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STRONTIUM-90: A REVIEW OF MEASUREMENT TECHNIQUES INCLUDING RELEASES TO THE ENVIRONMENT, ITS CARCINOGENIC PROPERTIES AND U.S. EPA RECOMMENDED ACTIVITY LEVELS

Abstract

This article gives brief review of measurement technique of 90Sr. We firstly give the brief review of sources of ^{90}Sr in environment, and then we explore the hazard health effects of ^{90}Sr on human in particular vulnerable children, followed by lifetime cancer mortality risk coefficients, and overview of detection technique. We show in particular interest for further research in order to upgrade the multi-detector system based on Cherenkov radiation.

STRONCIJUM-90: PREGLED MJERNIH TEHNIKA UKLJUČUJUĆI NJEGOVU PRODUKCIJU, KANCEROGENA SVOJSTVA I EPA PREPORUČENE DOZE

Izvod

Rad prikazuje pregled metoda za mjerenje stroncijuma-90. Prvo ćemo prikazati načine produkcije stroncijuma-90, njegove toksičke i kancerogene efekte, EPA dozvoljene koncentracije i detekcione tehnike uključujući radiometriju. Poseban interes je dat korišćenju Čerenkovog zračenja u detekciji stroncijuma-90.

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INTRODUCTION

The US Baby Tooth Survey inspired a number of similar initiatives in other parts of the world [1]. The results of a more comprehensive study of the elements found in the teeth collected showed that children born after 1963 had levels of ^{90}Sr in their baby teeth that was 50 times higher than that found in children born before the advent of large-scale atomic testing. Such analyses deeply questioned the health hazard of nuclear weapon testing. A number of related studies by the US Radiation Public Health Project assert that levels of radioactive strontium, ^{90}Sr are rising in the environment and that these increased levels are responsible for increases in cancers, particularly cancers in children, and infant mortality. There are claims that radioactive effluents from nuclear power plants are directly responsible for the increases in Sr-90. In one study, researchers reported that ^{90}Sr concentrations in baby teeth are higher in areas around nuclear power plants than in other areas. This has also sometimes been referred to as "The Tooth Fairy Project." However, numerous peer-reviewed, scientific studies do not substantiate such claims. In spite of pro and against claim, the world is faced with increase of genetic mutations, bone cancers, cancer of nearby tissues, and leukemia. The «big picture» could be largely affected or even suppressed by government body due to demand for increasing the nuclear plants in the world due to problem with electricity supply and exponential population growth. At thus place we quote the statement placed on the official public site of Environmental Protection Agency (EPA) "Large amounts of Sr-90 were produced during atmospheric nuclear weapons tests conducted in the 1950s and 1960s and dispersed worldwide. …Strontium-90 is chemically similar to calcium, and tends to deposit in bone and blood-forming tissue (bone marrow). Thus, strontium-90 is referred to as a 'bone seeker.' Internal exposure to Sr-90 is linked to bone cancer, cancer of the soft tissue near the bone, and leukemia. Risk of cancer increases with increased exposure to Sr-90. The risk depends on the concentration of Sr-90 in the environment, and on the exposure conditions" [2]. We are also aware that almost every few decades there are nuclear plant accidents (Chernobyl in 1986., Fukushima in 2011) involving huge cancer risk, thus the extensive monitoring is needed.

Montenegro does not have accredited method for measuring activity of ^{90}Sr and mainly the monitoring is based on the «gross» beta activity which is not sufficient. The results published by N. Antovic [3] shows that source of $^{239+240}Pu$ and ^{137}Cs in Montenegro is deposition from both the atmospheric nuclear tests and Chernobyl accident. For obtaining precise information about the Chernobyl accident contribution to the Montenegrin territory contamination, the $^{238}Pu/^{137}Cs$ and $^{238}Pu/^{239+240}Pu$ activity ratio should be also analyzed. The fact that there is deposition from both the atmospheric nuclear tests and Chernobyl accident still in Montenegro, strongly support the need for Sr-90 Montenegro mapping and monitoring. This article is the first in row towards establishing Sr-90 monitoring in Montenegro. We firstly give the

brief review of sources of ^{90}Sr in environment, and then we explore the hazard health effects of ^{90}Sr on human in particular vulnerable children, followed by lifetime cancer mortality risk coefficients, and overview of detection technique. We show in particular interest to develop a new detector for ^{90}St/^{90}Y activity measurement.

BETA DECAY OF ^{90}SR

In β^- decay, the weak interaction converts an atomic nucleus into a nucleus with one higher atomic number while emitting an electron e^- and an electron antineutrino ν^e. The generic equation is:

$$ {}^{A}_{Z}X \rightarrow {}^{A}_{Z+1}X' + e^- + \bar{\nu}_e $$

where *A* and *Z* are the mass number and atomic number of the decaying nucleus, and *X* and *X'* are the initial and final elements, respectively.

The process where neutron is transformed into proton is shown by the following term:

$$ n \rightarrow p + e^- + \nu_e $$

Following above equations, ^{90}Sr decay is shown by following term:

$$ {}^{90}Sr \rightarrow {}^{90}Y + e^- + \bar{\nu} $$

A basic characteristic of the beta- decay process is the continuous spectrum of the emitted beta particles as illustrated in Fig 1.

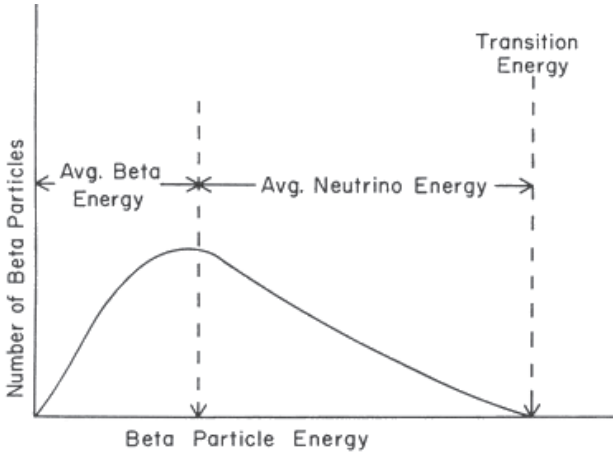

Figure 1. The illustration of spectrum of the emitted beta particles [4].

Simply caused by the fact that only the electron and the proton could be detected, a fixed energy of two body decay was expected. Thus, conservation of energy and momentum seemed to be violated. The problem was solved by Pauli in 1930 by suggesting three body decay under the assumption of a neutral not yet known particle. The name 'neutrino' for this particle was introduced later by Fermi. Due to the extremely low interaction probability of the neutrino, it took more than 25 years until it's existence could be confirmed experimentally by Cowan and Reines in 1956. Since the antineutrino shares the energy with the beta particle in the radioactive decay, the resulting beta radiation has a continuous distribution of energies from 0 to maximum decay energy (endpoint kinetic energy).

For most beta sources, this maximum value range from a few tens of KeV to few MeV. In case of ^{90}Sr decay the endpoint kinetic energy in the beta spectrum is equal E_{max} = 0.546 MeV [4]. Its daughter nucleus ^{90}Y is also unstable and in turn decays with half-life $t_{1/2}$ = 64 hours to ^{90}Zr which is stable. The decay scheme of ^{90}Sr/^{90}Y is shown in Figure 2.

