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Development of a New Technique of 4πβ-γ Detection System to Perform Primary Methods for the Measurement of Activity of Radionuclide

Heranudin

National Research and Innovation Agency of Indonesia (BRIN)

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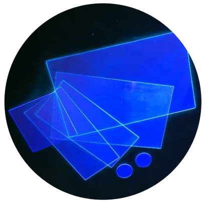
OUTLINES

- Introduction
- Objectives
- Methods
- Results
- Conclusion

INTRODUCTION

- There are a number of fields that require accurate measurement of radionuclides, including nuclear medicine, environmental monitoring, nuclear decommissioning, and nuclear forensics.
- The $4\pi\beta$ - γ coincidence counting method, utilizing proportional counter, pressurized proportional counter, or liquid scintillator for $4\pi\beta$ channel, has been established as an absolute method for radioactivity measurement for several decades.
- The use of plastic scintillators as a beta detector poses challenges such as unwanted gamma interference and the effect of after pulses.
- The offline analysis method has ability to extract and analyze valuable information from data that was previously missed in the conventional real time analysis.






OBJECTIVES

- To investigate effectiveness of the plastic scintillator as a β counter for the 4πβ-γ
- To devise an innovative offline analysis technique for the digital pulse processing system
- To obtain results with various parameters using only a single set of measurement data, saving both time and cost
- To perform absolute measurement of activity of radionuclides using the developed 4πβ-γ detection system

METHODS (Coincidence Counting Method)

- The coincidence counting method has proven to be a powerful technique for measuring radionuclide activity.
- To apply this technique, at least two independent detectors are required, each sensitive to one type of radiation. Additionally, at least one detector must have a 4π view of the source to meet the requirements of the method.
- In beta decay immediately followed by gamma emission, the coincidence counts are based on the disintegration of the same atom. We assume A is the source activity. In simple terms, the single channel count rates of β and γ are ρ_β and ρ_γ, respectively, while $\rho_{\beta\gamma}$ is the coincidence count rate. Whereas ε_β is the efficiency for detecting beta particles, and ε_γ is the efficiency for detecting gamma rays.

$$\rho_\beta = \varepsilon_\beta A \quad ,$$

$$\rho_\gamma = \varepsilon_\gamma A \quad , \text{and}$$

$$\rho_{\beta\gamma} = \varepsilon_\beta \varepsilon_\gamma A \quad .$$

→

$$A = \frac{\rho_\beta \rho_\gamma}{\rho_{\beta\gamma}} \quad ,$$

$$\varepsilon_\beta = \frac{\rho_{\beta\gamma}}{\rho_\gamma} \quad , \text{and}$$

$$\varepsilon_\gamma = \frac{\rho_{\beta\gamma}}{\rho_\beta} \quad .$$

METHODS (Correction and Efficiency Extrapolation)

- Using a formula to correct coincidence counting data for the effects of non-extendable deadtime and accidental coincidences.
- Utilizing a 4πβ detector, varying β efficiency and extrapolating it to unit value can derive an accurate value of the activity, without needing to know the precise values of the nuclear decay scheme parameters.

$$\rho_{\beta\gamma} = \frac{R_c - (r_\beta + r_\gamma)R_\beta R_\gamma}{(1 - R_\beta\tau_\beta)(1 - R_\gamma\tau_\gamma)\mathrm{X}(r_\beta, r_\gamma) + R_c\tau_m \mathrm{Y}}$$

$$\rho_\beta = A\left[1 - f_1\left[1 - \frac{\rho_{\beta\gamma}}{\rho_\gamma}\right]\right] \to A, \qquad as\ \frac{\rho_{\beta\gamma}}{\rho_\gamma} \to 1,$$

or equivalently

$$\frac{\rho_\beta\rho_\gamma}{\rho_{\beta\gamma}} = A\left[1 - f_2\left[\frac{1 - \frac{\rho_{\beta\gamma}}{\rho_\gamma}}{\frac{\rho_{\beta\gamma}}{\rho_\gamma}}\right]\right] \to A, \qquad as\ \frac{\rho_{\beta\gamma}}{\rho_\gamma} \to 1.$$

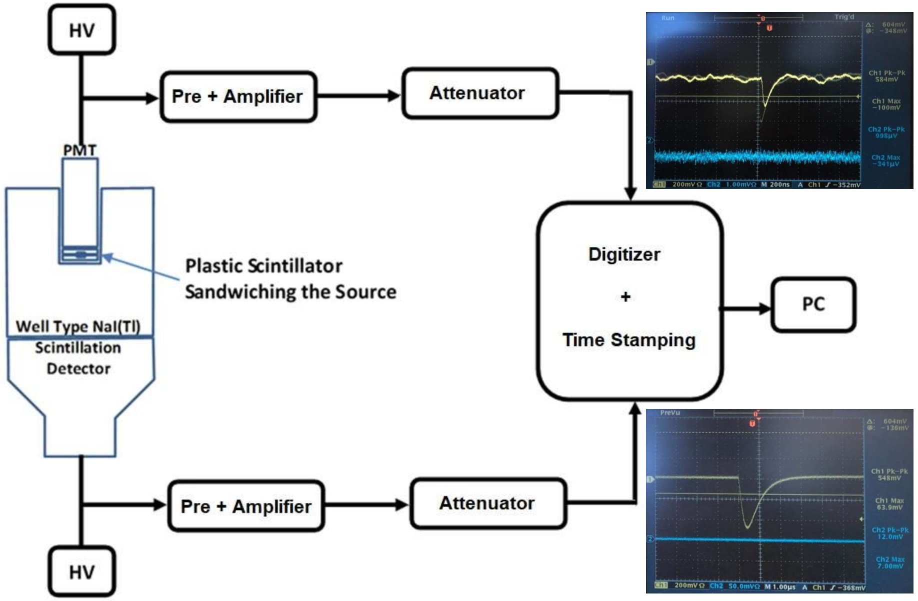
Practically, the functions of f_1 and f_2 are supposed to be polynomials in

