



HNPS Advances in Nuclear Physics

Vol 14 (2005)

HNPS2005



To cite this article:

Stamoulis, K. C., Ioannides, K. G., Karamanis, D. T., & Patiris, D. C. (2019). A rapid method for screening 90Sr activity in water and milk samples using Cherenkov radiation. *HNPS Advances in Nuclear Physics*, *14*, 113–118. https://doi.org/10.12681/hnps.2258

A rapid method for screening ⁹⁰Sr activity in water and milk samples using Cherenkov radiation

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Abstract

Strontium-90 is one of the most dangerous fission products that enter the food chain. Because of its similar chemistry to calcium, it is deposited to bones posing a high risk for human health. ⁹⁰Sr is a pure β -emitter and decays to ⁹⁰Y also a β -emitter. The fast moving electrons emitted by the ⁹⁰Y decay, produce Cherenkov radiation, which can be detected by a Liquid Scintillation Counter (LSC).

Water and milk samples spiked with various concentrations of 90 Sr/ 90 Y in equilibrium were measured in a LSC (Tricarb 3170 TR/SL). The derived efficiencies were 50% for water samples and 13 % for milk samples. The minimum detectable activities (MDA) for measuring 90 Sr in water and milk samples, without any pretreatment were calculated 240 mBq/L and 1 Bq/L respectively. The intervention levels for 90 Sr contamination in water and milk samples, established by the European Union are 125 Bq/L and 75-125 Bq/L depending on age, respectively.

The interference of 40 K, which is present in milk due to its natural potassium content, was also investigated.

1 Introduction

The radioisotope 90 Sr is a fission product which enters the environment from nuclear reactors wastes. Due to its chemistry similar to calcium's, it follows the paths of this element in the food chain and enters the human body posing a high risk to human health. In the case of a nuclear accident the concentration of 90 Sr

may be raised orders of magnitude. Then the contamination of food must be routinely monitored.

Radio-strontium decays to its daughter nucleus 90 Y, emitting beta particles and 90 Y also decays in beta mode emitting beta particles with $E_{max} = 2280$ keV and half-life of 64 hours. Because of its short half-life, it is in equilibrium with its parent nucleus 90 Sr. Thus measuring 90 Y activity in a sample, the 90 Sr activity of the sample can be estimated. Fast moving electrons that are emitted by the 90 Y decay, produce Cherenkov radiation in a liquid medium like milk or water. The energy threshold of the fast moving electrons to produce Cherenkov radiation is 256 keV [1]. The radiation produced lies in the optical and UV parts of the electromagnetic spectrum, so it can be detected by the photomultipliers of a LSC.

Milk contains natural potassium, which contributes with its radioisotope 40 K to the natural radioactivity of milk. One gram of natural potassium has radioactivity equal to 30 Bq. Thus milk containing about 1200 mg/L of potassium can have activity due to 40 K equal to 36 Bq/L. Because of the fact that 40 K decays producing also fast moving electrons ($E_{max} = 1312$ keV), it also contributes to the Cherenkov effect. This contribution can be estimated by assessing the efficiencies of measuring 90 Sr and 40 K in milk samples with a LSC.

Strontium-90 in milk can be measured either by using a gas proportional counter or a LSC [2,3]. The samples must be radiochemically pretreated in order to isolate 90 Sr or its daughter 90 Y. Some investigators have used Cherenkov counting to measure 90 Sr/ 90 Y in water samples [1,4–6].

The intervention levels established by the European Union for 90 Sr contamination are 125 Bq/L for water and 75-125 Bq/L for milk, depending on age. A screening level in the order of 1 Bq/L in milk and less for water samples is considered sufficient for a rapid method that does not require any pretreatment of the sample.

In the present work, the possibility of rapid screening water and milk samples for ${}^{90}\text{Sr}/{}^{90}\text{Y}$ contamination was investigated. Cur-

rent methods for assessing the 90 Sr activity in milk are laborious and time-consuming (at least 2 days for sample preparation). For this purpose, the efficiency for measuring 90 Sr/ 90 Y activity in water and milk samples by detecting Cherenkov radiation in a LSC was calculated. The efficiency for measuring 40 K in water and milk samples was also estimated.

2 Materials and methods

In order to assess the efficiencies for measuring 90 Y and 40 K activities in water and milk, plastic vials of 20 mL capacity were used. Milk and water of various volumes were added to the vials with known activities of 90 Sr/ 90 Y or 40 K. All vials were measured with a Tricarb 3170TR/SL LSC for Cherenkov radiation, at the Radiochronology Center of the University of Ioannina. The background for Cherenkov radiation was found 2 cpm for deionised water and 1.5 cpm for water mixed with acrylic paint. The acrylic paint was used in order to simulate the scattering and absorption effects of the milk matrix. Typical counting time was 1000 min in order to obtain satisfactory statistics.

