

Predicting Detection Limits and Accelerating Analytical Timelines using γ - γ Coincidence Spectrometry

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This work explores the use of gamma-gamma (γ - γ) coincidence spectrometry to reduce detection limits in high activity multi-radionuclide samples. The sensitivity of coincidence detection limits to contaminants at varying activities is investigated. The outcomes of this assessment can inform the aims of radiochemical separations. Finally, the benefits are presented of γ - γ coincidence in accelerating timelines by easing the requirements on radiochemistry.

Introduction

Many techniques exist that aim to improve detection limits for gamma spectrometry measurements. Two important techniques are:

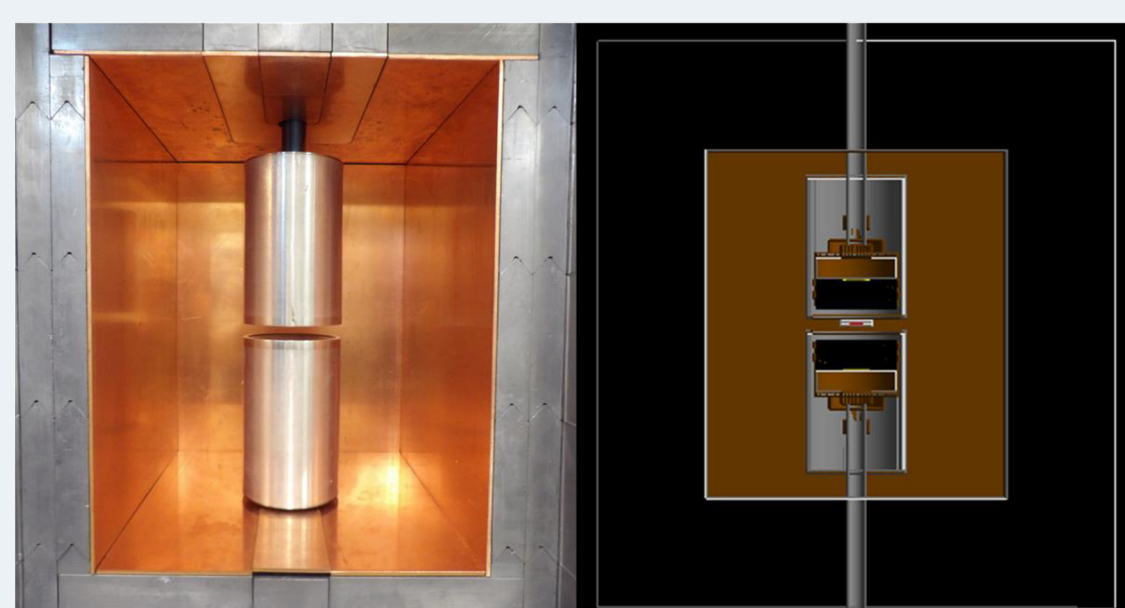
- Gamma-gamma (γ - γ) coincidence spectrometry
- Radiochemical separations

Separations are often performed with the aim of achieving elementally-pure samples, minimising the impurities that increase detection limits for the analytes of interest^[1].

The GEANT4 toolkit^[2] can be used to assess detection limit sensitivity in both coincidence and traditional “singles” gamma spectra to different impurities and activities. These ideas are expanded on by considering whether a combination of partial separations and γ - γ coincidence to accelerate timelines and reduce the requirements on radiochemistry.