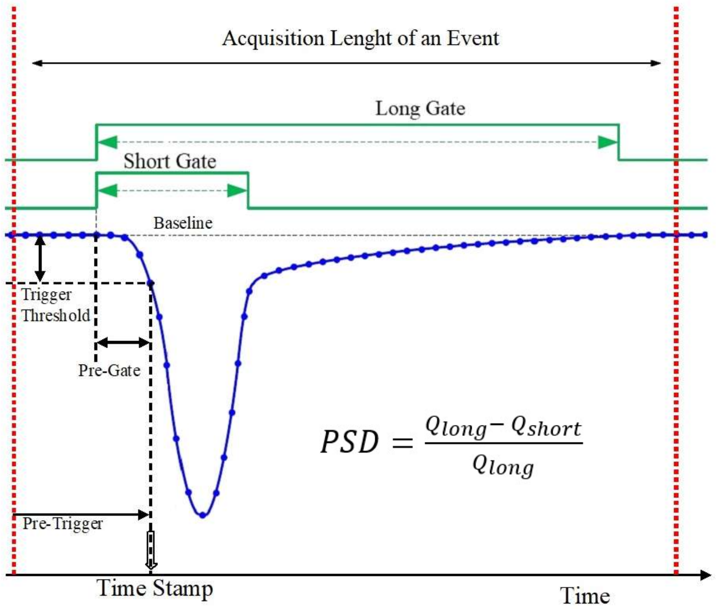
$$\left[1 - \frac{\rho_{\beta\gamma}}{\rho_\gamma}\right] \text{ or } \left[\frac{1 - \frac{\rho_{\beta\gamma}}{\rho_\gamma}}{\frac{\rho_{\beta\gamma}}{\rho_\gamma}}\right].$$

METHODS (Development of a 4πβ – γ)

- The gamma detector used a factory-made well-type NaI(TI) scintillator (dimensions of 127 mm in diameter and 127 mm in length with a 30 mm well diameter and a 75 mm well length), and the beta detector utilized plastic scintillators (EJ-212).
- The GEANT4 simulation was used to investigate the plastic scintillator that absorbs incoming electrons while minimizing photon interference.
- The simulation of betas and gamma rays with energy ranging from 50 keV to 2000 keV was conducted to inspect the electron detection probability and the unwanted gamma interference of the scintillator.
- The optical photon was also simulated to attain a design with optimal light collection from the plastic scintillator to the photomultiplier tube.
- A CAEN N6751 10-bit digitizer was used to acquire signals from both beta and gamma channel, employing Pulse Shape Discrimination (PSD) technique for signal identification.
- Several parameters such as duration of measurement, trigger threshold and gate times were set before data acquisition.

METHODS (Development of a 4πβ – γ)





- Upon exceeding the trigger threshold during the pre-gate time, the sample value is recorded as an event with a time stamp. Conversely, pulses with sample values below the threshold are disregarded as an event.
- The correlation of the charges in the two regions creates a ratio (PSD) that remains relatively constant for pulses possessing the same shape. Therefore, ensuring the correct charge parameter for each detector is crucial to obtain proper results.

METHODS (Development of a 4πβ – γ)

