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**Elemental Analysis of Brazing Alloy Samples
by Neutron Activation Technique**

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**ELEMENTAL ANALYSIS OF BRAZING ALLOY SAMPLES BY
NEUTRON ACTIVATION TECHNIQUE**

Two brazing alloy samples (CP2 and CP3) have been investigated by Neutron Activation Analysis (NAA) technique in order to identify and estimate the concentration of the constituent elements. The Pneumatic Irradiation Rabbit System (PIRS), installed at the First Egyptian Research Reactor (ET-RR-1), was used for short-time irradiation (30 s) with a thermal neutron flux of $1.6 \times 10^{11} \text{ n/cm}^2 \cdot \text{s}$ in the reactor reflector, where the thermal to epithermal neutron flux ratio is 106. Long-time irradiation (48 hours) was performed at the (ET-RR-1) reactor core periphery with thermal neutron flux of $3.34 \times 10^{12} \text{ n/cm}^2 \cdot \text{s}$ and thermal to epithermal neutron flux ratio of 79. Activation by epithermal neutrons was taken into account for the $(1/v)$ and resonance neutron absorption in both methods. A hyper pure germanium (HPGe) detection system was used for γ -ray acquisitions. The concentration values of Al, Cr, Fe, Co, Cu, Zn, Se, Ag and Sb were estimated as percentages of the sample weight and compared with the reported values.

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INTRODUCTION

Silver alloy solders or brazing materials were first used by jewelers and other similar trades. These had a silver content of approximately 80% and were white in color to match "sterling silver". Silver brazing is the joining of metals by means of heat, using a filler metal consisting of silver, copper and small percentage of other metals. The flow temperature of this filler metal alloy must be above (427°C), to classify as silver brazing⁽¹⁾.

Brazed joints are superior joints in a number of ways. They are strong, with tensile strengths greater than those of the brazing alloys themselves and approaching the strength of the metals joined. They are ductile-able to withstand the stresses of shock and thermal expansion and contraction. They are excellent conductors of electricity and heat, and they are leak tight, corrosion resistant, and smooth in appearance.

Brazing alloys are used extensively for joining copper, especially refrigeration and air conditioning copper tubing and copper electrical conductors. They may also be used on brass, with specialized application on silver, tungsten molybdenum⁽²⁾. The unit cost of brazed joints is relatively low, for several reasons, as very little brazing alloy is needed to make a joint and brazing is performed at low temperatures compared to welding.

Analysis of some complex industrial samples such as alloys, to obtain some information on the elemental constituents has a considerable interest in the last few decades⁽³⁻⁹⁾. Such analysis can be achieved by (NAA) technique which is considered as a very sensitive and non destructive method. This gives a good indication on the quality of the samples under investigation by direct determination of the concentration values of the effective elements.

EXPERIMENTAL

Two samples of brazing alloys (CP2) and (CP3) were provided in a wire form by the Egyptian General Engineering & Motor company which imports them from two

British companies, Johnson Matthey PLC MTD Metal Joining and Thessco Limited, respectively^(10,11). Each sample was prepared in a homogeneous granular form.

A specimen weighing 15 mg of each were packed in a clean aluminum foil of known weight. A gold foil monitor and an empty aluminum foil of known weights were included with the two specimens in the same irradiation can for qualitative and quantitative analysis purposes. The can was irradiated with 3.34×10^{12} thermal neutrons / cm². s. for 48 hours at the (ET-RR-1) reactor core periphery where the thermal to epithermal neutron flux ratio is 79. All of the can contents were left for 3 days to cool down post the end of irradiation and before γ -ray spectrum measurements were started. Gamma-ray acquisition was performed for an hour for each of the can contents and was repeated weekly for 3 weeks.

The (PIRS) installed at the (ET-RR-1) reactor was used for irradiating 45 mg from each of the brazing alloy specimens enveloped in cellophane paper of a known weight. A pure gold foil monitor was included with each specimen inside a polyethylene vial. The irradiation was performed for 30 s in the reflector near the reactor core with a thermal neutron flux of 1.63×10^{11} neutrons / cm². s where the thermal to epithermal flux ratio was 106. Gamma-ray acquisitions were performed for an hour after the elapse of 50 s in transporting the specimen post irradiation.

