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The Characterization of Radioactive Waste Drums Using Nondestructive Scanning Gamma Ray System

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Abstract
A non-destructive assay (NDA) for radioactive waste drum has been studied using a local manufacturing gamma scanning system. The gamma system has been designed and implemented using scanning system contains a high efficiency portable HPGe detector for characterization and surveying the radioactive waste drums at Al-Tuwaitha site- Baghdad. To achieve identification with non-homogenous radioactive waste drum, six parallel plastic pipes (2cm in diameter) were inserted inside the cement type Portland contain radioactive sources and located at different distances from the outer diameter of the drum. The efficiency calibration is measured by conventional technique, using five miscellaneous radio nuclides with drum. The efficiency trend effected strongly with distance between detector and hot spot (location of radionuclide inside drum) due to attenuation factor and geometry effects. The NDA procedure becomes more acceptable in radioprotection demands dealing with the management and inspection of radio-waste drums.

Keywords: NDA waste drums, scanning gamma, efficiency.

وصف حاويات النفايات المشعة بأستخدام منظومة مسح اشعة كاماغيرالإتلافية

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الخلاصه
تم دراسة التحليل غيرالإتلافي لحاوية نفايات مشعة بأستخدام منظومة مسح اشعة كاما المصنوعة محليا. حيث تم تصميم وانجاز منظومة المسح التي تحتوي على عداد جرمانيوم محمول عالي النقاوة وعالي الكفاءة لوصف واستقصاء حاويات النفايات المشعة في موقع التويثة-بغداد. لغرض تحقيق تطابق مع حاوية نفايات مشعةغير متجانسة، تم استعمال ستة انابيب متوازية من مادة البلاستك ذات قطر 2 سم داخل السمنت نوع بورتلاند وتحتوي على عناصر مشعة ومتمركزة عند مسافات مختلفة من القطر الخارجي للحاوية. لقدقيست معايرة الكفاءة بوساطة التقنية المألوفة بأستخدام خمسة انوية مشعة ذات الخصائص المختلفة مع الحاوية. لقد وجد ان ميل الكفاءة يتأثر بشدة مع المسافة بين العداد والمصدر الموجود داخل الحاوية بسبب تأثيرمعامل التوهين والشكل الهندسي. من طريقة استخدام التحليل غيرالإتلافي تصبح الاجراءات المتبعة اكثر قبولا لطلبات الحماية الاشعاعية مع التعامل والكشف عن الحاويات المشعة.

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Introduction

The radioactive waste (RW), arising from various sources, where the most of waste come from civilian nuclear activities, nuclear fuel cycle, nuclear weapon activities, naturally occurring radioactive materials (NORM) as well as from other sources include medical and industrial wastes. In Iraq the problem is aggravated because of many destroyed nuclear facilities, which is a legacy of old nuclear activities in different area. It is experienced two devastating wars in 1991 and 2003, where old nuclear equipment and materials are scattered randomly beyond an official storage areas and at different governments. In spite of difficulties faced the specialized expert teams at Iraqi Energy Commission this nuclear waste should have meet standards and admissible criteria defined by national regulatory and management authorities for the intermediate and final storage [1]. Therefore, RWs are packaged in drums and being placed and located in storage sites such as Al-Tuwaitha site. Also, they must be managed with special care, from generation to final disposal and protecting the environment to be safe to the present and future generations [2].

The characterization of the RW is an important compulsion in waste management and safeguards. The nondestructive gamma scanning system was used for characterization the RW drums. For this purpose and in order to evaluate RW drums, an energy calibration for the gamma scanning system should be employed with common radioactive sources and then the qualitative assay for radio nuclides determination is not difficult task to be obtain.

The quantitative analysis to determine the activity of each radionuclide is more difficult due to the efficiency calibration of this system. In principle, also, it should be carried out with calibration standard, which requires physical and chemical similarity with unknown sample. In fact, the calibration standard is not needed if the detector efficiency is accurately known as a function of source position, the counting geometry and the sample size and shape are accurately known, exact energy peak and the counting rate of emitted gamma radiation is determined for the isotope(s) being analyzed [3, 4].

The non-homogeneous distribution of radioactive waste inside drums poses additional problems, as there are no references sources with the required properties for direct calibration of the detection system [5].

