

Report

on a dissertation submitted for acquiring the educational and scientific degree "Doctor"
in the professional field 4.1 Physical sciences,
(Doctoral program "Nuclear Physics")

Author of the dissertation: Chavdar Chavdarov Dutsov, Doctoral student at the Faculty of Physics, Sofia University "St. Kliment Ohridski"

Topic of the dissertation: "Studies on the applications of the triple to double coincidences ratio method for primary activity standardization using liquid scintillation counting"

Reviewer: Professor Kiril Assenov Krezhov, Ph.D., D. Sc., member of the scientific jury according to the appointment of the Rector of Sofia University by Order RD 38-355 / 16.07.2021

Professional-biographical data about the doctoral student

The candidate Chavdar Dutsov completed his secondary education in 2012 in his hometown of Sofia and graduated with honours from the National High School of Natural Sciences and Mathematics, Academician "Lyubomir Chakalov", profile "Physics and Astronomy". He is a laureate of prestigious high awards from a number of national and international student competitions in physics and astronomy. He received higher education in physics from the Faculty of Physics (FzF) at Sofia University "St. Kliment Ohridski" with honours (6.00) bachelor's (2016) and master's (2018) diploma theses in the specialization "Medical Physics" under the guidance of Associate Professor Dr. Krassimir Mitev from the Department of Atomic Physics. Chavdar Dutsov worked as a physicist at FzF, he supervised students in the practical classes of the course "Metrology of ionizing radiation". His development as a specialist in the field of registration and metrology of ionizing radiation continues with his enrolment on April 1, 2019 in the doctoral program "Nuclear Physics" of the Faculty of Physics as a full-time doctoral student under the supervision of Assoc. Prof. Krassimir Mitev.

Reading the documents, I was left with the impression that the experience gained by Dutsov in his master's thesis on "Investigations on the application of the TDCR method" has gained in-depth, supplemented and further developed during his specializations in the metrological laboratory LNHB (Henri Becquerel National Laboratory) at LNE (French National Laboratory of Metrology) with targeted research on the absolute determination of radioactivity by liquid scintillation measurements. The obtained deductions and conclusions, combined with the results of hard work in the Department of Atomic Physics, served as the basis for the developed and presented in time dissertation.

Objectives and relevance of the problem

The title of the dissertation "Studies on the applications of the triple to double coincidences ratio method for primary activity standardization using liquid scintillation counting" correctly reflects its content. According to the doctoral student "*The primary measurement of activity using the TDCR method is the main focus of research in this dissertation.*" My opinion is that this dissertation describes step by step an active and creative overcoming of methodological and scientific problems in response to emerging challenges during the limited period of a full-time doctorate. The presented research and development is specified on the development and

validation of the method of triple to double coincidences in liquid scintillation medium as a method for absolute measurements of activity, including the creation and testing of components of a real functioning new specialized detector equipment implementing the Compton - TDCR method.

The topicality of the dissertation is beyond doubt, because the purpose of the experiments and their mathematical modelling as well as software and hardware support of the developed methodology is the accumulation of new knowledge directly related to the development and validation of innovative metrological methods and tools for determining radioactivity. I will note in general that solving the problems associated with obtaining reliable and proven results in the metrology of ionizing radiation is the subject of active international cooperation, often expressed through the participation of national and other selected laboratories in measurements on reference materials. The dissertation also presents results from the successful participation of the FzF metrological laboratory in inter-laboratory comparisons for tritium solution standardization, radon-containing water activity and measurements of radon behaviour in some polymers.

Structure and content of the dissertation

The dissertation is written in English and is presented on a total of 215 pages. It is structured in 12 chapters, divided into 3 parts. There are 3 Appendices included. It contains Cover Page, Acknowledgments, Contents (3 pages), Abbreviations Used (1 page), Introduction (3 pages), Part 1 (31 pages), Part 2 (55 pages), Part III (75 pages), section Scientific contributions (1 page), section Publications related to doctoral studies (2 pages), Bibliography of 159 titles (7 pages), including publications and reports with the participation of the doctoral student. In the document 69 figures and 12 tables are presented.

