector to avoid pile up effect. Both the irradiated glass sample and standard are counted alternatively. In the beginning number of short counting of the irradiated glass sample and standard was done alternately for 300 sec to follow the decay of short-lived product ²⁸Al. Then long counting of few hours for the glass sample and standard was also done alternatively. Several sets of counting for irradiated glass sample and standard were done. A typical γ -ray spectrum of the irradiated glass sample from short and long counting is shown in Fig. 1 and Fig. 2, respectively. From Fig. 1, the gamma-ray energy of ²⁸Al was clearly seen. In Fig. 2, the gamma-ray energy of ²⁸Al is absent but gamma-ray energy of ²⁴Na is clearly visible.

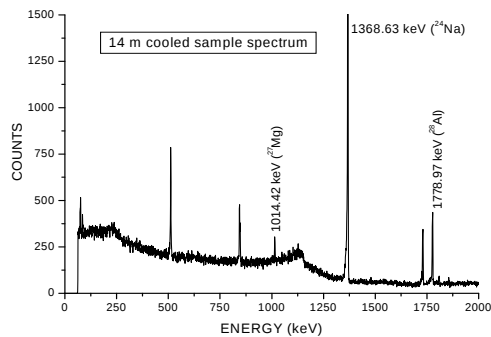


Fig. 1 Gamma ray spectrum of irradiated glass sample showing the γ -lines of ²⁴Na, ²⁷Mg and ²⁸Al

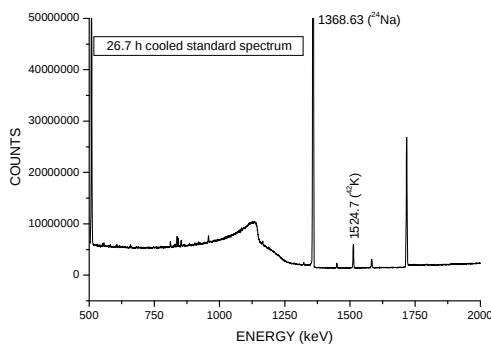


Fig. 2 Gamma ray spectrum of irradiated IAEA standard showing the γ -lines of ²⁴Na and ⁴²K

III. CALCULATION AND RESULTS

The photo peak areas of different gamma-rays of reaction products of interest of nuclide such as ²⁴Na and ²⁸Al were calculated by subtracting the linear Compton background from their total net peak areas. The number of detected gamma-rays (A_{obs}) under the photo peak of individual nuclide is related to the (n, γ) reaction cross-section (σ_R) as (Naik, H. et al., 2011)

$$A_{\text{obs}}(\text{CL/LT}) = n\sigma_R \phi I_\gamma \varepsilon b(1 - e^{-\lambda t}) e^{-\lambda T}(1 - e^{-\lambda \text{CL}})/\lambda \quad (1)$$

Where n is the number of target atoms with isotopic abundance b and ϕ is the neutron flux. The measurands t and T are the irradiating time and cooling time, whereas CL and LT are the real time and life time respectively. " λ " is the decay constant of the isotope of interest and I_γ is the abundance or branching intensity of the chosen γ -rays of the reaction products. The γ -rays energies and nuclear spectroscopic data such as half-lives and branching intensity of the reaction products are taken from references (Firestone, R.B. et al., 2004, Browne, E. et al., 1986, Blachot, J., 1981) and are shown in Table 1.

TABLE I NUCLEAR SPECTROSCOPIC DATA FROM REFS. (FIRESTONE, R.B. ET AL., 2004, BROWNE, E. ET AL., 1986, BLACHOT, J., 1981)

Nuclide	Isotopic abundance (%)	(n, γ) product of nuclide	Half-life (keV)	γ -ray energy (keV)	γ -ray abundance (%)
²⁷ Al	100	²⁸ Al	2.2414 m	1778.97	100
²³ Na	100	²⁴ Na	14.959 h	1368.63	100

The detector efficiency " ε " for the γ -ray energy at a fixed geometry was calculated as follows

$$\text{Ln}\varepsilon = \sum C_n \text{Ln}E \quad (2)$$

where C_n represents the fitting parameters and E is the γ -ray energies for ¹⁵²Eu standard source used in the present work. Since the γ -ray energies of ¹⁵²Eu are within 121.8 keV to 1408.0 keV, three parameters in Eq. (2) have been used for efficiency determination.

The numbers of detected γ -rays under the photo-peak of nuclide of the irradiated sample (A_{sample}) and the IAEA standard (A_{std}) are related as

$$\begin{aligned} \frac{A_{\text{sample}}}{A_{\text{std}}} &= \frac{n_{\text{sample}} \sigma_R \phi I_\gamma \varepsilon b(1 - e^{-\lambda t}) e^{-\lambda T(\text{sample})} (1 - e^{-\lambda \text{CL}(\text{sample})})/\lambda}{n_{\text{std}} \sigma_R \phi I_\gamma \varepsilon b(1 - e^{-\lambda t}) e^{-\lambda T(\text{std})} (1 - e^{-\lambda \text{CL}(\text{std})})/\lambda} \\ &= \frac{n_{\text{sample}} e^{-\lambda T(\text{sample})} (1 - e^{-\lambda \text{CL}(\text{sample})})}{n_{\text{std}} e^{-\lambda T(\text{std})} (1 - e^{-\lambda \text{CL}(\text{std})})} \\ n_{\text{sample}} &= \frac{n_{\text{std}} A_{\text{sample}} e^{\lambda T(\text{sample})} (1 - e^{-\lambda \text{CL}(\text{std})})}{A_{\text{std}} e^{\lambda T(\text{std})} (1 - e^{-\lambda \text{CL}(\text{sample})})} \end{aligned} \quad (3)$$

where n_{sample} and n_{std} are the amount of element in the sample and IAEA standard. The n_{std} for 44.96 mg of IAEA standard sample was obtained by using the Eq. (4)

$$n_{\text{std}} = \text{Conc. of the element in IAEA standard} \times \text{Wt. of the standard} \quad (4)$$

where Wt. is the weight of the IAEA standard mentioned above. Conc. is the concentration of the sodium and aluminum in mg/Kg of IAEA standard, which are shown in Table 2.