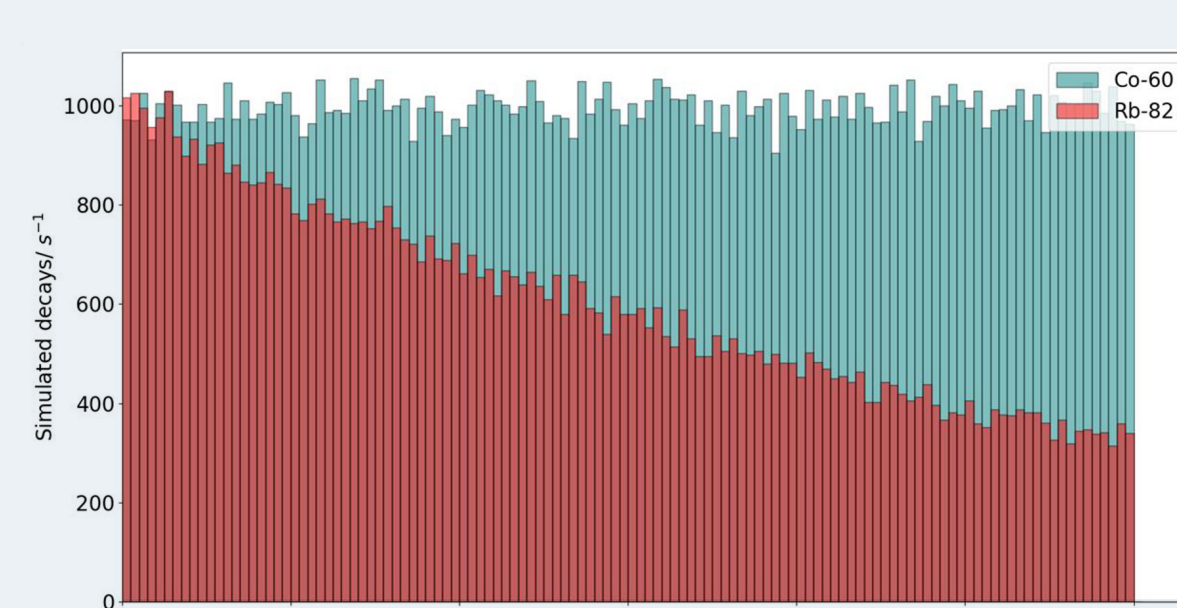
Methods

Simulation predicts energy captured in each detector

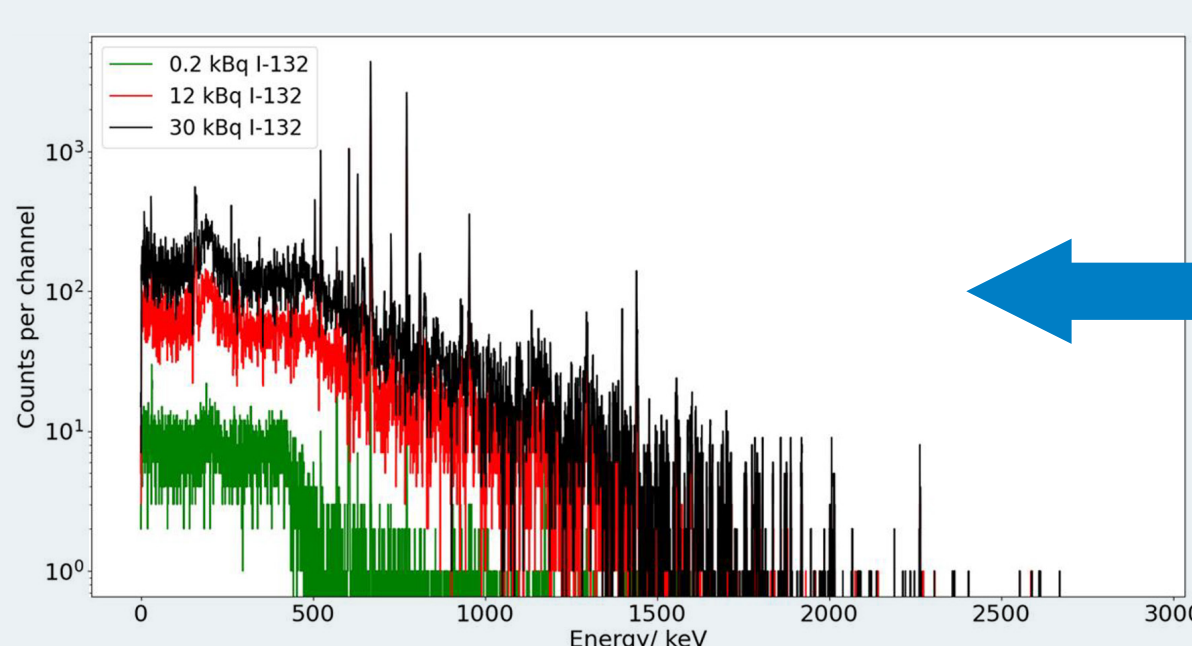


2 x BEGe5030 (carbon fibre window),
Mirion Technologies

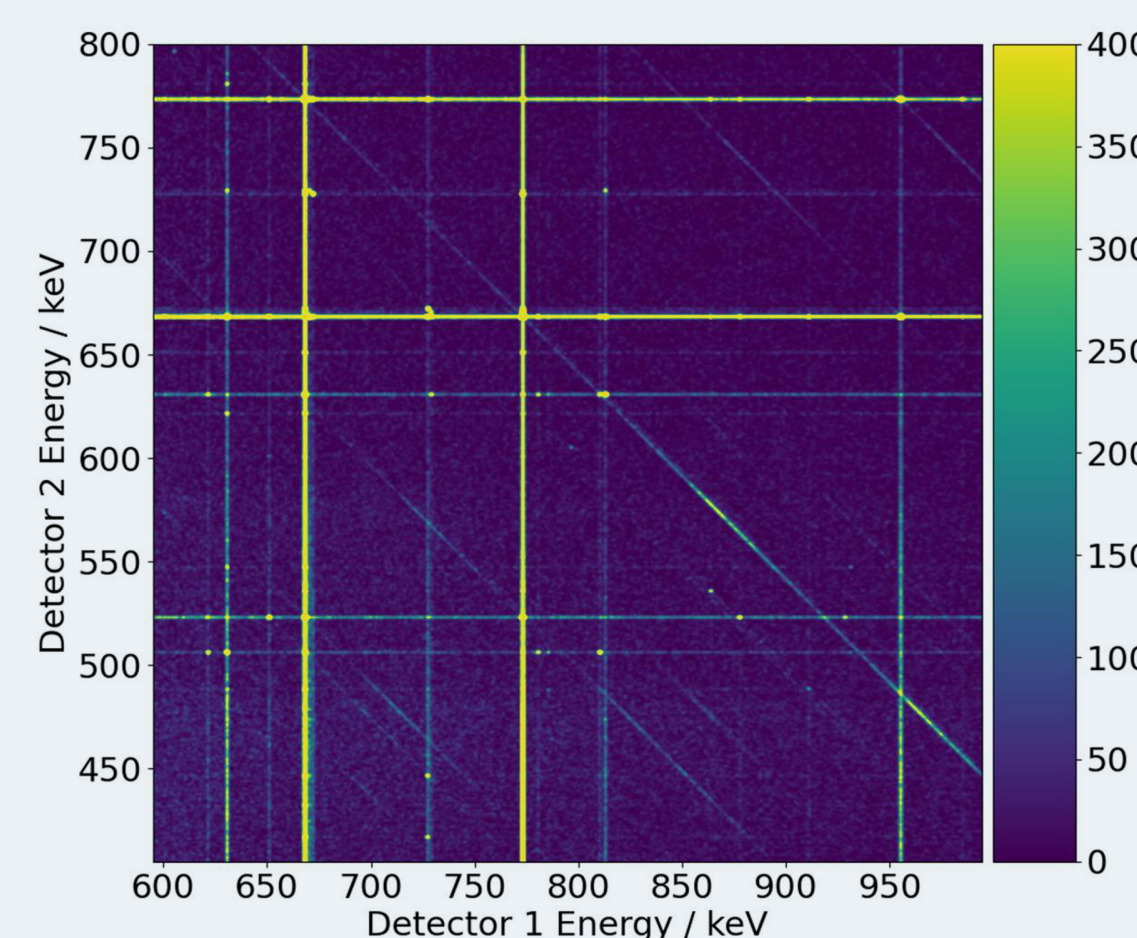
Decay times simulated generating T-List files



Summed energy-gated spectra outputted & analysed



Data sorted into a 2D coincidence matrix



Sensitivity study

2 Bq ^{134}Cs simulated with 0.2 – 100 kBq ^{137}Cs . Study then repeated using ^{132}I as the impurity. Spectra analysed for minimum detectable activity (MDA) of ^{134}Cs .

