Study of a Laboratory-based Gamma Spectrometry for Food and Environmental Samples

M. N. Islam[1], H. Akhter[2], M. Begum[3], Y. Mawla[4], M. Kamal[5]

[1,2,3,4] Electronics Division, Atomic Energy Centre, Bangladesh Atomic Energy Commission,
P.O. Box No. 164, Dhaka, Bangladesh.
[5]Radioactivity Testing and Monitoring Laboratory, Atomic Energy Centre, Bangladesh Atomic Energy Commission,
P.O. Box No. 1352, Chittagong, Bangladesh.

Abstract— *A comprehensive study on a laboratory-based Gamma Spectrometry has been presented in this paper for food and environmental samples. The system comprises of HPGe detector with proper cooling for minimizing thermal generation of charge-carriers and appropriate shielding to reduce background emission; associated processing electronics and acquisition as well as analysis software. The choice of HPGe detector for laboratory-based Gamma Spectrometry, its radiation interaction mechanism and system optimization have been presented.*
Keywords—*HPGe Detector, Radiotracers, Efficiency Calibration, Energy Calibration, Activity Concentration.*
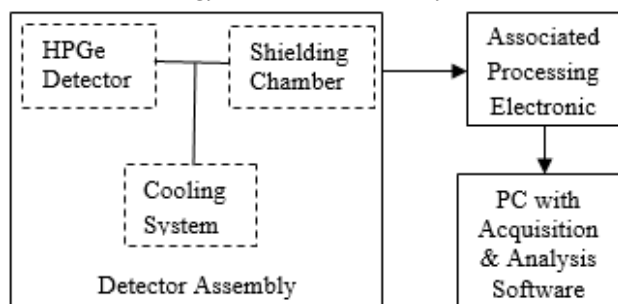
Fig.1: Block Shows the Configuration of Laboratory-based Gamma Spectrometry.

I. INTRODUCTION

Environmental Gamma Spectrometry is an essential tool for modeling spatio-temporal processes through coordination of data from different radiotracers. The two main advantages of environmental gamma spectrometry are: it is a nondestructive technique; and the measurement of different radiotracers is done simultaneously. The principal radiotracers of interest for environmental studies, measured via gamma spectrometry, are: ^{210}Pb, ^{226}Ra, ^{137}Cs, ^{241}Am, ^{238}U (^{234}Th), ^{235}U, ^{232}Th (^{228}Ac), ^{40}K, ^{7}Be, ^{212}Pb, ^{222}Rn, ^{214}Pb ^{214}Bi and ^{88}Y. [1]. Gamma-ray spectrometry has been widely used in various environmental and natural sciences by its ability to determine the concentrations of each radionuclide of the

samples, and also because of the easiness in sample preparation and measurement procedures [2]. The purpose of an HPGe detector is to convert gamma rays into electrical impulses which can be used, with suitable signal processing, to determine their energy and intensity. All HPGe radiation detectors are either coaxial HPGe, well-type HPGe or broad energy HPGe (BEGe) just large, reverse-biased diodes. The germanium material can be either "n-type" or "p-type". The type depends on the concentration of donor or acceptor atoms in the crystal. [3]. Liquid nitrogen (LN$_2$) and cryocooler (electric cooler) would be used to cool HPGe detectors. Detector assembly also contains lead shielding chamber to reduce the background radiation emission. The later part is Multichannel Analyzer (MCA) means a DAQ card along with acquisition and analysis software to measure the pulse height, type and activity of incoming radiation.

II. METHODOLOGY

Laboratory-based Environmental Gamma Spectrometry comprises of detector assembly, associated processing electronics and a PC with gamma acquisition and analysis software. The detector assembly contains High Purity Germanium (HPGe) detector, the cooling system and the shielding chamber.

2.1: Detector

The choice of the best HPGe detector in case of Gamma Spectrometry for one particular measurement situation is based on some basic concepts such as how the detection process works, how gamma rays penetrate materials and the principles of gamma-ray spectroscopy. On the basis of these concepts the following parameters would be taken into consideration while the best choice of a detector.

2.1.1: Minimum Detectable Activity (MDA)

One measure of the quality of a spectrum is the minimum detectable activity (MDA) of the detector system. The

resolution, background and efficiency of the detector are related to the MDA. This relationship may be simply stated as below

$$MDA(E) \sim \frac{\sqrt{R(E)B(E)}}{\varepsilon(E)} \qquad (1)$$

R(E) is the energy resolution of the detector as a function of energy; B(E) is the background counts per KeV (unit energy) as a function of energy and ε(E) is the absolute efficiency of the detector as a function of energy. The MDA varies with energy because the quantities on which it depends vary with energy. Here we have separated out all the factors in the MDA that only depend on the detector itself. The gamma rays per decay, the shield and count time affect the MDA, but will do so in the same way for all detectors.

