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Measurement of Natural Radioactivity in Cement of Tlemcen - Algeria by Using Well-Shape NaI(Tl) Detector

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Abstract.

The aim of this study was to characterize the concentrations of ^{40}K, ^{226}Ra and ^{232}Th in the cement samples manufactured in Tlemcen (Algeria) using a high-efficiency gamma-ray spectrometer. Measurements of radioactivity in the environment are of great importance to monitor and control the levels of radiation to which man is exposed directly or indirectly. The environment around people is radioactive due to ionising radiation from the sky, the Earth's crust, and various atoms existing in the water, food, building materials and metals. Indoor exposure arises from the soils on which the building stands and the building materials used during construction. This is because all building materials contain certain levels of natural radionuclides ^{238}U, ^{232}Th, and ^{40}K. The building materials like bricks, concrete, sand, cement etc. are earth based, containing uranium and thorium in varying amounts which are the biggest inescapable sources of natural ionizing radiation for human exposure. The objectives of this study were to determine the level of natural radioactivity and associated radiological hazard caused by natural radioactivity in local cement factory of Beni-Saf (province Tlemcen) in Algeria has been measured using NaI (Tl) 2x2 well detector (Canberra Inc.). These samples were analyzed using gamma-ray spectrometry technique. The spectra of samples were collected and measured using a multichannel analyzer (MCA) connected with measurement system for this purpose. Activity concentrations of ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K were found in the range of 1.0-4.2 Bq.kg^{-1}, 4.85-7.7 Bq.kg^{-1}and 115.3Bq.kg^{-1}, respectively. The average values of activity concentrations obtained for ^{226}Ra, ^{232}Th and ^{40}K in different samples of cement are lower than the corresponding global values reported in UNSCEAR publications. The results obtained in this study show no significant radiological hazard when Beni-Saf cement is used for construction of buildings.

Keywords: Activity Concentration, Cement Samples, Gamma-Ray Spectrometry Technique.

1- Introduction.

Human beings are continuously exposed to radiation of natural origin. The main sources of such exposure are the cosmic rays and the terrestrial natural radionuclide's (such as ^{238}U radionuclide) that are occurring at trace levels in the Earth's crust. It is estimated that the absorbed dose rate in air as a result of the exposure to cosmic radiation at the sea level is 30nGy/h (**1**). Building materials contain various amounts of natural radioactive nuclides. For example, materials derived from rock and soils contain mainly natural radionuclides of the uranium (U) and thorium (Th) series, and the radioactive isotope of potassium ^{40}K(**2**).Measurement of radioactivity in the building materials is very important to determine the environmental hazards on the health of humans and is very essential to set the standard radiation levels and national guidelines according to the international recommendations (**3**).The assessment of the population exposures due to indoor radiation is very important and therefore the knowledge of the concentrations of natural radionuclides in the construction materials is required. Construction materials are derived from both natural sources (e.g. rock and soil) and waste products (e.g. phosphogypsum, alum shale, coal fly ash, oil shale ash, some rare minerals, certain slugs etc.) and also from industry products e.g. power plants, phosphate fertilizer and oil industry (**4**).

It is necessary to measure the natural environmental radiation level provided by ground, air, water, foods, building interiors etc., for the estimation of the exposures to natural radiation sources. IAEA has published data for the doses accumulated by the human beings during their life activities. The exposures to cosmic radiation are about 0.38mSv/year, to terrestrial radiation 0.45mSv/year (this figure increases by nearly 20 % for brick and concrete buildings), to water, food and air 1.5mSv/year. The exposures to X-rays diagnostics are about 0.4mSv/year and to the other factors like color TV, air flights, nuclear power plants the exposure is about

0.1mSv/year. Thus, in total the human being receives about 2.7mSv/year from natural and man-made radiation sources. The dose limit for public exposure to man-made radiation sources is 1mSv/year (**5**). Cement is one of the important and expensive materials used by the building industry in Algeria. Most buildings in Algeria are constructed from cement blocks and cement concrete. Cement is used for making blocks, concrete, and for plastering the buildings made of bricks, blocks or concrete.

