

## **Determination of Aluminum in Copper-Aluminum Alloy Using Gamma-Ray Spectrometry**

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*In this work, a nuclear gauge, based on single and dual energy  $\gamma$ -ray transmissions, has been developed and used to determine the concentrations of the aluminum component in copper-aluminum alloy. Disk foils of 7.07 cm<sup>2</sup> area and thicknesses ranging from 0.48 to 0.834 cm have been prepared from the alloy to be used as targets for the  $\gamma$ -ray photons. Criteria for the best choice of the suitable photon energies for both single and dual energy  $\gamma$ -ray transmissions have been investigated. In this work, aluminum concentration values with an accuracy of 98.53% and 99.09% have been obtained by using both single and dual  $\gamma$ -ray energy transmission techniques, respectively. A comparison of the present results with those from the conventional chemical analysis indicates a fair agreement. A criterion for the best choice of the suitable  $\gamma$ -ray transmission technique for the aluminum determination has been investigated.*

## Introduction

Gamma-ray absorptiometry is widely used in non-destructive characterization of opaque materials [1-7]. Many other methods such as X-ray fluorescence spectrometry [8], prompt  $\gamma$ -ray neutron activation analysis [9], chemical analysis, and instrumental neutron activation analysis [10] could be used for material characterization. The main advantages of gamma-ray absorptiometry as an analytical tool are the following [2,3,7]:

- a) its simplicity for handling, instrumentation, and processing,
- b) it can provide representative analysis without the need of sample preparation procedures,
- c) it can give an early indication of the mineral quality by allowing direct determination of concentration of sensitive elements and the derivation of important economic parameters, such as ash content value in samples, in conveyor belts and in ores, and
- d) it is non-destructive, accurate, economic, and low cost analytical tool.

In this work, single and dual energy  $\gamma$ -ray transmission techniques are used to determine the aluminum component in copper-aluminum alloy. Criteria for the best choice of the suitable photon energies for both single and dual energy  $\gamma$ -ray transmissions have been investigated. In this work, aluminum concentration values with an accuracy of 98.53% and 99.09% (100 - fraction error) have been obtained by using both single and dual  $\gamma$ -ray energy transmission techniques, respectively. A comparison of the present results with those obtained from the conventional chemical analysis indicates a fair agreement. A criterion for the best choice of the suitable  $\gamma$ -ray transmission technique for the aluminum determination has been investigated. Preliminary results show that the suggested technique can be successfully applied in composition characterization of materials to give a rapid and economic determination of the concentration of certain elemental components.

## Gamma-Ray Transmission Techniques

The 1980s have seen the development and the widespread use of the industrial application of a new generation of nuclear gauges [11]. Among the measurement techniques used in this new generation of nuclear gauges are the following:

### 1. Single Energy Gamma-Ray Transmission Technique (SET):

The intensity  $I_0$  of a collimated  $\gamma$ -ray beam will be attenuated to an intensity  $I$  after it has passing through an absorbing material mixture of mass thickness  $x$  according to the following general absorption law [12]:

$$I = I_0 e^{-\mu_t x} \quad (1)$$

where  $\mu_t$  is the total mass absorption coefficient which can be given by

$$\mu_t = \sum_i \mu_i C_i, \quad (2)$$

with

$$\sum_i C_i = 1 \quad (3)$$

where  $\mu_i$  and  $C_i$  are the mass absorption coefficient and the weight fraction of the  $i^{\text{th}}$  component in the material mixture, respectively.

