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Inter-comparison of Accelerator Based-fast Neutron Sources for their Application in PGNAA Analysis

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ABSTRACT. The Prompt Gamma Ray Neutron Activation Analysis (PGNAA) technique is a multi-elemental analysis technique and is used in industrial process control. The performance of a PGNAA system depends upon neutron energy spectrum. Recently a PGNAA facility was designed and tested at King Fahd University of Petroleum and Minerals (KFUPM), Dhahran, Saudi Arabia to analyze bulk material samples. The facility was tested successfully with the analysis of cement and concrete samples. It is desired to improve the performance of the KFUPM PGNAA facility by replacing its D(d,n) reaction based 2.8 MeV neutron source with a more efficient neutron source. Several studies have been undertaken, in which the performance of the KFUPM setup has been calculated for the radioisotope-neutron sources-based PGNAA setup as well as accelerator-based PGNAA setup. In accelerator-based PGNAA setup, performances calculations were carried out for accelerator-based PGNAA setup utilizing ^{9}Li(p,n) and ^{3}H(p,n) and ^{3}H(d,n) reactions to produce fast neutrons. In the following, the results of the accelerator-based PGNAA setup are compared with each other.

Keywords. PGNAA, ^{3}H(p,n) and ^{3}H(d,n) reactions Neutrons, Cement sample, Monte Carlo calculations

Introduction

Prompt Gamma Ray Neutron Activation Analysis (PGNAA) technique is a fast and non-destructive technique used to measure elemental composition of bulk samples [1-12]. Particularly it is used in cement manufacturing plant to determine lime, silica, alumina, and iron oxide concentrations in cement raw materials [1-8, 11]. The development of a non-destructive elemental analysis for cement and concrete samples is also highly desired for corrosion studies of concrete building structures [13]. There are two types of prompt gamma ray analyzers namely; radioisotope neutron source-based prompt gamma ray analyzers and accelerator-based prompt gamma ray analyzers [11]. The radioisotope neutron source-based prompt gamma ray analyzers, which utilize Am-Be or ^{252}Cf [7-9]

neutron source to irradiate sample, have certain disadvantages namely; high gamma ray dose, frequent replacement of neutron source due to its finite half-life and permanent radiation hazards. An alternative to a radioisotope neutron source-based prompt gamma ray analyzer is accelerator-based prompt gamma ray analyzers, where an accelerator, which generates fast neutrons produced via a nuclear reaction. The fast neutrons are generally produced via D(d,n) or T(d,n) reactions[3, 11]. The accelerator-based prompt gamma ray analyzer has certain advantages over the radioisotope neutron source-based prompt gamma ray analyzers. Due to the controlled mechanism of neutron production, accelerator-based prompt gamma ray analyzers have radiation dose only, which is less radiation dose. Recently compact accelerators are produced with physical size comparable with those of radioisotope nentron source-based prompt gamma ray analyzer. In fact neutron flux in the accelerator-based prompt gamma ray analyzer is higher than those in radioisotope neutron source-based prompt gamma ray analyzers. In short an accelerator based prompt gamma ray analyzer is better or at least comparable with that of radioisotope neutron source-based prompt gamma ray analyzer.

Recently a PGNAA facility was designed and tested at King Fahd University of Petroleum and Minerals (KFUPM), Dhahran, Saudi Arabia to analyze bulk material samples [3]. The facility was tested successfully with the analysis of cement and concrete samples [1, 4]. The prompt gamma rays are produced via capture of thermal neutrons, produced by scattering of fast neutrons in a moderator surrounding the sample. The facility utilizes the 2.8 MeV neutrons from the D(d,n) reaction to produce fast neutrons to irradiate the bulk samples [3]. It is desired to improve the performance of the KFUPM PGNAA facility by replacing its D(d,n) reaction based 2.8 MeV neutron source with a more efficient neutron source. Several studies have been undertaken, in which the performance of the KFUPM setup has been calculated for the radioisotope-neutron sources-based PGNAA setup as well as accelerator-based PGNAA setup [2, 5, 6] In accelerator-based PGNAA setup, performances calculations were carried out for accelerator-based PGNAA setup utilizing ^{6}Li(p,n) [2] and ^{3}H(p,n) [6] and ^{3}H(d,n) [5] reactions to produce fast neutrons. For the radioisotope-neutron sources-based PGNAA setup, calculations were carried out for ^{252}Cf neutron source [5] based PGNAA setup and ^{241}Am-Be source based setup [12]. These studies have revealed very interesting feature of these PGNAA setups. In the following, the results of the accelerator-based PGNAA setup are compared with each other.