A large number of studies on the level of radioactivity in building materials have been completed in various parts of the world. In Algeria, a study on the radioactivity in building materials commonly used in the country showed that radium equivalent activities for all the building were all below the criterion limit of gamma-radiation dose 1.5mSv/yr. As is known, cement is the most important construction material of today civilization. The radioactivity content of cement varies considerably depending upon the geological characteristics of the initial raw materials from which the cement is processed. This paper outlines the methodology used for determining the type and specific activity of the naturally occurring radionuclides found in commonly used building materials (cement) in the Algeria.

2. Materials and Methods.

2.1. Sampling and sample preparation.

Many samples were collected from production plants of local cement (Beni-Saf- Tlemcen, Algeria) for analysis. The sample, each about 1 kg in weight, was dried in an oven at about 110^{o}C to ensure that moisture is completely removed. The samples were then sieved through a 2 mm mesh, which is the optimum size enriched in heavy minerals. The powdered samples were stored in tight plastic containers (fit to well volume of our detector) for more than one month to allow for radioactive equilibrium to be reached (secular equilibrium where the rate of decay of the daughters becomes equal to that of the parent). This step is necessary to ensure that Radon gas confined within the volume and the decay products also remain in the sample.

2.2. Radiometric measurement.

Radiometric measurements were performed for qualitative identification as well as quantitative determination of radionuclides present in cement. The gamma-spectrometric measurements were performed with NaI(Tl) well detector 2x2 inch with its electronic circuits Canberra Inc. (**6**). The emphasis was on the determination of specific activity concentration of ^{226}Ra, ^{232}Th and ^{40}K. Every sample of cement was measured for 48h and its spectrum was stored in a PC-based multichannel analyzer (MCA). The analysis of ^{40}K was based upon its single peak of 1460.8 keV, whereas the analysis of ^{226} Ra and ^{232}Th depended upon the peaks of the daughter products in equilibrium with their parent nuclides, the concentration of ^{226}Ra was determined from the average concentrations of ^{214}Pb (352keV) and ^{214}Bi (609, 1120 and 1765keV), and that of 232Th was determined from average concentrations of ^{212}Pb (239keV), ^{208}Tl (583,2615keV) and ^{228}Ac (338.3,911,969.11keV) in each sample under study (**Table 1**). In the uranium series the decay chain segment starting from radium (Ra) is radiological the most important and, therefore, reference is often made to Radium instead of Uranium.

2.3. Method of calculations.

The efficiencies for each radionuclide were calculated and used to estimate the activity concentration of each of the radionuclide in the samples. The detection efficiency of the system was determined using several calculations including linear attenuation coefficient, geometric and intrinsic efficiencies for well type 2x2 NaI(Tl) detector. The well shaped detectors are of higher efficiency for the same volume of detector, this particular characteristic allows almost a 100 percent efficiency (so called 4π geometry) for low gamma-emitting test sources that can fit the well shape (**7,8,9**).

2.3.1. Calculating of detector counting efficiency DE.

There are three factors, G, I and M, that affect the efficient absorption of the photons emitted by the source. Their product is the detector counting efficiency DE.

$$\mathbf{DE = G \times I \times M} \qquad \mathbf{(1)}$$

G = fraction of all space that the detector subtends. Unless the detector completely surrounds the source, the geometrical solid angle factor is less than 1.

I = fraction of the photons transmitted by the intervening materials that reach the detector surface. There are losses due to absorption by material in the path of the photon. Air, detector housing materials and light reflectors around the detector are possible absorbers.

M = fraction of the photons absorbed by the detector. The detector material is not always sufficiently thick to stop the radiation.

