Abstract

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with title:

STUDIES ON THE APPLICATIONS OF THE TRIPLE-TO-DOUBLE COINCIDENCES RATIO METHOD FOR PRIMARY ACTIVITY STANDARDIZATION USING LIQUID SCINTILLATION COUNTING

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CONTENTS

ACRONYMS

SI	Système International d'unités/International System of Units
TDCR	triple-to-double coincidences ratio
LS	liquid scintillation
PMT	photomultiplier tube
EC	electron capture
FOM	figure of merit
C-TDCR	Compton coincidences and TDCR
ZoMBieS	zero model by using coincidence scintillation
CSET	Compton spectrometer efficiency tracing
CNET	CIEMAT/NIST efficiency tracing[1]
CdTe	cadmium telluride
CDT	common dead-time
IDT	individual dead-times
FWHM	full width at half-maximum

1 The name comes from the two institutions that worked on the method – CIEMAT (Spain) and NIST (USA).

FPGA	field-programmable gate array
CFD	constant fraction discriminator
MAC3	module d'acquisition de coïncidences triples
nanoTDCR	front-end digital pulse processing device for TDCR counting[2]
RCTD1	the French primary TDCR counter[3]
TDCR-SU	TDCR counter developed and operated at Sofia University
ICRU	International Commission on Radiation Units
DDEP	Decay Data Evaluation Project
BIPM	The International Bureau of Weights and Measures
LNHB	*Laboratoire National "Henry Becquerel"*
MIL	"Metrology of Ionizing Radiation" laboratory at Sofia University

2 It is developed by Valentin Jordanov from the labZY company.
3 The acronym comes from the french *rapport de coïncidences triple à double* meaning triple to double coincidences ratio.

1

ABSTRACT

METROLOGY is the science of measurement, embracing both experimental and theoretical determinations at any level of uncertainty in any field of science and technology[1]. It has its roots as far back as Ancient Egypt, where the success of the large construction projects of the time relied on consistent measurements. This was achieved by standard rods of equal length – the *royal cubit*, the earliest attested standard measure for length.

Throughout history the need to synchronize measurements was always present, and so, many local standards and units appeared and disappeared all over the world. However, up until the 18$^{\text{th}}$ century there was no universal measurement system. This was changed in 1795 with the creation of the decimal-based metric system in France by the law "on weights and measures". It followed the newly formed definition of the *metre* as the length of one ten-millionth part of the distance from Earth's equator to its north pole – the first unit which in its determination was neither arbitrary nor related to any particular nation on the globe. The adoption of the metric system

1 Source: BIPM website (Archived)

expanded with time, and, to ensure conformity between the different countries, the The International Bureau of Weights and Measures (BIPM) was formed [1].

The metric system has evolved into the International System of Units, abbreviated as SI from the French *Système International d'unités*. It consists of seven base units, which are the second (time, s), metre (length, m), kilogram (mass, kg), ampere (electric current, A), kelvin (thermodynamic temperature, K), mole (amount of substance, mol) and candela (luminous intensity, cd). As of 2019, the magnitude of all SI units are defined by exact numerical values for seven constants when expressed in terms of the corresponding SI units. These are the speed of light in vacuum c, the hyperfine transition frequency of caesium $\Delta\nu_{Cs}$, the Planck constant h, the elementary charge e, the Boltzmann constant k, the Avogadro constant N_A, and the luminous efficacy K_{cd}. Except the seven base units, there are 22 derived units with special names and symbols, which can also be used. One of these units is the unit for radioactivity – the becquerel Bq. It corresponds to the mean number of radioactive decays per unit time of a radioactive source. One becquerel is equal to one decay per second [1].