After different waste management predisposal steps (e.g. pretreatment, treatment and conditioning), an opening drums and containers for Characterization are not practical from either a radiological safety or cost requirements, because it is more labor and time, in addition, wastes may be generated in location or the hot-cell (if exist), where the assays take place[6, 7]. For all above reasons, non-destructive assays (NDA) by gamma spectroscopy without opining drums are applied preferably, because a representative sampling and a time consuming preparation of appropriate specimen for different analytical techniques is avoided. That would regard characterization is a part of the larger waste management strategy. Therefore, the NDA of gamma spectrometry is widely used for the characterization of drums containing radioactive waste [8, 9].The technique was performed by measurement of the drum as a whole or segments, a methodology utilizing efficiencies calculated with reference sources consolidated with theoretical method was done for getting the efficiency curve for varying sources measured with a NaI(Tl) detector , utilizing Monte Carlo code MCNP the efficiency simulation of an HPGe coaxial detector was fulfilled in other researches[10-12].

In the present work, the radiological characterization of the radioactive waste drums was performed by gamma scanning geometry technique, by determination the efficiency calibration of gamma spectroscopy system, which leads to determine the quality and quantity activity (specific activity) for each isotope in radioactive waste drums. The gamma spectroscopy system consists of portable high purity germanium (HPGe) gamma detector, equipped with scanning system that designed, manufactured and employed for this purpose, and has the capability to estimate the quality and quantity radioactivity levels in radioactive waste drums, where the drum is moved vertically and horizontally during the scanning**.**

Five radioisotopes sources ^{241}Am, ^{60}Co, ^{137}Cs, ^{192}Ir and ^{22}Na, were prepared to serve the conditions of experiment.

Experimental

The calibration storage and collecting drums are considered in the present work have 85 cm height, upper and lower internal diameter2R=57 cm. These drums were made of iron with 0.7 mm wall thickness. Iron drum, volume of 220 liter, was used for efficiency calibration, which is similar to that used for collecting radioactive waste. It is filled partially with Karasta Portland cement (ρ= 1.57g/cm^3) and six parallel plastic pipes are inserted at different distances from the outer diameter of the drum. These pipes were made 6 hollow of 2 cm diameter. As shown in Figure-1 the hollow cylindrical gaps were made in the calibration drum at different radial distances R_i (i= 1to 6)from the drum centerline as follows:H6 (center), H5 (7 cm), H4 (12 cm), H3 (16 cm), H2 (21 cm), H1 (24.5cm) from center, respectively. After removed the plastic pipes, the hollow cement tubes are used to contain the radioactive sources into the drum to perform the efficiency calibration.

Low radiation background shielded HPGe detector with LN2 cooling was used to measure the activity of the radioactive sources. A feature of this detection system was studied by using ^{152}Eu reference source. As shown in Figure-2 the calibration of energy and resolution sketch were measured experimentally for the present counting system. In order to minimize the effect of non-homogeneous matrix and sources distribution in typical measurements of waste drums, the drum was rotated and translated (a double translation) in front of an HPGe collimated detector, while the measurement was performed.