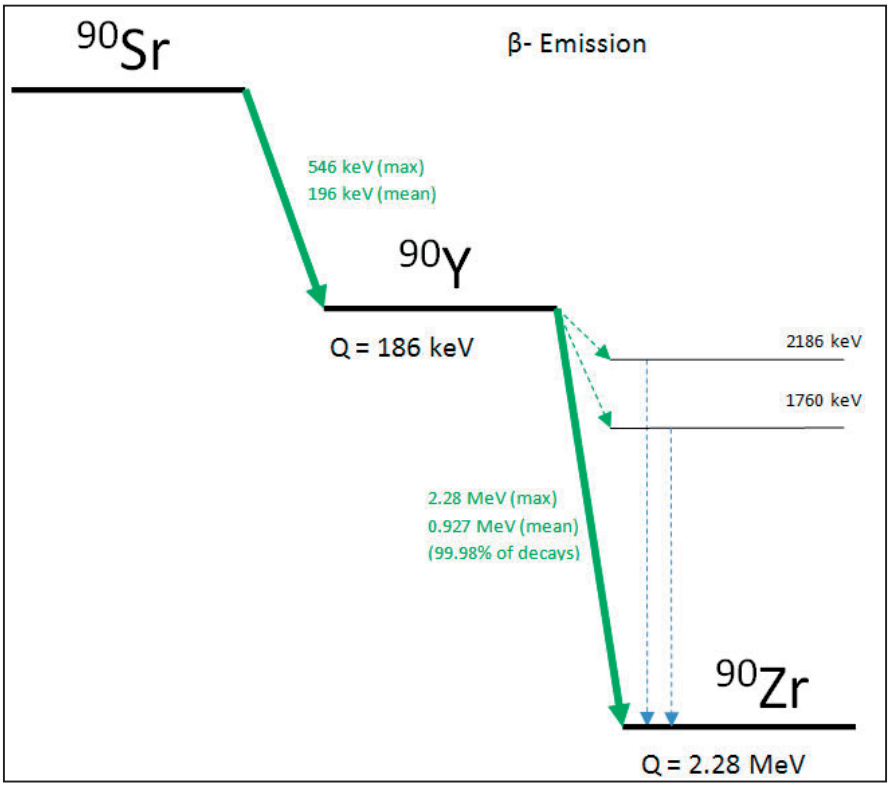

Figure 2.The decay scheme of ^{90}Sr/^{90}Y

POTENTIAL TO HUMAN EXPOSURES

Stable Strontium

Elemental strontium (atomic number 38) occurs naturally in the Earth' mantle as a mixture of four stable isotopes, ^{88}Sr, ^{86}Sr, ^{87}Sr, and ^{84}Sr, and is present everywhere in very dilute concentrations. It is very similar to calcium in its environmental and physiological behavior. There is no extensive study done in Montenegro, thus at this place we quote the U.S. EPA report [5], and expect that levels of 90Sr are lower in Montenegro due to cleaner environment: "Strontium naturally occurs in the earth's crust and is released into the atmosphere as a result of natural processes such as entrainment of dust particles, resuspension of soil by wind, and sea spray. It is generally found in molecular compounds with other elements. Commercially important strontium minerals include celestite (SrSO4) and strontianite (SrCO3). Strontium is used in the manufacture of ceramics and glass products, primarily in the faceplate glass of televisions and other cathode-ray-tube devices, where it serves to block x-ray emissions. The general population is exposed to stable strontium primarily by ingestion of food and water, and to a lesser degree, by inhalation. The strontium content in air averages 20 ng/m3, with higher concentrations resulting from stack emissions from coal-burning plants. Strontium is present in nearly all fresh waters in amounts generally ranging between 0.5 and 1.5 mg/L, with higher levels occurring where there are celestite-rich limestone deposits. The average concentration of stable strontium in soil is approximately 240 mg Sr/kg, but agricultural soils may be treated with phosphate fertilizer or limestone, which contain ~610 mg Sr/kg. Because strontium is chemically similar to calcium, it is taken up from the soil by fruits and vegetables. The average concentration of strontium in fruit produce ranged from 0.0416 to 2.232 μg/L. The total estimated daily exposure to stable strontium is approximately 3.3 mg/day (0.046 mg/kg/day): 400 ng/day from inhalation, 2 mg/day from drinking water, and 1.3 mg/day from the diet. Assuming a reference body weight of 70 kg, the typical daily strontium exposure is 46 μg/kg body weight. The strontium content of the human body is approximately 4.6 ppm by weight, 99% of which is localized in bones and teeth. Blood concentrations of strontium are in the range of 20–31 μg/L".

Radioactive Strontium

The radioactive isotopes of strontium do not occur naturally but are produced as a by-product of nuclear fission of ^{235}U, ^{238}U, or Pu-239.. The most significant isotopes are ^{90}Sr (half-life of 29 years [4]), ^{89}Sr (half-life of 51 days [4]), and ^{85}Sr (half-life of 65 days [4]), which decay by the emission of beta particles. ^{90}Sr is currently found in spent fuel rods in nuclear reactors and is considered a waste

product. Other radioactive strontium isotopes have been employed for medical uses: ^{89}Sr (as Metastron™) as a cancer therapeutic for the relief of bone pain and ^{85}Sr in the radiological imaging of bone.

RELEASES TO THE ENVIRONMENT

Nuclear weapon testing

Radioactive strontium (e.g., ^{90}Sr) was released into the atmosphere from above-ground testing of nuclear weapons during the period of 1945–1980. Nuclear weapon testing injects radioactive material into the stratosphere, which results in wide dispersal of radionuclides. However, atmospheric deposition of ^{90}Sr has steadily decreased from a high in 1963 of approximately 1.10x10^{8} GBq (3.0 MCi) to <3,000 Ci in 1990, which suggests that global concentrations of ^{90}Sr in the atmosphere have declined., ref. [6]

Chernobyl disaster in the Ukraine and Mayak plutonium complex in Ural

Other sources of regional contamination from radiostrontium include large-scale nuclear power plant accidents such as the Chernobyl disaster in the Ukraine (April 1986), which resulted in releases of about 2.2 MCi (8.1x10^{7} GBq) of ^{89}Sr and 0.22 MCi (8.1x10^{6} GBq) of ^{90}Sr into the atmosphere, ref. [5]. However, although some ^{90}Sr reached the upper atmosphere and was subsequently transported around the world, most of the radiostrontium was deposited as regional fallout in eastern Europe. The total annual releases of radiostrontium from nuclear power plants in the United States (75.4 mCi or 2.79 GBq) are insignificant compared to releases of ^{90}Sr from the testing of nuclear weapons. In the former Soviet Union between the years 1949 and 1956, large-scale environmental contamination occurred in the region surrounding the Mayak plutonium production complex in the Ural region of Russia. Releases of radioactive liquid wastes into the Techa River, both planned and accidental, of about 10^{17} Bq (2.7 MCi) resulted. ^{90}Sr contributed about 12% (or 0.23 MCi) to the total activity released. Other minor releases of ^{90}Sr have involved accidents with rockets or satellites that have disintegrated in the atmosphere. The Soviet satellite Cosmos 954 powered by a plutonium fueled nuclear reactor released 3.1x10^{3} GBq (83 Ci) of ^{90}Sr to the regional atmosphere in northern Canada in 1978. According to EPA report ^{90}Sr is found in nearly all soils in the United States. Between 1954 and 1989 at the Savannah River Site, 105 Ci (3.9x103 GBq) of ^{89}Sr and 299 Ci (1.1x104 GBq) of ^{90}Sr were released into onsite seepage basins. The total content of ^{90}Sr on surface soil in the 30 km contaminated zone around Chernobyl accident site in the Ukraine (not including the reactor site and waste storage) was about 8.1x10^{14} Bq (2.2x10^{7} Ci) in 1997, which corresponds to about 0.4–0.5% of the Chernobyl reactor inventory at the time

of the accident (Kashparov et al. 2001). Ten years after the accident, about 95% of the ^{90}Sr activity is associated with the upper 10–20 cm layer of soil for most of the soils in this area. Mean ^{90}Sr activity in soil at a Chernobyl contaminated field site in the Ukraine was 36 Bq dry weight (0.97 nCi dry weight) (Malek et al. 2002). Levels of ^{90}Sr in soils from Belarus situated at a distance of ~40 km from the Chernobyl accident site ranged from 50 to 640 kBq/m^2 (1.4–17 μCi/m^2), while levels at a distance of 200–250 km ranged from 10–80 kBq/m^2 (270–2,200 nCi/m^2) (Sokolik et al. 2001). The mean activity of ^{90}Sr in lacustrine and marine sediments from Antarctica in 1989–1996 ranged from 0.17 to 0.76 Bq/kg dry weight (4.59–20.5 pCi/kg dry weight) and from <0.10 to 0.21 Bq/kg dry weight (<2.7–5.78 pCi/kg dry weight), ref[6].. As additional information we give in Table 2 the comparison of total atmospheric releases of 137Cs and the most abundant natural radioisotopes in the ocean.