2. Geometry of the PGNAA setup

The basic geometry of the simulated PGNAA setup, is the one used for design of 2.8 MeV neutron based PGNAA setup at KFUPM [3]. It mainly consisted of a cylindrical sample enclosed in a cylindrical high-density polyethylene moderator. The sample fits into a hollow cavity, created at one end of the moderator. The moderator-sample assembly is placed between a γ-ray detector and a 2.8 MeV neutrons source. In order to prevent undesired gamma rays and neutrons from reaching the detector, lead and paraffin shielding are inserted between the moderator and the detector. Figure 1 shows a schematic representation of the experimental PGNAA setup [3]. The neutron shielding is made of a mixture of paraffin and lithium carbonate mixed in equal weight proportion. Fast neutrons are thermalized in two regions of the moderator, the first region is located between the neutron source and the sample front end (front moderator) and second region is the moderator reflector collar surrounding the sample along its length. The fast neutrons that

escape from the front moderator region, are reflected by the collar into the sample region. The length of the moderator is defined by the sum of the sample length and the front thickness of the moderator. The outer radius of the moderator is defined by the sum of sample radius and the thickness of the collar.

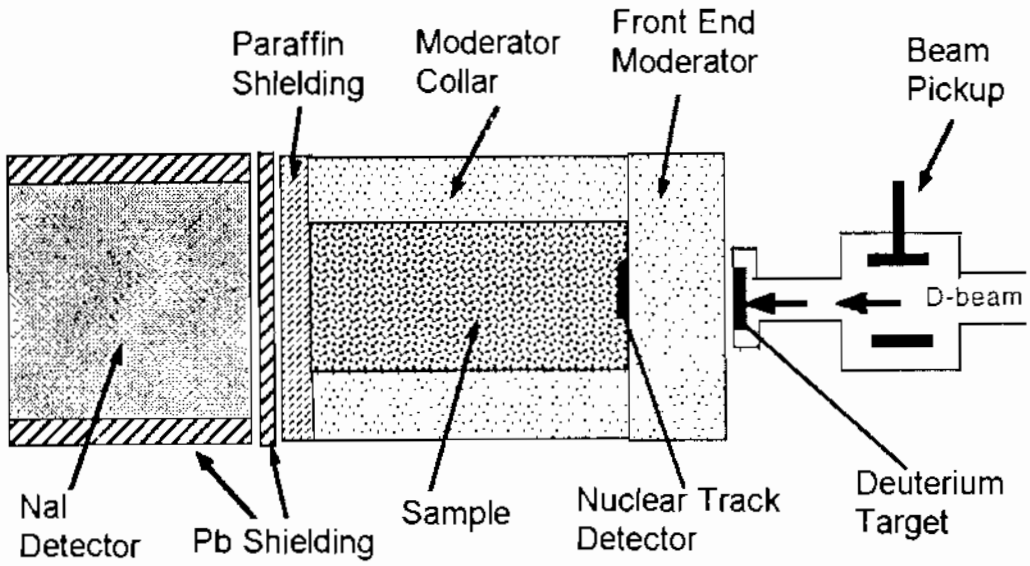

Fig. 1. Schematic representation of the 2.8 MeV based PGNAA setup [4] used to measure the thermal neutron and prompt gamma ray yield. In thermal neutron measurements, the sample is replaced by a Nuclear Track Detector (NTD) placed behind the front moderator [3].