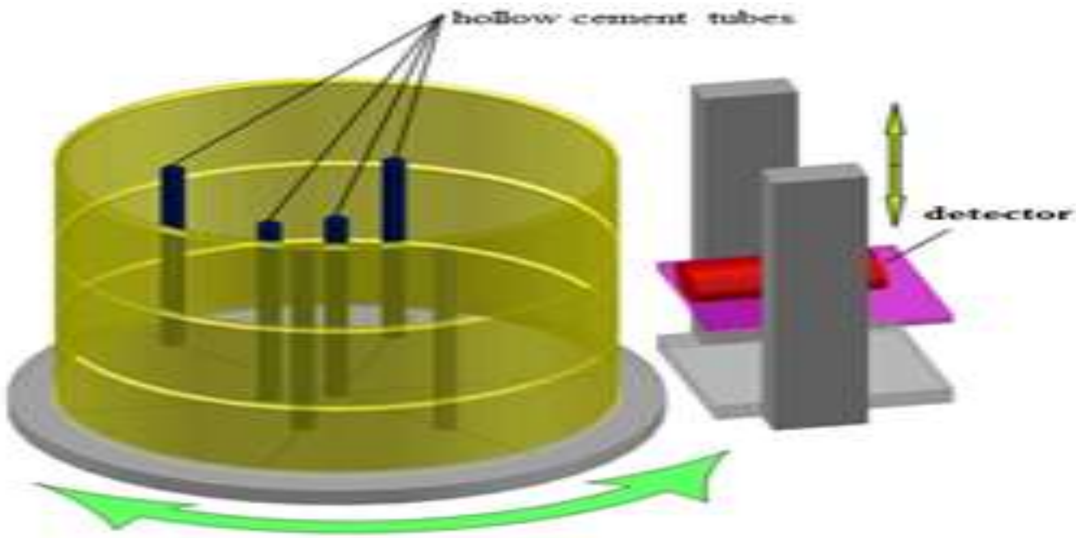

Figure 1-The transparency view of drum with detector.

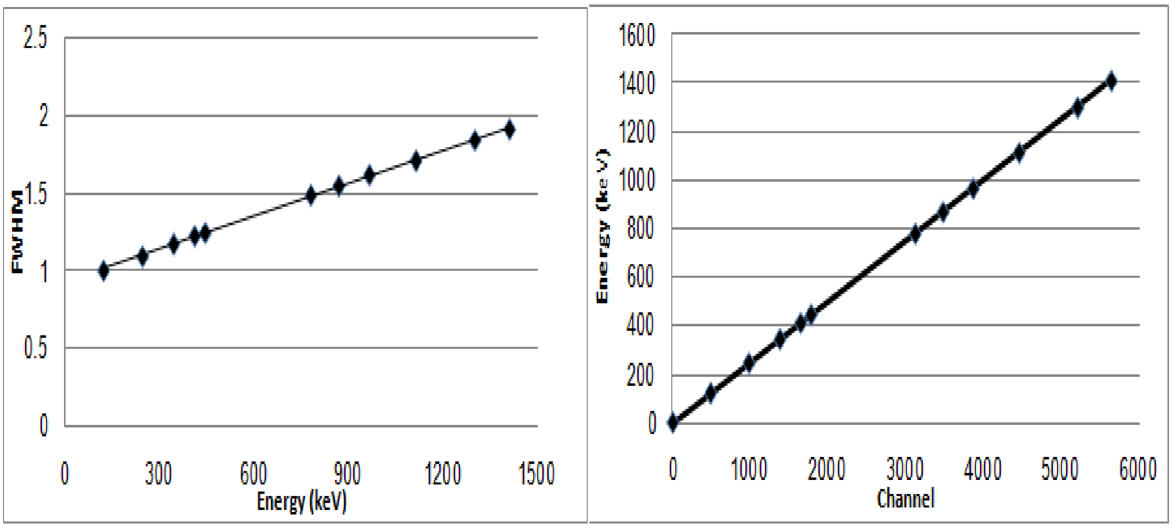

Figure 2- The energy calibration and FWHM for the shielded HPGe detector.

In general the efficiency value of a specific detector depends upon several factors: the geometry arrangement of detector, radioactive source, the incident photon energy, the content of detector material around the crystal and the parameter of the detector crystal. So the absolute full energy peak efficiency can be calculated by [13]:

$$\varepsilon(E) = \frac{A(I)}{T\,\Lambda\,Y(E)} \quad \text{….(1)}$$

where ε(E) is the absolute full energy peak efficiency of a gamma ray of energy, A(I) is the net counts gamma ray under the photo peak, Y(E) is the emission probability of radionuclide for identified photo peak energy E, Λ is the activity of the radio nuclide and T is the collection life time in seconds.

The collected count rate with respect to radial distance for twelve photo peaks and for six holes can be represented by [14]:

$$A_T = \sum_{i=1}^{6} \frac{A_i\, S_i\, L}{S\, L} \quad \text{…(2)}$$

where Ai: Net area under specific photo peak of gamma energy (counts/sec).
i: 1,2…..,6 number of holes.
S: drum base area, $S = \pi R^2$; R = 28.5 cm, $S_i = \pi(R^2 - R_i{}^2)$.
L: height of drum, V = $S \times L$, Vi = $S_i \times L$, V: volume of drum.
Vi: longitudinal volume section of drum and Si is the base area for specific longitudinal volume section of drum.

Five miscellaneous radioactive nuclides ^{241}Am (370000Bq), ^{60}Co (7400Bq), ^{137}Cs (70300Bq), ^{192}Ir (22200Bq) and ^{22}Na(5550Bq) were used and these nuclides covered the principal required range of gamma energies spectrum within the twelve peaks as: 59(0.36), 1173(1), 1332(1), 662(0.85), 295(0.287), 308(0.298), 316(0.83), 468(0.477), 588(0.0457), 604(0.82) and 1274(0.99) keV respectively.

