CTH-NT-229

Thesis for the degree of Doctor of Philosophy

Safeguards: Modelling of the Detection and Characterization of Nuclear Materials

Andreas Enqvist

Nuclear Engineering
Department of Applied Physics
Chalmers University of Technology
S-412 96 Göteborg, Sweden 2010

Safeguards: Modelling of the Detection and Characterization of Nuclear Materials
ANDREAS ENQVIST
ISBN 978-91-7385-356-9

Doktorsavhandling vid Chalmers tekniska högskola
Ny serie nr 3037
ISSN 0346-718X

Nuclear Engineering
Department of Applied Physics
Chalmers University of Technology
S-412 96 Göteborg
Sweden
Telephone +46 (0)31 772 1000
Fax +46 (0)31 772 3872

Cover: Experimental setup for measuring coincident radiation in form of neutrons and gamma rays from fissile samples. Also shown in Fig. 7.1(b).

Chalmers Reproservice
Göteborg, Sweden 2010

Safeguards: Modelling of the Detection and Characterization of Nuclear Materials

ANDREAS ENQVIST
Nuclear Engineering
Department of Applied Physics
Chalmers University of Technology

ABSTRACT

Nuclear safeguards is a collective term for the tools and methods needed to ensure nonproliferation and safety in connection to utilization of nuclear materials. It encompasses a variety of concepts from legislation to measurement equipment.

The objective of this thesis is to present a number of research results related to nuclear materials control and accountability, especially the area of nondestructive assay. Physical aspects of nuclear materials are often the same as for materials encountered in everyday life. One special aspect though is that nuclear materials also emit radiation allowing them to be qualitatively and quantitatively measured without direct interaction with the material. For the successful assay of the material, the particle generation and detection needs to be well understood, and verified with measurements, simulations and models.

Four topics of research are included in the thesis. First the generation and multiplication of neutrons and gamma rays in a fissile multiplying sample is treated. The formalism used enables investigation of the number of generated, absorbed and detected particles, offering understanding of the different processes involved.

Secondly, the issue of relating the coincident detector signals, generated by both neutrons and gamma rays, to sample parameters is dealt with. Fission rate depends directly on the sample mass, while parameters such as neutron generation by alpha decay and neutron leakage multiplication are parameters that depend on the size, composition and geometry of the sample. Artificial neural networks are utilized to solve the inverse problem of finding sample characteristics from the measured rates of particle multiples.

In the third part the interactions between neutrons and organic scintillation detectors are treated. The detector material consists of hydrogen and carbon, on which the neutrons scatter and transfer energy. The problem shares many characteristics with the area of neutron moderation found in reactor physics.

Finally the last part of the thesis consists of measurement systems. Measuring coincident neutrons and gamma rays using fast scintillation detectors and data acquisition systems, can enable development of new types of methods for interpreting material signatures.

Keywords: nuclear safeguards, master equations, number distribution, multiplicities, fissile material, scintillation detectors, light pulse distribution, cross correlations

Appended papers

This thesis consists of an introduction to and a summary of the work published in the following papers:

Paper I

A. Enqvist, I. Pázsit and S.A. Pozzi, "The Number Distribution of Neutrons and Gamma Photons Generated in a Multiplying Sample"
Nuclear Instruments & Methods ***A***, **566**, 598–608 (2006).

Paper II

A. Enqvist, I. Pázsit and S.A. Pozzi, "The Detection Statistics of Neutrons and Photons Emitted from a Fissile Sample"
Nuclear Instruments & Methods ***A***, **607**, 451–457 (2009).

Paper III

I. Pázsit, A. Enqvist and L. Pál, "A note on the multiplicity expressions in nuclear safeguards."
Nuclear Instruments & Methods ***A***, **603**, 541–544 (2009).

Paper IV

A. Enqvist, I. Pázsit and S. Avdic,"Sample Characterization Using Both Neutron and Gamma Multiplicities"
Accepted for publication in Nuclear Instruments & Methods ***A***, (2010).
doi:10.1016/j.nima.2010.01.022

Paper V

S. Avdic, A. Enqvist and I. Pázsit, "Unfolding sample parameters from neutron and gamma multiplicities using artificial neural networks."
ESARDA Bulletin, **43**, 21–29 (2009). Invited paper.

Paper VI

S.A. Pozzi, M. Flaska, A. Enqvist and I. Pázsit, "Monte Carlo and Analytical Models of Neutron Detection with Organic Scintillation Detectors."
Nuclear Instruments & Methods ***A***, **582**, 629–637 (2007).

Paper VII

A. Enqvist and I. Pázsit, "Calculation of the light pulse distributions induced by fast neutrons in organic scintillation detectors."
Submitted to Nuclear Instruments & Methods ***A***.

Paper VIII

A. Enqvist, M. Flaska and S.A. Pozzi, "Measurement of Neutron/gamma-ray Cross-

Correlation Functions for the Identification of Nuclear Materials"
Nuclear Instruments & Methods ***A***, **595**, 426–430 (2008).

Paper IX

A. Enqvist, M. Flaska and S.A. Pozzi, "Initial Evaluation for a Combined Neutron and Gamma-ray Multiplicity Counter"
Submitted to Nuclear Instruments & Methods ***A***.

Related works not included in this thesis

A. Enqvist, I. Pázsit and S.A. Pozzi, "The Number Distribution and Factorial Moments of Neutrons and Gamma Photons Generated in a Multiplying Sample"
Journal of Nuclear Materials Management, **XXXV**, No. 1, 29 (2006). Invited paper.

I. Pázsit and A. Enqvist, "Theory of periodically pulsed Feynman- and Rossi-alpha methods."
Trans. Am. Nucl. Soc. **97**, 656 - 657 (2007)

I. Pázsit and A. Enqvist, "Neutron Noise in Zero Power Systems."
Lecture series at the IAEA Workshop on Neutron Fluctuations, Reactor Noise, and Their Applications in Nuclear Reactors. Lecture notes 171 pages. Hosted by the International Centre for Theoretical Physics (ICTP), Trieste, Italy, 22 to 26 September 2008, I2-TR-35632.

A. Enqvist, I. Pázsit and S.A. Pozzi, "Calculation of the pulse height distribution induced by fast neutrons in a scintillating detector."
Proceedings of Joint International Topical Meeting on Mathematics & Computation and Supercomputing in Nuclear Applications (M&C + SNA 2007). Monterey, CA, USA April 15-19 (2007).

CONTENTS

CHAPTER

1

INTRODUCTION

This introduction describes the objective of the work which led to this thesis, and outlines the contents of the following chapters.

1.1 Objective

Nuclear materials are a subcategory of radioactive materials, which are normally harmful or even lethal to human beings when exposed to them without precautions. At the same time, many cancer patients will associate the radiation to life saving procedures. In older generations many have associations to the cold war and the race to nuclear arms that it meant. The atomic bomb may be the pinnacle of man-made destruction and horror, and thus a constant fear in the minds of many people. The climate change has renewed focus to nuclear energy both in fission and fusion format as a source of non-carbon emitting energy. The concept of nuclear means fear and hopes, ideas and questions, all coexisting together in the collective minds of people.

This opens a great possibility for research, understanding and knowledge: how can the positive sides of a phenomenon or concept be utilized without insurmountable drawbacks and potential risks? In the specific area of nuclear materials, the key issue is safeguards. A concept including areas such as physical protection, legislation and measurement systems.

A very prominent source of nuclear materials is what is expelled as "waste" from commercial nuclear reactors. The materials and elements contained within show great differences, where some quickly decay into stable harmless isotopes, while others stay radioactive and pose a very real threat for thousands of years. Due to the mixed contents, some of the used fuel from reactors is put through reprocessing

and separation, leading to materials with new signatures and uses. Among those plutonium, unused uranium and long lived actinides are some examples of the more dangerous nuclear materials.

The risk of nuclear proliferation is well-known and has been visible and imposing ever since the first countries acquired nuclear weapons. The International Atomic Energy Agency (IAEA), which is part of the United Nations (UN), attempts to address this issue by urging all countries to sign nonproliferation agreements, and agreeing to implement additional safeguards measures, so that peaceful uses of nuclear materials within energy production, agriculture and health care can be sustained.

Naturally, having rules and legislations will mean nothing if there are no methods of detection and understanding of what was the source of the signals detected. This marks a dependence on research done within the areas of detectors, nuclear physics, data analysis etc. to provide the means to enforce that no clandestine activities or unintentional mistakes leads to proliferation or to exposure of persons to unmotivated health hazards.

Present and future nuclear energy needs public acceptance for successful operation and expansion. The acceptance in turn, depends on how the question of radioactive waste is solved. what will be the final solution, how is the handling of such material monitored to ensure the public safety? This means that expanding and developing nuclear safeguards is very important to the nuclear industry as well as for the authorities. The proposed solutions to the issue of nuclear waste include repositories, reprocessing or other techniques. All of these require that the handling of the material has to be guaranteed to be safe and with a minimal probability of accidents and emissions of dangerous materials. Safeguards is a key parameter to ensure all these things. The objective of the work presented here is to give contribution to the development of methods in specific areas of nuclear safeguards to achieve the above goals.

1.2 Outline of the thesis

Nuclear safeguards is a very wide field, of which only selected parts have been the subject of the work underlying this thesis. The work is oriented towards the nuclear physics, and specifically that of modelling nuclear materials and understanding the physical interactions which allows for characterization and detection of nuclear materials. The process of particle generation is normally through nuclear fission in which a certain number of neutrons and gamma rays are produced. Since the particle generation is not deterministic, instead it is described by random variables, i.e. a stochastic process. Even when the particle parameters and the material constituents are perfectly known, the result of a fission can only be described by a set

of probabilities. The stochastic aspects and the resulting probability theories are addressed in the presented results.

This thesis starts with an introduction into safeguards and its history in Chapter 2 to give a overview of the area. Existing methods and tools will be detailed to some extent in Chapter 3, to exemplify what the field of safeguards contains in form of materials control and accountability. The publications that have been the result of the work are then the foundation of the following chapters. Chapter 4, based on Papers I-II, investigates the number distribution of particles generated in and detected from fissile samples. A deeper survey into the multiplicity theory and sample parameter unfolding is offered in Chapter 5 (Papers III-V). A way of detecting the emissions from fissile samples is by using scintillation detectors, the interactions between neutrons and such detectors are contained in Papers VI-VII which give the foundation of Chapter 6. Chapter 7 then outlines some experimental work and actual measurements, which have been the basis of Papers VIII-IX. Finally, Chapter 8 summarizes the work and details possible future work and extensions.

It is the aim of the thesis to take a full journey from the motivation of safeguards to the existing methods and tools and how they could be understood and improved. Later it addresses some issues and models of the more specific scenarios dealt with in safeguards. Finally a connection is made between theory and reality, when the problem of interpretation of data by solving the inverse problem from the measurables to sample parameters is studied, and finally some measurements are detailed that investigate the applicability of some of the models.

CHAPTER

2

SAFEGUARDS - BACKGROUND

Nuclear Safeguards is a term for tools and concepts used to quantify, detect and monitor nuclear materials. Nuclear materials themselves can range from hospital radioisotopes and nuclear batteries to spent nuclear fuel and weapons grade material. To understand the needs of safeguards an initial foundation and motivation for the area will be given.

2.1 The origin

The origins and foundation of safeguards stem from the rapid development of the nuclear age in connection to and after the Second World War. It quickly became apparent that the possibility to acquire nuclear weapons was open to many nations. Both the knowledge of designing such devices and the material needed for it was quickly spreading around the world. The first attempt to address this issue was D. Eisenhower's "Atoms for peace"-speech to the United Nations assembly in 1953. It outlined a desire to limit the number of nations who gained nuclear potential, by offering them an incentive in the form of technology and knowledge needed for using nuclear energy for peaceful purposes.

As the idea matured and the number of countries involved in the program increased, it eventually led to the birth of the International Atomic Energy Agency (IAEA). In the statute of IAEA the goals of the organization are outlined; promoting nuclear development and peaceful technology, while also establishing safeguards for the purpose of assuring that no nuclear assistance from IAEA was used to further the development of nuclear weapons [1].

Also in the present and future utilization of nuclear power the concept is very important. Much focus is currently aimed at developing the next generation nuclear reactors, often referred to as Generation-IV reactors. Some of the main aspects of

those designs are proliferations resistance, inherent safeguards, as well as destroying some of the waste produced in previous generations reactors. So while safeguards was born as a way to deal with an existing problem, it has now evolved to being one of the driving forces of future innovations and safety considerations.

It is seen from the statute of the IAEA that safeguards have both a technical and a political side. To give a broader view of what the challenges were and still are, we can take the current divisions of the Institute of Nuclear Materials Management which well exemplifies the different areas where safeguards are used [2]:

- International Safeguards - Exchange of information on the development, implementation and effectiveness of nuclear safeguards. Promoting publications and workshops on the spread of advancements in safeguard technologies.
- Materials Control and Accountability - Technology and knowledge needed by professionals within the area of materials management.
- Nonproliferation and Arms Control - Efforts and support of nonproliferation and arms control by treaty verification, transparency measures and nuclear materials management aimed at furthering international stability.
- Packaging and Transportation - Promoting the safe and controlled packaging and transportation of radioactive materials found in e.g. spent nuclear fuel, materials containing induced radioactivity, radioactive waste and contaminated materials.
- Physical Protection - Advancements and implementations of technology and systems for the physical protection of nuclear materials and facilities, such as surveillance, protection barriers etc.
- Waste Management - Radioactive waste management technology such as storage, handling, processing and disposal of nuclear waste produced around the world.

By understanding the needs which the IAEA has, or more precisely the needs of inspectors connected to IAEA or other agencies monitoring radioactive materials, equipment and routines can be constructed for detecting and monitoring radioactive materials. In the following the area of materials control and accountability (MC&A) will be the focus.

2.2 Radioactive materials

The nuclear materials mentioned above are a subset of radioactive materials, and those are characterized by the emission of radiation. This radiation can take a number of forms: photons, neutrons, electrons and alpha particles. Radionuclides

can be divided into a number of different groups depending on their usage and occurrence [3]:

- Nuclear materials: ^{233}U, ^{235}U, ^{239}Pu, ^{241}Pu.
- Industrial radionuclides: ^{60}Co, ^{137}Cs, ^{192}Ir, ^{226}Ra, ^{241}Am, ^{238}Pu.
- Medical radionuclides: ^{18}F, ^{57}Co, ^{67}Ga, ^{99m}Tc, ^{201}Tl, ^{123}I, ^{125}I, ^{111}In, ^{192}Ir.
- Naturally occurring radioactive materials (NORM): ^{40}K, ^{226}Ra, ^{232}Th, ^{238}U.

As can be noted there are several radionuclides, many of which are in regular use in industry, medicine or energy production. The American Nuclear Regulatory Commission defines those nuclear materials that could be used in the creation of nuclear weapons as "special nuclear material".

However, also the other radionuclides need to be monitored and verified to avoid risks. The dangers of sources that are accidentally or intentionally lost are many and some examples of incidents include: unintentional incorporation of radioactive material into recycled steel [4, 5]; theft of radioactive material [6] and recovery of lost sources by unsuspecting individuals [7]. Discovery of such sources might be best implemented at points of transport, such as airports, stations, seaports etc.

2.3 Special nuclear materials

To more accurately address the needs of the IAEA, and to create quantifiable goals of what the safeguards are aimed at achieving, a number of special terms need to be defined. In the regime of nonproliferation, it is especially material that could be used for creation of nuclear weapons that needs to be safeguarded. The IAEA differentiate between "direct-use" and "indirect use" materials, by referring to the time needed to develop nuclear weapons.

Direct-use material such as highly enriched uranium (HEU) has a timeliness of as little as one month. Timeliness here is defined as a measure of how fast a nuclear device could be created using that specific material. Indirect use material, such as uranium enriched below 20%, has a timeliness longer than of one year. However it is not only the time aspect of certain materials that put conditions on the goals of safeguards, but also the quantities. For this purpose the term significant quantity (SQ) has been defined, which corresponds to the total amount of material needed to create a nuclear device (see Tab. 2.1).

