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Faculty of Environmental Science and Engineering

Alpha spectrometry sources and environmental applications of alpha and beta spectrometry

- THESIS -

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TABLE OF CONTENTS

THEORETICAL PART

1. Introduction
  - **1.1. Background**
  - **1.2. Aim of the study**

2. Activity measurements of alpha radionuclides
  - **2.1. Alpha spectrometry technique - principle of the method**
  - **2.2. Recent status of the source preparation steps**
    - **2.2.1. Sample dissolution**
    - **2.2.2. Tracer addition**
    - **2.2.3. Preconcentration**
    - **2.2.4. Chemical separation**
    - **2.2.5. Source preparation**
  - **2.3. General description of the investigated radionuclides – radiochemistry and nuclear properties**
    - **2.3.1. Radium**
    - **2.3.2. Uranium**
    - **2.3.3. Polonium**

3. Activity measurements of beta radionuclides
  - **3.1. Beta spectrometry technique - principle of the method**
  - **3.2. Liquid scintillation counting**
    - **3.2.1. Scintillation process**
    - **3.2.2. Quench in LSC**
    - **3.2.3. Liquid scintillation methods**
      - **3.2.3.1. CIEMAT/NIST**
      - **3.2.3.2. TDCR**
  - **3.3. General description of the investigated radionuclides –uses and applications**
    - **3.3.1. Strontium: ^{89}Sr and ^{90}Sr**
    - **3.3.2. Nickel – ^{63}Ni**

ORIGINAL CONTRIBUTIONS

4. **Alpha spectrometry source preparation and activity measurements**
    - **4.1. Equipment used**
    - **4.2. Prevention of alpha detector contamination**
        - **4.2.1. Production of formvar films**
        - **4.2.2. Production of the sources with different energies**
        - **4.2.3. Measurements and uses of the protective films**
        - **4.2.4. Conclusions**
    - **4.3. Uranium alpha spectrometric sources preparation**
        - **4.3.1. Designing and optimizing an electrodeposition cell**
        - **4.3.2. Uranium determination from environmental samples**
        - **4.3.3. Preliminary uranium test measurements by spectrofluorimetry**
        - **4.3.4. Conclusions**
    - **4.4. Radium alpha spectrometric source preparation**
        - **4.4.1. Determination of ^{226}Ra by MnO_2 coated disc method**
            - **4.4.1.1. Investigation of a new material for radium adsorption**
            - **4.4.1.2. Preparation of MnO_2 coated discs**
            - **4.4.1.3. Description of analytical procedure**
            - **4.4.1.4. Results and discussions**
            - **4.4.1.5. Conclusions**
        - **4.4.2. Determination of ^{226}Ra by micro-coprecipitation**
            - **4.4.2.1. Description of analytical procedure**
            - **4.4.2.2. Results and discussions**
            - **4.4.2.3. Conclusions**
        - **4.4.3. Determination of ^{226}Ra by radon emanation technique**
            - **4.4.3.1. Sample preparation**
            - **4.4.3.2. Method description**
            - **4.4.3.3. Results and discussions**
            - **4.4.3.4. Conclusions**
    - **4.5. Polonium alpha spectrometric source preparation**

4.5.1. Description of analytical procedure
4.5.2. Results and discussions
4.5.3. Conclusions
5. Activity standardization of beta radionuclides
5.1. Equipments used
5.1.1. Hidex 300SL for TDCR method
5.1.2. Tricarb 3100TR for CIEMAT/NIST method
5.2. The investigated samples: ^{89}Sr, ^{90}Sr, ^{63}Ni
5.3. Activity calculation and interpretation of results
5.3.1. General scheme for data analysis
5.3.2. ^{89}Sr by TDCR method - results and discussions
5.3.2.1. Influence of coincidence resolving time
5.3.2.2. Influence of kB
5.3.3. ^{89}Sr by CIEMAT/NIST method - results and discussions
5.3.4. ^{90}Sr results- TDCR results and discussions
5.3.4.1. Influence of coincidence resolving time
5.3.4.2. Influence of kB
5.3.5. ^{63}Ni TDCR results and discussions
5.3.5.1. Influence of coincidence resolving time
5.3.5.2. Influence of kB
5.4. Conclusions
6. General conclusions
References

Keywords: alpha spectrometry, beta spectrometry, liquid scintillation counting, uranium alpha sources, electrodeposition cell, radium alpha sources, polonium alpha sources, strontium, nickel, coincidence time, kB

ABBREVIATIONS

ADC - Analog-To-Digital Converter

CIEMAT/NIST - Centro De Investigaciones Energeticas Medioambientales Y Technologicas And The National Institute Of Standards And Technology

CPM - Counts Per Minute

CRT- Coincidence Resolving Time

DVB - Divinylbenzene

EC - Electron Capture

EDTA - Ethylene Diaminetetraacetic Acid

EDXF - Energy-Dispersive X-Ray Fluorescence

ENEA-National Agency For New Technologies, Energy And Sustainable Economic Development

EXC - Extraction Chromatography

FC - Correction Factor

FD - Decay Factor

Formvar - Polyvinyl Formaldehyde

FWHM - Full Width Half Maximum

IAEA - International Atomic Energy Agency

ICPMS - Inductively Coupled Mass Spectrometry

INMRI – Italian National Institute of Ionizing Radiation Metrology

LET - Linear Energy Transfer

LNHB -Laboratoire National Henri Bequerel

LSC - Liquid Scintillation Counting

MARLAP - Multi-Agency Radiological Laboratory Analytical Protocols

MCA - Multichannel Analyzer

PIPS - Passivated Implanted Planar Silicon

PMT -Photomultiplier Tube

PTB - Physikalisch-Technische Bundesanstalt

ROI - Region of Interest

TDCR -Triple to Double Coincidence Ratio

TIMS - Thermal Ionization Mass Spectrometry tSIE - Spectral Index of The External Standard

Aim of the study

The thesis discusses two main fields of application of radionuclides. The first topic focuses on the measurement of the most significant alpha radionuclides for environmental research and radioprotection: the uranium, radium and polonium isotopes. Radioanalytical laboratories are often confronted with the need of accurate measurement of radioisotope quantities and isotopic ratios that are a challenge for the analysts and for the analytical methods. In this research, alpha spectrometry technique has been applied for the determination of alpha emitting radionuclides in various matrixes, which is capable of measuring the radionuclides content of samples with ultra-low concentration. Sample preparation methods for alpha spectrometry sources have been developed in our laboratory and the procedures, which imply a series of radiochemical steps, have been investigated along with their applicability in different matrices. Different procedures for preconcentration of the radionuclide of interest, diferent methods for separation of radionuclides to eliminate elements that may interfere with their measurement and different methods for source preparation have been tested. When a separation method is developed, a high chemical recovery and pure chemical fractions are important to be obtained. Most efforts were conducted to obtain both high yield recovery and good energy resolution of the alpha spectrum. Although there are many laboratories in Romania using alpha spectrometry, despite the obvious important application of this technique for dating purpose by uranium disequilibrium series, the method has not been thoroughly applied in a Romanian laboratory up to now for this purpose. Thus, developing and implementing this kind of measurements in our country is a great challenge. The research activities and the measurements were carried out in the radiochemistry laboratory of the Faculty of Environmental Science and Engineering, Babes-Bolyai University. The laboratory is well-equipped, with all necessary equipment for source preparation and measurements by alpha spectrometry.

The second topic of my thesis deals with the activity determination of beta radionuclides by liquid scintillation counting. This technique in combination with the TDCR method is a powerful method for activity standardization and can be applied to a great number of radionuclides. Up to now, all TDCR counters used by national metrology institutes for activity determinations are home-made systems. Hidex Oy (Finland) is the first company which has started the commercialization of TDCR counter. The Hidex 300SL is a revolutionary scintillation counter with a brand new design, incorporating the most advanced technology in liquid scintillation counting, Triple-to-Double Coincidence Ratio counting. The measurements performed in this work comprise an investigation of the applicability of this first commercial counter TDCR for reliable activity measurements. Within the scope of Work Package 6 of the EMRP[1] ENG08 project "Metrology for new generation nuclear power plants" (Metrofission) the only commercial TDCR counter from Hidex Oy (Finland) has to be tested by two participants. With this purpose, the Italian National Institute of Ionizing Radiation Metrology (INMRI) belonging to ENEA- Italy and the Physikalisch-Technische Bundesanstalt (PTB), Braunschweig, Germany organized a bilateral comparison to make various tests of this counter on a set of ^{89}Sr sources and to investigate the applicability of this new TDCR-LSC counter for reliable activity measurements of pure beta radionuclides. The radionuclide ^{89}Sr was selected since it is relevant for nuclear power plants. This yielded valuable information on adjustments and important test measurements for other potential users. Additionally, the activity results of measurements are compared with the CIEMAT/NIST (Centro de Investigaciones Energeticas Medioambientales y Technologicas and the National Institute of Standards and Technology) method, which is a tracing efficiency method developed by two NMIs: CIEMAT (Spain) and NIST (USA). Moreover, the measurements were extended to two kinds of radionuclides: the high energy beta emitter (^{90}Sr) and one low energy beta emitter (^{63}Ni). Attention has been focused on the influence of the coincidence resolving time and on the influence of the Birks' constant, kB on the activity measurements. The motivation of this study is user demand, from radiopharmaceutical industries and nuclear engineering, for accurate activity standards of different beta energy radionuclides. The measurements of beta radionuclides were carried out at the Italian National Institute of Ionizing Radiation Metrology INMRI, belonging to ENEA C.R. Casaccia Italy, over a period of six months research stage, during which Oana Alexandra Rusu (Dumitru) has worked under the supervision of Dr. Marco Capogni.

