Nucl. Sci. and Tech, Vol.4, No. 4 (2014), pp. 22-29

Nuclear Science and Technology

Journal homepage: https://jnst.vn/index.php/nst

Integration of auto analysis program of gamma spectrum and software and determination of elementcontent in sample by k-zeromethod

Trinh Quang Vinh[1], Truong Thi Hong Loan[2], Mai Van Nhon[2], Huynh Truc Phuong[2]

[1]Vietnam National University Ho Chi Minh City; [2]University of Sciencein Ho Chi Minh City

Email: tqvinh@vnuhcm.edu.vn

Abstract: Integrating the gamma spectrum auto-analysis program withelemental analysis software by k-zero method is the objective for many researchers. This work is the first stepin building an auto analysis program of gamma spectrum, which includes modules of reading spectrum, displaying spectrum, calibrating energy of peak, smoothing spectrum, calculating peak area and determining content of elements in sample. Then, the results fromthe measurments of standard samples by a low level spectrometer using HPGe detecotr are comparedto thoseof other gamma spectrum auto-analysis programs.

Key words: *gamma spectra, k-zero*

I. INTRODUCTION

For many researchers, the sample's elemental qualitative and quantitative analysis is essential, so the task of building anauto-analysis program of gamma spectrum integrated with anelemental analysis software is still the main goal.

The auto-analysis program of gamma spectrum VGSpec version 2.2 is based on the C # programming language, built with WPF interface (Windows Presentation Foundation) in Microsoft Visual Studio 2010 environment. It has some improvements inthe functions of elemental qualitative and quantitative analysis.

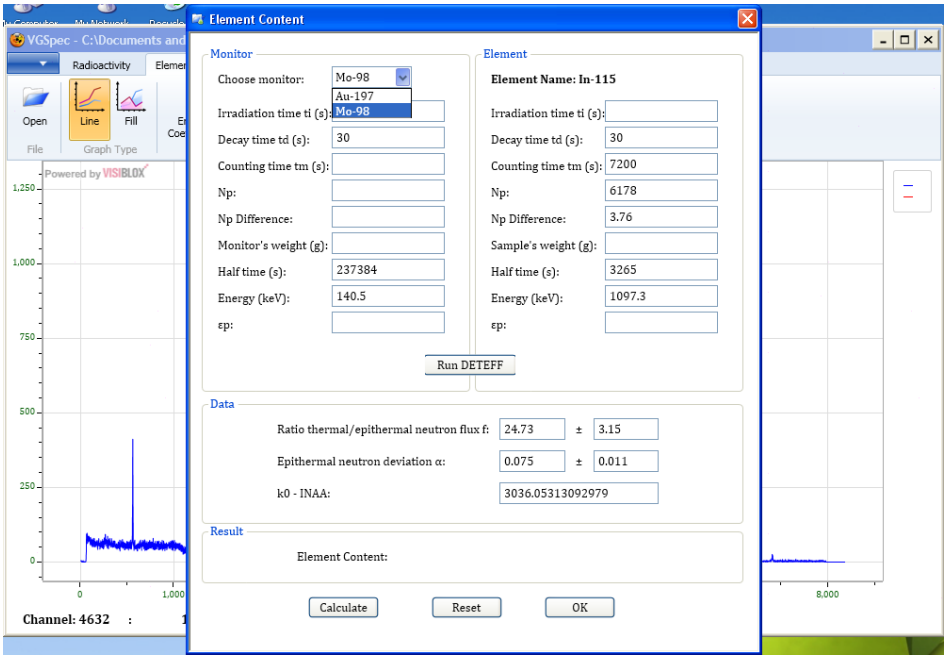

Fig. 1. Interface of quatitative analysis of sample^{115}In in VGSpec programme.

https://doi.org/10.53747/jnst.v4i4.240
Received 19 January 2014, accepted 5 February 2015

A block diagram of the program is described in Figure 2.

