Investigation of the Photo-activation Analysis on Concentrations of Various Materials

A Cesur¹, M Karakoc¹,², I Boztosun¹,² and H Dapo¹,²
¹ Akdeniz University, Department of Physics, 07058, Antalya, Turkey
² Akdeniz University, Nuclear Sciences Application and Research Center – NUBA, 07058 Antalya, Turkey
E-mail: alpcesur@akdeniz.edu.tr

Abstract. It is possible to excite atomic nuclei electromagnetically and even separate proton or neutron from a sample which is exposed to gamma rays by a clinical linac. Thus, the determination of the elemental concentration of a sample by photo-activation analysis is a reasonable attempt since it is related to the detailed analysis of the photonuclear reaction of an intended element. In this study, in order to get the proof of principle for photo-activation a titanium dioxide and a 10 kurus Turkish coin samples are irradiated with high energy bremsstrahlung photons at the end-point energy of 18 MeV and their zinc and copper concentrations are determined by a precise measurement with a high purity germanium detector.

1. Introduction
The determination of the elemental concentration in a given sample has taken a great deal of interest over the years. There are two ways to achieve this; destructive and non-destructive methods. Non-destructive methods are very important for the analysis of precious elements. These methods are known to be very reliable and accurate. While destructive methods like X-ray fluorescence, laser ablation and radiochemical neutron activation analysis (RNAA) provide accurate results, the analysed samples are often damaged due to the chemical treatment on samples. Neutron activation analysis (NAA) is one of the well-recognised non-destructive method. In general, thermal neutrons from nuclear reactors utilised for activation. However, it has many disadvantages. For instance, the determination of some low-Z elements such as C, N, O, F or several other elements such as Mg, Si, Ca, Ti, Ni, Sr, Y, Zr, Nb, Tl and Pb is not determined accurately or sometimes the determination is impossible at all [1]. In addition, the results may provide only the surface concentrations [2]. Photo-activation analysis (PAA) on the other hand is a complementary method to NAA. In PAA, the target nucleus is activated through photo-induced reactions such as single-neutron or proton knockout, usually at optimal photon energies between 10 and 30 MeV [3]. It is also shown that PAA allows the determination of elements with a better sensitivity compared to NAA [4,5,6].

In this paper, in order to verify the success of PAA method, we aim to investigate the determination of elemental concentration for a given sample by using PAA. In the next section, we present the experimental method and data analysis. Section 3 presents the results of our measurements and section 4 is devoted to our conclusions.
2. Experimental

We provide below the details of experimental method in order to determine the elemental concentration in a 10 kurus Turkish coin and titanium dioxide (TiO$_2$) sample by using a c-linac and HPGe detector in our PAA setup.

2.1. Sample and Reference Preparation

In order to obtain accurate results in PAA, it is important that the sample and reference material have the same geometry. As it is pointed out in the introduction, in contrast to destructive methods, there is no physical or chemical damage to sample under study in PAA method.

2.2. Activation Process

Samples are activated with a c-linac with an end-point energy of 18 MeV, which is already in use at Akdeniz University Nuclear Sciences Application and Research Center (NUBA). The irradiation surface is adjusted to 55 cm below the tungsten converter. Sample and reference materials are then placed to the central point of irradiation area carefully and exposed to high energy bremsstrahlung photons. Thus, photonuclear reactions are performed.

2.3. Spectrum Measurement and Analysis

After the irradiation process and the cooling time, the sample and reference material carried to the gamma spectroscopy laboratory safely in a lead container. Gamma-ray spectrometers with a HPGe coaxial detectors with relative efficiencies of 40% and resolution FWHM 1.85 keV for the 1332 keV photons of $^{60}$Co were used for PAA. Before counting process, a Job Control file is created with detector software Maestro [7]. Thus, spectrum files can be stored in a certain sequence of time. When counting is finished the spectrum is then analysed by gf3 Radware gamma spectroscopy software [8].

2.4. Calculation of Concentration

Both samples and reference material are exposed to the same amount photon flux during the irradiation process since the beam profile of our c-linac is very well known. It is assumed that all the samples that will be examined are present on earth, so natural abundance of these elements is the same for both sample and reference material [9]. According to these hypothesis, the relative concentrations of the investigated sample calculated as follows [10]:

$$C_S = \frac{C_R \frac{M_R I_S}{M_S I_R} e^{-\lambda T_{DR}}}{1-e^{-\lambda T_{DS}}} \frac{1-e^{-\lambda T_{CR}}}{1-e^{-\lambda T_{CS}}}$$  \hspace{1cm} (1)

In this equation, $S$ denotes the sample and $R$ denotes the reference material. $C_R$ and $C_S$ are concentrations, $M_R$ and $M_S$ are total masses, $I_R$ and $I_S$ are the net peak areas, $T_{DR}$ and $T_{DS}$ are cooling times and $T_{CR}$ and $T_{CS}$ are the measurement times in detector of referance and sample respectively, $\lambda$ is the decay constant of the isotope of the element whose concentration is to be determined.