Table1. Linear attenuation coefficients of gamma ray in Al. and NaI

U-238 (A)	Gamma Energy KeV	Probab. Of γ Emission%	μ_1(Na) Cm^2/g $W_1=0.153$	μ_2 (I) Cm^2/g $W_2=8.46$	$\sum \mu_i . w_i$ μ_m.cal. (NaI)	μ_m.(NaI) Ref.aver.	μ_m.(NaI) average	μ_l(NaI) $=\mu_m.\rho$ $\rho = 3.67$ g/cm^3	μ_l. (NaI)for low enegy.ref.	μ_l(Al) 1/cm Ref.	μ_l(Al) 1/cm average
226Ra	186.10	03.51	0.123	0.500	0.4188	0.425	0.420	1.541	1.450	0.343	0.343
	241.98	**07.12**	0.110	0.250	0.2173	0.278	0.245	0.899	0.860	0.310	0.310
	295.21	18.15	0.102	0.163	0.1486	0.153	0.151	0.554	0.629	0.290	0.290
	351.92	03.51	0.095	0.130	0.1151	0.130	0.122	0.448	0.480	0.260	0.260
	609.31	44.10	0.077	0.079	0.0762	0.078	0.077	0.283		0.205	0.205
	768.63	04.76	0.069	0.070	0.0675	0.068	0.068	0.249		0.188	
238U	49.50		0.244	11.20	9.1874	10.40	9.89	36.30		0.960	
214pb	295.10	19.24	0.102	0.160	0.1456	0.153	0.149	0.547	0.629	0.290	0.280
	325.00	37.20	0.099	0.145	0.1331	0.142	0.138	0.506	0.530	0.280	0.270
	351.93	35.34									
214Bi	609.30	46.36	0.077	0.079	0.0762	0.078	0.077	0.283		0.205	0.205
214Bi	1764.5	15.80	0.046	0.043	0.0420	0.043	0.043	0.158		0.126	0.126
214Bi	1120.3	15.1 0	0.057	0.053	0.0520	0.051	0.051	0.187		0.156	0.156
234Th	63.280	04.47	0.200	07.32	6.016	6.100	6.05	22.20	20.94	0.710	0.710
	92.370	02.60	0.155	2.600	2.1477	2.220	2.18	8.000	7.400	0.490	0.450
235U	185.70	57.25	0.125	0.510	0.4360	0.425	0.43	1.578	1.460	0.345	0.345
	143.70	10.96	0.128	0.600	0.5096	0.516	0.51	1.872	2.400	0.385	0.385
(B)	**232Th**										
228Ac	338.30	11.40	0.097	0.151	0.1379	0.140	0.139	0.510	0.500	0.274	0.265
	911.20	27.70	0.065	0.060	0.0589	0.062	0.060	0.220		0.174	0.174
	969.80	05.20	0.062	0.058	0.0565	0.059	0.058	0.213		0.171	0.171
212Bi	727.00	11.80	0.075	0.072	0.0709	0.071		0.257		0.197	0.193
212Pb	115.18	0.62	0.146	1.800	1.4913	1.500	1.495	5.487	4.300	0.430	0.430
	300.09	03.40	0.101	0.162	0.1475	0.153	0.150	0.550	0.600	0.285	0.275
	238.60	43.60	0.117	0.150	0.2200	0.190	0.205	0.752	0.866	0.293	0.300
208Tl	583.20	84.50	0.079	0.080	0.0731	0.081	0.077	0.283		0.215	0.215
	2615.0	99.79	0.038	0.039	0.0378	0.038	0.038	0.140		0.054	0.054
228Ra	338.32	11.26	0.097	0.151	0.1372	0.140	0.139	0.510	0.500	0.274	0.274
	911.07	26.60	0.065	0.060	0.0589	0.062	0.060	0.220		0.175	0.175
	969.11	16.23	0.062	0.058	0.0565	0.059	0.058	0.213		0.172	0.172
(c)											
60Co	1173.0	100	0.057	0.055	0.0527	0.054	0.0530	0.195		0.156	0.156
60Co	1332.0	100	0.052	0.050	0.0488	0.050	0.0495	0.182		0.145	0.145
134Cs	604.70	97.10	0.077	0.079	0.0763	0.079	0.078	0.286		0.210	0.210
	795.50	85.40	0.067	0.065	0.0649	0.065	0.065	0.239		0.183	0.183
137Cs	661.60	85.00	0.070	0.075	0.0720	0.075	0.0735	0.270		0.196	0.196
(D) k-40	1460.8	10.66	0.050	0.045	0.0384	0.042	0.040	0.147		0.137	0.137