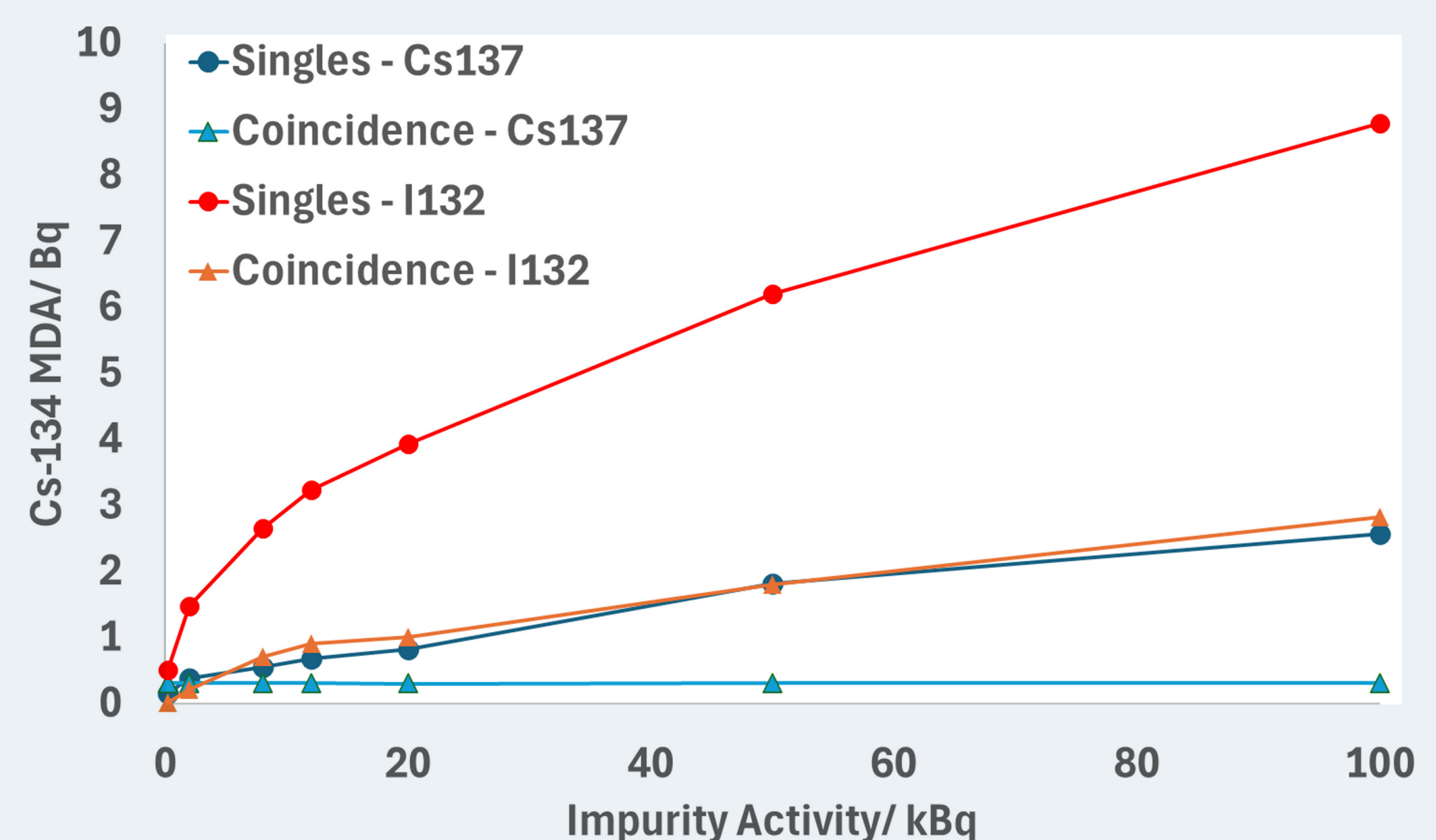
Timelines study

Key fission product lanthanides in quantities representative of approximately 2.3×10^{12} fissions (thermal neutrons on ^{235}U) simulated. 100% yield assumed for all radionuclides. Spectra analysed for MDA of ^{156}Eu .

All other measurement conditions remained constant.