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1 European Metrology Research Program

Outline of the thesis

The thesis is structured in six chapters. In the first part a literature study was performed and the results are described with respect to the principles of the both, alpha and beta spectrometry, methods for source preparation and the main properties of the radionuclides of interest. This theoretical part comprises the first three chapters. In this Introduction (**Chapter 1**) the background and motivation for the realization of this thesis are described, and the goals and purposes are discussed.

Chapter 2 and **Chapter 3** are designed to give an introduction to relevant topics concerning this thesis. In Chapter 2, the basic principle of alpha spectrometry technique is briefly presented, and the recent status of the alpha source preparation steps, together with a general description of the radionuclides, with relevance for this thesis, are discussed. Chapter 3 gives an overview of beta spectrometry, respectively liquid scintillation counting method. This chapter presents the principle of beta spectrometry technique, describes the recently developed techniques in liquid scintillation counting, TDCR and CIEMAT/NIST methods, for the activity standardization of pure β-emitters, together with a general description of the beta radionuclides analyzed in the experimental work.

In the second part the realization of the practical work of the thesis is illustrated in detail – the selection of samples, the source preparation techniques and the analytical methods, the results of the thesis concerning different purposes. This part comprises **Chapter 4** and **Chapter 5.**

In **Chapter 4**, the experimental work and original contributions related to alpha spectrometry measurements are presented. This comprises the investigations performed using formvar (polyvinyl formaldehyde) home- made films to protect the alpha detector. The influence of the use of these films on alpha spectrum characteristics was investigated in a large range of energy. Included in this chapter are also the sources preparation techniques of uranium, radium polonium. Regarding the uranium sources, an electrodepostion cell was developed and designed. Thereafter, uranium sources were prepared from environmental samples, respectively from travertine and coral samples, for dating purpose. As a complementary method, spectrofluorimetry was tested to compare its detection limit with alpha spectrometry. Radium has been determined by alpha spectrometry by 2 source preparation methods: selective adsorption, for which a new material has been identified and micro-coprecipitation which is a very frequently used method. A number of drinking water has been analyzed from two important geological sites of Romania, by radon emanation technique. Polonium has been determined from water samples by spontaneous deposition and the results obtained show a good resolution of the spectrum.

Chapter 5 deals with activity standardization of pure beta radionuclides by liquid scintillation counting. The measurements performed in this thesis include an investigation of the applicability of the first commercial TDCR counter - Hidex 300SL (Finland) for reliable activity measurements. Two kinds of radionuclides were taken into account: high energy beta emitters (^{89}Sr and ^{90}Sr) and low energy beta emitter (^{63}Ni). Additionally, the results are compared with the CIEMAT/NIST method, which is a $4\pi\beta$ Liquid Scintillation tracing efficiency method. Attention has been focused on the influence of the coincidence resolving time on the activity measurements performed with the TDCR technique.

Finally, the results of the thesis are summed up in **Chapter 6**. Final statements and conclusions to the results are given.

The references used within this thesis are listed in the **References**.

ORIGINAL CONTRIBUTIONS

4. Alpha spectrometry source preparation and activity measurements

4.1. Equipment used

The alpha spectrometer used in this research is an ORTEC SOLOIST alpha spectrometer with PIPS detectors. The theoretical resolution of the detectors is 19 keV (ORTEC operating manual) and the data acquisition was made by ASPEC-927 Dual Multichannel. The module contains two independent detectors (900 mm^2 and $1200mm^2$ size), each including a vacuum chamber.

4.2. Prevention of alpha detector contamination

The production of formvar films and the results of using these films between source and detector are described. One of the purposes of the present work was to investigate the influence of these protective films on energy shift, efficiency and peak resolution lost, using two PIPS detectors with different areas.

4.2.1. Production of formvar films

To prevent this type of contamination we placed thin formvar foils (polyvinyl formaldehyde, $C_{10}H_{18}O_{12}$ purchased from Sigma – Aldrich Co. Germany) between source and detector to stop recoil nuclei. In order to obtain the formvar films we used previously described method by Van der Wijk A. (Van der Wijk, 1987, Van der Wijk et al. 1987).

The preparation process was the following: a 30 g/L formvar solution is prepared in a solvent mixture of chloroform: dichloroethane = 2: 3 under stirring at 30°C and stored in a tightly sealed glass vessel to prevent evaporation. Instead of separation funnel used by other authors (Van der Wijk, 1987, Van der Wijk et al. 1987), in the process of making the films we used a dip-coater system for layers deposition. This system is capable of achieving a uniform coating and also, accurate speed control is obtained. A piece of thin optical glass, previously cleaned with acetone and ethanol, is firmly fixed in a dip-coater. The layered deposits are made by immersing the glass successively in the formvar solution, keeping them 5 seconds in solution and 60 seconds drying between each deposition step (Rusu et al, 2011a).

A thin formvar film of high homogeneity sticks to the glass surface. For recovering the film the glass is lowered into water under an angle of about 45°. Films have been caught on a special design frame and at the corners the films were fixed using tape power (Rusu et al, 2011a).

4.2.2. Production of the sources with different energies

For the investigation of the alpha spectrometry parameters we used three different sources with different alpha energies. The measurements were performed with an alpha standard source containing ^{241}Am, ^{244}Cm and ^{239}Pu (Radiochemical Centre Armesham, The Netherlands). Additionally ^{238}U – ^{234}U source obtained by electrochemical deposition in our laboratory and ^{212}Bi – ^{212}Po source obtained by implantation were also used to investigate the energy loss of alpha particles and the decrease of detector resolution.

4.2.3. Measurements and use of the films

a) Using Pu – Am – Cm source and 900mm^2 PIPS detector

The source of Pu-Am-Cm was measured by inserting separately each of four films between source and detector. An example of the change of the peak position and energy resolution due to stopper foil is presented in Figure 4.9. In this figure a comparison between the spectrum of the source without any stopper film (a) and the spectrum of the source with five films between the source and detector (b) is presented. By overlapping the mentioned spectra for the Pu – Am – Cm source, a shift of the peaks of about 150 keV for ^{239}Pu, 138 keV for ^{241}Am and 130 keV for ^{244}Cm is observed.

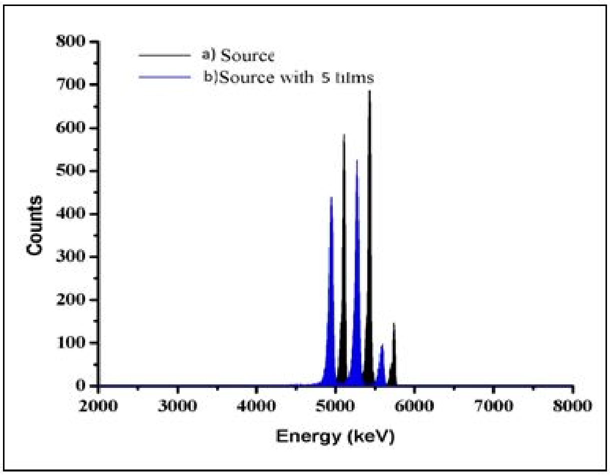


Figure 4.9. The shift of the spectrum of the standard Pu – Am – Cm source by using 5 stopper foils. a) source; b) source with 5 foils.

The decrease of peak height (~20%) for 5 layers of stopper foil is compensated by an increase of width of peak, thus the intensity of the peak is conserved. The intensity variation (1.37%) is much smaller than statistical deviations (5.4%) due to the phenomenon of disintegration. The use of films lowers the alpha particle energy, but the number of alpha particle reaching the detector is not changed significantly (Rusu et al, 2011a).

