Monte Carlo methods in spallation experiments

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Mitja Majerle

Branch: Nuclear Engineering
Specialization: Experimental Nuclear Physics

Supervisor: RNDr. Vladimír Wagner, CSc.
Affiliation: Department of Nuclear Spectroscopy
Nuclear Physics Institute
Academy of Sciences of the Czech Republic
Abstract

With new inventions in accelerator technologies, spallation process is being reconsidered as intensive source of neutrons. Apart from using spallation neutrons in basic research, some decades old idea of transmuting nuclear waste and catalyzing nuclear reactions is actual again - Accelerator Driven Systems.

Computer Monte Carlo calculations are the essential part at the design of spallation sources and experiments. The spallation process and subsequent high energy neutron transport are not studied in detail, and the computer codes are under development. This work should provide a measure of their usability and accuracy in spallation experiments.

The experimental and computational studies of two experiments with relativistic protons directed to thick targets are presented: Phasotron and Energy plus Transmutation experiment.

The first one is the experiment with 660 MeV protons directed to a bare, lead target, realized in the Laboratory of Nuclear Problems of the Joint Institute for Nuclear Research Dubna in December, 2003. Produced spallation neutrons were probed with small activation detectors at different places around the target. Monte Carlo codes MCNPX and FLUKA were used to study the systematic uncertainties and to predict the experimental results. Both codes described successfully most of the experimental results.

The Energy plus Transmutation setup consist of a lead target with the surrounding subcritical uranium blanket. The target was irradiated with relativistic protons (0.7-2 GeV) and deuterons (1.6 and 2.52 GeV). The analysis of the systematic uncertainties and the prediction of the experimental results performed with MCNPX and FLUKA codes are presented, together with comparisons with some experimental results.

Keywords:
spallation reactions, Accelerator-driven transmutation of nuclear waste, Monte Carlo simulations, MCNPX, FLUKA

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Chapter 1
Introduction

In recent years the world has registered obvious progress in the accelerator technique. The successful functioning of several high energy research accelerators (Berkeley, KEK), the construction of the LHC,... brought new technologies also to mid- and low- energy accelerators. Advances like superconductivity for magnets and RF cavities, ion sources have led to the practical realization of high-power beams.

When a high power accelerator of middle energy (few 100 MeVs) is coupled with spallation target, these systems offer a alternative method of producing neutrons. Neutrons produced in spallation have continuous energy spectrum up to the energy of the incident beam with the maximum at ca 1 MeV.

Spallation neutrons have huge potential in research, medical industry or energy production. There are several projects related to the construction of the spallation sources for research [1, 2] and the production of medicine radioisotopes [3], but, in this work the main focus is put to the high power spallation source coupled with a subcritical reactor, a so called Accelerator Driven System (ADS).

From the theoretical point of view, the description of the spallation and subsequent processes exist for many decades [4], however, extensive experimental tests are showing that our knowledge of these processes is not complete. From experiments with relativistic particles directed to thin and thick target we have learnt that our models predict the results of such experiments within the same order of magnitude, but, better accuracy is needed, especially for more complex systems, where the spallation is not the only worse known process.

Spallation experiments and Simplified ADS experiments are the topics in which Nuclear Research Institute in Řež performs experiments for some time in collaboration with the Joint Institute for Nuclear Research (JINR) in Dubna, Russia. Experimental data and their comparison with actual calculation codes are presented after the introduction, which should inform the reader about the essential ideas of the ADS topics.

1.1 Accelerator driven systems (ADS)

The long-term hazard of radioactive waste arising from nuclear energy production is a matter of continued discussion and public concern in many countries. By the use of partitioning and transmutation of the actinides and some of the long-lived fission products, the radiotoxicity
Figure 1.1: Closing the nuclear cycle with the Energy Amplifier, a sub-critical device with a Th-\(^{233}\)U fissile core fed with a supply of spallation neutrons. There is no criticality, no plutonium and no problem of actinide waste. At the end of the cycle, \(^{233}\)U and the other uranium isotopes are recycled to serve as the initial fissile part of a new load of fuel. Thorium is an abundant resource (much more than uranium) and supplies could last thousands of centuries [7].

of the high-level waste and, possibly, the safety requirements for its geologic disposal can be reduced compared with the current once-through fuel cycle. To make the technologically complex enterprise worthwhile, a reduction in the high-level waste radiotoxicity by a factor of at least one hundred is desirable. This requires very effective reactor and fuel cycle strategies, including fast reactors and/or accelerator driven, sub-critical systems. The Accelerator Driven Systems has recently been receiving increased attention due to its potential to improve the flexibility and safety characteristics of transmutation systems.

The idea of the ADS was born in 90’s with articles of C.D.Bowman and C.Rubbia, who independently proposed a new approach to the problems of radioactive waste and limited uranium resources: to introduce extra neutrons produced in spallation reaction to the core of the subcritical reactor (Fig. 1.1). They discussed this idea in two articles, C.D.Bowman in year 1992 [5] and C.Rubbia in year 1993 [6]. The main idea of their approach is to direct an intensive, relativistic proton beam to a heavy metal target, where tens of neutrons per 1 proton are produced in the spallation reaction. Spallation target is placed in a subcritical reactor core, where extra neutrons are used for sustaining the chain reaction, transmuting radioactive waste to short-lived and stable isotopes and breeding the fuel from \(^{238}\)U, \(^{232}\)Th and other isotopes. Heat from the reactor is used to produce energy, part of this energy (ca. 30%) is used to power the accelerator, and part (ca. 70%) can be sent to the electric grid - the cycle is closed. The articles are different in some technical details, Bowman suggested a thermal reactor, on the other hand, Rubbia considered fast reactor to suit better transmutation purposes, however, both demand a special accelerator, which is today the main obstacle in realizing the ADS technology. Apart from the accelerator (or possibly another intensive high energy neutron source), the detailed studies of spallation reactions and transport of neutrons of energies >20 MeV are needed.
1.1.1 Spallation

Spallation is a nuclear reaction that can take place when two nuclei collide at very high energy (typically 500 MeV per nucleon and up), in which the involved nuclei are either disintegrated into their constituents (protons and neutrons), light nuclei, and elementary particles, or a large number of nucleons are expelled from the colliding system resulting in a nucleus with a smaller atomic number (Fig. 1.2). This mechanism is clearly different from fusion reactions induced by heavy or light ions with modest kinetic energy (typically 5 MeV per nucleon) where, after the formation of a compound nucleus, only a few nucleons are evaporated. A spallation reaction can be compared to a glass that shatters in many pieces when it falls on the ground. The way how the kinetic energy is distributed over the different particles involved in a spallation reaction is otherwise well understood, but from the point of view of ADS the spallation process is not described enough accurately.

In the frame of the ADS, the spallation reaction in heavy nuclei (lead, bismuth) serves as the source of neutrons - proton with 1 GeV energy impinging to a thick target of Pb-Bi alloy produces ca. 30 neutrons in spallation.

Phases of the spallation reaction

Spallation is usually described as a two-step reaction: in the first step the target nucleus is heated, then comes the de-excitation of the target in the second step.

- The Intra Nuclear Cascade
  
  One can consider that the first step of the reaction consists in individual collisions between the nucleons. As a matter of fact, the reduced wavelength \( \frac{\lambda}{2\pi} \) of a few hundreds of MeV incoming nucleon is about \( 10^{-14} \) cm. Thus \( \frac{\lambda}{2\pi} \) of the incoming nucleon is smaller than the distance between nucleons, usually about 1 fermi \( = 10^{-13} \) cm, and the incoming nucleon "sees" the substructure of the nucleus, i.e. a bundle of nucleons.
The interaction leads to the ejection of some of the nucleons and to the excitation of the residual nucleus which will cool itself afterwards. The typical duration of the intra nuclear cascade is $10^{-22}$ sec.

- **The de-excitations modes**

  When the intra nuclear cascade is finished and the last nucleon has been ejected, the nucleus is being left in an excited state. Then the de-excitation of the residual nucleus can proceed in two main ways: evaporation and fission. The typical duration of the de-excitation process is $10^{-16}$ sec.

  The evaporation is the dedicated de-excitation mode for non fissile or hardly fissile nuclei which have been excited above the energy required for the separation of one neutron. In this case, the excited nucleus emits nucleons or light nuclei such as D, T, $^3$He, $\alpha$, Li, Be.

  Fission is the second important de-excitation channel. During the fission process, the nucleus changes its shape to reach firstly the so called saddle point at which the fission is due to occur, then a second point, the scission point, at which the nucleus is cut into two fragments with different masses.

  During de-excitation, emission of photons is also possible. The nucleus emits particles until its energy of excitation goes below the binding energy of the last nucleon. At this state, about 8 MeV are remaining. They will be evacuated out of the nucleus by gamma radiation.

  The ending of gamma emission does not mean that the de-excitation process is at the end. As a matter of fact, the resulting nucleus after gamma decay is often a radio-isotope. This radio-isotope will decay until the corresponding stable nucleus is reached.

**1.1.2 Accelerator**

Different beam performance levels are envisioned to satisfy the requirements of an XADS (experimental) facility and an ADS (industrial-scale) plant. In an XADS facility, the blanket power needs to be high enough to be representative of a full-scale ADS burner; a value between 80 MW\textsubscript{th}\textsuperscript{1} and 100 MW\textsubscript{th} is considered adequate. Nominal parameters for the accelerator driving such an XADS facility are a beam power of 5 MW to 10 MW at an energy of 600 MeV or more, so that subcritical multiplier operation over a large range of $k_{eff}$ can be evaluated [9].

For an industrial-scale ADS plant, on the other hand, the nominal fission power would be about a factor of 10 greater than in XADS, on the order of 500 MW\textsubscript{th} to 1500 MW\textsubscript{th} per burner. The ultimate beam specifications for both an XADS facility and ADS industrial systems will be dependent on the range of $k_{eff}$ desired for operation of the subcritical assemblies.

The optimum proton energy for production of neutrons by spallation in a heavy metal target, in terms of costs, target heating, and system efficiency, lies in the range 600 to 1 000 MeV.

\textsuperscript{1}MW\textsubscript{th} - thermal power
MeV. Although specific neutron production efficiency (neutrons per unit of beam power) continues to increase up to about 1.5 GeV, a minimum-cost, performance-optimized facility is generally obtained at somewhat lower energies due to other factors, such as the beam current, the accelerating gradient, and the accelerator electrical efficiency. For XADS power levels, the optimum energy in terms of minimizing the accelerator cost would be about 400 MeV, but target considerations drive the practical lowest beam energy up to 600 MeV. At lower beam energies, the power deposition density in the spallation target is too high, and the energy loss in the beam entrance window becomes significant. For the industrial ADS plant, the range 800 MeV to 1000 MeV is optimum, with lower energies matched to lower beam powers and vice versa.