In the SET technique [2,7], an unknown concentration of a material component is determined by measuring the intensities of a narrow beam of low or high-energy  $\gamma$ -ray that is transmitted through an absorbing material of mass thickness  $x$  ( $\text{gm}/\text{cm}^2$ ). In the case that the absorbing material is a binary alloy such as Cu-Al alloy, equations 1,2 and 3 can be combined to give an expression for aluminum concentration,  $C_{\text{Al}}$ , in the alloy as follows :

$$C_{\text{Al}} = \left\{ \ln \left( \frac{I_0}{I} \right) / x - \mu_{\text{Cu}} \right\} / (\mu_{\text{Al}} - \mu_{\text{Cu}}) \quad (4)$$

where  $\mu_{\text{Al}}$  and  $\mu_{\text{Cu}}$  are the mass absorption coefficients of aluminum and copper, respectively. Eqn. 4 represents a straight-line relation between  $C_{\text{Al}}$  and  $\left\{ \frac{1}{x} \ln \left( \frac{I_0}{I} \right) \right\}$

## 2. Dual Energy Gamma-Ray Transmission (DET) Technique :

In the DET technique [2,7], the content of a specific component is determined by combining measurements of intensities of narrow beams of low and high-energy  $\gamma$ -rays transmitted through the sample. According to the formula derived by Watt and Steffner [12], the percentage concentration of aluminum in copper –aluminum alloy is determined by using the following expression :

$$C_{\text{Al}} = (\mu''_{\text{Cu}} - R \cdot \mu'_{\text{Cu}}) / \{R (\mu'_{\text{Al}} - \mu'_{\text{Cu}}) + (\mu''_{\text{Cu}} - \mu''_{\text{Al}})\}, \quad (5)$$

The superscripts ‘ and ‘‘ refer to the lower and higher  $\gamma$ -ray energies, respectively.  $R$  is the ratio of the mass absorption coefficients for the material sample at the higher and lower  $\gamma$ -ray energies; it is given by

$$R = \ln(I_o/I)'' / \ln(I_o/I)' \quad (6)$$

$\mu''_{Al} \approx \mu''_{Cu}$  ( $\mu''_{Al} = 0.097$  and  $\mu''_{Cu} = 0.101$  cm<sup>2</sup>/g), therefore the term ( $\mu''_{Cu} - \mu''_{Al}$ ) in Eqn. (5) should be omitted. Accordingly, Eqn. (5) should be rewritten in the following from:

$$C_{Al} = \{\mu''_{Cu} / R (\mu'_{Al} - \mu'_{Cu})\} - \mu'_{Cu} / (\mu'_{Al} - \mu'_{Cu}) \quad (7)$$

which represents a straight line between  $C_{Al}$  and  $1/R$ .

### Error Analysis

In SET technique, the errors in the measured aluminum concentration,  $C_{Al}$  may be due to errors in the measured  $\gamma$ -ray intensities, material density ( $\rho$ ) and the linear thickness of the target ( $d$ );  $x = \rho d$ . The error in the concentration of the determined component can be estimated by the partial differentiation of Eqn.(4).

$$\delta C_{Al} = \frac{\left( (I \delta I_o - I_o \delta I) / (I I_o) - \frac{1}{d\rho} (d\delta\rho + \rho\delta d) \ln(I_o/I) \right)}{d\rho(\mu_{Al} - \mu_{Cu})} \quad (8)$$

The relative error in  $C_{Al}$  has been estimated to be in the order of magnitude of 1.5% .

In the present study, the errors in  $d$  and  $\rho$  are negligible, so that  $\delta d$  and  $\delta\rho$  may be omitted in Eqn. (8) to be in the from :

$$\delta C_{Al} = \frac{1}{d\rho(\mu_{Al} - \mu_{Cu})} ((I \delta I_o - I_o \delta I) / (I I_o)) \quad (9)$$

or

$$\delta C_{Al} = \frac{\delta \mu_t}{(\mu_{Al} - \mu_{Cu})} \quad (10)$$

where  $\delta\mu_t$  is the error in the total mass absorption coefficient  $\mu_t$  and it is given by:

$$\delta\mu_t = \frac{1}{d\rho} \left( \frac{I\delta I_o - I_o\delta I}{I I_o} \right)$$

The relative error in  $\mu_t$  has been estimated; it was found to be  $\leq 1.49\%$ .