2.1.2: Detector Resolution

Energy resolution is the dominant characteristic of a germanium detector. Gamma-ray spectrometry using high purity germanium detector is enhanced by the excellent energy resolution which can help to separate and resolve various close energy gamma-ray peaks in a complex energy spectrum. The full width at half maximum of the full energy peak known as FWHM and sometimes referred as a measure of energy resolution. The units of FWHM are expressed in KeV for Ge detector and are defined at specific, characteristic full energy peaks associated with standard sources such as 662 KeV for a ^{137}Cs source or 1332 KeV for a ^{60}Co source. The energy resolution of the germanium detector can affected by the number of electron-hole pairs created in the detector, incomplete charge collection and electronic noise contributions. The effect of these three factors depends on the properties of the detector and the gamma-ray energy [4].

2.1.3: Detector Efficiency

The detector efficiency in Eq.2 will potentially have the most effect on MDA. In this Eq.2, ε(E) is the absolute efficiency at the specified energy. ε(E) will depend on the detector-to-sample geometry, and many other energy dependent factors, including gamma-ray absorption in matrix, detector dead layers and the intrinsic efficiency of the detector. The IEEE-325 relative efficiency is no longer a suitable indicator.

The absolute detector efficiency at that energy is calculated by dividing the net count rate in the full-energy peak by the decay corrected gamma-ray-emission rate of the standard source. Efficiency curves were constructed from these full-energy-peak efficiencies.

ε_{abs}

$$= \frac{\text{Total number of counts recorded under the photo peak}}{\text{Total number of photons emitted by the standard sources}}$$

$$\varepsilon_{abs} = \frac{\text{cps experimental}}{\text{cps theoritical}}$$

$$\varepsilon_{abs} = \frac{\left(\frac{C}{S}\right)_{std} - \left(\frac{C}{S}\right)_{sample} - \left(\frac{C}{S}\right)_{BG}}{\left(\frac{C}{S}\right)_{theo} Exp\left(-\ln(2) \cdot \frac{t}{t_{\frac{1}{2}}}\right)} \qquad (2)$$

$\left(\frac{C}{S}\right)_{std}$ Count of soil sample with standard solution, $\left(\frac{C}{S}\right)_{sample}$ Count of soil sample without standard solution, $\left(\frac{C}{S}\right)_{BG}$ Count of background, $\left(\frac{C}{S}\right)_{theo}$, Counting of gamma ray of used standard solution, t the time of decay, $t_{1/2}$ half-life of the radionuclide [5].

2.1.4: Compton Suppression System (CSS)

For a given HPGe detector, a Compton Suppression System (CSS) will always reduce Compton Background. It is also called an "active shield." It reduces the cosmic background because a cosmic ray produces events (counts) in detectors.

2.1.5: Dead Layers, Windows and Absorption

Any gamma rays stopped in the dead layer do not produce an output. Below 150 KeV, the GMX has higher efficiency and below 100 KeV, the difference increases rapidly as one go down in energy. This is because the dead layer of the GEM (~600 microns) is much larger than that of the GMX (~0.3 microns).

Thin window PROFILE X series GEM-FX8530, compared to a GEM80 76 mm diameter x 87 mm depth. The much higher absolute efficiency of the FX85 at all energies below 160 KeV.

The absorption processes are a function of energy and described by the exponential attenuation equation below:

$$I = I_{0e^{-\mu(E)X}} \qquad (3)$$

Where I_0 is the unattenuated gamma-ray flux, I is the flux after passing through the material and μ is the linear attenuation coefficient of the absorber and x is the thickness.

2.1.6: Detector Cooling System

A High Purity Germanium (HPGe) detector is required a High Voltage for performing its proper operation and hence the detector should be cooled sufficiently in order to reduce the thermal generation of charge carriers to an acceptable level. Otherwise, the noise due to leakage current would destroy the energy resolution of the detector. Liquid Nitrogen (LN$_2$) which has temperature

77^0K (-196^0 C) is the common cooling medium of the detector. The Liquid Nitrogen (LN_2) Dewar served as a reservoir of liquid nitrogen [4]. Electric Coolers offer compared to liquid nitrogen (LN_2) to cool HPGe detectors, the limited cooler life time, higher initial investment and need for periodic maintenance have always been major drawbacks. With the introduction of pulse tube coolers in CANBERRA's Cryo- Pulse 5 and Cryo-Pulse 5 plus, these issues are no longer present [6].