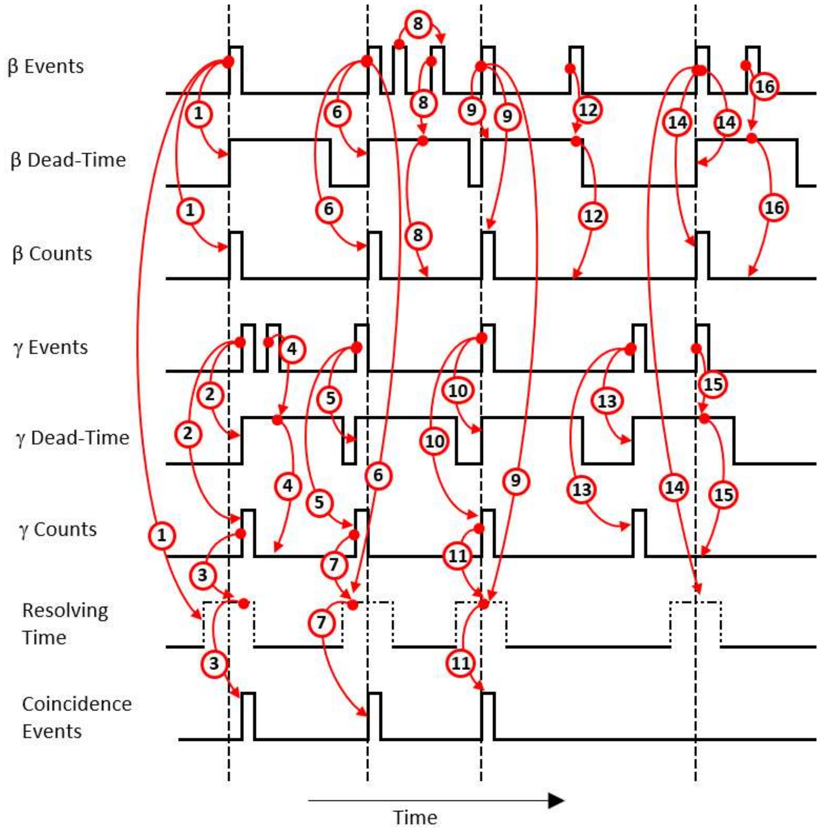
- Upon completion of the measurement, the data is saved as binary list-mode files (each 24 bytes), including the timestamp, pulse waveform, and gate charge. A highly effective means of data storage.
- Then, the recorded data can be analyzed without the need to continually collect new data, thereby eliminating signal distortion caused by temperature and environmental conditions.
- Python codes were developed to deconstruct the raw binary data.

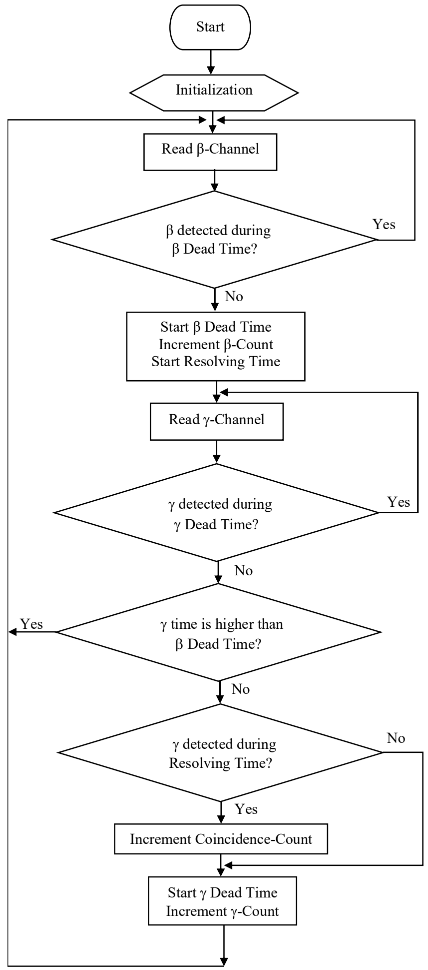
```
        … True:

    #Scan every single data of the measured gamma
    try:
        j0, j1, j2, j3, j4, j5, j6, j7, j8, j9, j10, j11 = fromfile(filej, dtype='<u2', count=12)
    except ValueError:
        j_result = False

        … j2 + (j3 << 16) + (j4 << 32) + (j5 << 48)      #Get time-stamp of a data signal of …
                                                          #Get the charge(amplitude) …
```

byte 24	N samples
byte 23	
byte 22	
byte 21	
byte 20	Flags
byte 19	
byte 18	
byte 17	
byte 16	Q Short
byte 15	
byte 14	Q Long
byte 13	
byte 12	Time stamp in pico seconds
byte 11	
byte 10	
byte 9	
byte 8	
byte 7	
byte 6	
byte 5	
byte 4	Channel ID
byte 3	
byte 2	Board ID
byte 1	

METHODS (Offline Analysis Method)

- Python program also was used to conduct offline analysis method.