Eqn.(10) can be reformulated on the following form to give the sensitivity of  $C_{Al}$  to the relative error in  $\mu_t$ .

$$\delta C_{Al} / (\delta \mu_t / \mu_t) = \frac{\mu_t}{(\mu_{Al} - \mu_{Cu})} \quad (11)$$

In DET technique, the relative errors in the measured aluminum concentration,  $C_{Al}$ , are due to the errors in the experimental measurements of R, which in turn depends on the measured intensity. Partial differentiation of Eqn.(6) gives the error in R as follows:

$$\delta R = \frac{1}{\ln(I_0/I)} \sqrt{\left( \frac{(\mu' \delta I_0'' - I_0'' \delta \mu')}{I'' \cdot I_0''} \right)^2 + R^2 \left( \frac{(\mu' \delta I_0' - I_0' \delta \mu')}{(I' \cdot I_0')} \right)^2} \quad (12)$$

The relative error in R has been estimated to be in the order of the magnitude of 3%. Partial differentiation of Eqn. (5) gives the error of the aluminum concentration, which is due to the inaccuracy in the measurement of R as follows:

$$\delta c_{Al} = \frac{\{(\mu'_{Cu} + C_{Al}(\mu'_{Al} - \mu'_{Cu}))\} - \delta R}{\{R(\mu'_{Al} - \mu'_{Cu}) + (\mu''_{Cu} - \mu''_{Al})\}} \quad (13)$$

The relative error in  $C_{Al}$  was estimated to be of the order of magnitude of 0.9%. the sensitivity of  $C_{Al}$  to the relative error in R can be given by the reformulation of Eqn. (13) to be in the form:

$$\delta C_{Al} / (\delta R / R) = \frac{-((\mu'_{Cu} + c_{Al}(\mu'_{Al} - \mu'_{Cu}))R}{(R(\mu'_{Al} - \mu'_{Cu}) + (\mu''_{Cu} - \mu''_{Al}))}. \quad (14)$$

## Experimental

### I. Target foils

Disk foils of 7.07  $\text{cm}^2$  area and thickness ranging from 0.48 to 0.834  $\text{cm}$  have been prepared from the Cu-Al alloy to be used as targets with different Al concentration for the  $\gamma$ -ray photons. In this work, it was found that when the Al concentration exceeds 31%, the prepared sample would be destructed.

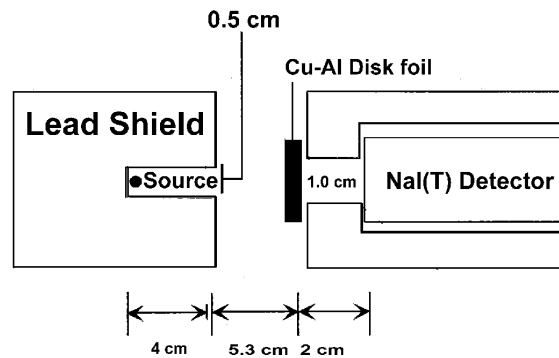


Fig. (1) Experimental set up and arrangement

## II. Experimental Set-Up

The experimental setup consists mainly of a CANBERRA 3" x 3" NaI (Tl) crystal (model 2007P) coupled to a NUCLEUS PCA 8192 multichannel analyzer. The whole experimental setup is shown in Fig. (1). The samples were prepared in the form of cylindrical discs of 3 cm diameter and thicknesses in the range of 0.39 to 0.83 cm at 0.081 and 0.356 MeV  $\gamma$ -ray energies.  $^{133}\text{Ba}$  as a  $\gamma$ -ray source of strength  $\cong 4 \mu\text{Ci}$  "AMERSHAM" was used in the experiment.

## III. Experimental precautions

Using a high accuracy digital balance will help in minimizing the errors in measurements. The background and peak centroids were checked carefully during the  $\gamma$ -ray spectrum measurements. The  $\gamma$ -ray beam was checked using a laser beam (HELIUM-NEON/0.95 mW laser source, SPECTRA PHYSICS type-model 155). Effects due to temperature, applied voltage, and detector efficiency were minimized by stabilizing the voltage source and by air conditioning the laboratory. In addition to these necessary precautions, the photo-peak windows that gave experimental mass absorption coefficient values for Cu and Al in fair agreement with the theory were selected to determine the total photo-peak area.