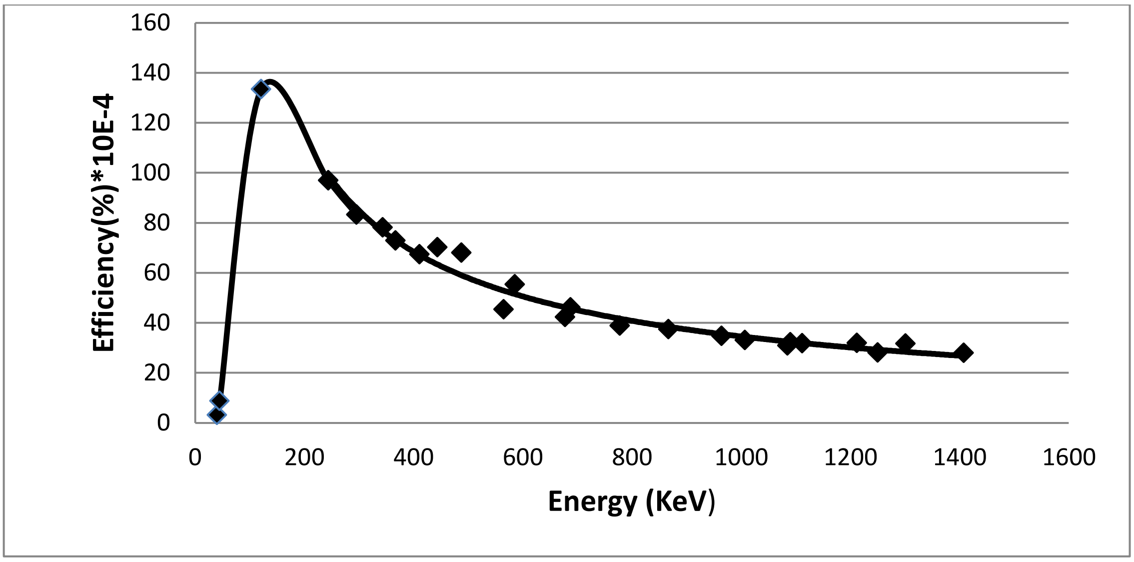

Figure 3 -The measured efficiency as a function of gamma-ray energy for the shielded HPGe detector with specific geometry used in the present work.

Some conditions must be exist and verified in the features of radioactive sources diameter, activity and energy, which makes the present work safer in selecting an appropriate radioactive sources. In the present work, employed radioactive sources represent the main component of the radioactive waste and the activity, dose and exposure rates are measured.

The detector of scanning system

A portable and remote control with windows tablet personal computer wired or wireless external control (as little as apparatus and cables in contaminated area)Falcon 5000 High Purity Germanium spectrometer system has been used for experimental measurements of scanning gamma. The detector model is a Canberra with Falcon software interfaced with Canberra genie 2000 software.
The main performance specifications of the detector are as follows:

Relative efficiency of 1.33 MeV ^{60}Co source is 18.7%,FWHM resolution of 1.78 keV, FWHM resolution of 3.29 keV both at 1.33 MeV ^{60}Co, and the peak-to-Compton ratio for ^{60}Co is 44.7:1. The crystal diameter is 61mm and the length is 25.5 mm. The core hole diameter is 8 mm, and the core hole length is 15.5 mm. The end cap to crystal distance is 5 mm. The cryostat window material is aluminum with 1.5 mm thickness. within it electrical cooler Type – Pulse Tube Cooler, time to cool – 3 to 4 hours at 25 °C (77 °F).

The drum is placed in to an electrical rotating platform, which can be rotate in two opposite directions, In order to minimize the effect of non-homogeneities (matrix density and radioactive source distribution). The total and the sectional surface scanning that can be got to obtain a complete survey of the drum. The system is provided with an electromechanical system, which assures the simultaneous rotation and horizontal translation of the drum, where the vertical up and down translation of detector can be achieved. As shown in Figure-4 the present scanning system with supplied control system consisting sensors and times switches.