Table 2. Atmospheric Releases of 137Cs (Total) vs. Most Abundant Natural Radioisotopes in the Ocean

Atmospheric Releases of 137Cs (Total)	
Global nuclear weapon testing 1950-1960:	36 million Curies
Chernobyl	1-3 million Curies
Three Miles Island	10 Curries
Most Abundant Natural Radioisotopes in the Ocean	
Uranium 238	1 billion Curies
Potassium 40	400 billion Curies

SUMMARY OF HEALTH EFFECTS

Stable Strontium

There is no direct evidence that stable strontium is toxic to humans under normal environmental exposures. The primary toxicological effect of absorbed strontium in laboratory animals is abnormal skeletal development (rickets), which occurs only at relatively high oral doses.

Radioactive Strontium

"Exposure to radioactive strontium can result in health consequences that vary depending on the dose, the route of exposure, and the chemical form. Both ^{90}Sr and ^{89}Sr emit beta particles, which, in tissue, may ionize cellular molecules within a range of 1 cm, resulting in tissue damage and disruption of cellular function if the capacity of natural repair mechanisms is exceeded. Adverse health effect occurs at high levels of exposure that significantly exceed background levels encountered by the general population. It should be noted that no discernable adverse health

effects were detected in the general population from chronic low-level exposure to ^{90}Sr in fallout during the period of aboveground weapons testing. ^{90}Sr represents the most significant isotope of concern because of its relatively long half-life (29 years [4]) and because of the bone-seeking properties of strontium. The most serious effects of oral exposure to absorbed radioactive strontium are necrotic lesions and cancers of bone and the adjacent tissues. High level acute exposures can destroy hematopoietic bone marrow, leading to acute radiation syndrome (see below), the primary cause of mortality in the short term. At lower doses, irradiation of bone marrow may lead to chronic suppression of immune function." [7].

MAXIMUM CONTAMINED LEVEL (MCL) AND LIFETIME CANCER MORTALITY RISK

The extensive list of regulations and advisories for 90Sr can be found in ref [8]. We quote extracted parts:" Derived intervention levels (DIL), which are based on food intake rates, are calculated for different age groups and the DIL for the most vulnerable group is then adopted to provide a conservative margin of safety for the entire population. For ^{90}Sr, with a half-life of 29 years, the DIL is based on the dose to the bone surface in 15-year-old individuals, who have the highest rate of bone growth. For ^{89}Sr, with a half-life of 50.5 days, 3-month-old infants represent the most sensitive group because of the higher doses to the lower intestine from milk consumption. Lifetime cancer mortality risk coefficients have been calculated for nearly all radionuclides, including strontium-90" (see Table 1).

Table 1. Lifetime cancer Mortality Risk coefficients:

Isotope	Lifetime Cancer Mortality Risk	
	Inhalation (pCi^{-1})	Ingestion (pCi^{-1})
Strontium-90	1.0×10^{-10}	7.5×10^{-11}

"The risk is associated with the high energy beta particle emitted by yttrium-90. While the risk coefficient for ingestion is lower than for inhalation, ingestion is generally the most common means of entry into the body. Similar to other radionuclides, the risk coefficient for tap water is about 80% of that for dietary ingestion. In addition to potential radiogenic effects, strontium has been shown to inhibit calcification and cause bone deformities in animals, notably at high doses. The EPA toxicity value for estimating the potential for non-cancer effects is termed a reference dose. This is an estimate of the highest dose that can be

taken in every day without causing an adverse non-cancer effect. The reference dose used to estimate non-cancer effects for strontium from oral exposure is 0.6 milligrams per kilogram body weight per day (mg/kg-day). This value was developed by studying test animals given relatively high doses over their lifetimes, then adjusting and normalizing those results to a mg/kg-day basis for humans. EPA recommends that drinking water levels of stable strontium should not be more than 4 milligrams per liter of water (4 mg/L)", ref [8,9].

"Department of Energy (DOE) in US established derived air concentrations (DAC) for workplace exposure to radiation at DOE facilities. The DAC ranges from 0.000000002 microcuries per milliliter (μCi/mL) ($2x10^{-9}$ μCi/mL of air = 70 μBq/mL of air) for radioactive particles remaining in the lung for 100 days to 0.000000008 μCi/mL ($8x10^{-9}$ μCi/mL of air = 300 μBq/mL of air) for radioactive particles remaining in the lung for less than 10 days. The USNRC established an annual intake limit of 20 μCi (7 MBq) for on-the-job exposure to ^{90}Sr in air", [8]. "A set standards for the concentration of ^{90}Sr in community water supplies. The average annual concentration of ^{90}Sr in water supplies should not exceed 8 pCi/L (0.3 Bq/L). EPA also established maximum contaminant levels (MCLs) in drinking water for radionuclide activities to protect against harmful effects of ^{90}Sr. For beta particles like strontium, the MCL is 4 mrem per year (4x10-5 Sv per year). The USNRC set a workplace value of 31 μCi (1.1 MBq) for the amount of ^{90}Sr that can be taken in by mouth in a year without any harmful effects", ref …With regard to risk assessment, reference is usually to the equivalent dose to specified organs or to the effective dose. ICRP recommended effective dose limit is 1mSv per year. Detection limits for man-made beta particle and photon emitters for ^{90}Sr is 2pCi/L. The USNRC recommended dose to an embryo/fetus (dose equivalent during the entire pregnancy) is set up to 0.5 rem (5 mSv)".

CHEMICAL TECHNIQUES

We shall discuss methods for strontium-90 and strontium-89 determination in air, water, milk, in other media. Either of two types of measurements it commonly required: In one type, a quantitative measurement of strontium-90 specific activity (e.g., pCi/g) it made, while in the second type it is necessary to determine an upper limit of activity to assure safety. In the second type, a gross-beta measurement is usually perforated at a first screening step, since it is easier than elaborate strontium-isolation chemistry. Only when the gross-beta measurement exceeds guidelines, or when a strong suspicion of strontium contamination is present, is specific strontium analysis called for. There is important request: strontium must be determined at activity levels well below the permissible gross-beta levels in strontium's absence.

A goal summary of the various type of chemical separation techniques has been given by Bowen in Ref. [10]. There is an extremely wide variety of methods for radiostrontium, but all follow a similar outline: chemical extraction, followed by purification and sample preparation, followed by counting. Separate treatments are applied to the various media: air, water, milk (principally foodstuffs and soil). The separate treatment is necessary because of the differences in the chemical techniques: for example, in both milk and urine yttrium forms an anionic complex which makes possible its separation by direct anion exchange. In air filters, the chemical techniques are quite different. The more common technique for strontium-^{89}Sr/^{90}Sr separation uses the ingrowth of the daughter ^{90}Y. Counting rates (corrected for efficiency) can be compared from two measurements, the first immediately after strontium chemical reparation (within a few hours, say) and the second 3 to 6 days later. Alternatively, chemical separation of the yttrium daughter can be performed after allowing for ingrowth. Both techniques are frequently used. Measurements of „total radiostrontiun" usually refer to the total activity of a strontium fraction immediately after yttrium is separated chemically. The time elapsed before counting is critical, of course, because after strontium-90 and yttrium-90 reach parent/ daughter equilibrium the „gross beta" counting rate represents exactly twice the activity of the parent strontium-90 itself. We do not want to go into details since it is already done in paper we referred to, but for the reader's better understanding of complexity of chemical methods we give at this place quote from ref [11]: „Strontium is separated from calcium, other fission products, and natural radioactive elements. Nitric acid separations remove the calcium and most of the other interfering ions. Radium, lead, and barium are removed with barium chromate. Traces of other fission products are scavenged with iron hydroxide. After the strontium-90/yttrium-90 equilibrium has been attained, the yttrium-90 is precipitated as the hydroxide and converted to the oxalatelor counting. Strontium chemical yield is determined gravimetrically as strontium carbonate, 0.5 pCi strontium-90 can be determined by this method with an error of less than $\pm 10\%$ at the 95% confidence level under usual conditions (counter background of less than 1 cpm and a 60-minute count)."

