Determination of Window Analysis and Full Spectrum Analysis Method of Gamma Spectrometry Measurements in Low Level Activity

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Abstract. *Analysis of radionuclides at low level activity with the gamma spectroscopy requires high precision. The common methods used to determine radionuclide concentrations are Window Analysis (WA) and Full Spectrum Analysis (FSA). The purpose of this study was to obtain information in determining methods to be used when performing spectrum analysis because presumably there are differences in the results of the analysis and to ensure they are appropriate for low level radionuclide spectrum analysis. Low level activity radionuclides of ^{137}Cs and ^{60}Co with known activities between 2 Bq up to 6 Bq were used as sample; and ^{137}Cs, ^{60}Co and ^{152}Eu were as standard sources. The sample were counted for short time and long time. The results from comparative studies showed that FSA and WA had 11% to 24% and 1% to 20% of error, respectively. Otherwise there were a correlation between long measurements and short measurements with $R^2=0.9907$ and $R^2=0.7723$, respectively. Based on these data it can be concluded that WA method was more accurate and efficient in time required for counting. On other hand FSA is better in estimating the value of uncertainty. However by the correlation analysis results, the longer counting time would result in no significant difference.*

Keywords: Window Analysis, Full Spectrum Analysis, Gamma Spectrometry, Measurements, Radionuclide

Introduction

Gamma spectroscopy enables identifying and quantifying gamma ray emitters. The intensity of radiation is depended of the amount of the radionuclide. When gamma ray emissions from sample are collected and analyzed with gamma spectroscopy system, a gamma energy spectrum is produce radiation, gamma spectroscopy has the ability to determine both the level of radioactivity and radiation energy. There are several types of detectors are used to determine the concentration of gamma-emitting radionuclides. The HPGe (High Purity Germanium) detector is well known has advantages in resolution. In order to find out determination of window analysis and full spectrum analysis, the result were compared.

Environmental radioactivity monitoring has become an important activity that is frequently performed. The environmental presence of this radionuclide has been measured by High Purity Germanium detector and the content in the Indonesian marine environment is below MDA to 013 Bq/m^3 (Suseno et al., 2015) and classified as low level activity. This require an effective analysis method to identifying and quantifying low level activity concentration of radionuclide. In determining the concentration of radionuclides all types of detectors have their own characteristics, but it has the same basic principles. All radiations interact with the detector will be converted into electrical pulses. Measurement of gamma-emitting radionuclide concentrations in environmental samples by gamma ray spectroscopy system is The relative measurement techniques. The samples measured directly and comparing it with a standard source.

^{137}Cs has often been chosen as the most significant representative of anthropogenic radionuclides found in the marine environment. It is the most abundant anthropogenic radionuclide, and it is the largest contributor to dose among the anthropogenic radionuclides (Povinec, 2003).

The accuracy of gamma-ray spectrometric measurements is ultimately influenze by spectrum analysis. There are two methods for fitting the gamma-ray and determine concentration of radionuclide. These are the Window Analysis (WA) and Full Spectrum Analysis (FSA). The most widely used are FSA than WA because enable to automatically locate and fit the peaks. The FSA is a powerful tool for γ-spectra analysis, its reduction in required statistic and its increase in analyzable radionuclide (Caiolli et al., 2012). On other hand, Maphoto (2004) compared two analysis methods using an HPGe detector. The result were shown that two methods have the differences of 1% - 61 % depending in the samples. The WA method is known as a conventional method because analysed manual by individual gamma-ray. Furthermore, many gamma-ray lines associated with the primordial radionuclides are not resolved in energy and therefore form overlapping peaks. With the WA approach these overlapping peaks have to be deconvoluted by means of a peak-fitting procedure (Newman RT et al., 2007).

Present day, WA method is rarely used sinced the FSA method much shorter time frame, saving time and manpower. In some studies, analysis is performed twice to ensure the results. Based on ratio of percentage deviation (Mahmood Hafiz S., et al.,

2013) successfully using gamma-ray spectroscopy when combined with either the FSA and the WA data analysis method. All mention above, the comparison results of two analysis methods and produce different results. Therefore the purpose of this study to compare two methods of analysis using known activity in order check if they generate reliable results, as needed for analytical spectroscopy applications. Furthermore the sample were counted in short time measurement and long time measurement to determine how far the difference if the sample was measured with different measurements duration. At the end we can decide the advantage of each method and using appropriate methods for several case.