A coaxial (HPGe) detector of diameter 56.5 mm and 81.3 mm length with an active volume facing the window of 200 cm³ and a dead layer of thickness 600 microns was used with the associating electronic units and a MYLEX PCA. A lead shield was surrounding the detector and a lead γ -ray collimator was located above the (HPGe) detector to achieve a good detection geometry. The dependence of the absolute full-energy peak (FEP) on the γ -ray energies was determined by means of ²²Na, ²⁴Na, ⁶⁰Co and a multigamma-ray standard sources⁽¹³⁾. A fortran computer program was constructed for calculating the elemental concentration of the samples by TANDY 3000N PCA.

RESULTS AND DISCUSSION

The sensitivity tables reported by Eissa et al^(14,15) for long- and short-time irradiation were used in interpreting the γ -ray spectra of the (CP2) and (CP3) brazing alloys.

Background γ -ray lines resulting from contaminants with the lead shield and collimator surrounding the (HPGe) detector are mainly due to γ -ray lines from ^{40}K , ^{60}Co , ^{137}Cs , ^{226}Ra and ^{232}Th .

The trace elements in the aluminum envelopes of the specimens are Na, K, Sc, Fe, Co, Zn, Ga, As, Br, Mo, Sb, Hf, Au and Th which were considered as background γ -ray lines to be subtracted in case of interference with those of the brazing alloy specimens. The (FEP) area of the gamma-ray lines emitted from the empty Al envelope were normalized to the weight of the envelopes containing the brazing alloy samples as well as the decay time before subtraction.

Cellophane paper is insensitive to activation by short-time irradiation, so that the only γ -ray background lines are due to contaminants in the lead shield and collimator.

The epithermal neutron contribution to the sample activation was taken into account for the (1/v) and resonance neutron absorption in both methods by using the following expression:

$$C = \frac{A}{[F L_{\gamma} \epsilon_{\gamma} N^{\circ} / M \lambda] \sigma_{th} \phi_{th} [1 + (\sigma_{epi} \phi_{epi} / \sigma_{th} \phi_{th})] (1 - e^{-\lambda T}) e^{-\lambda t} (1 - e^{-\lambda t_m})}$$

.....(1)

where

- C is the concentration of the element as a fraction of the sample weight,
- A is the net (FEP) area, which represents the number of characteristic γ -rays accrued during a time interval t_m (=3600 s) per gram of the specimen,
- F is the fractional isotopic abundance of the target nuclide,
- L_{γ} is the absolute intensity of the characteristic γ -ray line,
- ϵ_{γ} is the absolute (FEP) efficiency of the (HPGe) spectrometer for a characteristic γ -ray line,
- N° is Avogadro's number in atoms / gm . atom,
- λ is the decay constant of the product nuclide in s^{-1} .

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- M is the atomic weight of the trace element in gm / gm atom,
 σ_{th} is the thermal neutron capture cross section in $cm^2 / atom$,
 σ_{epi} is the integral cross section, in $cm^2 / atom$, due to $(1/v)$ and resonance absorption
over the reactor $(1/E)$ epithermal neutron spectrum,
 ϕ_{th} is the thermal neutron flux in $n / cm^2 \cdot s$,
 ϕ_{epi} is the epithermal neutron flux in $n / cm^2 \cdot s$,
 T is the irradiation period in seconds,
 t_i is the transportation time interval in seconds elapsed post irradiation until γ -ray measurement is started, and
 t_m is the measurement time in seconds.

Values of thermal neutron capture cross section and resonance integral cross section over the epithermal neutron spectrum were taken from ref⁽¹⁶⁾. The denominator of Equation (1) can be defined as the total sensitivity^(14,15) corrected for activation by epithermal neutrons. This is the number of accrued characteristic γ -rays during t_m due to activation by a thermal reactor neutron spectrum per unit elemental concentration. Table (1) shows the concentrations estimated in percentage for the constituent elements of the two brazing alloy samples compared with the previously reported values^(10,11).

In the present work the presence of Cr, Fe, Co, Se and Sb in the two brazing alloy samples is reported for the first time. The concentrations of Zn and Cu in the present work are in fair agreement with the reported values. The aluminum concentration of the present work is 1.46 times the reported value for (CP2) while it is nearly fourth the reported value for (CP3). The present work gives an upper concentration limit of 0.71% for silver in the (CP2) sample which is much lower than the reported value 1.8%. Also an upper concentration limit of 0.12% of silver for the (CP3) sample which was reported as a silver free sample.

