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A thalium-doped sodium iodide well counter for radioactive tracer applications with naturally-abundant ^{40}K

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ABSTRACT

The use of a thallium-doped sodium-iodide well-type scintillation detector for the assay of the low-activity radioisotope ^{40}K, in open-source potassium chloride aqueous solutions, is described. The hazards, safety concerns and radiowaste generation associated with using open-source radioactive isotopes can present significant difficulties, the use of hot cells and escalated costs in radioanalytical laboratory research. A solution to this is the use of low-hazard alternatives that mimic the migration and dispersion characteristics of notable fission products (in this case ^{137}Cs). The use of NaI(Tl) as a detection medium for naturally-abundant levels of ^{40}K in a range of media is widespread, but the use of ^{40}K as a radioactive tracer has not been reported. The use of such low-activity sources is often complicated by the ability to detect them efficiently. In this paper a scintillator detector designed to detect the naturally-abundant ^{40}K present in potassium chloride in tracer applications is described. Examples of the use of potassium chloride as a tracer are given in the context of ion exchange and electrochemical migration studies, and comparisons in performance are drawn from literature with hyper pure germanium semiconductor detectors, which are more commonly utilised detectors in high-resolution counting applications.

1. Introduction

Anthropogenic radionuclides provide one of the most challenging hazards associated with remediating environments and industrial facilities in the nuclear industry. Of these radionuclides the fission product ^{137}Cs is a particularly difficult isotope to remove, as its chemical and physical properties make it highly mobile in aqueous environments and its radiological properties render it a significant danger to human health [1].

The nature of the hazards involved with handling ^{137}Cs extends to laboratory settings where preliminary remediation experiments and decommissioning techniques are developed. Extensive risk management and personal protective equipment (PPE) are required to protect against the hazard of open-source use of the isotope in the laboratory, due to the high-energy γ-ray emitted at 662 keV. Such PPE and risk management strategies are often time consuming, cumbersome and expensive to implement. The development of protocols requiring significant quantities of material contaminated with ^{137}Cs often requires the use of a hot cell and a remote handling equipment, escalating the cost of these procedures by several orders of magnitude. As a result suitable analogues that mimic the characteristics of ^{137}Cs on chemical, physical and radiological levels without the safety concerns offer significant advantages in terms of cost and ease of use for the development of radioanalytical methods. By removing ^{137}Cs experimental risk is controlled at the first stage of the Hierarchy of Hazard Control, Elimination [2], where the Hierarchy of Hazard Control is a health and safety philosophy used to reduce, manage and control experimental risk.

One such analogue is the naturally-occurring isotope of potassium, ^{40}K. Like caesium, potassium is a Group I element that exhibits very similar chemical and physical properties. ^{40}K can also be considered radiologically similar to ^{137}Cs as it boasts a single, prominent γ-emission in the same energy range as that of ^{137}Cs, i.e. 0.5–2 MeV, thus with similar, highly-penetrating characteristics albeit at significantly lower activity. Indeed, unlike ^{137}Cs its specific activity is so low that it can be considered non-radioactive, and therefore safe, despite being radiometrically detectable [3]. These features render ^{40}K a suitable safe alternative to ^{137}Cs in remediation method development.

A paradox in the use of ^{40}K as an analogue is that its low specific activity can make it difficult to detect at naturally occurring concentrations with sufficient efficiency to provide easy measurements particularly in high background environments, given that ^{40}K is a main constituent of natural background radiation. The successful use of ^{40}K as a radio-analogue of open-source ^{137}Cs is critically dependent on the following specific spectrometry considerations:

- extended counting times,
- background attenuation, and
- the use of large source samples.

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While background attenuation cannot be considered in the detector design *per se*, steps can be taken to optimise it to reduce source sample sizes and counting times.

In this paper, we report on the use of a thallium-doped sodium-iodide, NaI(Tl,) well-type scintillation detector, based on optimal design choices to improve the detector efficiency to count low-activity sources. The primary purpose of the detector is the radiometric study of ^{40}K in aqueous potassium chloride solutions as an analogue for ^{137}Cs in open-source developmental experiments. Such a device may be used in the future for *in situ* radiometric measurements of radionuclides of concern in the cleanup of nuclear sites.

2. Low activity counting and NaI(Tl) scintillators

Various detection technologies have been developed for low-activity counting applications, most often doped metal halide scintillators such as sodium iodide and caesium iodide, whilst the efficiency of these is amongst the best for γ-ray counting, the energy resolution can limit the use of these compounds where a fine degree of discrimination is needed between lines with very similar energies. Where high-resolution γ-ray spectrometry is desired such as for the case of ^{40}K where contamination can obscure the 1460.8 keV line, the *de facto* detector of choice for low-activity counting is typically the hyper pure germanium (HPGe) semiconductor. Numerous examples exist in the relevant literature where both standard geometry and well-type HPGe detectors have been utilised to measure low-activity samples containing ^{40}K, with minimal detectable activities (MDA) in the range of Bq/m^3. For the sake of brevity only a small number of instances will be given here; the instances include determining potassium content in food produce [3,4], calculating the activity of naturally occurring radioactive material (NORM) in building material, of which ^{40}K is a constituent [5,6], and studying the levels of ^{137}Cs in soil and grain samples [7]. Significant drawbacks exist to the use of HPGe semiconductors, they are expensive to purchase and maintain given that they require cooling to 77 K, usually requiring the use of liquid nitrogen (LN2) dewars [8]. The use of LN2 is often restricted in many industrial environments due to concerns over potential spillage which can be hazardous. Mechanically-cooled HPGe detectors are available which provide a means around LN2 restrictions, but again they can be considerably more expensive [9].