Material and Methods

Counting was performed by Gamma Spectroscopy system consists of High Purity Germanium (HPGe) detector, high voltage power supply, preamplifier (as part of the detector), amplifier, ADC and multichannel analyzer. HPGe detector with p-type coaxial 20% relative efficiency is used and GENIE 2000 as analysis software by canberra.

Canberra p-type coaxial HPGe detector (relative efficiency = 20%; FWHM = 0,801 keV at 1173 keV and = 1,78 keV at 1332 keV). It has a 0.5 mm thick carbon epoxy entrance window and low background lead castle (15 cm thick Pb). There are weekly fillings with liquid niterogen wich is mounted in dewar.

The detector mounted in vacuum enclosure (cryostat) included liquid niterogen Dewar and there are weekly fillings with liquid niterogen (capacity of 30 liters). Standar spectra for this study using IAEA-TEL-2011-03 as reference material. Further detail of IAEA reference materials are given in Tables 1.

In order to determine both of method we use reference material from IAEA with known radio-nuclide and activity concentration as a sample. The geometry and volume for the sample as same as with the standard source. In the case of different counting time, its sure that will be the difference results but with this case will show the ablity both of method and concluded the advantages of each method.

Energy and Efficiency Calibration

The essential requirements of a calibration are to establish an energy, efficiency and channel number by ADC system. The energy calibration of spectros-copy gamma performed by measuring mixed standar sources of known radionuclides provide by IAEA. The standar used for calibrating the detector is IAEA 2011-03 in liquid matrix with 500 ml volume. During the energy calibration procedure the HPGe detector system is used to acquire a γ-ray spectrum within energy range 121 keV to 1408 keV. The spectrum energy calibrated by setting regions of interest (ROI) around a number of peaks of interest (Table 1). The selected peaks are then manually calibrated by entering in the known energies corresponding to the ROI centroids. A linear fit equations was used to obtain the energy calibration parameters and this can be written as follows :

$$E = 0.4956\,x - 1.1173 \qquad (1)$$

Where E is the energy in keV and x is the channel number in the spectrum.

The standar sources has to be measured for long enough in order to get sufficient accuracy for calibration. The relationship between energy and channel number very sharp is shown in figure 1.

Table 1. The gamma ray lines used and their activities that have been corrected

Nuclide	Energy (keV)	Branching ratios	Activity	Uncertainty
152Eu	121	0.284	9 .025	0.2
152Eu	244	0.075	9.025	0.2
152Eu	344	0.266	9.025	0.2
137Cs	662	0.850	4.151	0.1
152Eu	964	0.146	9.025	0.2
152Eu	1112	0.135	9.025	0.2
60Co	1173	0.998	6.755	0.2
60Co	1332	0.999	6,755	0.2
152Eu	1408	0.205	9.025	0.2