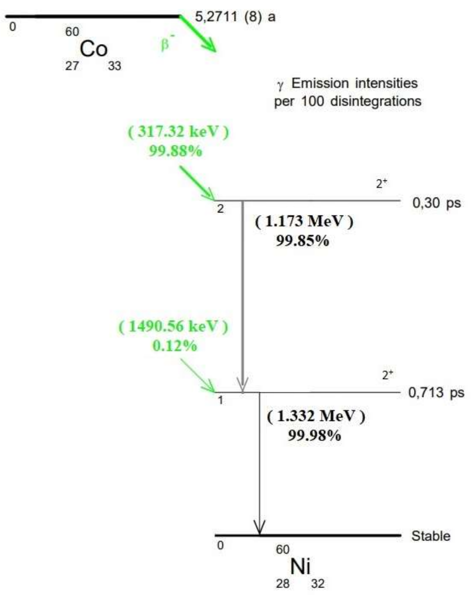


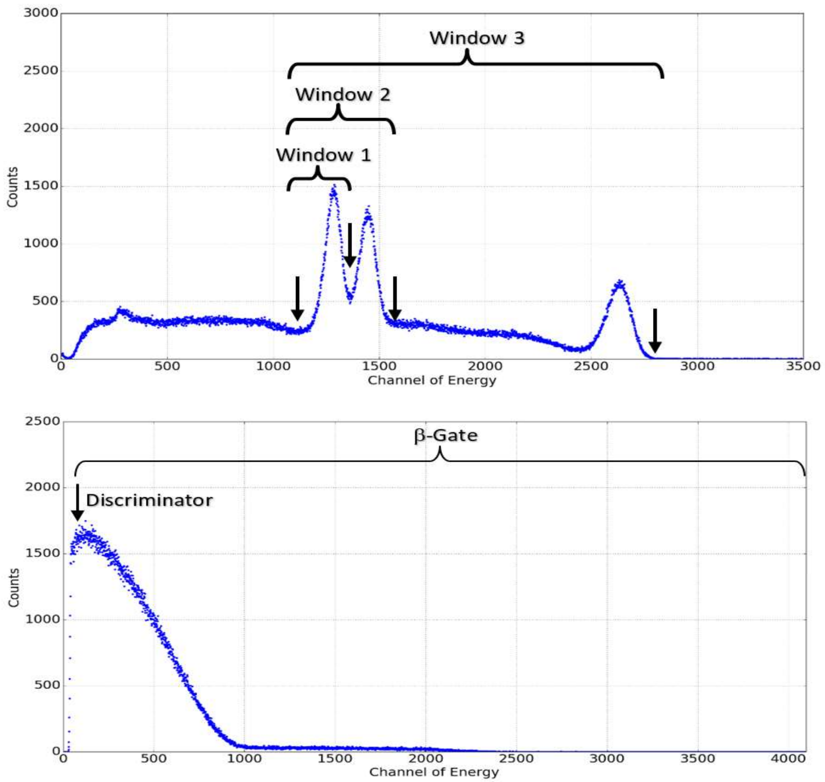


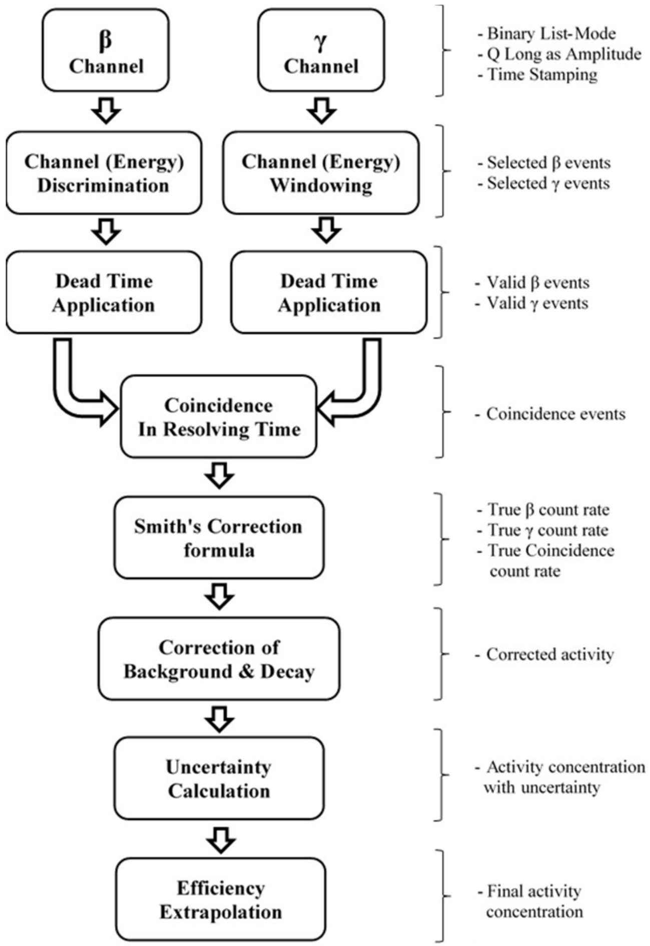

```
        == 0:
   gth=i_Tstamp

  valid signal of beta
 stamp > i_DT and (i_Qlong
 DT = i_Tstamp + (DT*1000)
i_RT = i_Tstamp
i_count = i_count + 1
j_result = True
#check the coincidence of beta-
#it means previously time stamp
if (j_OV_flag == True):
    if (i_DT >= j_Tstamp):
        if ((i_RT - (RT*1000))<=
            if j_Qlong > j_ld and
                coinc_count = coi
                j_result = False
        j_OV_flag = False

    else:
        j_result = False

#looping to find valid gamma even
while j_result is True:

    #Scan every single data of
    try:
        j0, j1, j2, j3, j4, j5,
    except ValueError:
        j_result = False

    j_Tstamp = j2 + (j3 << 1
    j_Qlong = j6
```

METHODS (Offline Analysis Method)

- Three γ-window settings for Cobalt-60 applied to the γ channel of the NaI(Tl) detector.

