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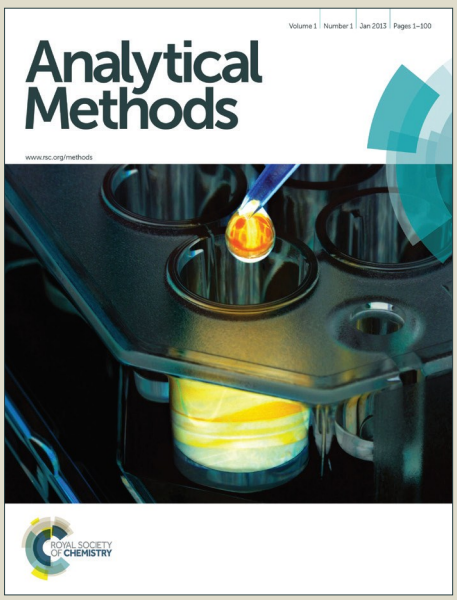

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Neutron Activation Analysis for Cement Elements Using IECF Device as a High Energy Neutron Source

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Abstract- Neutron Activation Analysis (NAA) is an important technique for quantitative and qualitative multi-element analysis. Inertial Electrostatic Confinement Fusion (IECF) device is known as a fast and monoenergetic neutrons generator. In this study, NAA for cement elements using IECF facility as a high energy neutron source was investigated. The Iranian IECF device was simulated using MCNPX code version 2.7 and 'ACT card' was used to consider the induced delayed gamma-ray spectra during delayed gamma NAA (DGNAA). The peaks related to Si, Fe, Ca and Al were distinguished precisely which show the applicability of IECF as an appropriate neutron source in order to DGNAA analysis for cement elements.

Keywords: IECF Device, Modeling, Monte Carlo Simulation, Neutron Activation Analysis.

1- Introduction

Iranian Inertial Electrostatic Confinement Fusion (IR-IECF) device is a compact and simple structure for nuclear fusion researches by electrical discharge.[1] It can operate in

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the pulsed or continuous mode and consists of two concentric (or coaxial) electrodes that usually the central one (cathode) is negatively high-voltage-biased and the outward electrode (anode) is grounded as shown in Fig. 1.

In this configuration, strong electric fields between electrodes lead to ionization of filling gas and then accelerate the created ions toward the center, where the electrons are placed in the opposite direction. As a result, rather hot and dense plasma is formed in the center of cathode. If using deuterium or mixture of deuterium-tritium gas, the nuclear fusion reactions occur and therefore neutron is produced, which are the results of the beam-target and beam-beam interactions.

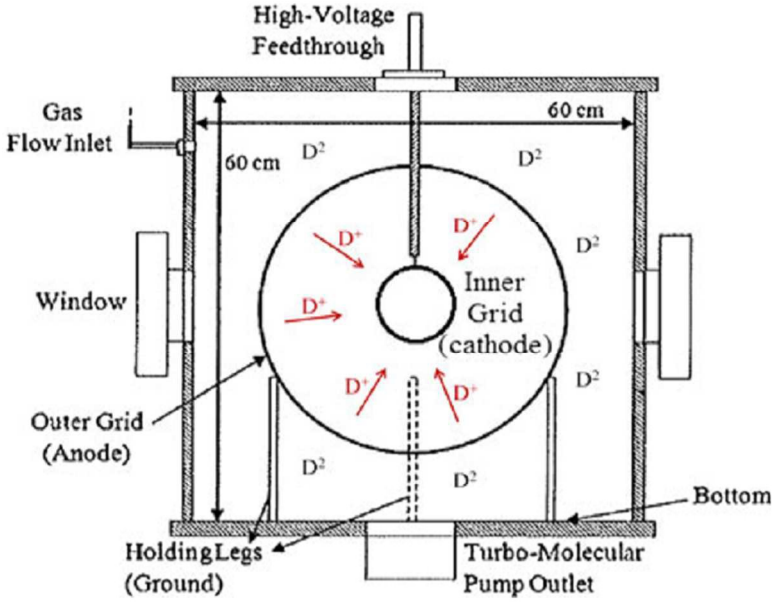

Fig. 1. Schematic of IR-IECF device.[2]