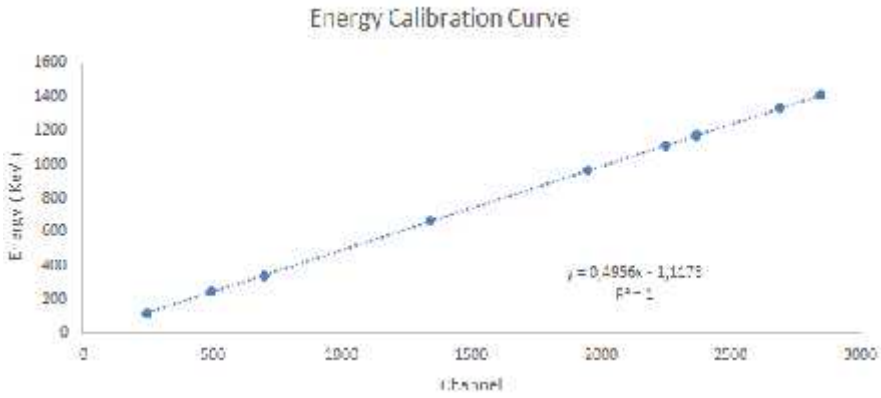

Figure 1. Energy calibration curve

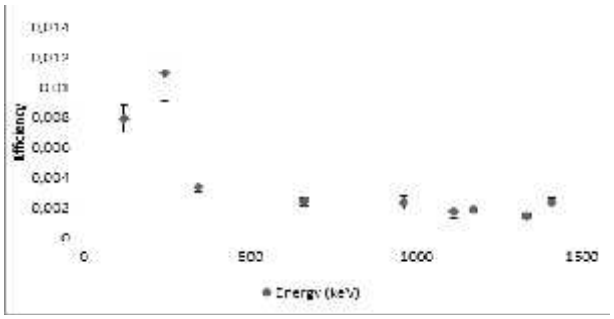

Figure 2. Efficiency calibration curve

To establish an energy calibration, we have located of datasource's peaks, which correspond to the energy of the entries in the calibration file. The mixture of three sources with known energies was made to ensure that the calibration covers the entire energy is to be used and plotted in curve (Figure 1).

The peak efficiency must be calculated for a set of well defined single peaks (or as close to a single peak as possible) using a standard source of the same geometry and count rate as is expected for the actual samples. Furthermore, the peaks should cover the entire energy range of interest.

Sample Measurement

To produce valid data in measurements we use the same matrix, volume and geometry refered to standar. The sample is measured with short time measurement and long time measurement in 9128 second and 54000 second respectively. The spectra marked at 662 keV, 1173 keV and 1332 keV then analyzed using Window Analysis and Full Spectrum Analysis. After that all the results obtained will compare each other.

Window Analysis

Window analysis peak locate will assign the peak locations based on the library energies. During the locate phase, the first step is the determination of the continuum backgroud. In this step, the photo-peaks are eroded until they subside into the continuum. The summary formulation of net peak area for a single peak is calculated as follows.

$$S = G - B \qquad (2)$$

Where S is the net peak area, G is the sum of gross counts in the peak ROI and B is the continuum. A Linear continuum, B, ilustrated in figure 2, is calculated from the spectrum using the equation

$$B = \left(\frac{N}{2n}\right)(B1 + B2) \qquad (3)$$

Where N is the number of channels in the peak ROI, n is the number of continuum channels on each side, B1 is the sum of counts in the continuum region to the left of the peak, and B2 is the sum of counts in the continuum region to the right of the peak.

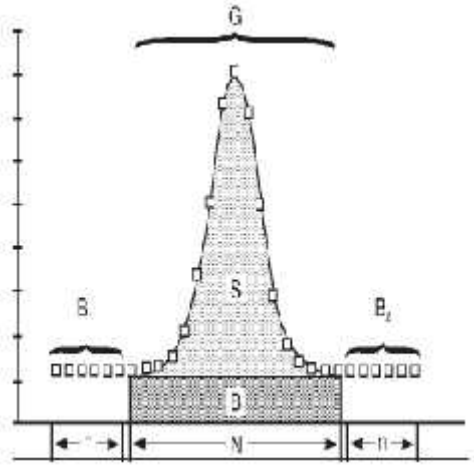

Figure 3. Peak Area for singlet peak

Unlike the Full Spectrum Analysis The specific activity (in $Bq.l^{-1}$) of a radionuclide in a sample with weight in liter measured during a period t (in seconds) is given by the equation (4) :

$$Act = \frac{N\ f\varepsilon}{W\ .I\gamma .\ \varepsilon} \qquad (4)$$

Where N is the net number of counts in the photopeak, W is weight, Iγ is the gamma intensity, ε is the peak detection efficiency.

In WA method after a pulse height spectrum has been recorded, the peaks of interest must be identified. The idenified peaks is marked on the graphics display and determine the number of counts in the peak region. The activity concentration is calculated by finding the same ROI peak in the spectrum of measurement sample and applying the previously obtained energy dependent nuclide efficiency of standard source.