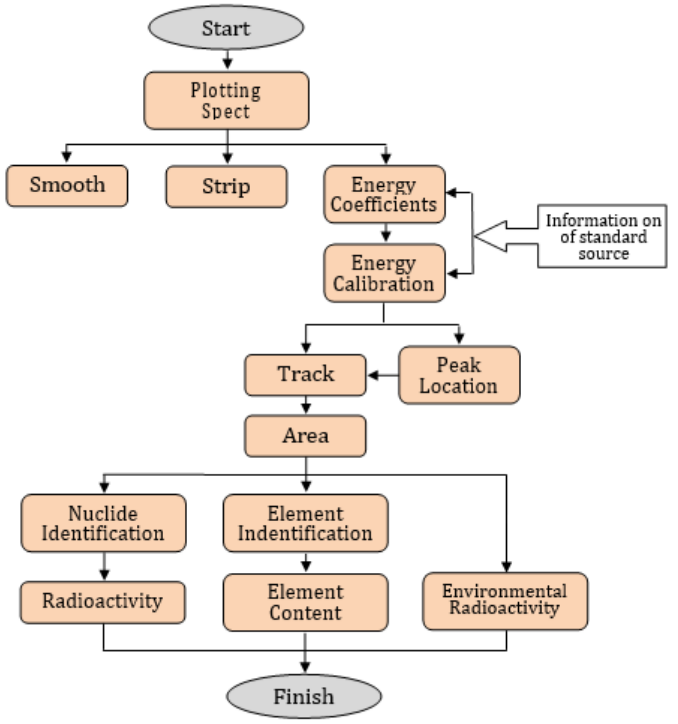

Fig. 2. Block diagram of an auto analysis program of spectrum VGSpec.

II. METHODS

Method of determining element contents by standardized k-zero - INAA is one of the most significant developments of the NAAappeared in the 70s of the last century. Compared with the other standardization methods, k-zero standardization method has some advantages such as simpler than the relative method, more accurate than the absolute method; flexible as condition changesand measurement changesagainst standard method, suitable for application and computerization [1].

$$\rho(\mu g/g) = \frac{A_{sp}}{A^*_{sp}} \cdot \frac{1}{k_{0,Au}} \cdot \frac{G^*_{th} f + G^*_e Q^*_0(\alpha)}{G_{th} f + G_e Q_0(\alpha)} \cdot \frac{\varepsilon^*_p}{\varepsilon_p} .10^6 \quad (1)$$

The concept of standardization k_0 in the NAA based on the sample irradiation simultaneous with one neutron flux monitor or some ones denoted as n, and the using of composed nuclear constant $k_{0,n}(a)$ determined by experiment [2]:

$$k_{0,n}(a) = \frac{M_n \theta_a \sigma_{0,a} \gamma_a}{M_a \theta_n \sigma_{0,n} \gamma_n} \quad (2)$$

Where M is the atomic mass; θ is the abundance of isotopes; σ_0 is the neutron cross section at the neutron velocity of 2200m/s,γ is the probability of the gamma-ray emission.

Neutron flux monitor is often used being gold.The coefficients k_0 of concerned elements "a" compared to gold $k_{0,Au}$ has been determined by independent experiments for the most important reaction (n,γ) given by De Corte, as follows:

$$k_{0,Au}(a) = \frac{M_{Au} \theta_a \sigma_{0,a} \gamma_a}{M_a \theta_{Au} \sigma_{0,Au} \gamma_{Au}} \quad (3)$$

However, the k_0 method is not limited to gold, it can also be used for other n monitor by definition:

$$k_{0,n}(a) = \frac{k_{0,Au}(a)}{k_{0,Au}(n)} \quad (4)$$

$f = \frac{\phi_{th}}{\phi_e}$ is the ratio of thermal neutron flux and epithermal one [3].

$Q_0(\alpha)$ is the ratio of the resonance integral cross section for thermal neutron spectrum distribution and $\sigma_0$follow rule of $1/E^{1+\alpha}$[7]:

$$Q_0(\alpha) = \frac{I_0(\alpha)}{\sigma_0} = \frac{Q_0 - 0,429}{\overline{E_r}^{\alpha}} + \frac{0,429}{(2.\alpha+1)E_{Cd}^{\alpha}} \quad (5)$$

With $Q_0 = \frac{I_0}{\sigma_0}$ is the ratio of the resonance integral and $\sigma_0$at the neutron velocityat 2200 $m.s^{-1}$; I_0 is the resonance integral of the distribution of epithermal neutron flux in the ideal case of 1/E, α is the coefficient of thermal neutron spectrum deviation; $\overline{E_r}$ is the average effective resonance energy (eV), E_{Cd} is the energy at threshold cadmium (E_{Cd} = 0.55 eV).

G_{th}, G_e are the correction factors of the self-shielding for the thermal neutron and epithermal one, respectively.