IECF is considered as a source of hot and dense plasma, highly energetic ions and fast neutrons (when using deuterium or mixture of deuterium-tritium gas). IECF device is an excellent apparatus because of its ability to generate fast and monoenergetic neutrons with high-flux from a small source for many applications, such as: medicine (e.g. boron neutron capture cancer therapy),[3-5] radiography or tomography of thick materials, space

propulsion system,[6] inspection system and explosive landmine detection,[7] neutron activation analysis, mine and petroleum exploration, security screening, etc.

Neutron Activation Analysis (NAA), discovered in 1936, is an important technique for quantitative and qualitative multi-element analysis of major, minor and trace elements.[8] The practical and technical foundation of the NAA doesn't depend on chemical properties of the element but on the nuclear properties of the isotopes. This point could be the most distinctive property for this method. So, elements with similar chemical properties, which are difficult to separate and distinguish them by usual chemical methods, could be analyzed qualitatively and quantitatively by NAA hastily and accurately.[9] Sequential instrumental neutron activation analysis allows quantitative measurement of up to about 35 elements in small samples of 5 to 100 mg. Additionally the lower detection limit is in parts per million or parts per billion, depending on the element.[10] Some of the advantages of NAA are no need for sample preparation, is not destructive, several elements are measured and the precision is larger than other methods.

Determination of concentration of certain elements in cement is very useful in evaluating their suitability as building material. Worldwide, the demand and use of cement is very large, thus several studies using NAA have been carried out to assess the elemental concentration; the NAA has been carried out with a nuclear reactor and with an isotropic neutron source.[9,11,12] Since simulating delay gammas is a new feature in MCNPX, version 2.7, there is no work in this field of study. In this work, the IR-IECF facility was simulated and the NAA about the cement certain elements on the basis of produced neutrons was investigated.

2. Monte Carlo Simulation

MCNPX is a Fortran 90 Monte Carlo radiation transport computer code that transports nearly all particles at nearly all energies for nearly all applications. It has capabilities that include three-dimensional (3D) geometry modeling, continuous-energy transport,

and transport of 34 different type of particles, a variety of source-term and tally options and interactive graphics. The new capabilities of the latest version, MCNPX 2.7.E, are beyond those in MCNPX 2.6.0.[13] In this work the new feature, 'ACT card', of MCNPX version 2.7 has been used to simulate the induced delayed gamma-ray spectra during DGNAA analysis.

The MCNPX activation capability enables the study of neutron induced reactions that result in delayed gamma-ray emission. This feature permits the automated execution of radiation transport simulations of delayed gamma-ray emission spectra at discrete (line) energies created by the products of neutron fission and activation.[14] Delayed gamma rays spectra produced along thermal neutron capture during (n, p) reactions, were estimated with the MCNPX code.

Elements of cement material have been defined separately at first, and then totally with together to form a compound in the MCNPX input file with a specific weight fraction for each element and related density. Table 1 shows isotopes, abundance, and density of each cement raw element separately. Schematic of the geometry used for producing delay gammas is depicted in Fig.1. Only photons and neutrons were considered and importance factor 1 was assigned for all cells. A cylindrical sample with dimensions of 15 cm diameter and 3 cm height irradiated by IECF neutron source with 14.1 MeV and 2.45 MeV respectively for comparison. Delayed gamma-ray spectra up to 4 MeV was computed using F1 tally (current on surface) on the sphere centered at the target with 50 cm radius. Statistical uncertainty associated with the Monte Carlo transport simulation results presented in this paper, with 10 million histories per simulation run, is less than 5% in each bin. The photon spectra were calculated in 4000 energy channels, which is sufficient to simulate a typical multichannel analyzer connected to a HPGe detector. The detector simulated in this study was a GMX series HPGe coaxial detector system. The resolution (FWHM) at 1.33 MeV, ^{60}Co is 2.02 KeV and peak-to-Compton ratio, ^{60}Co is 59:1 and relative efficiency at 1.33 MeV, ^{60}Co is 40%. The crystal has a diameter of 60.6 mm and length of 66.9 mm and also has a 0.3 micron Ge/B dead layer and 700-