Two completely different kinds of machines can be considered for acceleration of high currents of protons to an energy of 600-1000 MeV: linear accelerators (linacs) and cyclotrons. For an industrial-scale ADS system, the logical accelerator choice would be a linac. The present status of cyclotron technology extrapolates to maximum beam powers and energies for a single cyclotron to about 10 MW at 1 GeV. Linac beam theory and recent technology advances have confirmed that a linac capable of delivering up to 100 MW at 1 GeV is a relatively direct extension of existing technology. Well-supported designs for this class of linac were completed several years ago at Los Alamos and Saclay, and operation of a 100 mA (continuum wave) 6.7 MeV prototype front end for such a machine has been demonstrated [10]. Another factor favoring a linac is that the system reliability and fault minimization will lead to a design requirement that will require the accelerator operating point to be well below the maximum subsystem limits.

However, at present time, the development of suitable accelerators is in the first stage. The costly design is left for the time when all other aspects of ADS are reliably studied.

1.1.3 Subcritical reactor

A subcritical reactor is a nuclear fission reactor that produces fission without achieving criticality. Instead of a sustaining chain reaction, a subcritical reactor uses additional neutrons from an outside source. Such a reactor coupled to a particle accelerator to produce neutrons by spallation is called an ADS.

While originally thought that an ADS would be a part of a light water reactor design, other proposals have been made that incorporate an ADS into other generation IV reactor concepts. One such proposal calls for a gas cooled fast reactor that is fueled primarily by Plutonium and Americium. The neutronic properties of Americium make it difficult to use in any critical reactor due to neutronic properties that tend to make the moderator temperature coefficient more positive, decreasing stability. The inherent safety of an ADS, however, would allow Americium to be safely burned. These materials also have good neutron economy, allowing the pitch-to-diameter ratio to be large, which allows for improved natural circulation and economics.
1.2 Experimental ADS

In 90’s, mainly optimistic views about ADS existed and several ambitious projects of sub-critical systems and accelerators were planned. But, at the beginning of the 21st century it is being realized that the same technical, physical and financial problems as 20 years ago exist, which make it so far impossible to make a considerable progress in this field. In next paragraphs are mentioned the most important ADS activities.

1.2.1 European research

In European scale, the research are coordinated within special framework programmes [11]. "The Fifth Framework Programme - Euroatom” is a part of the FP5 programme, which is concerned also about the research in ADS, and within it the following activities concerning ADS research were performed:

- MEGAPIE (CERN), the project has recently fulfilled its goal and demonstrated the feasibility of safely running a liquid heavy-metal Pb-Bi target in the 1 MW proton beam [12].
- THORIUM CYCLE (Holland), CONFIRM (Sweden) projects were focused on the nuclear data for thorium-cycle reactors and for ADS construction materials.
- PDS-XADS (France) was a theoretical study focused on realization, safety, licensing and price of the construction of European XADS facility.
- ADOPT (Belgium) network was created to coordinate research activities of the whole Fifth framework programme.
- Experimental project HINDAS (Belgium) used several European accelerators in order to obtain experimental cross-section data needed for ADS experiments.
- nTOF (CERN) was another project focused on the cross-sections measurements for materials which are supposed to be used in ADS.
- MUSE experiments were performed in order to provide basic understanding of the behavior of subcritical systems driven with the outside neutron source.

The ”Sixth Framework programme - Euroatom” which followed after the closing of the previous one is focused on the research of nuclear fission and radiational protection. Its main activities are:

- EUROPART (EUROpean Research Programme for the PARTitioning of Minor Actinides)
- EUROTRANS (EUROpean Research Programme for the TRANSmutation of High Level Nuclear Waste in an Accelerator Driven System)
Research centers CIEMAT (Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas), project ESS (European Spallation Source) and research center ITEP in Moscow are also involved in the program of developing ADS and data needed for their functioning.

The experimental facilities directly connected to ADS which currently exist or are planned in Europe are the following:

- **Project IREN - Intensive Resonance Neutron Source** is being built in the JINR Dubna. It should be used as a neutron source for a large spectrum of applications, some of them concerning ADS.

- **MYRRHA (Multi-purpose hYbrid Research Reactor for High-tech Applications)** is an experimental ADS being built in Mole (Belgium). It is composed of the proton accelerator (350 MeV), liquid Pb-Bi target and a subcritical blanket in which are in a hexagonal lattice inserted 45 rods with MOX fuel (30% Pu), with $k_{eff} \approx 0.95$.

- **YALINA in Minsk** is composed of a subcritical uranium-polyethylene target-blanket to which high intensive neutron generator NG-12-1 (14 MeV, intensity $10^{10}$-$10^{12}$ n/s) provides neutrons. The system is used for the studies of subcritical systems with external source.

- **TRASCO (TRAsmutazione SCOrie)**, prepared following C. Rubbia’s suggestions, was focused on the studies of physics and technologies needed for ADS development. It consists of a linear proton accelerator (1 GeV) and Pb-Bi target, and was used for the research in ADS fields connected with accelerators, spallation and transport of neutrons.

- **TARC (Transmutation by Adiabatic Resonance Crossing)**, which was running in years 1996-1999, demonstrated the efficiency of the ”Adiabatic Resonance Crossing” method in the liquidation of Long Lived Fission Fragments in the ADS. It was sited at the CERN PS accelerator and precise measurements of distributions of spallation neutrons were performed in the lead cube with 3m side.

- **TRADE (TRiga Accelerator Driven Experiment)** came with an interesting idea - to couple existing, well studied, low-power reactor with the spallation target and accelerator. The idea is supposed to be realized on TRIGA reactor in ENEA Casaccia Centre (Italy) in years 2007-2008. The core of the reactor is supposed to have $k_{eff}$ in the range between 0.9-0.99 and a constant proton beam (few hundred $\mu$A) will be provided by accelerator to Pb target.

Intensive research in the field is performed in the Joint Institute for Nuclear Research in Dubna, Russia, to which is dedicated the section 1.2.3.

### 1.2.2 Research outside Europe

In the USA the program Advanced Accelerator Applications (AAA) was funded for the research of nuclear problems connected to energetics and waste, which is supposed to manage
different projects connected to accelerator technologies as for example: Accelerator Produc-
tion of Tritium (APT) and Accelerator Transmutation of Waste (ATW). The program has
3 main aims:

- The transformation of the APT project to the Accelerator Demonstration Facility
  (ADF).
- The construction of the ADF.
- Testing and study of technologies connected to transmutation systems.

The project Spallation Neutron Source (SNS) was funded in Oak Ridge, Tennessee, USA
with the collaboration of six USA laboratories: Argonne, Lawrence Berkeley, Brookhaven,
Jefferson, Los Alamos a Oak Ridge [1]. In May, 2006, it produced first neutrons after 7
years of construction. It is supposed to provide the world’s most intensive neutron beams
for scientific research and industrial use.

An thermal neutron accelerator-driven transmutation system - Tier - has been proposed
in USA by Bowman [13]. Tier 1 (k_{eff} = 0.96, neutron flux 2 \cdot 10^{14} \text{n cm}^{-2} \text{s}^{-1}) is a graphite
assembly with circulating molten salt (NaF-ZrF_4) fuel and liquid Lead spallation target.
This ATW system with 750 MWth power generated by fission of 300kg/y of Pu and minor
actinides corresponding to annual pressure water reactor production of these elements would
be a once-through transmuter with 80% efficiency. Tier 2 (k_{eff}=0.95, neutron flux 4 \cdot 10^{14} \text{n}
\text{cm}^{-2} \text{s}^{-1}) system would be then a back-end option transmuting the spent fuel from 4 Tier 1
units.

Accelerator-Driven Transmutation research in Japan is by any standards best integrated
into the broad programme of fundamental and applied nuclear physics. From October
1988 research on partitioning and transmutation in Japan has been conducted in the frame
OMEGA programme^1. In more details Accelerator-Driven Transmutation research is and
will be focused on the development of high power accelerators: a superconducting high inten-
sity proton accelerator with energy 1 - 1.5 GeV, current several 10s mA is under development.
The accelerator is expected to be supplemented by an experimental transmutation facility
and taken into operation after the year 2008.

In South Korea, Since 1992, a long-term research project has been in progress at KAERI
(Korean Atomic Energy Research Institute) with the aim of developing a method for reducing
the radiotoxicity of high-level waste [15]. This programme comprises the evaluation of data,
the study of the possibility of transmuting heavy actinides in PWRs, the development of
codes for the calculation of transmutation rates and the design of transmutation systems.
Conventional reactors, fast reactors and hybrid systems consisting of a subcritical reactor
and an accelerator are being studied. In 1997, the programme was reviewed and a decision
was made that research on accelerator-driven systems will be one of KAERIs main areas of
work up to the year 2007.

^1OMEGA stands for ”Options for Making Extra Gain of Actinides and fission products generated in the
nuclear fuel cycle”) [14]
1.2.3 Russian research

Since 1994, several Russian research institutes have been involved in research on accelerator-driven systems as a possible means of eliminating weapons-grade Plutonium and nuclear waste. The studies have comprised the realization of a linear proton accelerator to drive a specially designed subcritical transmutation core as well as reprocessing processes which can be applied in an integrated transmutation facility. The work has largely been financed by International science & technology center Russia. The transmutation facility which was studied comprised a target of liquid Lead-Bismuth as well as a subcritical core with two zones. Important results which have emerged include the recommendation of using lighter material such as titanium and graphite as window material for the proton beam as well as the conclusion that problems with the accumulation of $^{210}\text{Po}$ in Lead-Bismuth-cooled systems are less important than previously believed.

There are also several other projects related to ADS which are currently financed through ISTC. Part of these projects are focused on cross-section measurements and evaluations for different reactions on neutrons and protons in energy ranges from GeV down to meV. Also basic studies important for the solid spallation target construction are underway in Russia.

In the Joint Institute for Nuclear Research, a long term tradition of cross-section measurement on the "Phasotron" cyclotron exists. This program was quickly adopted for the measurements of the cross-sections for ADS purposes and later extended on studies of neutron production on thick targets in energy ranges 0.5-2 GeV. The experiments performed so far are:

**Cross-section measurements**

The measurements of the cross-section of 660 MeV protons with different fission products ($^{129}\text{I}$), natural uranium and higher actinides ($^{237}\text{Np}$, $^{241}\text{Am}$, $^{239}\text{Pu}$) were realized with specially prepared samples irradiated by the protons extracted from the Phasotron accelerator. After the irradiation the yields of the produced isotopes were determined by the means of the gamma spectrometry. The results contributed data to EXFOR cross-section database and are also used to check the theoretical models for cross-section predictions [16].

**GAMMA-2**

GAMMA-2 was the experiment focused to the studies of production of neutrons in spallation process and their moderation and transport in the neutron moderator. Gamma-2 consisted of a thick, lead target (2$r$=8cm, $l$=20cm) surrounded by a paraffin moderator of 6 cm thickness to slow down fast spallation neutrons to resonance energies. Slow neutrons were detected through $(n,\gamma)$ reaction by radiochemical detectors placed on top of the polyethylene along the whole setup. Gamma-2 was a simple setup providing results which are very useful for comparison with computer codes predictions [17].

**Phasotron experiment**

While GAMMA-2 was focused on the overall neutron production, the Phasotron experiment was mostly concerned about the spatial distribution of high energy neutrons ($E > 10 \text{ MeV}$)
produced in spallation process and the transmutation of radioactive iodine $^{129}$I in such neutron spectrum. The intensive beam of 660 MeV protons from the Phasotron accelerator was directed to a bare, lead target (2r=9.6cm, l=45.2cm). Small radiochemical detectors (metal foils $2 \times 2 \times 0.1$ cm$^3$) were placed on top of the target together with the iodine transmutation samples. The gamma-spectra measurements were performed quickly enough after the irradiation to study the transmutation of iodine to short-lived isotopes. The results from this geometrically simple setup are again very useful for computer codes tests as will be shown in the next chapters.

