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# Determination of $^{90}$Sr–$^{90}$Y activity in urine samples by using Cherenkov counting

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## HIGHLIGHTS

- ▸ A new color quenching correction method (ESAR) was tested for Cherenkov counting.
- ▸ It uses the spectrum of the $^{152}$Eu outer source of a Quantulus 1220$^{TM}$ system.
- ▸ The method was applied for $^{90}$Sr–$^{90}$Y determination in urine samples.
- ▸ Results within the range of 11.5% from target values were obtained in blind tests of urine samples.

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## ABSTRACT

Cherenkov counting of the $^{90}$Sr–$^{90}$Y pure beta emitters in aqueous samples is an attractive method; but color quenching correction is needed, this being especially significant for urine which is characterized by a strong coloration. A quench correction method based on the external source of some liquid scintillation systems (named ESAR—External Source Area Ratio) was proposed for aqueous solutions. In the present work, the application of the ESAR method for determination of $^{90}$Sr–$^{90}$Y in human urine samples is described.

## 1. Introduction

$^{90}$Sr is an important fission product, being present in the environment as a result of atmospheric nuclear weapons testing. Its concentration may increase due to releases from nuclear facilities and nuclear accidents, and it is also a candidate for "dirty bombs" and emergency concerns resulting from terrorist attacks. $^{90}$Sr is a high-risk radionuclide due to its long physical half life (28.79 years), its accumulation in bone tissue and the high energy beta particles from its daughter nuclide $^{90}$Y. $^{90}$Sr decays by beta particles with an $E_{max}$ of 0.546 MeV to $^{90}$Y, which has a half life of 64 h and decays into $^{90}$Zr by beta particles with an $E_{max}$ of 2.284 MeV. Because of the short life time of the $^{90}$Y daughter, $^{90}$Sr–$^{90}$Y is in secular equilibrium after several days from production or separation.

From the environment, the uptake process of $^{90}$Sr to the body may occur through foodstuff, especially milk. $^{90}$Sr may be released also in nuclear facilities, and the radiation laboratories must be able to evaluate $^{90}$Sr uptake in workers. Since both $^{90}$Sr and $^{90}$Y are pure beta emitters, their presence in the body cannot be monitored directly, and is performed by bioassay methods, mostly urine analysis. Based on international safety standards of ICRP it can be shown that the derived recording level in urine for $^{90}$Sr–$^{90}$Y for quarterly routine monitoring (90 days monitoring interval) is about 2 Bq/l for intake of most strontium compounds. As the Minimum Detectable Activity (MDA) for direct urine measurement methods is higher than this level and as other radioisotopes may be also present, treatment of the urine samples must be employed. Several classical methods have been developed during the past years, containing several stages of chemical treatment. They may include strontium selective precipitation, ashing, ion exchange and extraction (Cahill and Lindsey, 1966; Kramer and Davies, 1982; Scasnar, 1984; Dietz et al., 1991). These steps are laborious and time consuming and use additional, sometimes expensive materials such as specific resins. The samples are then counted, usually by gas ionization, or scintillator detectors. Due to possible emergency situations requiring fast time schedules for $^{90}$Sr–$^{90}$Y determination, several rapid and automated bioassay

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methods were published recently (Plionis et al., 2009; Sadi et al., 2010).

The quantitative determination of radio-strontium is usually performed by beta counting using proportional gas detectors and liquid scintillation systems. Due to high counting efficiency and sample uniformity the liquid scintillation counting (LSC) is mostly preferred. Beta particles with energy in excess of 263 keV produce Cherenkov photons in aqueous solutions, which can be detected and quantified by using the light detection features implemented in LSC systems. Cherenkov counting has a lower efficiency relative to LSC, but there are some outstanding advantages as no adding scintillator material is required, saving time and expenses in sample preparation and less interference from lower energy beta and gamma radiation emitters. When handling urine samples, acids are used to dissolve sediments, which will inhibit the scintillation process when using LSC, therefore using Cherenkov counting for high energy beta emitters in urine samples seems to be a preferred choice. The practical use of this method is described in several publications, as (Francois, 1973) and (L'Annunziata, 2012). When measuring $^{90}$Sr–$^{90}$Y activity by Cherenkov counting, most of the Cherenkov radiation is produced by $^{90}$Y (about 98.6%), due to the much higher energy of its beta particles relative to these from $^{90}$Sr (see above). There is no chemical quenching when employing Cherenkov counting, however, the counting efficiency varies strongly with color quenching, at a greater extent than in standard liquid scintillation counting (L'Annunziata, 2012), and therefore proper quench correction techniques must be employed.