$\varepsilon_p$is performance of detector at the peak of the energy.

III. EXPERIMENTS

The System of neutron activation and analysis has been studying at the Laboratory of Nuclear Physics, Faculty of Physics – Engineering Physics, University of Science, Vietnam National University – Ho Chi MinhCity includes the following devices: i) Am–Be neutronsource; ii) automatically sample transfer system; and iii) HPGe spectrometer; and iv) gamma spectrum analysis software.

A. Am–Be neutron source

Neutron source was ^{241}Am – Be isotopic one. Neutrons are produced by the following mechanism: radioactive isotopeof Am^{241}(Half-life of 432 years) decaying α particles was mixed with Be powder to increase neutronradiation with nuclear reaction as follows:

$$\alpha + {}_4Be^9 \rightarrow {}_6C^{12} + {}_0n^1$$

Neutron sources are generated in this way with activity of 7Ci, thermal neutron flux of about $1,5.10^7$ $n.cm^{-2}.s^{-1}$ and neutron energy of 4.7 MeV in average. To create the thermal neutron flux, paraffin was used as inhibitor. The configuration of Am–Be source used for neutron activation consists of two channels: one channel for activation with fast neutrons and the other for thermal neutron activation (Figure 3). Thermal neutrons existdue to the layer of paraffin using to slow neutrons around the sources.

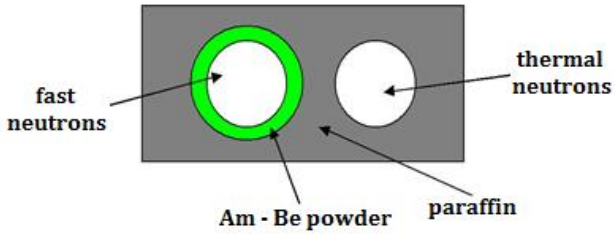

Fig. 3. Configuration of Am–Besource.

B. Automatically sample transfer system

Automatically sample transfer system is shown in Figure 4

Fig. 4. Automatically sample transfer system.

C. Gamma spectrometer offered by Canberra Industries company [8]

The system consists of the following main components: HPGe Detector GC2018 with attached devices, including high-voltage power supply for detector, preamplifier, amplifier, and MCA unit; shielding lead chamber surrounding the detector. When modeling spectrometer, we are only interested in the configuration of the probe, sources and lead shielding chamber.

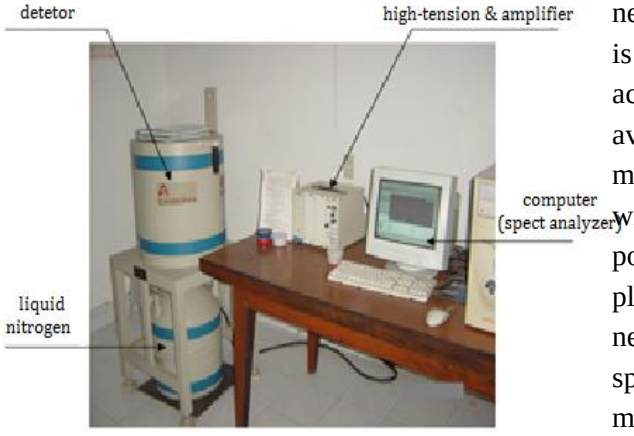

Fig. 5. Gamma spectrometer with using HPGe detector 2018.

D. Sample preparation

Sample preparation consists of standard samples of Au^{197} thin foil in the form of Al-Au alloy (99.99% Au), In^{115} pure foil (element 100%), Mn^{55}in the formMn_2O_3 oxides, Mo^{98}in the formMoO_3 oxides, Br^{81}in the form KBr. The sample with corresponding elementsgiven in Table I.

The sample preparation must be implemented with high precision, and relatively small sample volume to limit the influence of neutron self-shielding. Therefore, the analysis is required for preparing carefully to ensure the achievement of a highly homogeneous sample, avoiding contamination to reduce errors to a minimum. The samplewas weighed accurately with electronic balance, put into a 20 x 40mm^2 polyethylene bag and placed in a cylindrical plastic box, then irradiated in the thermal neutron channel and measured with the spectrometer. In the process of irradiation and measurement, it is needed to interest in the parameters as follows: the irradiating time, measuring time and decaying one and as well as sample.