micron Ge/Li dead layer. In Monte Carlo MCNPX code, Gaussian energy broadening (GEB) is a special treatment for tallies to better simulate a physical radiation detector in which energy peaks exhibit Gaussian energy broadening. GEB is called by entering FTn card in the input file of MCNPX to account for any possible noises that may have influence on FWHM in order to create a nearly real simulation. The desired FWHM that is specified by the user-provided constants, a, b, and c, shows a nonlinear response:

$$FWHM = a + b\sqrt{E + cE^2} \qquad (1)$$

Where E is the incident γ -ray energy. The units of a, b, and c are MeV, $MeV^{½}$, and MeV^{-1}, respectively. The following parameters in the GEB option [15] have been used for this purpose:

a= 5.86828E-4 MeV b= 3.95113E-4 $MeV^{½}$ c= 7.46793 MeV^{-1}

A cooling and counting time of 30 seconds and 1 hour have been considered for post calculations respectively. It is important to note that the HPGe detector simulated here with its specific features which are described in the reference 16 has the resolution to resolve the two 843 keV and 846 keV peaks related to Mg-27 and Mn-56 respectively,[16] which are occurred in real experiment.

Table 1. Elemental properties of cement material [15]

Element	Isotopes	Abundance %	Density (g/cm^3)
Silicon	Si-28	92.23	2.33
	Si-29	4.67	
	Si-30	3.1	
Iron	Fe-54	5.8	7.87
	Fe-56	91.72	
	Fe-57	2.2	
	Fe-58	0.28	
Calcium	Ca-40	96.94	1.54

	Ca-42	0.647	
	Ca-43	0.135	
	Ca-44	2.086	
	Ca-46	0.0004	
	Ca-48	0.187	
Aluminum	Al-27	100	2.69

The results of simulations are illustrated in Figures 2, 3, 4 and 5 for cement raw elements including Al, Fe, Si, and Ca respectively and in Figure 6 for a typical cement sample.

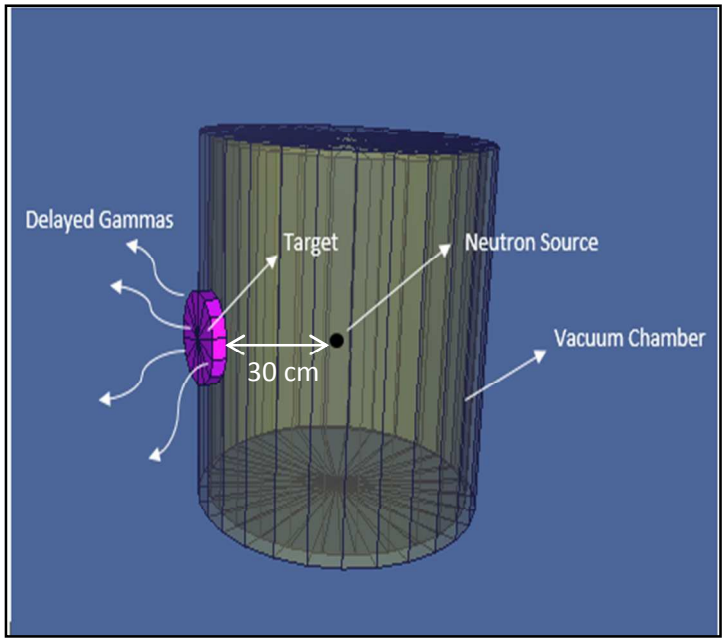

Fig. 2. Schematic of the simulated IECF source and sample geometry in MCNPX used for generating delayed gamma spectra