In our well detector, the sample placed in the hole of the detector, we have a specific conception for dealing with this fractions. The dimensions of 2x2 NaI(Tl) detector are 2-inch diameter with 2 inches high (crystal) and a 0.75 inch diameter by 1.44 inch deep well (hole), for these properties of well-detector the previous fraction is seen as follows:

To calculate the fraction of space not subtended and then to subtract that value from 1 to get the fraction G subtended. The fraction not subtended is the area of the hole of 0.75 inch diameter at the end of the well at distance of 1.44 inches. The (absolute) total efficiencies for a right cylinder and a well-type are presented as functions of the source position and the photon energy. When the sources are located on the surface, the total efficiencies for low energy photons are 0.5 and ~ 1, respectively. This means every photon incident on the detector produces an output pulse considering the solid angles of both geometries (2p for right cylinder, and nearly 4p for well type), regardless of the energy deposited (**9**).

$$\mathbf{1 - G = (\pi r^2) / (4\pi R^2)} \tag{2}$$

Where: πr^2 = area of hole in detector face, and $4\pi R^2$ = area of sphere with a radius equal to the distance from the source to the hole.

1 - G = (π x 0.375 inch x 0.375 inch) / (4 x π x 1.44 inch x 1.44 inch) = 0.017, and G = 0.983

This detector subtends or intercepts 98% of all space (A great advantage of the well geometry is, of course, the large solid angle (nearly 4π **sr**), which leads to a high efficiency.

To calculate **I**, we have

$$\mathbf{I = exp^{-(\mu l \times d)}} \tag{3}$$

Where: μ_l, the linear attenuation coefficient for gamma ray in aluminum.

d = 0.025 cm (0.010 inch), the thickness of the aluminum container.

The fraction of the photons absorbed by the detector M is calculated by subtracting the fraction that pass through the detector from 1:

$$\mathbf{M = 1 - exp^{-(\mu l \times d)}} \tag{4}$$

μ_l = the linear attenuation coefficient for gamma ray in NaI(crytal).

d = 1.422 cm (0.56 inch), the minimum distance traveled in NaI(Tl) at the bottom of the well,

2.3.2. Linear attenuation coefficient calculations

For calculation of detecting efficiencies we try to find the values of μ_l for each aluminum and NaI(crytal). Firstly we investigate the references in this item and do a comparison between them to take the its main values, then we calculate μ_l for mixture NaI using the following formulas:

$$\mu_m(\mathrm{NaI}) = \sum \mu_i . w_i = (\mu_1 . w_1)_{Na} + (\mu_2 . w_2)_I \tag{5}$$

$$\mu_l(\mathrm{NaI}) = \mu_m(\mathrm{NaI}) . \rho \tag{6}$$

$$A = N / (T . I_\gamma . \varepsilon . W) \tag{7}$$

Where N is net peak counts (background subtracted), T is the measured time (sec.), ε is the efficiency of detector, I_γ is the branching ratio of gamma emission for decay mode and W is the sample weight.

where **ρ** is the density of NaI =3.7g/cm^3, the calculation result of μ_l (**NaI**) in **Table 1** are compared with the values from references (**9,10**), to view the fit value with the graph of linear attenuation coefficient, this leads us to chose proper solution to this calculations. Finally we calculate the **DE** of our detector for each gamma energy in the cement samples under study (**Table 2**). Using the above work, the activity concentrations for the ^{40}K, ^{232}Th, ^{238}U and ^{226}Ra radionuclides were calculated using the detected photopeaks in the spectra.

Table 2.Efficiencies of well-shaped 2x2 NaI(Tl) detector.