The foregoing outlines the quantitative and qualitative goals associated with safeguards for special nuclear material. The goal is to be able to detect whether 1 SQ of material has been diverted within a time frame that is less than the timeliness of the specific material in question. This also describes the tools needed for this goal: systems able to correctly quantify material such that diversion of material can be

Material category	SQ	Comment
"Direct-use" Pu	8 kg	Total amount of the material
^{233}U	8 kg	Total amount of the isotope
U (^{235}U $\geq$ 20 %)	25 kg	^{235}U
"Indirect use" U (^{235}U $<$ 20 %)	75 kg	^{235}U
Thorium	20 000 kg	Total amount of the material

Table 2.1: List of different types of materials and their respective significant quantities (SQ) [8].

detected. Since all measurements have statistical uncertainties it means that those need to be reasonably small, such that deviations between verifications really depend on material unaccounted for, rather than uncertainties of the tools available to do the verification.

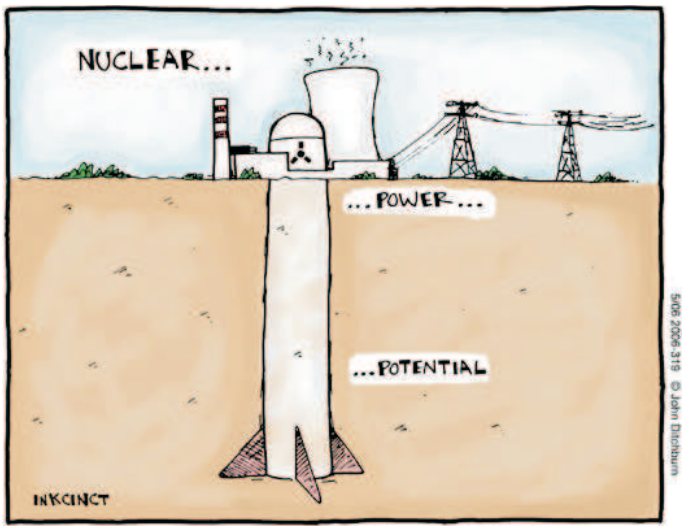


Figure 2.1: The good and the bad sides of the situation? Nuclear power offers a solution to very dire climate problems, but not without also solving the back-end situation of how to handle nuclear waste. [9]

CHAPTER

3

EXISTING METHODS AND TOOLS

The type of tools needed by safeguards depends on the radiation emitted, the type of material, the mass and a number of other parameters. An overview of those tools will be given here and it will also serve as an introduction to the work presented in the following chapters.

3.1 Material assay

Safeguards for analyzing materials can usually be divided into two subcategories:

- Destructive Assay (DA)
- NonDestructive Assay (NDA)

Destructive assay constitutes an intrusive handling of the material, and for that to be possible, direct access to the material is needed. In a laboratory it can give very precise information about the investigated material. For assaying large quantities of material or packed materials that one finds in large volumes at seaports or airports, this method is hardly suitable. Nondestructive assay on the other hand relies on the material sending out radiation that can be detected: either by just passively observing the inherent radiative signature of the material (passive interrogation) or irradiating the material to induce emission of radiation and detecting that (active interrogation). Passive interrogation means that the physical processes and interactions of the material needs to be well understood, since that is what can be used to apply inverse tasks such as finding source properties (type of nuclide, mass of the sample) from the radiative signature.

In the following sections a few such methods will be described. The goal of the work was to develop these methods further, to enhance the understanding of the processes and to elaborate more sensitive and precise monitoring methods.

3.1.1 Gamma measurements

Most nuclear materials emit characteristic gamma rays, which could be used for identification. Enriched uranium has a strong 186 keV gamma ray associated with the alpha decay of ^{235}U, so gamma spectrum measurements of uranium can give information on the enrichment level. Similarly plutonium samples tend to contain a number of Pu isotopes, which gives rise to a characteristic mix of gamma ray energies. This can be used to find the isotopic composition of said samples. Also spent fuel can be assayed by a similar method utilizing the decay of ^{137}Cs, to determine the discharge date from the reactor.

Both scintillation detectors and solid state detectors are used for gamma ray spectroscopy. They have different properties and tend to complement each other depending on the situation and the task performed [10]:

- Sodium iodide (NaI) detectors can be made very large and thus have a high detection efficiency. They can be used for enrichment measurements, but have a rather poor energy resolution, meaning they can not be used to distinguish gamma rays from nearby peaks. The time resolution is very high with the signal carrier being photons in a scintillation detector.
- High purity germanium (HpGe) detectors have excellent energy resolution as their main advantage, but they require low temperatures for optimal operation, usually achieved by liquid nitrogen cooling. The time resolution for the solid state HpGe detector is worse than that of a scintillation detector.
- Cadmium zinc telluride (CZT) detectors exhibit features such as room temperature operation and an energy resolution in between NaI and HpGe detectors. They also exhibit very high intrinsic efficiency (internal detection efficiency), but are normally limited in size compared to the other detectors.

An example of a detection system for gamma ray measurements is the hand-held HM-5 (fieldSPEC), which is normally based on a NaI-detector, but can also use a higher resolution CZT detector. It represents a type of device that can be used for a wide range of tasks: dose rate measurements, source searches, isotope identification etc. in a wide range of situations enabled by the portability of the device. Some examples of detectors can be seen in Fig. 3.1.

3.1.2 Neutron measurements

Neutron measurements are popular especially for determining Pu mass. With the penetrability of neutrons through high-Z material they do not experience the problem of self-shielding that gamma rays exhibit in larger samples. Most neutron measurement systems are based on coincidence or multiplicity counters measuring mostly plutonium or uranium. Since neutrons are produced especially in fission events, they

Figure 3.1: The handheld HM-5 (right) shown together with an active well neutron counter (left).

are normally a good indicator of fissile material. However they are also produced in (α, n) events, so searching for the coincident neutrons from fissions enables better verification of fissile material in the sample.

3.1.3 Example of implementation

Implementing safeguards, means using a number of different techniques and detectors. Table 3.1 lists examples of the techniques used at different plants by IAEA during 2002. In total over 700 NDA systems were used and a total of 2430 inspections were performed. In addition to the systems mentioned below, video cameras for optical surveillance are extensively used together with seals on containers and areas. Finally, also environmental sampling is being used, both at sites such as enrichment plants but also in and outside other installations not mentioned in the table.

3.2 Multiplicity counters

To give a flavour of the methods used in the research, neutron multiplicity measurements are presented here. It is a proven method implemented at many facilities. The properties dealt with in the physics of fission are neutron and gamma ray distributions, together with the changes introduced by having a multiplying sample. Measurable units however are detection rates of neutrons in neutron assay. The measurable units can be derived from factorial moments that can be calculated from the particle distribution related to fission in different isotopes. A generalization of concepts and equations mentioned below is a core part of the research presented in the following chapters.

Plant	Materials	Main techniques deployed
Enrichment plant	UF_6	γ-ray spectroscopy, Weighing
Fuel fabrication plant	U and Pu oxides, MOX	γ-ray spectroscopy, Neutron counting, Destructive analysis, Isotopic determination
Power reactors	Spent fuel	Cherenkov glow detection, Gross γ-ray and neutron detection
Reprocessing plant	U and Pu nitrates	Destructive analysis, Neutron counting

Table 3.1: Sample of implementation scheme of safeguards in the case of facilities connected to nuclear power [11].

Multiplicity counters have been in use for some time now and which have evolved since the original designs. Both passive and active mode are used in different multiplicity and coincidence designs, depending on how high of a fission rate the material exhibits. They represent a type of system that closely connects the area of safeguards to the specific research mentioned in the subsequent chapters.

Modern multiplicity counters use almost exclusively ^{3}He-detectors for neutron multiplicity measurements. However, originally fast plastic scintillation detectors were used in a type of system called fission multiplicity detection. Where ^{3}He-detectors depend on moderated neutrons to undergo the $^3\mathrm{He}\,(n,p)\,^3\mathrm{H}$ capture reaction, which means that they are sensitive to neutrons only, the scintillation detectors are also sensitive to high energy gamma rays [12]. The problem with this approach is that, as will be demonstrated later, the so-called point model can be used to correlate the important neutron parameters to sample parameters related to mass and composition etc. but for gamma rays this equivalence did not exist. A total multiplicity of the sample was measured rather than a neutron multiplicity, unless special measures were taken to suppress the gamma rays.

3.2.1 Factorial moments

To understand the basis of the equations used the concept of factorial moments needs to be introduced. Starting with a certain distribution of a discrete variable $P(\nu)$, one can create a probability generating function (PGF) [13,14] in the form of:

$$Q(z) = \sum_{n}^{max} P(n) z^n. \tag{3.1}$$

The n-th factorial moment ν_n is taken from the relation:

$$\nu_n = \left.\frac{d^n Q(z)}{dz^n}\right|_{z=1}, \tag{3.2}$$

thus the first factorial moment i.e. the expectation of the variable ν is

$$\nu_1 = \sum_n^{max} \nu P(\nu). \tag{3.3}$$

This can be applied very straightforward to the probabilities associated with the number of neutrons generated in a spontaneous or induced fission event. This type of distribution is a nuclear constant depending on which isotope(s) the sample consists of:

$$\nu_{sf,n} = \left.\frac{d^n p_{sf}(z)}{dz^n}\right|_{z=1}, \quad \nu_{i,n} = \left.\frac{d^n p_i(z)}{dz^n}\right|_{z=1}. \tag{3.4}$$

Table 3.2 lists the neutron emission probabilities associated with the sample composition used for quantitative work in Papers I-II.

number of neutrons	spontaneous fission, $p_{sf}(n)$	induced fission, $p_i(n)$
0	0.0638	0.0071
1	0.2316	0.0674
2	0.3325	0.2283
3	0.2533	0.3263
4	0.0987	0.2510
5	0.0181	0.0958
6	0.0020	0.0208
7	0	0.0029
8	0	0.0005
$\sum p_x(n)$	1.000	1.000

Table 3.2: The nuclear data for the probabilities of emitting n neutrons from spontaneous and induced fission in a sample of 20 wt% ^{240}Pu and 80 wt% ^{239}Pu.

3.2.2 Detector measurables

Formulae for the factorial moments belonging to a PGF for the number of neutrons generating from a single source event in a *multiplying* sample were derived by Böhnel [15] and Hage & Cifarelli [16]. By correlating those expressions to the process of detection, analytical formulae for the first three detection rates (singles, doubles and triples) can be obtained in the form of [17]:

$$S = F\varepsilon_n \mathbf{M}\nu_{sf,1}(1+\alpha), \tag{3.5}$$

$$D = \frac{F\varepsilon_n^2 f_d \mathbf{M}^2}{2}\left[\nu_{sf,2} + \left(\frac{\mathbf{M}-1}{\nu_{i,1}-1}\right)\nu_{sf,1}(1+\alpha)\nu_{i,2}\right], \tag{3.6}$$

$$\begin{aligned} T = \frac{F\varepsilon_n^3 f_t \mathbf{M}^3}{6}\Bigg\{ & \nu_{sf,3} + \left(\frac{\mathbf{M}-1}{\nu_{i,1}-1}\right)\left[3\nu_{sf,2}\nu_{i,2} + \nu_{sf,1}(1+\alpha) + \nu_{i,3}\right] \\ & + 3\left(\frac{\mathbf{M}-1}{\nu_{i,1}-1}\right)^2 \nu_{sf,1}(1+\alpha)\nu_{i,2}^2 \Bigg\}. \end{aligned} \tag{3.7}$$

In the above ε_n stands for the neutron detection efficiency, the neutron leakage multiplication, $\mathbf{M}$, and the α-ratio are defined in the following chapters, f_d and f_t are gate fractions for the detectors and finally F is the spontaneous fission rate in the sample. In the case of Pu assay, it is taken as 473 fissions per second per gram of effective ^{240}Pu mass. The effective mass depends on the isotopic composition and is defined as the mass of ^{240}Pu that would give the same number of doubles as obtained from all even isotopes of Pu:

$$^{240}\mathrm{Pu}_{\mathrm{eff}} = 2.52\,^{238}\mathrm{Pu} + {}^{240}\mathrm{Pu} + 1.68\,^{242}\mathrm{Pu}.$$

By measuring the first three multiples rates (singles, doubles and triples), It is possible to deduce the three unknowns F, $\mathbf{M}$, and α, when the detector efficiency is known. A schematic of a design for a ^{3}He based multiplicity counter is found in Figure 3.2.

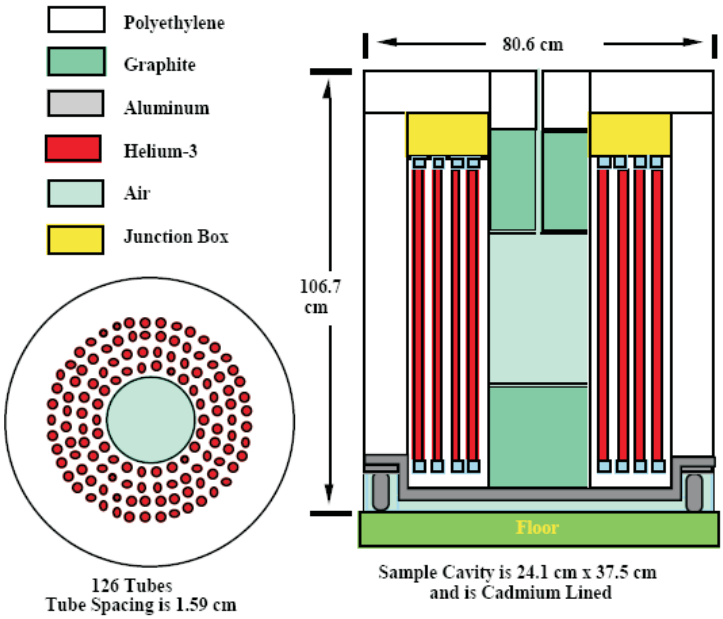


Figure 3.2: Design schematic of the In-plant (Pyrochemical) Multiplicity Counter (from [17]).

CHAPTER

4

PARTICLE NUMBER DISTRIBUTIONS

When dealing with neutrons branching processes are often encountered. The understanding of the number of particles generated and how they are connected offers a deep insight into the physics of fissile samples. This chapter details the mathematics needed as well as the basic concepts behind the processes governing particle generation.

4.1 Branching processes

In fissile samples neutrons can be absorbed into a nucleus and generate a fission event. In that process a number of new neutrons are born each with the same ability to generate new fissions. However, not only fissions can take place but also absorptions and leakage from the sample, which means that the number of neutrons in the chain is reduced. The full behaviour of the neutrons in the sample can be depicted as a branching process like the one in Fig. 4.1.

Mathematically it can be described in a number of ways depending on what assumptions and approximations are made. From now on it is assumed that the particles and sample have no "memory", implying that a particle born later in the branching process will has no altered behaviour depending on the previous history of the branching process, such that the new particle have the same probability to undergo reactions as a previous particle in the sample. Mathematically it can be expressed using conditional probabilities in the form of:

$$Pr(X_{n+1} = x | X_1 = x_1, X_2 = x_2, \ldots, X_n = x_n) = Pr(X_{n+1} = x | X_n = x_n), \quad (4.1)$$

where x_i is the state of the random process X_i at different discrete steps. The

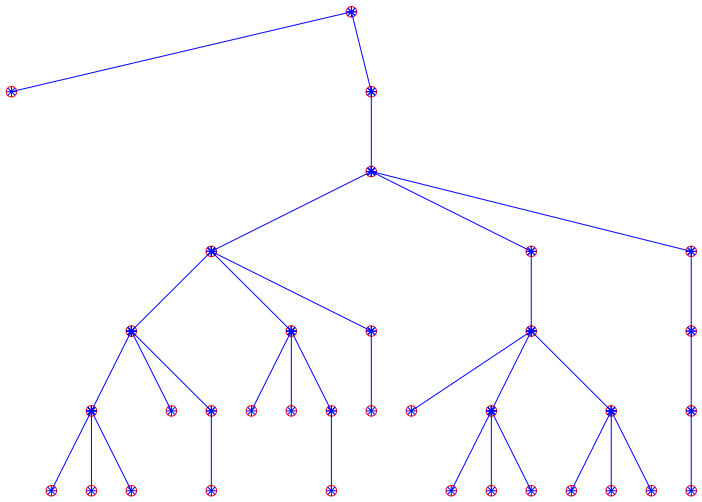


Figure 4.1: A branching process, where the event can lead to both a multiplication of the number of particles but also the removal of a particle (cf. absorption).

generation of neutrons (and also gamma rays) is considered to follow this behaviour and the process is then called a *Markov* process [18].