Perhaps the key point to make is that, at the low levels, the only method for *specific radiostrontium* analyses involve *analytical chemistry* followed by beta counting. The difficulty in comparing different chemical techniques is that rest too much in mind and hands of analyst. A choice may mostly depend more on the particular skills and equipment available in laboratory then on the difference in techniques themselves. This point has been made extremely well by Bowen, from whom we quote (Ref. [10]): „Strontium-90 in sea-water choice among these various possibilities has been made partly on the basis of prejudice, partly on economics, partly on the basis of which other radionuclides were to be sought on

the same samples, partly on the basis of which alternative used fewest reagents known or suspected to be significant contributors to the radiochemical blank and partly on the basis of safety. In most of these variations the decisions have been legitimate ones, but we feel of even more importance have been questions which one can still only describe as those of art: in the hands even of very competent and experienced analysts, no one method, whether of trace element or of low level radiochemical analysis, appears ever to have been uniformly superior in quality of results. For this reason, we strongly resist the suggestion that a single 'standard' method be selected and recommended." Furthermore, we quote Bowen here again"... it is clear, we believe, that in suitable hands each method was capable of delivering data of high quality; it is also clear from some of the entries ... that these well-established methods were capable in unsympathetic hands of yielding some pretty bizarre sets of numbers, indeed." The author of this article is not chemist but is nuclear and particle physicist, with strong interest in developing technique for the measurement of ^{90}Sr activity where the results would depend less upon a specific skill of the analyst and more upon the general competence of method and technique itself. To our opinion much can be done in the development of these or other methods to make them less dependent upon individual person. We also refer to [11] for more details. As pointed out in ref [12]: „..further research and development in this field is most needed in simplifying, standardizing, and automating the methods; but finally, that the market for a fully automated system (even if available) is probably insufficient to justify its development. „We agree and want to emphasize that chemical methods involved in determination of ^{90}Sr activity seems to be unavoidable in the case of low-activity of ^{90}Sr, thus special attention has to be addressed to analyses of systematic errors which determine the reliability of method and accuracy of measured ^{90}Sr level. The strontium analyses is subject of variety of possible errors which need to be understood if reliable results are to be achieved. The accuracy of ^{90}Sr activity measurement depends strongly on accuracy of chemical procedure used for strontium separation as well as on the accuracy of beta-counting and good knowledge of scientist who is dealing with data in order to extract information from beta-spectra and present the result. Unfortunately many results are published without giving transparently full range of systematic errors (mostly statistical errors are presented). The errors are related to a few steps: analytical separation, radiometry, method performance (decontamination, chemical yield, radiometric assay, method validation). As many published papers reported, (for details we refer to [13]) and we quote it here „Incomplete extraction of the determination nuclides from the matrix of environmental materials represents a common source of error." It was suggested that complete extraction of strontium nuclides can be achieved by leaching with dilute nitric acid after the sample was ashed in a muffle furnace to destroy organic matter. And as it was

point out previously, the analyst is the one who is making decision which procedure to apply (publishing does not make the result more reliable) and we quote the following from Ref. [13]: „However, some refractory materials may need total dissolution in order to free all of the strontium from the sample matrix and ones is on the analyst to decide which method to use". Regarding radiometric assay, the most obvious errors are likely to arise as result of inadequate decontamination from other beta-emitting nuclides when these are not eliminated to a sufficient degree by the sequence of separation steps [13]. The environmentally important interfering radio-nuclides which might contaminated sample are the following: ^{45}Ca with $t_{1/2}$ = 163d and b = 0.257 MeV, ^{91}Y with $t_{½}$ = 58.6 d and b =1,544 MeV, ^{95}Zr/^{95}Nb with $t_{1/2}$ = 64d (3,5d) and b = 0.366 MeV and 0.386 MeV respectively, ^{106}Ru/^{106}Rh with $t_{1/2}$=368 d (30s) and b = 0.0394 MeV and 3.54 MeV respectively, ^{140}Ba/^{140}La with $t_{1/2}$=12.6d (40.2h) and b = 1.03 MeV and 1.68 MeV respectively, ^{210}Pb/^{210}Bi/^{214}Bi) with $t_{1/2}$=138.4d (5.0d/20min) and b = -, 0,74 MeV and 3.28 MeV. The list is not exhaustive and analyst should carefully check their presence. Another counting error which is not necessarily obvious occurs in the measurement of the 85Sr yield tracer. Unlike the standard solution of ^{85}Sr, which is to be compared, the prepared sample solution (after chemical separation of strontium from matrix) contains varying amount of relatively high-energy b -emitting nuclides- notably ^{89}Sr and ^{90}Y. Such high energy b particles cause Bremsstrahlung radiation and often the counting system is not capable to distinguish pulses from Bremsstrahlung radiation coming from ^{89}Sr and ^{90}Y from those coming from ^{85}Sr what lead to systematic errors and inaccuracy of final result. In radiometric assays ^{90}Sr is quite easy unambiguously via its short-lived daughter 90Y due to high endpoint kinetic energy of 2.28 MeV. Also standardized solutions of ^{90}Sr/^{90}Y are readily available and the choice of Cherenkov or end-window beta counting is a matter of choice and preferences. Surely the detection limit of apparatus affects the choice. Low background (< 0.5 counts/min) gas-flow beta counters provide a detection limit up to 5 times better than Cherenkov counting. ^{89}Sr cannot be assayed uniquely in the environmental samples. The radio ^{89}Sr/^{90}Sr depends of the time when sampling is done (time after nuclear accident is realized). Cherenkov counting is undoubtedly the best approach in particular when proper choice of refractive index of radiator is done since all radiation bellow the required energy threshold (we will give special attention to it later in the article) could be significantly suppressed allowing good efficiency for 90Y ($E_{threshold}$ = 2.28 MeV). Following Fig. 3, ^{89}Sr is likely to be of interest only when fresh fission product is in the environment. The sensitivity of detector on background radiation plays important part. Polythene liquid scintillations counters are preferred in comparison to glass scintillators. Very often the great part of natural radiation is removed using anticoincidence and coincidence units in multidetector systems (more attention will be given to this issue later).

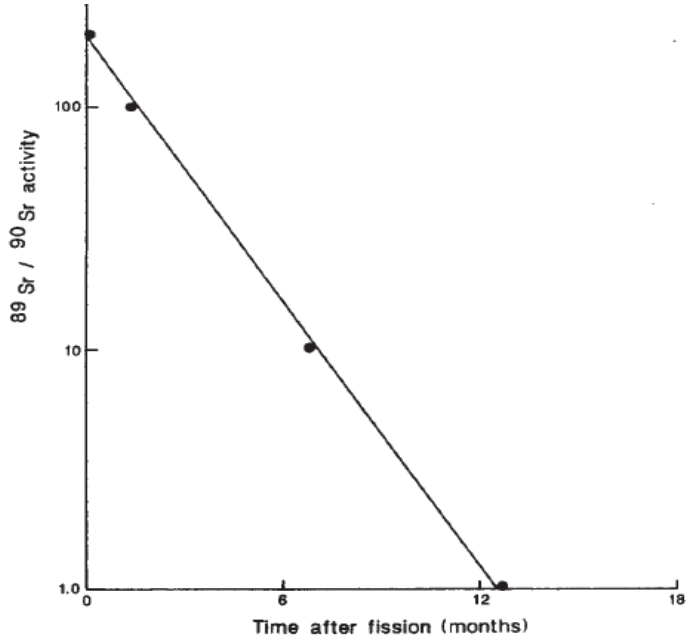

Figure 3: Relative activity of ^{89}Sr and ^{90}Sr in fission products assuming equal mass production of the two nuclides [13]