The becquerel can be regarded a special unit, due to the nature of the radioactive decay, which is the process by which unstable or excited nuclei lose energy by emission of one or more subatomic particles. There is a large variety of possibilities for the type of the emission resulting from a radioactive decay: β^+ and β^-, α-particles, neutrons, fission products, X-rays and γ-rays, Auger electrons and neutrinos. Some radioactive decays result in a single emission and some in a cascade of emissions, each of a different type. Of the possible particles, there are some that have electric charge and some that are neutral. Their masses span from the massless photons and almost-massless neutrinos to the heavy α-particles and even heavier fission products. The effect of the emission on the surrounding medium can be felt from micrometers to hundreds of meters. Moreover, there is a huge variety in the radioactive isotopes themselves – from the lightest elements to the heaviest nuclei – which can be found in numerous chemical compounds and in all states of matter. For these reasons, there could be no single universal realization of the standard for the becquerel unit. Thus, the standardization techniques are usually specifically tailored to the radionuclide of interest.

One largely used measurement platform, however, is liquid scintillation (LS) counting since many radionuclides can be incorporated into a mixture with a liquid scintillator. This provides a 4π geometry for detecting the emitted radiation and significantly increases the detection efficiency. The exact variant of liquid scintillation detection technique used for primary standardization depends on the properties of the radionuclide. For γ-ray emitting radionuclides, coincidence methods can be used to determine the activity. For pure β, electron capture (EC) or mixed decays, the triple-to-double coincidences ratio (TDCR) method is applied, which uses ratios of measured coincidences to produce robust results. This is precisely the main focus of this thesis – the primary standardization of radioactivity using the TDCR method, what are some well known or newly discovered aspects of it, what difficulties are encountered and how they can be solved.

1.1 STRUCTURE OF THE DISSERTATION

The thesis is structured in three parts and twelve chapters. The first part serves as an overview of the field of radionuclide metrology using LS counting. The second and third parts contain the original work in the scope of the thesis. A short overview of the contents of each part is as follows:

1.1.1 *Overview of Part I*

In Part I, an overview of liquid scintillators and liquid scintillation counting in general are given. The emphasis is placed on radioactivity standardization methods and the TDCR method in particular.

1.1.2 *Overview of Part II*

Part II focuses on the time domain in LS counting and the distribution of the time intervals between detected scintillations in a LS detector in particular. First, a program, that has been developed for the analysis of digitizer list-mode data, is presented in Chapter 3. The following Chapter 4 describes a comparison of two algorithms to process coincidences and deadtimes in a detector with three photomultiplier tubes (PMTs). Chapter 5 contains a derivation of the statistical distribution of the time intervals between detected prompt fluorescence events. The derived equations were verified using a dedicated Monte Carlo code, developed in the framework of the thesis and presented shortly in Appendix A. It is shown how this distribution could be used to estimate the detection efficiency in LS measurements using the free parameter model. The next chapter, Chapter 6, deals exclusively with experimental studies on the derived equations and their application to real measurements. The possibility to use the newly developed theory to supplement standardizations using the TDCR method is discussed. Some applications of the time interval distributions to routine LS counting measurements are also shown.

1.1.3 *Overview of Part III*

Part III focuses on how the information from the distribution of the time intervals between detected events can be used in radionuclide metrology and the TDCR method in particular. Chapter 7 presents methods for precise measurements of the half-life of some nuclear excited states, namely the long-lived states in ^{57}Co and ^{237}Np, using LS counting and analysis of the resulting time distributions. The time information from LS measurements is utilized to develop an experimental method to evaluate the counting rate of accidental coincidences in a TDCR detector is presented in Chapter 8. Besides the experimental method, theoretical equations to evaluate the accidental coincidences are also developed and verified in the same study. Chapter 9 deals with the influence of the delayed fluorescence on the activity calculated by the TDCR method. The studies demonstrate the problems encountered in the standardization of low-energy emitters such as ^{3}H and ^{55}Fe. Chapter 10 shows the development and characterization of a new Compton coincidences and TDCR (C-TDCR) system for primary standardization of LS samples. The system was used to study the response of commercial liquid scintillators to electrons with energies in the range from 2 keV to 8 keV. The possibility to use this information in ^{3}H standardizations is also discussed. Chapter 11 outlines some practical applications of the TDCR method which are used in the "Metrology of Ionizing Radiation" laboratory at Sofia University to ensure metrological assurance of LS measurements.