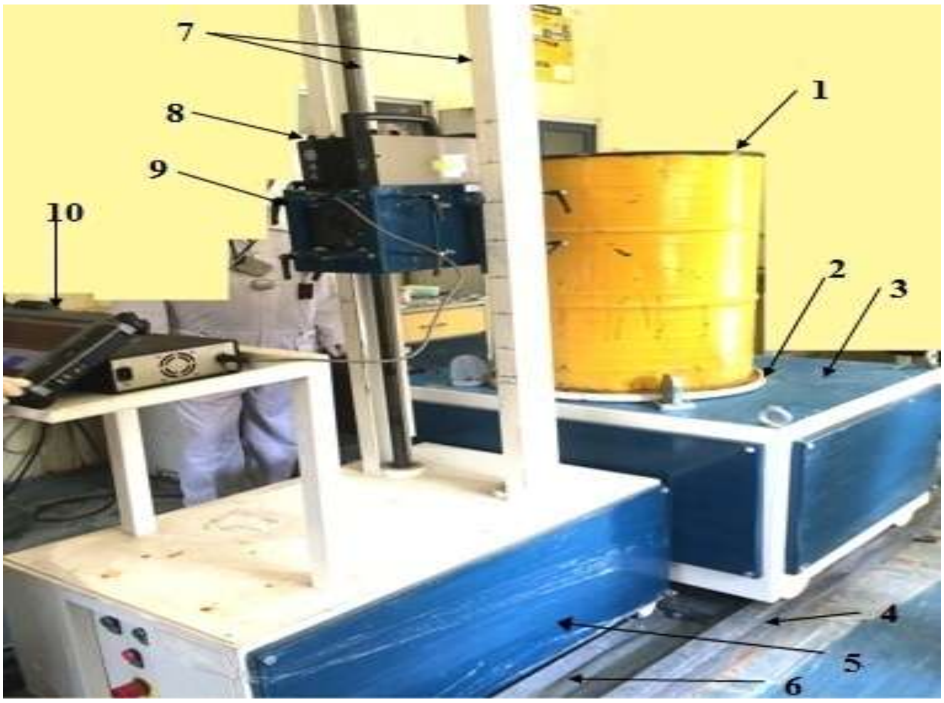

Figure 4-illustrate main parts of local manufacturing scanning system.

(1)Radioactive waste drum (container), (2)Rotating disk (platform) changeable speed and direction, (3)Motors cabinet and electrical control of rotating platform that carries the waste drum (can rotate horizontally in two opposite ways with ability to change speed of rotation per time, (4)Iron slide rail for motors cabinet mentioned in 3, (5) Motors cabinet and electrical control of rotating screws supplied with control system (sensors and timers),(6) Iron slide rail motors cabinet mentioned in 5, (7) Vertical rotating screws to move up and down for detector carrier box, (8) Portable and wireless HPGe detector, (9) Unshielded carrier box and holder for detector, (10)The tablet of remote control and software analysis.

Results and discussion

The set of calibration radio nuclides includes ^{241}Am,^{60}Co,^{137}Cs,^{192}Ir and ^{22}Na have been taken together in the present work, to cover the range of principal interest region of routine energy spectrum from 60 to 1332 keV**.** The Collected counts assumed that the activity per unit volume is constant and these are analyzed by using Genie 2000 software.In the present work the distant between ends of the detector cup to the outer surface of the waste drum is fixed at 5 cm.

The count rates of two radio nuclides spectra have collected individually for each. Figure -5 illustrated the attenuation factor effect and inverse square law of the radio nuclides spectra. However, for all energies it is clear that the count rate of spectra in location H1decreases relative to its level in

H2 due to increase the thickness of the cement and the radial distance between detector and radio nuclides location.