Usually uncertainty related to decay constant, cooling time and measurement times in detector are much smaller than other uncertainties as given in Eq. (1). Hence, these parameters may not be included in error propagation. We will also assume that the mesurement time was recorded without any uncertainties. Error propagation [10] for Eq. (1) is given by:

$$\Delta C_S = C_S \left[ \left( \frac{\Delta C_R}{C_R} \right)^2 + \left( \frac{\Delta M_R}{M_R} \right)^2 + \left( \frac{\Delta M_S}{M_S} \right)^2 + \left( \frac{\Delta I_R}{I_R} \right)^2 + \left( \frac{\Delta I_S}{I_S} \right)^2 \right]^{1/2}$$  \hspace{1cm} (2)

Various elements concentrations in a sample can be obtained by characteristic gamma ray transition energies and also the error propagation i.e. reliability of the obtained data could be calculated along with those equations.
In order to find the zinc and copper concentrations in the TiO$_2$ and 10 kurus coin sample, a zinc pill and a 1 kurus coin used as reference materials respectively.

![Titanium spectrum]

**Figure 1.** Gamma Energy Spectrum of TiO$_2$ $^{64}$Zn($\gamma$,n)$^{63}$Zn reaction at 669 and 962 keV energies has been observed and analysed.

3. Results

As described above, we have irradiated 10 kurus sample together with the reference material by using a c-linac with an end-point energy of 18 MeV and counted the samples with a HPGe detector. The spectrum is shown in Fig. 1. As it can be seen from this figure, we observe not only ($\gamma$,n) reactions but also ($\gamma$,p) reactions. However, since we are interested in determining the concentration of zinc, we only use $^{64}$Zn($\gamma$,n)$^{63}$Zn reaction data of 669 and 962 keV energies. The results of analysis are given in Table 1-2. As it can be seen from the results presented in Tab. 2, the results of concentration analysis for coins are in a good agreement with corresponding literature values. The results for both TiO$_2$ and 10 kurus samples of $^{63}$Zn at 669 and 962 keV energies are also in a good agreement with each other. Sample concentration values are represented by $C_S$ and their respective uncertainty of the concentration values are represented by $\Delta C_S$.

The reliability of coin results lead us to investigate the zinc concentration in a TiO$_2$ sample as a trace element. Results of this experiment is clearly better owing to the fact that they have smaller uncertainties. It should be noted that the counting time for TiO$_2$ sample was much greater than 10 kurus coin sample. All parameters in Eq. (1) are nearly the same except counting time and so the net peak area. It is then easy to see in order to have minimal uncertainty and more accurate results there should be sufficient amount of counts in an energy peak which should be investigated further.
Table 1. Concentrations and their uncertainties in TiO₂ sample.

| Name | Isotope | Energy (keV) | Cᵣ (%) | Cₛ(%) | ΔCₛ |
|------|---------|--------------|---------|-------|------|
| TiO₂ | ⁶³Zn    | 669          | 0.333   | 0.269 | 0.013|
| TiO₂ | ⁶³Zn    | 962          | 0.333   | 0.274 | 0.014|

*Our Zn reference material

Table 2. Concentrations and their uncertainties in 10 Kr sample.

| Name | Isotope | Energy (keV) | Cᵣ (%) | Cₛ(%) | ΔCₛ |
|------|---------|--------------|---------|-------|------|
| 10 Kr| ⁶³Zn    | 669          | 29.0    | 22.8  | 3.3  |
| 10 Kr| ⁶³Zn    | 962          | 29.0    | 24.8  | 4.3  |
| 10 Kr| ⁶³Cu    | 1172         | 65.0    | 49.8  | 18.4 |

*Turkish General Directorate of Mint and Stamp Print House [11]

4. Summary and Conclusions

In this paper, we have shown the application of PAA method to determine the elemental concentration of an intended sample by using a c-linac and a HPGe detector. Therefore, we performed series of experiments on a 10 kurus Turkish coin and TiO₂ sample in order to get a proof of principle for photoactivation analysis with our experimental setup. Our findings are in good agreement with the literature values. Through this study, we also show the inherent non-destructive nature of PAA method. This property can be used to analyse samples that are too precious to be harmed in any way, like archaeological samples. It is important to note that PAA analysis is used to determine the elemental concentration in a given sample first time in Turkey. Further applications will be made in the future.

In order to improve the results further, counting process has to be handled more carefully since most of the uncertainty in calculations come from the net peak area of the sample and reference material. Effect of dead time, shielding issues and others also have to be taken care of in the future.

Acknowledgements

This work is partly supported by the scientific and technological research council of Turkey (TÜBİTAK) with grant number: 114F220.

References

[1] Randa Z, Kucera J, Mizera J and Frana J 2006 J. Radioanal. Nucl. Ch. 271 589
[2] Green J, Wells D, Benson B, Zaijing S and Maschner H 2011 AIP Conf. Proc. 1336 497
[3] Wells D, Segebade C and Cole P 2010 AIP Conf. Proc. 1265 379
[4] Lutz G J 1971 Anal. Chem 43 93
[5] Randa Z, Spacek B, Kuncir J and Benada J 1981 Nuclear Information Centre, Prague
[6] Segebade C, Weise H P and Lutz G J 1988 Photon Activation Analysis (Berlin, New York: W. de Gruiter)
[7] ORTEC 2012 Maestro Application Software http://www.ortec-online.com/download/MAESTRO.pdf
[8] Radford D C 2000 Notes on the use of the program gf3 http://radware.phy.ornl.gov/gf3/gf3.html
[9] Boztosun I et al. 2014 Turk. J. Phys. 38 1-9
[10] Sun Z J, Wells D, Segebade C and Green J 2011 AIP Conf. Proc. 1336 473
[11] http://www.darphane.gov.tr/tr/content.php?parent_id=182&content_id=735