STANDARD MEASUREMENTS OF ^{90}SR ACTIVITY

Among methods, used for the measurement of ^{90}Y activity after its chemical extraction, are liquid scintillation counting, Cherenkov counting, and Bremsstrahlung solid scintillation counting with NaI(Tl) detectors. Due to the low penetrative power of beta radiation, the detection efficiency of beta emitter, especially the low energy beta emitters, are quite low. The best possible contact is achieved when the sample is dissolved in the scintillation solution. By the counting the photons produced in the reaction of beta particles with the scintillator, the quantification of beta emitting isotopes can be easily carried out. In LSC, the radionuclide is mixed with a cocktail, which is consist of solvent and scintillator (fluor), the decay energy will be transferred to the cocktail, and converted to photons, by counting the photons using a PMT (photomultiplier tube), the activity of radionuclides is measured. PM tubes are not sensitive to the fluorescence wavelength of the aromatic solvents. A fluor is therefore used to capture the solvent energy and emit light at a wavelength more easily detected. Unfortunately, the early PM tubes were not sensitive in the region of fluorescence emission of the primary scintillators. To solve this problem a wavelength shifter (secondary scintillator) was introduced. This absorbs energy from the primary scintillator and emits light at a wavelength more suitable to the PM tubes. The photons of light are collected by the optical reflector system

of the scintillation counter to the light sensitive photocathode of the PM tubes and multiplied. The electrical charge pulse obtained is proportional to the original beta particle energy. A single photon striking the photocathode has at most a 30% probability of producing an electrical pulse. It is thus of prime importance that the energy transfer process is as effective as possible in order to maximise the counting efficiency, as there are losses associated with the full light generation process (quench). At the end of journey of radioactivity detection is a Multi Channel Analyser (MCA) that is is a memory that stores the electrical pulses from the PM tubes. Each individual decay event is stored at a location in the MCA depending upon its pulse height (decay energy). A digital picture of the energy distribution of the isotope is formed as more events are accumulated. Luminescence such us bioluminescence and photoluminescence are the usual side effects of interaction of photons with medium and they have to be controlled and eliminated.

Generally, the published papers report (i) high liquid scintillation counting efficiencies for 90Y (B99.7–99.75%) over a wide range of quench levels, (ii) moderate counting efficiencies over the range of 39–68% using Cherenkov counting depending on instrumentation, sample geometry and color quench level, and (iii) low counting efficiencies with Bremsstrahlung counting (B9.9–18%) depending on sample counting geometry and detector efficiencies. Cherenkov counting has certain advantages over liquid scintillation analysis, namely, (i) it is less expensive as no scintillation fluor cocktails are used, (ii) it is carried out generally in aqueous solution without the addition ofany reagents that could destroy the sample (e.g. radiopharmaceutical) leaving the sample suitable for furthers tests (e.g. spectroscopy, chromatography, electrophoresis, bioassays, etc.), and (iii) no interference is caused by other radionuclides in the sample with decay emissions that cannot produce the Cherenkov effect, such as, ^{3}H, ^{14}C, ^{35}S, 33P, ^{45}Ca, etc. The significant number of analyses uses Cherenkov radiation in aqueous solutions. However the problem arises when Cherenkov photon yield of other β emitting isotopes interfere with photons produced by β from ^{90}Sr, since E_{max}^{β} is up to 1.5 MeV (above Cherenkov threshold). However, the E_{max}^{β} of the β from ^{90}Sr is higher than E_{max}^{β} of β from background isotopes (several exceptions – can be identified by gamma - spectrometry). The solution is to use Cherenkov radiation in aerogel and discriminate electron with different energies.

METHODS BASED ON CHERENKOV RADIATION

Cherenkov radiation

Cherenkov detectors belong to a category of radiation detectors based on the light which is emitted by fast charged particles passing through an optically transparent medium with index of refraction greater than one. An extensive coverage

of the origins and applications can be found in Ref. [14]. The light is emitted whenever the velocity of charged particle exceeds that of light in the medium through which is passing, or bn > 1. Cherenkov detectors bear some similarity to common scintillation detectors, in that the emitted light is converted into an electrical signal by a photomultiplier tube in optical contact with the Cherenkov medium. However, several important properties are quite different:

As indicated by above equation, a minimum particle velocity is required in a given medium is required in order to generate Cherenkov radiation. The required minimum particle velocity depends on the refractive index of medium (Cherenkov radiator). As a consequence, Cherenkov detectors have an inherent discrimination ability which is unique among radiation detectors. Their response is limited to electrons whose energy exceed a minimum threshold given by

$$E_{th} = m_0 c^2 \left(-1 + \sqrt{1 + \frac{1}{n^2 - 1}} \right)$$

where m_0c^2 represents the electron rest mass energy (0.5111 MeV). The threshold electron energy is plotted in Figure 4. as a function of the index of refraction n.

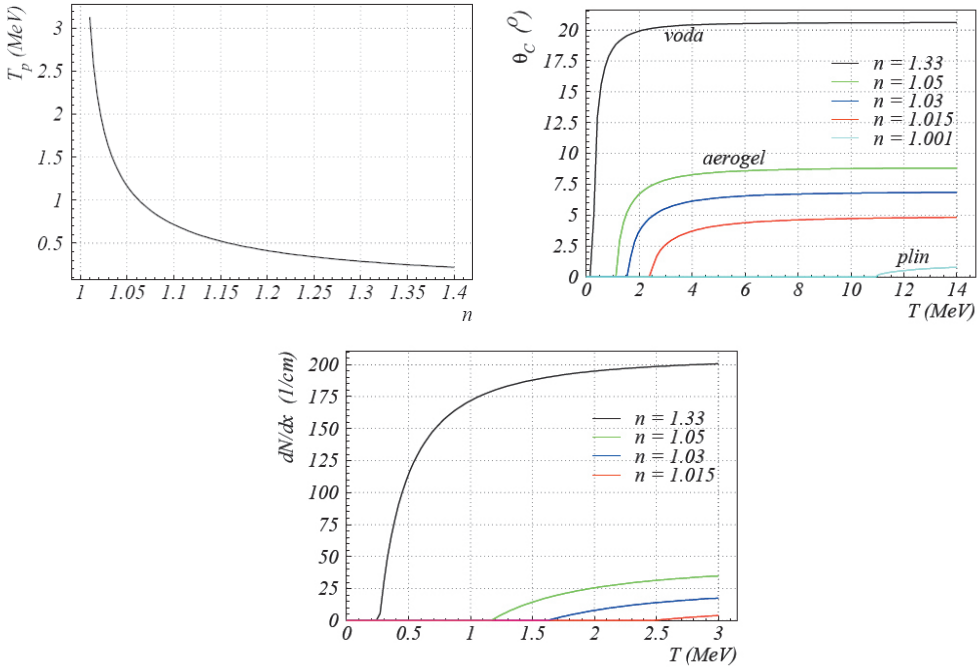

Figure 4: a) Energy threshold as a function of refractive index, b) Cherenkov angle as a function of electron kinetic energy for different refractive indices, c) the number of photons detected in full solid angle [15].

The Cherenkov angle as a function of electron kinetic energy for different refractive indices and the number of photons detected in full solid angle as a function of energy threshold for Cherenkov radiation is displayed also in Fig. 4. This inherent energy discrimination can be a very useful feature in situations in which a high rate of low-energy events would otherwise be recorded. Potential problems arising from pulse pile-up or excessive rates can be avoided if these unwanted events are eliminated at the start. By different geometry arrangement using slices of radiators with different indices of refraction, various detection thresholds can be chosen and thus the accuracy of measurement could be significantly improved. In connection to ^{90}Sr measurement, the inherent (energy threshold) discrimination feature of Cherenkov radiators is very usefull and has many advantages in comparison to other types od detection, since ^{90}Y (daughter of ^{90}Sr) has much higher endpoint kinetic energy (Emax = 2.28 MeV) and thus with proper refractive index the contributions from other pure beta and/or beta-gamma emitters could be completely avoid or drastically reduced. That way uncertainty due to the overlapping of beta spectra is reduced. Unfortunately, this also leads to lower detection efficiency and thus lower sensitivity.