Finally, Chapter 12 contains a brief summary of what has been presented in the thesis and highlights the important conclusions of the work. Some future directions for study are also stated. Additionally, some complementary information and measurements are shown in three appendices placed at the end of the thesis.

2 SUMMARY OF THE RESULTS IN PART II

The main focus of the second part of the thesis falls on the studies of the distribution of light detection events.

2.1 DEVELOPMENT OF THE LIST_MODE_ANALYSIS SOFTWARE

The LIST_MODE_ANALYSIS software for the off-line processing of CAEN digitizer data was presented in Chapter 3. The main contribution of the code is the definition of the time interval distributions not only for two channels in coincidence, but also for the logical sum of double coincidences and for the triple coincidences. There are three main modes of operation of the LIST_MODE_ANALYSIS program:

1. obtain the single and coincidence counting rates in the various channels
2. obtain the time distribution of events within a coincidence channel
3. obtain the counting rates in all channels as a function of the energy deposited in the γ channel.

Besides the three main modes of operation, the program has a number of options that can be used to filter specific events or to manipulate the input and output. The software does not have a graphical user interface and it is operated within the command line with a set of options. A full list of the options is given in the Appendix in listing C.1. The source code of the program is published on https://gitlab.com/cdutsov/cdt_logic.

2.2 COMPARISON OF TDCR COUNTING ALGORITHMS

Another main feature of the program is that all signals that are considered are subject to the same coincidence window and dead-time logical rules. This includes the case when the time distribution histograms are constructed. The dead-time logic that was chosen for the LIST_MODE_ANALYSIS code is the common dead-time logic. Its performance was compared to the recently proposed individual dead-times counting algorithm. The comparison was presented in Chapter 4. The individual dead-times (IDT) logic seems to lead to a reduced dead-time of the system when measurements of high activity ^{3}H sources are performed. It also shows very similar performance as the common dead-time (CDT) in ^{63}Ni, ^{14}C and ^{90}Sr/^{90}Y measurements for activities in the range of 2 to 3 kBq. While having some benefits, the improvements in performance of the IDT logic seem to be at the expense of misclassification, in rare cases, of T events as D events. This was especially evident in ^{222}Rn measurements, where the ^{222}Rn activity obtained with IDT counting rates is 0.4% higher than with CDT counting rates. The results of the comparison were published in the journal *Applied Radiation and Isotopes* [2].

The comparison of the counting algorithms was performed also on artificially generated time sequences of events using a Monte Carlo code, which samples the timing of detected scintillations from a pre-measured time interval distribution. This code also served as an inspiration to develop another Monte Carlo simulation program that uses physical assumptions about the time dependence of the scintillation light of the prompt and delayed fluorescence and of the time response function of a LS detector. The latter code, described in Appendix A, outputs digitizer-like list-mode data that can be analyzed by the LIST_MODE_ANALYSIS program. The time distributions that were obtained from Monte Carlo simulation data were compared to real measurements and an excellent agreement was obtained.

2.3 THE TIME INTERVAL DISTRIBUTIONS IN LS COUNTING

The exploration of the time interval distributions were continued further in chapters 5 and 6. Firstly, an analytical model of the cross-correlation distribution of prompt fluorescence was developed. The various steps in the derivation were validated through comparisons with the Monte Carlo code. The final equation gives identical results as the Monte Carlo simulation in the case of a nuclide with a given energy spectrum, a LS cocktail with a given prompt decay constant and a detector system with given relative PMT efficiencies, static delays and standard deviations of the Gaussian time jitter.

As the analytical equation depends on the figure of merit (FOM) of the measurement, it was postulated that it could be used to fit experimental data and be used to obtain the detection efficiency of a two PMT system. However, the complexity of the theoretical equation required a fast computer code for its calculation. Such code was developed in the framework of this thesis, and it is presented in Appendix B. The code was optimized to be as fast as possible, and it can be used to fit measurement data with the analytical equation in order to extract the underlying parameters.