Both elements Be and P form a single stable nuclide (9Be and ^{31}P) which are activated to the pure beta emitters ^{10}Be (2.5×10^6 years) and ^{32}P (14.28 days). This is the reason for their undetectability in the present work.

Bismuth forms a single stable nuclide ^{209}Bi which can be activated in the long-time irradiation method to ^{210}Bi of half-life time 5.01 days, but it was undetectable, probably because of its low concentration and poor sensitivity.

Natural Lead has four isotopes ^{204}Pb (1.4%), ^{206}Pb (25.1%), ^{207}Pb (21.7%) and ^{208}Pb (52.3%). The very long lived product nuclide ^{203}Pb (3.0×10^7 years) as well as the stable product isotopes ^{207}Pb and ^{208}Pb , in addition to the pure beta-emitter ^{209}Pb keep lead undetectable even with 100% concentration.

Naturally occurring Cadmium has eight isotopes ^{106}Cd (1.25%), ^{108}Cd (0.89%), ^{110}Cd (912.5%), ^{111}Cd (12.8%), ^{112}Cd (24.1%), ^{113}Cd (12.2%), ^{114}Cd (28.7%) and ^{116}Cd (7.5%). ^{113}Cd has an extremely high thermal neutron capture cross section (19910 barns) leads to a stable product isotope ^{114}Cd . This is in addition to the stable product isotopes ^{111}Cd and ^{112}Cd will be undetectable. Similarly the extremely long lived product isotope ^{113}Cd (9×10^{15} years) and its isomer $^{113\text{m}}\text{Cd}$ (14 years) will be undetectable. The product isotopes $^{111\text{m}}\text{Cd}$ (48.6 minutes), ^{113}Cd (53.4 hours), ^{117}Cd (2.4 hours) and its isomer $^{117\text{m}}\text{Cd}$ (3.4 hours) were not detected in the short-time irradiation method probably because of their poor sensitivities and the low content of cadmium in the samples. Similarly the product isotopes ^{109}Cd (453 days), ^{115}Cd (53.4 hours) and its isomer $^{115\text{m}}\text{Cd}$ (44.8 days) were not detected in the long-time irradiation method because of their poor sensitivities and the low concentration of cadmium in the samples.

Finally, one may say that (NAA) technique with the facilities mentioned in this work was able to make a good elemental analysis with high accuracy for such industrial samples.

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Table (1): Concentration (in %) of elements in the two brazing alloy samples compared with the previously reported values.

Element	Isotope or Isomer	Present work concentration(%)		Reported values ^(10,11)	
		CP2	CP3	CP2 max	CP3 max
Beryllium	⁷ Be	undetectable	undetectable	0.0005
Aluminium*	²⁸ Al	0.0146±0.0015	0.0024±0.0006	0.0100	0.0100
Phosphorous	³¹ P	undetectable	undetectable	6.9000	7.8000
Chromium	⁵¹ Cr	0.0010±0.0001	0.0036±0.0002
Iron	⁵⁶ Fe	2.300±0.0490	2.1200±0.0490
Cobalt	⁶⁰ Co	0.0035±0.00014	0.0030±0.0001
Zinc	⁶⁵ Zn	0.0486±0.0017	0.0472±0.0015	0.0500	0.0500
Copper*	⁶⁶ Cu	92.0561±3.9634	95.4200±2.2100	90.6500	93.0000
Selenium	⁷⁵ Se	0.0049±0.0002	0.0037±0.0002
Silver	^{110m} Ag	≤ 0.7100±0.0078	< 0.1200±0.0030	1.8 - 2.2
Cadmium	¹¹³ Cd	undetectable	undetectable	0.0250	0.0250
Antimony	¹²² Sb, ¹²⁴ Sb	0.0060±0.0002	0.0087±0.0005
Lead	²¹⁰ Pb	undetectable	undetectable	0.0200	0.0200
Bismuth	²¹⁴ Bi	undetectable	undetectable	0.0010	0.0010

* Elements which were analyzable only by the short-time irradiation method.