Table I. Parameters for sample irradiation and measurement

Sample form	Element	Mass(mg)		Content (%)	Time (s)		
		Sample	Element		t_i	t_d	t_m
Au(foil)	Au^{197}	14.000	13.998	99.99	247980	30	7200
In (foil)	In^{115}	31.100	31.100	100.00	247980	30	7200
Mn_2O_3	Mn^{55}	558.600	194.384	34.80	3780	30	4298
MoO_3	Mo^{98}	586.100	393.289	67.10	495360	30	36201.3
KBr	Br^{81}	105.400	71.063	67.42	238200	30	11379

E. Energy calibration, peak width

The energy calibration is usually prepared prior for the measurement to get spectrum and often considered as a part of the measuring system startup. The calibration features include the following steps: measuring the spectrum of a radioactive source emitting know gamma energics; identifying gamma spectrum peaks in the order, supplying corresponding power to the peak determined. Then establishingthe relationship between the

gamma energy and the channel of the first derivative as follows:

$$E = A + B.K \quad (6)$$

Similarly, the calibration of the peak width will contribute to improving the accuracy of the peak area calculations and determine the peak shock. The gamma peak is usually approximate in the form of Gaussian and Full width at half max (FWHM) of the peak which is often matched according to energy in the following form:

$$FWHM = A + B\sqrt{E} \quad (7)$$

F. Activation and analysis data

The selected monitor types include Au-197 or Mo-98, ratio of the thermal neutron flux to the thermal one, the coefficient of thermal neutron spectrum deviations are shown in Table II, and energy, half-life, the k_0, the integral ratio of the resonant elements shown in Table III.

Table II. The score on the thermal neutron flux neutron flux monitor the temperature of Au - 197 or Mo-98 and coefficient of thermal neutron spectrum deviation [4].

Monitor	Coefficient of thermal neutron spectrum deviation, α	Radio of thermal neutron flux toepithermal one, f
Au-197	0.075 ± 0.011	24.73 ± 3.15
Mo-98		25.73 ± 2.20

Table III. Energy, half-life, k_0 coefficient, ratio of the element resonance integral

Element	Isotope	Energy, E (keV)	Half-life, $T_{1/2}$ (h)	$k_{0,Au}$	Ratio of the element resonance integral, $Q_0(\alpha)$
Au^{197}	Au^{198}	411.8	64.68	1.00E+00	13.8012
In^{115}	In^{116m}	138.3	0.9068	1.01E-01	16.2242
		416.9		7.54E-01	
		818.7		3.36E-01	
		1097.3		1.60E+00	
		1293.5		2.29E+00	
		1507.4		2.69E-01	
Mn^{55}	Mn^{56}	846.8	2.579	4.96E-01	0.7836
		1810.7		1.35E-01	
Mo^{98}	Mo^{99}	140.5	65.94	5.27E-04	35.2978
Br^{81}	Br^{82}	554.3	35.3	2.38E-02	13.3368
		619.1		1.45E-02	
		698.4		9.38E-03	
		776.5		2.76E-02	
		827.8		7.99E-03	
		1044		9.14E-03	
		1317.5		8.91E-03	
		1474.9		5.42E-03	

Method of neutron activation analysis using the k_0 INAA was used in determining the element concentration in the sample. But it is determined by the performance of

experimental probes suffering from a number of difficulties, such as some mono-energy gamma emitters with short half-life or Eu^{152}, Ra^{226} isotopes induced coincidence effect or the sources having not enough energy peak for the entire region of interest from 50 keV to 2000 keV. These difficulties can be solved by Monte Carlo simulation method, which ispopularly used to simulating devices, measuring system, the process of nuclear physics. Deteff program is used to simulate the peak performance of the total energy of the detector connected with VGSpec program (Figure 6). The simulation parameters should include: the first, the technical parameters of the detector provided by the manufacturer are shown in Table 4;the second, the source parameters such as geometry (cylindrical shape, form box, Marinelli format), the source material of the tire structure, substrate composition, mass, effective density should be provided;and the third, the filter parameters for the high activity sources [5].