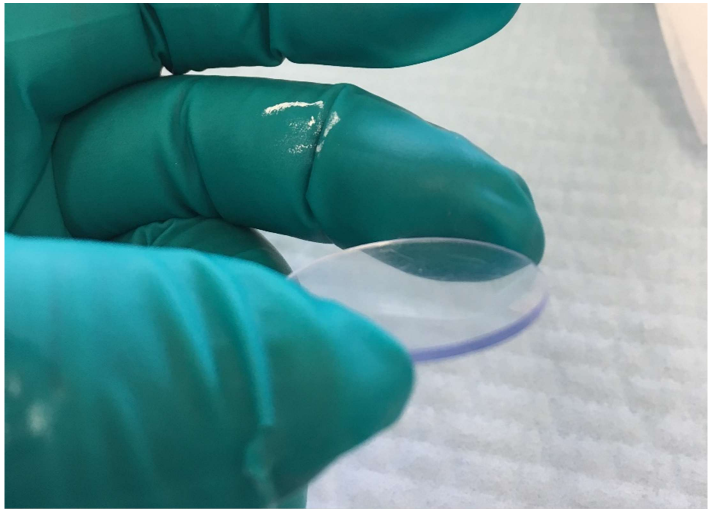


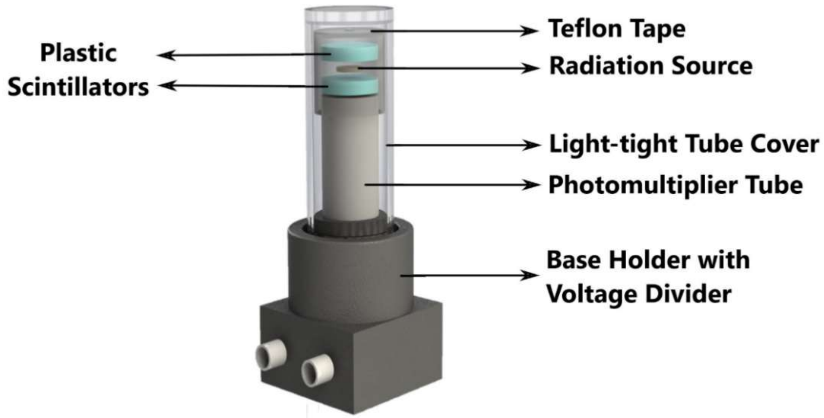




METHODS (Source Preparation)

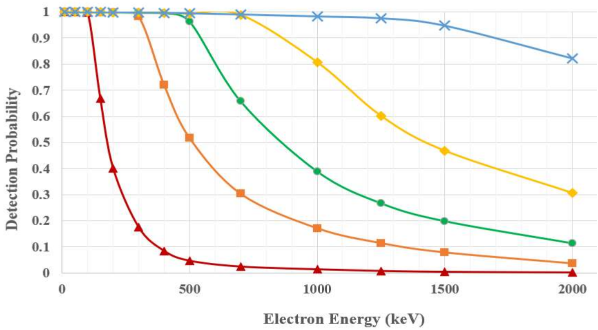
- A required amount (~0.5 mL) of ^{60}Co solution was drawn into the pycnometer and dispensed on the plastic scintillators. Once dried, the scintillator with the dropped source was glued to another blank plastic scintillator by BC-600 optical cement.
- The plastic scintillators, which was wrapped by Teflon reflector tape, were placed directly onto the PMT surface