## Results and Discussion

From Eqn. (4) it's clear that if  $\mu_{\text{Al}}$  approaches  $\mu_{\text{Cu}}$  the dominator in the equation tends to zero and  $C_{\text{Al}}$  will tend to infinity. In such a case it will not be possible to construct an analytical calibration curve in its normal meaning. For this reason, the most important criterion in this work is that the absorption coefficient of one of the material components should be much smaller than the absorption coefficient of the other one. In such a case, the total absorption is mainly due to this absorbing component, and a calibration curve using its variation is possible. Table (1)

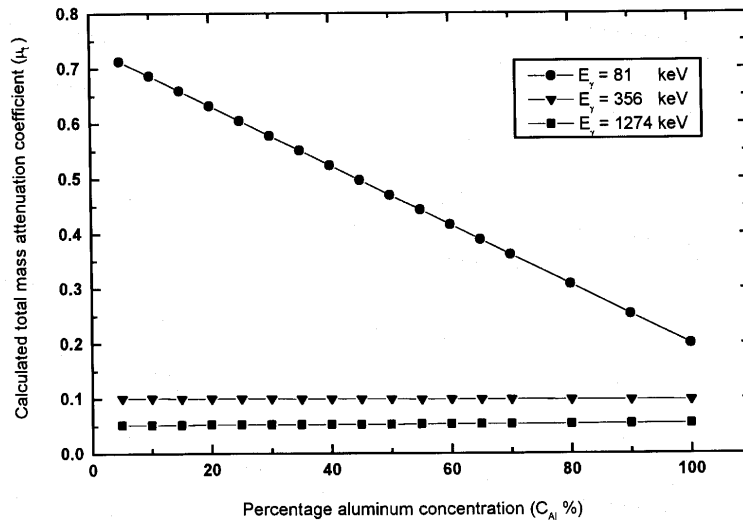
gives the values of the absorption coefficients  $\mu_{Al}$  and  $\mu_{Cu}$  at 81 and 356 keV  $\gamma$ -ray energies [13].

From Table (1), it's clear that the mass absorption coefficient of Cu is nearly 4 times the mass absorption coefficient of Al, which satisfies the previously mentioned criterion.

**Table (1): The mass absorption coefficients of Cu and Al components in the Cu-Al alloy at 81 and 356 keV  $\gamma$ -ray energies**

Mass absorption coefficients of the Cu-Al components (cm <sup>2</sup> /gm)	$\gamma$ -ray energies (keV)	
	81	356
$\mu_{Cu}$	0.74	0.101
$\mu_{Al}$	0.20	0.097

The value of the  $\gamma$ -ray energy that satisfy the above mentioned criterion has been investigated. Fig. (2) shows the variation of the total mass absorption coefficient,  $\mu_t$ , with the percentage concentration of Al,  $C_{Al}$ , at the  $\gamma$ -ray energies 81 and 356 keV. From the figure, it can be seen that the  $\gamma$ -ray absorption in the alloy is clearly sensitive to the variation in the Al concentration at 81 keV  $\gamma$ -ray energy while at the other energy the response is not remarkable.



**Fig. (2)** Variation of both calculated and measure total mass absorption coefficients with Al concentration at 81 and 356 keV  $\gamma$ -ray energies.