4.2 Master equations

A way of describing the generation of neutrons and gamma rays using master equations has been shown by Böhnel [15] and Pázsit & Pozzi [19]. Considering an absorption-free system (or alternatively: investigating the total number of *generated* particles), the only reaction a neutron can be involved in is an induced fission, which can be assigned a certain probability, p, and likewise the probability not to induce fission is $(1-\mathrm{p})$. The first collision probability is closely linked to the sample mass, the larger the sample the more likely a neutron will suffer a collision. Using the tool of probability generating functions it is possible to define the following generating functions for the probabilities of the number of neutrons generated in the sample by a single neutron or by a source event (spontaneous fission), $h(z)$ and $H(z)$, respectively:

$$h(z) = \sum_n p_1(n) z^n \tag{4.2}$$

and

$$H(z) = \sum_n P(n) z^n. \tag{4.3}$$

Here $p_1(n)$ and $P(n)$ are the probabilities to generate n neutrons starting with a single neutron or a spontaneous fission respectively. By using the PGFs of the

neutron distribution from spontaneous and induced fission:

$$q_{sf}(z) = \sum_n p_{sf}(n) z^n \quad , \quad q_i(z) = \sum_n p_i(n) z^n, \tag{4.4}$$

a derivation of master equations can be performed by writing down the probability balance equations. For the probability $p_1(n)$ of neutrons induced by a single starting neutron, one has, by summing up the probabilities of the mutually exclusive events of not having or having a first collision in the sample, the following:

$$p_1(n) = (1 - \mathrm{p})\delta_{n,1} + \mathrm{p} \sum_{k=1}^{\infty} p_i(k) \sum_{\{n_1 + n_2 + \ldots + n_k = n\}} \prod_{i=1}^{k} p_1(n_i) \ . \tag{4.5}$$

By doing the same also for $P(n)$, and converting the equations into those for the probability generating functions, a pair of coupled backward master equations can be derived:

$$h(z) = (1 - \mathrm{p})z + \mathrm{p} q_i[h(z)] \tag{4.6}$$

and

$$H(z) = q_{sf}[h(z)], \tag{4.7}$$

where according to Eq. (4.4), $q_1[h(z)] = \sum_n p_i(n)[h(z)]^n$. In Eq. (4.6) the first term represents the event that the neutron does not induce fission, and the process then ends there. The second term however, indicates that a number of new neutrons are generated in the induced fission event, each of which will be treated independently and thus have the chance to generate new neutrons themselves according to the generating function $h(z)$ itself.

4.2.1 Gamma ray equations

For gamma rays the situation is very different compared to neutrons. The reason is that gamma rays are not self-multiplying but rather depend on the neutrons for multiplication through induced fissions. This means that to create master equations similar to the case of neutrons the formulae needs to be connected to neutrons rather than gamma rays. The distributions of gamma rays from induced and spontaneous fissions are also needed, and their PGFs are defined as follows:

$$r_{sf}(z) = \sum_n f_{sf}(n) z^n \quad , \quad r_i(z) = \sum_n f_i(n) z^n, \tag{4.8}$$

where $f_{sf}(n)$ and $f_i(n)$ are the probabilities of generating n gamma rays in a spontaneous or induced fission, respectively.

The next step is to define the probabilities $f_1(n)$ and $F(n)$ for the numbers of gamma rays generated by a single neutron or by a source event, respectively, and

their PGFs $g(z)$ and $G(z)$. The master equations for the generating functions can now be expressed as:

$$g(z) = (1 - \mathrm{p}) + \mathrm{p}r_i(z)q_i[g(z)] \tag{4.9}$$

and

$$G(z) = r_s(z)q_s[g(z)]. \tag{4.10}$$

If the neutron fails to undergo fission, no gamma rays will be created, while if the neutron does induce a fission, a number of gamma rays will be generated according to the induced fission distribution $f_i(n)$. Further, each of the neutrons that are generated in the same event needs to be accounted for and independently treated again in the same type of equation. In the source event, one has both the initial neutrons that can later create more gamma rays, as well as a number of gamma rays created directly with probabilities described by the number distribution of spontaneous fission, $f_{sf}(n)$.

In Paper II the same equations are also derived from a single joint formalism treating both the neutrons and the gamma rays together. An additional benefit then is that the joint moments of neutrons and gamma rays can be directly derived. For a longer and more complicated derivation of neutron and gamma ray factorial moments (which is valid only for the individual moments but not for the mixed ones), and the effect of multiplication on them, we refer to Oberer's thesis [20].

4.3 Probability distribution

As was described in Section 3.2.1, the factorial moments of the probability distribution $P(n)$ can easily be obtained by repeated derivations of the generating function $H(z)$. Whereas the usage of factorial moments is well documented the calculation of the probability distribution on the other hand is a new type of analysis. such probabilities were previously calculated only for a few low order terms. Here we embark on the calculation of the full probability distribution.

Using the definition of Eq. (3.1) one can also find an analogous way to express the probabilities as the Taylor expansion of the PGF:

$$P(\nu = n) = \frac{1}{n!}\frac{Q^n(z)}{dz^n}\bigg|_{z=0}. \tag{4.11}$$

The main difference is that, compared to the factorial moments, the expression is evaluated at z equal to zero rather than at $z = 1$. For the higher order terms, this will create additional terms as compared to the expressions for the factorial moments. Since PGFs have the property that $Q(z = 1) = 1$, many terms will be unity, especially when encountering multiple internal derivatives. In the case of the probabilities however, this is not the case when the implicit functions such as Eq. (4.6) are evaluated at $z = 0$. A great benefit however is that factorial moments can

only be used to reconstruct the probability distribution once the moments up to high orders are known [21], however from the expressions for the probability distribution the expressions for the same order factorial moments can be found as a simplified limiting expression.

A large difference when comparing the factorial moments and the probabilities is that the 0-th factorial moment is just equal to unity, while the lowest order probability is actually a very important quantity that needs to be calculated for the sake of completeness. The probability $P(n)$ will contain terms corresponding to all previous probabilities $P(m),\ \forall\ \ m < n$. Consequently, the lowest order probability, corresponding to $h(0)$ in Eq. (4.6), needs to be calculated. Inserting $z = 0$ gives the following:

$$p_1(0) = (1 - \mathrm{p})z + \mathrm{p}q_i[h(z)]\big|_{z=0} = \mathrm{p}q_i[p_1(0)] = \mathrm{p}\sum_{n=0}^{N} p_i(n)[p_1(0)]^n, \tag{4.12}$$

where $p_1(0)$ is the probability to generate 0 neutrons when starting with a single neutron. Intuitively it might seem that Eq. (4.12), which is a N-th order polynomial equation in $p_1(0)$, is impossible to solve since no generated fission would lead to keeping the single neutron, and a fission would normally lead to even more neutrons. However, in a fission process it is possible to generate also zero neutrons. As can be seen in the above equation the probability $p_1(0)$ is closely linked to the first collision probability p.

In the case of ^{240}Pu N is limited to 8 and Eq. (4.12) has a single positive root which can be found. A more detailed investigation and a derivation of the expressions for the probabilities of the number of generated neutrons and gamma rays can be found in Paper I. Also in the case of gamma rays a high order polynomial equation needs to be solved for the initial probability $f_1(0)$, the order of that equation is once again decided by the number of *neutrons* that can be generated in an induced fission event.

The important first collision probability, p, is listed in Table 4.1, for the metallic plutonium samples that were used in a comparison between the analytical model and simulations made using the Monte Carlo code MCNP-PoliMi [22].

Sample	Mass (kg)	p	σ
1	0.335	0.0852	< 0.0002
2	2.680	0.1678	< 0.0002
3	9.047	0.2461	< 0.0002

Table 4.1: Probability to induce fission, p, for one neutron depending on the mass of the sample. The metal spheres have a composition of 80 wt-% ^{239}Pu and 20 wt-% ^{240}Pu, and a density of 15.9 g/cm^3.

The probabilities $P(n)$ and $F(n)$ for neutrons and gamma rays, respectively,

needs to be calculated up to high order to ensure that the probability distribution is complete. This can be achieved by calculating the accumulated probability and stopping the calculation once a certain threshold is reached such that $\sum_{n=0}^{N} P(n) \geq 0.99$, if 99% of the probability distribution is desired. For the number of generated particles this can go as high has $N = 100$ depending on the size and composition of the sample.

In Figure 4.2 excellent agreement between simulations done using the code MCNP-Polimi and the analytical model is shown. To ensure that the comparison is fair the value of p is taken from the MCNP-PoliMi simulations, and the nuclear data constants are taken from the same database. Compared to the distribution of the spontaneous fission, which is the limiting case for an infinitesimal sample, the probability distribution acquires a tail, which means that there is a increasing probability to have larger neutron bursts generated by an increased amount of induced fissions.

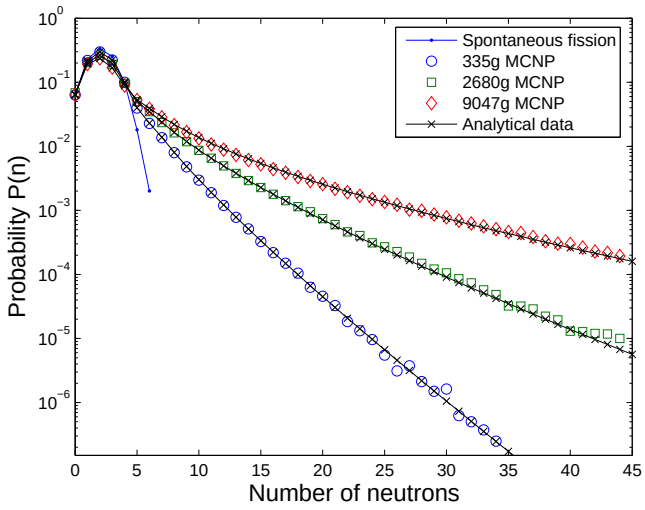


Figure 4.2: Probability distribution of neutrons generated for three samples of Pu-metal spheres. The results of the analytical model are compared to Monte Carlo simulations using MCNP-PoliMi.

Figure 4.3 shows the probability distribution for gamma rays for the same samples as used in the neutron analysis. Similarly to neutrons also the gamma rays acquire a tail to the distribution. A major difference though is that the gamma rays have a wider distribution from fissions, as many as 20 can be generated in a single fission. Considering the fact that larger numbers of gamma rays are generated also in small samples, they could be a good candidate for multiplicity assay compared to the neutrons. However, these are the number of generated particles, and when also accounting for absorption and detection the situation can change drastically.

It can be noted that for the high order probabilities especially for neutrons, the

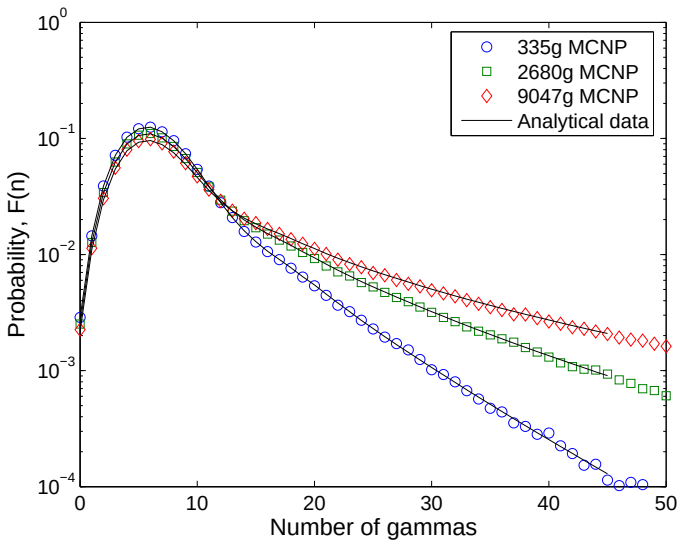


Figure 4.3: Probability distribution of gamma rays and the comparison with MCNP-PoliMi for three sample masses.

simulated results show some statistical scattering. This can be amended by running longer simulations, but that can become rather time consuming. In the same spirit, the analytical model is well suited for doing parametric studies to investigate the dependance for example on the first-collision probability. Once the formulae have been derived, it is straightforward to evaluate them with different values of the parameters such as done for p in Fig. 4.4.

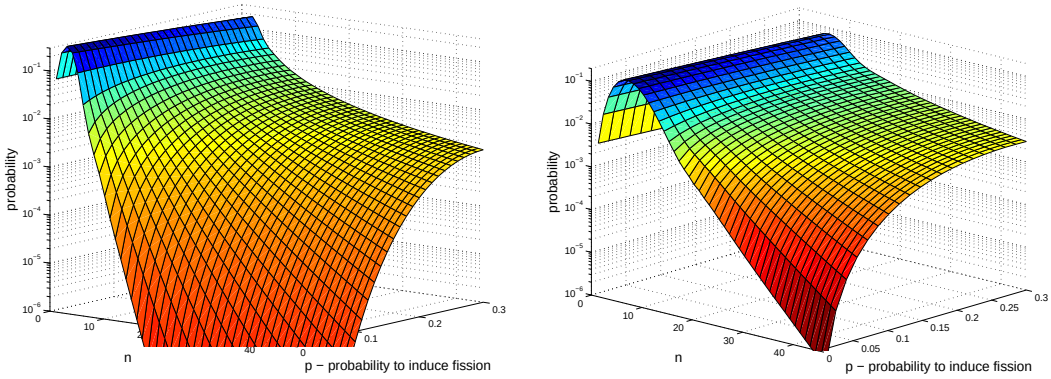


Figure 4.4: The dependance on the first-collision probability, p, for neutrons and gamma rays, respectively.

The value of p was changed in a range corresponding to tens of grams up to multiple kilogram quantities. The high order probabilities continue to increase until we reach a critical system at $\mathrm{p} = 1/\nu_{i,1}$; beyond that point, the calculations become irrelevant.

4.3.1 What about absorption?

The process of absorption can be modelled in different ways yielding the same result but different equations. In the case of neutrons, the probability of having an internal absorption in the sample is very small. The absorption can be modelled together with the induced fission as one single reaction, yielding a certain number of new neutrons (zero in the case of absorption). To use this approach the first-collision probability, p, needs to be changed to be the probability of either absorption of fission p′:

$$\widetilde{p}_i(n) = \frac{\mathrm{p}' - \mathrm{p}}{\mathrm{p}'}\delta_{n,0} + \frac{\mathrm{p}}{\mathrm{p}'}p_i(n). \tag{4.13}$$

The benefit of such an approach is that the master equations remain the same, while only some of the input parameters change. For gamma rays the same procedure does not work, and the effect of absorption is two-fold: direct absorption of gamma rays; and also absorption of neutrons which will reduce the number of induced fissions and thus reduce the number of generated gamma rays.

The alternative approach is to define generating functions also for the process of absorption which can be described with a leakage probability, l, and the absorption is then the reciprocal probability $(1 - l)$. Formulating it as a probability generating function in the case of neutrons and gamma rays respectively gives the following equations:

$$\ell_n(z) = l_n z + (1 - l_n), \tag{4.14a}$$

$$\ell_\gamma(z) = l_\gamma z + (1 - l_\gamma), \tag{4.14b}$$

with l_n and l_γ being the leakage probability of neutrons and gamma rays, respectively.

4.3.2 Detection probabilities

Detection is a process that occurs to a generated particle which escaped absorption and then was detected with a certain detection probability, ϵ. Like in Eqs. (4.14) this can be implemented using PGFs:

$$\varepsilon_n(z) = \epsilon_n\, z + (1 - \epsilon_n), \tag{4.15a}$$

$$\varepsilon_\gamma(y) = \epsilon_\gamma\, z + (1 - \epsilon_\gamma). \tag{4.15b}$$

The benefit of this approach is that new master equations for the detection statistics are easily formulated in accordance with:

$$h_d(z) = h[\ell_n\{\varepsilon_n(z)\}] \quad , \quad H_d(z) = H[\ell_n\{\varepsilon_n(z)\}], \tag{4.16}$$

and in an analog manner for the gamma ray equations. Note can be made that the equations needed for the 0-th order probability and the higher order derivatives changes since new terms are introduced.