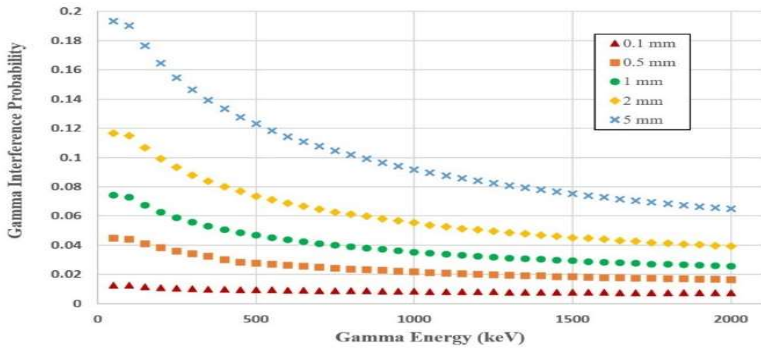


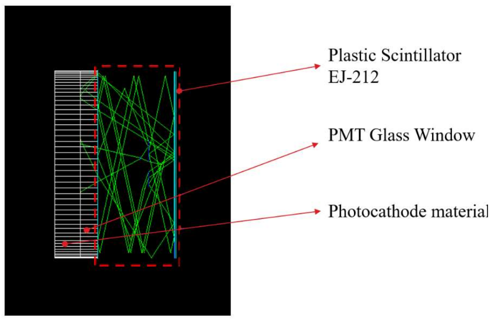


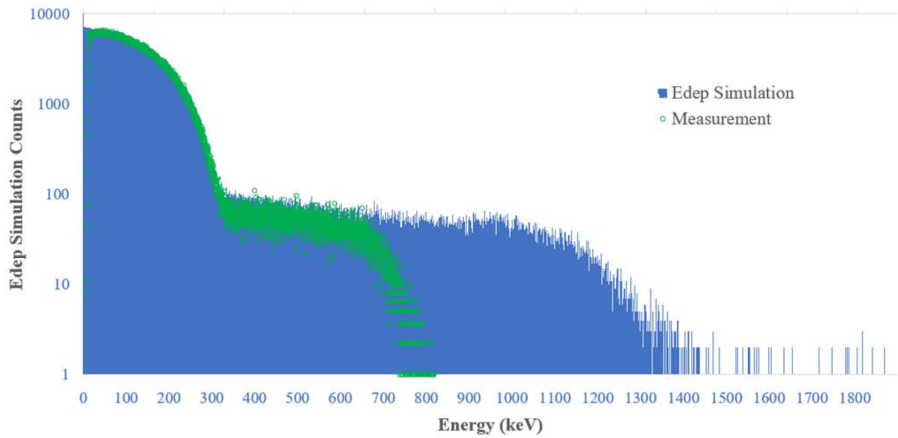
RESULTS (GEANT4)

- For the β detector, the simulated spectrum for the EJ-212 plastic scintillator corresponds closely with the measured spectrum of Cobalt-60.



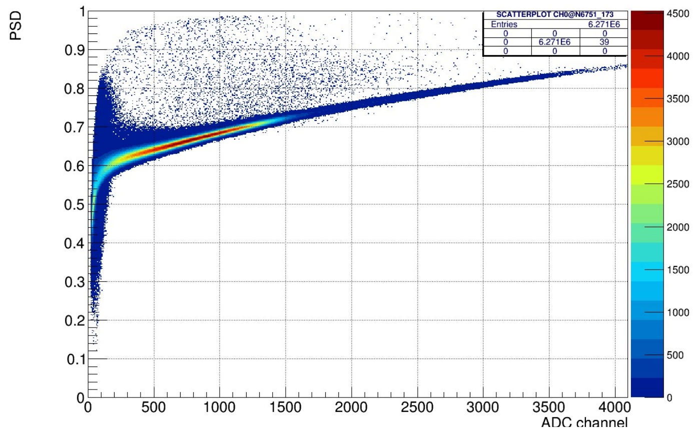


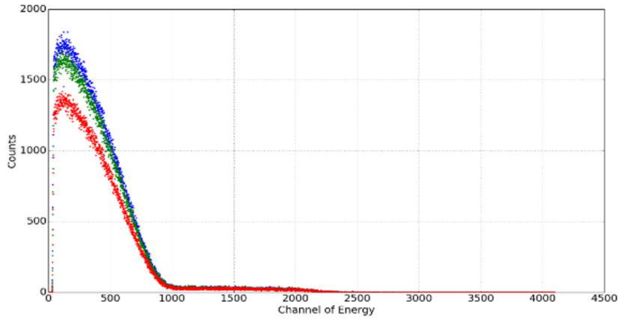


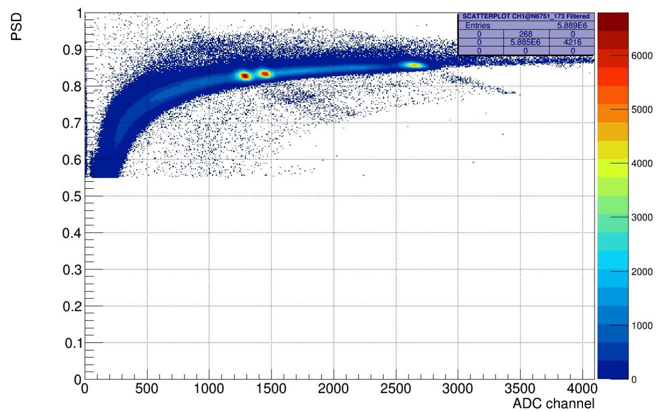


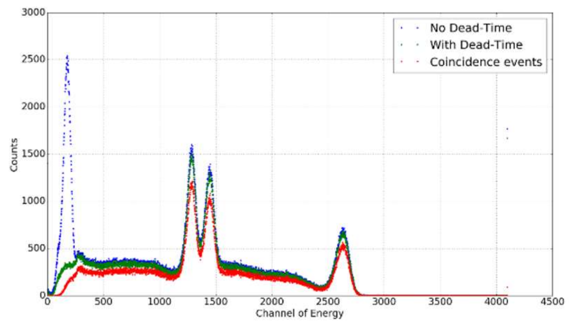
RESULTS

- The results presented a two-dimensional histogram of the β-γ channel for the ^{60}Co source. In the scatter plot, we can clearly see the area with high frequency corresponding to the main energy of beta (Left) and gamma (Right).