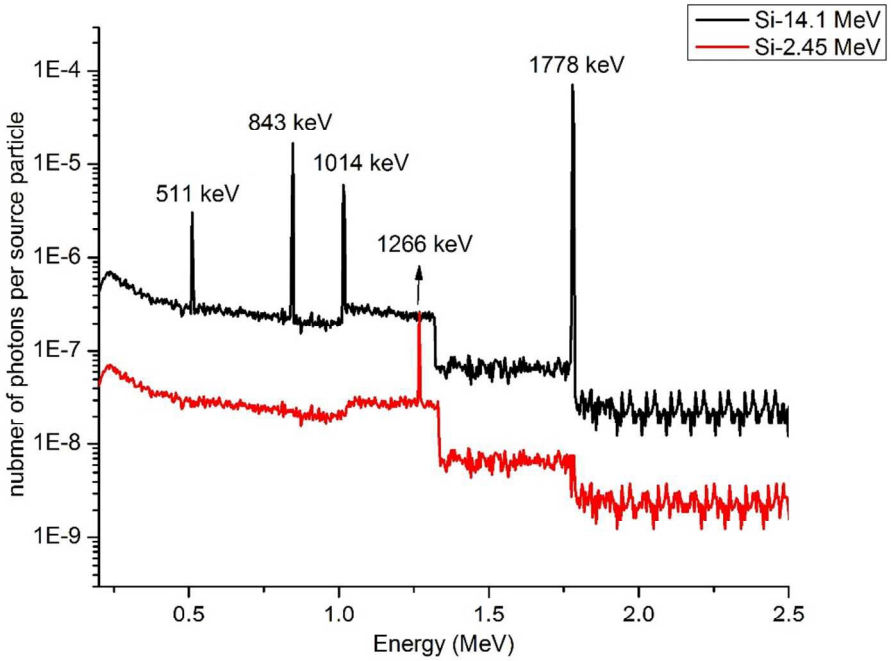

Fig. 3. Gamma spectra of Silicon using 14.1 MeV and 2.45 MeV neutron source

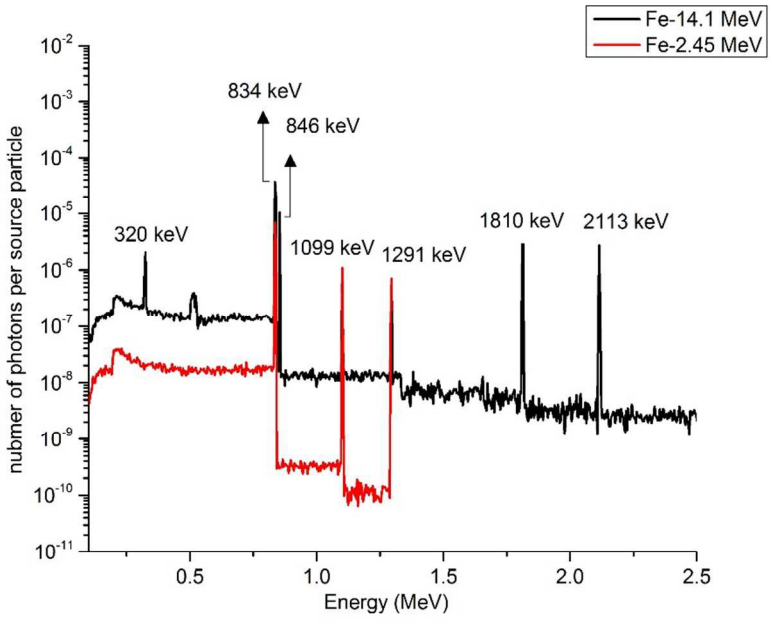

Fig. 4. Gamma spectra of Iron using 14.1 MeV and 2.45 MeV neutron source

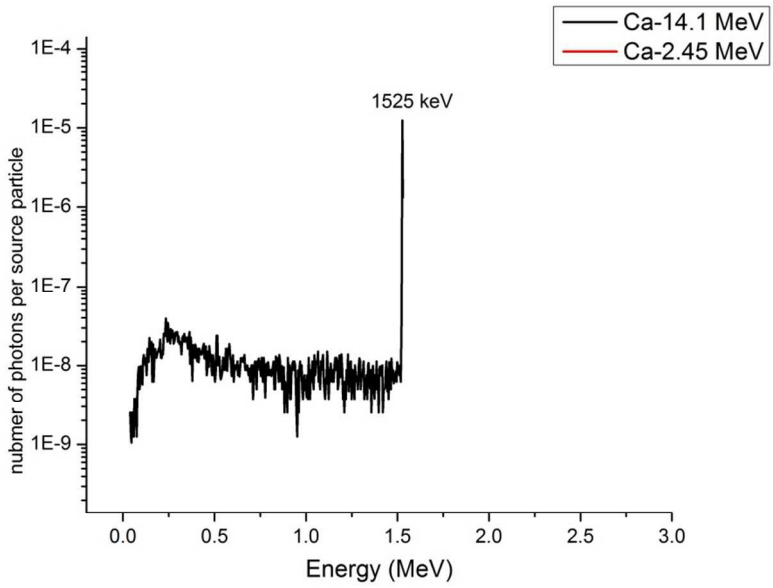

Fig. 5. Gamma spectra of Calcium using 14.1 MeV and 2.45 MeV neutron source

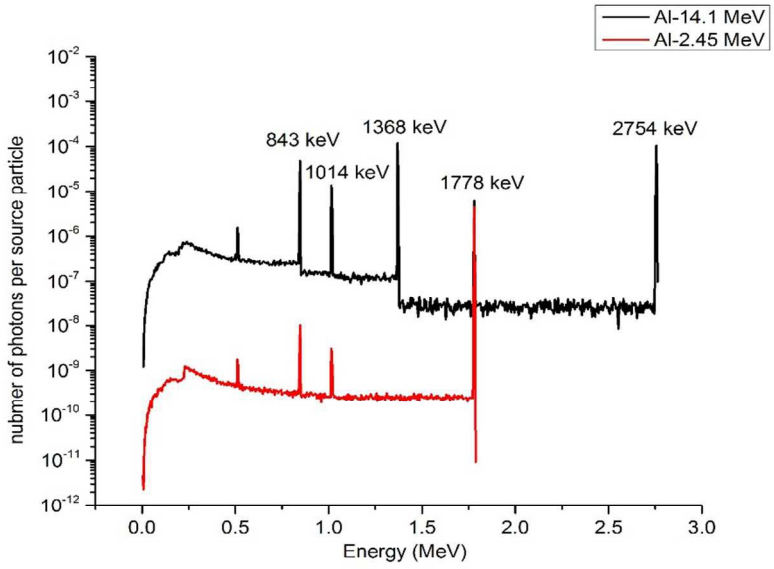

Fig. 6. Gamma spectra of Aluminum using 14.1 MeV and 2.45 MeV neutron source

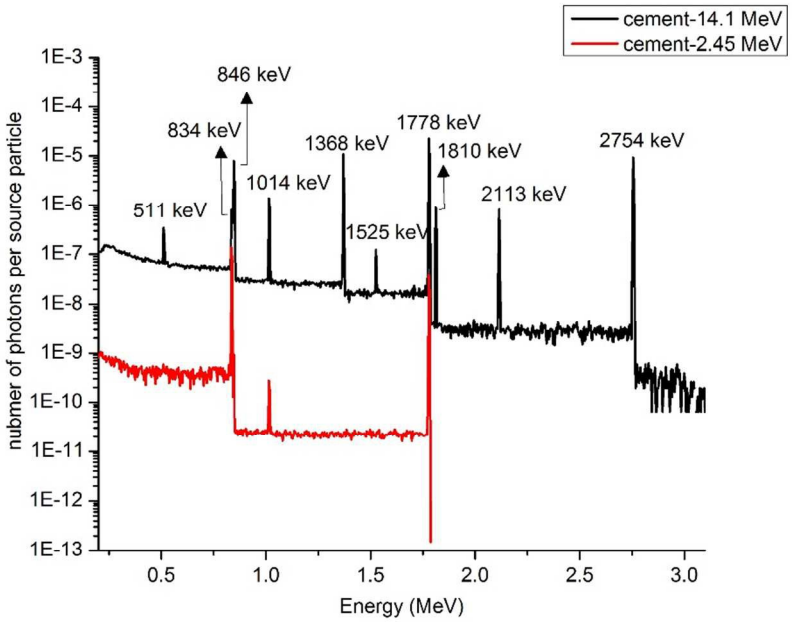

Fig. 7. Gamma spectra of a typical cement sample using 14.1 MeV and 2.45 MeV neutron source

3. Results and Discussion

Table 2 shows isotopes, cross sections, production modes, product half-lives and their relevant gamma ray energies of each cement element. Since for the interactions such as (n, p) and (n, α), neutron should have at least a minimum energy so that the interactions take happen, all of the threshold energies related to the elements have been extracted. Table 3 shows the required energy which is needed for threshold reactions.