TABLE II ENVIRONMENTAL REFERENCE MATERIALS MEASURED FOR TRACE ELEMENT AS SORTED BY MANUFACTURER CODE

MEASURED	CONC mg/kg
Al	24500
Na	6690

After knowing the value of n_{std} , the amount (n_{sample}) of particular element (Na and Al) in the sample per 100 g of glass was determined using Eq. (3) and is given in the second column of Table 3.

TABLE III COMPARISON OF EXPERIMENTALLY VALUE OF NA AND AL WITH THE THEORETICAL VALUE

Element	Experimental (g in 100g of glass)	Manufacturer specification (g in 100g of glass)
Na	10.838±0.070	10.0657
Al	0.48301±0.02325	0.07176

The amount of Na and Al in the float glass sample was also calculated using manufacturer specification given in molar percentages and standard molar to weight conversion factor (Table 4). The value of Na and Al calculated from manufacturer specification using above technique is given in third column of the Table 3 for comparison.

TABLE IV FLOAT GLASS AND TYPICAL CONTAINER GLASS COMPOSITION, MOL% AND CONVERSION FACTORS

	Float Glass	Typical	Mol%	Wt%
	Container glass			
Al ₂ O ₃	0.08	0.75	0.59	1.0
Na ₂ O	13.13	12.9	12.58	13.0

IV. RESULT AND DISCUSSION

The amount of Na and Al per gram of sample obtained experimentally in the present work based on neutron

activation and off-line gamma-ray spectrometric technique is given in second column of Table 3. The uncertainties shown in the measured value are the precision from two determinations. The overall uncertainty represents contribution from both random and systematic errors. The random error in the observed activity is due to counting statistics and is estimated to be 5-10 % which can be determined by accumulating the data for an optimum time period that depends on the half line of nuclide of interest. On the other hand, the systematic errors are due to uncertainties in the irradiation time (~2%), the detection efficiency calibration (~3%), the half life of nuclides of interest and the γ -ray abundance (~1%), which are the largest variation in the literature (Firestone, R.B. et al., 2004, Browne, E. et al., 1986, Blachot, J., 1981). The overall systematic error is about 4%. An upper limit of 6.5% -11% has been carried out for the amount of Na and Al in the glass sample based on 5-10% random error and 4% systematic error.

In order to compare the present estimate, amount of Na and Al in the glass sample was calculated from the manufacturer specification given in molar percentage (Table 4). In the calculation, the standard molar to weight conversion factor has been taken care using the Table 4. The calculated value of Na and Al is given in the third column of the Table 3 for comparison. It can be seen from Table 3 that the experimental values for Na are in good agreement with the calculated value, which shows the correctness of the present approach. However, in the case of Al, the experimental value is seven times higher than the calculated value based on manufacturer specification.

During the irradiation, the radioactive ^{24}Na is usually formed from the following reactions (Penev et al., 1985)

a (%)	Reactions	E_{th} (MeV)	σ (mb)
100	$^{23}\text{Na}(n, \gamma) ^{24}\text{Na}$	-	40
78.99	$^{24}\text{Mg}(n, p) ^{24}\text{Na}$	4.0	200
100	$^{27}\text{Al}(n, \alpha) ^{24}\text{Na}$	3.249	100

Similarly in the case of Al the radioactive nuclide ^{28}Al is usually formed from the reaction

a (%)	Reactions	E_{th} (MeV)	σ (mb)
100	$^{27}\text{Al}(n, \gamma) ^{28}\text{Al}$	-	230
92.23	$^{28}\text{Si}(n, p) ^{28}\text{Al}$	4.0	550
100	$^{31}\text{P}(n, \alpha) ^{28}\text{Al}$	2.8	130-300

From the above reactions, it can be seen that ^{28}Al can form from $^{28}\text{Si}(n, p)$ and $^{31}\text{P}(n, \alpha)$ reactions besides $^{27}\text{Al}(n, \gamma)$ reaction. However, from these reactions it can be inferred that the formation probability of ^{28}Al from the $^{28}\text{Si}(n, p)$ and $^{31}\text{P}(n, \alpha)$ reaction is insignificant due to higher reaction

threshold value. Thus the lower evaluated value of Al may be due to the incorrect specification of 0.08 mol% of Al_2O_3 quoted for float glass compared to the value of 0.75% for typical container glass. Thus the estimation of different elements based on the activation and off-line gamma-ray spectrometric technique will help glass industry to classify the glass composition accurately.

V. CONCLUSION

1. In the present work, the amount of Na and Al per 100 gram of glass sample was estimated using neutron activation and off-line gamma-ray spectrometric technique.
2. The experimentally determined value for Na per 100 gram of glass sample from present work is in agreement with the value calculated based on the quoted data of composition, which shows the correctness of the present approach. However, in the case of Al, the experimental value per 100 gram of glass sample is about seven times higher than the estimated value based on the quoted data of composition.
3. The estimation of different element of glass sample by neutron activation and off-line gamma-ray spectrometric technique will help glass industry to classify the glass and their composition accurately.

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