Full Spectrum Analysis

The Genie 2000/Genie-PC architecture currently provides two different algorithms for calculating peak areas in a spectrum : "Library (Gamma-M)" and "Sum/Non-Linear LSQ Fit". As a general rule, the "Library (Gamma-M)" method is best suited for situations where only specific nuclides are of interest. The "Sum/Non-Linear LSQ Fit" algorithm is best suited for Full Spectrum Analysis and when the spectrum contents are unknown. After the ROI determination completes the calculation of the boundaries of the ROI, the number of continuum background channels to be used to the left and right of the peak ROI is then determined. The peak fits locate in the spectrum are done to a peak region whose limits parameter using a peak model consisting of a pure Gaussian.

$$F_G(x_i) = he^{-\frac{(x_i-E)^2}{z^2}} \qquad (5)$$

Where h is the height of the peak, xi is the energy at channel I, E is the energy of the incident gamma ray, and Z is a measure of the peak width.
In this algoritm, we calculate the photopeak areas by fitting the Gaussian model to the data in the least squares sense, which requires that the quantity

$$x^2 = \sum_i W_i\ [Y_i\ \ -F(x_i,P_j)]^2 \qquad (6)$$

Where W_i is the weighting factor assigned to the i^{th} data pont, Y_i is the net counts (original data minus the background continuum) and P_j is the j^{th} parameter to be fit.

Full spectral shape is used in the full spectrum analysis. The automatically peak search will be performed on the region between peak seacrh start and end channels. The sensitivity threshold (i.e. the number of standard deviations above background a feature must be to be considered a peak) controls the sensitivity of the peak locating algorithm. For example, if the sensitivity is set to 5, and the average background in a region is 10000 counts, any feature with a height smaller than about 500 counts above background will be ignored. (One standard deviation of 10000 is 100.) typically the sensitivity should be between 3 and 10. Determining the ROI limits will also make the determination whether adjacent peaks are going to be analyzed as two singlets or a multiplet.

Results and Discussion

In order to determine the activity concentration of gamma-ray spectrum we use GENIE 2000 software and the detector is calibrated with known activity measured in the same geometry as the sample. (S. Zahn Guilherme et al., 2009). To performed evaluation of software for gamma spectrum analysis and GENIE 2000 performed very well all around and reliable for FSA as well as WA method. Furthermore will show the results of each method and compare both of them at the end. The channel dependence on the energy is shown in Fig. 3. The straight line fitted to the points shows a very high degree of correlation, and therefore we can conclude the spectroscopy gamma is operated well.

Windows Analysis Results

Gamma spectroscopy measurements produce a lot of peaks entire of gamma energy. In attempt to determine region of interest (ROI) from the spectrum, WA method only specified the area to be analyzed. As general rule, the "User Specified" method is best suited for an application where only specific regions of the spectrum are of interest.

As we know difference in duration of counting time it mean difference result but that is important think to see how much the difference. The largest is 27% percentage difference value by WA that is indicate the method is poor in control the statistical processes in involving the duration of counting time. In this method, only ROI peak is considered, thus, it takes longer to measure to achieve statistical significance and also a relatively long time is required for data analysis. In the case single pleak analysis WA produce lower value than the certificate due to ignoring the compton continuum in the WA that implies throwing away some counts that can be used for data analysis.

Full Spectrum Analysis Results

Unlike the WA method, FSA uses count numbers from full-absorption peaks including the contribution from Compton scattering, thus, background energy spectrum and the reference energy spectrum of measurement nuclides were used (Jeong Meeyoung et al., 2014).

The activity concentration calculated with full spectrum analysis method are shown in Table 3, where the percentage is much lower than the WA method. The largest is 10% percentage value by FSA that is indicate the method is quite good involving the duration of counting time into statistical processes. From table 2 and table 3, it is clear that measuring times have to be increased for WA and FSA short measurement in order to obtain good counting statistics with these methods.

Comparison between the FSA and the WA Methods

The FSA method yields concentrations of radionuclides (i.e., ^{137}Cs and ^{60}Co) just like WA methods, which makes the comparison between the two methods easy. Were plotted on the histogram to show the variations in activity concentrations for various samples and duration of counting time.