Equation (11) has been used to clarify the correlation between the variation in the percentage Al concentration ( $\delta C_{Al}$ ) and the variation in the relative error ( $\delta\mu_t/\mu_t$ ) in the total mass absorption coefficient. Fig. (3) chose the dependence of the variations ( $\delta C_{Al}$ ) in the percentage Al concentration on the

normalized relative error ( $\delta\mu_t/\mu_t$ ). A percentage error of one percent in  $\mu_t$  corresponds to variations of 0.01, 0.26 and 0.23 in the percentage Al concentration at the  $\gamma$ -ray energies 81, 356, and 1274 keV, respectively. This indicates that the variation of the percentage Al concentration with the percentage error in  $\mu_t$  at the photopeak line 81 keV is clearly smaller than that at the other two-photopeak lines. From the above discussion, it is conclusively evident that the line 81 keV is more preferable to be used as a SET gauge for Al determination in Cu-Al alloy. A calibration curve between  $C_{Al}\%$  and  $1/x \ln(I_o/I)$  shows that, the Al concentration should not exceed 31% because as mentioned before when the Al concentration exceeds 31% the target foil will be destructed.

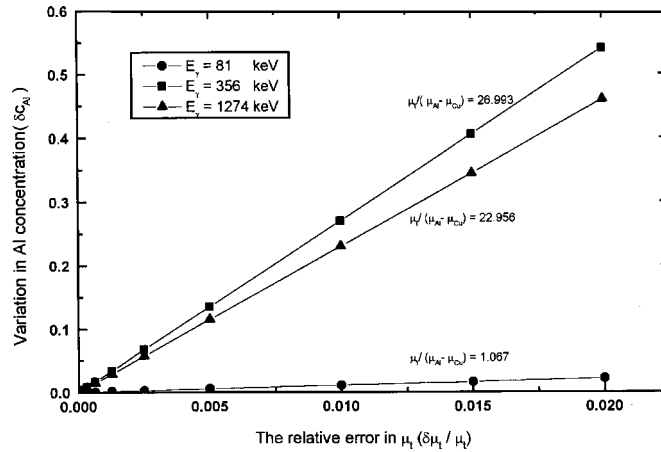


Fig. (3) Effect of the error in the total mass absorption coefficient on the variation in aluminum concentration in an alloy with 69.64% Cu and 30.36% Al at 81,356 and 127 keV  $\gamma$ -ray energies

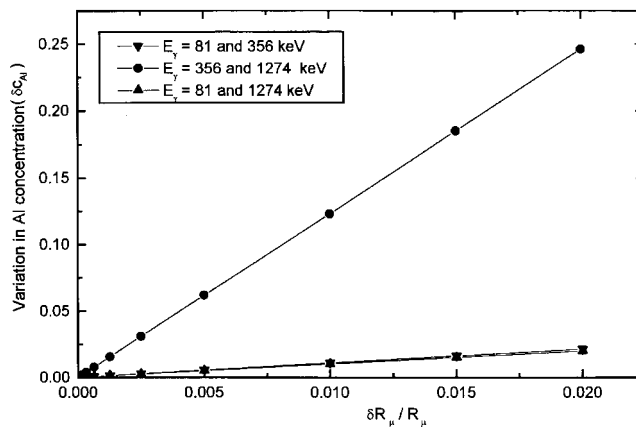


Fig. (4) Effect of the error in  $R_\mu$  on the variation in Al concentration in an alloy of 69.64% and 30.36% at three different energy couples.