Urine is a complex material containing soluble and organic compounds, with a strong coloration which causes a significant quenching effect. Its composition varies from person to person, and also for the same person during the day. Decolorization using activated carbon can be attempted, but about 50% of the activity will also be removed from the solution (Sadi et al., 2010). Mostly, quench correction factors are applied when using LSC, based on mathematical indicators calculated from the distorted scintillation spectra, but these indices are not optimal for Cherenkov spectra. In a previous publication (Tsroya et al., 2009) it was shown that a quench correction method based on the external source of some liquid scintillation systems can be applied. The method (named ESAR—External Source Area Ratio) uses an indicative parameter based on the integral area of the spectra, and it was shown that it is superior to the common methods which use conventional indexes (Tsroya et al., 2012). The ESAR method applied to Cherenkov counting is described also in a recent review (L'Annunziata, 2012). In the present work, the application of the ESAR method for determination of $^{90}$Sr–$^{90}$Y in human urine samples is described.

## 2. Materials and methods

A Quantulus 1220$^{\text{TM}}$ LSC system containing a $^{152}$Eu standard source, operated at Soreq Nuclear Research Center/ Israel, was used for the present work. The ESAR method described elsewhere (Tsroya et al., 2009) was employed for quench correction. This method consists of two stages. In the first stage, the pulse height spectrum with and without the external $^{152}$Eu source is measured. By subtraction, the net spectrum due to the external source is obtained. This spectrum area, relative to an unquenched sample, is used to determine the quench correction factor. In the second stage, the sample is counted for a preset time, chosen to obtain the desired statistical accuracy. The activity is calculated from the spectrum obtained in the second stage, by using the quench correction factor obtained in the first stage from a previously prepared calibration curve.

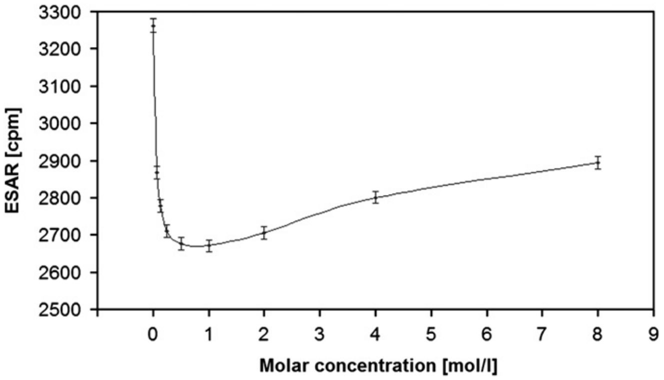

**Fig. 1.** External source count rate as a function of the sample molarity in an aqueous sample.

### 2.1. Preparation of the samples

Part of the analytical method is based on oxalate precipitation described in the modified Volchok method (Volchok, 1984). The beta-emitting material from 250 ml urine samples was precipitated as oxalate, using ammonium oxalate in an acid medium. The urine was placed in a glass beaker and Sulkovitz solution was added. The Sulkovitz solution was prepared from 25 g oxalic acid, 25 g ammonium oxalate and 50 ml concentrated acetic acid. The volume was adjusted to 750 ml with distilled water. After a few minutes of stirring, the precipitate was allowed to settle overnight. The next day, the supernatant was decanted and discarded to waste. The precipitate was transferred to a 50 ml centrifuge tube and the centrifuge was operated at 2000 rpm for 5 min. Using the centrifuge was not a part of the original procedure. It was added to improve the separation efficiency. After this stage, the supernatant was decanted and discarded to waste.

The sediment was dissolved in $HNO_3$. The molarity of the solution influences the counting efficiency by two different phenomena. On one hand it acts as a quenching factor, reducing the counting efficiency, but on the other hand the refraction index of the solution is changed, improving the Cherenkov radiation production efficiency. The dependence of the count rates due to the external source obtained in aqueous solutions for different molarities of added $HNO_3$ is presented in Fig. 1. It can be seen that there is a significant drop in the efficiency when increasing the molarity up to about 1 mol/l, but the efficiency improves constantly with increased molarity thereafter for the checked range.

A molarity near 8 seems to be a good choice for complete dissolution of the sediment and minimizing of the quench effect. 7.8 M was chosen, and the sediment was dissolved with 20 ml of 7.8 M $HNO_3$ and allowed to dissolve totally for 24 h. After 24 h the sample was transferred to a counting vial made of polyethylene, as it is generally accepted that they are superior to glass vials for Cherenkov detection (L'Annunziata and Passo, 2002; Takiue et al., 1996). The efficiency curve was prepared in a similar way as described in (Tsroya et al., 2009), by adding varying amounts of sediments up to four times the normal sediments content in 250 ml urine. All reagents used were of analytical grade. A calibrated liquid standard with specific activity of 4070 Bq/g of $^{90}$Sr in 0.1 M HCl was used, produced by Amersham Int.

## 3. Results

### 3.1. The contribution of $^{40}$K

Natural potassium contains 0.017% $^{40}$K. Potassium is present in the environment and the average intake into the human body,