The light is emitted over the very short time required for the electron to slow from its initial velocity to below the threshold energy. As a practical matter, their timing properties are normally limited by the photomultiplier tube. The most important drawback of Cherenkov detectors is the low level of the light which is produced. As shown in Figure 4 the number of photons emitted per electron in common Cherenkov media is only several hundred per MeV, and is thus about a factor of 100 smaller than in the scintillator.

In contrast to scintillation light, which is emitted isotropically, Cherenkov photons are emitted preferentially along the direction of the particle velocity. The light is confined to a cone with vertex angle θ. The yield of Cherenkov photons per unit wavelength λ is proportional to $1/\lambda^2$. The emission is therefore concentrated in the short wavelength region of spectrum. Thus to get better detection efficiency by photomultiplier using a wavelength shifter could be of great use.

When we measure the activity of a certain radioisotope, for example ^{90}Sr, we deal with difficulties arising from the mixture of radioisotopes in the sample which could overlap beta spectra of Sr. Thus, the choice of Cherenkov radiator has to improve signal to background ratio. According to above considered, in order to increase the signal contribution, namely to maximize photon yield we need to: maximize refractive index, maximize photon collection area, and to maximize photon detector sensitivity. Oppositely, in order to minimize background contribution the chosen Cherenkov radiator has to minimize yield for low energy β and at the same time to minimize refractive index, see Figure 4.

Rayleigh scattered light contributes also to the signal and it has to be takn into account. Additionally, the following requirements for the thickness has to be satisfied: all β should be absorbed in the radiator; and Cherenkov photon path to the photon detector should be minimized. As already mentioned, if the ratio of ^{90}Sr to ^{90}Y is known (e.g. the sample is in the radioactive equilibrium or after chemical separation or the time after nuclear release is known) then the activity of ^{90}Sr can be determined from measurement of Cherenkov radiation produced by electrons from ^{90}Y, which has higher endpoint energy of beta spectrum than parent nucleus ^{90}Sr. In this case the choice of lower index of refraction of radiator is favorable. This way it is possible to eliminate or at least strongly suppress the contribution of other β-emitters with lower β - energy spectra to the signal, since lower index of refraction implies higher threshold for production of Cherenkov radiation. On the other side, as we have seen from Fig. 4, the number of emitted photons decreases with decreasing the index of refraction. Since the materials with lower densities ionization losses are smaller, lower density of the radiator implies longer path on which charged particles radiate, and thus increases the number of photons. The more energetic electrons not only radiate more Cherenkov photons per unit path length, but they also radiate on a longer path. Thus, a better separation using Cherenkov radiation in water than with ordinary energy measurements (scintillation or semiconductor detectors) could be expected. However, the refractive index of water (n=1.33) with its low threshold for Cherenkov radiation of electrons (Eth= 0.263 MeV) allows many isotopes to contribute to the signal. For measurement of beta particles from ^{90}Y decay the radiator with refractive index lower than water (n = 1.33) is required since water with its relatively low threshold for Cherenkov radiation (E_{thr}=0.263 KeV) can not strongly decrease the contributions of other beta-emitters to the signal. The radiators should have refractive index about 1.005 (E_{thr} – 1.16 MeV), lower density and high transparency. Traditional gas and liquid radiators have a refractive index either smaller than 1.0018 (C_5F_{12}) or larger than 1.27 (liquid C_6F_{14}). The gap in refractive index is covered by so called aerogels which can be produced with range n=1.004 to n=1.1. Commercially available aerogels on the other hand, with refractive indices in the range 1.005 to 1.06, correspond to electron Cherenkov thresholds between 4.62 MeV and 1.025 MeV. The low density of aerogels (0.1-0.3 g/cm3) offers additional benefits, such as reduced absorption of gamma rays and a longer path on which beta-particles radiate Cherenkov photons.

Photon detector

The photon detector has to satisfy the following criteria: high quantum efficiency (QE); position sensitive; good single photon resolution; stability during operation; low noise in order to and to minimize the electronic background contribution.

The experiences from high energy physics, in particular from developing RING imagining Cherenkov detector for the KEK and CERN, shows that the most promising candidates among photomultipliers are multi-anode Hamamatsu photomultipliers such as Hamamatsu H8500 and R5900-M16. R5900-M16 has very low noise rate and higher PDE. Ather specifications are as follows: bi-alkali photo-cathode, Q.E. - up to 25 % at 400 nm; segmented anode: 16 channels with total active area 18mm x 18mm and good single photon resolution.

Background sources

Possible contributions to the background are the following: β electrons of other radioactive sources; cosmic rays; γ rays from the sample (photoefect in the PMT). How to deal with it? We need medium for Cherenkov radiation (Cherenkov radiator) with low enough refractive index and scintillator veto (plastic scintillators in anticoincidence with Cherenkov detector) and another detector in coincidence with Cherenkov detector which would have high sensitivities to beta and low sensitivities to gamma radiation (multiwire proportional chamber MWPC).

Sample preparation

The proper preparation of sample is very important. Special care should be focused into the preparation of the sample: due to stopping and absorption of the electrons in the sample; only part of them reaches the aerogel and; with lower energies. The detected rate of β electrons as a function of sample thickness is illustrated in Fig. 5.

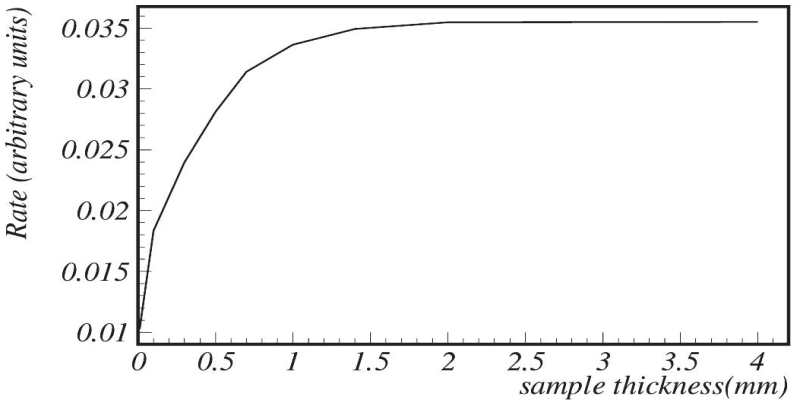

Figure 5: The detected rate of β electrons as a function of sample thickness

MEASUREMENT OF ^{90}SR/^{90}Y ACTIVITY USING EXPERIMENTAL SET UP OF CHERENKOV RADIATOR AND MULTIWIRE CHAMBER:

OVERVIEW THROUGH 20 YEARS

In 1994, a completely new method of determination of 90Y ($E_{\beta max}$ = 2.27 MeV, $t_{½}$ = 64 h) , the daughter of 90Sr ($E_{\beta max}$ = 0.546 MeV, $t_{1/2}$ = 28.5 yr) , on the basis of Cherenkov radiation of the β particles in a silica aerogel has been developed [15]. The set up was originally explored and developed in [16]. For more details we refer also to [16, 17, 18]. The experimental setup is shown in Figure 6.