2.1.7: Detector Shielding Arrangement

The detector shielding arrangement is usually fabricated by using locally available lead. Because of high density (11.4gm/cc), large atomic number (Z=82) and comparatively low cost, lead is the most widely used material for construction of the shields. The shielding of the detector from the environment radiation is an absolute necessity for low level measurement of activity. The shielding not only reduces the background resulting from cosmic radiation, natural radioactive traces in the building material or in the surface of the earth but also from nearby nuclear facilities and other radiation sources like the ambient air, which presumably contains trace amounts of radioactive gases such as Radon.

2.1.7.1: Background Radiation

The counting system must have a background as low as is attainable with a minimum spectral lines originating from natural radionuclides which may be present in the system components and the surrounding environment, i.e., the walls, floors, furnitures etc of the counting facility. Construction materials such as concrete, plaster and paints which contains barites (barium sulphate) will tend to cause elevated backgrounds due to natural radionuclides [4].

2.1.8: Detector Interaction Mechanism

Four major interaction mechanisms play an important role in the measurement of photons. These mechanisms are: photoelectric effect, coherent scattering, incoherent scattering and pair production. The photon energy of major interest for environmental spectrometry studies ranges between a few KeV to 1500 KeV. The term "low energy" will be used here for the energy range 1 to 100 KeV, "medium energy" for energies between 100 and 600 KeV and "high energy" for energies between 600 and 1500 KeV.

2.1.8.1: Photoelectric Effect

In the photoelectric effect, there is a collision between a photon and an atom resulting in the ejection of a bound electron. The photon disappears completely, i.e. all its energy, Ep, is transferred to the electron. The amount of energy, Ee, which is transferred to the electron, can be calculated if the binding energy, Eb, of the ejected electron is known:

$$Ee = Ep - Eb \qquad (4)$$

2.1.8.2: Coherent Scattering

In the coherent scattering process no energy is transferred to the atom. The electromagnetic field of the photon sets atomic electrons into vibration. The electrons then re-emit radiation of the same magnitude as the interacting photon and mainly in the forward direction. The cross-section for coherent scattering decreases rapidly with increasing photon energy. This process can be neglected for photon energies above 100 KeV. The differential cross section per atom for this process as a function of scattering angle θ is written as follows:

$$\frac{\partial \sigma_{coh}}{\partial \theta} = \frac{Z r_0^2}{2} \left(1 + \cos^2(\theta)\right)[F(x,Z)]^2 . 2\pi . \sin(\theta)$$
(5)

with the parameter x defined as:

$$x = \frac{\sin(\theta/2)}{\lambda} \qquad (6)$$

Where r_0 is the classical electron radius, F(x,Z) is the atomic form factor and λ the photon wavelength.

2.1.8.3: Incoherent or Compton scattering

In the incoherent or Compton scattering process, only a portion of the photon energy is transferred to an atomic electron. The remaining energy appears as a secondary photon. The direction (scattering angle theta) and energy of the secondary photon, Ep', are related by the following equation:

$$E'_p = \frac{E_p}{1 + \alpha(1 - \cos(\theta))} \qquad (7)$$

With α defined as

$$\alpha = \frac{E_p}{m_0 c^2} \qquad (8)$$

Where m_0 is the rest mass for an electron and c is the speed of light in vacuum.

2.1.8.4: Pair Production

The pair production mechanism only occurs for photon energies above 1022 KeV. Here photons are converted to electron-positron pairs under the effect of the field of a nucleus. Since one electron and one positron are formed, the photons must have energies equivalent to at least two

electronic masses (2 x 511 KeV) and the excess photon energy is shared between the created electron and positron pair. The annihilation of the positrons produces two photons in opposite directions, each with 511 KeV. The total cross-section for this process increases with energy above the threshold energy.

$$\sigma_{Total} = \sigma_{Photoelectric} + \sigma_{Pair\ Production} + \sigma_{Coherent\ Sc.} + \sigma_{Incoherent\ Sc.} \quad (9)$$

The total probability of interaction per unit path length for a photon is proportional to the sum of the total individual cross-sections [1].

2.2: Processing Electronics
The electrical charge output from the detector would be acquired and processed through an Electronic System consists of preamplifier and high-voltage filter, a main amplifier, a count-rate meter, a pulse height analyzer, a precision pulse generator, a detector bias supply and an oscilloscope[7].