In order to estimate the thickness of formvar films, the ASTAR web databases for helium ions which calculate stopping powers, ranges and related quantities in 74 materials between 1 keV and 1 GeV was used (Berger et al, 2005). The results are presented in Table 4.3, where ΔE represents the average values calculated in Table 4.3 for energy shifts and LET is calculated using ASTAR web databases.

Table 4.3. Stopping power and thickness of formvar foils.

Radionuclide	**Energy (MeV)**	**LET ($MeV/g/cm^2$)**	**Δ E (keV)**	**Thickness (g/cm^2)**
^{239}Pu	5.157	724.70	30.27	5.43E-05
^{241}Am	5.486	694.10	29.85	5.58E-05
^{244}Cm	5.805	667.20	25.91	5.04E-05

Considering that the effect of each film is similar, the following measurements were performed placing by addition increased number of films between the source and the detector. Figure 4.10 shows the correlation between the energy shift ΔE (keV) due to the increased multi coulomb interactions of alpha particles as foils are added, measured as a function of number of stopper foils.

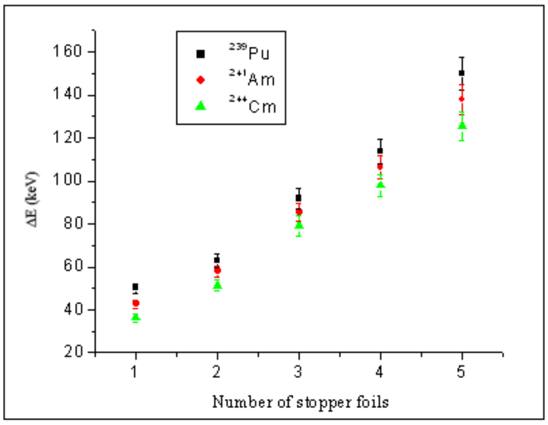


Figure 4.10. Energy shift versus number of stopper foils for the energy of the standard Pu – Am – Cm source.

We find that as the foil thickness increases, the alpha particle energy will decrease, and also the full width half maxima of the peaks will widen.

b) Using ^{238}U *–* ^{234}U *source and a PIPS detector of 1200* mm^2

The results obtained in this case are shown in Table 4.4. As it can be seen from this table for both energies of uranium, 4.196MeV (^{238}U) and 4.777 MeV (^{234}U), the energy shifts linear increase with the number of foils from 42.8 keV in the case of ^{238}U for single foil to 186.1 keV in the case of 5 foils, while for ^{234}U this increasing is from 41.3 keV to 165keV. Decreasing of resolution (FWHM) is also linear from 4.81keV for a single foil to 22.91 keV for 5 foils in the case of ^{238}U and similar for ^{234}U from 6.46 keV to 33.46 keV (Cosma et al, 2012).

c) Using ^{212}Bi*–*^{212}Po *source and PIPS detector of 1200* mm^2

The experimental conditions were the same as for the previously measurements but we used another set of formvar foils, more bulk as in the first two experiments due to the evaporation of the initial formvar solution. The energy shifts increase from 72 keV, for a single foil used, to 206.1 keV for 5 foils and the resolution decreases from 7.54 keV to 35.42 keV for ^{212}Bi. For 8.85 MeV energy peak of ^{212}Po the increasing of energy shifts were from 41.83 keV to 154.5 keV and the resolution decreased from 13.71 keV for a single foil used to 48.5 keV for 5 foils.

4.3. Uranium alpha spectrometric sources preparation

4.3.1. Designing and optimizing an electrodeposition cell

The design and the development of an electrodeposition device for the uniform plating of uranium which is leak proof and simple in operation, has been reported. The present study was conducted to verify and optimize, as well, the electrodeposition process working conditions. The goal was to find the optimal parameters to be used to achieve uranium alpha sources with good spectral properties and maximum yield in the electrodeposition process. A series of preliminary experiments were conducted to establish the main parameters that have a significant influence on the deposition efficiency.

Designing the electrodeposition device

A new device with two cells working simultaneously, to avoid any possible contamination between the two radionuclides was developed. The electroplating cell was made from a plastic tube of 30 ml as cell body, Figure 4.17. The cell is covered with a spiral shaped copper pipe through which the circulation of water of a constant temperature is done in order to keep deposition at the required temperature. Moreover, at the screw-cap side the thread of the vial was covered with Teflon tape in order to make the cell leak tight. The cell was checked for leakage, by filling it with water. The filling test proves that the vial is wtertight enough to support the operational processes. The new improved electrodeposition system is displayed below.

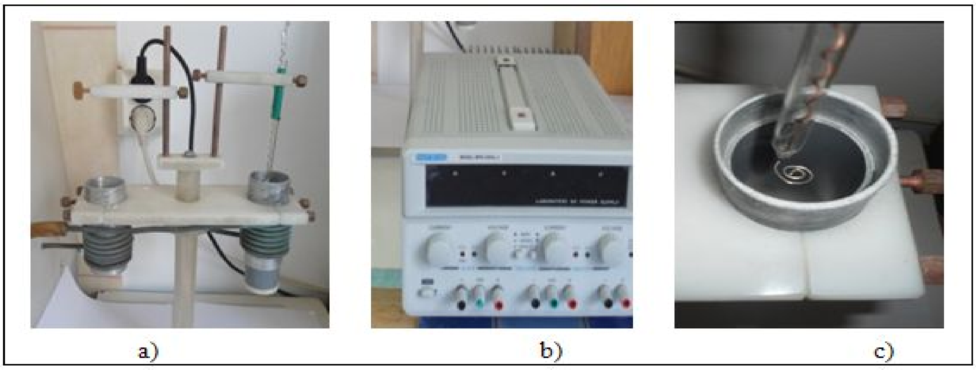


Figure 4.17. a) The improved electrodeposition device with a cooling option and with two cells; b) the power supply; c) the platinum anode.

Sample preparation and electrodeposition

The cell was first tested and evaluated using samples of known activity. The experiments were carried out using the same stock solution prepared by dissolution of uranyl nitrate hexahydrate ($UO_2(NO_3)_2*6H_2O$) in deionised water. Working solutions were prepared by transferring a known weight of uranyl nitrate hexahydrate followed by volumetric dilution to an appropriate working concentration (~1 Bq/ml). This low activity was chosen to be comparable with the level of uranium in environmental samples.

The deposition time plays an important role in the spectral quality and it was varied to examine its effect on uranium yields. Six uranium sources were prepared at different electrodeposition times of 10, 20, 30, 40, 50 and 60 min. We found that for 1 hour electrodeposition time the highest yield of uranium was obtained. We choose this electrodeposition time as a potential optimum for the electrodeposition process conducted in the designed device. Finnaly, the optimum parameters, corresponding to the highest obtained yeld, are presented in Table 4.7.

Table 4.7. The optimal parameters of electrodeposition.

Optimal parameters of electrodeposition	
Anode	Spiral platinum wire
Cathode	Polished stainless steel disk
Electric current	1.5 A
Time	1h
pH of electrolyte	2.3
Volume	25 ml

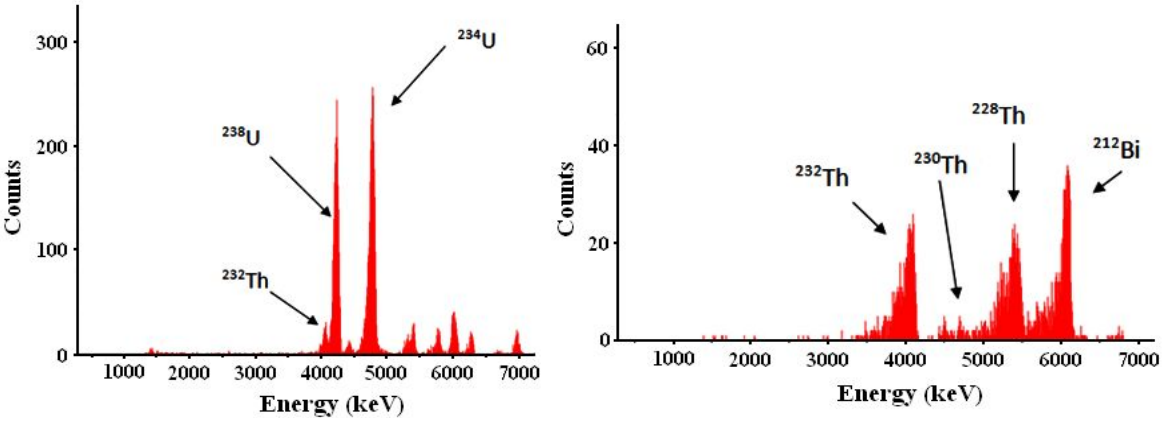
Our determinations prove that the parameters described in Table 4.7 allow the best experimental setting for yield measurement. The energy resolution of the sources must to be taken into account, especially when the activity ratios between different uranium isotopes are to be determined.