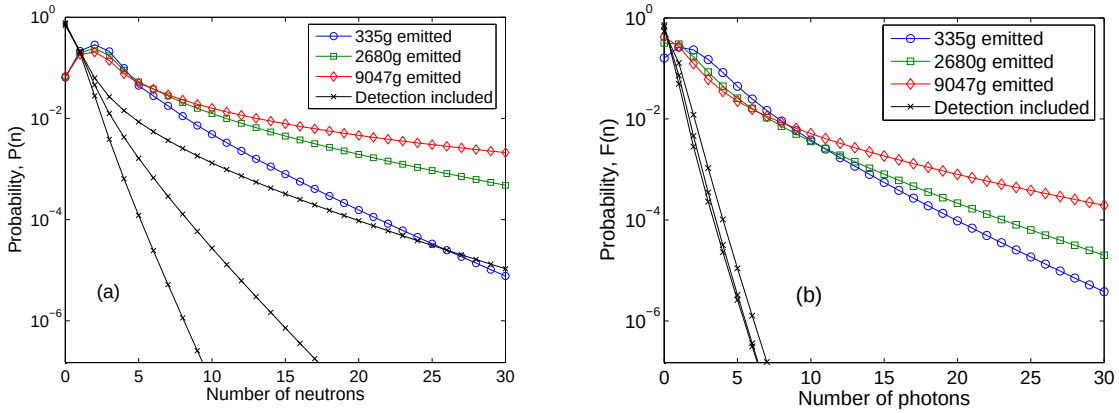


Figure 4.5: Probability distributions with detection included. The results are obtained from the analytical models. The shape of the curves will be dependent on the detector efficiency ϵ_x, which was taken as 10% for neutrons (a) and 20% for gamma rays (b) in these plots.

The effect of including absorption is mostly visible for the gamma rays since a high-Z material such as plutonium is highly self-shielding for the gamma rays while the neutrons are to large extent unaffected by absorption. When also adding the process of detection, the probability distributions reduce even further as is seen in Fig. 4.5. The "emitted" lines are the result when absorption is taken into account, the large change for gamma rays is apparent when comparing to Fig. 4.3. It is very interesting to note that the probability to detect the first few multiples is higher for a small sample than for a larger one. The probabilities $P(n)$ and $F(n)$ are the probability per source event though, and in a larger sample the rate of such events will be higher. it is also apparent that although the gamma rays have higher initial multiplication the neutrons exhibit larger probability of detecting higher order multiples for this type of sample.

It is worth noting that the detection probabilities could be connected to detection rates, by correlating to the number of source events per mass unit of the specific material. It means that the probability distribution offers an alternative route to finding the detection rates compared to using the factorial moments investigated in References [15,16,19,23,24]. Compared to the theory mentioned in Section 3.2, there is no mention of alternative source events other than spontaneous fission. Especially in materials in oxide or fluoride form there are usually a large number of neutrons created through (α, n) events. As was noted by Böhnel though, this can also be simply modelled by creating a compound source and that approach will be utilized in the next chapter.

CHAPTER

5

MULTIPLICITY THEORY AND PARAMETER UNFOLDING

Detection rates for neutrons have already been briefly presented (Sec. 3.2). Here a deeper investigation will be presented which also derives the detection rates of gamma rays and even mixed multiples. Due to the complexity of the resulting multiplicity rate expressions, artificial neural networks were applied for the unfolding problem.

5.1 Multiplicity theory

Multiplicity theory has long been applied to neutron counting for assay of nuclear materials or nuclear waste. In the previous chapter similar theory for gamma rays was outlined as the basis for possibly enabling also gamma ray multiplicity counting. This is achieved by converting factorial moments into detection rates depending on parameters which vary with the sample mass, because the distribution corresponding to one source event cannot be measured. By doing a novel extension of the formalism also to joint moments as will be demonstrated, there are nine auto- and cross factorial moments when counting up to third order for neutrons and gamma rays. The main problem when extending the theory is the complexity of the equations which becomes more advanced with the additional parameters that are introduced especially for the gamma rays. Simple analytical inversions of the formulae are no longer available; therefore, the introduction of artificial neural networks (ANN) [25] is outlined in Sec. 5.2. The non-linearity of the equations and the redundancy in the number of measured rates compared to the number of output parameters is well handled by the ANN.

5.1.1 Neutrons, gamma rays and combinations

When measuring the detection rates of neutrons and gamma rays together with the joint multiples there are a total of three neutron rates (S, D, T), three gamma ray rates (S_γ, D_γ, T_γ) and also three combined rates ($D_{n\gamma}$, $T_{nn\gamma}$, $T_{n\gamma\gamma}$). A big challenge of measurements of this type is how to detect the different types of radiation. Traditionally ^{3}He-detectors were used for the neutron assay, and while they remove the neutron from the system avoiding the risk of a single particle giving pulses in more than one detector, they are on the other hand not capable of detecting gamma rays.

Within nuclear safeguards the prospects of using organic liquid scintillation detectors have recently been investigated [26, 27]. This type of detector is capable of detecting not only both neutron and gamma rays, but they are also giving different pulse signatures such that one can discriminate which particle caused each pulse. Other systems using plastic scintillation detectors [28] were also sensitive to both types of radiation, but they did not offer the possibility to see which type of radiation caused each pulse, which would make full joint multiplicity counting impossible.

Now that it is possible to detect both types of radiation with a single detector setup in a relatively easy manner, the motivation for doing so must also be investigated. One reason for finding and using also the joint moments can be illustrated by the covariance between neutrons and gamma rays. As shown in Fig. 5.1 there is a very strong relationship between the two particle types, for increasing values of the probability to induce fission (p). The probability to induce fission is not normally the parameter found in the formulae for the detection rates as seen in Sec. 3.2, instead one usually converts it to the so called neutron leakage multiplication, $\mathbf{M}$, through:

$$\mathbf{M} \equiv \frac{1-\mathrm{p}}{1-\mathrm{p}\nu_{i,1}}; \qquad \mathrm{p}\nu_{i,1} < 1. \tag{5.1}$$

5.1.2 Mixed sources

In fissile material there may be other sources present besides spontaneous fission. Neutrons produced through (α, n) reactions can also give rise to reaction chains producing higher order multiples caused by the multiplication process of the neutrons. To account for the presence of single neutron producing events, one introduces the statistics of the total source events as a weighted average of the two processes of spontaneous fission and (α, n) reactions [15], [29]. Denoting the generalized source event by a subscript s, the source moments of neutrons can be described as follows:

$$\nu_{s,n} = \frac{\nu_{sf,n}(1+\alpha\delta_{1,n})}{1+\alpha\nu_{sf,1}}. \tag{5.2}$$

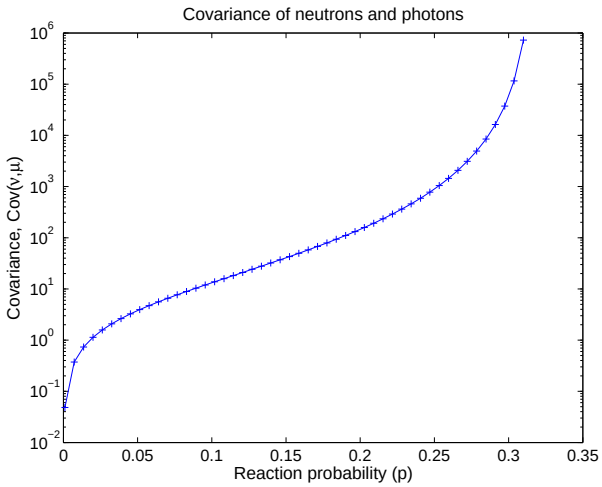


Figure 5.1: Covariance between neutrons and gamma rays, defined as: $\mathrm{Cov}(\nu, \mu) = \langle \nu\, \mu \rangle - \langle \nu \rangle \langle \mu \rangle$.

The factor α is defined as

$$\alpha = \frac{Q_\alpha}{Q_f \nu_{sf,1}}.$$

Here Q_f and Q_α are the intensities of spontaneous fission and (α, n) processes, respectively, and hence α stands for the ratio of (α, n) neutrons to spontaneous fission neutrons. Similarly the source moments for gamma rays can be modified to account for gamma rays produced in the (α, n) events. A recent investigation into the derivation of the effect of (α, n) events on the neutron detection rates can be found in Paper III.

An additional benefit of having a formalism for other neutron events is that it could be used as basis for a formalism for an actively interrogated sample where a neutron accelerator producing neutrons is used to induce reaction chains in the sample.

5.1.3 Formulae for detection rates

To find the detection rates such as those listed in Eqs. (3.5)-(3.7), one needs to first derive the factorial moments of the same order. For neutrons they are just the n-th derivative of Eq. (4.7), evaluated at $z = 1$, giving the following expression for the first three moments:

$$\nu_1 = \frac{\mathbf{M}}{(1 + \alpha \nu_{sf,1})} \nu_{sf,1}(1 + \alpha), \tag{5.3}$$

$$\nu_2 = \frac{\mathbf{M}^2}{(1 + \alpha \nu_{sf,1})} \left[\nu_{sf,2} + \left(\frac{\mathbf{M} - 1}{\nu_{i,1} - 1} \right) \nu_{sf,1}(1 + \alpha) \nu_{i,2} \right], \tag{5.4}$$

$$\begin{aligned}\nu_3 \;&= \frac{\mathbf{M}^3}{(1+\alpha\nu_{sf,1})}\Bigg[\nu_{sf,3} + \left(\frac{\mathbf{M}-1}{\nu_{i,1}-1}\right)\left[3\nu_{sf,2}\nu_{i,2} + \nu_{sf,1}(1+\alpha)\nu_{i,3}\right] + \\ &\qquad + 3\left(\frac{\mathbf{M}-1}{\nu_{i,1}-1}\right)^2 \nu_{sf,1}(1+\alpha)\nu_{i,2}^2\Bigg].\end{aligned} \tag{5.5}$$

Use have been made of the combined source moments defined in Eq. (5.2).

Similarly for gamma rays the equivalent expressions can be found from derivations of Eq. (4.10). This leads to the expression:

$$\mu_1 = \frac{\mu_{sf,1} + \alpha\nu_{sf,1}}{(1+\alpha\nu_{sf,1})} + \frac{\nu_{sf,1}(1+\alpha)}{(1+\alpha\nu_{sf,1})}\mathbf{M}_\gamma, \tag{5.6}$$

with

$$\mathbf{M}_\gamma \equiv \frac{p\mu_{i,1}}{1-\mathrm{p}\nu_{i,1}} \tag{5.7}$$

being the *gamma leakage multiplicity per one initial neutron*. Note that $\mathbf{M}_\gamma$ is not strictly a new parameter since it depends foremost on the fission probability p which occurs similarly in the expression for the neutron leakage multiplication $\mathbf{M}$. The second and third factorial moments are:

$$\begin{aligned}\mu_2 = \;& \frac{\mu_{sf,2}}{1+\alpha\nu_{sf,1}} + 2\frac{(\mu_{sf,1}+\alpha\nu_{sf,1})}{(1+\alpha\nu_{sf,1})}\,\frac{\nu_{sf,1}(1+\alpha)}{(1+\alpha\nu_{sf,1})}\mathbf{M}_\gamma + \\ & + \frac{\nu_{sf,2}}{(1+\alpha\nu_{sf,1})}\mathbf{M}_\gamma^2 + \frac{\nu_{sf,1}(1+\alpha)}{(1+\alpha\nu_{sf,1})}\,g_2\end{aligned} \tag{5.8}$$

and

$$\begin{aligned}\mu_3 = \;& \frac{1}{(1+\alpha\nu_{sf,1})}\Bigg[\mu_{sf,3} + 3\mu_{sf,2}\frac{\nu_{sf,1}(1+\alpha)}{(1+\alpha\nu_{sf,1})}\mathbf{M}_\gamma + \\ & + 3(\mu_{sf,1}+\alpha\nu_{sf,1})\left\{\frac{\nu_{sf,2}}{(1+\alpha\nu_{sf,1})}\mathbf{M}_\gamma^2 + \frac{\nu_{sf,1}(1+\alpha)}{(1+\alpha\nu_{sf,1})}\,g_2\right\} + \\ & + \nu_{sf,3}\mathbf{M}_\gamma^3 + 3\nu_{sf,2}g_2 + \nu_{sf,1}(1+\alpha)g_3\Bigg].\end{aligned} \tag{5.9}$$

In Eqs. (5.8)-(5.9) g_n are the factorial moments of the gamma ray distribution initiated by a single neutron, which are defined as [29]:

$$g_n = \left.\frac{d^n g(z)}{dz^n}\right|_{z=1}. \tag{5.10}$$

Factorial moments alone are not enough to relate to the measurable quantities such as singles, doubles etc. First it is needed to find the factorial moments $\widetilde{\nu}_k$

of the *detected* neutrons per one initial event. This requires the introduction of the detector efficiency and for the first few moments these factorial moments are obtained as:

$$\widetilde{\nu}_1 = \varepsilon_n \nu_1, \tag{5.11a}$$

$$\widetilde{\nu}_2 = \varepsilon_n^2 \nu_2, \tag{5.11b}$$

$$\widetilde{\nu}_3 = \varepsilon_n^3 \nu_3. \tag{5.11c}$$

Additionally it is important to account for the fact that a measured doublet could be the result of detecting two particles from a higher order multiplet, and that the detection *rates* C_k, $k = 1, 2, 3$ are also related to the total source intensity. Using C_k as the notation for the k-th order multiplet (such that $C_1 = S$, $C_2 = D$ etc.) and the total neutron source rate $Q_s \equiv Q_f + Q_\alpha$, it follows that

$$C_k = Q_s \left\langle \binom{\widetilde{n}}{k} \right\rangle = Q_s \varepsilon_n^k \sum_n \frac{n!}{k!(n-k)!} P(n) = Q_s \frac{\widetilde{\nu}_k}{k!}. \tag{5.12}$$

Using the α ratio it is possible to rewrite the source rate as depending only on the rate of fission, F: $Q_s = F(1 + \alpha\nu_{sf,1})$. Using the equation (5.12) together with (5.11) on the factorial moments of neutrons, gives the previously mentioned neutron detection rates, Eqs. (3.5)-(3.7).

For gamma rays a parameter for a single gamma ray generating source can be introduced in an analogue way as for the neutron case. The gamma ratio, γ, is defined as the ratio of the single gamma ray source strength, Q_γ, and the neutron source strength, Q_s. The gamma detection rates are now given by the following expressions:

$$S_\gamma = \left[\gamma F(1 + \alpha\nu_{sf,1}) + F(1 + \alpha\nu_{sf,1})\widetilde{\mu}_1\right], \tag{5.13a}$$

$$D_\gamma = F(1 + \alpha\nu_{sf,1})\frac{\widetilde{\mu}_2}{2}, \tag{5.13b}$$

$$T_\gamma = F(1 + \alpha\nu_{sf,1})\frac{\widetilde{\mu}_3}{3!}. \tag{5.13c}$$

For mixed detection rates the formulae grows longer and more complex and can be found in Paper IV.

5.2 Parameter unfolding

The application of artificial neural networks on the unfolding of the sample rates was the main focus of Paper V, while the derivation of the required formulae can be found in Paper IV. This section will investigate some of the results and findings of the prospect of using ANNs for parameter unfolding from neutron and gamma ray detection rates.

5.2.1 Artificial neural networks

Since the neutron rates can be analytically inverted they present a good opportunity for validating the design and layout of the neural network [30]. Figure 5.2 shows the schematic structure used when trying to unfold three sample parameters (fission rate, alpha ratio and neutron leakage multiplication).

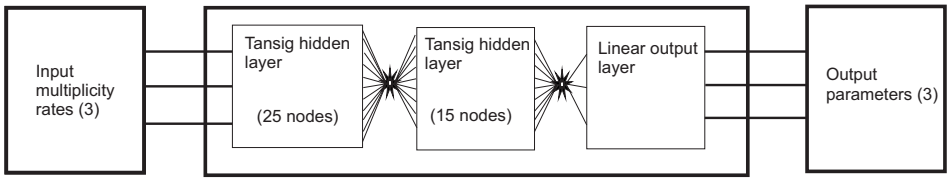


Figure 5.2: ANN schematic.