Associated with each size of the moderator, is an optimum value of the sample radius that can produce maximum yield of the prompt γ-rays which can be determined from Monte Carlo calculations. Due to its high hydrogen concentration and easy machining properties, high-density polyethylene was chosen as the moderator material. Photons were detected through use of a cylindrical 25.4 cm x 25.4 cm (diameter x height) NaI γ-ray detector. For initial calculations, the sample length was assumed to be 14 cm, corresponding to the saturation length at which the sample produces maximum yield of γ-rays.

These calculations were carried out using Monte Carlo code MCNP4B2 [16] utilizing a 1 GHz Pentium III PC. For simulation study, the moderators and sample cells were divided into sub-cells of 1 cm thickness. This allowed study of the transport of the neutrons of appropriate statistical weight to the next adjacent cell, without any loss. The simulations were carried out for a dry Portland cement sample. The bulk density of the sample was taken as 1.6 g/cm^3. Notwithstanding the fact that the moisture content of dry cement generally amounts to up to 1 wt %, due to its absorbency the moisture content of cement samples were assumed to be 5 wt % [7]. To attain a statistical uncertainty of the order of 1%, some 45 to 50 minutes were required in order to calculate neutron yield, while for prompt gamma ray calculations about 2 hours were required to calculate the yield of the most prominent calcium line.

3. Comparison of geometry parameters of the accelerator based PGNAA setups

The Monte Carlo calculations were carried out for the four neutron sources given in the Table 1. The difference between them is mainly in the mean energy of their neutrons. Also neutrons from the ^{7}Li(p,n) [15] and ^{3}H(p,n) [16] reactions near the reaction threshold energy are emitted in forward cone with an energy distribution similar to Maxwellian distribution for kT values corresponding to energies of 25 and 52 keV respectively. This type of keV energy neutron source with a collimated beam makes it useful for their application in PGNAA studies, especially if the elemental analysis is carried out through thermal neutron capture. In such a setup neutrons are moderated in external moderators. With the collimated beam of keV neutrons, the external moderator required would be of a smaller size, and less shielding would be needed in the PGNAA setup. The moderation efficiency of the moderator is higher for keV neutrons than for MeV neutrons. This will result in a higher yield of thermal neutrons, thereby increasing the sensitivity of the PGNAA setup.

Table 1. Important features of various neutron producing nuclear reactions

Reaction	Energy Spectrum	Neutron Mean Energy	Function used for Energy Spectrum
^{2}H (d,n)	Mono-energetic	2.8 MeV	-
^{3}H(d,n)	Mono-energetic	14 MeV	-
^{7}Li(p,n)	Maxwellian	25 keV	$Ee^{[E/kT]}$, with kT=25 keV
^{3}H(p,n)	Maxwellian	52 keV	$Ee^{[E/kT]}$, with kT=52 keV

A comparison of the optimum geometry parameters for the ^{7}Li(p,n) [2] , ^{3}H(p,n) [6] , ^{3}H(d, n) reaction [5] and the 2.8 MeV neutrons [3] based PGNAA setup is given in Table 2. It shows that the optimum radius of the sample for all the setup studied, are not much different from each other. Thus said, the maximum yield of the prompt gamma rays from the samples has been observed for a sample with a radius of 7-9 cm. The effect of neutron source energy on the geometry of a PGNAA setup is reflected in the smaller values of the front moderator thickness and the sample length calculated for the ^{7}Li(p,n) reaction based setup. As shown in Figure 2 [2] , the distribution of the prompt gamma rays as a function of the front moderator thickness of the ^{7}Li(p,n) reaction based setup is much sharper than that of the 2.8 MeV neutrons based PGNAA facility. Its optimum thickness is almost 25 % smaller than that of the 2.8 MeV neutrons based facility. This may be due to the fact that lower energy neutrons can be moderated in the shorter thickness of the absorber material. The optimum length of the sample of the ^{7}Li(p,n) reaction based setup, as shown in Figure 3 [2] , is smaller than that of the 2.8 MeV neutrons based setup. It supports the idea of the shorter sample length for lower neutron energy based PGNAA setup. Since the sample also acts as an additional moderator of the neutrons, one expects shorter length of a bulk sample of a PGNAA setup utilizing neutrons with lower energy, such as the ^{7}Li(p,n) reaction based PGNAA setup. As shown in Figures 4 and 5 [6], similar trends are also observed for ^{3}H(p,n) reaction based PGNAA setup.