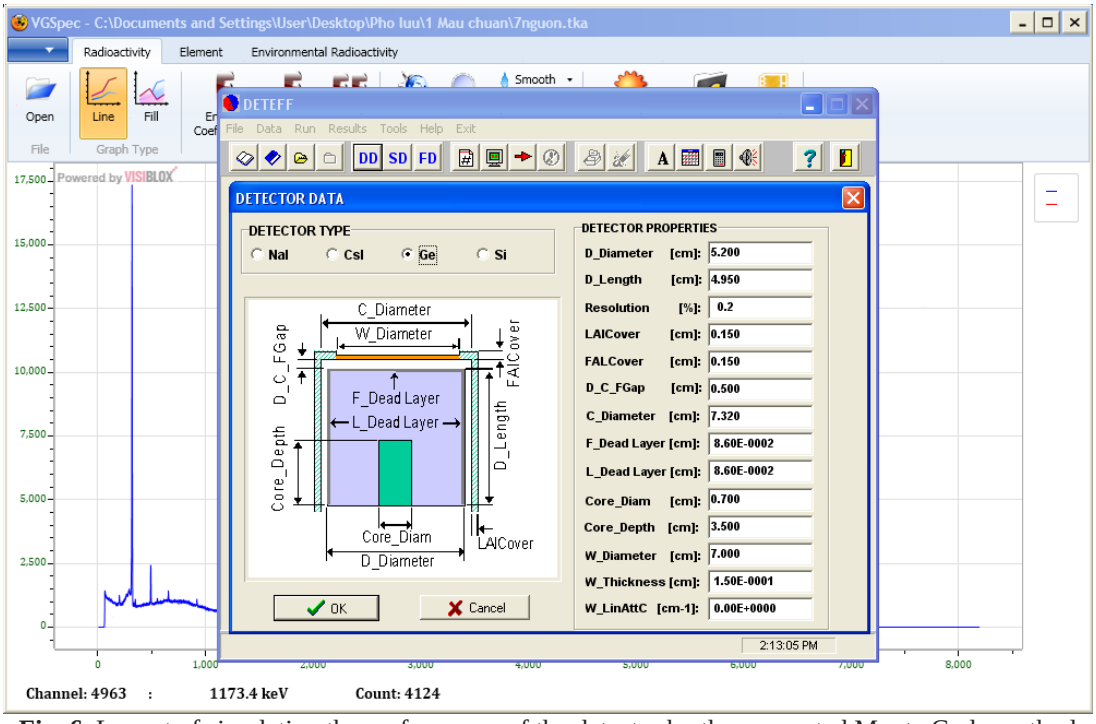

Fig. 6. Layout of simulating the performance of the detector by the connected Monte Carlo method in the VGSpec program.

Table IV. Specifications of HPGe - detector GC2018 provided by the manufacturer [6].

Describe	Size (mm)	Material
Diameter of germanium crystal	52	Germanium
Height of germanium crystal	49.5	Germanium
Diameter of hole	7	
Height of hole	35	
The distance from the window to crystal	5	
Windowthickness	1.5	Aluminum
Outer diameter of detector	76.2	
External junctions	0.86	Li

Internal junctions	0.0003	Bo
External protection class	1.5	Aluminum

IV. RESULTS AND DISCUSSION

Automatic peak Search Package, the elements quality of sample package, the quantitative analysis of sample package forspectrum processing program automaticallyof VGSpec made for elements of Au^{197}, In^{115}, Mn^{55}, Mo^{98}, Br^{81}gave results as follows:

– Forthe monitor of Au^{197} thin foil in the form of Al-Au alloy (99.99% Au):

• Find 6/6 peaks automatically;qualitative determination of Full Match at peak of 6: 138.3 keV, 416.9 keV, 818.7 keV, 1097.3 keV, 1293.5 keV and 1507.4 keV; and element contents are not exceed 1% difference to the levels of in fact form In^{115} foil of pure (100 % component) when provide neutronactivation.

• Find 2/2 peaks automatically;qualitative determination of Full Match at peak of 846.8 keV and 1810.7 keV; and element contents are not exceed 1% difference to the actual content of Mn^{55}sample in the form of Mn_2O_3 oxides when provide neutron activation.

– For Mo^{98} monitor in the form of MoO_3oxides: Find 8/8 peaks automatically;qualitative determination of Full Match at peak of 8:554.3 keV, 619.1 keV, 698.4 keV, 776.5 keV, 827.8 keV, 1044.0 keV, 1317.5 keV and 1474.9 keV; and element contents coincide perfectly with the actual content in the form of KBr salt when provide neutron activation.

Table V. Results of auto spectrum searching, and qualitative determination of elements in the sample by VGSpec programs for elements ofAu^{197}, In^{115}, Mn^{55}, Mo^{98} and Br^{81}.