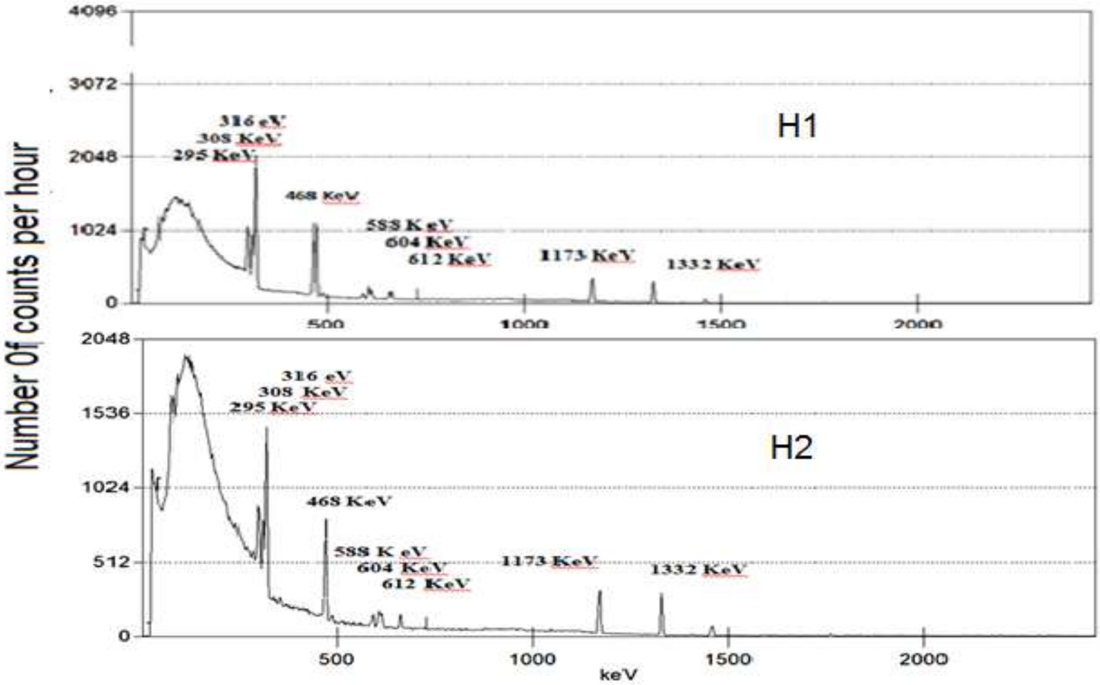

Figure 5- the reduction of the ^{192}Ir and ^{60}Co radioactive sources spectrum from the hollow cement tube H1and H2.

It follows up and according to the radial distance via solid angle between the detector and the location of radionuclide source; the efficiency progressively is changed between the cement tube hollows and the surface of the waste drum, where the increasing is occurred at the nearest point to the surface of the drum and decreasing appeared at locations near the center of the waste drum. As shown in Figure -6 several energies illustrated this behavior.

Generally, the expected response of HPGe detector in the range had used twelve photo peaks energy as shown in Figure -7, which indicates changing the efficiency curve relative to varying the radial distance between detector and radioactive waste drum for an average efficiency.

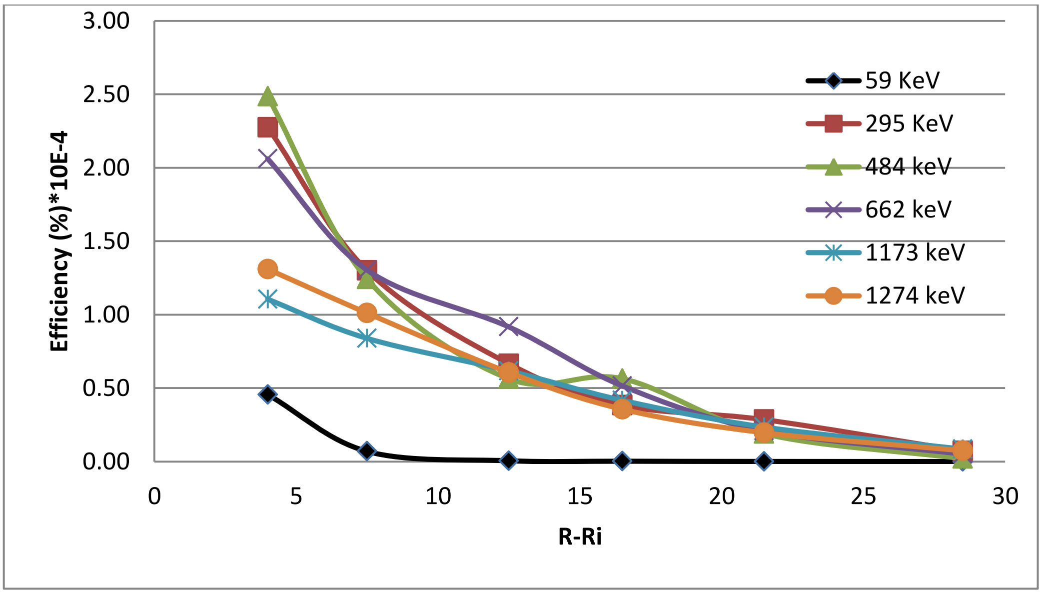

Figure 6-The measured average efficiency for each individual gamma energy at specific volume with radius R-R_i.