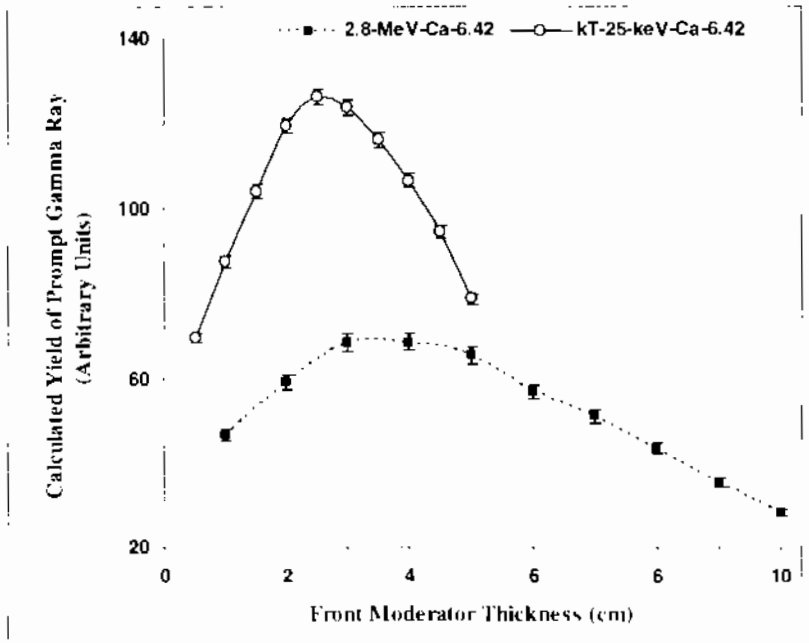

Fig. 2. Calculated yield of 6.42 MeV prompt gamma rays from calcium in a Portland cement sample for a ^{7}Li(p,n) reaction [2] and 2.8 MeV neutron [3] based PGNAA setup plotted as a function of the cement sample radius. The data points are connected by lines to show the trend.

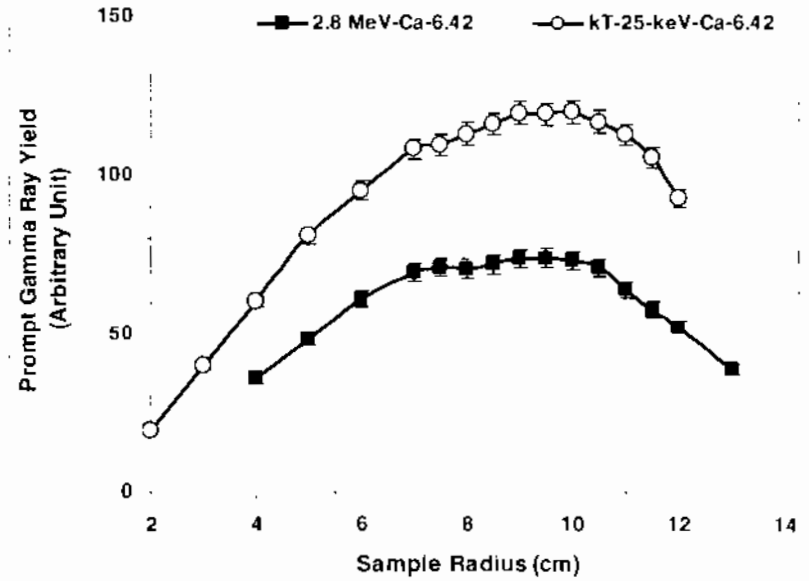

Fig. 3. Calculated yield of 6.42 MeV prompt gamma rays from calcium in a Portland cement sample for a ^{7}Li(p,n) reaction [2] and 2.8 MeV neutrons based PGNAA setup [3] plotted as a function of the front moderator thickness. The data points are connected by lines to show the trend.