Table 2. Radionuclide concentration short measurements

	Nuclide	Activity (SM)	Activity (LM)	Percentage Difference
IAEA 2011-02	^{137}Cs	2.287 ± 1.137	2.829 ± 0.54	21.188
	^{60}Co	5.048 ± 1.55	4.489 ± 0.71	11.722
IAEA 445	^{137}Cs	5.579 ± 1.61	6.717 ± 0.842	18.51
	^{60}Co	3.148 ± 1.50	2.394 ± 0.47	27.21

Table 3. radionuclide concentration sample 2 Short Measurements

	Nuclide	Activity (SM)	Activity (LM)	Percentage Diffrence
IAEA 2011-02	^{137}Cs	3.560 ± 1.08	3.200 ± 0.272	10.650
	^{60}Co	5.250 ± 1.33	5.100 ± 0.484	2.898
IAEA 445	^{137}Cs	8.129 ± 0.73	7.677 ± 0.393	5.719
	^{60}Co	3.280 ± 0.574	3.197 ± 0.287	2.562

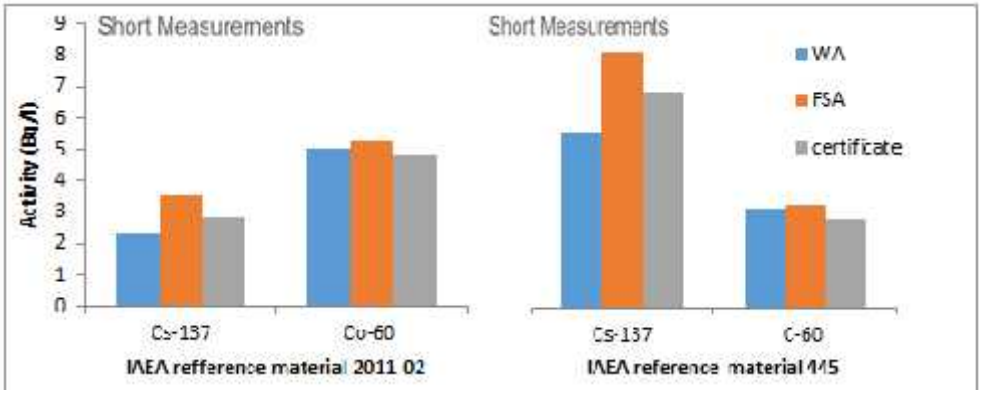

Figure 4. The comparison of activity concentration (short measurement) in both Window Analysis and Full Spectrum Analysis

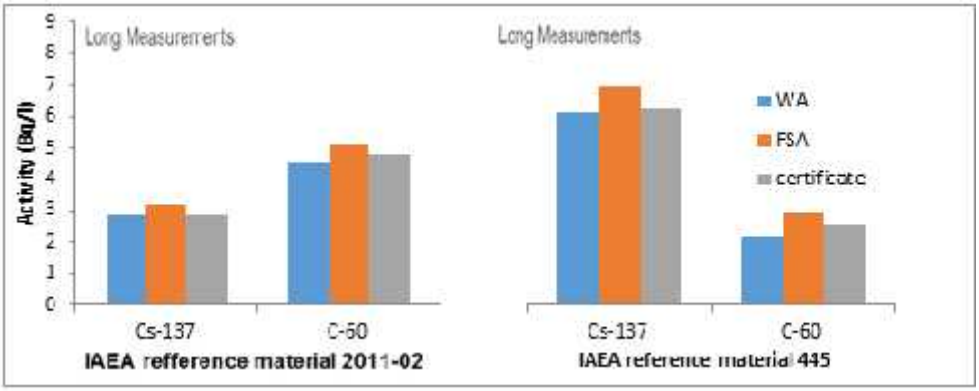

Figure 5. The comparison of activity concentration (long measurement) in both Window Analysis and Full Spectrum Analysis