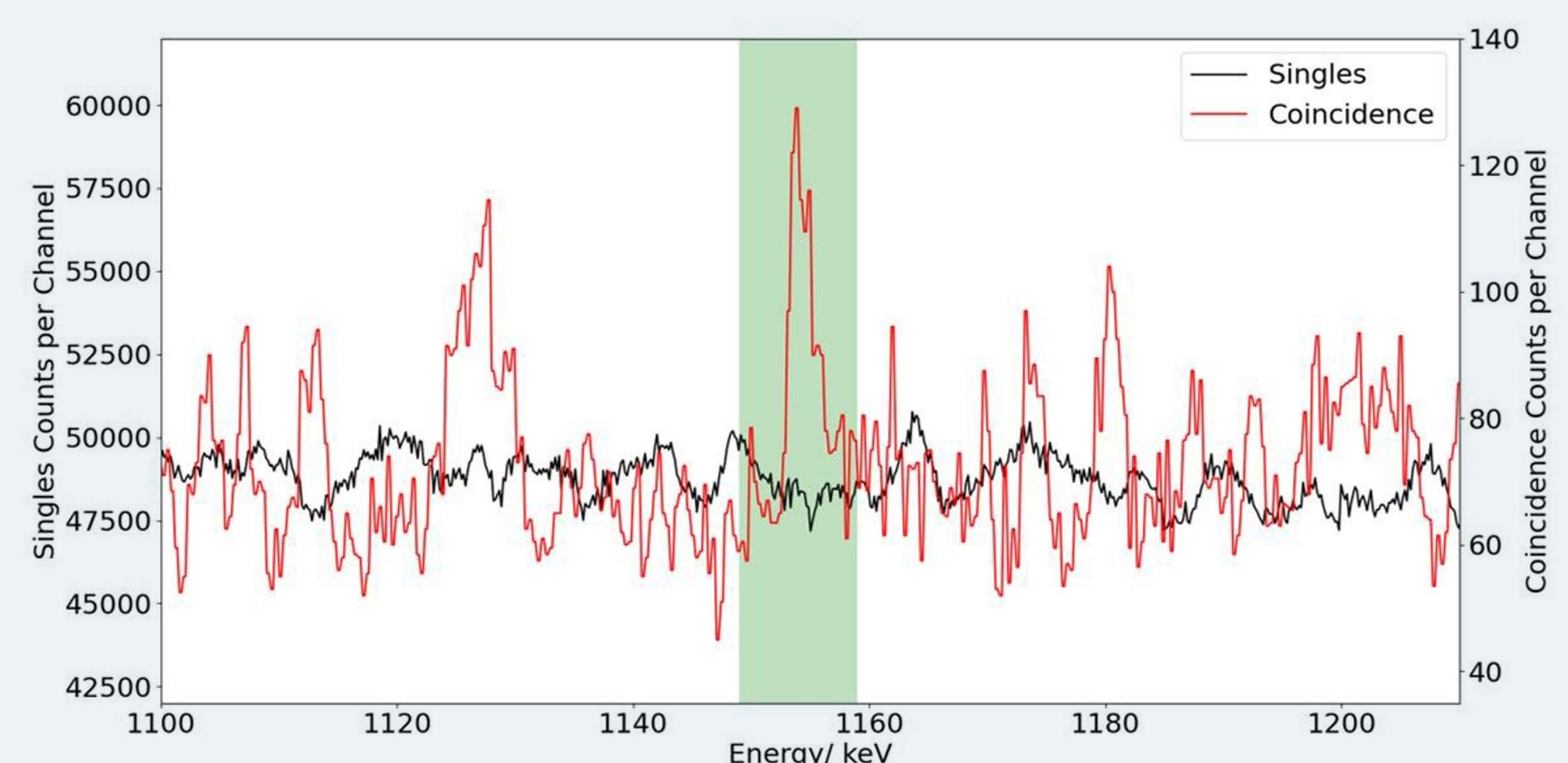
Results

Sensitivity Study



These simulations predict γ - γ coincidence spectrometry has increasing benefits on detection limits relative to singles measurements in samples of higher impurity activity. It is demonstrated that the MDA of ^{134}Cs MDA is significantly affected by the ^{132}I activity, whilst being insensitive to the activity of ^{137}Cs . This is due to ^{132}I emitting high energy gamma-rays in coincidence, causing scattering interactions that contribute to the coincident Compton background. The opposite is true for ^{137}Cs . This work represents the first assessment of the relative sensitivity gain of γ - γ coincidence across a range of contaminant activities.

Timelines Study



Analysis of a fission product lanthanide simulation, representing a partial separation, suggests ^{156}Eu (186 Bq) is likely detectable in a 12-hour count using γ - γ coincidence (MDA 112 Bq) but not singles (MDA 301 Bq). Whilst consideration should be given to the impact on measurement of other analytes and yielding techniques, implementation could accelerate quantification timelines by >12 hours.

Conclusion

A validated GEANT4 model of a high-resolution γ - γ coincidence spectrometer is reported and used to assess detection sensitivity with varying impurity radionuclide content. The potential to accelerate timelines through partial radiochemical separations is also proposed. These results provide semi-quantitative support to inform future analytical strategies involving radiochemical separations and radiometric detection.

References

1. Stokes, T. C., Goodwin, M. A., Jackson, M. J., & Boston, A. J. (2025). A modelling approach to defining radiochemical separation requirements for gamma spectrometry analysis. *Radiation Physics and Chemistry*, 113348.
2. Agostinelli, S & Geant4 Collaboration. (2003). Geant4—a simulation toolkit. *Nuclear instruments and methods in physics research section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 506(3), 250-303.