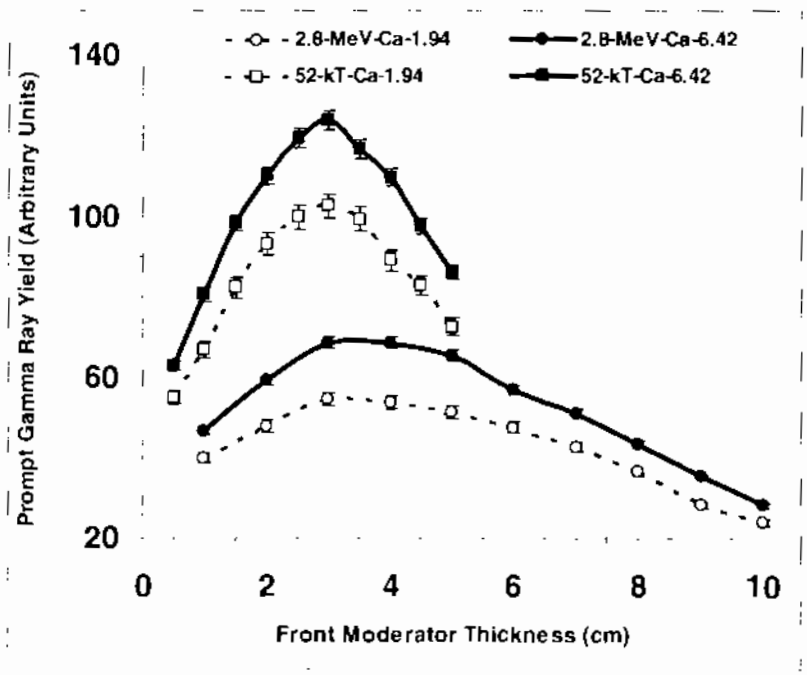

Fig. 4. Calculated yield of 1.94 MeV and 6.42 MeV prompt gamma rays from calcium for a ^{3}H(p,n) reaction [6] and 2.8 MeV neutrons based PGNAA setup [3] plotted as a function of the cement sample radius. The data points are connected by lines to show the trend.

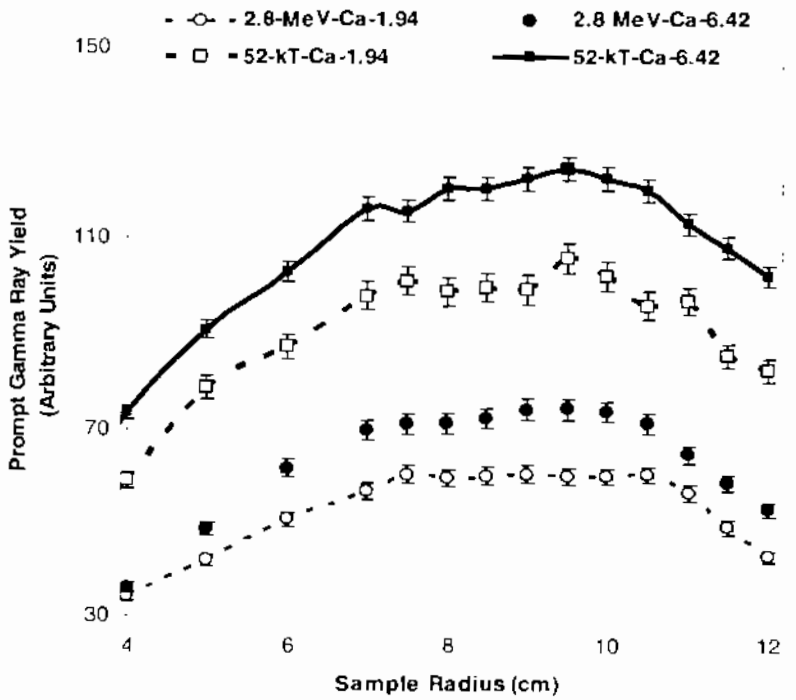

Fig. 5. Calculated yield of 1.94 MeV and 6.42 MeV prompt gamma rays from calcium for a ^{3}H(p,n) reaction [6] and 2.8 MeV neutrons based PGNAA setup [3] plotted as a function of the front moderator thickness. The data points are connected by lines to show the trend.