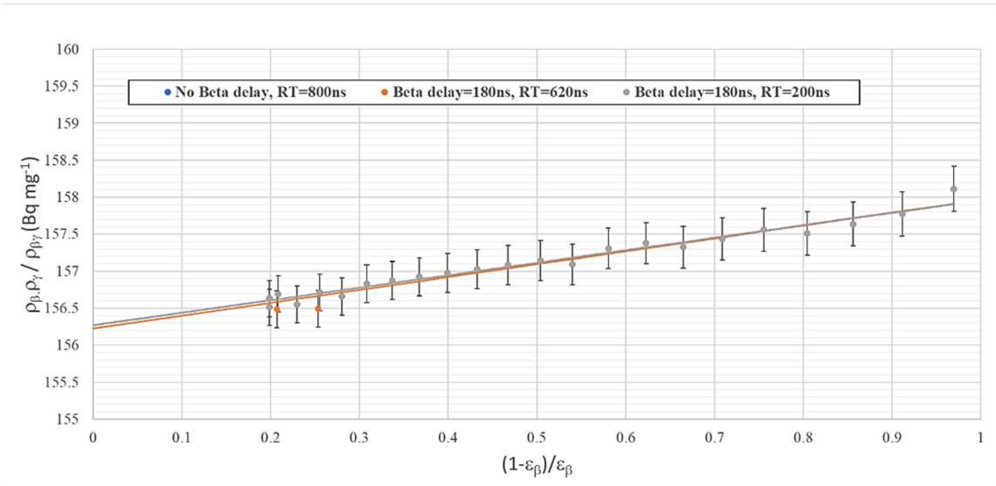


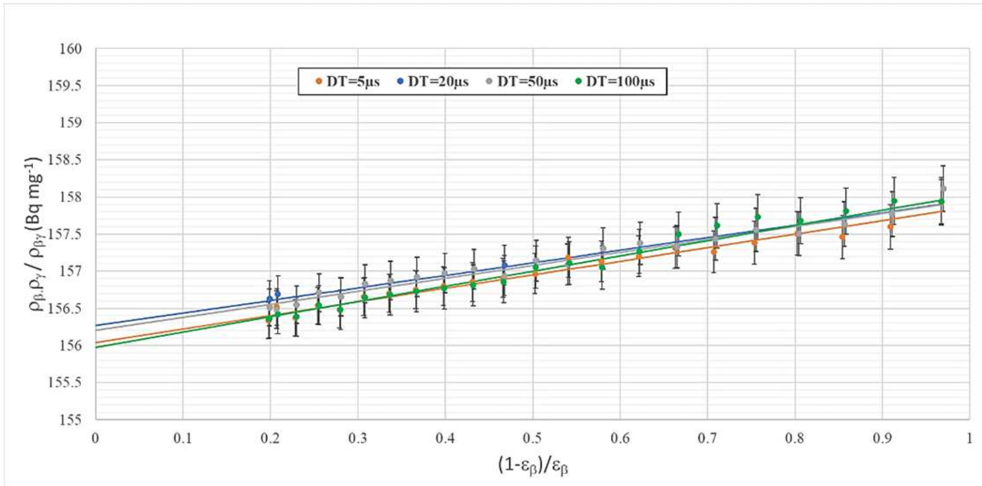




RESULTS

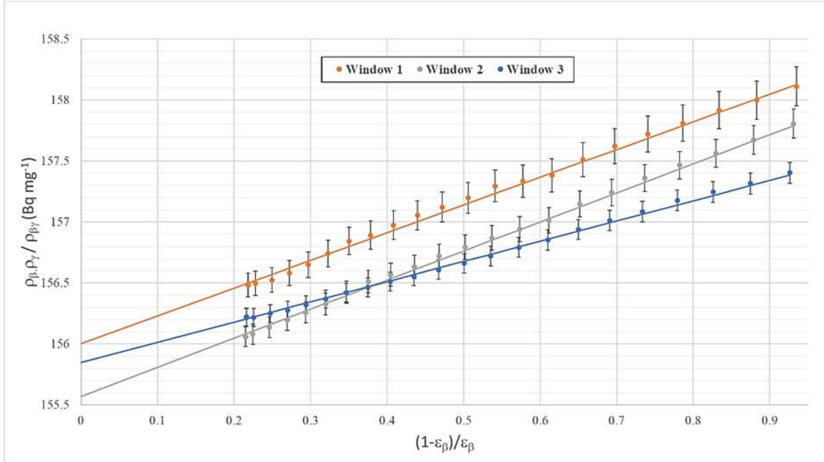
- The correction of background and decay were applied before the calculation of activity concentration. Efficiency extrapolation was performed to obtain the final value of activity concentration.
- Setting variation of beta delay and resolving time can be addressed by the offline analysis.
- Only one set of experimental measurement data was used to find sensitivity of the efficiency extrapolation using several different settings of dead time.