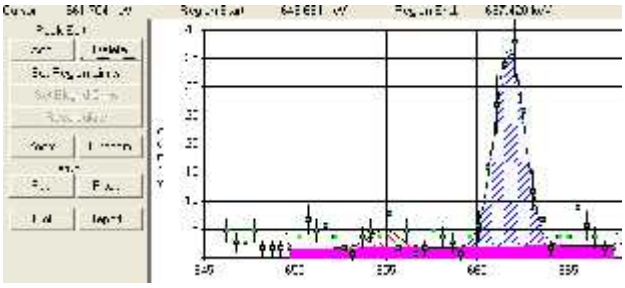

Figure 6. in the case of peak fit ^{137}Cs by Full Spectrum Analysis

Figure 4 shown there are difference value the differences because of the counting duration is not enough to meet the good statistic. As we know short time duration in measurement produce large error in unsufficient statistic therefore both of methode produce a result with poor accuracy. The best result in short duration measurement is achieve by window analysis with 5% error percentage and the worst result getting from full spectrum analysis in 25% error percentage. In case of short measurement FSA and WA method cannot achieve best accuracy, both of method produce arround 20% error.

Prediction accuracies of long time measurements in the WA methods were comparable with FSA method, from this results it can be notice that the FSA produce the activity concentration higher than the certificate in all condition measurements and at the same time WA produce lower than the certificate.

The value in the net peak area establishes the continuum under the peak ROIs. If you have two peaks that are close together, reducing the number of continuum channels may give better results. If you have poor peak statistics and there are no other peaks nearby, increasing the number of continuum channels establishes the continuum more accurately but makes it more likely that close lying peaks will be considered as a multiplet instead of as a singlet.

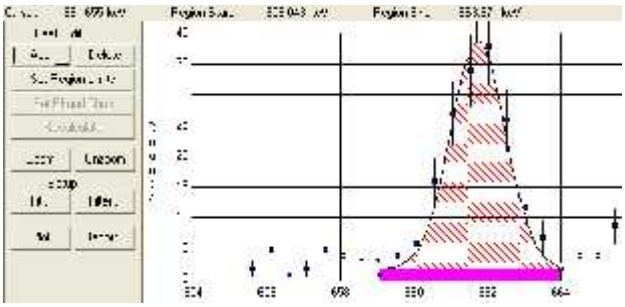

Figure 7. in the case of peak fit ^{137}Cs by Window Analysis

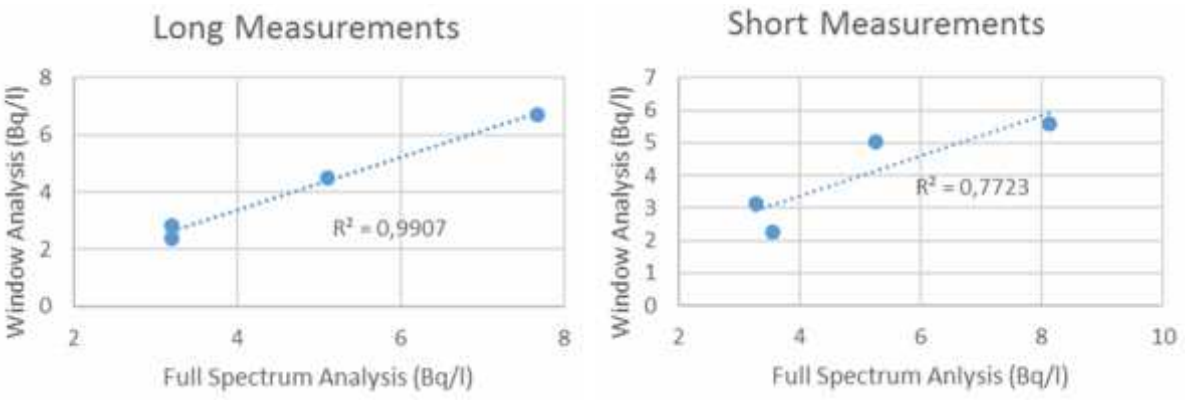

Figure 8. The comparison WA and FSA plotted as a linear function

FSA determine the area of interest by estimate the baseline through iteratively filtering the spectrum to remove that resemble peaks according to the current shape calibration. The width of the filter is a function of local peak width times the specified erosion width (typically 1) and the entire continuum is changed. Figure 4 show the FSA is over in determine peak continuum both on the right and on the left. More over unlike the WA method, FSA uses count numbers from full absorption peaks including contribution from Compton scattering, that probably effected the measeurement level is higher. Furthermore the higher concentration by Full Spectrum Analysis is probably cause by coincidence summing effect. There are posible to recalculated the area of FSA method, the results are called "corrected spectra" and an example is shown in Fig. 7 all the quantities used in the data analysis are obtained from these corrected spectra.