**Energy Plus Transmutation**

The "Energy plus Transmutation" (EPT)setup consists of a thick, lead target (2r=8.4 cm, l=48cm) surrounded with a uranium blanket (206.4 kg) and placed in a polyethylene box. In series of experiments, relativistic protons and deuterons of energies from 0.7 to 2.52 GeV were directed to the target. Produced neutron flux and its transmutation capabilities were studied at different places of the setup with activation, solid state nuclear track, $^3$He and other detectors. Closer to the real reactor core geometry, the EPT setup consist of several parts, which all have influence on the neutron field, and is therefore not appropriate for direct tests of accuracy of computer codes. However, in many studies, the possible sources of systematic uncertainties of obtained experimental data were analyzed using computer codes simulations to justify the use the experimental data for comparison with the results of the MCNPX simulation code [19].

**Subcritical Assembly at Dubna**

The Subcritical Assembly at Dubna (SAD) is planned project that should consist of a replaceable spallation target (Pb, W) with a subcritical MOX blanket ($\text{UO}_2+\text{PuO}_2$). The studies of neutron production, power release, fission rates of higher actinides and transmutation rates of fission products are some of the motivations for this complicated setup [20].

**1.3 Calculations**

The development of a transmutation program using ADS requires accurate spallation simulation tools in order to design such systems. The simulation tools developed for nuclear reactors cannot be applied immediately to externally driven sub-critical systems. Indeed, the spatial distribution of the neutron flux is expected to be radically different in the two cases. While in a critical reactor the flux distribution inside the volume is determined essentially by the boundary conditions, in an Energy Amplifier the effect of the initial high-energy cascade is dominant. In fact, in a sub-critical arrangement the neutron flux along any radial direction starting from the center must fall-off in an approximately exponential manner. The corrections to the exponential behavior depend primarily on the shape of the source and they are important when close to it.

Such a complex simulation has to be validated. The validation was one of the main goals of many of mentioned projects (section 1.2), and detailed comparisons of the neutron mul-
tiplication factor, of energy and space distributions of the neutron fluence were performed. From most point of views, good agreement with simulation was obtained. These sets of experiments confirm in particular that the spallation process is correctly predicted and validates the reliability of the predictions of the integral neutronic parameters of experimental ADS facilities.

Computer programs used for neutron multiplying systems fall into two broad categories: (a) deterministic and (b) Monte Carlo codes.

- **Deterministic codes** are based on the solution of the neutron transport equations. To make the problem amenable to a computer solution, a discretisation is introduced both in space and in energy. These codes operate on a spatial grid and on a fixed number of energy "groups". While this approach has shown its viability in many applications, and is widely used to simulate critical reactors, it suffers from some drawbacks that become important in the case of a sub-critical device coupled to a particle accelerator. Most probably, the required complete analytical model would not provide a solution in a time shorter than with a well implemented Monte Carlo. In summary, deterministic model codes are well adapted to the simulation of relatively well known critical systems, but they cannot be easily used in their present form to explore the new domain of sub-critical accelerator driven systems. They are usually useful after tuning as they tend to represent a parametrization of the system rather than a true simulation.

- **Monte Carlo codes.** The second major type of approach to the simulation of nuclear fission systems is the Monte Carlo method. When point-wise cross-sections are used, the Monte Carlo is free from almost all the drawbacks of deterministic codes, but its precision varies inversely with the square root of the number of events processed. This represents a potentially large problem of CPU time, particularly when the simulation must span the entire lifetime of a power producing system.

  Fully analogous Monte Carlo simulations with point-wise cross-sections however provide a host of information not easily available to deterministic codes: "infinite" spatial resolution; full treatment of resonances (correct account of selfshielding effects) and "on line" full 3-D calculation of activation and spectrum-dependent transmutation effects.

  The main limitations of the Monte Carlo method are:

  - The correctness of the neutron cross-sections, but this is common to all transport codes.
  - The physical model used, but for low energy neutron transport this is mainly expressed by the partial reaction cross-sections, double differential cross-sections, etc.
  - Its intrinsic imprecision, due to the random nature of the events generated, though it may be reduced by increasing the number of trial events N, now possible with the help of fast parallel computers which can generate many events simultaneously.

A number of Monte Carlo and deterministic codes are available for the purpose of ADS simulations and some details on their functioning are given below.
1.3.1 Simulation of spallation reaction

Most existing codes used for high energy ion-nucleus reactions are based on the intranuclear cascade (INC) model for the first stage of the reaction, the final steps being described by an evaporation (EVAP) model [21]. The philosophies of the INC and EVAP models are very different: The INC calculations follow the history of individual nucleons in a classical or semi-classical manner, while the EVAP calculations follow the deexcitation of the whole nucleus while it decays from one nuclear level to a lower one. The connection between the two approaches is one of the delicate points of high (or intermediate) energy simulations of ion-nucleus reactions. In principle the single particle approach of INC should be justified as long as the wavelength of the incident nucleon is smaller than the nucleon radius ($\lambda \leq r_{\text{nucleus}}$ or $E > 160$ MeV). On the other hand, the evaporation approach is valid as long as the energy of the nucleon does not exceed too much the nuclear potential depth ($\approx 40$ MeV [22, 23]). Thus, the transition energy between the INC and EVAP calculations cannot be specified rigorously. For that matter several codes have added an intermediate step whose domain of validity is expected to overlap on the INC and EVAP domains. This step is the preequilibrium phase.

• **During the Intra-Nuclear Cascade (INC)** - $E \geq \approx 160$ MeV - the incident particle collides with one or several nucleons of the target nucleus. The struck nucleons, in turn, collide with the unperturbed nucleons. A cascade develops. The INC calculation for a specific nucleon stops whenever its energy falls below a specified value, related to the depth of the nuclear potential well ($\approx 40$ MeV).

• **Preequilibrium phase**: The INC model lacks justification for nucleon energies (inside the nucleus) below around 160 MeV. Preequilibrium models have, since long, been used in nuclear physics in this energy domain. These models follow a population of quasi-particle excitations of the nuclear Fermi gas by means of a master equation. Quasi-particle states are characterized by their particle escape and damping widths. Angular distributions are associated to the escaping particles. In a sense, preequilibrium models allow an easier phenomenological adjustment of angular distributions than does the intra nuclear cascade. There are many versions of preequilibrium models, but, unhappily, no clear criteria to choose among them, except their ability to reproduce experimental data.

• **Evaporation phase** - $E \leq \approx 40$ MeV: The compound nucleus is formed and the energy is uniformly distributed throughout it. The nucleus is in a highly excited state and loses its energy by evaporating neutrons, by fission or $\gamma$ emission. The most important ingredients of the calculations of this phase are the level densities. It is important to account for the influence of shell effects on the level density parameters and of their washing out with nuclear temperature.

**Modelling of Intra Nuclear Cascade**

The INC model, first proposed by Serber [24], is used to describe the interaction between high energy hadrons (pions, protons, anti-protons...) or light nuclei with a target nucleus. The nucleus is considered under a statistical point of view. When the nucleus is at rest,
it is regarded as a degenerated Fermi gas at zero temperature. All the particles which are scattered or produced during the cascade are treated in the field of the classical mechanics, they are defined by their velocity and their position. Every scattering which would lead to an already occupied energy level is forbidden because the nucleons are fermions. As a matter of fact, only one fermion can be in a given state according to the Pauli exclusion principle.

There are two main approaches to describe the intra nuclear cascades (see fig. 1.3). In the Bertini approach [4, 25], the incoming particle hits the target material (target nuclei) which is regarded as a continuous medium. The particles have a specific mean free path $\lambda = (\rho \sigma)^{-1}$ in this medium (i.e. inside a target nucleus). After each path, the particle scatters on a nucleon with which it exchanges energy. In the Cugnon approach [26, 27], the incoming particle is propagating freely in the target material (i.e. inside a target nucleus) until it is at its minimum distance of approach from a nucleon $d_{\text{min}}$. The particle is scattered if $d_{\text{min}} \leq \sqrt{\frac{\sigma_{\text{tot}}}{\pi}}$.

Modelling of de-excitation

In the de-excitation phase three processes compete: evaporation, fission, $\gamma$-emission. The last one is of negligible influence, evaporation and fission are in most cases equally probable.

There are several models of neutron evaporation which are all based on calculations of highly excited states of nucleus and de-excitations to ground state. Most often used are Dresner [29] and ABLA [30], which is more sophisticated as it takes into account several corrections left out by Dresner model (nuclear collective states, ...).

Two models of fission are available for describing high-energy fission, the ORNL model (from Oak Ridge National Laboratory) [31] and the RAL model (from Rutherford Appleton Laboratory) [32]. The ORNL model simulates only fission for actinides with $Z > 90$, while the RAL model allows fission from $Z > 71$. The ABLA fission-evaporation model uses its own fission model.

The $\gamma$-emission is not very important when other de-excitation channels are open.
1.3.2 High energy cross-section libraries

While the spallation models are believed to be quite reliable above $\approx 150$ MeV and, on the other hand, the behaviour of neutrons with energies $<20$ MeV in reactor systems is also well studied, there exist an energy gap from 20 up to 150 MeV, where we do not know how to successfully model the reactions of neutrons in different materials.

Suites of evaluated reaction cross-section files (LA150 library [33], NRG-2003 library [34]) have been developed in support of ADS design. They cover the energies from 20 MeV up to 150 MeV (200 MeV for NRG) for neutrons and from 1-150 MeV (200 MeV for NRG) for protons. Evaluations are completed for isotopes of the structural, shielding, and target-blanket materials.

The primary motivation for using these evaluated data is the accuracy improvements that one can expect to obtain in the below 200 MeV energy region. In most previous transport simulations, intranuclear-cascade methods have been used for neutrons above 20 MeV and for protons at all energies, even though the semiclassical assumptions do not hold at lower energies. By developing evaluated cross-section libraries, one can expect to have the most accurate possible representation of the nuclear cross-sections.

The nuclear models used for LA150 cross-sections are based on the theoretical approaches that are appropriate for the energies in the few-MeV to 150 MeV range: the Hauaer-Feahbach compound nucleus theory; preequilibrium calculations based on the Feshbach-Kerman-Koonin theory or the exciton model; direct reactions calculated from the optical model using collective excitation form factors; and elastic scattering from the optical model. The GNASH code was demonstrated to be one of the most accurate codes available for model calculations below 150 MeV in a Nuclear Energy Agency code intercomparison [35]. The optical model is used for predictions of the total, reaction, and elastic scattering cross-sections, making use of nucleon potentials at higher energies. It is particularly useful for accurately representing the angular distributions in elastic scattering, allowing more accurate neutron transport simulations.

1.3.3 MCNPX

The MCNPX code [36] is a coupling of two previous calculations codes: LAHET [37] and MCNP [38]. MCNPX only needs one input file for both codes and avoids the transfer of large data files. It allows the treatment of transport problems in a large range of energies, from thermal energy (25 meV) to a few GeV.