The first part of the dissertation (Chapters 1 and 2, 31 pages in total) shows the doctoral student's ability to synthesize and rank in importance the information from the analysed bibliographic sources in historical aspect regarding the measurement of radioactivity with liquid scintillators in the form of a mixture (cocktail) and used detectors systems with one or more photoelectron multipliers (PMTs). This part is a useful introduction to the metrology of ionizing radiation with liquid scintillators. A careful description of the triple-to-double-coincidences (TDCR) method is given and conditions are formulated to evaluate the random coincidences of this method and to calculate the registration efficiency in determining the activity of the sample.

Part II (Chapters 3, 4, 5 and 6, 55 pages in total) presents important main results of the dissertation in developing a new completely original approach for analysing the temporal aspects of scintillation events and for rejecting false events by accounting for the dead time of the detector complex. A detailed description of the distribution of scintillation events over time is given, the role of fast and delayed light emission of the scintillator is compared, and the different algorithms for registration and evaluation in the methods of coincidences when using liquid scintillators are compared. The correlation distribution and the application in TDCR are discussed in detail. At the end of this part, the doctoral student has made a brief review of the information presented in the individual chapters and critically addresses the main challenges for the successful application of the cross-correlation function in radionuclide metrology using liquid scintillation counting.

Part III (Chapters 7, 8, 9, 10 and 11, 75 pages in total) is devoted to the use of time distribution information in liquid scintillation measurements. The possibility of measuring the

half-life of excited nuclear states by means of time distribution analysis is shown. The new approach has been successfully applied to ^{57}Fe and ^{237}Np nuclei. Methods have been developed for estimating the contribution of random coincidences and making corrections in counting with a liquid scintillation system with three PMTs. Delayed fluorescence has also been shown to contribute to the accuracy of the estimation of activity calculated by the TDCR method and poses a problem for the correct choice of the coincidence window for low-energy radiation measurements analysed by Monte Carlo simulations. Chapter 10 describes the development of a new detector system for the primary standardization of liquid scintillation samples, which consists of an external source of monoenergetic gamma radiation and a Compton photon detector operating in a coincidence circuit with a TDCR detector system. With the new Compton-TDCR system, interesting results have been obtained regarding the behaviour of commercial liquid scintillators / cocktails when irradiated with electrons with energies in the range from 2 keV to 8 keV and in particular for standardization of tritium and ^{55}Fe . At the end of Part III the doctoral student synthesizes the presented information and the main conclusions in the separate 5 chapters.

The main claims of the doctoral student for the scientific and methodological contributions of the dissertation are formulated in Chapter 12 (2 pages) entitled Conclusions.

Three appendices are presented giving explanations and details about the programs created by the doctoral student: Appendix A (6 pages) on the developed Monte-Carlo approach for modelling scintillation events, Appendix B (6 pages) on the calculation of the cross-correlation function) also describing the efforts to optimize the computational processes, Appendix C (2 pages) with additional information marked C1-on possible selection of specific modules for analysis and classification of detected light pulse events and C2 - with additional figures for results of experiments with the newly developed Compton-TDCR detector.

Mastering by the doctoral candidate of the state of knowledge on the topic of the dissertation

The dissertation describes and presents in-depth analyses of the problems that accompany the absolute measurement of radioactivity by liquid scintillation methods and in the design and implementation of modern liquid scintillation detector systems. Both the main processes in the liquid scintillation media and the main means for recording ionizing radiation by liquid scintillation measurements are addressed. The doctoral student shows in the presentation a good knowledge of the facts and results of other authors, and critically comprehends and interprets the necessary information. The cited literature sources of 159 titles reflect the achievements and considered promising directions in the metrology of ionizing radiation in view of the progress in simulating the processes in liquid scintillation media and in the algorithms for registration and evaluation in the methods of coincidences. The publications on the dissertation also present a historical aspect and analyse current scientific and practical achievements in the field.

Characteristics of the material on which the contributions of the dissertation are built

The original experimental and theoretical material is set out in Part II and Part III, where the texts show an in-depth understanding of the physical basis of processes in scintillation media and reflect a widely discussed metrological perspective on the possibilities for absolute

measurements of radioactivity. My general impression is that the dissertation features high-level readability and has the quality to present the information in a logical sequence.