4.3.2. Uranium determination from environmental samples

The aim of our work was to develop the source preparation procedures for alpha spectrometry measurements in our laboratory for dating purpose.

Summary of the method: The samples were digested in nitric acid, spiked (^{229} Th/^{233}U), equilibrated by H_20_2 oxidation and boiled for several hours. U and Th were pre-concentrated by scavenger precipitation on hydroxide. Iron was then removed by ether extraction in 9M hydrochloric acid, and U and Th separated by ion exchange chromatography on Dowex 1x8 resin. The purified fractions of U and Th were then electroplated on stainless steel disc (Ivanovich, 1992; Constantin and Lauritzen, 1999; Lauritzen and Onac, 1999; Watanabe and Nakai, 2006; Choukri et al, 2007; Nassef et al, 2008; Labidi et al, 2010).

The procedure described here was used to analyze the content of U and Th isotopes in travertine and coral samples. Besides the very low chemical recover, a very bad resolution of the spectrum was obtained in the first set of measurements; the peaks are overlapped and no calculations are possible. The preliminary results evidenced the fact that the sources must be thinner, in order to avoid the self-absorption. This would lead to a better resolution of the spectrum. After improving each step of the procedure, a significantly better resolution of the spectrum was obtained, and also a better recovery. The peaks can be easily identified. One of the spectra is displayed in the Figure 4.30.



a) b)

Figure 4.30. Alpha spectrum of: a) uranium from travertine; b) thorium from travertine.

4.3.3. Preliminary uranium test measurements by spectrofluorimetry

The present study aims at developing a highly sensitive and accurate method for the determination of uranium concentrations, to be compared thereafter with alpha spectrometric results. The uranium concentration in water samples was determined by fluorimetric method using aliquots prepared from uranyl nitrate stock solution (uranyl nitrate hexahydrate) diluted at different concentrations covering range between 10^{-3} – 10^{-13} g/l. All fluorescence spectra were recorded using an ABLE & Jasco V 6500 spectrofluorimeter with xenon lamp as the source, applying an excitation wavelength of 415 nm and monitoring the emission wavelength over the 460–800 nm range.

Figure 4.34 shows the plot of the fluorescence intensity of UO_2^{2+} against concentration, over the UO_2^{2+} concentration range 10^{-3} g/l to 10^{-7} g/l. As mentioned above, the measurements were performed over the UO_2^{2+} concentration range between 10^{-3} g/l to 10^{-13} g/l. The recorded spectra show that the method is sensitive for samples in the range of 10^{-3} – 10^{-6} g /l.

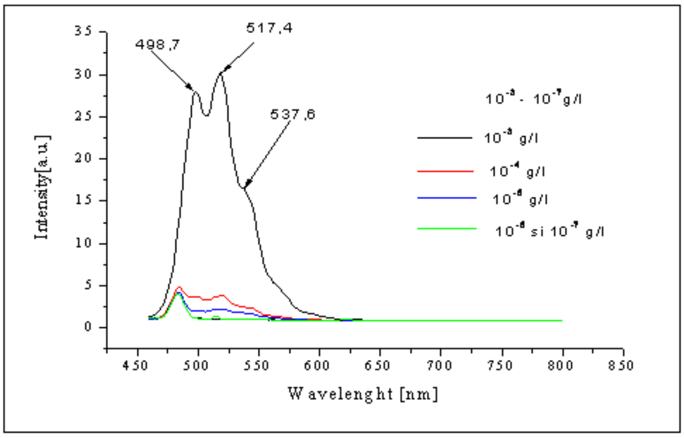


Figure 4.34. Fluorescence spectra of uranyl aqueous solutions in the uranium concentration range of 10^{-3} – 10^{-7} g/l.

4.4. Radium alpha spectrometric source preparation

4.4.2. Determination of ^{226}Ra by MnO_2 coated disc method

The determination of radium by sorption on manganese dioxide coated discs followed by alpha spectrometry is described and our attempt on new substrate for the alpha spectrometry source preparation is presented.

4.4.2.1. Investigation of a new material for radium adsorption

In our study we used different disc material, like polyamide, glass, but the best results were obtained onto a new material based on polyvinyl polymers. Analysis and identification of this polymer material composition was determined by measuring their infrared spectra using a Fourier transform infrared (FT-IR) spectrometer and then comparing the results with a specifically prepared spectral data base (Rusu et al, 2011c).

4.4.2.2. Preparation of MnO_2 coated discs

In our investigations, determination of ^{226}Ra was carried out by adsorption of radium on MnO_2-coated discs prepared as follows: discs of a polyvinyl polymer based substrate of 35mm in diameter were washed successively with ethanol, hydrochloric acid and isopropyl alcohol. After this cleaning process they were immersed under stirring in a $KMnO_4$ solution at 70 °C which causes coating of the disc with a thin MnO_2-layer. The discs were then washed with distilled water, dried in air and further used in the radium sorption experiments. All discs were prepared according to this procedure.

4.4.2.3. Description of analytical procedure

In a typical ^{226}Ra sorption on a thin film experiment the MnO_2 coated discs were dipped in a glass beaker containing the sample. All experiments were carried out taking 50 ml of sample. The amphoteric behavior of manganese dioxide enables to control the adsorption of radium on its surface by regulating the pH conditions. Taking into account this behavior a pH value of 7.5 was selected to ensure a good adsorption of radium. Sorption of radium is performed at room temperature over about 24 h. An exposure time of about 24 hours is sufficient for almost complete sorption of Ra onto the MnO_2-layer of the disc. Following the sorption step, samples were washed with distilled water and left to dry at room temperature. Radium was determined directly by measuring the completely dried disc with α-spectrometer

4.4.2.4. Results and discussions

The influence of the preparation time of MnO_2 films

The influence of the preparation time of MnO_2 films on adsorption yield of radium was studied. Therefore, to determine the optimal time of preparation of manganese dioxide films, measurements at different preparation times, maintaining constant the concentration of $KMnO_4$ solution, were performed. With this purpose, a series of experiments was carried for different preparation times: 30, 60, 90, 120 and 150 min. The graph from the Figure 4.42 indicates that, as an optimum for both, spectral resolution and energy intensity, preparation time of MnO_2 films of two hours is recommended for the determination of Ra isotopes. Therefore, in all measurements we used this preparation time. The influence of preparation time of MnO_2 on radium sorption is presented in the Figure 4.42 (Rusu et al, 2011c).

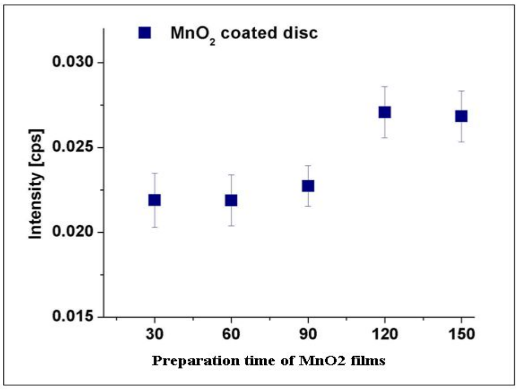


Figure 4.42. Effect of the preparation time of MnO_2 films on adsorption yield of radium.

The spectrum obtained for two hours preparation time is shown in Figure 4.43. ^{226}Ra produces a doublet at 4.602 MeV (5.55%) and 4.785 MeV (94.45%). Also contained in the spectrum are the ^{226}Ra daughters, ^{222}Rn, ^{218}Po and ^{214}Po (Rusu et al, 2011b).