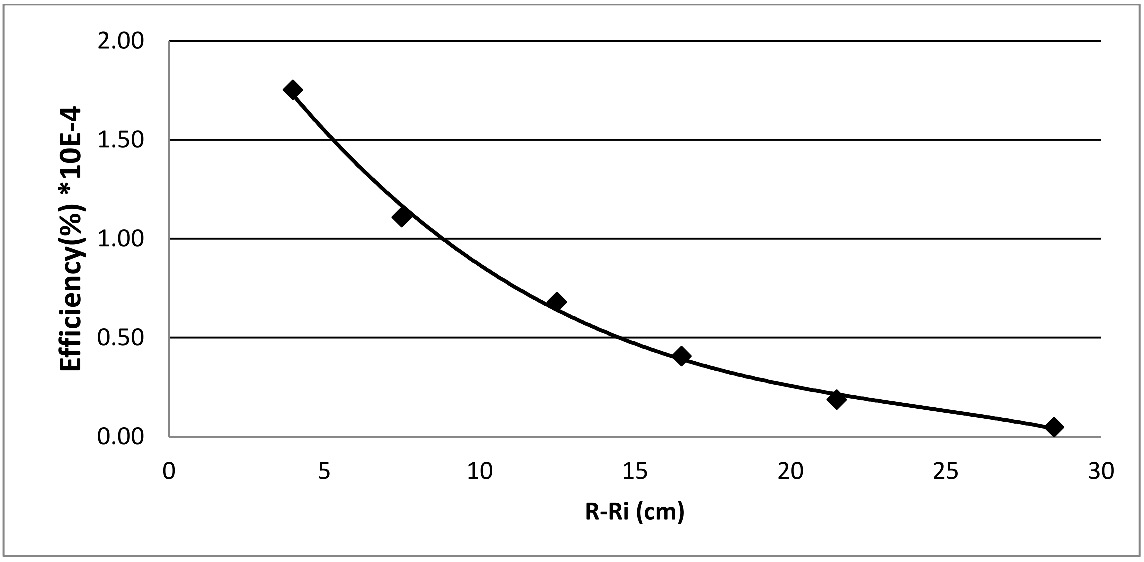

Figure 7- The HPGe detector average efficiency response of spectrum range of energy relative to specific volume of drums content with polynomial fitting order 4.

The efficiency altered for twelve gamma photo peaks and for six hollow cement tubes in drum. As shown in Figure-8 it was illustrated the effects of attenuation factor and inverse square law on the value of efficiency due to changing the distance between detector and the hot spot inside cement drum, which represents the location of radionuclide in one of six hollow cement tube. As a result the geometry changes successively and follows up a gradually change in the efficiency. The decreasing starts from H1, which is the nearest location of cement tube to the detector, to the H6 hole at the center, where the location of the radio nuclides. The hole H6 represents the greater distance of cement tube to the detector location.