For energies lower than 20 MeV, quite complete sets of cross-sections are available for the major part of the stable nuclei. International cross-sections libraries such as ENDF [39], JEFF [40], JENDL [41], are available and are regularly updated. To treat the transport, MCNP uses data deduced of these libraries after processing them with NJOY/ACER [42].

For energies larger than 20 MeV, there are less cross-section data. Presently the LA150 and NRG-2003 libraries, which cover around 50 isotope (most common in ADS) up to 150-200 MeV, are included with the MCNPX code package and the preparation of complete data files up to 150 MeV is in progress in several projects.

After running an MCNPX-job, several evaluations can be performed with an auxilliary
code, HTAPE3X, to obtain specific informations (neutron spectrum, energy deposition, residual nuclei...)

Most options from older LAHET are used in MCNPX. They include the following models:

- the BERTINI [25] and the ISABEL [44] INC models as in LAHET, and a third INC model: the CEM model [43],
- the INCL4 [45] model based on Cugnon INC approach (see 1.3.1),
- a multistage pre-equilibrium model,
- DRESNER [29] and ABLA [30] evaporation model,
- RAL and ORNL fission models (see 1.3.1),
- a nucleon elastic scattering model,
- a gamma production model.

MCNPX code package is the main simulation tool used in our work. The spallation reactions and subsequent neutron transport is simulated with several models. As a result, the neutron, proton and spectra of other particles at the places of the activation detectors used in the experiment are calculated. Those are in the next step convoluted with appropriate cross-sections.

1.3.4 FLUKA

FLUKA [46, 47] is a general purpose tool for calculations of particle transport and interactions with matter, covering an extended range of applications spanning from proton and electron accelerator shielding to target design, calorimetry, activation, dosimetry, detector design, ADS, cosmic rays, neutrino physics, radiotherapy etc.

The highest priority in the design and development of Fluka has always been the implementation and improvement of sound and modern physical models. Microscopic models are adopted whenever possible, consistency among all the reaction steps and/or reaction types is ensured, conservation laws are enforced at each step, results are checked against experimental data at single interaction level. As a result, final predictions are obtained with a minimal set of free parameters fixed for all energy/target/projectile combinations. Therefore results in complex cases, as well as properties and scaling laws, arise naturally from the underlying physical models, predictivity is provided where no experimental data are directly available, and correlations within interactions and among shower components are preserved.

Fluka can simulate with high accuracy the interaction and propagation in matter of about 60 different particles, including photons and electrons from 1 keV to thousands of TeV, neutrinos, muons of any energy, hadrons of energies up to 20 TeV (up to 10 PeV by linking Fluka with the Dpmjet code) and all the corresponding antiparticles, neutrons down to thermal energies and heavy ions. The program can also transport polarised photons
(e.g., synchrotron radiation) and optical photons. Time evolution and tracking of emitted radiation from unstable residual nuclei can be performed on line.

The PEANUT (PreEquilibrium Approach to NUclear Thermalization) [59] is used for the simulation of hadron-nuclear interactions from GeV region down to 20 MeV, through more steps (Generalized IntraNuclear Cascade, Preequilibrium stage, FLUKA evaporation model). The cross-section libraries used in FLUKA are imported from ENDF/B-VI [39].

1.3.5 TALYS

TALYS [48] is a software package for the simulation of nuclear reactions that is used to calculate total and partial cross-sections, for the detector materials used in our experiments. TALYS is a deterministic code, it implements various physical models and can quite reliably reproduce various cross-sections in the energy range 1keV-250 MeV.

In this work, partial cross-sections for reaction channels \((n,\alpha)\), \((n,xn)\), \((n,fis)\) for detector materials (Au, Al, Bi, I...) are calculated with TALYS from 1-150 MeV. From 150 MeV, the cross-sections calculated with MCNPX code package and CEM03 nuclear model normalized to TALYS values at 150 MeV were used.

1.4 Motivation for Dubna ADS experiments and this work

The main focuses of the existing studies on the neutron production on lead targets [49, 50] are either the overall neutron production in thick targets or the angular distribution of spallation neutrons in thin targets. The current Dubna ADS program combines both - thick targets in which the spatial distribution of neutrons is studied. This is the intermediate level between the pure spallation studies and the studies of spallation together with the transport and moderation of neutrons. These experiments present a step towards the real ADS devices, and enable the studies of materials, detectors, and computer models under ADS working conditions.

This work’s main interest are two experimental setups: a bare, lead target which was irradiated with 660 MeV protons from the Phasotron accelerator in November, 2003; and a complex setup Energy Plus Transmutation, which was from the year 2003 several times irradiated with protons (0.7-2 GeV) and deuterons (1.6 and 2.52 GeV). The geometrical properties of the experimental setups were determined by their similarity to the real ADS devices and neutron detectors adapted to such conditions were used. Around \(10^{13}\) incident particles were directed to the targets during few hours and activation detectors with masses around 1 g were activated enough to be measured with the HPGe detectors after the experiment. The experimental results carry information about the total number and about the spatial and energetic distribution of produced neutrons.

Both experimental setups were simulated by Monte Carlo codes MCNPX and FLUKA. The codes had the key role at the estimation of the systematic uncertainties of the experiments. The experimental parameters were measured with finite accuracy, and by changing the simulation parameters within these limits, the impact of such inaccuracies on the results
was studied. It was found out that the systematical inaccuracies of the experimental setups are in orders of tens of percents.

The calculated results were compared to experimental values. In studies mentioned above [49, 50], the simulations quite reliably reproduced the overall neutron production (with 10% accuracy) or angular distribution of neutrons. It is expected that the codes would describe more complicated setups less accurately than simple experiments - the measured quantities are the averages from multiple processes (spallations, transport) and the inaccuracies of the description of single processes are summed. However, for both described experiments the disagreements were in general only slightly bigger than the systematical inaccuracies, within 50%. Also the comparisons between different models of spallation reaction was performed. In most cases, all models predicted similar results, within the limits of inaccuracy of the experiments it was impossible to qualify different models.
Part I

SPALLATION NEUTRONS ON BARE TARGET
Chapter 2

The Phasotron experiment

2.1 The experimental setup and results

2.1.1 Experimental setup

The setup consisted of a cylindrical lead target with the radius 4.8 cm and length 45.2 cm, placed at the end of the concrete corridor with length of 20 m, height and width 2 m and 2 m thick walls (Figure 2.1). The target was separated in two cylindrical parts (with lengths 12.3 cm and 32.9 cm) and 0.7 cm air gap between them. Both parts were made of smaller segments (in cm: 4.7, 3.8, 3.8, gap 0.7, 3.3, 4.6, 4.3, 4.2, 3.9, 4.8, 3.8, 4).

Activation detectors, made of Al, Au, and Bi thin foils (dimensions 2 cm × 2 cm × 0.05 mm for Al and Au foils and 2.5 cm × 2.5 cm × 1 mm for Bi foils), were placed on top of the setup along its whole length. Au and Al detectors were placed every 2 cm from the beginning of the target and Bi detectors were placed on the 1st, 9th, 21st, 31st, and 43rd cm. Five sets of Al and Au activation detectors were placed in the gap, one detector set on the target central axis and four sets around it, forming a cross with ca. 3.5 mm space between the foils, as seen in Figure 2.2a. In front of the target were placed detectors for the measurement of the beam integral made of bigger Al and Cu foils (8 cm × 8 cm × 0.1 mm).

High energy neutrons produced during the irradiation were at the same time used for the studies of transmutation of radioactive isotope iodine $^{129}$I. Four iodine samples were placed on top of the setup, two samples at 9th cm and other two samples at 21st cm. Each pair of the samples contained a sample with natural iodine ($^{127}$I) and another with iodine from the nuclear waste (mixture of 17% $^{127}$I and 83% $^{129}$I) in the form of NaI. $^{127}$I samples were in the form of solid cylindrical tablets ($r = 1.05$ cm, $h = 0.3$ cm), and $^{129}$I samples were prepared from NaI powder packed in Al shielding [53].

After 10 minutes of irradiation with the proton beam of approximate intensity ($10^{13}$ protons/s), the detectors and samples were collected from the setup. Their gamma spectra were measured with HPGe spectrometers. The detectors were measured twice, soon after

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1Shortly before the experiment, these foils were manipulated and it is possible that they were not centered after that.
2The aluminum shielding was remodeled for this experiment to provide reasonable safety at the minimum of Al used.
the irradiation for a short time, and after the decay of short living isotopes for a longer time. The iodine samples were measured \( \approx 10 \) times. Gamma spectra of radioisotopes with decay times from few minutes up to some days were registered. The spectra were analyzed using the standard method (described in [54]) and the amount of activated/transmuted material in the detectors was determined. The quantity introduced as the production rate \( B(A) \) - the mass of activated isotope A per one incident proton and per 1 g of detector material - is used to present the results.

### 2.1.2 Experimental data - beam parameters

During the irradiation, the beam was monitored with the wire chamber placed at the end of the beam tube. The wire chamber showed that after ten minutes of constant irradiation, the beam centered to the central target axis, with the intensity of ca. \( 10^{13} \) protons/s and with horizontal and vertical diameters of 1.6 cm - 1.9 cm was produced.

Independently, the beam integral was measured with the activation detectors, big Al and Cu foils, placed in front of the target. Their analysis showed that isotopes \(^{24}\text{Na}\) (not used for the determination of the beam integral, part of it is produced by neutrons) and \(^{7}\text{Be}\) are found in Al foils and \(^{7}\text{Be}, \(^{24}\text{Na}, \(^{42}\text{K}, \(^{43}\text{K}, \(^{44}\text{mSc}, \(^{46}\text{Sc}, \(^{47}\text{Sc}, \(^{48}\text{V}, \(^{51}\text{Cr}, \(^{52}\text{Fe}, \(^{52}\text{Mn}, \(^{54}\text{Mn}, \(^{55}\text{Co}, \(^{56}\text{Co}, \(^{57}\text{Co}, \(^{58}\text{Co}, \(^{57}\text{Ni} \) in natCu foil. From the production rates and the cross sections for this isotopes (taken from EXFOR [55] and extrapolated to 660 MeV if necessary) the number of protons was calculated. Mean weighted average value of the integral proton flux was determined to be \( 1.58 \cdot 10^{15} \) protons with the accuracy of 6%.

The beam diameter and displacement from the central axis were measured with the cross of five sets of Al and Au detector sets placed in the gap between the target sections. The production rates are shown in the Figure 2.2b. Comparing the rates in different foils, one can conclude that the beam had the elliptical shape (the ratio between the horizontal and vertical axis was 0.6:1) and that the center of the beam was somewhere between the central
and the top foil. Shortly before the experiment the cross with the detectors was manipulated and it is possible that it was displaced from the target center.

2.1.3 Experimental data - longitudinal neutron field

In the detectors used for the measurement of longitudinal distribution of high energy neutrons were found the following isotopes: in Al detectors $^{24}\text{Na}$, in Au detectors $^{189}\text{Au}$-$^{196}\text{Au}$, $^{198}\text{Au}$ and in Bi detectors $^{201}\text{Bi}$-$^{206}\text{Bi}$. The production rates against the position along the target are plotted in Figures 2.3a, 2.3b, and 2.3c for all three types of detector foils. The error bars are only statistical uncertainties of the gamma peak approximation with Gaussian curve.