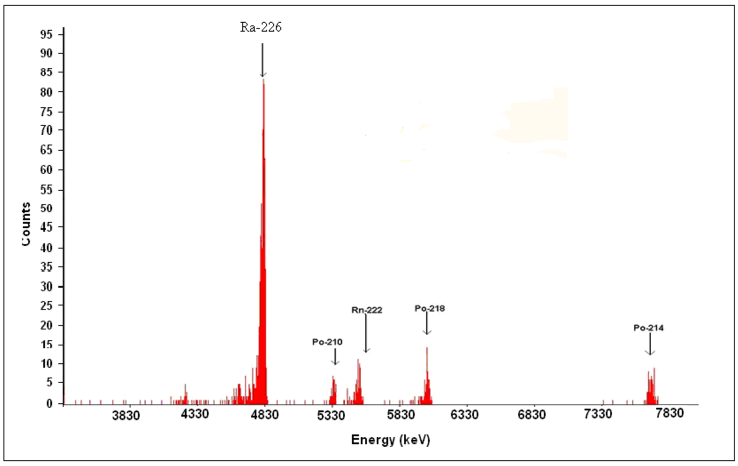


Figure 4.43. The alpha spectrum obtained for 2 hours preparation time of MnO_2 films.

4.4.1. Determination of ^{226}Ra by micro-coprecipitation

4.4.2.1. Description of analytical procedure.

The analytical procedure here consists in co-precipitating radium with lead sulphate, using $BaCl_2$ as a carrier, to separate it from the gross sample constituents. Using a disposable pipette the ^{133}Ba is added as tracer. As ^{133}Ba is no alpha-particle emitter, therefore interferences between tracer and analyzed radionuclides are not created in the alpha spectrum. The pH of solution was changed to 9 by addition of ammonia solution. Then, concentrated (98%) H_2SO_4 sulfuric acid solution was added until the pH was changed to 1. In this situation radium and lead was precipitated as $Ra/PbSO_4$ (Hosseini and Fathivand, 2004). The barium carrier solution is then added and to the beaker and mixed with magnetic stirrers. Precipitation of $Pb(Ba)(Ra)SO_4$ takes place by adding $Pb(NO_3)_2$ solution through a dripper while stirring. The solution is stirred for 2 hours, the beaker is covered with watch glass, allowing the precipitate to settle overnight. The supernatant is discarded and the precipitate is centrifuged for five minutes at 3500rpm (Repinc and Benedik, 2001; Medley et al, 2005).

4.4.2.2. Results and discussions

One of the spectra obtained by mirco-coprecipitation is presented in the Figure 4.52.

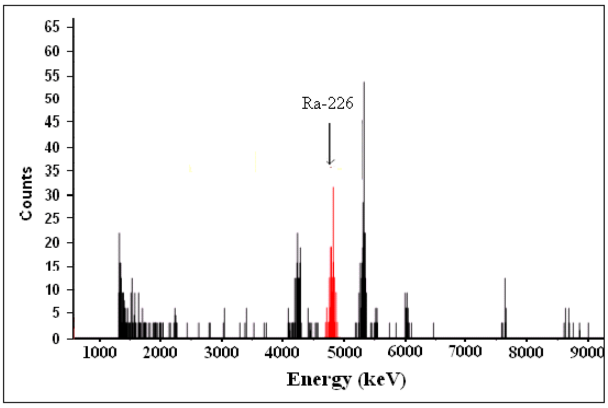


Figure 4.52. Alpha spectrum of radium obtained by micro-coprecipitation method.

Results from alpha spectrometry of samples suggest that micro precipitation, as a sample preparation method is suitable for determination of ^{226}Ra in natural water samples.

4.4.2. Determination of ^{226}Ra by radon emanation technique

Another method used for radium determination from water samples is the radon emanation technique, which is the oldest and the most popular method for determination of ^{226}Ra activity. Our measurements by using this technique have as basic objectives the radioactivity control of the drinking waters according to existing standards and have evaluated the levels of radium in some waters from Romania which are used as potable water reservoirs. The aim of this study was to measure the ^{226}Ra activity concentration in water samples from two important geological sites of Romania. Population worldwide provides their water from surface water and ground water. Ground water is more radioactive than surface water since it passes through rock and soil formations, dissolves many compounds, minerals and radioactive substances (Begy et al, 2012).

Radioactivity measurements in some spring water samples from Romania collected from 2 counties in Romania, Harghita and Bihor, were carried out. Bihor county is famous for its geothermal activity and because of this there can be found many thermal water springs and Harghita county with many post volcanic activity and more than few hundred spring waters.

Table 4.9. Activity concentration of ^{226}Ra [mBq/l] in the measured water samples.

Harghita county		
1	Lóvész	1600 ± 190
2	Csikszentmiklós	650 ± 78
3	Csikrákos	1300 ± 150
4	CsiksomLyó	810 ± 97
5	Csilszépviz	150 ± 18
6	Csikzsögöd	86 ± 10
7	Csikszentkirály	404 ± 48
8	Homoród	54 ± 6

Bihor county

1	Marghita	609 ± 73
2	Chişlaz	127 ± 15
3	Săcuieni	172 ± 21
4	Ciocaia	428 ± 51
5	Sânnicolau de Munte	154 ± 18
6	Chiraleu	226 ± 27
8	Felix 1	367 ± 44
9	Felix 2	2550 ± 306
10	Salonta	87 ± 10
11	Tăşnad	166 ± 20

The results from the table show a range of the value between 54 to 1600mBq/l in Harghita county and 87 to 2550 mBq/l in Bihor county.

4.5. Polonium alpha spectrometric source preparation

A polonium spectrum with ^{209}Po used as spike is shown in Figure 4.59. The spectrum consists of two singlet peaks. Once the count rates in the ^{209}Po and ^{210}Po have been obtained, the background correction must be applied. The net count rates in the two peaks are used to calculate the activity concentration of ^{210}Po in the sample.

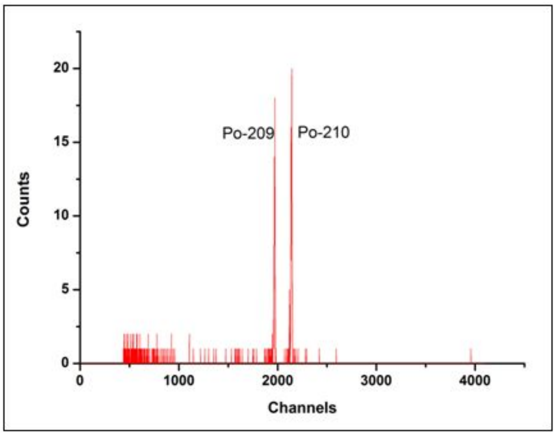


Figure 4.59. Alpha spectrum of ^{210}Po obtained by spontaneously deposition.

5. Activity standardization of beta radionuclides

5.1. Equipments used

This work comprises measurements performed with both methods mentioned above, TDCR and CIEMAT/NIST. As described earlier, for the TDCR method a system with three photomultipliers is used, with this purpose, measurements with Hidex 300SL "Metro" version were performed, and for the CIEMAT/NIST method a system with two photomultipliers was used, namely Packard Tricarb 3100TR. A short description of both systems used is presented below.

5.1.1. Hidex 300SL for TDCR method

Up to now, all TDCR counters used by metrology Institutes for activity determinations have been custom built. Only one commercial company has up to now started the production of TDCR devices. The Hidex 300SL system is the first commercial TDCR counter and is equipped with an automatic sample changer which makes measurements of a large number of samples very convenient. The Hidex 300SL is a liquid scintillation analyzer which uses a three photomultiplier detector design aligned at 120 degrees from each other, enabling triple-to-double coincidence ratio counting. The system is computer controlled and runs with embedded user interface software "Commfiler.csv" supplied by Hidex. This software controls all the instrument functions, as well as providing data processing and analysis. All the measurements were performed in a special counting mode, denotes as „METRO" mode which delivers all the necessary information for applying this methods in a metrological sense. This model is specially designed to meet the requirements on metrology applications.

5.1.2. Tricarb 3100TR for CIEMAT/NIST method

For CIEMAT/NIST method, the measurements were carried out with a Packard TriCarb 3100TR consisting of two photomultipliers operated in coincidence, with linear amplification. It has automatic sample changers, allowing the automatic measurement of a set of LS sources over a long period of time. This instrument is versatile and easy to use, but its main drawback, for their application in precise radionuclide metrology, is the fact that they behave like black boxes and the user cannot control important parameters like the PMT discriminator threshold or the measurement timing (Broda et al, 2007).

5.2. The investigated samples: ^{89}Sr, ^{90}Sr, ^{63}Ni

The radionuclides were selected to cover a wide energy range, therefore measurements were performed with radionuclides with high and low energy of the □ emissions. Samples of ^{89}Sr, ^{90}Sr, ^{63}Ni were measured with both TDCR and CIEMAT/NIST methods and, were it was possible, the results are compared with previously calibrated values. The scintillation cocktail was chosen to be suitable for the chemical composition of the active solution and to give the best compromise in terms of detection efficiency and stability.

Table 5.4. The analysed radionuclides, the different level of quench and the maximum beta energy.