The main result in this part is the study of the applications of the cross-correlation distribution. It was shown that the height of the distribution could be used to calculate the mean number of detected photons, if the parameters of the scintillator and detection system are known. Attempts were made to fit the analytical cross-correlation distribution directly to experimental data. Very good results were observed for ^{3}H and satisfactory results were observed for ^{14}C. These studies demonstrate that if the decay constant of the prompt component of the scintillator is known, it is possible to estimate the FOM of the measurement, and from it the detection efficiency. These results are an important first step towards the use of the cross-correlation distribution for primary activity measurements.

The use of cross-correlation data in routine LS analysis was also explored. It was shown that the height of the experimental distribution depends linearly on the FOM for the low-energy emitters ^{3}H and ^{55}Fe. This would allow to record the height as a function of the FOM for a calibration source and then use this information in subsequent measurements of unknown sources. The relationship between the height scaled to the FWHM and the mean number of detected photons was shown to be linear for all studied nuclides. The possibility to characterize the distribution using its kurtosis was also explored by plotting it with respect to the mean number of detected photons. The results show that, under certain conditions, the same linear relationship is observed for ^{3}H, ^{63}Ni and ^{14}C when in the same LS cocktail.

2.4 CHALLENGES

There are several challenges in front of the successful application of the cross-correlation method for primary activity standardization by LS counting. Perhaps the largest obstacle is in the form of the delayed scintillation component. Thus far there does not seem to be a reliable model of its timing properties and attempts to describe it include many unknown parameters. Moreover, it is diffusion controlled and its emission rate depends on the temperature of the scintillator. As will be shown in Chapter 9, the delayed fluorescence is also problematic for the TDCR method. In this regard, it would be beneficial to develop LS cocktails with less pronounced delayed component.

Another challenge would be that the rise time of the prompt scintillation component is not included in the analytical model. For some cocktail it is possible that this rise time is a significant fraction of the decay time and would influence the overall shape of the cross-correlation distribution. The inclusion of another exponent to describe the scintillation rise time can be done, but it would not permit the analytical evaluation of the cross-correlation between the responses of the two PMTs.

From experimental point of view, high-quality PMTs have to be used for cross-correlation measurements, i. e., high quantum efficiency and single photon sensitivity. In addition, matched PMTs with similar efficiencies and timing response would be preferable. The analysis of the time of arrival of PMT signals requires fast digitizers with constant fraction discrimination or fast circuits with timing resolution better than 200 ps. The time-response properties of the system, described by λ, μ and σ in the cross-correlation distribution $D(\Delta t; \varphi, \lambda, \epsilon_A, \epsilon_B, \mu, \sigma)$, have to be carefully characterized.

The parameters μ and σ are specific to the detector system and depend on the PMTs and the associated electronics. The decay constant λ is a property of the LS sample. Unfortunately, the decay constant λ and the figure of merit φ cannot be determined from a single fit of the cross-correlation distribution because they are strongly correlated. The solution is to use additional measurements, as it is done in this work, or to use other techniques, such as time-correlated single photon counting, to determine the decay constant λ. The latter option is very promising as it allows completely independent determination of λ for a particular LS sample and thus will largely facilitate the determination of the FOM. This approach will be explored in future studies.

From a theoretical point of view, a challenge in front of the cross-correlation method as a method for primary activity measurement is that it determines the mean number of detected photons or the FOM. In order to go from detected photons to detection efficiency one is obliged to go through the free parameter model. Thus, the cross-correlation method inherits all the deficiencies of the free parameter model, such as the unknowns in the light output as a function of the deposited energy. The same problems, however, apply to all other currently used methods in radionuclide metrology using LS counting, e. g., TDCR and CIEMAT/NIST efficiency tracing (CNET). In this regard, it is possible that the cross-correlation method gives new information that could be used to reduce the uncertainties associated with the free parameter model.

Finally, a full uncertainty budget of the detection efficiency derived from the cross-correlation method should be established for the application of this method in radionuclide metrology. Currently, only the fit uncertainty was considered, but in order to be precise, many other contributions would have to be considered, such as: the effect of the digitizer's finite sampling rate, the walk of the constant fraction discriminator, the influence of temperature on the decay constants, the influence of delayed fluorescence, uncertainty in the β-spectrum, uncertainties associated with the non-linearity of the light output of the scintillator with respect to the deposited energy and possibly others.