Measurement system for determination of ^{90}Sr ($E_{\beta max}$=0.546 MeV) activity is based on detection of Cherenkov photons emitted in silica aerogel by beta particles. Since ^{90}Y which is the daughter nucleus of ^{90}Sr, has a relatively high endpoint energy of beta ($E_{\beta max}$=2.28 MeV), a proper choice of aerogel refractive index offers considerable reduction of interferences of other beta emitters in the sample. The interference of gamma emitters is is reduced by using a thin, transmission multiwire proportional chamber in coincidence with the aerogel Cherenkov detector. Furthermore, using the plastic scintillators in anticoincidence with multiwire chamber and aerogel+photomultiplier system, the background is significantly eliminated. The contribution of both, ^{90}Sr and ^{90}Ymay then be determined from ^{90}Y if their ratio is known. The efficiencies defined as the measured coincidence count rate divided by the activity, are shown as a function of the end-point energy. Among the sources used, ^{32}P is the only pure, one-branch beta-emitter. The measured pulse height distributios are displayed in Fig 7.

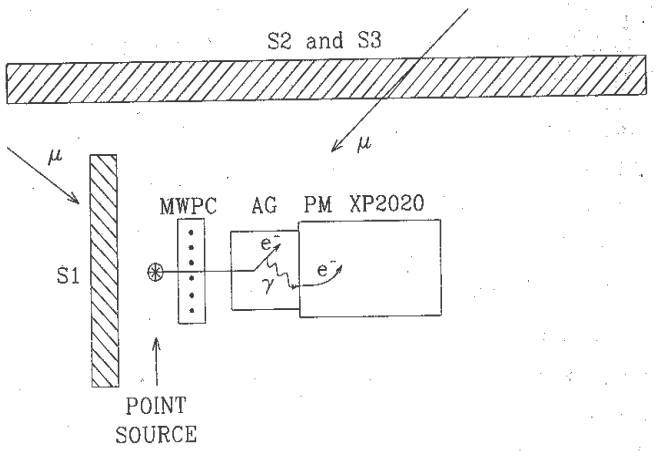

Figure 6. The experimental set up [16, 17]

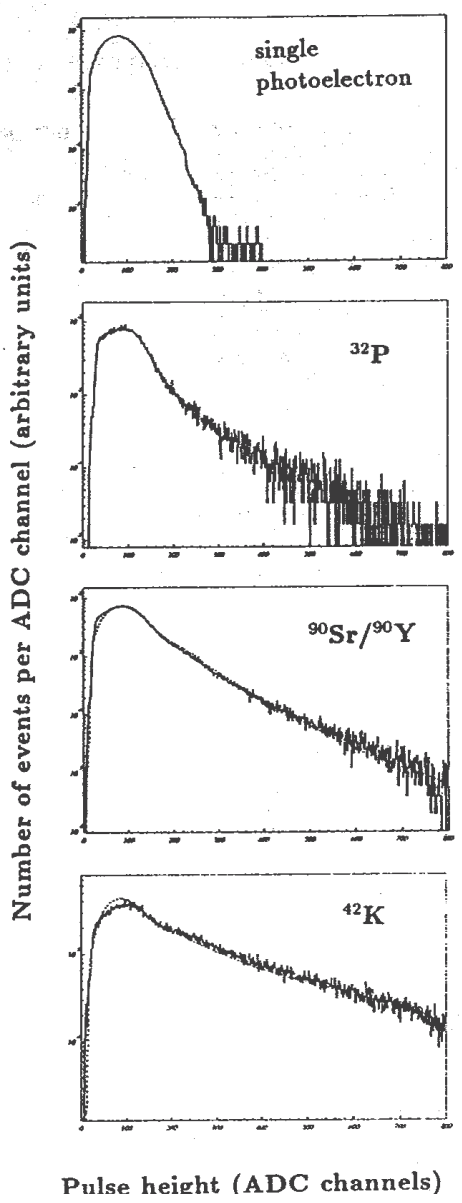

Figure 7: The pulse height distributions for calibrated sources ^{137}Cs, ^{32}P, ^{90}Sr/^{90}Y and ^{42}K [16]

Radionuclide relative detection efficiencies were measured by dividing the number of Cherenkov photon events with the number of events registered by the MWPC. Measurements of efficiencies with a silica aerogel radiator (n=1.055), described in [16,17] show a steep dependence on the β end-point energy above threshold (E_β^{th}=1.09 MeV).

Possible contributions from β-emitters other than ^{90}Y in the sample have been detected by the shape of the pulse height spectrum. An efficiency of 5 x 10^{-3} for ^{90}Y was obtained. The background counting rate of 15 x 10^{-3} s^{-1} is stable (sigma = 1 x 10^{-3} s^{-1}) and allows the determination of 1 Bq of ^{90}Y activity in a few hours of measurement of a thin sample. If the ratio of ^{90}Y to ^{90}Sr activities known (e.g. in equilibrium), the activity of ^{90}Sr may be obtained. The efficiencies obtained are however also low, which is due mainly to the low coverage of the aerogel area. This can be improved by either increasing the number of photomultipliers or by a photon concentrator such as a Winston cone.

Recent improvements in the transparency and stability of silica aerogels [19] and advances in commercially available single photon detectors [20] have led authors of article [21] to reinvestigate a method for detection of ^{90}Sr in environmental samples. Firstly, the new geometrically modified set up involving 3 slices of silica aerogels, Fig. 8.

Two different possibilities have been investigated for the radiator. In the first case the radiator consists of five 1 cm thick 10 x 10 cm2 tiles of silica aerogel. The refractive index of the first two is n = 1.03 (p = 0.106 g/cm3) and the last three n = 1.05 (p = 0.175 g/cm3). In the second case a 27 mm thick cylindrical aerogel of diameter of 42 mm is used. The entrance and side walls of the cylindrical aerogel are covered with aluminum foil to achieve higher collection efficiency. The refractive index of the aerogel in this case is n = 1.055. Finally, the aerogel radiator is viewed by a R5900 Hamamatsu M16 multi anode photomultiplier (the first multianode photomultiplier with little cross-talk), see Figure 9.

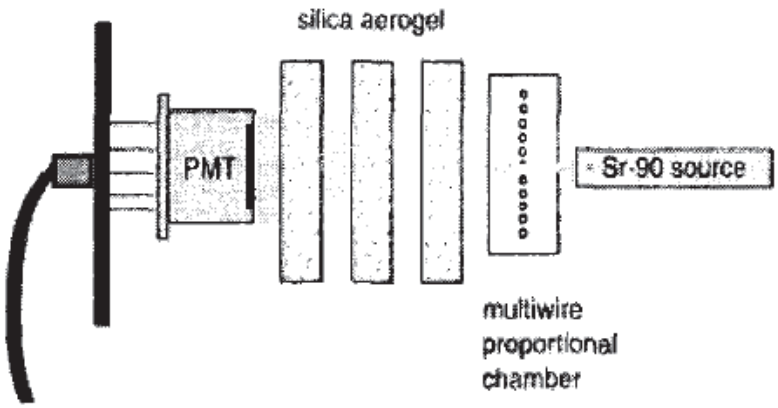

Figure 8: The 3-source, the MWPC, the aerogel radiators and the multi anode PMT are enclosed in a light tight box., set-up published in [21].

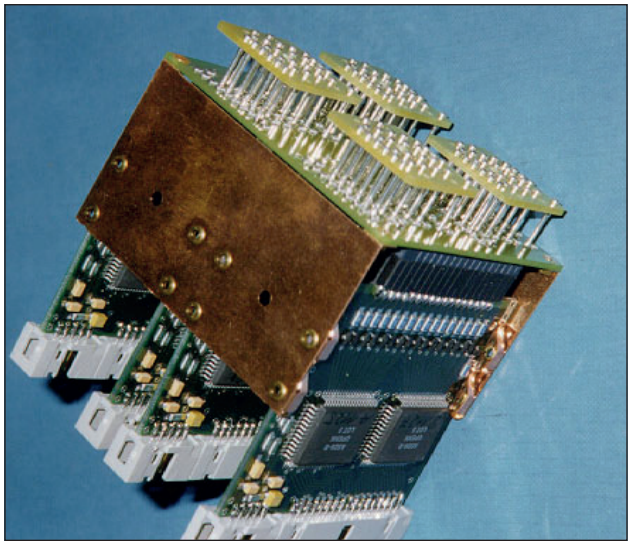

Figure 9. Multianode PMT with ASD8 board and voltage divider [22]

Interestingly. the same type of PMT was used in high energy physics HERA-B experiment for RICH (Ring Cherenkov Imagining Detector) for identification of particles such as pion, kaons and electrons. For the purpose of the present investigation, only one PMT was used. The photo cathode sensitive area of 17 x 17 mm2 thus results in a relatively poor coverage of the aerogel surface. To emphasize, in the setup published in [16-18], the author used single-channel photomultiplier, while authors in [22] used multichannel/multianode photomultiplier. So, instead to measure the height spectrum and compare it to the distribution of single photon the authors in article refereed to [22] investigate the measurement of distributions of events as a function of the number of detected Cherenkov photons. The obtained efficiency for ^{90}Sr detection was estimated to 0.0175 (1+-0.01).