Radio nuclides	Decay series	Photopeak Energy	**I**	**M**	**G**	**DE**
Ra^{226}	Bi^{214}	609.3	0.975309912	0.336304179	0.983	0.32236311
		1120.3	0.996107595	0.246494955	0.983	0.24113213
		1764.5	0.996854956	0.185569411	0.983	0.18182091
	Pb^{214}	295.2	0.992776217	0.542600005	0.983	0.529571059
		351.9	0.993024400	0.47247744	0.983	0.461100321
Th^{232}	Pb^{212}	238.6	0.992701762	0.656766395	0.983	0.640889613
	Ac^{228}	338.3	0.993173407	0.51378197	0.983	0.5015525
		911.6	0.995649447	0.268633078	0.983	0.262920121
		969.1	0995734124	0.261316696	0.983	0.25577852
	Tl^{208}	583	0.994639419	0.343270417	0.983	0.33512621
		2614	0.99865091	0.180515668	0.983	0.174875589
K^{40}		1460.8	0.996854956	0.223101565	0.983	0.20225655
Cs^{137}		661.7	0.995111985	0.318827703	0.983	0.31187569
Co^{60}		1173.2	0.996107595	0.242165306	0.983	0.237121914
		1332.5	0.996381562	0.228025684	0.983	0.22333897
U^{235}		143.8	0.990421172	0.930190419	0.983	0.905618519
		185.7	0.991412088	0.893957571	0.983	0.871213117

3. Results and Discussion.

Cement is an important construction material for houses and buildings in urban areas of Algeria. It is used for blocks and concrete manufacturing as well as for plastering the buildings walls, which made of bricks. However, detailed information of the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in cement and other building materials used in Algeria are not readily available in the literature with the exception of the work by Amrani and Tahtat (**11**). This study is a continuation of our ongoing project related to the measurement of specific activity of ^{238}U (^{226}Ra), ^{232}Th and ^{40}K in environmental samples from the vicinity of the city of Tlemcen (Algeria) using gamma-ray spectrometric technique. Using the above formula of specific activity with values of efficiency for well-shaped 2x2 NaI(Tl) detector for each gamma ray emitted by radionuclide under the study, the activity concentrations due to ^{226}Ra,^{232}Th and ^{40}K have been determined as present in **Table 3**.

As can be seen (**Table 3**), the average activity concentrations of the three radionuclides (Ra, Th,and K) are 1.0-4.2 Bq.kg^{-1}, 4.85-7.7 Bq.kg^{-1}and 115.3Bq.kg^{-1} for ^{226}Ra**,** ^{232}Th and ^{40}K respectively. It is important to point out that these values are not representative values for the countries mentioned but are specific to the regions from where the samples were collected. Radium, thorium and potassium are not uniformly distributed in soil or rocks, from which building materials are derived, but the radioactivity varies, often greatly, over a distance of some meters. The measured values of radium and thorium contents show only the average radioactivity in building materials (cement) used in province of Tlemcen. The mean values are lower than the corresponding world-wide average values which are 35, 30 and 400 Bq.kg^{-1} for ^{226}Ra**,** ^{232}Th and ^{40} K respectively(**1**). The specific activity of ^{40}K,^{226}Ra and ^{232}Th determined in the present study for cement have also been compared with values reported for other countries (**Table 3**).

Table3. Comparison between the activity concentrations(in Bq/Kg) of our building materials (cement) with that of other countries of the world.