The challenges in front of the application of cross-correlation data to more routine LS measurements seem to be quite fewer. The preliminary results described in this part seem to suggest that the height of the cross-correlation distribution could be used as a proxy for the detection efficiency if the detection system is calibrated beforehand.

3 SUMMARY OF PART III

The main focus in the second part of the thesis falls on the studies of the applications of the time distribution of scintillations, and digital list-mode acquisition in general, to radionuclide metrology using LS counting.

3.1 MEASUREMENT OF THE HALF-LIFE OF NUCLEAR STATES

Chapter 7 explores the possibility for precise measurements of short half-lives of nuclear isomeric states using LS counting. The half-lives of two excited nuclear states in ^{57}Fe and one in ^{237}Np were measured by means of LS-LS and LS-γ coincidences. The half-life of the 59.54 keV level of ^{237}Np obtained by LS-γ coincidences is 67.60(25) ns, which is in good agreement with already published decay data. The measured half-life of the 14.4 keV level of ^{57}Fe is 97.9(3) ns which is consistent with some of the published results, however a significant discrepancy exist between the values found in the literature. The half-life of the shorter-lived 136.47 keV level of ^{57}Fe was measured also by LS-LS coincidences and the result is 8.78(4) ns.

The result is consistent with the published data and comes with a significant improvement in the uncertainty.

The studies presented in Chapter 7 show that liquid scintillation fast timing measurements can be a useful tool to refine some already known decay times. The LS-LS coincidences method could allow more precise studies of half-lives of excited states of short-lived nuclides, where long measurements are difficult to perform. It could be valuable also when measuring isotopes in liquid phase. The studies show that LS-LS coincidences could be used for precise measurements of half-lives as short as 8 ns.

3.2 EVALUATION OF ACCIDENTAL COINCIDENCES IN TDCR

In Chapter 8, an experimental method to evaluate the counting rate of accidental coincidences in TDCR measurements was proposed. The method was used to derive and validate analytical expressions that can be used to directly calculate corrections for random coincidences with data from standard TDCR systems. The work was published in the journal *Nuclear Instruments and Methods, Section A* [2].

Off-line analysis of TDCR measurements allowed the development of an experimental method to evaluate the counting rate of accidental coincidences. These studies were central to the derivation of a set of analytical equations that can be used to evaluate random coincidences in any existing TDCR counter. The analytical method developed in this thesis was adapted in the extended international reference system for pure β-emitting radionuclides operated by the BIPM [3].

The analysis of the list-mode data shows that the analytical and experimental evaluations provide comparable results for ^{3}H, ^{55}Fe and ^{14}C measurements. The contribution of random coincidences increases with reduction of the counting efficiency. The counting rate of true coincidences does not seem to depend significantly on the choice of coincidence resolving time for the studied 23 kBq ^{3}H LS-source if the analytical approach is used.

Both methods were used to evaluate corrections for accidental coincidences for Monte Carlo generated ^{3}H measurements with activities from 2 kBq to 200 kBq. The correction according to the analytical method is consistent with the Monte Carlo reference to within 0.29% up to 100 kBq. The experimental method deviates less than 0.06% from the reference counting rates up to 60 kBq and less than 0.21% up to 200 kBq.

A note should be made here, that the corrections for accidental coincidences should be applied not only to the measurements of the radioactive source, but also to the measurements of the blank sample that is used for background correction. Due to the usually low counting rates in the coincidence channels of the blank sample, the single counting rates are relatively high. For example, a typical blank measurement with a TDCR detector at Sofia University has 12 cps in the double coincidences channels and around 400 cps in the single channels. Therefore, the contribution of the accidental coincidences to the double counting rates is significant.

The analytical equations can be used to evaluate the rate of accidental coincidences in measurements performed with most existing TDCR acquisition systems, the only requirement being that they output the single, double and triple coincidence counting rates. The correction for random coincidences provides the opportunity to study long coincidence windows. The corrected time distributions indicate that the use of short resolving times leads to a loss of coincidences in low-energy radionuclides. The accidental coincidences corrections will improve the non-linearity of TDCR systems used for studies of short half-life medical nuclides line ^{11}C, ^{15}O or ^{18}F. They will allow TDCR measurements of sources with high activities, where the contribution of accidental coincidence cannot be neglected.