Given the issues surrounding the use of HPGe detectors, well-type NaI(Tl) detectors have found use as a means of counting low-activity samples where the use of HPGe is prohibited and in some instances demonstrating a lower MDA [10,11], thereby competing with HPGe detectors on performance as envisaged by Theodorsson [12]. Other scintillation materials exist that, perhaps, offer better physical characteristics in the detection of low-activity, these include caesium iodide (CsI), bismuth germinate (BGO) and lutetium oxyorthosilicate (Lu_2SiO_5) [8]. The higher atomic density of thallium-doped caesium iodide CsI(Tl), for example, provides a higher degree of stopping power for energetic γ-rays than NaI(Tl), like the 1460.8 keV emission from ^{40}K [13], and is a competing choice of material. However, the presence of trace amounts (upwards of 60 mBq/kg) of the fission product ^{137}Cs within the CsI(Tl) crystals provides a significant hindrance to its use as a means of low-activity counting [14,15]. A variety of exotic, but increasingly common, scintillator materials are becoming affordable; however in terms of cost, performance, and ease of manufacture NaI(Tl) constitutes perhaps the best compromise.

Given the poor energy resolution of NaI(Tl) relative to alternatives such as HPGe, well detectors based on this are perhaps best used for single-isotope detection where the isotope is usually known beforehand. The detection of ^{225}Ac using a 20×20 cm^2 NaI (Tl) crystal with a 13 cm deep 2.5 cm diameter well by Pommé et al. is one such example of NaI(Tl) low counting [16]. In this work ^{225}Ac samples were counted to measure the half-life of the isotope using six different counting methods. The results obtained from the NaI(Tl) well detector were consistent with those from other methods, namely a HPGe detector, a CsI sandwich spectrometer amongst others, thereby validating NaI(Tl) well counters as a low-activity counting technique against higher-resolution alternatives.

The efficiency of the NaI(Tl) well detectors can be further improved with the addition of a counting 'lid' to the detector, commonly found in Positron Emission Tomography (PET). Utilising a second detector inserted into or over the detector well provides almost 4π counting geometries. The use of two sodium iodide scintillators, as described by Rieppo, provides for a 4π geometry, though no substantial increase in efficiency is noted over a well-type crystal [17]. The major complicating factor with these detector arrangements is often the extra electronics and additional analysis of the two spectra that is required [8,17].

The relative ease with which large NaI crystals are manufactured has also led to arrays of large NaI(Tl) detectors being used in whole-body counters [18]. Whole-body counters are used throughout the nuclear industry as a means of checking personal exposure to radiation; they have also been used as a means of determining the bodily content of ^{40}K in humans and other animals in nuclear medicine studies [18–20]. In these studies, ^{40}K is naturally occurring within the tissues of the examples being counted. This is in contrast to the intended use of potassium in this work where ^{40}K is specifically introduced in the form of potassium chloride (KCl).

NaI(Tl) well detectors are perhaps most commonly utilised in the field of nuclear medicine, where they are utilised as a means of validating radiation levels in PET and Single-Photon Emission Computed Tomography (SPECT) analysis [21]. However, the isotopes used in this analysis are typically low volumes of short half-life isotopes, ^{99m}Tc being the most common example, with comparatively high activities upward of 30 MBq [21].The NaI(Tl) well detectors used in nuclear medicine are often built into automated multi-sample detection rigs; examples include the Wallac Wizard and Packard Cobra II systems which are unsuitable for our purposes [22].

Whilst the use of NaI(Tl) detectors in well counter systems is widespread for the assay of contamination in a variety of environments and applications, there have been few if any reports of their use specifically for the detection of ^{40}K, specifically where potassium has been introduced as a radioactive tracer. In addition to its use in whole-body counters, there are numerous instances where sodium iodide has been used to detect inherent levels of ^{40}K [23,24], but its use for the assay of quantities of introduced ^{40}K tracer has not been described, often it appears because of the very low activity of ^{40}K coupled with their often being no restriction on the use of potassium i.e. due to its chemical significance in a given tracer application *per se*: thus there are usually higher activity alternatives that need less counting time. The only evidence in the literature of a potassium isotope being utilised specifically as a radioactive tracer is the use of ^{38}K for medical imaging by Takami et al. Necessarily, ^{38}K is radiologically very different from its ^{40}K cousin in that its half-life is only 7.5 min and has a far higher specific activity [25]; it is these radiological properties that cause it to be selected for this application as opposed to the choice of element. In this paper a alternative scenario is reported where the choice of element is crucial in replicating the chemical properties of a given adsorbent, whilst the natural radioactive properties present in the natural isotopic abundance of that element renders it traceable on a radiological basis.

3. Detector description and experimental setup

The NaI(Tl) well detector, manufactured by Scionix, Bunnik, the Netherlands, used for the measurements performed in this work