In order to determine the suitability of a  $\gamma$ -ray energy couple to be used as a dual  $\gamma$ -ray energy beam, Eqn. (14) has been applied to calculate  $\delta C_{Al}$  for a sample of known concentrations in terms of  $\delta R/R$ . Fig. (4) shows  $\delta C_{Al}$  versus  $\delta R/R$  as obtained from Eqn. (14). It is obvious that the relation is a straight line of a slope, which is dependent on the value of  $\mu$  and  $R$ . It could be easily noted that there is no an observable difference between the energy couples (81, 356 keV) and (81, 1274 keV), while for the energy couple (356, 1274 keV)  $\delta C_{Al}$  shows a large sensitivity to  $\delta R/R$ . a percentage error of 1% in  $\delta R/R$  corresponds to the variations of 0.011, 0.0101 and 0.123 in the Al concentration percentage ( $C_{Al}$ ) at the energy couples (81, 356 keV), (81, 1274 keV) and (356, 1274 keV), respectively. So, it could be concluded that Fig. (4) does not recommend the energy couple (356, 1274 keV) as a partner in the present proposed dual energy transmission technique. Accordingly, the energy couple (356, 1274 keV) should be excluded in this work. More over, the experimental setup is based on the using of narrow beam geometry, so that Compton continuum is not effective. The energy couple of the two photolines 81 and 356 keV is emitted from a compact radioactive source  $^{133}\text{Ba}$ , therefore a good beam geometry with a good source positioning and a beam alignment could be attained. The choice of the energy couple for DET technique should be governed by all these previous criteria. Accordingly, the energy couple (81, 356 keV) is chosen as a dual energy beam to be used in the present work. Although, Fig. (2) demonstrates that the DET technique is of no value, since at the 356 keV energy there is no response, and the use of this technique adds nothing new to the SET technique, the DET technique has the advantage of its independence on the target thickness. This enables us to avoid any errors due to thickness measurements, which by role reflects on accurate determination of Al concentration. According to Eqn. (8) a calibration curves between  $C_{Al}\%$  and  $1/R$  which is equal to  $\ln(I_0/I)' / \ln(I_0/I)''$  has been investigated.

The accuracy of the present Al determination by using both single and dual  $\gamma$ -ray energy transmission techniques has been tested by analyzing the measured samples using the conventional chemicals analysis technique. Average relative errors of the order of magnitudes  $\pm 1.69\%$  and  $\pm 1.25\%$  have been assigned to the results from both single and dual  $\gamma$ -ray energy transmission techniques, respectively. In Figs. (7,8), the percentage aluminum concentration values from the SET gauge (81 keV) and the DET gauge (81, 356 keV), respectively, are compared with those values obtained from conventional chemical analysis. Both figures show linear relationships between the aluminum concentration values as obtained from the  $\gamma$ -ray absorption measurements and those obtained from chemical analysis. A linear regression has been made for the data in both figures. Linearity with r.m.s estimating error of  $\pm 0.4$  and  $\pm 0.37$  for both figures, respectively, has been attained.

The errors in the mass thickness  $x$  and density  $\rho$  in addition to the errors in  $I_0$  and  $I$  are the main sources of the errors in the SET gauge. On the other hand  $I_0'$ ,  $I'$ ,  $I_0''$  and  $I''$  are the main sources of the errors in the DET gauge. As the conclusion, the values of these errors stand behind the decision which gauge (SET or DET) can be used for the determination of Al concentration in Cu-Al alloy. In some details, if  $x$  is large and  $\rho$  is also large, the DET gauge is recommended. While, if  $I_0'$ ,  $I'$ ,  $I_0''$  and  $I''$  values are not accurately enough due to, e.g., Compton continuum and low  $\gamma$ -ray counting rates and if

there is no significant error in the thickness x, the SET gauge is the best technique and strongly recommended.

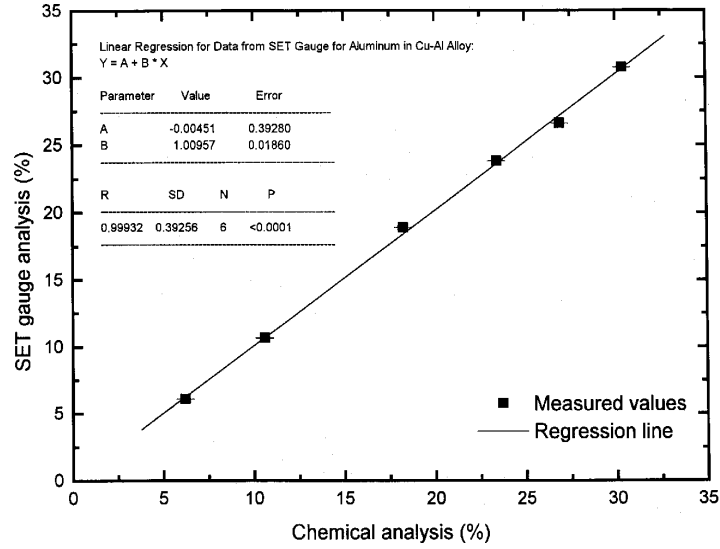


Fig. (5) A comparison between results from the SET gauge for Al in Cu-Al alloy and the those results from chemical analysis.