As shown in the Table 2 and Figures 6 and 7 [5] , the optimum radius of the sample for 14-MeV neutron-based PGNAA system is comparable with those of the other three PGNAA setups but the optimum values of its front moderator thickness and sample length are larger than those of ^{7}Li(p, n) reaction, ^{3}H(p, n) reaction and 2.8 MeV neutrons based

PGNAA setups. The Figures 6 and 7 also show the data for a ^{252}Cf source based PGNAA setup [5]. Due to different neutron energy used in each PGNAA system, it is expected that front moderator thickness will be different for each PGNAA system. The PGNAA system utilizing low energy neutrons will require thin front moderator while the thick front moderator will be required by the PGNAA system utilizing the higher energy neutrons. This is supported by the fact that the calculated average thickness of the front moderator of the ^{7}Li(p, n) / ^{3}H(p,n) reaction, 2.8-MeV neutron and 14-MeV neutron-based PGNAA system was found to be 3, 4 and 5 cm respectively.

Table 2: Optimum values of the geometry parameters of the ^{7}Li(p,n), ^{3}H(p,n), ^{3}H(d,n) reactions, and 2.8 MeV neutron based PGNAA setup.

Sample Parameters	**PGNAA setup based upon**			
	^{7}Li(p,n) reaction [2]	**^{3}H(p,n) reaction [6]**	**^{3}H(d,n) reaction [5]**	**2.8 MeV neutrons [3]**
Radius (cm)	7-10.5	7-10	7.5-9.0	7.0-10.5
Front Moderator Thickness (cm)	2.5-3	2.5-3	4-6	3-5
Length (cm)	9-12	9-13	12-18	11-13
Moderator Radius (cm)	12.75	12.75	12.75	12.75

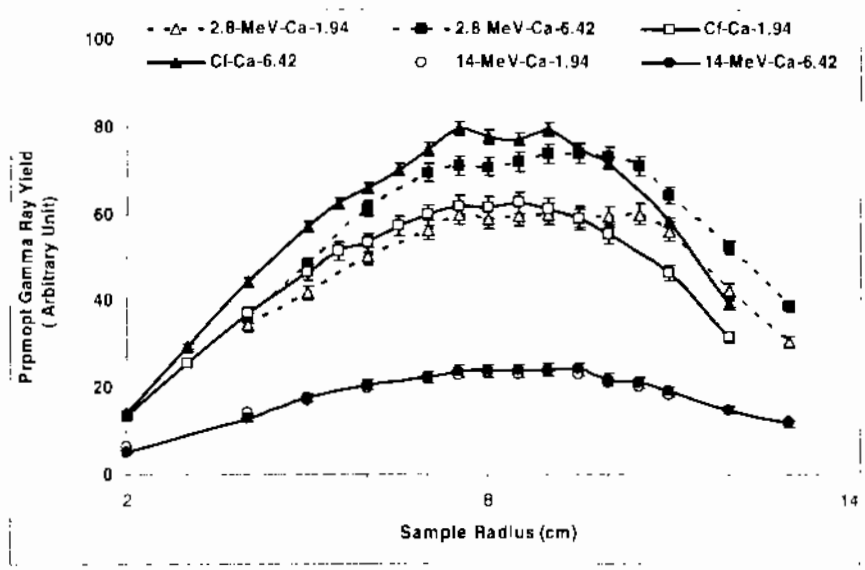

Fig. 6. Calculated yields of 1.94 MeV and 6.42 MeV prompt gamma-rays from calcium for a ^{252}Cf neutron [5], 14-MeV neutron [5] and 2.8-MeV neutron [3]-based PGNAA system plotted as a function of the cement sample radius. The data points are connected by lines to show the trend.