The network contains two hidden layers, each with a number of nodes which connect to the nodes of the next layer. In the end a number of output parameters are produced according to the design of the network. It can be noted that although the fission probability p is an important parameter, it might be more straightforward to unfold the neutron leakage multiplication, $\mathbf{M}$, due to the way the two terms appear in the formulae for the detection rates. These types of considerations were tested with trial and error to achieve optimized networks.

5.2.2 Validation and training

The application of ANNs require large sets of training and validation data. In the case of unfolding the neutron detection rates it is the three detection rates that are the input data while the parameters p, α and F are varied over ranges of values corresponding to different sample masses and compositions. Figure 5.3 shows a sample of the generated training data.

	fission rate (F)	α	p
max. abs. rel. error (%)	0.0001	0.0021	0.0001
mean error of training (%)	-1.98e-9	-4.14e-7	4.96e-9
standard deviation of training (%)	7.56e-6	2.11e-4	3.24e-5
mean error of test data (%)	1.28e-6	-7.15e-6	2.56e-6
standard deviation of test data (%)	9.34e-6	1.42e-4	3.29e-5

Table 5.1: Training results of ANN, using the neutron equations to simulating large plutonium samples in the kg-range.

The training and testing data show that for the neutron rates extremely low errors are to be expected when unfolding the sample parameters (Tab. 5.1). The

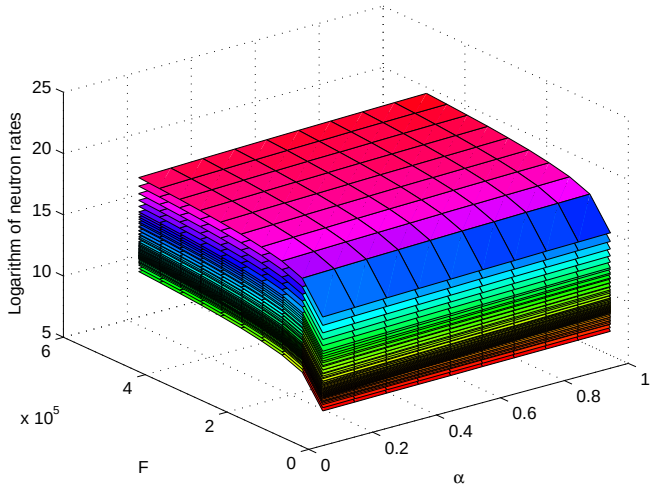


Figure 5.3: Training data that are used to calculate F, α and p. The surfaces correspond to different values of p while F and α are represented on the x, y-axis, respectively.

shown inaccuracies are small enough to warrant usage of ANNs for unfolding also of neutron rates measured in neutron multiplicity counters. A motivation for using the ANNs rather than the analytical unfolding is that the neutral network is less sensitive to error propagation from measurement uncertainties, provided that the network has been properly trained and validated.

5.2.3 Results

As more detection rates are added to the neural network it also introduces new parameters. When using both neutron and gamma ray rates, there are a total of 6 inputs, but the number of sample parameters is also increasing with the parameters γ, $\mathbf{M}_\gamma$ and ε_γ being introduced. For the network to perform adequately there is a need to increase the complexity of the internal structure, while also spending a bigger computational effort of training. As shown in histograms in Fig. 5.4, the errors in unfolding of the added parameters from the gamma ray detection rates are also kept promisingly low.

For successful application of neural networks on measurement data there is a need to investigate the sensitivity of the network. The neutron rates were perturbed by random noise of different levels to investigate how the unfolding of the most important parameter, the fission rate F, is affected. Figure 5.5 shows the effect of 1%, 5% and 10% noise on the input data. It is significant to note that it is especially the singles rate that will induce errors in the predicted fission rate. The extent of the error though is quite small in comparison to the introduced perturbation. In measurements it is especially the triples rate that has the largest statistical un-

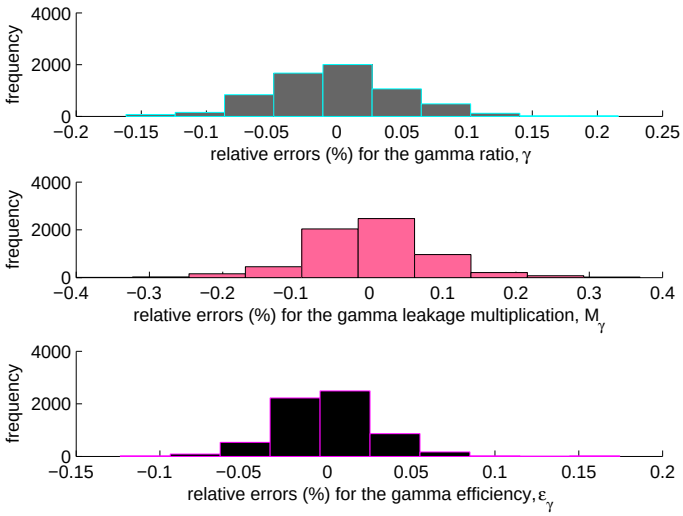


Figure 5.4: Histograms of the relative errors of some of the parameter occurring in the expressions for the gamma ray detection rates.

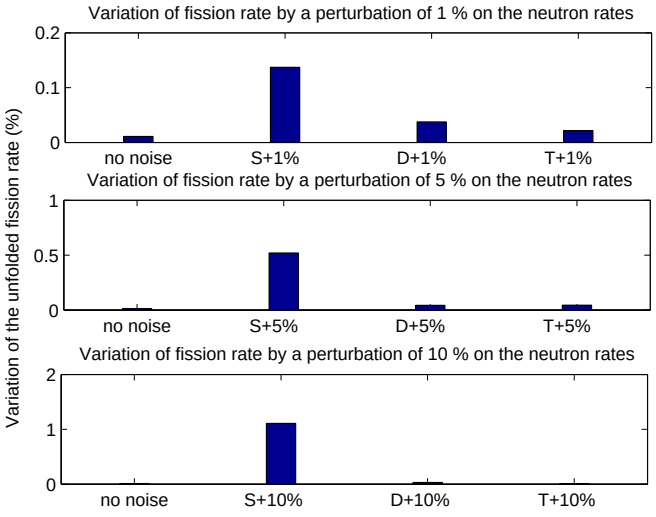


Figure 5.5: Influence of noise on different neutron rates, when unfolding the fission rate.

certainties [31, 32], fortunately the results show that it is the triples that have the smallest effect on the unfolded fission rate. This is a very promising feature of the neural network approach, especially if considering applying it for the widespread neutron multiplicity analysis.

Sensitivity analysis was also performed for the case of using all 9 neutron, gamma ray and mixed rates. Since this is the case of the maximum number of available

inputs it implies large requirements on the accuracy of the measurement. That all the multiplicity rates would be accurately acquired is unlikely. Therefore, the ANN was tested when omitting different inputs. An additional benefit of such an approach is that it can show the relative importance of different inputs, by showing which introduces the largest uncertainties when it is omitted.

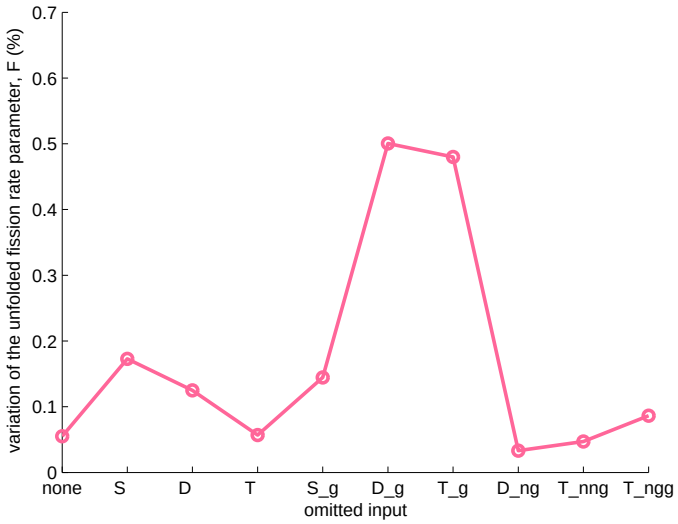


Figure 5.6: Maximal variation of the unfolded fission rate after omitting one of the inputs.

The results shown in Fig. 5.6 indicate that the variation in the unfolded fission rate is relatively small when omitting most of the input rates. For the case of doubles and triples for gamma rays the generated variation is much larger though. This also confirms the importance of measuring the gamma rates when attempting unfolding, since almost all parameters occur in the expression of those rates.

While very promising results are shown for the application of neural networks, it is also important to understand the complexity of properly designing and using them to solve the problem at hand. Choosing which parameters to unfold, such as $\mathbf{M}$ and $(1-\alpha)$ rather than p and α, can improve the accuracy. Likewise choosing the inputs in a sophisticated way is of great importance. Using S, D/S, T/S rather than S, D, T means that the input parameters are more independent and once again better accuracy in achieved. The expertise within neural networks belongs to the co-author of Papers IV-V: *Senada Avdic*, who performed the analysis described here.

CHAPTER

6

SCINTILLATION DETECTORS AND NEUTRONS

This chapter presents an analytical model of how the interaction between neutrons and organic scintillation detectors results in light pulses. The idea is to create additional understanding of the interaction physics while also giving new ideas in how to best use scintillation detectors when trying to understand the incident neutron radiation. In the words of the neutron: "Let there be light!"

6.1 Theory

A way of detecting and identifying nuclear materials in nonproliferation applications is by use of organic scintillation detectors in both liquid and plastic form. The mechanism of neutron detection is light generated by scatterings on hydrogen (H) and carbon (C) nuclei, which are the main constituents of the detection material. These detectors are as previously mentioned also sensitive to gamma rays, but by using pulse shape discrimination those pulses can be discarded and a pure neutron pulse analysis is then achievable [33, 34].

The light produced in the scintillation material is scattered inside the detector until it reaches a photo multiplier tube (PMT) - Fig. 6.1. This device transforms the light quanta to electrons via a photocathode. The electrons are then multiplied through a cascade process, and thus generate an electric signal. In the following it will only be the parts prior to the PMT that will be modelled, meaning the initial generation of light from energy transferred by neutrons. A first investigation into finding individual neutron collision contributions was reported by Hermsdorf et al. [36]. Since this problem was so far only handled by Monte Carlo methods we

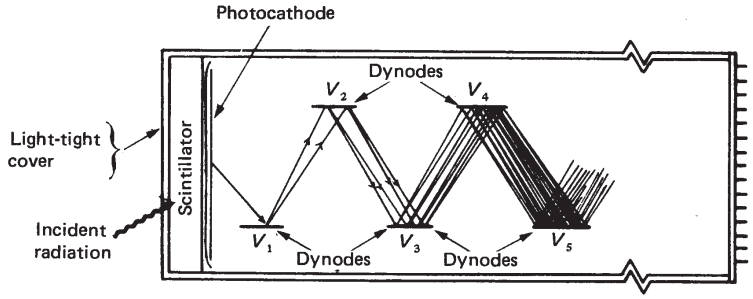


Figure 6.1: Photo multiplier tube (from [35]).

wanted to have also an analytical model for comparison. The basis of this chapter is the results found in PAPERS VI-VII, together with some additional work [37,38].

6.1.1 Collision probabilities

It is intuitively clear that the distribution of the light pulse amplitudes is strongly depending on the probability of the neutron to undergo certain reactions in the detector. In an analytical model this step, which depends on the material cross section and the system geometry, presents a challenge which is hard to handle without approximations. The collision probability for an isotropic neutron source distributed uniformly in a sample of volume V, is given by the so-called Peierls-formula [39]:

$$P = \frac{\Sigma}{V}\int_V d\mathbf{r}\int_{\mathbf{V}} \mathbf{dr}'\frac{\mathbf{e}^{-\mathbf{\Sigma}|\mathbf{r}-\mathbf{r}'|}}{\mathbf{4}\pi|\mathbf{r}-\mathbf{r}'|^{\mathbf{2}}}. \tag{6.1}$$

For homogeneous samples, which we will assume here, and for some regular geometries such as a sphere, this expression can be brought to either a closed form, or to a form sufficiently amenable for numerical calculations. This approximation works best when the neutron undergoes several collisions which will force them to approach such a uniform isotropic source before the next collision in the sequence. For a cylinder an expression was given for this probability by Carlvik [40] in the form :

$$\begin{aligned} P_c = 1 - \frac{1}{h}\left[E_3(0) - E_3(h)\right] + \frac{4}{\pi d^2 h}\int_0^d dt\left\{E_3(t) - E_3\left[(t^2+h^2)^{1/2}\right]\right\}(d^2+t^2)^{1/2} \\ -\frac{4}{\pi d^2 h}\int_0^h du(h-u)\int_0^d t^2 dt\frac{e^{\left[-(t^2+u^2)^{1/2}\right]}}{(t^2+u^2)^{3/2}}(d^2-t^2)^{1/2} \equiv P(E), \end{aligned} \tag{6.2}$$

where

$$E_n(z) = \int_1^\infty \frac{e^{-zu}}{u^n}du$$

is the exponential integral. The height and the diameter is expressed in mean free paths, and this is where the energy dependence of the collision probability enters. However, it is not only the geometry that decides $h(E)$ and $d(E)$, but also the exact composition. In the quantitative work the composition chosen is that of the scintillation liquid BC-501A, which is commonly used in liquid scintillation detectors. Table 6.1 shows the detector dimensions in mean free paths for neutrons of energy 1 MeV.

Detector dimensions: h = d	
cm	MFPs
1	0.308022
5	1.54011
10	3.08022
15	4.62033

Table 6.1: Detector dimensions for a right cylinder, expressed in centimeters and in mean free paths for a neutron energy of 1 MeV.

6.1.2 Light pulse distributions

Since the energy transferred from the neutron inside the scintillation material is what creates the light pulses, the first step it to create a model for the energy transfer from a neutron. As can be seen in Figure 6.2, a neutron with initial energy E_0 can undergo a series of collisions, each reducing the remaining energy of the neutron which affects the amount of energy that is transferrable in later collisions.

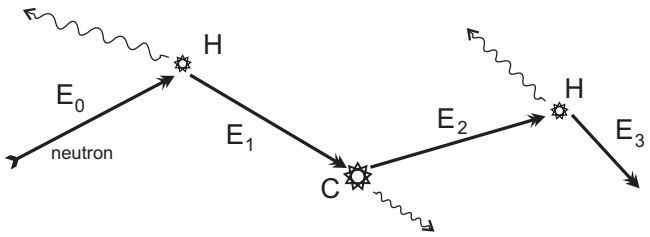


Figure 6.2: Collision sequence for e neutron of initial energy E_0. Collisions on hydrogen (H) and carbon (C) are resulting in scintillation light.

For collisions on hydrogen the energy distribution for the neutron is a simple flat distribution since the hydrogen nuclei (proton) has practically the same mass as the neutron itself, and thus a full energy transfer is possible while still maintaining conservation of momentum in the collision. The distribution in transferred energy, T, for a neutron of initial energy E_0 is:

$$p(T, E_0)dT = \frac{dT}{E_0}. \tag{6.3}$$

The transferred energy is then transformed into light, and the amount of light produced will depend on the nuclei involved and for the following calculations is taken as the empiric relation

$$L = aT^2 + bT \equiv L_h(T), \tag{6.4}$$

with L being the produced light in MeVee (MeV electron equivalent), and a, b are constants. Combining these formulae and calculating the distribution of a function of a random variable with a known distribution, the light distribution for a neutron colliding once with hydrogen is as follows:

$$f_{1h}(L, E_0) = \frac{1}{E_0\sqrt{b^2 + 4aL}}. \tag{6.5}$$

Similarly for collisions on carbon, the same type of derivation can be made. The light generation from energy transferred to carbon however is simpler and follow the relationship $L_c(T) = cT$, with c being a constant. A neutron colliding with a carbon nuclei can only transfer a fraction α of its energy in each collision, and the following light distribution is found for a single collision on carbon:

$$f_{1c}(L, E_0) = \frac{\theta(L_{max,c} - L)}{c\,(1-\alpha)\,E_0}, \tag{6.6}$$

with θ being the Heaviside function.