The graphs show the specific shape: the maximum at around the 8th cm, and the point near the 30th cm, where the neutron field starts to decrease faster. The second point coincides with the range of 660 MeV protons in lead - protons with such energy are stopped due to ionization losses after 31 cm of lead material according to calculation (Fig. 2.3d) [56]. After ca. 30 cm of the lead, there is no more spallation by primary particles, what is seen as a fast decrease of production rates after this point. The graph for $^{198}\text{Au}$, which is produced through $(n,\gamma)$ reaction channel by low energy neutrons shows constant production along the target. The neutrons from the target were moderated and partly reflected back by concrete walls, resulting in a almost homogenous low energy neutron field around the target, which is seen as a flat distribution of production rates of $^{198}\text{Au}$.

2.1.4 Experimental data - transmutation of iodine

The main interest in the experiment was to measure the production rates of higher order reactions in iodine - $(n,5n)$, $(n,6n)$,... reactions. Actually, the yields of produced isotopes up to $^{118}\text{I}$ - $(n,10n)$ - were determined with the accuracy of 10%, and the products decayed from iodine isotopes up to $^{110}\text{I}$ - $(n,12n)$ - were detected. The yields of produced isotopes for $^{129}\text{I}$
Figure 2.3: B-values for $^{24}$Na in Al foils (a) and different isotopes in Au (b) and Bi (c) foils along the target (errors at the graphs are only statistical errors). In (d) is shown the proton range in lead in dependency of the energy [56].
Figure 2.4: B-values for different isotopes in $^{127}$I and $^{129}$I. Samples were placed at the 9th (a) and 21st cm (b).

were calculated with the substraction of $^{127}$I contribution in the samples with the mixture of radioactive and stable iodine.

The graphs in Fig. 2.4 show the production rates of measured iodine isotopes at the 9th cm and the 21st cm for $^{127}$I and $^{129}$I. The production rates lie in the range between $10^{-8}$ g$^{-1}$proton$^{-1}$ and $10^{-5}$ g$^{-1}$proton$^{-1}$.

2.2 Simulations - systematic uncertainties of experimental results

The experimental setup was simulated using the MCNPX v2.6.0f code package [57] and the FLUKA [46, 47] code. In the simulations, the target was approximated with the lead cylinder with its real dimensions and the concrete with 2 m thick walls. The beam tube, the beam stopper and the table on which was placed the setup were approximated with an evacuated iron tube ($r = 10$ cm, $d = 0.5$ cm, ends 30 cm before the target), full iron tube ($r = 10$ cm, starts 55 cm after the target) and iron plate (1 cm thick, 1 m $\times$ 0.5 m), respectively. The detectors were approximated with thin foils with realistic dimensions and the samples were approximated with realistic thin cylinders (enveloped in aluminum shielding - $^{129}$I samples).

In the simulations with the MCNPX code package two cascade models (CEM03 [58], INCL4/ABLA [45, 30]) and LA150 [33] libraries were used. In FLUKA, the preequilibrium-cascade model PEANUT [59] and FLUKA’s own cross section libraries (for materials used at this experiments imported from ENDF/B-VI) were used.

Neutron and proton fluences in the detectors and iodine samples binned in 1 MeV energy intervals (1-150 MeV, 50 MeV bins above 150 MeV) were calculated. These fluences were convoluted with the (n,xn) cross sections calculated with the TALYS-1.0 [48] code (and MCNPX code using CEM03 model for energies higher than 150 MeV) in order to obtain the production rates $B(A)$. 

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2.2.1 The influence of the setup parts and experimental uncertainties to the results

In the first step, the MCNPX (CEM03 cascade model) simulation was done with the thin, central beam and the obtained results were compared to the experimental measured values. Most experimental values were described well, with the two points mentioned in 2.1.3 (maximum at 10\textsuperscript{th} cm and faster decrease of production rates after 30\textsuperscript{th} cm) at the right places, and with the differences between experimental and simulated production rates which were within 30%.

In the next step, a set of MCNPX simulations with changed setup parameters was performed in order to study the influence of the setup parts such as concrete walls and iron components on the experimental results, and to estimate the systematic uncertainty of the experimental results (mainly because of the beam parameters).

Concrete walls and iron parts

Concrete walls moderate neutrons coming from the target and reflect part of moderated neutrons back to the setup place, thus they produce an almost homogenous field of low energy neutrons around the target. Calculated neutron spectra along the target for the case with the walls included and for the case without walls are seen in the Figure 2.5 together with the ratios between them. It is important to stress that the high energy part of the produced neutron spectrum is not changed due to the walls (the ratio between the spectra is 1 within the error bars), there is no physical mechanism how high energy neutrons could be reflected back to the setup. The same conclusion applies also to protons. Calculations of production rates of threshold reactions for the setup with and without walls confirmed that the results do not differ outside the statistical uncertainties which were 2%. One can conclude that the walls have no influence on high energy neutron field (and on production rates in threshold detectors). However, they change significantly the neutron field for neutrons with energies < 100 keV, neutrons scattered from the wall contribute from 20%-90% to the production rates of \textsuperscript{198}Au (at the beginning and at the end of the target, respectively), as seen in the Figure 2.6.

In the iron parts of the setup, a mechanism that could change the high energy neutron field exists. Heavy Fe nuclei can scatter neutrons, and additionally, in spallations or (n,xn) reactions in iron, more neutrons can be produced. Calculations were performed to estimate the importance of these effects. A simulation was performed with the iron parts approximated as described in the introduction of Section 2.2, and another where iron parts were replaced with air. It was found out that iron parts have negligible influence on threshold and (n,\gamma) reactions. With the iron parts included, the reaction rates after 30\textsuperscript{th} cm are 5% lower than without iron (statistical uncertainties are 5%).

Beam parameters

Several simulations with miscentered beams and different beam profiles were compared. The beam was at first approximated with a point beam, directed to the center of the target. In the following five simulations, the beam was displaced every time for 0.3 cm upwards.
Figure 2.5: The neutron spectra along the target length calculated with MCNPX CEM03. The left upper figure shows the case without the concrete and the right upper figure shows the case, where concrete walls moderated fast neutrons and reflected part of them back to the setup creating almost homogenous low energy neutron background. On the lower figures are plotted the ratios between the calculated spectra for the setup without and the setup with concrete walls. On the left are the ratios for the whole energy spectrum along the target, and on the right are shown the ratios between spectra at energies 0.1-660 MeV at positions 10 cm and 30 cm from the beginning of the target. Concrete walls have no influence on the neutron spectra in the energy range 1-660 MeV.
This direction should have the biggest influence on the results - it is the direction towards
the detectors placed on top of the target. In the detectors on top of the setup and iodine
samples the increase in production rates for non-threshold reaction \((n,\gamma)\) was 10% for each
displacement. For the threshold reactions, the increase at every displacement was 10% and
15% for \((n,2n)\) and \((n,10n)\) reactions, respectively. The differences are significant only up to
the 30th cm, which the range of the proton beam.

Then another direction of the displacement was chosen, the beam was displaced to the
left. It was found out, that displacement to the left has much lower influence on the results:
the displacement of the beam for 1.5 cm lowered the production rates (threshold and non-
threshold reactions) for less than 10%.

Finally, the calculations with the beam approximated with the Gaussian profile with
FWHM = 2 cm and FWHM = 4 cm were compared to the first simulation with the point
beam. The reaction rates increased with the width of the beam. For the beam with FWHM
= 4 cm, the increase is 15% and 40% for \((n,2n)\) and \((n,10n)\) reactions respectively. The
changes are significant only in the detectors at the beginning of the target, after the 10th
cm, the changes are much smaller. For the beam with the FWHM = 2 cm (the experimental
beam had smaller FWHM than 2 cm), no changes were observed from the point beam. The
beams with the cylindrical shape behave similarly as those with the Gaussian profile.

As the profile and horizontal displacements of the beam do not influence the experimental
results significantly, the only source of systematic uncertainty is the beam displacement on
the vertical axis. The accuracy of the beam position was 3-4 mm what brings 15% systematic
uncertainty in the experimental results of the top detectors and iodine samples.

**Detectors and samples displacement**

The calculation with the detectors displaced to the left for 0.3 cm was performed. The
reaction rates did not differ from the reaction rates for not displaced detectors within the

Figure 2.7: The production rates for $^{197}$Au($n,7n$)$^{191}$Au and $^{197}$Au($p,6np$)$^{191}$Au reactions (and their sum) along the target calculated with MCNPX. Around the 30th cm, the protons contribute 50% to the total production rate. Scale on Y axis is linear, so that the second peak is better seen. Statistical errors (ca. 15%) are not shown on the graph.

limits of statistical uncertainties (5%). The displacement of the detectors upwards for 2 mm produces for ca 5% lower reaction rates in detectors at the beginning of the target, the difference decreases to 0 around the 20th cm.

Another calculation with the detectors displaced for 0.3 cm along the target showed that in the detectors placed far from the 30th cm, the reaction rates are not sensible to such displacement. For the isotopes produced through ($n,xn$) and ($p,\text{(x-1)np}$) reactions with $x>4$, there is another peak in production rates around the 30th cm, see Figure 2.7. It is caused by primary protons, which are deviated from their initial direction by coulomb interactions and reach the target surface around this point. They contribute up to 50% to the production of isotopes from higher ($n,xn$) reaction. The peak maximum moves to the neighbor detector if detectors are displaced along the target for 0.3 cm. This is also observed if the target is simulated with extra 0.5 mm air gaps inserted between the segments. The production rates in the peak change for 50% when detectors are displaced along the target or the target is extended with gaps between segments comparing with the normal setup.

Apart from the foils near the 30th cm, the detectors are not sensible to small displacements along the target. The detectors and target positions are known with the accuracy ca. 1 mm, the systematic uncertainty is below 5%.

Similar calculations were performed for iodine samples. The accuracy of placement of these samples was not so good and 0.5 cm displacement along the target or in the upward direction are possible. The systematic uncertainty of the experimental results in the iodine samples was calculated to be 30%.

**Proton and pion induced reactions**

Part of the radioactive material in the detectors is produced by protons and pions (only in threshold reactions). The calculations showed that the production of radioisotopes in reactions with pions is at least three orders of magnitude lower than the production in
Figure 2.8: a) Ratios between experimental and simulated B-values in beam monitor foils placed in the gap. The beam in this simulation was approximated with the Gaussian profile with FWHM in the X and Y direction 0.7 and 0.8 cm and displaced for 1.1 cm upwards and 0.1 to the right. b) Ratios between experimental and simulated B-values in Au and Al detectors placed along the target. INCL4/ABLA models were used to simulate B-values.

reactions with neutrons and thus negligible. Protons influence mainly the production rates of \((n,xn)\) reactions with higher \(x\), and their influence is the biggest around the 30th cm of the target (the point of rapid decrease of the neutron field). At that point their contribution to the total production rate was 10% for \((n,2n)\), 40% for \((n,6n)\), and 50% for \((n,9n)\) reaction, see Figure 2.7.