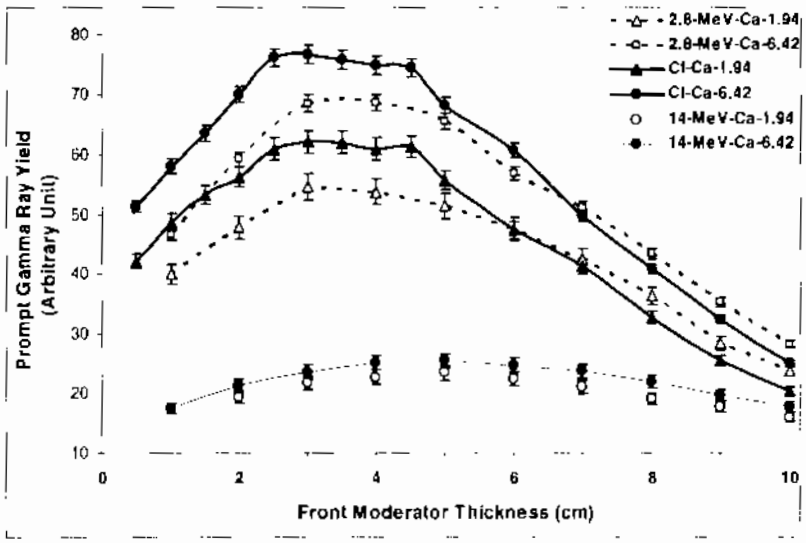

Fig. 7. Calculated yields of 1.94 MeV and 6.42 MeV prompt gamma-rays from calcium for a ^{252}Cf neutron [5], 14-MeV neutron [5] and 2.8-MeV neutron [3] -based PGNAA system plotted as a function of the front moderator thickness. The data points are connected by lines to show the trend.

Similarly optimum length of PGNAA sample was expected to depend upon the neutrons energy because neutrons are also moderated in the sample as well. The higher is the neutron energy, larger is the optimum length of the sample. The average length of sample of the ^{7}Li(p, n) / ^{3}H(p, n) reaction, 2.8-MeV neutrons and 14-MeV neutron-based system was calculated to be 11, 12 and 14 cm respectively.

4. Comparison of the Prompt gamma ray yield from ^{7}Li(p,n) reaction, ^{3}H(p, n) reaction, the 2.8 MeV neutrons and 14 MeV neutrons based PGNAA setup

The maximum yield of prompt gamma rays from Ca, Si, Fe and Al in a Portland cement sample for the ^{7}Li(p,n) reaction [2] , ^{3}H(p,n) reaction [6] , 2.8 MeV [3] and 14 MeV neutrons [5] based setup are listed in the Table 3. The yield was calculated for a sample with an average optimum value of sample radius and sample length as well as front moderator thickness given in the Table 2. The prompt γ-ray yield from the ^{7}Li(p,n) reaction based setup is comparable with that from ^{3}H(p,n) reaction based setup [2]. The maximum yield of prompt gamma rays for Ca, Si and Fe in a Portland Cement sample for the ^{3}H(p,n) reaction based setup is respectively 71-75 %, 45-59 % and 72 % higher than that of the 2.8 MeV neutrons based facility. Moreover the maximum yield of the prompt gamma rays from aluminum in a Portland Cement sample for the ^{3}H(p,n) reaction based setup is about 76 % higher than that from the 2.8 MeV neutrons based facility. This higher yield will result in a higher sensitivity for the ^{7}Li(p,n) reaction and ^{3}H(p,n) reaction based setup in comparison with the 2.8 MeV neutrons based facility. The performance of the 14-MeV neutron-based system is poorest among the four setups studied. The calculated yield of prompt gamma rays from the 14-MeV neutron-based system is almost one-third of that from the 2.8-MeV based system.

Table 3. Calculated yield (arbitrary unit) of the prompt γ-rays from Ca, Si, Fe and Al from ^{7}Li(p,n) reaction, ^{3}H(p,n), ^{3}H(d,n), and 2.8 MeV neutron based PGNAA setup.