3.3 DELAYED FLUORESCENCE AND TDCR COUNTING

In Chapter 9 it is shown that both the detected coincidences and the TDCR estimated activities depend significantly on the choice of coincidence window. This dependence is attributed to the unequal loss of double and triple coincidences as well as to the different ionization quenching properties of the delayed scintillation component. Studies of Monte Carlo generated data shows that if the used cocktail has a non-negligible delayed fluorescence contribution, then, even for short coincidence resolving times, the activity of a low energy β-emitter would be overestimated. Despite the developed corrections for accidental coincidences in TDCR counting, the obtained results show that it is not yet advisable to use longer coincidence resolving times in TDCR measurements of low energy β-emitters. A primary method, different from the TDCR method, would be needed to measure the activity of a sample, so as to have a reference value. A good candidate for such a method is the Compton coincidences and TDCR method discussed in Chapter 10.

3.4 FIRST RESULTS WITH A NEW COMPTON-TDCR SYSTEM

The characterization and first results with a new C-TDCR system are shown in Chapter 10. The detector was designed in such a way as to allow changing the angle between the external source and the detector, thus changing the useful energy range of Compton electrons. An excellent agreement between the experimentally obtained scintillation response at 40° and at 90°. This is a good indication that the system is not biased towards a certain PMT. The system was used to measure the light output as a function of deposited energy for three commercial cocktails – UltimaGold, UltimaGold LLT and HionicFluor, and a home-made Toluene + PPO cocktail.

The response of the cocktails was approximated with Birks' ionization quenching formula and a good agreement was observed. The measurements were made also with a set of grey filters, which allows to fix the kB parameter in the ionization quenching function more precisely. The results, however, lead to a much larger kB parameter than is commonly used in the TDCR method and what is found by the efficiency variation technique.

The C-TDCR system was also used to perform a comparison between three primary activity measurement methods – TDCR, Compton spectrometer efficiency tracing (CSET) and C-TDCR. The activity obtained by the C-TDCR method was evaluated with four different assumptions for the function describing non-linearity of the light output of the scintillator in order to estimate the uncertainty on the calculated detection efficiency. The comparison between the three methods shows a large discrepancy between the TDCR activity at the kB parameter found by efficiency variation with grey filters and the C-TDCR activity. The results confirm the expectations laid out in Chapter 10, that the kB parameter could be underestimated by the efficiency variation technique due to the influence of the delayed fluorescence.

3.5 APPLICATIONS OF TDCR AT SOFIA UNIVERSITY

Chapter 11 presents the validation of the TDCR system developed and operated at Sofia University through a participation in an international key comparison of ^{3}H activity measurements organized by the BIPM. It is also concerned with the validation of the front-end digital pulse processing device for TDCR counting (nanoTDCR), a TDCR acquisition device developed by the labZY company. The chapter also describes shortly some of the practical applications of the TDCR technique for the purposes of the studies performed at the "Metrology of Ionizing Radiation" laboratory at Sofia University (MIL), namely the standardization of ^{222}Rn activity. Overall, Chapter 11 summarizes some applications of the TDCR method for calibration and quality assurance of the activity measurements performed in the MIL laboratory at Sofia University.

4 FINAL REMARKS

In conclusion, the cross-correlation distribution, that describes the time intervals between the first detected events in each PMT within a given coincidence window, has been derived. The feasibility of using the derived theoretical model to fit experimental cross-correlation distributions and extract information about the FOM was evaluated. The preliminary results show that, if the decay constant of the scintillator is known, it would be possible to calculate the FOM and, from there, the activity of the sample. This constitutes a new approach at primary activity measurements using LS counting.