The reflections of the doctoral student on the possibilities for specifying the obtained solutions and formulating future guidelines for his research in the field of metrology of ionizing radiation leave a good impression. The possibility to perform cross-correlation measurements and obtain the light output with the developed Compton-TDCR system is already shown in the dissertation, but separately - section 6.1 for cross-correlation measurements with the Compton-TDCR system and section 10.4 for the light output of the scintillator. These are prerequisites for combining the results of the two types of measurements to obtain more comprehensive information about the scintillator.

Scientific contributions

The contributions in the dissertation formulated by the doctoral student in seven points are supported by convincing arguments and facts and I accept them. I also accept his classification of achievements as of fundamental, methodological, and applied science nature in the field of liquid scintillation methods.

Among the contributions received with the crucial role of the doctoral student, I would like to note:

1. Methods for estimating the rate of counting random coincidences in TDCR measurements have been developed.
2. The cross-correlation distribution of the time intervals between detected scintillation events is derived and it is shown how it can be used to determine the efficiency for registration and the activity of liquid scintillation samples.
3. The effect of delayed fluorescence on the activity calculated by the TDCR method is shown.
4. A Compton-TDCR system has been developed with the possibility for quantitative characterization of the light yield of liquid scintillators.

To achieve these contributions, as well as the other contributions noted by the doctoral student, he has demonstrated and applied creatively his software knowledge and skills.

Publications on the dissertation and candidate's personal contribution

The activities on the experiments, the peculiarities of the developed and / or applied methods for extraction of reliable physical information as well as the obtained results with the participation of the doctoral student are presented widely enough at international and national scientific forums and are summarized in 9 published scientific papers in specialized journals. The author teams of the articles and reports are from 4 to 7 members and in all articles the co-author is the scientific supervisor of the doctoral student associate professor Krassimir Mitev. In 4 articles the doctoral student is on the first place in the list of authors. I accept the distribution as indicated by Chavdar Dutsov (out of the 9 publications, 5 are with his significant contribution).

The doctoral student did not provide information about independent citations of the publications with his co-authorship included in the dissertation.

Abstract

The abstract is presented in two versions - in Bulgarian (58 pages) and in free translation into English (36 pages). The version in Bulgarian is in an expanded form and in practice is an independent review of the doctoral student on the dissertation as the numbering of the included figures and tables differs from those used in the text of the dissertation. The structure of the presented text is also different. Chapter 1 in the abstract presents Part I in the dissertation, Chapter 2 synthesizes part of Chapter 4 in the dissertation, Chapter 3 combines the exposition in Chapters 3,4,5 and 6 of the dissertation, Chapters 4 to 9 summarize Chapters 7 - 12 of the dissertation. A similar inconsistency in structuring is found in the English text.

Despite the inconveniences of reading and comparing the facts, my assessment is that both versions of the abstract clearly and accurately reflect the content of the dissertation, the conclusions and the contributions formulated by the dissertation.

Critical remarks and recommendations

The dissertation is very well illustrated, the presentation is concise and without repetitions. I do not find any contradictions and shortcomings in the logical sequence of the presentation of facts, the performed analyses and the drawing of conclusions, which would cast a shadow over the serious work and achievements of the doctoral student on the topic and question the achieved professionalism. In short, I have no objections in principle to the content and structure of the dissertation and to the reliability of the presented results and conclusions.

Conclusion

The presented dissertation satisfies the requirements for obtaining the educational and scientific degree "Doctor" defined in the Law for development of the academic staff in the Republic of Bulgaria, the regulations for application of this law, as well as in the internal regulations for its application of Sofia University and the Faculty of Physics. And in scientometric terms it exceeds them. The doctoral student has actively participated and has shown its analytical skills in the preparation and implementation of important stages in the development and validation of the method of triple to double coincidences for absolute measurements of radioactivity. He has made significant contributions to the ideas and in some specific methodological and key software developments has shown initiative and creative independence. It can be concluded that the doctoral student has developed competence and convincingly proven potential for future development, including independently.

Based on the above, I most strongly call on the esteemed members of the Scientific Jury to vote with YES for the award of the educational and scientific degree "DOCTOR" to Master Chavdar Chavdarov Dutsov.

Sofia, August 24, 2021.

Reviewer:

Prof. Dr.Sci. Kiril A. Krezhov