2.3 Simulations - comparison of code predictions with experimental results

2.3.1 Determination of the beam parameters by simulations

The exact conclusions about the beam shape and position were not possible from the experimental data. Few MCNPX simulations (CEM03 cascade model) with different beams were performed to find the approximation of the beam, that would produce the reaction rates in the monitor foils and in the top foils close to the experimental ones.

The beam data from the cross of monitor foils suggested that the beam was displaced upwards, so that the center is somewhere between the central and the top foil, and that the beam FWHM is 0.7 and 0.8 cm in the X and Y direction. Such a beam describes the reaction rates in the monitors well, but predicts 1.6 times higher values in the top detectors (Fig. 2.8).

Obviously, the beam was not displaced upwards (also the data from the wire chamber show that the beam was centered to the target axis). The simulation with the centered beam (FWHM in the X and Y direction were 0.7 and 0.8 cm) predicts the values in the top detectors well. It predicts well also the values in the cross of the monitor detectors, assuming that the cross was displaced downwards for 0.5-1 cm.
The conclusion about the exact beam position could not be made, because the data from the wire chamber and from the cross of monitor detectors do not agree. From the simulations and the experimental data we assume that the beam was centered, but its position uncertainty is ca. 3 mm. The simulations from the section 2.2.1 concerning the beam parameters showed that the systematic uncertainty of the experimental results on the top of the target is therefore 15%.

### 2.3.2 Simulations of neutron fluences in detectors on top of the setup

**Simulations with CEM03 cascade model**

The complete setup was then simulated with the beam parameters which were determined above. The calculations were successful in describing the spatial distributions and the absolute values of production rates along the target.

The distribution of low energy neutrons along the target which was calculated predicts an almost homogenous distribution (as the experiment), but experimental values for $^{198}$Au are ca. 1.5 higher than calculated production rates. However, the experiment was not focused on low energy neutrons, the structure details about concrete walls were not known accurately, and this underestimation is explained with the material and geometry uncertainties of concrete walls which influence low energy neutrons significantly, see Figure 2.6.

The calculated production rates of threshold reactions (high energy neutrons) describe the experiment well: there is a maximum at around 8$^{th}$ cm, and near the 30$^{th}$ cm the values start to decrease faster. The absolute values are described well except for some isotopes ($^{191}$Au, $^{202}$Bi), see Figure 2.9.

A sharp peak for some isotopes ($^{191-192}$Au, $^{202-205}$Bi) in experimental/calculation ratios around the 30$^{th}$ cm is also visible in the graph. This is the point, where the protons exit the target material and produce radioactive isotopes in the detectors and the peak can be
Figure 2.10: Ratios between experimental and simulated B-values in Au and Al detectors (a), and in Bi detectors (b). INCL4/ABLA models were used to simulate B-values.

explained with the systematic uncertainties of the experimental data (see Section 2.2.1). The results around this point are very sensitive to two parameters of the setup that could not be controlled enough precisely: the displacement of the detectors along the target (uncertainty 1-2 mm) and small gaps between the target segments (1 mm). The additional simulation with extra 1 mm gaps between the target segments showed that the peak is reduced while the other ratios stay unchanged.

Simulations with INCL4/ABLA cascade model and FLUKA code

Simulations were repeated using the INCL4/ABLA model from MCNPX code package. The comparisons between the experimental and calculated values in the beam monitors and detectors on top of the setup are shown in Figure 2.10. INCL4/ABLA predicts similar results as CEM03, with some ratios closer to 1 and with a bit decreased peak around the 30\textsuperscript{th} cm. It is worth noting that both simulations predict similar ratios for isotopes $^{196}$Au and $^{24}$Na, but disagree in the ratios of isotopes with higher thresholds ($^{191-192}$Au, Bi).

Using the same setup approximations as for the MCNPX simulations (see 2.2), the neutron and proton fluences were calculated with the FLUKA 2006.3b code. The numbers of neutrons/protons were convoluted with the same cross-sections as for MCNPX simulations.

In the Figure 2.11 it is seen that the ratios for different isotopes in FLUKA calculation are closer to 1 than in MCNPX calculations and also that the peak around the 30\textsuperscript{th} cm is reduced. Only in the detectors at the beginning of the target, experimental values are significantly higher than FLUKA prediction.

Comparison between codes/models

The neutron and proton spectra in the detector foils on top of the setup were calculated with MCNPX models CEM03 and INCL4/ABLA and with the FLUKA code and were compared with each other. In the Figure 2.12 are compared the neutron spectra in the detector foil at the 9\textsuperscript{th} cm. The biggest disagreement between spectra is in the energy region below 3 MeV.
Figure 2.11: Ratios between experimental and simulated B-values in Au and Al detectors (a), and in Bi detectors (b). FLUKA 2006.3b code was used to simulate B-values.

and above 30 MeV and is up to 50%. This disagreement is observed in different predictions of high threshold reaction rates by different codes (e.g. $^{191}$Au in Figures 2.9, 2.10, 2.11). The neutrons with energies above 30 MeV present less than 10% of all produced neutrons. Concerning the total number of produced neutrons per one incident proton, the codes are in good agreement. The FLUKA code and MCNPX INCL4/ABLA predict values 11.8 and 11.7 produced neutrons per one primary proton and MCNPX CEM03 predicts slightly higher value 12.6 produced neutrons per one primary proton.

### 2.3.3 Simulations of neutron fluences in iodine samples

The neutron and proton fluences in iodine samples were calculated with the MCNPX code package using the INCL4/ABLA models. The fluences were convoluted with cross sections which were also calculated with TALYS/MCNPX. In the Figure 2.13 are shown the ratios between the experimental and simulated production rates in iodine samples. In a rude approximation, one can see that MCNPX overpredicts the production rates. It must also be noted that the systematical uncertainties of the experimental data in the samples was close to 50% because of the samples and beam position uncertainty. The simulations with other models and with the FLUKA code predict similar results.
Figure 2.12: The neutron (a) and proton (b) spectra in the detector foil on the 9th cm calculated with the MCNPX CEM03, MCNPX INCL4/ABLA and the FLUKA code, and the ratios between the calculated neutron spectra (c). In (d) are the cumulative reaction rates (in relative units, normalized to 1) calculated with MCNPX CEM03. It can be seen that $^{24}\text{Na}$, $^{194}\text{Au}$, $^{192}\text{Au}$ and $^{201}\text{Bi}$ are produced mainly with 10, 30, 60 and 90 MeV neutrons, respectively.

Figure 2.13: Ratios between experimental and simulated B-values for different isotopes in $^{127}\text{I}$ and $^{129}\text{I}$. Samples were placed at 9th (a) and 21st cm (b). INCL4/ABLA was used to simulate B-values.
Part II

SPALLATION NEUTRONS ON TARGET WITH URANIUM BLANKET
Chapter 3

Energy Plus Transmutation experiments

3.1 Experimental setup

The target-blanket part of the EPT setup [60] is composed of four identical sections. Each section contains a cylindrical lead target (diameter 8.4 cm, length 11.4 cm) and 30 natural uranium rods (diameter 3.6 cm, length 10.4 cm, weight 1.72 kg) distributed in a hexagonal lattice around the lead target. The lead target and uranium rods are enclosed in aluminum claddings of thicknesses 2 mm and 1 mm, respectively. The target and uranium rods in each section are secured in hexagonal steel container with a wall thickness of 4 mm. The front and back of each section are covered with a hexagonal aluminum plate of thickness 5 mm. The four target blanket sections are mounted along the target axis, on a wooden plate (thickness 6.8 cm) covered with 0.4 cm thick steel sheet. There are 0.8 cm gaps between the blanket sections which are used for placement of activation and other detectors. The four target blanket sections mounted on the wooden plate are placed in a wooden container filled with granulated polyethylene, density of which was measured to be 0.8 g cm$^{-3}$. The inner walls of the polyethylene box are covered with 1 mm thick cadmium layer. The floor wall of the polyethylene box is a textolite plate of thickness 3.8 cm. The polyethylene box and cadmium are used to modify the neutron spectrum as will be discussed in this paper. The geometrical arrangements and dimensions of the EPT setup are shown in Fig. 3.1.

Several experiments have been carried out using the EPT setup and its target was irradiated with relativistic protons of energies in the range of 0.7 to 2 GeV. In these experiments the neutron flux was measured using activation foils (radiochemical detectors) that were placed between the blanket sections. The radiochemical detectors (aluminum, gold, bismuth, yttrium, and other monoisotopic materials) with dimensions of 2 cm × 2 cm and the thickness of ca. 0.1 mm were used. Various nuclear reactions, e.g. (n,$\gamma$), (n,xn), (n,$\alpha$), occur in the radiochemical detectors. The production rates of the reaction products were determined from their gamma spectra and characteristic peaks.

For thermal, epithermal, and resonance neutrons, the dominant reaction is the neutron capture (n,$\gamma$) process for which cross-sections are large (in the range of hundreds to thousands of barns). The others are threshold reactions for which cross-section are in range of mbarns.
to barns. At the end of the irradiation, the activities of detectors were measured by the means of HPGe detectors and activities were converted into production rates $B(A)$, which give the number of produced nuclei of the isotope A normalized to 1 g of the activation detector and to 1 primary proton [60]. We will talk about these in Sec. 3.7.

3.2 Simulation procedure

MCNPX v2.6.c [62] and FLUKA [46, 47] Monte Carlo codes were used to simulate the behavior of neutrons and other secondary particles in the experimental setup. The EPT setup was defined in the codes with the characteristics given in Fig. 3.1. Fig. 3.2 illustrates the EPT setup as seen by the MCNPX code. Fig. 3.2a shows the MCNPX plot of the XY (a plane normal to the target axis, Z) cross-section of the EPT setup, while in Fig. 3.2b the YZ cross-section of the EPT is shown. In Fig. 3.2b the control detectors with their corresponding reference numbers are added to the MCNPX plot.

In order to obtain $B(A)$ production rates at different places of the setup, the simulated spectra of neutrons, protons and pions were convoluted with the cross-section for the specific reaction. The missing cross-sections were previously simulated (calculated with TALYS up to 150 MeV, for higher energies was used MCNPX and CEM03 intranuclear cascade model, extracted with FT8 RES card). The simulated spectra at the places of the detectors were convoluted with the cross-sections. In the case of the $(n,\gamma)$ reactions the cross-sections from the MCNPX libraries were used (these are ENDF-VI), in FLUKA $(n,\gamma)$ reactions were not simulated.

In order to investigate the role of different parts of the experimental setup on the obtained results, simulations with changed geometrical and physical properties of the setup were
Figure 3.2: a) The front cross-section of the target placed in the polyethylene box (MCNPX plot).

b) The side cross-section of the target placed in the polyethylene box (MCNPX plot) with enumerated control detectors (bigger than in simulation).

Figure 3.3: lot of the target placed in the polyethylene box (SABRINA plot of the MCNPX input file, provided by Jaroslav Šolc).
performed in the MCNPX code. The influence of the Intra-Nuclear Cascade model and the cross-section libraries was estimated by series of calculations with different libraries and INC models. MCNPX was also used to calculate the criticality of the experimental setup, as well as the number of produced neutrons per one incident proton. These two parameters are crucial when comparing the EPT setup with similar setups. The simulated results by both codes, MCNPX and FLUKA, were at last also compared to some experimental results of the 1.5 GeV experiment.