Table 2. Nuclear data related to cement elements including their isotopes.[15]

Target Isotope	Cross Section			Production Modes	Product Half-life	Gamma Energy (keV)
	th (b)σ	RI(b)	$\bar{\sigma}$(mb)			
Si-28	-	-	6.4	(n, p) Al-28	2.24 m	1778
Si-29	-	-	3.01	(n, p) Al-29	6.56 m	511-1273
Si-30	0.108	0.106	-	(n, γ) Si-31	2.62 h	1266
Si-30	-	-	0.155	(n, α) Mg-27	9.46 m	843-1014
Fe-54	-	-	82.5	(n, p) Mn-54	312 d	834
Fe-54	-	-	0.6	(n, α) Cr-51	27.7 d	320
Fe-56	-	-	1.07	(n, p) Mn-56	2.58 h	846-1810-2113

Fe-58	1.31	1.28	-	(n, γ) Fe-59	44.5 d	142-192-1099-1291
Ca-46	0.62	0.81	-	(n, γ) Ca-47	4.54 d	807-849-1297
Ca-48	1.12	0.5	-	(n, γ) Ca-49	8.72 m	3084-4072
Al-27	0.226	0.16	-	(n, γ) Al-28	2.24 m	1778
Al-27	-	-	0.725	(n, α) Na-24	14.96 h	1368-2754
Al-27	-	-	4	(n, p) Mg-27	9.46 m	843-1014

Table 3. The required energy for threshold reactions which occur during activation analysis.[17]

Isotope	Interaction Mode	Threshold Energy (MeV)
Si-28	(n, p)	4
Si-29	(n, p)	3
Si-30	(n, α)	4.34
Fe-54	(n, p)	0.699
Fe-54	(n, α)	3
Fe-56	(n, p)	2.96
Al-27	(n, α)	3.247
Al-27	(n, p)	1.896

Gamma spectra of cement elements are nearly in a good agreement with nuclear data in table 2. There are some minor discrepancies in the peaks from the table 2 which could be explained as follows:

1- Neutron energy for the specific reaction is lower than the threshold or required energy. for example, in Aluminum spectrum (Figure 4), 1368 keV and 2754 keV peaks related to the Al-27 (n, α) Na-24 interaction with 3.247 MeV threshold energy of neutron only can be seen in curve related to 14.1 MeV neutron source which suitably satisfies it.

2- In the case which there is small amount of target element/isotope, obviously there would be no related peak(s). For example isotopic abundance of Ca-46 is 0.0004 percent in a total bulk sample of Calcium, so it just could be seen in high thermal neutron fluxes such as in a reactor neutron source which reaction rate of this isotope with thermal neutron increases dramatically as a result of rich amount of thermal neutrons in a reactor.