Radionuclide	Quench (µl)	E_{max} (keV)
^{89}Sr	0, 10, 20, 40, 70	1495
^{90}Sr	0, 20, 40, 60, 80, 100	2280
^{63}Ni	0, 20, 40, 60, 80, 100	67

5.3. Activity calculation and interpretation of results

5.3.1. General scheme for data analysis

In the analysis of data, the necessaries corrections were taken into account.

5.3.2. ^{89} Sr by TDCR method - results and discussions

It is to be mentioned that the measurements of ^{89}Sr were performed within the framework of a bilateral comparison with Physikalisch-Technische Bundesanstalt (PTB), Braunschweig, Germany. The samples were prepared at PTB and consist of 15 ml of Ultima Gold Scintillator and 1 ml bidistilled water. Different aliquots (0, 10, 20, 40 and 70 µl) of CH_3NO_2 were added as quenching agent to a set of 5 vials to cover a wider efficiency range. Blank sources were prepared with the same materials and procedures, but replacing radioactive solutions with distilled water.

This work reports the application of both TDCR and CIEMAT/NIST scintillation techniques to the high-energy pure β-emitter, ^{89}Sr, which were measured within the framework of a bilateral comparisons with PTB, Germany. Since the same sources are measured in both laboratories it can be excluded the difference due to different primary standardization or due to different source preparation techniques. Moreover, the performance of measurements with the HIDEX 300SL counter under defined conditions assures that potential differences are not due to different settings.

The efficiency computation was done with a program which has been used in several previous works, TDCR07c.for, provided by LNHB-CEA, France.

5.3.2.1. Influence of coincidence resolving time

Unlike the home-made TDCR systems which used a fixed coincidence resolving time, the coincidence time of the Hidex 300 SL can be selected. Therefore, to find the optimum value of CRT, measurements with different coincidence time of 15, 20, 35, 40, 50, 60, 80, and 100 ns are preformed. The individual results for the activity concentration are then used to calculate a mean value. Then the deviation form the mean value of each activity, Δ (%), is presented versus different CRT in Figure 5.9. From measurements carried out according to various resolving times, the highest difference of about 0.3 % was obtained between the activities calculated at 15 ns and the mean value of the activities for all the coincidence times investigated. A general conclusion can be drawn, that the shorter the coincidence resolving time, the bigger the difference from mean value of the activities for all the coincidence times. A coincidence resolving time of 40 ns appears to be the optimum value for measurements of ^{89}Sr. After setting the CRT 40 ns, measurements were carried out for the reproducibility with an individual duration of counting time of 900s.

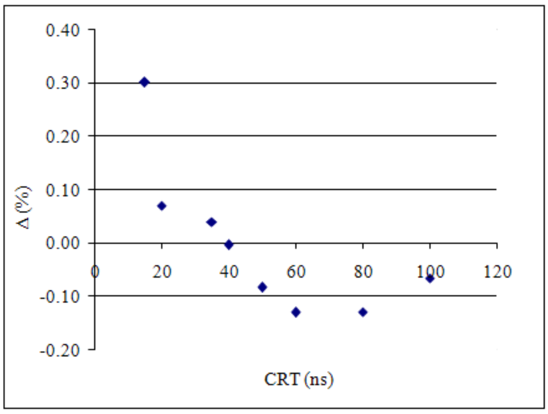


Figure 5.9. The deviation from the mean value of the activity measured at different coincidence resolving time.

5.3.2.2. Influence of kB

Determination of the activity of a source by TDCR involves the best estimation of the ionization quenching parameter, *kB* value. A precise evaluation of the *kB* value is difficult but a best estimate can be obtained by a calculation for various values of the detection efficiency. The program TDCR07c.for gives the required data for a value of kB in a range of 0.007-0.015 cm/MeV. From the plot of the activity versus experimental TDCR in Figure 5.10, is has been shown that for high energy beta pure emitter the *kB* does not have a significant influence.

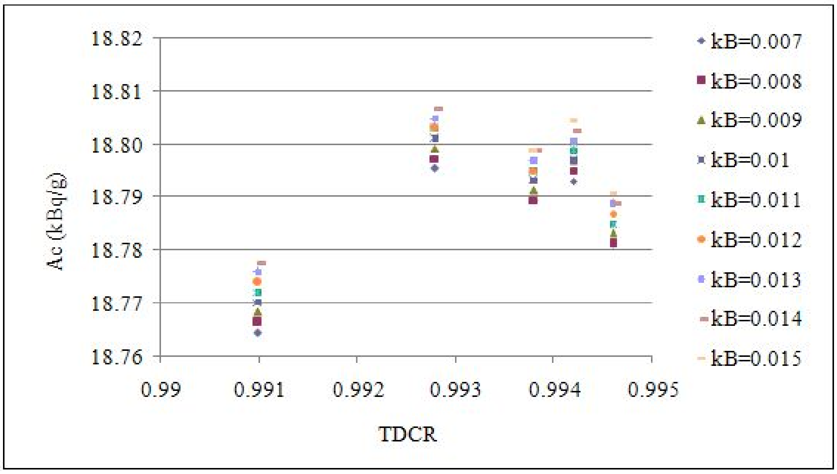


Figure 5.10. Activity versus experimental TDCR at different kB value in the range of 0.007-0.015 cm/MeV.

When using a coincidence resolving time of 40 ns and a *kB* value of 0.0075 cm/MeV to compute the ionization quenching function, the activity concentration of the ^{89}Sr solution on the reference date (2011-11-01, 0 h CET) was found to be (18.78 ± 0.07) kBq g^{-1} (mean value from all the 5 samples).

5.3.3. ^{89}Sr by CIEMAT/NIST method - results and discussions

A confirmatory activity determination was carried out by LS counting with CIEMAT/NIST efficiency tracing. The five LSC sources were repeatedly measured collecting a number of 40 measurements for each of them. Visual Basic CN2004 program was used for calculating the counting efficiency of the ^{89}Sr, where tritium is used as a tracer. The input parameters for the calculations using the CN2004 program are the parameters for beta radiation, the probabilities for all decay pathways and an atomic data part.

Applying the CIEMAT/NIST method, a set of quenched tritium standards sources containing different degrees of chemical quench was measured. As the beta radiation of ^{3}H is of low energy, the characteristics of the equipment and of the sample have a great influence on the efficiency. The calibration curve of ^{3}H provided the counting efficiency and the chemical quench parameter. The program computes the efficiency for different values of free parameter in the range of 1-2.35, form 0.05 step. A value of the Birks constant of 0.0075 cm/MeV was assumed. Second-order polynomials were used for the efficiency point interpolations required by the CIEMAT/NIST method.

The radioactivity concentration of the ^{89}Sr solution, obtained by CIEMAT/NIST method, on the reference date (2011-11-01, 0 h CET) is (18.70 ± 0.07) kBq g^{-1}.

After computing the efficiency and knowing the mass of the samples, the activity concentration of the ^{89}Sr sources was found and expressed in kBq/g. In the Table 5.7 are presented the mass of each sample, the quantity of the quench agent added and also the activity concentration for each source.

Table 5.7. Activity concentration by CIEMAT/NIST method for each source of ^{89}Sr.

Sample No.	Mass (mg)	Activity (kBq)	Activity concentration by CIEMAT/NIST (kBq/g)
2	148.320	2.7751	18.71
3	150.103	2.8069	18.70
4	143.675	2.6867	18.70
5	144.626	2.7045	18.70
6	149.327	2.7909	18.69

An example of the beta spectrum obtained with the Tricarb 3100TR counter is displayed in the Figure 5.13, obtained for the sample 2.

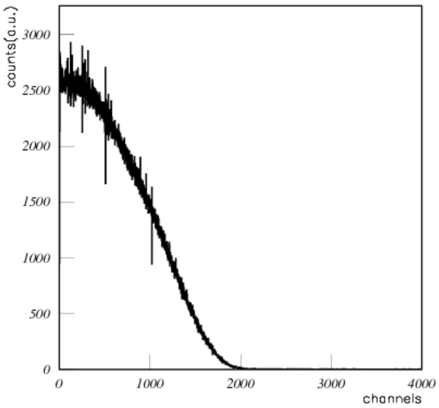


Figure 5.13. Beta spectrum obtained for the sample 2.

The final result, is is the arithmetic mean with of the single result obtained by the CIEMAT/NIST and TDCR method and gives the radioactivity concentration of the ^{89}Sr on the reference date (2011-11-01, 0 h CET) (18.74 ± 0.05) kBq g^{-1}.