2.2: Gamma Acquisition and Analysis
The Genie™ 2000 is Gamma Acquisition and Analysis (GAA) Software. The window in fig.2 is it's user interface for acquiring and analyzing nuclear spectra. It consists of the Title Bar, the Menu Bar, the Toolbar and the Display Status Line at the top of the screen, the Control Panel, the Spectral Display, the Status Pages, and the Report Window in the main part of the window and at the bottom of the screen, the Analysis Status Line [8].
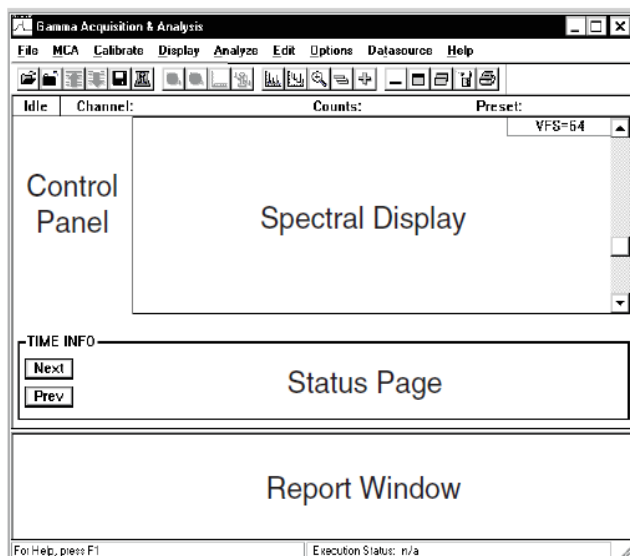

Fig.2: The Basic Gamma Acquisition and Analysis (GAA) Software Window.

2.3.1: Energy Calibration
Energy calibration establishes a linear relationship between the spectrum's channels and their energy levels.

By calibrating two peaks, one at each end of the spectrum, the energy of any other peak can be estimated fairly accurately [7]. For instance, the ^{40}K and ^{137}Cs radionuclides would be measured from their respective γ-ray energies 1460 and 661.66 KeV respectively [9, 10].
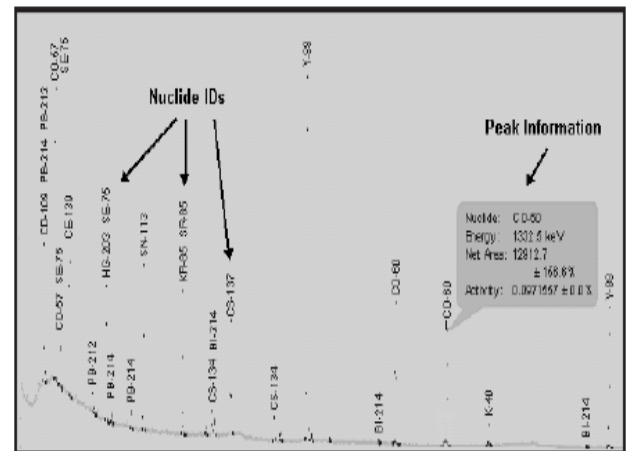

Fig.3: Nuclide IDs and Peak Information

2.3.2: Efficiency Calibration
For radioactivity measurement, the gamma-ray spectrometry with a high-purity germanium (HPGe) coaxial detector is widely used. In the method, a detection efficiency curve, that is, a set of photopeak efficiencies over the energy region of interest must be known in advance. The detection efficiency curve depends not only on a detection system but also on a sample shape and a sample matrix with different density and height of environmental samples.
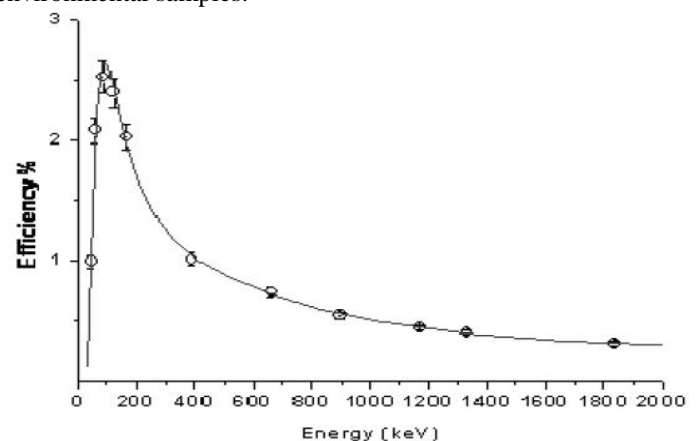

Fig.4: Efficiency calibration curve obtained from the reference geometry bottle 250 ml with different gamma-ray energies.