Neutron production rate of IECF device for deuterium- tritium is 10 times more than deuterium-deuterium.[18] According to the obtained spectrums and neutron production rate of IECF, using of deuterium- tritium mixture instead of deuterium as fuel of IECF device can increase precision and accuracy of NAA for cement elements. Also, because of mono-energetic neutrons, absence of interference gamma rays and time independent neutron yield, using of IECF device instead of other common neutron sources (radioisotope and nuclear reactor) increases the precision of NAA method.

4. Conclusions

The IECF device was used in order to DGNAA for cement elements. The neutrons of D-D (2.45 MeV) and D-T (14.1 MeV) reactions were considered for activating of target elements. According to the (n, α) and (n, p) threshold reactions, produced neutron from D-T reaction is better than D-D reaction. Usage of the IECF device instead of radioisotope source in order to NAA is safer, easier and more effective.

References

1. V. Damideh, A. Sadighzadeh, A. Koohi, A. Aslezaeem, A. Heidarnia, N. Abdollahi, F. Abbasi Davani, R. Damideh, J Fusion Energy, 2012, 31, 109-111.

2. E. Haji Ebrahimi, R. Amrollahi, A. Sadighzadeh, M. Torabi, M. Sedaghat, R. Sabri, B. Pourshahab, V. Damideh, J Fusion Energy, 2013, 32, 62–65.

3. G. L. Kulcinski, Fusion Technology, 1998, 34, 477.

4. R. P. Ashley and the UW IEC Team, 6th U.S.-Japan IEC Workshop, Japan, 2003.

5. R. P. Ashley, G. L. Kulcinski, J. F. Santarius, S. Krupakar Murali, G. Piefer, and R. Radel, Fusion Technology, 2001, 39, 546.

6. H. Momota, G. H. Miley, J. Nadler, J. Appl. Phys, 2004, 96.

7. K. Yoshikawa, K. Masuda, T. Takamatsu, S. Shiroya, T. Misawa, Y. Takahashi, E. Hotta, K. Yamauchi, M. Ohnishi, H. Osawa, The 2nd Joint International Conference on Sustainable Energy and Environment, Thailand, 2006.

8. D. T. Win, Assumption University Journal of Technology, 2004, 8, 8-14.

9. R. J. Muhyedeen Bahjat, F. I. Kanbour, S. M. Al-Jubori, *Iraqi J. Sci*, 2001, 42, 1.

10. Neutron Activation Analysis information document, Oregon State University, USA, 2003, http://ne.oregonstate.edu/facilities/radiation_center/naa.html.

11. M. Ali-Abdallah, N. A. Mansour, M. A. Ali, M. Fayez-Hassan, Advances in Applied Science Research, 2011, 2, 613-620.

12. Y. S. Khrbish, I. O. Abugassa, N. Benfaid, A. A. Bashir, Journal of Radioanalytical and Nuclear Chemistry, 2007, 271, 63–69.

13. D. B. Pelowitz, MCNPX 2.7.E Extensions, Los Alamos National Laboratory Report LA-UR-11-01502, 2011.
14. J. W. Durkee Jr, M. R. James, G. W. McKinney, H. R. Trellue, L. S. Waters, W. B. Wilson, Progress in Nuclear Energy, 2009, 51, 828–836.
15. Practical aspects of operating a neutron activation laboratory, International Atomic Energy Agency IAEA-TECDOC-564, 1990.
16. E. Eftekhari Zadeh, S. A. H. Feghhi, E. Bayat, G. H. Roshani, Journal of Experimental Physics, 2014, 4.
17. Evaluated Nuclear Data File (ENDF), Software Version of 2014.10.27, ENDF/B-VII.1.
18. G. H. Miley, S. Krupakar Murali, Inertial Electrostatic Confinement (IEC) Fusion Fundamentals and Applications, Springer, 2014.