3.2.1 Control detectors
Five thin gold foils were used as control detectors in simulations. Foils 1 and 2 were placed in the first gap between the target blanket sections, at the radial distances of 3 and 11 cm from the target axis. The foils 3 and 4 were at the same radial positions as the foils 1 and 2, but in the third gap. The foil 5 was in the horizontal position on the top of the second blanket section. The positions of the control detectors are shown in Fig.3.2b.

In the control detectors (foils) two reactions of $^{197}$Au($n,\gamma$)$^{198}$Au and $^{197}$Au($n,2n$)$^{196}$Au were simulated. The reaction $^{197}$Au($n,\gamma$)$^{198}$Au is sensitive to the low energy neutrons ($E_n < 0.1$ MeV) while the $^{197}$Au($n,2n$)$^{196}$Au reaction has a threshold energy of 8 MeV and therefore shows the behavior of the high energy neutrons ($E_n > 8$ MeV) in the EPT setup. The variation of the production rates of these two isotopes by altering the parameters of the experimental setup was investigated. The calculations were performed when the incident proton energy was 1.5 GeV.

3.3 The influence of the setup parts and experimental conditions on the neutron spectrum

3.3.1 The influence of the polyethylene box and cadmium layer
The polyethylene box around the target-blanket moderates part of the neutrons and reflects them back inside the box. The 1 mm thick cadmium sheet that covers the inner walls of the polyethylene box absorbs most of the reflected slow neutrons. A set of simulations (without box, with box but no cadmium, and with both - box and cadmium) showed that only reflected neutrons with energies less than $10^{-6}$ MeV are stopped by the cadmium layer (Fig. 3.4a). The box and cadmium do not affect by more than a few percent high energy ($E_n > 10$ MeV) part of the neutron spectrum (Fig. 3.4b). From Fig. 3.4 is evident that the low energy part of the neutron spectrum in the blanket area has been produced by the combined effects of the polyethylene and cadmium around the target blanket system. The spectra shown in Fig. 3.4 were calculated on top of the second section of the target-blanket.

High energy neutrons which are produced in spallation reactions present a risk for the environment. Therefore, a box filled with granulated polyethylene was designed around the target-blanket assembly having function of biological shielding with little influence on high energy neutron spectrum inside the box. To achieve that, the inner walls of the box were covered with 1 mm cadmium layer, low energy neutron absorber. A set of simulations
Figure 3.4: a) The simulated neutron spectra on top of the second section of the target-blanket are shown for three cases: for the target-blanket without the polyethylene box, for the target-blanket with the box but no cadmium, and for the target-blanket with both, the box and the cadmium. Small thermal peak in the case of tb+box+Cd is caused by the moderation effect of the wood.

b) The ratios of the spectra from the left figure from the energy 0.1 MeV. From these ratios it can be concluded that the polyethylene box affects significantly only neutrons with energies lower than 10 MeV. The increase of the ratios 1-10 MeV range is caused by the fission of $^{235}$U with moderated neutrons.

(without box, with box but no cadmium, and with both - box and cadmium) of neutron spectra at the place of the target blanket were compared, showed that only neutrons with energies less than 1 eV are stopped by the cadmium layer, and that field of neutrons ranging from 1 eV to 0.1 MeV inside the polyethylene box is produced by the combined effect of the polyethylene and cadmium [63].

Figure 3.5 shows calculated neutron spectra emitted to the environment for the target-blanket alone and for the target-blanket placed in the biological shielding. In order to obtain the spectra, neutrons crossing the surface of the virtual sphere surrounding the whole setup were counted. As can be seen, the polyethylene box essentially decreases the flux of emitted high energy neutrons by moderating them to lower energies. Calculations suggest that from 50 neutrons that are produced per one proton at 1.5 GeV, 42 would escape to the environment in the hypothetical case without the shielding, but with the shielding only 10 neutrons escape, 8 from these through front and back openings in the polyethylene box.

3.3.2 The influence of other setup parts (metal parts, wood)

Experimental data have shown that at the bottom part of the target-blanket system there are more low energy neutrons than at its upper part [64]. To verify if this is due to the wooden and textolite plates under the target-blanket system, the following three simulations were performed:

1. both wooden plate and polyethylene box were present,

2. only wooden plate was present,

3. only polyethylene box was present.
Figure 3.5: Calculated neutron spectra emitted to the environment from the target-blanket only and from the target-blanket surrounded by the polyethylene box.

Figure 3.6: The $^{197}$Au(n, $\gamma$)$^{198}$Au production rates in detectors placed along the vertical axis, Y in the first gap. The MCNPX calculations were performed for three different material compositions of the EPT setup as shown in the figure inset.

Fourteen $^{197}$Au detectors were placed in the first gap along the vertical axis Y in the interval of -14 to 14 and $^{196}$Au, $^{198}$Au production rate in each detector was determined. The wooden and textolite plates were approximated with the wood from the MCNPX materials library [65] and atomic fractions of 51%, 23%, and 26% were used for H, C and O respectively. The same density of 0.5 kg/l was used for the wood and textolite. The calculation results are shown in Fig. 3.6. In the case of the high energy neutrons, no asymmetry beyond the 5% was observed between the $^{196}$Au production rates in the Au-detectors in +Y direction as compared to their corresponding detectors in the -Y direction. However, in the case of the low energy neutrons the $^{198}$Au production rate is dramatically affected by the presence of the wooden and textolite plates. The polyethylene box alone (in absence of the wooden and textolite plates) produces almost homogenous, low energy neutron field in the first gap. This is expected due to the geometrical and material symmetry of the EPT setup in absence of wooden and textolite plates.
The metallic materials (steel and aluminum) used in the target blanket sections (as described in Sec. 3.1) do not have significant effect on the neutron spectrum within the blanket. In general, the effects of these parts on the reaction rates in the control detectors do not exceed the statistical uncertainties of the calculations which were about 3%.

3.3.3 The influence of detector selfshielding

The detectors that were used in the experiments had small dimensions and thus, no significant neutron flux selfshielding is expected. However, some extreme cases where the detectors could influence the experimental results were studied.

With the detector type and dimensions used in the experiments, in principle the detectors in one gap should have negligible influence on detectors in other gaps or on those detectors outside of the target-blanket assembly. This was proved by placing gold foils with thicknesses 2 and 4 mm in the first gap (extended over the whole gap) and calculating the reaction rates in the detectors in the third gap (i.e., foils 3 and 4 in Fig. 3.2b). No significant effects on the reaction rates outside of the 3% statistical uncertainties were observed.

A gold strap of 2 cm wide and 0.1 mm thick, stretching over the whole gap was placed in front of the detectors in the first gap. Subsequent simulations showed that the rate of the \(^{197}\text{Au}(n,\gamma)^{198}\text{Au}\) reaction in the detectors behind gold strap was reduced by up to 15%, while the rate of the \(^{197}\text{Au}(n,2n)^{196}\text{Au}\) reaction did not change within the statistical uncertainties (3%). The strap should not have any significant effect on the high energy part of the neutron spectrum, as neutrons at that energy have small cross-sections for the reactions with the gold. Only the influence of the low energy neutrons with large cross-section resonances with the gold is expected.

Calculations also showed that when gold foils were covered on both sides with bismuth foils of thickness 1 mm the production rates of the threshold reactions do not change beyond the calculation uncertainties. On the other hand, absorption in gold has significant effect on reactions with low energy neutrons, i.e., \(^{198}\text{Au}\) production rates in 50 \(\mu\)m thick gold foils are 50% lower due to self-absorption. Self-absorption for threshold reactions is negligible. This suggests that the threshold detectors can be mounted one after another within the gaps.

In the earlier experiments with the EPT setup, the activation detectors were mounted on a thick, plastic plate, and then placed in the gaps between the blanket sections. Such an arrangement may affect the low energy section of the neutron spectrum in the gaps. MCNPX calculations of the neutron spectrum in the gap in which a polyethylene plate of thickness 2 or 6 mm is inserted showed that such a plate has no effect on the high energy neutrons \((E_n > 10\text{ MeV})\), but, changes the low energy part of the spectrum, see Fig. 3.7.

Another source of the systematic experimental error is the displacement of the detectors. By simulations it was estimated that a displacement of detectors for 0.5 cm results in reaction rates that are ca. 20% different from the reaction rates with not displaced detectors.

3.3.4 The influence of beam parameters on the reaction rates

The beam parameters in our experiments were experimentally determined. The beam is usually approximated with the Gaussian distributions in X and Y directions, and its dis-

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placement is known with an accuracy of 3 mm. In reality, the beam is Gaussian with the extending tails. To estimate the systematic uncertainty resulting from Gaussian beam approximation and the beam displacement, a set of MCNPX simulations was performed and the reaction rates in the control detectors were computed.

To avoid the influence of the neutrons reflected from the polyethylene box around the target-blanket system, calculations were performed without the polyethylene box. Three calculations were made with two circular and homogenous beams of diameters 3 mm and 3 cm and with a beam of Gaussian profile for which the FWHM in both X and Y directions were 3 cm. In all three cases the beam directions were parallel to the target axis and the beams and target centers coincided. The induced reaction rates in the control detectors for these three proton beam profiles were the same within the statistical uncertainties of the calculations (i.e., 5%). This suggests that in our experimental setup the beam profile is not of a great importance as long as it is symmetric. The tails of the beam are for ca. three orders less intensive than the beam central part and have negligible influence on the control detectors.

In a series of calculations without the polyethylene box, the center of the Gaussian proton beam as described above, was displaced by 3, 5, 8, and 10 mm from the target axis and along the positive direction of the Y axis. The reaction rates in the control detectors showed a strong dependency on the beam displacement. The displacement of the beam 5 mm changes $^{197}$Au(n,2n)$^{196}$Au and $^{197}$Au(n,$\gamma$)$^{198}$Au reaction rates by up to 20% and 30% respectively.

With the presence of the polyethylene box (i.e., the case of the actual experiments) and as a result the contribution of the reflected low energy neutrons, the difference in the $^{197}$Au(n,$\gamma$)$^{198}$Au reactions rates for the cases of centered and displaced beam decreases to about 10% as compared with about 30% when the box was absent. The polyethylene box has no effect on high energy induced reaction rates (i.e., $^{197}$Au(n,2n)$^{196}$Au). A beam centre displacement of 3 mm results in a systematic error of up to 15%. Fig. 3.8a shows the difference between the reaction rates for centered and displaced proton beams (see the figure caption for details).