5.3.4. ^{90}Sr by TDCR method - results and discussions

The TDCR efficiency calculation technique has been effectively extended to the direct activity measurement of a set of ^{90}Sr samples with different quantities of quench agent. ^{90}Sr decays to yttrium 90 (^{90}Y), which in turn decays by beta radiation so that wherever ^{90}Sr is present ^{90}Y is also present. Due to their respective half-lives ($T_{1/2}$=28.6 years for ^{90}Sr and $T_{1/2}$=64.1h for ^{90}Y), we have considered both radionuclides in the computation of detection efficiency. The data acquisition and data correction followed the same procedure as explained above. Due to its high beta energy, this radionuclide should in principle be capable of being measured to high accuracy with high counting efficiency. The radioactivity concentration of the ^{90}Sr solution was previously standardized by CIEMAT/NIST on the reference date (2011-03-30, 7.00 AM CET) and the standardized value obtained is (8.48 ± 0.04) kBq g^{-1}.

5.3.4.1. Influence of coincidence resolving time

A number of measurements of each source were undertaken at difference coincidence resolving time. This is an important setting, because working with a value too low of coincidence resolving time events can be lost and therefore an incorrect efficiency is obtained. The individual results for the activity concentration are used to calculate a mean value. Similar results with those obtained in the case of ^{89}Sr measurements were found; if the energy of the radiation is high, the detection efficiency is high, so CRT has a very little influence on the detection efficiency. A small deviation was found, which can be attributed to the higher endpoint of this radionuclide which causes a higher number of emitted photons and hence, a higher detection efficiency, leading to a better signal.

5.3.4.2. Influence of kB

The TDCR method requires efficiency variation for the best estimation of the ionization quenching parameter kB value. As mentioned in the case of ^{89}Srmeasurements, the TDCR07c.for program gives the required data for a *kB* value in the range of 0.007-0.015 cm/MeV. In the Table 5.9, the measurements results for one of the sources, are presented as function of the kB value.

Table 5.9. TDCR results vs kB values used for the calculation of the Birks formula.

kB (cm/MeV)	**TDCR**	**ε_D ^{90}Sr**	**ε_T ^{90}Sr**	**Ac (kBq/g)**
0.007	0.9907	0.9919	0.9827	8.44
0.008	0.9907	0.9917	0.9825	8.45
0.009	0.9907	0.9916	0.9823	8.45
0.01	0.9907	0.9914	0.9822	8.45
0.011	0.9907	0.9913	0.9820	8.45
0.012	0.9907	0.9911	0.9819	8.45
0.013	0.9907	0.9910	0.9818	8.45
0.014	0.9907	0.9909	0.9817	8.45
0.015	0.9907	0.9908	0.9816	8.45

As expected, the activity is not significantly influenced by the kB. The detection efficiency decreases by a factor of less than 1% if kB value is changed from 0.007 to 0.015 cm/MeV. If the energy of the radiation is high, the detection efficiency is high, so any little shift in the calibration process has a very little influence on the detection efficiency.

It can be seen that the efficiencies obtained for ^{90}Sr are very similar to those for ^{90}Y. This result is in excellent agreement with the previously certified activity concentration value of (8.48 ± 0.04) kBq g^{-1}, the deviation is lower than 0.8 %.

5.1.1. ^{63}Ni TDCR results and discussions

Additional measurements were carried out with radionuclides with low energy. The nuclide ^{63}Ni decays by pure β-emission having a well-known β-spectrum maximum endpoint energy of $E_{\beta(max)}$ = 66.945 keV. The low-energy beta emitter is difficult to standardize for activity because of the short range of the beta particles. The absolute light emission yield is quite low in LSC, so the expected mean number of photons emitted per disintegration can be very small for low-energy radionuclides. This necessitates collecting light with maximum efficiency. The measurements on ^{63}Ni were carried out on samples which were previously standardized by CIEMAT/NIST and the activity is (12.56 ± 0.07) kBq g^{-1} at the reference date (2011-03-30, 7.00 AM CET).

5.1.1.1. Influence of coincidence resolving time

The measurements were extended to the study of the influence of the coincidence resolving time variation on coincidence counting in the case of low beta energy emitters. For that purpose, measurements with ^{63}Ni were performed. When the number of scintillation photons generated per disintegration is low, as for low-energy β-emitters, coincidence counting becomes sensitive to the time distribution of photons between PMTs. The plots in Figure 3.18 represent the variation of the coincidence counting rates as a function of the resolving time. A significant variation of coincidence counting, much higher than in the case of high energy emitters, is clearly observed when the resolving time was is set at 15 ns, that leads to a difference of about 3%.

5.1.1.2. The influence of kB

Ionization quench function introduces an important correction when measuring low energy □-emitter. Several papers have been carried out to improve the ionization quench corection, by obtainig a good agreement between the experimental counting and the theoretical ones. Ordinarily, one would expect the optimal value of *kB* to be that for which the calculated activity is independent of the value of the TDCR. Different values of Birks' parameter, *kB*, have been reported by various authors. kB parameter is known to be in the 0.007-0.015 cm g/MeV, kB values between 0.011 and 0.014 cm/MeV for most of scintillators have been reported by Broda et al. (2002).

The measurements results for a ^{63}Ni source are presented in Table 5.11, as a function of kB value in the range 0.007-0.0015 cm/MeV. The detection efficiency for low-energy beta emitters varies more greatly with the *kB* than high-energy emitters.

Table 5.11. Activity results for the unquenched sample versus kB value.

Q (µl)	**Mass (g)**	**kB (cm/MeV)**	ε_D	ε_T	**Ac (kBq/g)**	$(a_{TDCR}-a_{CN})/a_{TDCR}$ **(%)**
0	0.0369	0.007	0.832	0.691	12.41	1.13
0	0.0369	0.008	0.829	0.689	12.45	0.82
0	0.0369	0.009	0.827	0.687	12.49	0.53
0	0.0369	0.01	0,824	0.685	12.52	0.25
0	0.0369	0.011	0.822	0.683	12.55	0.01
0	0.0369	0.012	0.820	0.682	12.58	-0.23
0	0.0369	0.013	0.819	0.680	12.61	-0.45
0	0.0369	0.014	0.817	0.679	12.64	-0.67
0	0.0369	0.015	0.815	0.677	12.66	-0.87

In the Figure 5.19, discrepancy between the results that we obtained and the standardized value is plotted for different levels of the quench agent, as function of the different kB values. Our results showed that for a kB of 0.011 cm/MeV the minimal discrepancy was obtained.

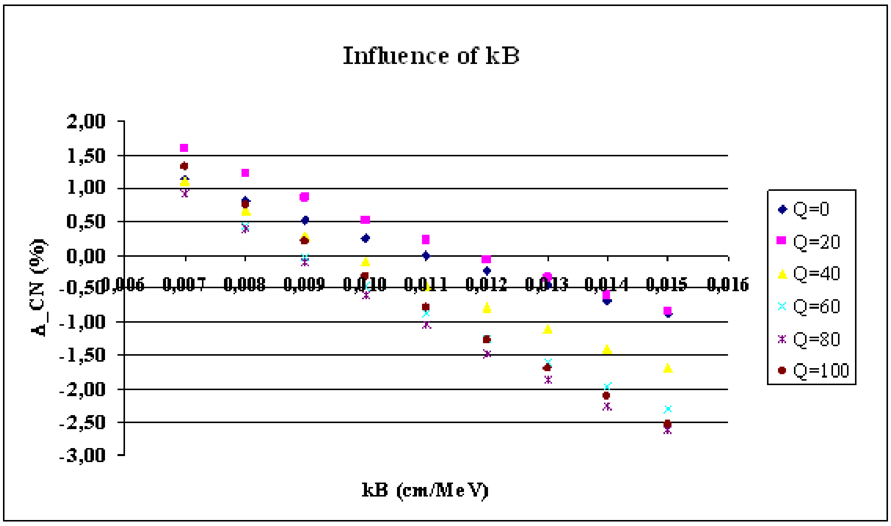


Figure 5.19. Discrepancy from the standardized value for different levels of the quench agent as function of the different kB values.

In the Table 5.12 the activity concentration is computed at 40 ns coincidence resolving time and for kB=0.011 cm/MeV. Under this conditions, the minimum discrepancy between the activity found by TDCR and the standardized value is achieved.

Table 5.12. Results of activity measurements on ^{63}Ni samples as given by the TDCR method.