When calculating the amount of light produced by a certain energy transfer from two collisions the specific energy transfer in each is an unknown variable, and the formulae generated will be in the form of convolution integrals of higher and higher order when adding more collisions. The manner of creating such formulae is found in Paper VI. In the case of collisions on carbon, the linear relation between transferred energy and light means that the problem is very closely linked with the slowing down problem in a graphite moderator. Figure 6.3 show the distributions after each added collision on carbon.

Since the collision probability is given via Eq. (6.2), it is possible to use that factor to get correctly weighted light distributions for a specific detector geometry and composition. Weighting factors depending on if the collisions was on hydrogen or carbon can be created as:

$$c_H(E) = P(E)\frac{\Sigma_H(E)}{\Sigma_T(E)}, \tag{6.7a}$$

$$c_C(E) = P(E)\frac{\Sigma_C(E)}{\Sigma_T(E)}, \tag{6.7b}$$

where $\Sigma_T = \Sigma_H + \Sigma_C + \Sigma_a$ are all macroscopic cross sections. The probability that the neutron history is terminated by either absorption or leakage, is given as

$$W(E) = 1 - c_H(E) - c_C(E).$$

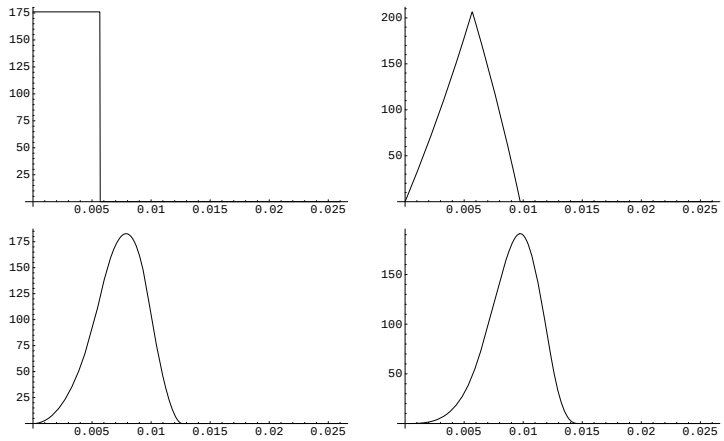


Figure 6.3: Light pulse distributions normalized to unity, for 1-4 collisions on carbon.

$W(E)$ is a quantity that has to be used when calculating the probability that a neutron history will end after a certain number of collisions of a given sequence.

A proper collision probability weighted distribution of light when colliding once with hydrogen or carbon respectively, reads as:

$$f_H(L, E_0) \equiv c_H(E_0) f_{1h}(L, E_0), \tag{6.8a}$$

$$f_C(L, E_0) \equiv c_C(E_0) f_{1c}(L, E_0). \tag{6.8b}$$

Since the equations are given in terms of light generated, L, there is a need to find the inverse relation of Eq. (6.4), giving the transferred energy as a function of the produced light, $T_h(L)$ and the equivalent for carbon. Probability distributions for any given collisions sequence can now be formulated (see Paper VII), where an example in the form of the three-collided H-C-H distribution is shown below (the last index indicating first collision):

$$f_{HCH}(L, E_0) = \int_0^{L-l_1} \int_0^{L} f_H \left[L - (l_1 + l_2), E_0 - (T_h(l_1) + T_c(l_2))\right] \times$$

$$f_C \left[l_2, E_0 - T_h(l_1)\right] f_H[l_1, E_0] W \left[E_0 - T_h(l_1) - T_c(l_2) - T_h(L - (l_1 + l_2))\right] dl_1 dl_2. \tag{6.9}$$

A real light pulse distribution which takes into account all different collisions sequences, will be the sum of the once-, twice-, etc. collided neutrons in the form of a collision number expansion:

$$f(L, E_0) = f_1(L, E_0) + f_2(L, E_0) + f_3(L, E_0) + \ldots \tag{6.10}$$

A truncation can be made at a higher number of collisions either based on the lower probability of having a high-order collision history, or alternatively, based

on the fact that additional collisions are likely to only produce minor light when the neutron is already slowed down, and thus the next order distribution will be approximately the same shape as the previous order. The separate formulae do however provide an opportunity to study the effects of collision order and sequences as will be demonstrated in the next section.

6.2 Collision history importance

The light pulse distributions without collision probability normalization have been validated and compared to simulations made using the code MCNP-PoliMi, and very good agreement was found. Considering the case of twice collided neutrons which

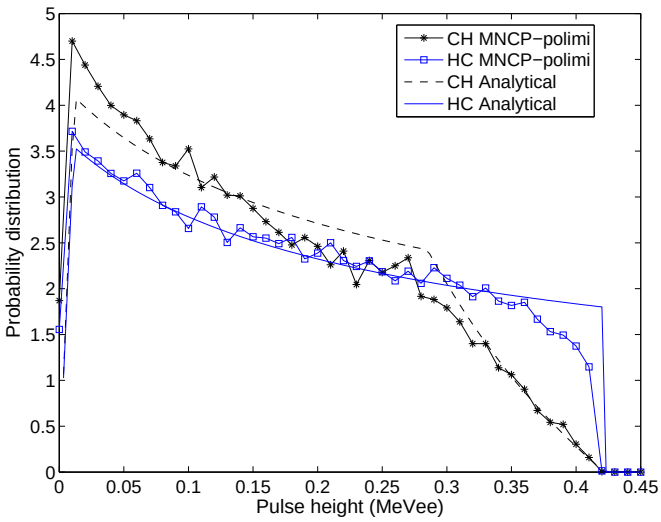


Figure 6.4: Double collision comparison with MCNP-PoliMi for neutrons of initial energy 2 MeV.

had one collision each on carbon and hydrogen, one could expect a very similar light pulse distribution for the same initial energy and light output. However, as can be seen in Fig 6.4, there are actually quite notable differences for different collision sequences, primarily arising from the limited energy transfer when colliding with carbon.

Similarly when comparing three times collided neutrons (Fig. 6.5) the differences grow even larger, and it is notable to see that the light pulse distribution might have peaks corresponding to a light output different from what could be expected from the maximum transferable energy. Also in this case the comparisons with Monte Carlo show a good agreement. It is also notable that the main simplification in the analytical model comes from the empiric formulae of light generation on different nuclei, and it seems probable that without the statistical scatter on the Monte Carlo results the shape of the curves would converge even closer to the analytical model.

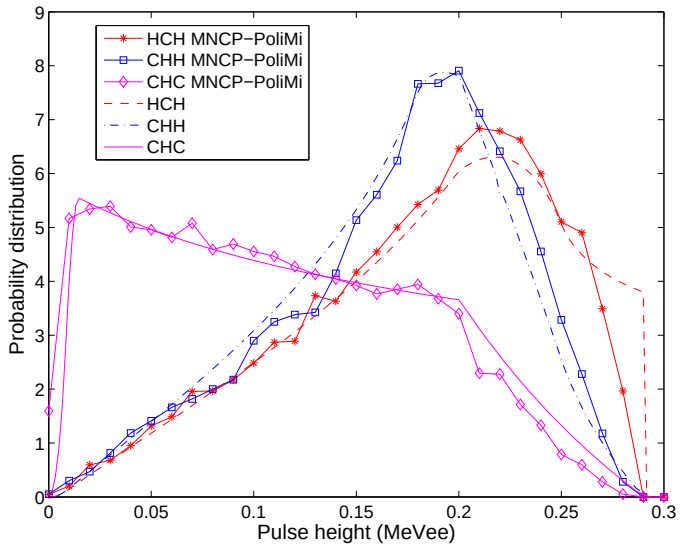


Figure 6.5: Triple collision comparison with MCNP, for neutrons of initial energy 1.5 MeV.

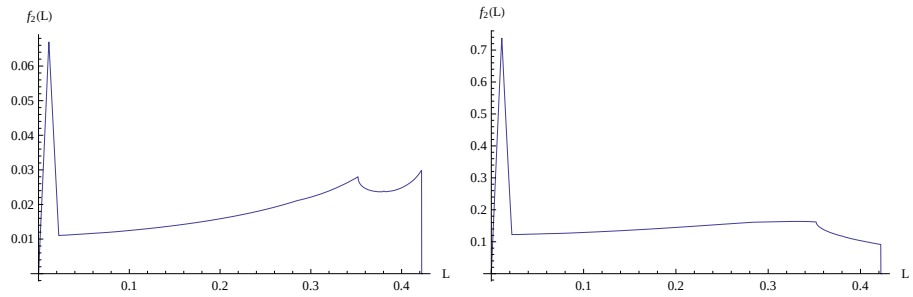
6.3 Detector models

The initial validation was made without proper collisions weighting, which is achievable by using Eqs. (6.2)-(6.7), and the termination probability $W(E)$. Such results will be presented below.

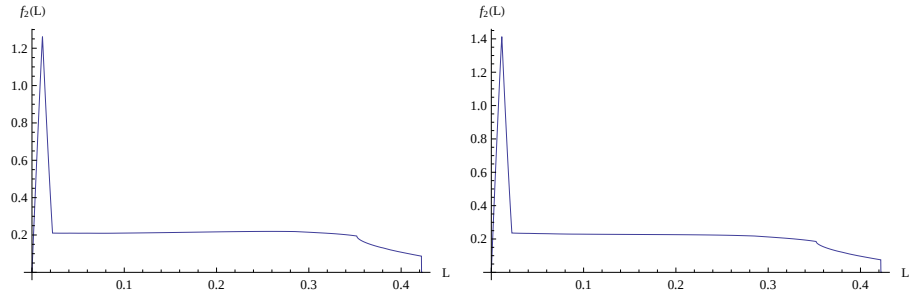
6.3.1 Impact of detector sizes

It is apparent that the order and amount of collisions greatly affect the scintillations pulses, but what about the detector size? The geometry of the detector will have a large effect on the likelihood of different collisions sequences, when some detectors are seen as "thin" by the neutron and others are seen as "thick", meaning a small or large probability to suffer a high number of collisions. In Fig. 6.6 this effect is shown for the distributions of all twice collided neutrons for 4 different detector sizes.

The fact that the scattering cross sections are not constant for different energies also changes the distributions. This can be seen especially when comparing individual collision histories for different detector sizes, such as done in Fig. 6.7. At about 0.145 MeVee light output the distribution shows a peak (Figs. 6.7(a)-6.7(b)) corresponding to a minor peak in the cross section of carbon. While such small effects would be very hard to detect in measurements, they offer insight to and understanding of the interaction process between neutrons and scintillation detectors.

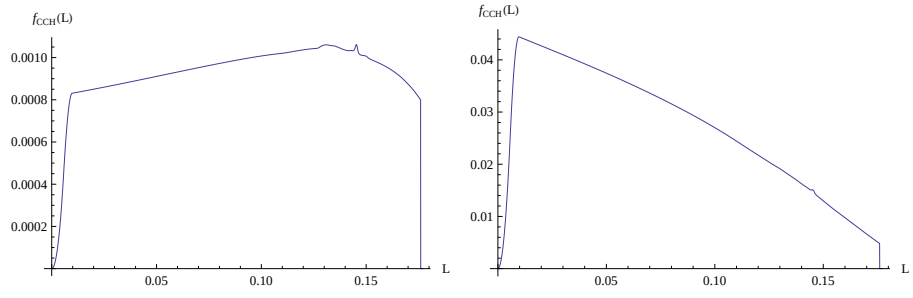


(a) 1 cm. (b) 5 cm.

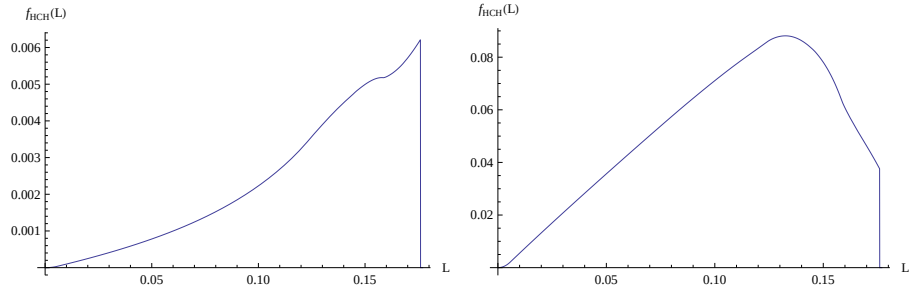


(c) 10 cm. (d) 15 cm.

Figure 6.6: Light pulse distribution for twice collided neutrons of initial energy 2 MeV for four different detector sizes. Light output L is given in MeVee.



(a) CCH, 1 cm. (b) CCH, 10 cm.



(c) HCH, 1 cm. (d) HCH, 10 cm.

Figure 6.7: Light pulse distribution for two different collision histories both in a 1 cm and a 10 cm scintillation detector (left and right figures, respectively). Incident neutron energy is 1 MeV.

CHAPTER

7

CORRELATED PARTICLES AND MEASUREMENTS

By correlated particles we refer to particles created in correlated events, such as a "superfission" as defined by Böhnel [15]. They are generated in reaction chains started with a single source event. In a very fast measurement system it is possible to measure only true coincidences of neutrons and gamma rays, thus simplifying the analysis and enhancing the nuclear materials signatures that such a system can provide. This is an example of the practical applications that the research in the previous chapters aimed at modelling and improving.

7.1 Turning theory into practice

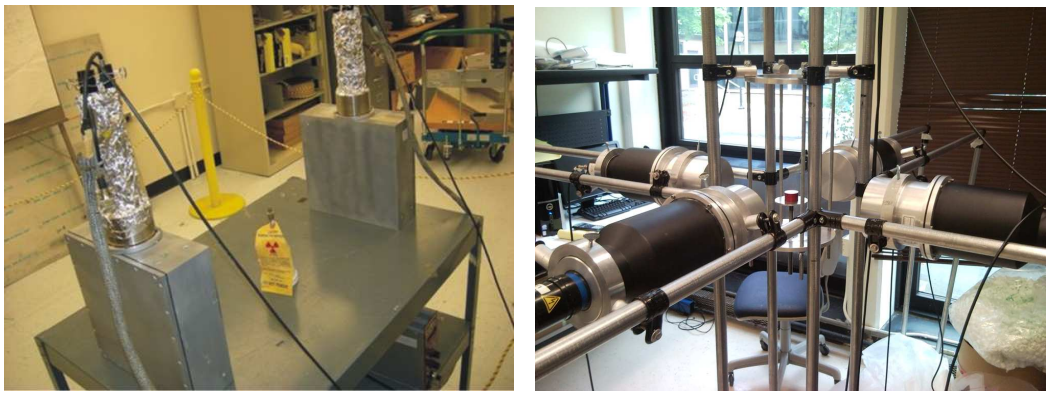
While much of the current research within safeguards concerns simulations and models (the latter being the main focus of this thesis), the work must aim at developing methods and systems that can be used in realistic situations for assuring safeguard performance. The vital part of testing the theories and seeing how they measure up to reality is best done in experimental work. In this chapter two methods: cross-correlation measurements; and mixed multiplicity counting will be detailed.

7.1.1 Which detectors?

When creating or using measurement systems for detection of nuclear material, a key issue is the type of detectors chosen. Proportionality counters, semiconductor detectors, scintillators or capture detectors are just some of the examples available. The use of scintillation detectors is a interesting one, due to the inherent property of utilising the speed of light they can often offer excellent timing characteristics.

Especially liquid organic scintillation detectors are suitable for use when detecting dual radiation, i.e. both neutron and gamma rays, while also being able to determine which pulse came from what as previously mentioned.

With fissile samples sending out large amounts of *both* neutrons and gamma rays, the scintillation detectors are suitable for assaying this type of material. The detectors can also be found in a variety of sizes and geometries, some of which can be seen in Fig. 7.1 showing the detection setups for cross-correlation measurements and multiplicity counting as used in Papers VIII-IX.



(a) Oak Ridge national Labs. (b) University of Michigan.

Figure 7.1: Measurement setups used for cross-correlation measurements and other experiments.