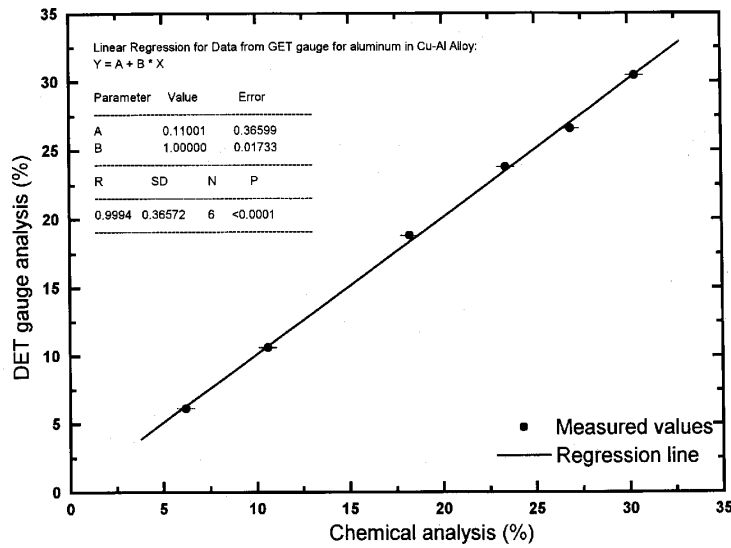


Fig. (6) A comparison between results from the DET gauge for Al in Cu-Al alloy and those results from chemical analysis.

**Table (2) : Aluminum concentration values in Copper-Aluminum alloy as obtained from chemical analysis and  $\gamma$ -ray transmission techniques.**

Chemical analysis % (standards)	SET gauge analysis % (81 keV)	DET gauge analysis % (81, 356 keV)
6.2	6.07±0.089	6.13±0.055
10.6	10.65±0.081	10.62±0.051
18.23	18.89±0.086	18.81±0.050
23.4	23.81±0.097	23.79±0.058
26.9	26.61±0.05	26.57±0.024
30.36	30.74±0.074	30.43±0.034

In this work, the linear relation between the results from the DET gauge and those from the standard chemical analysis gave a slope of one Fig. (8), which indicates a good agreement between both results. Since  $C_{Al}$  values as obtained by using the DET gauge were in good agreement with those obtained from the standard chemical analysis, we will conclude that the DET gauge is the best one for aluminum characterization. A slope with a slight shift far from 1 is obtained by the linear regression between the results from the SET gauge and those from the standard chemical analysis Fig. (7), which indicates some deviation between the nuclear and chemical results. Another reason for the preference of DET gauge on the SET gauge as an aluminum analyzer in this work is that the total error in  $C_{Al}$  values in case of DET gauge is smaller than that in case of the SET gauge.

## Conclusion

A nuclear gauge, based on single and dual energy  $\gamma$ -ray transmissions, has been developed and used to determine the concentration of the aluminum component in copper-aluminum alloy. The gauge is portable and uses a  $^{133}\text{Ba}$   $\gamma$ -ray microsource as the main source radiation. The gauge does not require special shielding and does not expose the user to unacceptable levels of radiations. Aluminum concentration values with an accuracy of 98.53 % and 99.09 % have been obtained by using both single and  $\gamma$ -ray energy transmission techniques, respectively. In addition one can conclude the following:

1. For binary alloys, the most important criterion in this work is that the absorption coefficient of one of the material components should be much smaller than the absorption coefficient of the other one.
2. The present gauge has been successfully applied to binary alloys and it can be extended for the characterization of multiple component alloys with a dominated component.

3. Although, the DET technique does not add any new to the SET technique, it still has the advantage of its independence on the target thicknesses, which enable us to avoid any errors due to thickness measurements.

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