The efficiency curve in fig.4 shows the energy range 46.54-1836 KeV as well as two regions of different behavior because distinct attenuation and absorption processes dominate. At low energies, the efficiency rises rapidly because of abrupt reduction in the attenuation in

radioactive source, detector cap or inner dead layer. A maximum is reached for an energy value which depends on the detector and source characteristics. Above a few hundreds KeV the efficiency decreases monotonically.

2.3.3: Calculation Specific Activity

Absolute Efficiency, ε_{abs} would be used to calculate the specific activity in Bq.kg $^{-1}$. Therefore, A_E *γ, i* of a radionuclide i and a peak at energy Eγ, is given by

$$A_E\ (\gamma, i)\ =\ \frac{NP}{t_c\ .I_\gamma(E_\gamma)\varepsilon(E_\gamma).M} \qquad (10)$$

Where NP is the number of counts in a given peak area corrected for background peaks of a peak at energy *Eγ*, ε (E_γ) *the* detection efficiency at energy *Eγ* , t_c is the counting lifetime in second, $I\gamma$ (E_γ), the number of gammas per disintegration of this nuclide for a transition at energy *Eγ*, and M is the weight of the measured food or environmental samples in kg [6].

2.4: Discussion and Analysis

The Laboratory-based Environmental Gamma Spectrometry is a multidisciplinary research tool for detection and measurement of activity and energy in different radionuclides. The state of the art Environmental Gamma Spectrometry is High Purity Germanium (HPGe) detector, Cooling System, Shielding, the associated processing electronics, Gamma acquisition and analysis software. Although there are many other components that constituent the system, the performance and ability of the system mainly depends on the right choice of the detector. Therefore, the characteristic parameters such as MDA, depends on detector resolution, background energy and detector efficiency, appropriate cooling and shielding arrangement should be taken in consideration while selecting a detector for a specific application. Moreover, detector types like coaxial, well-type or broad energy as well as dominant carriers n-type or p-type also essential. The system optimization can be done through CSS, thickness of windows, dead layers and absorption process. The efficiency of the detector depends not only on gamma abundance but also on geometric effects and coincidence summing. Monto Carlo simulation of the system for incomplete charge collection module may enhance the system performance. The sample metrics and sample geometry affect the performance of the Gamma Spectrometry in the same way. The associated processing electronics must be wide flexibility and good agreement with detector system. The standardization and accreditation of the acquisition and analysis software should be maintained.

III. CONCLUSION

An ample study of laboratory-based Gamma Spectrometry for food and environmental samples has been presented in this paper. Syntheses of the system components, system evaluation and optimization have been provided. User interface regarding Gamma Acquisition and Analysis, efficiency calibration, energy calibration and factors affecting the efficiency calibration also have been described briefly. The system is suitable for identifying concentration of low to high energy environmental radiotracers.

REFERENCES

[1] F. J. Hernandez "Optimization of environmental gamma spectrometry using Monte Carlo methods" Ph.D. thesis, Uppsala University, Sweden. 2002.

[2] Jeong Hee Han and Jeong-Heon Choi, Broad Energy HPGe Gamma Spectrometry for Dose Rate Estimation for Trapped Charge Dating, J. of Analytical Sci. & Technol., Vol. 1(2), pp.98 -108, 2010.

[3] ORTEC, The Best Choice of High Purity Germanium (HPGe) Detector, www.ortec online.com, Available on-line 2013.

[4] Nurul Absar "Study of the Radioactivity in Soil and Tea Leaf and Transfer Factor of Radionuclides", M.Phil. thesis, Chittagong University, Bangladesh, 2012.

[5] D. Willems, H. van der Weijden, Thales Cryogenics B.V.- J. Douwen, State-of-the art cryocooler solutions for HPGe detectors, www.canberra.com, Available on-line 2013.

[6] S. Harb, K. Salahel Din and A. abbady, Study of Efficiency Calibrations of HPGe Detectors for Radioactivity Measurements of Environmental Samples, Proc. of the 3rd Envi. Phys. Conf., Aswan, Egypt-207, 19-23 Feb. 2008.

[7] ORTEC Solid-State Photon Detector Operator's Manual, Revision B, USA, 2012.

[8] Genie™ 2000 Tutorials, 3.1 Basic Spectroscopy Software and Documentation, Canberra Industries, Inc, USA, 2006.

[9] IAEA, Measurement of Radionuclides in Food and the Environment, Technical Reports Series 295, 1989.

[10] ICRP, Radionuclide Transformation, International Commission on Radiological Protection, Oxford: Perganon Press, 1983.