A few years later, in 2008, another paper with slightly modified experimental set up involving two Hamamatsu multi-anode photomultipliers photomultipliers in order to increase of the coverage of the aerogel surface was published [23].They used an aerogel with great transparency for Cherenkov photons in the wavelength of highest photomultiplier sensitivity and lower index of refraction, n = 1.047 (the aerogel with n=1.055 was previously used), with 40 nm transmission length at lambda=400 nm. At the index of refraction, n = 1.047, the threshold energy for the production of Cherenkov photons by beta particles is 1.213 MeV which further discriminates against 89Sr beta particles (energy threshold for aerogel with n = 1.055 was Ethr = 1.16 MeV).

Since Sr90 efficiency increases as reflectivity increases, see Figure 11, it become obvious that an additional reflector would improve the efficiency for 90Y detection.

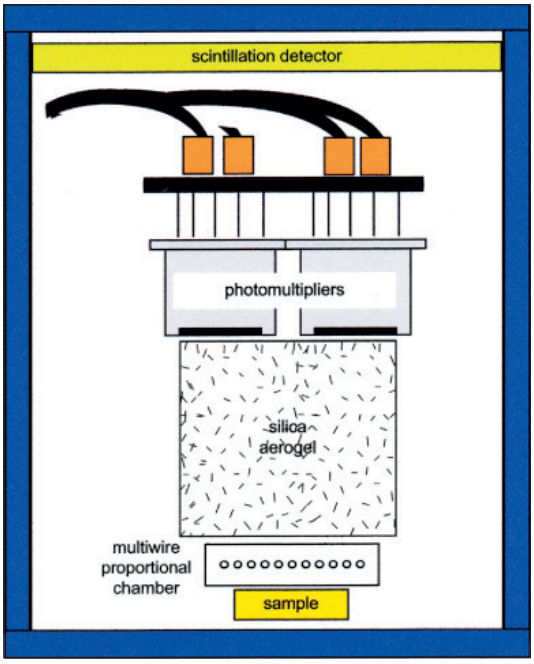

Figure 10. Set up for 90Sr measurement [23].

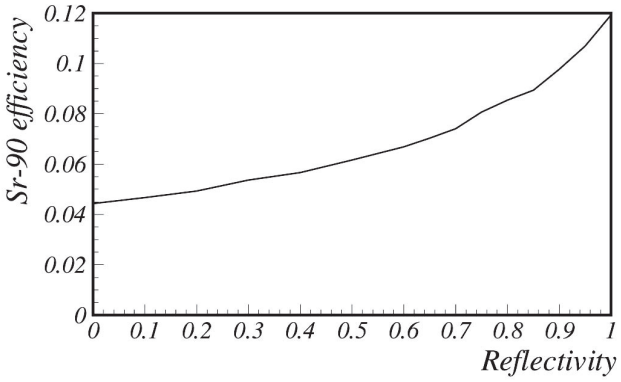

Figure 11. 90Sr efficiency as a function of reflectivity

Thus, in order to improve detector performances, a mylar foil reflector was additionally constructed to reflect photons hitting entrance and side walls of the aerogel block, increasing the efficiency by a factor of 2 (Figure 12).

Figure 12: A mylar foil reflector

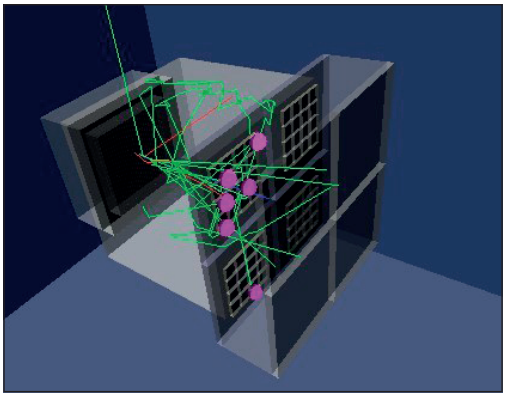

Figure 13. Illustration of Geant4 simulation

To optimize the detector parameters Geant4 simulation of the system response was used: (electron EM processes, Cherenkov photon generation in aerogel and in the PMT window, Rayleigh scattering in the aerogel, photon tracking). Cherenkov photon detection efficiency of the aerogel Cherenkov counter definedes a count rate divided by appropriate activity as a function of endpoint kinetic energy is displayed in Fig 14 [23]. The relative detection efficiency for 90Sr/90Y was 0.024.

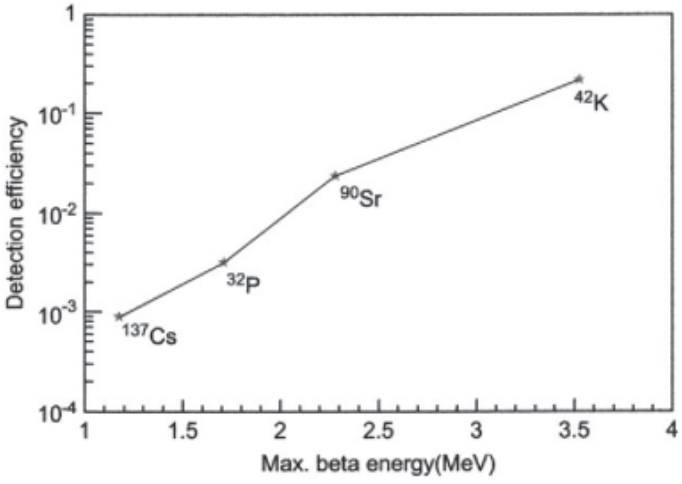

Figure 14. Cherenkov photon detection efficiency of the aerogel Cherenkov counter as a function of endpoint kinetic energy [23].

FUTURE IMPROVEMENT OF METHODS WITH CHERENKOV RADIATION

Cherenkov radiation shows advantages in comparison to other types of interaction of radioactivity with material of detector. Using better performances of aerogel or introducing wave-shifter would improve performances of detection and thus sensitivity of determination of Sr-90 activity. Multi-coincidence of multi-anode Hammatsu photomultipliers and different geometry of detectors would also bring better sensitivity. Different geometry would cover wider angle and thus increase detection efficiency. Anticoincidence arrangement is not yet fully exploited and extensive studz should be performed in future.

CONCLUSIONS

This aim of article was to collect information about Sr90 in order to support future monitoring in Montenegro. Monitoring of Sr-90 was never done. The concentration of Sr-90 in Montenegro after Chernobyl was never determined, and its level is mostly estimated using information from Croatian, Serbia and Slovenia where extensive research studies have been done in last decades. We show that chemical methods can not be avoided. Unfortunately, Montenegro does not have established and standardized method for the chemical separation of strontium from matrix (sample) and it could be one of the priorities in Montenegro's policy of radiation protection. The important part of article is determination of Sr-90 using an aerogel Cherenkov detector in coincidence with multiwire chamber where technique from particle physics has been applied in low-energy and environmental physics. The method was firstly developed in 1994 and latest version was published in 2008. Considering latest development in technology and new materials produced for the purpose of particle identification and for research in astrophysics we strongly support further investigation in this field.

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