Another calculation was performed with the beam which was not parallel to the target
Figure 3.8: a) The difference between the reaction rates for centered and displaced proton beams. The proton beam was displaced along the positive Y-axis with the amount given in the figure inset, and calculations were performed when the polyethylene box was present. Foils were placed as seen in Fig 3.2b. The abbreviations (n,2n) and (n,γ) refer to \(^{197}\text{Au}\,(n,2n)^{196}\text{Au}\) and \(^{197}\text{Au}\,(n,γ)^{198}\text{Au}\) reaction respectively.

b) The difference between the reaction rates for the beam parallel to the target axis and for the beam entering at 3 degrees. The abbreviations Au-196 and Au-192 refer to \(^{197}\text{Au}\,(n,2n)^{196}\text{Au}\) and \(^{197}\text{Au}\,(n,6n)^{192}\text{Au}\) reaction respectively.

axis. The beam and the target centers coincided, but the direction of the beam was deflected from the target axis for 3° upwards, exiting the target 2.5 cm from its center. Simulation showed that the deflection of the beam causes the increase of the reaction rates for up to 60% and 40% in reactions \(^{197}\text{Au}\,(n,2n)^{196}\text{Au}\) and \(^{197}\text{Au}\,(n,6n)^{192}\text{Au}\) respectively (Fig. 3.8b).

### 3.4 Isotope production in reactions with protons, pions and photons

Radioactive isotopes in the detectors can also be produced by other particles, mainly by protons, pions, and photons. To estimate the contributions of these particles to the reaction rates in activation detectors, the corresponding reaction cross-sections were evaluated using the MCNPX. The neutron, proton, pion and photon spectra in the control detectors were calculated and then were convoluted with the evaluated cross-sections. It was found that up to 20% of reaction products could be produced by proton induced reactions, suggesting that the influence of protons cannot be neglected. The isotope production by pions and photons is at least one order of magnitude lower than isotope production with neutrons and their influence can be neglected. Most of this contributions are proton induced reaction with protons from the primary beam. The contribution of radioisotopes produced by protons decreases very quickly with increasing radial distance and is strongly dependent on the proton beam profile and position of the beam center on the target.
3.5 Parameters of the simulations: Effects of different physics models and cross-section libraries

The setup was simulated with different combinations of INC (CEM03, BERTINI, ISABEL, INCL4) and evaporation models (DRESNER, ABLA) included in MCNPX, in order to check if these combinations of built-in models predict similar reaction rates.

In the case of $^{197}\text{Au}(n,2n)^{196}\text{Au}$ reaction, different INC models predict reaction rates similar within 10% when using the same evaporation model. These reaction rates differ for 40% from the reaction rates calculated with another evaporation model. The situation for the reaction $^{197}\text{Au}(n,6n)^{192}\text{Au}$ with higher threshold ($E_{thr}=39$ MeV) is inverse, only the use of different INC model changes the results significantly, while the results are not changed if another evaporation model is used.

Cross-section libraries NRG-2003 [34] are available up to 200 MeV in MCNPX code package, apart from the standard LA150 libraries [33], which are available up to 150 MeV. Simulations confirmed that the reaction rates calculated using the cross-sections from NRG-2003 libraries are the same as the reaction rates calculated using the standard LA150 libraries within few percents.

3.6 Simulation of the global characteristics of the EPT setup

Two important parameters of the EPT setup were determined with MCNPX simulations: the criticality ($k_{eff}$), and the number of produced neutrons per one incident proton. Using KCODE, the criticality of the EPT setup was calculated to be $k_{eff}=0.20247$. At the energy $E_p=1.5$ GeV, the overall neutron production per incident proton $m$, is 50 which includes neutrons from spallation process, uranium fission, and (n,xn) reactions. But in Fig. 3.9 the ratio of $m/E_p$ is shown as a function of incident proton energy ($E_p$). As it can be seen the optimal energy for the neutron production in the EPT setup is around 1 GeV.
Figure 3.10: The radial (a) and longitudinal (b) distributions of the experimental reaction rates (B-values) in gold detectors placed in the first gap. The lines are drawn to guide the eyes. The statistical uncertainties of the points are not visible on this scale.

3.7 Comparison of experimental data and calculation results

Almost all EPT experimental results were already published [60]. The experiment with 1.5 GeV proton irradiation was chosen as at this energy (and at the energy 2 GeV), interesting discrepancies between experiment and simulation were observed.

Figure 3.10 shows the spatial distribution of some threshold reaction rates (the B(A) values) in the gold at the incident proton energy of 1.5 GeV. The gold detectors were placed within the first gap at radial distances 3, 6, 8.5 and 13.5 cm, and in other gaps, as well as in front of and behind the target at the radial distance 3 cm. The threshold energy for $(n,xn)$ reactions, $(x=2$ to $7)$ are in the range of $8$ MeV to $40$ MeV. The $B$-values for all reactions rapidly decrease with increasing distance from the target axis. The decrease in longitudinal direction is slower, with the maximum of the $B$-values in the first gap (12 cm after the beginning of the target).

The reaction rates were calculated with MCNPX and FLUKA codes. The INCL4/ABLA and CEM03 models and LA150 cross-section libraries were used in the MCNPX code. In the FLUKA code, its own model of spallation reaction PEANUT and cross-section libraries imported from ENDF/B-VI were used. The input file of the MCNPX EPT setup was translated to FLUKA input file and simulated with the FLUKA code.

The ratio between the experimental and calculated results ($\kappa = B_{\text{exp}} / B_{\text{sim}}$) increases slightly with the increasing longitudinal distance, while in the radial distance the increase is much more dramatical, see Figures 3.11. Reasons for such large discrepancies are under investigation.

The ratios for isotopes $^{193}\text{Au}$ and $^{191}\text{Au}$ differ from other ratios. This is probably due to the systematical error at the spectrometry analysis. The calculations would predict differently also the $B$-values for $^{192}\text{Au}$ if that was the error in the calculated neutron spectrum.

The CEM03 model from the MCNPX code package and the FLUKA code were used to simulate the same setup. In the Figures 3.12 and 3.13 are the $\kappa$ values calculated with CEM03 and FLUKA. The results predicted by FLUKA seem to be similar to INCL4/ABLA predictions, while CEM03 predicts a bit different values.
Figure 3.11: The ratios between the experimental values and simulated B-values in the radial (a) and in the longitudinal (b) directions. The INCL4/ABLA models from the MCNPX code package were used in the calculation.

Figure 3.12: The ratios between the experimental values and simulated B-values in the radial (a) and in the longitudinal (b) directions. The CEM03 model from the MCNPX code package was used in the calculation.

Figure 3.13: The ratios between the experimental values and simulated B-values in the radial (a) and in the longitudinal (b) directions. The FLUKA code package was used in the calculation.
Chapter 4

Conclusion

The Phasotron experiment with a thick, bare, lead target provided a large set of experimental data, useful for the benchmark of Monte Carlo codes. Neutrons, produced at the irradiation of the target with relativistic protons, were probed with many small activation detectors, which provided information about high and low energy neutrons. The focus of the experiment was on the production of neutrons with energies higher than 10 MeV (representing one tenth of all produced neutrons), the energy region where the predictions of various Monte Carlo codes are not yet accurate. The parameters of the setup were not appropriate for the measurements of the low energy part of the produced neutron spectrum and the results concerning low energy neutrons are useless for benchmark tests.

The simulation procedure was based on convolution of the calculated neutron and proton spectra with the pre-computed cross sections (TALYS code). With the comparison of the simulation results (setups with changed parameters were simulated using MCNPX) it was found out that the systematic experimental errors are 15%, with the exception of few particular detectors (detectors around the 30\textsuperscript{th} cm). The biggest systematic uncertainty arises with the uncertainties in the beam position, which should therefore be controlled with most attention. Unfortunately, the additional monitor detectors in this experiment were obviously misplaced, which caused the mentioned systematic uncertainty. However, reliable data on high energy neutron and proton production and transport was obtained. The results concerning the transmutation properties of $^{129}$I in high energy neutron field are less accurate, because of geometrical and material uncertainties of the samples.

The benchmark tests with several cascade/evaporation models included in the MCNPX code package and with the FLUKA code showed consistent results. The codes successfully predict the general trends of the results and with some exceptions (which could be the systematical error) also the absolute values. The differences between the codes are minimal in the prediction of the production isotopes with lower threshold, but they become significant for some isotopes with threshold above 30 MeV. From the comparison with experimental data, it seems that the FLUKA code describes the neutron/proton spectrum after the 10\textsuperscript{th} cm better than models included in MCNPX. Concerning the total number of produced neutrons in the setup, the calculations by various codes are in good agreement and predict 11.7-12.6 neutrons per one primary proton.

The "Energy plus Transmutation" setup is used for the studies of neutron production...
and transport, transmutation of radioactive materials and other aspects of accelerator driven systems. The neutronics of the system is studied with activation radiochemical detectors. In this paper, it is shown that the experimental data for higher energies \((E > 10 \text{ MeV})\) from this type of detectors are not influenced (within the accuracy of 5\%) by the polyethylene box, the material of different holders, other construction details, or by the detectors. The systematic uncertainty mostly depends on the beam and detector displacement - the inaccuracy 3 mm in beam or detector position brings ca. 15% systematic uncertainty in the production rates. The dominant sources of epithermal, resonance, and thermal neutrons are moderation and scattering of the neutrons in the polyethylene box. Therefore, the flux of low energy \((E < 0.1 \text{ MeV})\) neutrons is almost homogenous at the place of the target-blanket assembly. The experiments with the EPT setup provide valuable data with experimental uncertainties within the range of 30\% for high energy part of the neutron spectrum \((E > 10 \text{ MeV})\).

The differences between the experimental values and the values calculated with MCNPX are within the limits of the experimental uncertainties for the experiments with 0.7 GeV and 1 GeV proton beams, but, the experimental values for the experiments with 1.5 GeV and 2 GeV proton beams were few times higher than the simulated values. These differences are not within the limits of the experimental uncertainties and are being investigated.

Several experiments with the “Energy plus Transmutation” setup were performed. Radiochemical and solid state nuclear track detectors are used to study the neutronics of the system. Calculations for radiochemical detectors are being performed for some time, and their behavior in various experimental conditions is well known. They mainly interact with 10-100 MeV neutrons through \((n,zn)\) and \((n,\alpha)\) reaction channels and are less sensitive to deviations of experimental conditions than SSNT detectors [63]. Studies performed for solid state nuclear track detectors with lead irradiator showed that SSNT detectors detect mostly primary protons and 100-300 MeV neutrons, and are essentially influenced by experimental conditions (mainly beam deviations). However, the comparison between experimental and calculated reaction rates for 1.5 GeV experiment shows that the shape of reaction rate distribution is satisfactory described by simulations with discrepancies up to 50\%. In the case of radiochemical detectors sensitive to neutrons with energies 10-100 MeV, these discrepancies are of the factor of few hundreds percent. These facts imply that the beam parameters are well measured and are not responsible for big discrepancies in the case of radiochemical detectors. Also, it is evident that neutrons with energies higher than 100 MeV are simulated reliably, and discrepancies are caused mainly by neutrons of energies 10-100 MeV. The reasons are under investigation.

The studies of the polyethylene box (primary designed as biological shielding) showed that it decreases the number of neutrons escaped to the environment for 75\%. Most neutrons escape through front and back openings of the box. To lower the number of escaped neutrons, it would be useful if two polyethylene walls were added in front and in the back of the target-blanket. Next improvement would be another cadmium layer at outer walls of the polyethylene that would prevent the emission of thermal neutrons to the environment.
4.1 Discrepancies

show here $2^{over05}$, 2 for different models, absolute results incl/abla, put attention to incl/abla having the lowest increase $2^{over05}$ say that in frames of EPT the answer is not possible
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