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An automated Monte-Carlo based method for the calculation of cascade summing factors

M.J. Jackson *, R. Britton, A.V. Davies, J.L. McLarty, M. Goodwin

AWE, Aldermaston, Reading, Berkshire RG7 4PR, UK

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ABSTRACT

A versatile method has been developed to calculate cascade summing factors for use in quantitative gamma-spectrometry analysis procedures. The proposed method is based solely on Evaluated Nuclear Structure Data File (ENSDF) nuclear data, an X-ray energy library, and accurate efficiency characterisations for single detector counting geometries. The algorithm, which accounts for γ–γ, γ–X, γ–511 and γ–e^- coincidences, can be applied to any design of gamma spectrometer and can be expanded to incorporate any number of nuclides. Efficiency characterisations can be derived from measured or mathematically modelled functions, and can accommodate both point and volumetric source types. The calculated results are shown to be consistent with an industry standard gamma-spectrometry software package. Additional benefits including calculation of cascade summing factors for all gamma and X-ray emissions, not just the major emission lines, are also highlighted.

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1. Introduction

When performing radiometric measurements via gamma-spectrometry it is often advantageous to count samples in close proximity to the detector in order to maximise the counting efficiency. This practice has the disadvantage of often being susceptible to complications due to the effects of true-coincidence summing. Such phenomena occur when multiple emissions are emitted in cascade and are measured within the resolving time of the detector system. The magnitude of these effects are entirely dependent on the structure of the radionuclide decay scheme and the detector-source geometry; they are inherently independent of the activity of the radionuclide.

The effect of true-coincidence summing is particularly important during quantitative measurements, such as those employed in Comprehensive Nuclear-Test-Ban Treaty (CTBT) verification [1], nuclear forensics and environmental monitoring. Such measurements are typically performed using large high-resolution HPGe detector systems, which often incorporate extensive, multi-layered, passive and active shields to reduce background radiation [2,3].

As part of the certified laboratory network, the GBL15 UK CTBT radionuclide laboratory (operated by AWE) has developed a generic and versatile method to calculate cascade summing factors for single detector gamma-ray spectrometry systems, based solely on the probability of decay cascade occurrences and detection efficiencies. This paper provides details of the applied methods together with a selection of validation results.

2. True-coincidence summing effects in gamma-spectrometry

In most radioactive decays, the parent nucleus decays via the emission of an alpha or beta particle, or via electron capture, to an excited energy level in the daughter nucleus. The excited daughter subsequently undergoes a transition to the ground state through the competing processes of either gamma-ray emission or internal conversion. The ejection of an electron during internal conversion leaves a vacancy in the an atomic electron shell, which when filled with an electron from a higher shell, results in the emission of X-rays or Auger electrons.

The lifetime of most excited states is typically of the order of picoseconds, far shorter than the time resolution of most gamma spectrometry detector systems (microseconds for HPGe). As a consequence, if two or more gamma-rays are emitted in cascade from a single decay, there is potential for γ–γ summing during spectrometry measurements. By extension, the same phenomenon is also true for γ–X, γ–511 and γ–e^- summation during internal conversion, electron capture and e^-–e^+ annihilation processes.

True-coincidence summation phenomena are generally categorised according to their effect on the measured spectrum. A summation resulting in a loss of counts in the photo-peak is known as “summing-out”. A summation resulting in a gain of counts in the photo-peak is known as “summing-in”. Using the β

* Corresponding author.
E-mail address: mark.j.jackson@awe.co.uk (M.J. Jackson).

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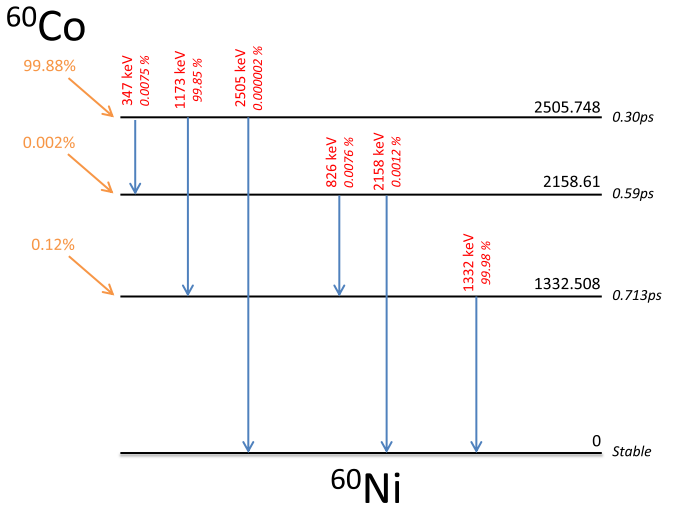

Fig. 1. Radioactive decay scheme for the β decay of ^{60}Co to ^{60}Ni. Gamma emission data given is shown in red. Branching ratio or level feed data is shown in orange. Data taken from [4]. (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this paper.)

decay of ^{60}Co to ^{60}Ni (Fig. 1) as an example, summation of the 1173 keV and 1332 keV gamma-ray emissions during a cascade results in a loss of counts in the two respective photo-peaks and gain of counts in a sum-peak at 2505 keV (1173 keV + 1332 keV); this is an example of *summing-out*. Similarly, summation of the less intense 347 keV and 826 keV gamma-ray emissions would produce a gain in the single 1173 keV photo-peak, and is hence an example of *summing-in*.