Prompt γ-Rays Energy (MeV)	PGNAA setup based upon			
	^{7}Li(p,n) reaction [2]	^{3}H(p,n) reaction [6]	2.8 MeV neutrons [3]	^{3}H(d,n) reaction - [5]
Ca(1.94)	105.6±1	104.9±1	60±1	23.6±0.5
Ca (6.42)	126.4±1	124.3±1	74±1	24.1±0.5
Si (3.54)	15.5±0.5	14.9±0.5	9.4±0.5	3.7±0.2
Si (4.94)	16.4±0.5	14.8±0.5	10.2±0.5	4.4±0.2
Fe(7.63,7.65)	12.6±0.4	13.6±0.4	7.9±0.4	3±0.2
Al (7.72)	3.6±0.2	4.4±0.2	2.5±0.2	0.83±0.11

5. Conclusion

An inter-comparison of the performance of the ^{7}Li(p,n) reaction and ^{3}H(p,n) reaction based PGNNAA setup as well as 2.8 MeV and 14 MeV neutrons based PGNAA has revealed the best performance for the ^{7}Li(p,n) reaction and ^{3}H(p,n) reaction based setup. Mostly the geometry parameters of the ^{7}Li(p,n) reaction and ^{3}H(p,n) reaction based PGNNAA setup as well as 2.8 MeV and 14 MeV neutrons based PGNAA have comparable values except the remarkably smaller values of the front moderator thickness for the ^{7}Li(p,n) reaction and ^{3}H(p,n) reaction based PGNNAA setup. The poorest performance has been observed for the 14 MeV neutrons based PGNAA setup. The prompt gamma yield from the 14 MeV based PGNAA setup is one-third of the 2.8 MeV neutron based setup while the prompt gamma ray yield from the ^{7}Li(p,n) reaction and ^{3}H(p,n) reaction based PGNNAA setup is 70-75 % higher than that of 2.8 MeV neutron based PGNAA setup. This study has shown that by replacing the 2.8 MeV neutron source in the existing PGNAA setup at KFUPM with a Maxwellian neutron source of ^{7}Li(p,n) or ^{3}H(p,n) type, an increase by 70-75 % in prompt gamma ray yield can be achieved.

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مقارنة تطبيقات تحليل المواد بأشعة جاما الفورية الناتجة عن التنشيط الإشعاعي لهذه المواد بواسطة النيوترونات السريعة الصادرة عن معجلات و مصادر أخرى

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المستخلص. تستخدم تقنية تحليل المواد لمعرفة عناصرها بواسطة أشعة جاما الفورية الناتجة عن تنشيط هذه المواد بواسطة النيوترونات في الصناعة كعملية ضبط. فكفاءة نظام التحليل هذا تعتمد على طاقة النيوترونات. ولقد تم حديثا في جامعة الملـك فهـد للبترول والمعادن بالظهران في المملكة العربية السعودية تصميم واختبار نظام التحليل المذكور في فحص العينات الكبيرة. أيضا فإن هذا النظام قد فحص بنجاح عينات من الأسمنت والخرسانة. وعليه فإن هناك رغبة في تحسين أداء هذا النظام أكثر وذلـك باستبدال مصدر النيوترونات القائم على التفاعل D(d,n) والذي يعطي نيوترونات بطاقة MeV ٢,٨ بمصدر نيوترونات أكثر كفاءة. ولقد أجريت دراسات عديدة على مـستوى أداء تجهيزات نظام التحليل المذكور في جامعة الملك فهد للبترول والمعادن في حالـة كون مصدر النيوترونات هو النظائر المشعة وفي حالة كون المصدر هو المعجـلات. وفي حالة المعجلات، فقد أجريت الدراسة باعتبار التفاعلات $^{3}H(d,n)$ ،$^{3}H(p,n)$ ،$^{7}Li(p,n)$ لإنتاج النيوترونات السريعة حيث تتم مقارنة نتائجها مع بعضها في هذا البحث.