Using fast detectors is only optimized if they are coupled to a data acquisition system capable of handling the fast pace of information. Such equipment is available in the form of fast digitizers able to read and output the pulse information in digital format at rates approaching the GHz-range, i.e. sampling at close to nanosecond intervals.

With particle identification being a key issue in environments containing both neutron and gamma ray radiation, there is another type of detector which also has interesting properties: capture-gated detectors. In such a detector, a pulse is produced by neutrons and gamma rays interacting with the scintillation material. In addition to that, neutrons can also undergo capture and thus create an additional pulse. This pulse is very distinct, and can be used to verify that a previous pulse was generated by a neutron. This property is achieved by doping the detector material with a neutron absorber such as boron or lithium. A more detailed description and discussion of such detectors can be found in References [41–44]. The drawback of the design though is that for the capture to be likely the neutron needs to be moderated to much lower energies, which normally require a large number of scatterings. For this to occur frequently the detector volume needs to be quite large, and the process of slowing down, capture and the subsequent generated capture pulse will be on the

order of microseconds which is much longer than the pure scintillation pulse lasts. Having a high count rate would then be impairing due to the large probability that an additional particle interacts with the detector within the time frame of the neutron capture.

7.1.2 Pulse shape discrimination

With pulse shape discrimination (PSD) being of utmost importance in the proposed applications, it is important to understand how the PSD works. In an organic scintillator most of the observed scintillation light comes as a prompt florescence, but also a minor longer-lived component can be observed. While the prompt part has a decay time on the order of nanoseconds, the delayed part has a decay time in the order of hundreds of nanoseconds. The slow component depends primarily on the energy loss dE/dx of the exciting particle and is higher for large values of dE/dx. This makes it possible to distinguish heavier particles such as α-particles, from neutrons (recoil protons) and gamma rays (recoil electrons). The behaviour of the scintillation pulses generated by different types of radiation can be seen in Figure 7.2.

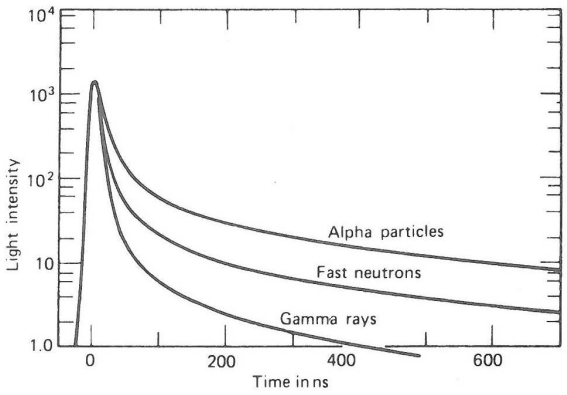
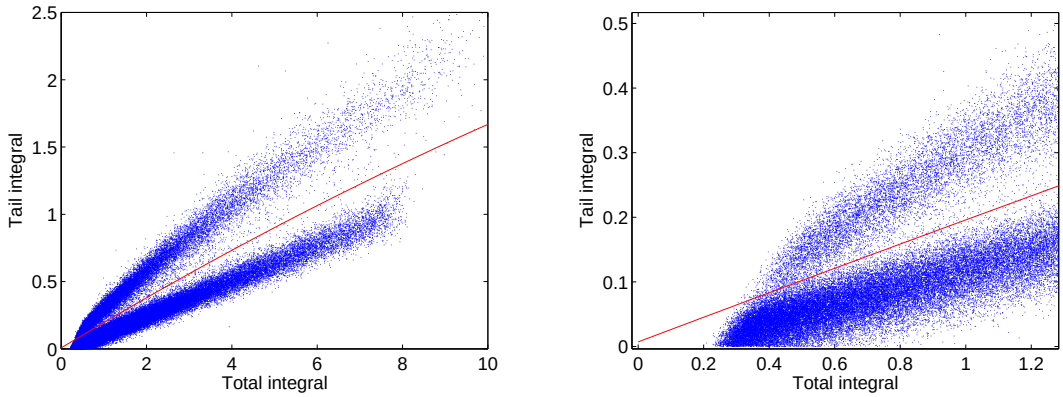


Figure 7.2: Particle pulse dependence (from [45]).

Quantitatively, PSD can be performed either by differentiation or integration [46, 47]. In the following the integration method will be detailed as implemented in some of the measurements in Sections 7.2-7.3. By integrating the total pulse and comparing it to the integral of the pulse tail a characteristic ratio is found. By observing the plot of total integral versus tail integral (Fig. 7.3) it is clear that neutron and gamma ray pulses divide into two distinct regions, where the neutrons are indicated by larger tail integrals. A discrimination curve can then be used to discriminate the neutrons from gamma rays in the analysis.



(a) Full range. (b) Zoomed in.

Figure 7.3: Tail versus total integrals for a ^{252}Cf measurement. The data shown are for a single EJ-309 detector, and the discrimination line indicates which pulses are created by neutrons (above the line) and by gamma rays (below the line).

7.2 Cross-correlations

Cross-correlations are measurements where two coincident pulses in the detectors are used to create a data point which depends on which detector each pulse was recorded in and the relative time difference. Since both neutrons and gamma rays occur it means that 4 different pairs can be detected: (n,n), (γ,γ), (n,γ), (γ,n), where the difference between (n,γ) and (γ,n) lies in which particle was detected in the "reference" detector. The analysis could be extended also into bi-correlations where three detector pulses are correlated to find a two-dimensional time difference distribution (see Ref. [20]). The cross-correlation measurements were performed using the setup displayed in Figure 7.1(a) and are presented in Paper VIII.

7.2.1 Measurements and comparisons

The measured cross-correlation curves have a unique shape for each material-geometry configuration. Most notably detecting two gamma rays, (γ,γ), will result in a peak well centred around zero time difference since both particles travel at the speed of light, and for the configurations used, it means a equal delay of around 1 ns from the source event. The neutron doublet will also have a distribution that is symmetric around zero, but it will be much wider due to the variation in arrival time depending on the neutron energy. The (n,γ) and (γ,n) peaks occur on negative and positive time differences, respectively.

A ^{252}Cf source was used to show the cross-correlations from a fissile source. Figure 7.4 shows the results with a 30 cm source–detector distance.

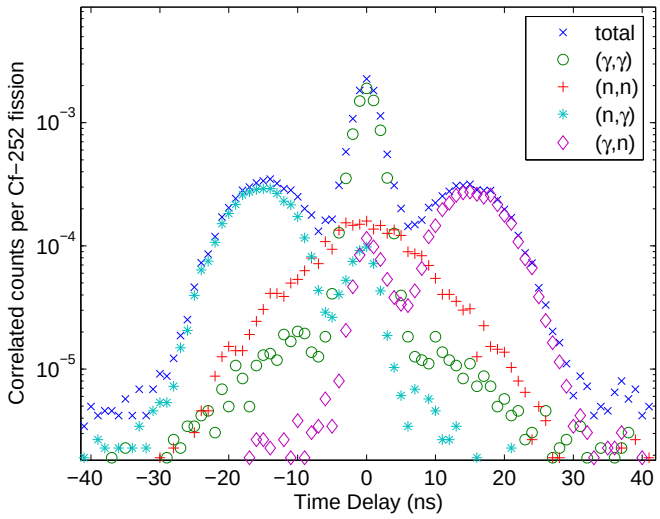


Figure 7.4: The measured cross-correlation functions for a 30 cm symmetric bare case.

Since nuclear materials could potentially be concealed by shielding, the effect of 2.2 cm Pb shielding was also investigated and is shown in Fig. 7.5. Neutrons will mostly be unaffected by the added shielding, but the gamma rays on the other hand are heavily reduced in number and that affects especially the (γ, γ) peak which has reduced in magnitude by approximately a factor 20. Also the (n,γ) and (γ,n) doublets are affected by the reduction in gamma rays that reach the detectors successfully.

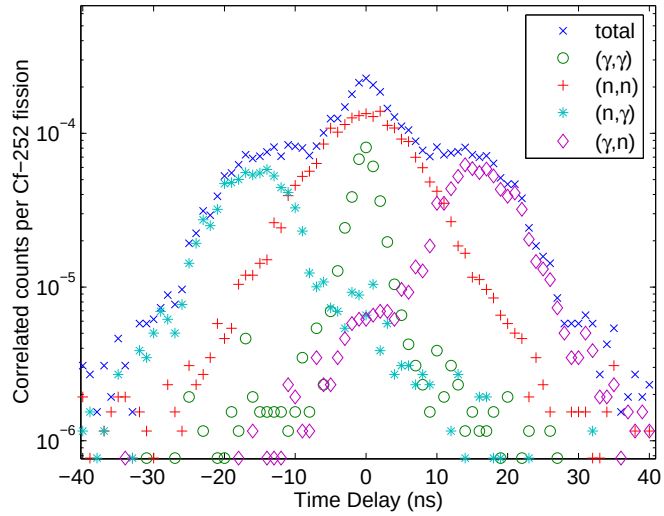


Figure 7.5: Measured cross-correlation functions for a 30 cm case with 2.2 cm lead shielding.

Comparisons were also made with simulations performed with MCNP-PoliMi. A good general agreement is found in Figure 7.6. Among the differences it is notable that the shape of the (γ, γ) peak is much narrower in the Monte Carlo simulations. A possible explanation here is that that simulation did not account for any uncertainty and delay that might exist in the electronics that acquires and stores the pulses. The digitizer in the experiment was run at 250 MHz sampling, extrapolation was used to determine pulse timings down to 1 ns time scale in the measurement. Comparing the width of the total cross-correlation is the equivalent of comparing the applied thresholds in the simulation and experiment. The lowest energy neutrons are those that generate the correlations on the highest positive and negative time delays.

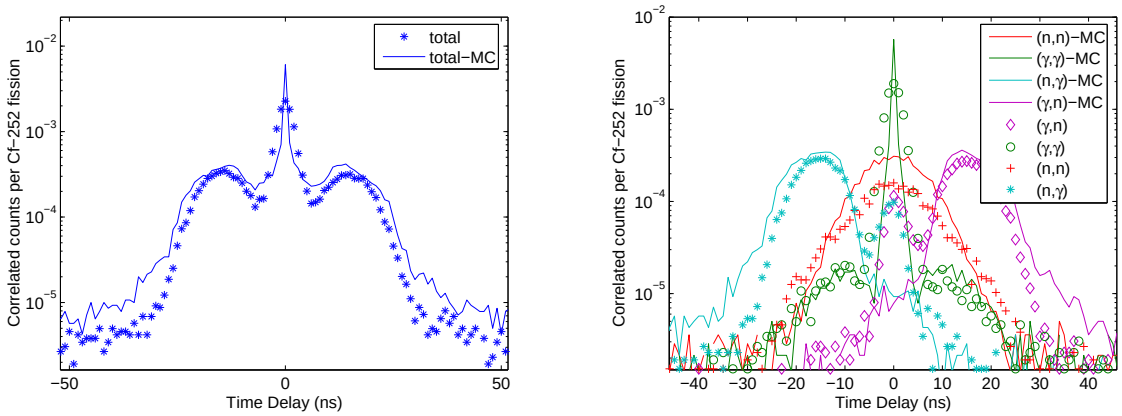


Figure 7.6: Comparisons of experimental and Monte Carlo results for the total and the partial contributions to the cross-correlation functions in the case of 30 cm source-detector distance. The most notable differences can be seen in the shape of the (γ, γ) and (n,n) peaks.

The time needed to obtain sufficient data in the measurements depends on the total detector efficiency, which rapidly increase with decreasing distance. The downside of shorter source–detector distance is that cross-talk between the detectors will increase.

7.3 A mixed multiplicity counter

Now that the theory behind mixed multiples has been mentioned in Chapter 5 and the cross-correlation measurements have shown that specific particle multiples (doubles) can be successfully measured, it is natural to attempt to extent the measurements into a full mixed multiplicity counting. The extension from the pure neutron counting is far from trivial and an initial evaluation of the capabilities of such a system is the subject of Paper IX.

7.3.1 Count rates

When doing multiplicity counting up to the third order, there are 9 different multiples whose detection rates can be measured. Measuring third order multiples naturally requires a minimum of three detectors. In the system used here (Fig. 7.1(b)), 4 detectors were used. The increased number of detectors means that the data analysis becomes all the more important to effectively determine which pulses are from correlated events. Having high PSD performance is also very important such that a neutron triplet, nnn, is not mistaken for an $nn\gamma$ triplet. Due to the high number of measurables though, correction factors could be found by comparing the ratios of neutrons and gamma rays, R_n/R_γ, to correct higher order measured rates such as R_{nnn}.

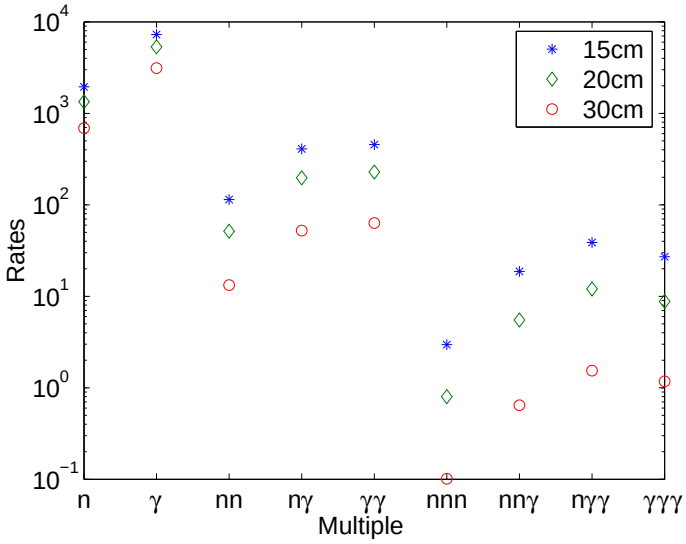


Figure 7.7: Detection rates of different multiples at different distances for the ^{252}Cf source.

Figure 7.7 shows the measured rates of all multiples up to third order, for 3 different detector distances to a ^{252}Cf source of strength 17 μCi. The higher detection rates seen for shorter distances means that decreased statistical uncertainty can be achieved, or alternatively, that the measurement times could be reduced without loss of statistical accuracy compared to larger detector distances. The measurements show that with measurement times on the order of a few minutes very good accuracy for the singles and doubles rates can be achieved. The rates and statistical errors are listed together with the measurement times in Table 7.1.

It is very encouraging to see that a relatively simple setup could be used for the type of measurement detailed here. The results show the feasibility of the method, and while improvements are possible with more sophisticated detector setups or

Count rates (s^{-1}) and statistical errors:						
	30cm	$\pm1\sigma$	20cm	$\pm1\sigma$	15cm	$\pm1\sigma$
R_n	690.4	0.63	1338.6	0.95	1947.4	1.36
R_γ	3119.9	1.34	5335.6	1.90	7256.6	2.62
R_{nn}	13.3	0.09	51.5	0.19	114.3	0.33
$R_{n\gamma}$	52.3	0.17	197.3	0.37	409.3	0.62
$R_{\gamma\gamma}$	63.4	0.19	227.7	0.39	455.0	0.66
R_{nnn}	0.1	0.01	0.8	0.02	3.0	0.05
$R_{nn\gamma}$	0.6	0.02	5.5	0.06	18.7	0.13
$R_{n\gamma\gamma}$	1.5	0.03	12.0	0.09	38.7	0.19
$R_{\gamma\gamma\gamma}$	1.2	0.03	8.8	0.08	27.0	0.16
	Measurement time (s):					
T	1735.8		1475.3		1054.6	

Table 7.1: ^{252}Cf count rates for different source–detector distances and all possible neutron, gamma ray multiples up to the third order. The statistical error is calculated as the square root of the total counts divided by the total measurement time.

altered data acquisition the initial findings are very promising for a continued evaluation of mixed multiplicity counters.

CHAPTER

8

SUMMARY AND FUTURE WORK

This last chapter will summarize the work presented in the thesis and also outlines some of the prospective future extensions which were not hitherto included into the research.

8.1 The presented work

The work has been divided into four main areas each with a dedicated chapter (4-7). Summaries of the areas are presented below:

1. Number distributions.

   The work presented in Papers I-II builds on the concept of describing a multiplying sample by master equations both for neutrons and gamma rays. The novelty lies in the derivation of probability distributions, rather than factorial moments, for the generated, emitted or detected particles.