		Measured CPS			**Extracted from data analysis**			
	Mass (g)	**D (cps)**	**T (cps)**	**TDCR**	**εD**	**εT**	**Activity (kBq/g)**	**(a_{TDCR}-a_{CN})/a_{TDCR} (%)**
1	0.0369	926.71	770.40	0.8309	0.832	0.691	12.55	0.01
2	0.0399	944.39	740.07	0.7833	0.788	0.617	12.53	0.24
3	0.0351	795.46	590.48	0.7419	0.751	0.557	12.61	-0.46
4	0.0395	838.70	576.51	0.6871	0.701	0.482	12.66	-0.86
5	0.0383	763.18	488.77	0.6402	0.659	0.422	12.69	-1.03
6	0.0295	548.52	326.85	0.5957	0.618	0.368	12.66	-0.79

6. General conclusions

As described in this thesis, alpha spectrometry sample preparation, chemical separation and sample mounting involve many steps leading to potential mistakes throughout the process. Our most fundamental concern in obtaining quality results is high recovery and good energy resolution of the spectra. The most important factors in obtaining quality results are proper sample preparation, reproducible radiochemical separation, and mounting techniques that produce thin, uniform samples. Even though the measurements by alpha spectrometry may be time consuming and costly to prepare, they are absolutely necessary in radionuclides determination at low activity. The separation methods developed in this study yielded good chemical recovery for the elements investigated and adequately pure fractions for radiometric activity determination. Attention has been given to avoid losses of the sample.

To prevent alpha detector contamination by recoil nuclides, which is a serious problem in specialized laboratories, the production and characteristics of protective formvar films are described. An improvement in the realization of formvar films consists in using a dip coater system for film deposition onto glass surface. The thickness of the protective films was estimated by using the ASTAR web databases. The influence of formvar film thickness on energy, resolution and detection efficiency has been studied in a wide range of energy using three different sources (a standard source containing ^{239}Pu, ^{241}Am and ^{244}Cm, and ^{238}U – ^{234}U and ^{212}Bi – ^{212}Po sources). The results showed that the alpha spectra characteristics are not significantly influenced by using our home- made films. An additional advantage of using protective films is the possibility to place the source closer to the detector, improving in this way the efficiency. Thus, by using these protective films, one can significantly reduce recoil, while losing very little in terms of performance.

Three of the most significant alpha radionuclides for environmental research and radioprotection, the uranium, radium and polonium isotopes, have been investigated through alpha spectrometry.

For uranium determination, an electrodeposition cell has been designed and developed. After setting the optimal working conditions, the method was successfully applied to coral and travertine samples. Complementary, measurements by spectrofluorimetry have been performed.

The results show a sensitivity of the method to 10^{-7} gU/l, which means that the detection limit is much higher that the one for alpha spectrometry.

Radium has been determined by alpha spectrometry by 2 methods: selective adsorption and micro-coprecipitation. A new material has been identified for the selective adsorption of radium. Besides the simplicity of its radiochemical procedure, a good efficiency of the method was obtained. Due to the thinness of the MnO_2 layer the resolution of the spectra is very good (~23 keV). The results obtained by micro-coprecipitation of radium suggest that this method is more suitable for determination of radium isotopes from water samples.

A number of drinking water have been analysed by radon emanation technique. The radon emanation technique, described here, is a simple and convenient method. ^{226}Ra activity concentration in water samples was determined from two important geological sites of Romania: Bihor county, which is famous for its geothermal activity, and the second site is Harghita county, with many post volcanic activity. The results obtained are in a range between 54 to 1600mBq/l in Harghita county and 87 to 2550 mBq/l in Bihor county.

Polonium has been determined from water samples by spontaneous deposition. A good resolution of the spectrum has been obtained.

In this research, activity concentrations of beta pure radionuclides were determined by TDCR and CIEMAT/NIST methods. Investigations were performed to check the applicability of the only commercial TDCR counter purchased from Hidex Oy (Finland) on measuring beta pure nuclides. The results were found to be satisfactory indicating the usability of the counter for reliable activity measurements with low uncertainties. The results provide valuable information and important tests measurements about potential usage of this counter for other potential users.

Measurements with three relevant radionuclides with high and low energy were carried out. The measurements of ^{89}Sr sources were carried out within the scope of work package 6 of the EMRP project ”Metrology for new generation nuclear power plants” (Metrofission) in a bilateral comparison between National Institute of Ionizing Radiation Metrology INMRI, ENEA Italy and Physikalisch-Technische Bundesanstalt (PTB), Braunschweig, Germany.

The influence of coincidence resolving time on activity determination was investigated and, it was found that, if the energy of the radiation is high, the coincidence resolving time has a very little influence on the detection efficiency. The influence of CRT is more prominent for low beta energy and a significant variation of coincidence counting is observed.

As a consequence of ionization quenching, the model takes also into account the non-linearity of the light emission through the Birks formula, which is a function of the linear energy transfer of the ionizing radiation in the scintillator using the kB factor. The influence of kB was also studied in this work. For beta radionuclides emitting high energy electrons the detection efficiency is close to unity and the influence of the *kB* parameter is not significant. This is not the case for beta radionuclides emitting low-energy electrons, when the calculated counting efficiency is highly dependent on the *kB* value.

LIST OF PUBLICATIONS

This thesis is based on the work contained in the following papers and submitted manuscripts:

1. **O. A. Rusu**, S. Dreve, C. Cosma, R. Cs. Begy, D. C. Nita, L. D. Bobos (2011) “Characteristics of formvar films used to prevent alpha-detector contamination”, *Journal of Radioanalytical and Nuclear Chemistry,* 290 (2), 241-245 (**Impact factor: 1.520**)

2. Cosma, C. **Rusu, O. A**. Cosma, V. Nita, D. Begy, R. Cs. Timar Gabor, A. Astilean, A. (2012) „Protection of alpha spectrometry detectors using thin formvar films and influence on detection characteristics” *IEEE Transactions on Nuclear Science,* 59 (4), 1175 - 1179 (**Impact factor: 1.447**)

3. R. Cs. Begy, S. Dreve, A. Timar-Gabor, **O. A. Rusu**, C. Cosma (2012) „Measurements of radium content in some spring waters from Romania*”, Environmental Engineering and Management Journal,* 11 (2), 247-251 (**Impact factor: 1, 004**)

4. D. C. Nita, **O. A. Rusu**, L. D. Bobos, C. Cosma (2011) „Radiochemical determination of uranium from environmental samples by open tubular liquid chromatography and alpha spectrometry” STUDIA UBB CHEMIA, LVI, 4, p. 41 - 47 (**Impact factor: 0,129**)

5. **O. A. Rusu**, R. Cs. Begy, L. D. Bobos, S. Dreve, C. Cosma „Determination of long-lived radionuclides in water by alpha spectrometry” *Advances in Environmental Sciences,* 3 (2), 148-155 (**BDI**)

6. D. C. Niță, C. Cosma, M. Moldovan, **O. A. Rusu**, B. Papp (2010) “Masuratori de radon in sol in zona Clujului” *Environment. & Progress* 14, p. 159-163

7. **O. A. Rusu,** R. Cs. Begy, D.C. Nita, L.D.Bobos, C. Cosma (2011) „Alpha spectrometry using MnO_2 coated disc on the polyvinyl polymer substrate” *Földkérgi radioizotópok a Környezetünkben,* Veszprém, Hungary, ISBN 978 615 5044 05 2, pp. 65-71

8. **O. A. Rusu**, R. Cs. Begy, D. C. Niță, C. Cosma „Determination of radium from environmental samples using manganese dioxide coated discs followed by alpha spectrometry”, *Ecoterra, Journal of Environmental Research and Protection* 25, ISSN 1584-7071, p. 175-180

9. C. Cosma, **O. A. Rusu**, V. Cosma, D. Nita, R. Begy, A. Astilean ”Protection of alpha spectrometry detectors using thin formvar films and influence on detection characteristics”

Proceedings on Radiation Effects on Components and Systems, RADECS 2011, pp. 729 – 733, ISSN: 0379-6566, Print ISBN: 978-1-4577-0585-4

10. M. Capogni, **O. A. Rusu**, “Tecniche a scintillazione liquida a confronto nella misura dell’attività dello ^{90}Sr”, submitted and accepted for oral presentation to the XXXV Congresso Nazionale di Radioprotezione of the Italian Association of RadioProtection (AIRP) that will be held in Venice on 17th-19th October 2012

11. R. Cs. Begy, J. Somlai, T. Kovacs, **O. A. Dumitru (Rusu)**, C. Cosma “The activity concentration of ^{210}Po in romanian commercial cigarettes and the radiation exposure estimation derived from their regular consumption” Submitted Manuscript at *Radiation Protection Dosimetry*