The degree to which true-coincidence summing affects gamma-spectrometry measurements is entirely dependent on the structure of the radionuclide decay scheme and the detector-source counting geometry. Complex decay schemes are more susceptible to true-coincidence summing effects and the energy level structure dictates which summations are allowed in a cascade. The solid angle between source and detector for a particular counting geometry has a large influence; close counting geometries will experience a high degree of true-coincidence summing compared to extended geometries. It is often preferable, however, to favour close counting geometries to maximise detection efficiencies [5]; in these situations it is important to fully account for true-coincidence summing effects when performing quantitative analysis. These corrections are performed using an additional term in the activity calculation defined in Eq. (1), where $C_{photopeak}$ is the net counts in the measured photo-peak, $\gamma_{\%}$ represents the gamma-ray abundance (the proportion of decays that emit the gamma radiation of interest), ϵ_{γ}^{peak} is the absolute full-energy peak efficiency, *LT* is the count live-time, and *CSF* is the cascade summing factor to correct for true-coincidence summing effects. The exponential term is used to correct the activity to a specific time, where λ is the decay constant for the given nuclide and t is the decay time between T_0 and the start of counting:

$$\text{Activity(Bq)} = \frac{C_{photopeak}}{\gamma_{\%}\epsilon_{\gamma}^{peak}\, LT\, CSF} e^{-\lambda t} \tag{1}$$

Traditionally, cascade summing factors are calculated through the measurement of a radionuclide sample at both close and extended geometries. A ratio of the two measured activities for a specific gamma emission line gives rise to a value of *CSF* suitable for use in Eq. (1). This process must be repeated for each gamma emission line and detector geometry, and as a consequence can be extremely time consuming for large laboratories.

More modern approaches use mathematical modelling to calculate these factors based on the decay scheme of a radionuclide. Such models are often incorporated in commercial software packages and greatly simplify the calculation process. While often acceptable for common radiometrology applications, these models have several limitations which can restrict more advanced applications. The following section highlights some of the limitations in these software packages and presents a newly developed method which mitigates against them.

3. Commercial software availability

Many radiometrology laboratories make use of commercially available software packages (e.g. Canberra ISOCS [6] and Canberra LabSOCS [7]) to mathematically calculate detection efficiencies as a function of energy for a given counting geometry. These approaches typically rely on an initial detector response characterisation, performed by the manufacturer, together with an explicit description of the counting geometry. This data is subsequently used in a Monte-Carlo model to estimate the peak and total efficiency of the detector as a function of energy. Such methods can provide significant time and cost savings compared to traditional experimental methods.

The geometry models used in these practices can be incorporated in to an automated spectral analysis code e.g. Canberra Genie2000™ [8] or Canberra APEX-Gamma [9], within which cascade summing factors are calculated for specific radionuclide emission lines. This allows the analyst to accurately quantify radionuclide activities for samples counted in close geometries. Such tools present a relatively simple method to account for true-coincidence summing effects compared to traditional, more experimental, approaches. However, the closed nature of the mathematical models and the commercial status of these products present some inherent limitations.

Since cascade summing factors depend on the decay scheme of the radionuclide in question, validated nuclear data evaluations are an essential component in their calculation. Decay data libraries featured in commercial software packages are usually limited to a pre-described collection of common radionuclides, which is often sufficient for most laboratory operations. However, should users wish to update this library with additional radionuclides or updated nuclear data, it typically requires a software update from the manufacturer, which may cause delays and have financial repercussions. A better solution would be to allow the user to add or update the evaluated nuclear data used in the calculations, although this functionality is rarely afforded to the user to protect commercial intellectual property.

The physics models adopted in these methods often have several limitations as a result of code simplifications. For example, the maximum number of gamma emissions per cascade or the maximum gamma emissions allowed in a summing-in event may be less than the maximum in the decay scheme. This simplification could result in the neglection of allowed cascades from cascade summing factor evaluations or the omission of complex coincidence signatures.

Most quantitative analysis methods preferentially adopt the most abundant gamma emission line(s) to maximise the observed signature. As a consequence, cascade summing factors are often only available for the most abundant gamma lines. When identifying and quantifying radionuclides, coherent agreement between multiple gamma emissions, including the minor emission lines, can often provide additional confidence in the final result. Since cascade summing factors are not always available for the less abundant lines, potentially valuable information has to be excluded from the analysis procedure.

The discussed limitations create an opportunity to develop a

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