Isotope	Peak, K	Energy, E (keV)	Search	Quantilative	Peak area, N_p		Content, ρ (%)		Remarks
					N_p	ΔN_p(%)	ρ	Δρ(%)	
Au^{198}	1722	411.8	x	Au^{197}	6491	3.60	99.99		Monitor
In^{116m}	563	138.3	x	In^{115}	2106	10.24	99.16	10.76	
	1745	416.9	x	In^{115}	5656	4.03	99.96	5.40	Countρ
	3451	818.7	x	In^{115}	1321	17.72	94.90	17.16	
	4632	1097.3	x	In^{115}	6178	3.67	100.81	5.18	Countρ
	5465	1293.5	x	In^{115}	7400	0.82	100.83	3.72	Countρ
	6375	1507.4	x	In^{115}	817	3.61	103.06	5.25	
					Average content		100.53	4.77	
					Factual content		100.00		
					Ratio		**1.01**		
Mn^{56}	3569	846.8	x	Mn^{55}	12935	0.46	35.02	1.27	Countρ
	7660	1810.7	x	Mn^{55}	1582	2.56	33.13	1.46	
					Average content		35.02	1.27	
					Factual content		34.80		
					Ratio		**1.01**		
Mo^{99}	562	140.5	x	Mo^{98}	8501	1.18	67.10		Monitor
Br^{82}	2320	554.3	x	Br^{81}	3992	1.80	66.27	4.62	Countρ
	2594	619.1	x	Br^{81}	2600	2.34	68.03	3.40	Countρ

	2929	698.4	x	Br^{81}	1137	3.52	66.94	3.27	
	3259	776.5	x	Br^{81}	3513	1.83	68.40	2.02	Countρ
	3477	827.8	x	Br^{81}	1089	3.60	65.59	10.25	
	4391	1044	x	Br^{81}	847	4.26	69.66	9.25	
	5548	1317.5	x	Br^{81}	824	4.02	70.69	7.30	
	6214	1474.9	x	Br^{81}	449	16.58	64.90	8.94	
						Average content	67.57	3.35	
						Factual content	67.42		
						Ratio	**1.00**		

V. CONCLUSION

The qualitative and quantitative determination of elementin the sample by the program was made to VGSpec with Au^{197} element, In^{115}, Mn^{55}, Mo^{98} and Br^{81} elements with selected monitors is Au^{197} thin foil in the form of Al-Au alloy (99.99% Au), Mo^{98} in the form of MoO_3 oxides. The results show that the element quantitative determination of elements in sample package matches well with In^{115} pure leaf sample (element 100%), Mn^{55} in the form of Mn_2O_3 oxides, Br^{81} in the form of KBr when happening neutron activation (less than 1% difference).

REFERENCES

[1] F. De Corte, "The k_0 – Standandardization Method: A Move to the Optimization of Neutron Activation Analysis", *Ph.D. Thesis*. GENT University, (1987).

[2] F.D. Corte, A. Simonits, "Recommended nuclear data for use in the k_0 standardization of neutron activation analysis", *Atomic Data and Nuclear Data* Tables 8, pp. 47–67, (2003).

[3] H.T.Phuong, M.V.Nhon, "A fast Method of Determination of and f Factors in the k_0-INAA Standardization", *7th National Conference on Nuclear Science and Technology*, Da Nang City, (2007).

[4] Huynh T. Phuong, Mai V. Nhon, Luu D.H. Oanh, "Development of k_0-INAA standardization method by neutron activation with Am-Be source", *Applied Radiation and Isotopes*, Vol. 70, pp. 478–482, (2012).

[5] N.Cornejo Diaz, M. Jurad Vargas, "DETEFF: An improved Monte Carlo computer program for evaluating the efficiency in coaxial gamma-ray detectors", *Nuclear Instruments and Methods in Physics Research*,Vol. 586, pp. 204–210, (2008).

[6] Tran Thien Thanh, Chau Van Tao, Truong Thi Hong Loan, Mai Van Nhon, "Research curve peak performance of the total energy use DETEFF program ", *Journal of Science, HCMC University of Pedagogy*, No. 33, pp. 88-93, (2012).

[7]http://www.en.wikipedia.org/wiki/Neutron_activation_analysis

[8] http://www.canberra.com

[9] www.laraweb.free.fr