The investigation was divided in two parts. Firstly, in order to estimate the efficiency of the LSC in detecting Cherenkov radiation produced by ${}^{90}\text{Sr}/{}^{90}\text{Y}$ in water samples, a series of vials with deionised water and known activities of ${}^{90}\text{Sr}/{}^{90}\text{Y}$ solution ranging from 0.065-13.1 Bq were prepared.

Due to the fact that milk is not a transparent medium, part of the Cherenkov radiation is absorbed within the milk. In liquid scintillation counting, this phenomenon is known as the color quenching effect. Modern LS Counters can automatically estimate this quenching effect for each sample by calculating a quenching index, the spectral index of sample (SIS). This quenching factor decreases as the sample gets more quenched and increases as the sample is less quenched. Thus a sample of milk gives lower values for SIS factor compared to the SIS factors calculated for water samples. The quenching effect decreases the efficiency of the LSC. This influence to the efficiency of the counter was determined by preparing another set of vials containing milk and known activities of 90 Sr/ 90 Y solution, also ranging from 0.065-13.1 Bq with total volume of 20 mL.

As mentioned previously, milk contains considerable quantities of natural potassium and thus elevated natural radioactivity due to 40 K. In order to estimate the contribution of 40 K decay to the total Cherenkov radiation, the efficiency of LSC for measuring 40 K in water was investigated. A series of vials containing water and known activities of 40 K ranging from 0.75 to 2.6 Bq was measured for Cherenkov radiation. Finally, a series of vials with 20 mL of milk and known activities of 40 K, ranging from 0.75 to 2.8 Bq, were prepared and measured in order to estimate the efficiency of the LSC for 40 K Cherenkov counting in milk.

3 Results and discussion

The linear regression of the 90 Sr/ 90 Y count rate with the activity added to the vials containing deionised water resulted the estimation of an efficiency of 50.8%. This is in agreement to other investigators. Vaca et al. found a 47.5% efficiency for Cherenkov counting, using a Quantulus LSC [4]. Also Rao et al. reported a value of 50.0% efficiency in measuring ⁹⁰Y produced Cherenkov radiation with a LSC [1]. Minimum detectable activity (MDA) for measuring 90 Sr/ 90 Y in water samples was calculated 0.240 Bq/L. The linear regression of 90 Sr/ 90 Y count rate vs activity added in milk resulted to an estimated efficiency that falls to 13.6% for milk samples. The about fourfold decrease of the efficiency may be attributed to color quench. The fact that SIS values for water samples varied from 21 to 31 and the respective SIS values for milk samples varied from 11 to 18 can also support this explanation. The corresponding MDA calculated for milk samples, was 1.2 Bq/L.

The contribution of 40 K to the Cherenkov effect was estimated by assessing the efficiency of measuring 40 K activity in water and milk samples. The efficiency was derived by linear regression of the data and its value was calculated equal to 32.2% for water samples and 5 % for milk samples. It is figured out that a sixfold drop of the efficiency for ⁴⁰K measurements is observed from water to milk samples and this can also be attributed to the color quench. This is more pronounced in the case of ⁴⁰K, compared to ⁹⁰Y. This could be due to the difference of beta energy maxima. The efficiency determined for ⁴⁰K Cherenkov counting in water is in very good agreement with the values 33% and 30% that are presented by Rao et al. [1], for conventional and ultra low background LSC respectively. Taking account these efficiency values, the MDA for ⁴⁰K Cherenkov counting calculated was 0.39 Bq/L for water samples and 2.7 Bq/L for milk samples.

From the discussion above, it is obvious that measuring a milk sample that is contaminated with 90 Sr at the level of 1 Bq/L, count rates detected by the LSC in Cherenkov counting mode, should be attributed mostly to the ⁴⁰K content of the milk. Measurements performed in pure milk samples revealed that count rates exceeded water-acrylic paint background of about 2.2 cpm. If we assume that milk contains natural potassium at about 1200 mg/L, the ⁴⁰K activity of milk is 36 Bq/L and with an efficiency 5% of Cherenkov detection the expected count rate due to 40 K for a 20 mL milk sample, is about 2.16 cpm. If the count rate due to ⁴⁰K of milk is subtracted, the remaining count rate, about 0.04 cpm could be attributed to the 90 Sr/ 90 Y content of the milk. This count rate corresponds to 0.24 Bq/L 90 Sr/ 90 Y in milk. The calculated activity is in agreement with those measured in milk samples in Greece in the period 1987-1994 (0.04-1.25 Bg/L), due to Chernobyl fallout [7] and in milk samples from Alpine pastures in Austria, (0.06-0.76 Bq/L) recently [2].

4 Conclusions

Concluding, 90 Sr/ 90 Y Cherenkov radiation measurements of milk samples can be used for rapid screening of samples that are con-

taminated with activity more than 1 Bq/L, without any pretreatment of the sample. Also, 90 Sr/ 90 Y Cherenkov radiation measurements can be used for water samples with activity more than 0.3 Bq/L. The contribution of 40 K to Cherenkov radiation in milk samples can be considered as negligible if the 90 Sr/ 90 Y contamination is higher than 1 Bq/L. It is inferred that using this method it's not possible to measure activities lower than this level.

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