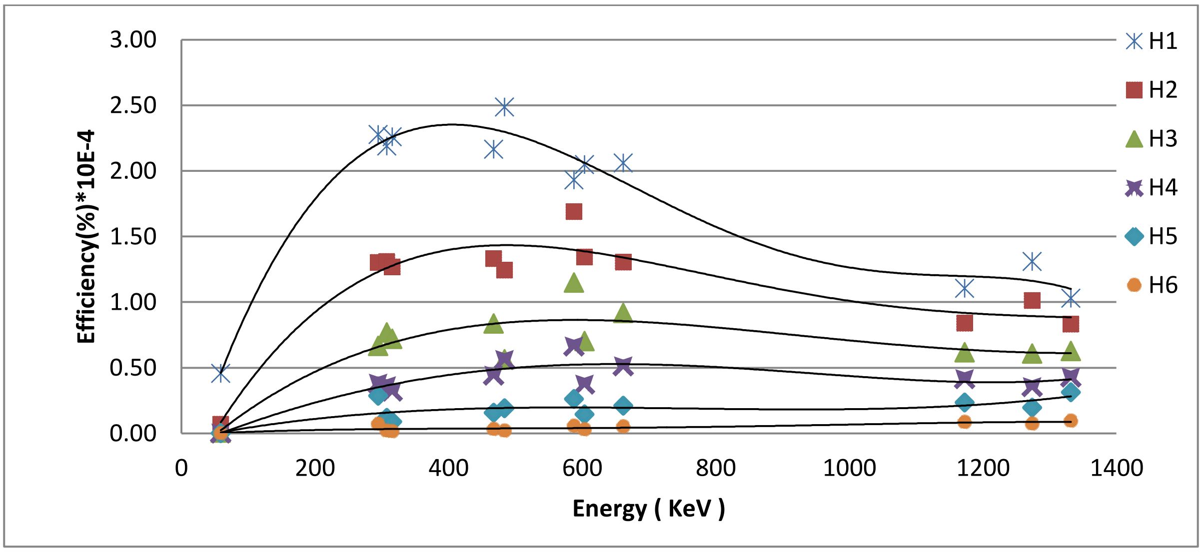

Figure 8-The HPGe efficiency behaviors for six holes (volume) for twelve photo peak with polynomial fitting order 4.

The average efficiency curve shown in Figure -9 for spectrum of twelve gamma photo peak energy, raised from the five radionuclide, as a radioactive material, distributed in a whole volume waste drum content.

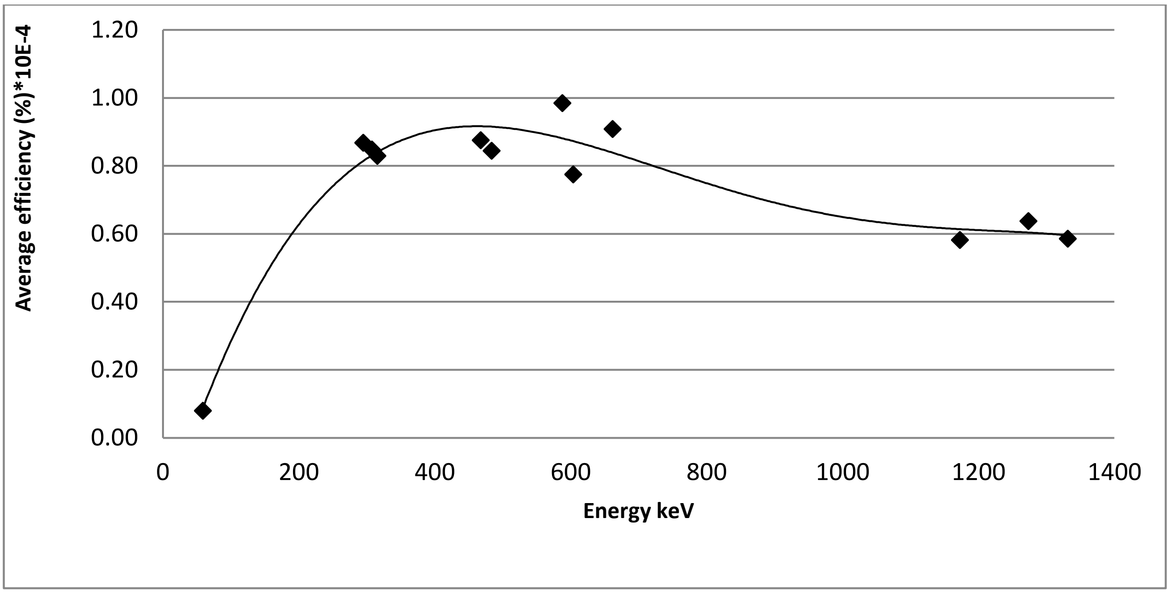

Figure 9-changing average efficiency of specific energy relative to whole volume of drums content with polynomial fitting order 4.

Conclusions

It was concluded that the behavior of the efficiency was affected strongly with distance between detector and hot spot (location of radionuclide inside drum) due to attenuation factor and geometry effects, where the radio nuclides spectra illustrated the effect of attenuation factor and inverse square law.Also, from the efficiency reduction of the scanning waste drum gives an idea that the characterization of drums containing low level activity of radioactive waste can be evaluated by the sample assay technique (or DA method) rather than NDA method. The DA method found more precise in procedure than NDA due to difficulties in time and efforts, in addition a radiation protection must be taken into account.

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