The Window analysis using manual marker, this set of controls specifying the number of channels to be used for the left background and the right background and wheter the continuum between the background is liniear or step.

The region of interest fitted the gaussian distribution properly as seen on Figure 4. Window analysis method have the advantage to determine the Area of interest on the gaussian distribution with more detail because fitted by manually. On other hand, the WA have large percentage difference in the case of measurement duration. FSA is powerfull to control a lot of parameters and have the advantages to resolve the problem.

In figure 8, the comparison of activity concentration were plotted as a linear function, for WA and FSA method in short time measurement and long time measurement.

High correlation between radionuclide concentrations measured by the FSA and the WA methods in long time measurement indicates that accumulation of gamma rays of each radionuclide in its represen-tative WA represents its distribution in the all of the energy spectrum. Figure 8 show that, to obtain a similar accuracy as with the WA, measuring times using FSA method is must be long time measure-ment. Other crucial parameter is the uncertain value produce by measurement, because that indicate how far an experimental quality might be from the "true value".

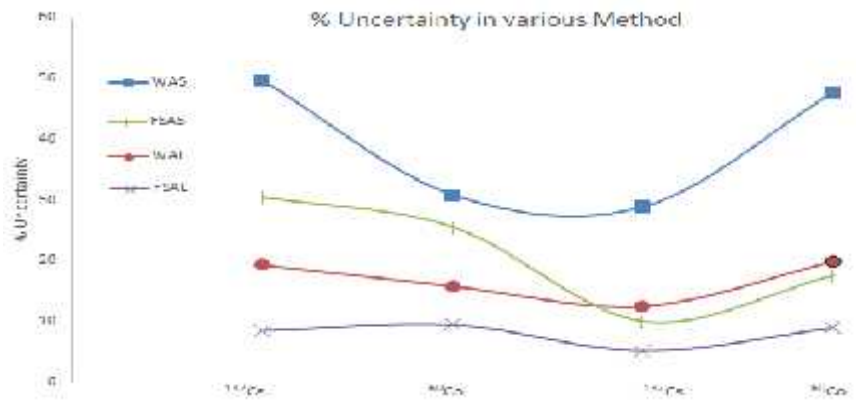

Figure 9. % Uncertainty in various method and sample

Figure 9 compared the uncertainty for both method and the largest uncertainty value obtained from window analysis in short duration measurement.

Contrary to WA, which only uses the data in a number of peaks for analysis, FSA includes all the spectral information in the data analysis. The increased amount of information will decrease the statistical uncertainties in the method, thus giving this method an advantage (Maphoto KP, 2004).

As mentioned earlier (Hendriks et al., 2001), significant improvements in gamma ray spectrum analysis have been obtained by implementing the full spectrum analysis method. Accumulation of good statistic is required for the reliability of this method. This was demonstrade by comparing the uncertainties associated with measurements of varying duration and radionuclide. Results from shorter measurements were more uncertainty than those from long ones.

Conclusions

Both of method was observed in different counting time and we can conclude that the Wa method yielded slightly better but at the same time WA method loses some counts by ignoring continuum area. FSA method showed the value is higher than te certificate may cause by compton effect and coincidence summing. It will be perfectly for using FSA after the gamma spectroscopy built in anti compton and coincidence. However by the correlation analysis results (R^2=0,997), the longer counting time would result in no significant difference. The window analysis with the marker method is recommended for spectra where there are other peaks close to the calibration peak. On other hand FSA is better in estimating the value of uncertainty. Although the system is powerful enough for low level activity measurements, it is important, nowadays, to have a great statistical in short time measurement: which is a challenge for scientists.

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