2. Multiplicity theory.

   While neutron multiplicity, based on factorial moments was already well established, the theory was revisited and extended not only to gamma rays but also to neutrons and gamma rays in combination. The idea of using neural networks to relate measurables to sample parameters was also presented - Papers III-V.

3. Neutron–scintillator interactions.

   Scintillation light pulses generated by fast neutrons is the subject of Papers VI-VII. While individual collision contributions to light generations cannot be measured, the validation and understanding of the physics behind neutron

energy transfer and light generation gives insight into the proper utilization and modelling of organic scintillation detectors.

4. Experimental measurement systems.

   The last chapter was dedicated to measurements of cross-correlations (Paper VIII), as well as an extension to all neutron and gamma ray multiples up to third order (Paper IX). The work serve as a initial evaluation of the prospect of using measurements to acquire novel sample characteristics of nuclear materials. It was done in accordance to the ideas in the analytical models presented in the previous works.

8.2 Future extensions

While the work aimed at exploring all the interesting aspects of each topic, there are a number of unanswered questions that have arisen from external input as well as from matured insights. Some of those unexplored areas are detailed below as suggestions for future work:

- How would the probability distribution of detected particles compare to the factorial moments as an alternative route to modelling detection rates and sample parameters?

- The multiplicity theory that was derived would be very interesting to apply to measured data, and also comparing to Monte Carlo simulations, to see also how the neural network approach holds up to non-ideal measurement data.

- While the models for light pulse distributions are expressed as functions of a discrete neutron energy, it could be extended to account for energy spectrums by superimposition of functions.

- The differences in light pulse distributions regarding the detector sizes would be very interesting to investigate further and to determine whether additional spectroscopic information could be obtained by using detectors of different sizes.

- The experimental measurements could benefit greatly by being extended to additional samples such as special nuclear material: HEU, Pu.

- The idea of a mixed multiplicity counter would be interesting to develop further using simulations and improved designs with the aim of creating a versatile, usable system.

8.3 Final words

Finally, I want to conclude with a short summary of what this work has meant to me and what I have learnt from it. The area of nuclear safeguards is a very interesting one, where many different disciplines meet and combine. When it comes to the actual methods and tools needed the work can be divided into three main areas as experimental work, analytical models and numerical models (mainly through Monte Carlo simulations). Though the main focus here has been analytical methods, the models have often been compared to Monte Carlo simulations, showcasing the weaknesses and strengths of both the models and the simulation codes. The hope is that the comparisons can lead to implementation of improvements in both areas, which can then lead to better tools and measurement systems. Being able to also do experiments and actual measurements have brought an additional understanding and appreciation of the theory and models described, while also removing the idea that idealized models and simulations could create perfect results without proper validation.

Lastly the whole experience has been utmost rewarding and educational, with benefits hopefully not only to the author but perhaps also for the reader of this thesis.

REFERENCES

[1] D. Fischer, *History of the International Atomic Energy Agency, The First Forty Years*. IAEA, Vienna, 1997.

[2] "Institute of nuclear materials management: INMM Technical Divisions." `www.inmm.org`, December 2009.

[3] ISO/FDIS, Geneva, *Monitoring for inadvertent movement and illicit trafficking of radioactive material*, ISO/FDIS 22188:2004(E), 2004.

[4] J. Lubenau and J. Yusko, "Radioactive materials in recycled metals," *Health Physics*, vol. 68, no. 4, pp. 440–451, 1995.

[5] J. Lubenau and J. Yusko, "Radioactive materials in recycled metals: an update," *Health Physics*, vol. 74, no. 3, pp. 293–299, 1998.

[6] Sectretaria de energía de minas e industria paraestatal, México, *Accidente por Contaminación con Cobalto-60*, CNSNS-IT-001, 1985.

[7] W. Chang, C. Chan, and J. Wang, "Co-60 contamination in recycled steel resulting in elevated civilian radiation exposure: Causes and challenges," *Health Physics*, vol. 73, pp. 465–472, 1997.

[8] R. A. Knief, *Nuclear Engineering: Theory and Technology of Commercial Nuclear Power*. Taylor & Francis, second ed., Washington, 1992.

[9] J. Ditchburn, "Inkcinct cartoons, australia." `www.inkcinct.com.au`, December 2009.

[10] G. Knoll, *Radiation detection and Measurement Computational methods in Engineering and Science*. Wiley, third ed., New York, 2000.

[11] IAEA, Vienna, *Safeguards Techniques and Equipment 2003 Edition*, International Nuclear Verification Series No. 1 (Revised), IAEA/NVS/1/2003, 2003.

[12] R. Sher and S. Untermyer II, *The Detection of Fissionable Materials by Nondestructive Means*. American Nuclear Society, 1980.

[13] T. Harris, *The Theory of Branching Processes*. Springer Verlag / Prentice-Hall Inc., Berlin / Englewood Cliffs, 1963.

[14] W. Feller, *An Introduction to probability Theory and Its Applications*, vol. II. John Wiley & Sons, New York, 1966.

[15] K. Böhnel, "The effect of multiplication on the quantitative determination of spontaneously fissioning isotopes by neutron correlation analysis," *Nuclear Science and Engineering*, vol. 90, pp. 75–82, 1985.

[16] W. Hage and D. Cifarelli, "On the factorial moments of the neutron multiplicity distribution of fission cascades," *Nuclear Instruments and Methods A*, vol. 236, no. 1, pp. 165–177, 1985.

[17] N. Ensslin, W. Harker, M. Krick, D. Langner, M. Pickrell, and J. Stewart, *Application Guide to Neutron Multiplicity Counting*, Report LA-13422-M. Los Alamos Natonal Laboratory, Los Alamos, 1998.

[18] S. Meyn and R. Tweedie, *Markov chains and stochastic stability*. Springer Verlag, London, 1993.

[19] I. Pázsit and S. A. Pozzi, "Calculation of gamma multiplicities in a multiplying sample for the assay of nuclear materials," *Nuclear Instruments and Methods A*, vol. 555, no. 1-2, pp. 340–346, 2005.

[20] R. B. Oberer, *Fission Multiplicity Detection with temporal Gamma-Neutron Discrimination from Higher-Order Time Correlation Statistics*. PhD thesis, School of Mechanical Engineering, Georgia Institute of Technology, 2002. BWXT Y-12 Report No. Y/LB-16,144.

[21] P. L. Richards, *Manual of Mathematical Physics*. Pergamon Press, London, 1959.

[22] S. A. Pozzi, E. Padovani, and M. Marseguerra, "MCNP-PoliMi: a monte-carlo code for correlation measurements," *Nuclear Instruments and Methods A*, vol. 513, no. 3, pp. 550–558, 2003.

[23] M.-S. Lu and T. Teichmann, "A generalized model for neutron coincidence counting," *Nuclear Instruments and Methods A*, vol. 313, no. 3, pp. 471–476, 1992.

[24] D. Cifarelli and W. Hage, "Models for a three-parameter analysis of neutron signal correlation measurements for fissile material assay," *Nuclear Instruments and Methods A*, vol. 251, no. 3, pp. 550–563, 1986.

[25] R. Rojas, *Neural Networks - A systematic Introduction*. Springer Verlag, Berlin, 1996.

[26] K. Frame, W. Clay, T. Elmont, E. Esch, P. Karpius, D. MacArthur, E. McKigney, P. Santi, M. Smith, J. Thron, and R. Williams, "Development of a liquid scintillator neutron multiplicity counter (LSMC)," *Nuclear Instruments and Methods A*, vol. 579, no. 1, pp. 192–195, 2007. Proceedings of the 11th Symposium on Radiation Measurements and Applications.

[27] S. Clarke, M. Flaska, S. Pozzi, and P. Peerani, "Neutron and gamma-ray cross-correlation measurements of plutonium oxide powder," *Nuclear Instruments and Methods A*, vol. 604, no. 3, pp. 618–623, 2009.

[28] J. T. Mihalczo, J. A. Mullens, J. K. Mattingly, and T. E. Valentine, "Physical description of nuclear materials identification system (NMIS) signatures," *Nuclear Instruments and Methods A*, vol. 450, no. 2-3, pp. 531–555, 2000.

[29] I. Pázsit and L. Pál, *Neutron Fluctuations - A Treatise on the Physics of Branching Processes*. Elsevier Science Ltd., London, New York, Tokyo, 2008.

[30] H. Demuth, M. Beale, and M. Hagan, *Neural Network Toolbox 6 User's Guide*. Release 2009b, The MathWorks, Inc., 2009.

[31] P. Peerani and M. M. Ferrer, "Sensitivity analysis of physical/operational parameters in neutron multiplicity counting," *Nuclear Instruments and Methods A*, vol. 577, no. 3, pp. 682–689, 2007.

[32] P. Peerani and M. M. Ferrer, "Assessment of uncertainties in neutron multiplicity counting," *Nuclear Instruments and Methods A*, vol. 589, no. 2, pp. 304–309, 2008.

[33] H. Klein and S. Neumann, "Neutron and photon spectrometry with liquid scintillation detectors in mixed fields," *Nuclear Instruments and Methods A*, vol. 476, no. 1-2, pp. 132–142, 2002.

[34] M. Flaska and S. Pozzi, "Identification of shielded neutron sources with the liquid scintillator BC-501A using a digital pulse shape discrimination method," *Nuclear Instruments and Methods A*, vol. 577, no. 3, pp. 654–663, 2007.

[35] N. Tsoulfanidis, *Measurement and detection of radiation*. McGraw-Hill, New York, 1983.

[36] D. Hermsdorf, K. Pasieka, and D. Seeliger, "EFFI-EIN monte-carlo-programm zur berechnung der nachweiseffektivität eines neutronendetektors mit organischem szintillator," *Nuclear Instruments and Methods*, vol. 107, no. 2, pp. 259–268, 1973.

[37] A. Enqvist, I. Pázsit, and S. Pozzi, "Calculation of the pulse height distribution induced by fast neutrons in a scintillating detector," in *Joint International Topical Meeting on Mathematics & Computation and Supercomputing in Nuclear Applications (M&C + SNA 2007)*, (Monterey, CA, USA), April 15-19, 2007.

[38] I. Pázsit, A. Enqvist, and S. Pozzi, "Analytical calculation and simulation of the light pulse distribution in organic scintillation detectors," in *INMM 49th Annual Meeting*, (Nashville, TN, USA), July 13-17 2008.

[39] K. M. Case, F. de Hoffman, and G. Placzek, *Introduction to the Theory of Neutron Diffusion*, Vol. I. Los Alamos Scfientific Laboratory Report, Atomic Energy Comission, 1953.

[40] I. Carlvik, "Collision probabilities for finite cylinders and cuboids," *Nuclear Science and Engineering*, vol. 30, pp. 150–151, 1967.

[41] D. Drake, W. Feldman, and C. Hurlbut, "New electronically black neutron detectors," *Nuclear Instruments and Methods A*, vol. 247, no. 3, pp. 576–582, 1986.

[42] J. B. Czirr and G. L. Jensen, "A compact neutron coincidence spectrometer, its measured response functions and potential applications," *Nuclear Instruments and Methods A*, vol. 349, no. 2-3, pp. 532–539, 1994.

[43] J. B. Czirr, D. B. Merrill, D. Buehler, T. K. McKnight, J. L. Carroll, T. Abbott, and E. Wilcox, "Capture-gated neutron spectrometry," *Nuclear Instruments and Methods A*, vol. 476, no. 1-2, pp. 309–312, 2002.

[44] S. A. Pozzi and I. Pázsit, "Neutron slowing down in a detector with absorption," *Nuclear Science and Engineering*, vol. 154, no. 3, pp. 367–373, 2006.

[45] L. M. Bollinger and G. E. Thomas, "Measurement of the time dependence of scintillation intensity by a delayed-coincidence method," *Review of Scientific Instruments*, vol. 32, no. 9, pp. 1044–1050, 1961.

[46] J. M. Adams and G. White, "A versatile pulse shape discriminator for charged particle separation and its application to fast neutron time-of-flight spectroscopy," *Nuclear Instruments and Methods*, vol. 156, no. 3, pp. 459–476, 1978.

[47] P. Sperr, H. Spieler, M. R. Maier, and D. Evers, "A simple pulse-shape discrimination circuit," *Nuclear Instruments and Methods*, vol. 116, no. 1, pp. 55–59, 1974.

ACKNOWLEDGEMENTS

The thank yous, gratitudes, miss yous and to all those who deserve a mention...

This thesis, but much more so this whole research and PhD, is due to one very inspiring, motivating, encouraging and supportive person. A person who in seconds formulates ideas and tasks that have kept me busy for weeks, months and years at a time. My supervisor Prof. Imre Pázsit, is someone to whom I cannot express my gratitude enough. I would not be where I am today without his supervision, insight, encouragement and suggestions for improvements. In short: Thank you!

I would also like to thank Prof. Sara A. Pozzi who has offered tons of answers, inputs and help. You made me feel very welcome in both my visits to US and I owe you a Grazie Mille!

Thanks to (all) my colleagues at the Department of Nuclear Engineering who have made my days more enjoyable and helped me both with work and non-work related matters. Thanks to old and present room mates for their discussions and laughters: Kalle, Valentin, Victor, Cheuk, and a number of short term desk-occupants. Thanks to Eva for always helping with arrangements and administrative matters. Lasse and Lennart for putting up with computer issues, late e-mails and other help. Viktor for being a worthy competitor in the fight for which mouth is the hardest to shut up. You won some of the fights, but I believe I won the war. For having a boss that might not have given me a single boss-like order during my whole employment: Thank you Anders! Past and present co-workers: Alexander, Anders J, Augusto, Berit, Chiara, Christophe, Davide, Irina, Ishtvan, Jan, Joska, Katarina, Larissa, Lembit, Nils-Göran, Mattias, Monika, Paolo, Petty, Tunde and many other students, inspiring visitors and persons - Thank you.

This PhD work was financially supported by the Swedish Radiation Safety Authority (SSM; formerly The Swedish Nuclear Power Inspectorate, SKI), which is gratefully acknowledged.

Thanks to all those who made me feel welcome during my visits to US, especially Shaun and Marek (and Maria) deserves special mention and thanks. I hope to see you all again very soon.

Thanks to my long time amigo, support and gother Daniel: Tack! Markus: May the wheels never stop spinning.

Thanks to family, relatives and friends for inspiring me, and helping me whenever I've needed it. I would not be here today without you. Especially not without the supportive words and surprising insights made me smile: Tack Syrran!

To those names that did not make the list: I owe you a beer and a thanks!

(Hidden section)

Words offering wisdom, amusement and astonishment...

Why should I care about future generations? What have they ever done for me?

- Groucho Marx.

The elusive erogenous zone said to exist in some women may be a myth, say researchers who have hunted for it.

- BBC headline.

There is not the slightest indication that (nuclear) energy will ever be obtainable.

- Albert Einstein.

A farmer noticed that his chickens were sick, and called in a biologist, a chemist, and a physicist to help diagnose the problem. The biologist observed the chickens, concluding, "I can tell you there's something wrong with your chickens, but I don't know what's causing it." The chemist took fluid samples from the chickens back to his lab, and returned saying, "I can tell you what's infecting your chickens, but I don't know how they got it." Meanwhile, the physicist had been sitting on the floor, scribbling madly on several notebooks worth of paper. Suddenly, he jumped up, exclaiming, "I have the answer, but it only works for spherical chickens in a vacuum.

- The big bang theory; TV show.

I named him Blix, because he is a Weapon of Mice Destruction (WMD).

- Canadian woman explaining the name of her cat in a letter to Hans Blix.

You can't be a real country unless you have a beer and an airline - it helps if you have some kind of a football team, or some nuclear weapons, but at the very least you need a beer.

- Frank Zappa.

Well you know boys, a nuclear reactor is a lot like women. You just have to read the manual and press the right button.

- Homer Simpson (Dan Castellaneta).

US intelligence gave 200 tips regarding nuclear activities in Iran. 36 unannounced IAEA inspections following the tips found nothing!

- Hans Blix wondering why it is called American "Intelligence"?

Papers I-IX