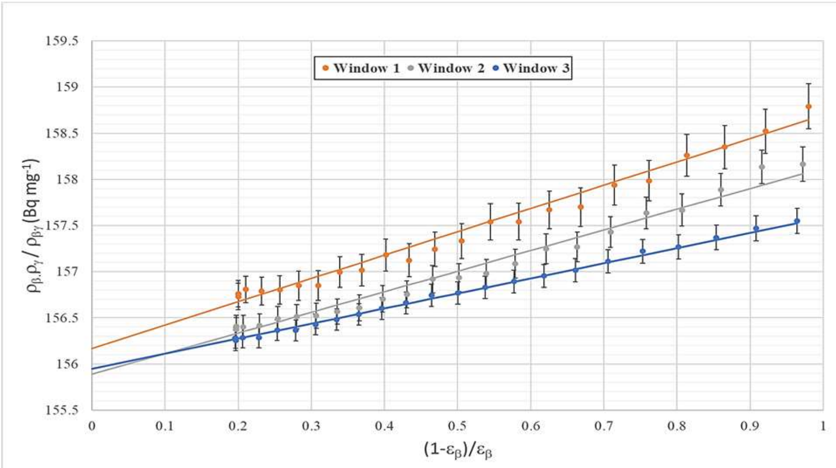


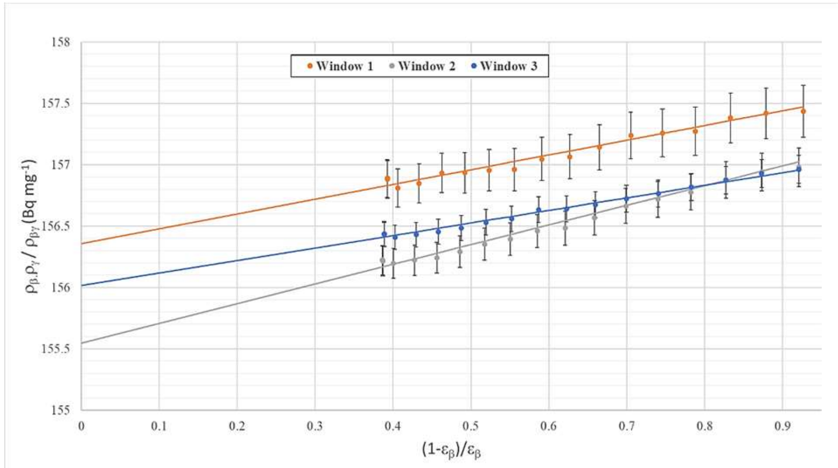


RESULTS

- The activity of three sample sources were calculated using three gamma windows. The correction of background and decay were applied before the calculation of activity concentration.
- Efficiency extrapolation using weighted fits was performed to obtain the final value of activity concentration with its uncertainty.

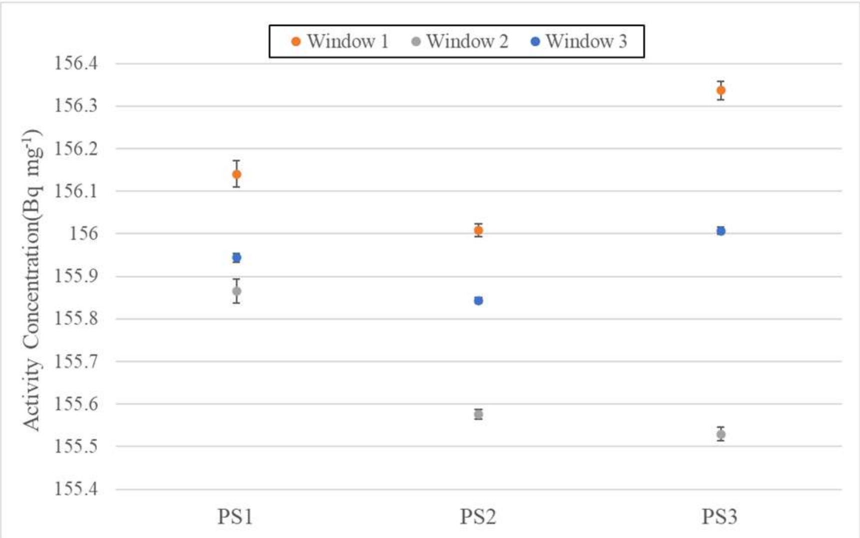






RESULTS

- The final result obtained was 155.9 ±0.5 Bq/mg agrees with the reference value of 155.4 ±0.4 Bq/mg to within one standard deviation.
- Several components of the measurements were taken into account to calculate the uncertainty budget.



Uncertainty component	Relative uncertainty (%)	Comment
Weighing and source preparation	0.15	Weighing uncertainty for the smallest source
Variation of resolving time and synchronisation	0.03	Variation of resolving time 200 - 800 ns
Variation of deadtime	0.19	Variation of deadtime 5 - 100 µs
Variation due to window setting	0.19	Maximum difference between different windows, rectangular distribution
Variation between sources	0.072	Maximum difference between different sources, rectangular distribution
Counting statistics	0.040	Standard deviation of the weighted mean for 3 sources and 3 windows each
Combined uncertainty	**0.320**	**Standard uncertainty (*k*=1)**

Conclusion

- The offline-analysis method for the coincidence counting with a 4πβ(Plastic Scintillator)−γ instrument was implemented successfully.
- Using this technique, accurate calculations of absolute radioactivity measurement can be performed since raw detector signals were first experimentally measured , without the need to continually collect new data, reducing measurement time and unwanted data fluctuations.
- The offline analysis provides a level of flexibility that is not possible with real-time data processing. Researchers can easily adjust and refine their analysis techniques based on their findings. This allows for a more iterative and exploratory approach to data analysis, which can lead to new insights and discoveries.

thank you