Perhaps the largest contribution of the thesis to radionuclide metrology using the TDCR method are the derived corrections for accidental coincidences. They can be used in any existing TDCR LS system that reports the single and coincidence counting rates. The corrections allow the measurement of radioactive sources with high activity, which can be encountered when performing *in situ* measurements of medical radioisotopes. Removing the influence of accidental coincidences gives the possibility to use long coincidence resolving times and study the time dependence of delayed fluorescence, which was shown to have a non-negligible effect on the TDCR calculated activity.

The studies of the influence of the coincidence window on TDCR measurements showed that, for some LS cocktails, there is a significant dependence of the calculated activity on the resolving time. Moreover, as the influence of delayed fluorescence depends also on the detection efficiency, the efficiency variation technique was shown to be potentially unreliable to determine the kB parameter of the ionization quenching function. This was emphasized by a discrepancy that was observed between the kb parameters obtained with efficiency variation with grey filters, as well as with chemical quenching.

As a possible solution to the problems related to the uncertainties regarding the light output of the scintillator, a Compton coincidences and TDCR system was investigated. It was shown that the non-linearity of the scintillator response can be studied in the interval between 2 keV and 8 keV energy of Compton electrons. The light output of several commercial scintillation cocktails was obtained and the optimal kB parameters in all cases were significantly higher that what is usually used in TDCR measurements. This indicates that conventional TDCR measurements of low-energy emitters such as ^{3}H and ^{55}Fe could underestimate the activity of the sample by as much as two or three percent. Perhaps, the accuracy of the standardization of low-energy emitters can be improved by the use of the Compton coincidences and TDCR technique.

Within this work, it was shown that LS counting and off-line data analysis can be useful tools for the study of half-lives of nuclear excited states as short as 8 ns. The half-lives of the long-lived excited states in ^{57}Fe and ^{237}Np were measured by LS-LS coincidences with 0.4% uncertainty.

Some applications of the TDCR method for calibration and quality assurance of the activity measurements performed in the MIL laboratory at Sofia University were shown.

A program called LIST_MODE_ANALYSIS was developed to analyze CAEN digitizer data, and it was used throughout the thesis. A Monte Carlo code to simulate the timing of detected scintillation events in a three-PMT detector was also developed. The code was used to validate a theoretical model of the timing of the prompt fluorescence. A fast software for the calculation of the model was developed in order to be able to fit the equations to real-world data.

CONTRIBUTIONS TO NEW KNOWLEDGE IN THE FIELD OF ACTIVITY MEASUREMENTS USING LS COUNTING

- Methods for the evaluation of accidental coincidence counting rates in TDCR measurements were proposed for the first time [P1].

- The cross-correlation distribution of the time intervals between detected scintillation events was derived, and it was shown how it can be used to determine the detection efficiency and activity of the sample [P2].

CONTRIBUTIONS TO METHODICAL ASPECTS IN THE FIELD OF LS COUNTING

- The effect of the delayed fluorescence on the activity calculated using the TDCR method has been demonstrated [P3].
- A comparison of two counting algorithms for TDCR measurements has been performed [P4]. The comparison demonstrates their advantages and disadvantages.
- The possibility to use digitized LS measurements for the precise determination of the half-life of some excited nuclear states has been demonstrated [P5].
- A software for the analysis of digitizer data has been developed. It has applications for TDCR measurements, cross-correlation measurements and C-TDCR measurements.
- A Monte Carlo code has been developed that gives the possibility to simulate the time interval distribution and the number of detected scintillation events in LS measurements.

PUBLICATIONS

PUBLICATIONS ON THE TOPIC OF THE THESIS

[P1] C. Dutsov, P. Cassette, B. Sabot, and K. Mitev. "Evaluation of the accidental coincidence counting rates in TDCR counting". In: *Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 977 (2020), p. 164292. DOI: 10.1016/j.nima.2020.164292 (cit. on p. 25).

[P2] Krasimir Mitev, Chavdar Dutsov, Philippe Cassette, and Benoît Sabot. "Time-domain based evaluation of of detection efficiency in liquid scintillation counting". In: *Scientific Reports* 11 (2021), p. 12424. DOI: 10.1038/s41598-021-91873-1 (cit. on p. 26).

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