country	226Ra	232Th	40K	$(226Ra)_{eq}$	References
Algeria	41	27	422.0	112	(11)
Albania	55 40-61	17 11-23	179 107-251		(13)
Australia	51.5	48.10	114.7	129.4	(4)
Australia	7-180	7-240	24-850		(5)
Austria	26.7	14.2	210.0	63.1	(23,38)
Bangladesh	29.7	54.3	523.0	148	(14)
Bangladesh	62.3	59.4	329.0	172.8	(33)
Bangladesh	120.2	132.4	505.0		(27,38)
Belgium	62	76	------		(38)
Brazil	61.7	58.5	564	188.8	(15)
China	56.5	36.5	173.2	122	(23)
China	52.0	103	310		(16)
Cameron	27	15	277	70	(33)
coroatia	129-131	23.1-25.4	252.4-307.3	182.6-187.4	(17)
Cuba	9-71	02-38	47-2511	73	(18)
Egypt	78	33.3	37	151	(23)
Egypt	31.3	11.1	48.6	50.9	(19)
Egypt	36.6	43.2	82	103	(33)
Egypt	3.84-8.4	1.63-5.13	4.53-89.17		(20)
Finland	44	26	241		(21)
Finland	40.2	19.9	251		(23)
Germany	52	52	147		(27)
Germany	15	22.9	325		(22)
Ghana	35.94	25.44	233	90.12	(23)
Greece	62.8	23.8	284.1	117	(23)
Hungary	0.6-228	0.6-199	7-709		(24)
Hongkong	19.2	18.9	127		(33)
India	37	24.10	432.2	104.7	(23,33)
India	54	66	490	186.20	(25)
India	59.2	14.8	107.4		(27)
India	28-54	23-41	149-233	76-122	(26)
Iraq	60	32	346		(27)
Iraq	217-3593	31-387	157-3145		(28)
Iran	39.6	28.9	290	103.32	(29)
Iran	31.1	12.4	121	58.1	(30)
Ireland	60	12	150		(31)
Italy	46	42	316		(38)
Italy	38	22	218	92	(33)
Japan	35.80	20.7	139.4	77	(23)
Jordan	43.21	11.23	265.12	79.9	(32)
K.S.A	38.4	45.3	86	108	(33)
Lebanon	73.2	09	79.7	92.2	(34)
Malaysia	51	23	832	188	(33)
Malaysia	81.4	59.2	203.5		(38)
Malaysia	7.8-83	9.8-49.4	82-337	35-135	(35)
Mexico	26	52.6	00		(36)
Netherland	27	19	230		(43)
Norway	30	18	241		(21)
Norway	29.6	18.5	259		(37)

Pakistan	26.1	28.7	272.9	87.9	(38)
Poland	48.1	48.1	185		(27)
Slovak	5.8-21.6	1.6-38.23	52-733		(39)
Spain	74	31	241		(27)
Sweden	55	47	241		(21)
Tunisia	21.50	10.10	175.5	49.7	(23)
Turkey	40	28	248.3	99.1	(23)
Turkey	24.7	20.7	2493.1		(4)
U.K	22	18	155		(21)
Vietnam	39.86	25.46	243.50		(40)
Yemen	35.79	18.41	147		(41)
Zambia	23	32	134	79	(42)
This study	1.0-4.2	4.85-7.7	115.3		

4. Conclusion.

Gamma spectroscopy method was used for assessment of the U-238 and Th-232 series and K-40 concentrations in cement samples manufactured in Beni-Saf (Algeria, province of Tlemcen), then they were compared with published results from other countries. The average activity concentrations of the calculated average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the cement samples were1.0-4.2 Bq.kg^{-1}, 4.85-7.7 Bq.kg^{-1}and 115.3Bq.kg^{-1}, respectively. The results obtained in this study compares well with data from Egypt, Germany, Hungary, Australia, Cameron, Malaysia, Slovakia, Turkey and the U.K while they showed some variations with values from other countries (**Table 3**). These indicated considerable variations in the activity concentration are due to the varying amounts of uranium, thorium and K contents as a result of different geological formations under the earth crust from where the raw material for particular brands of cement were obtained.

From this research, we have achieved the following:

1- A new performance table for linear attenuation coefficients of gamma ray in Al and NaI (for efficiency calculation) (**Table 1**)

2- Calculation the efficiencies of well-shaped 2x2 NaI(Tl) detector. The values of DE are shown in **Table 2** and **Figure 1**, these values are close to those reported in **Figure 2** (exactly between line 1.5 and 2.5), and so our work completed the previously reported data (new line which can be added to this figure).

3- Any study in this field interest to compare own results with that for all over the world, thus we sit new **Table 3** of natural radioactivity measurement in cement.

4- This paper used gamma spectrometry to assessment the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K.

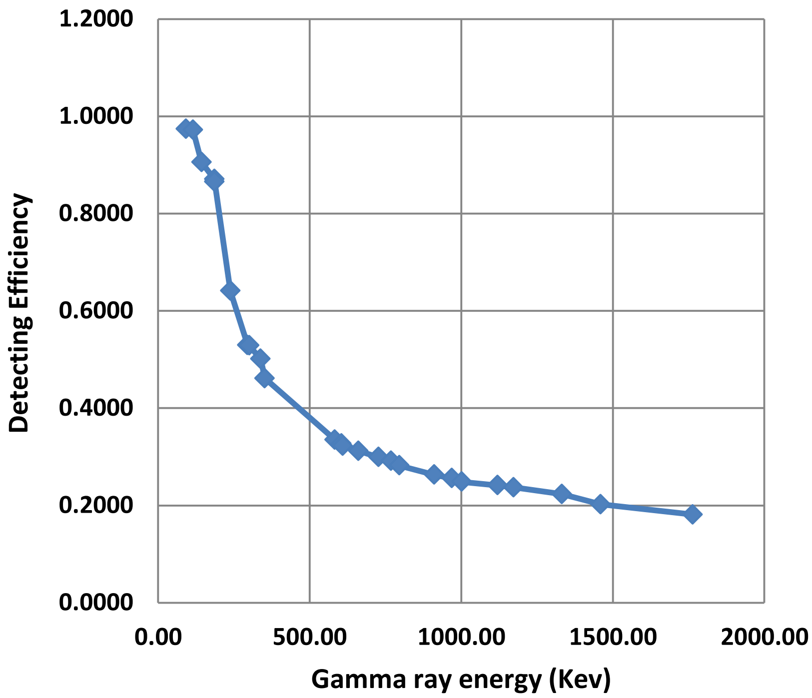

Figure 1.Efficiency of 2x2 NaI(Tl) detector.

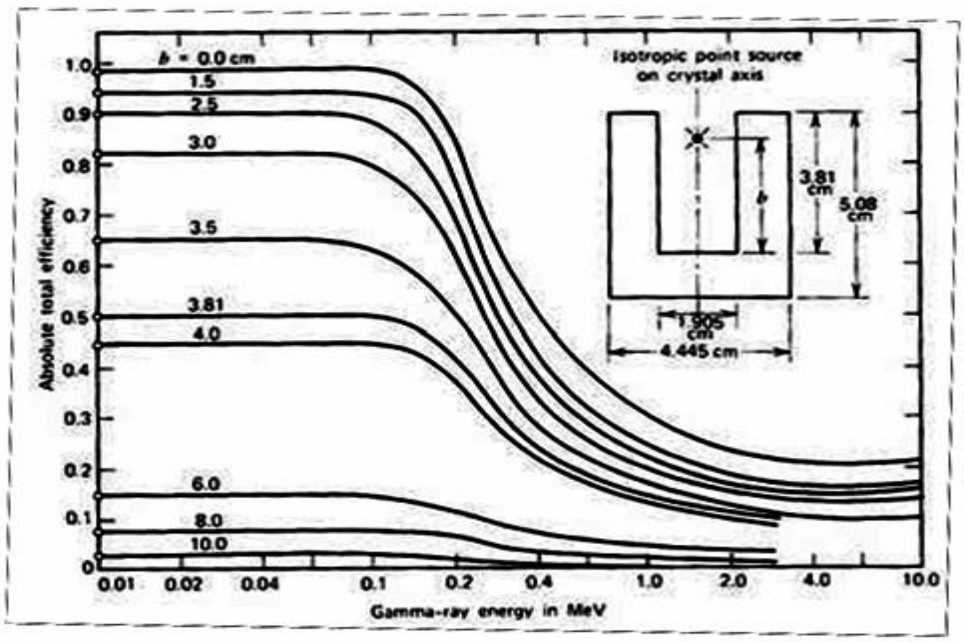

Figure 2. Absolute total efficiency for a well-type NaI(Tl).(Ref.12)

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