CZECH TECHNICAL UNIVERSITY IN PRAGUE FACULTY OF NUCLEAR SCIENCES AND PHYSICAL ENGINEERING

———— HABILITATION THESIS ————

Research of neutron fields and utilization of activation analysis in interdisciplinary approach

Commented set of scientific papers

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Abstrakt

Habilitačná práca, spracovaná formou komentovaného súbora odborných publikácií, sa zaoberá aplikáciami neutrónovej aktivačnej techniky na urýchľovačom riadenom neutrónovom zdroji NG-2 pri izochrónnom cyklotróne U-120M na ÚJF AV ČR v Řeži a na školskom jadrovom reaktore VR-1 prevádzkovanom na FJFI ČVUT v Prahe. Na neutrónovom zdroji NG-2 boli skúmané neutrónové polia zdrojovej reakcie p + Be pre protónové zväzky s energiami 35 MeV a 24 MeV a zdrojovej reakcie d + Be pre deuterónové zväzky s energiami 20 MeV a 15 MeV. Stanovené boli spojité neutrónové spektrá reakcií p(35) + Be, p(24) + Be, d(20) + Be a d(15) +Be. Novo vyvinuté neutrónové polia sú dôležité pre experimentálne úlohy validácie jadrových dát a testy radiačnej odolnosti elektroniky voči rýchlym neutrónom, pre úlohy materiálového výskumu a neutrónovú aktivačnú analýzu. Ďalej na školskom jadrovom reaktore VR-1 bola využitá metóda neutrónovej aktivačnej analýzy a bola posudzovaná možnosť využitia reaktora v kombinácii s aktivačnou analýzou ako nástroja v interdisciplinárnom prístupe pri skúmaní vzoriek zaujímavých z pohľadu humanitných smerov. Úspešne bolo analyzované zloženie európskych potravinových doplnkov, tibetských historických mincí, tibetských tradičných liečiv, ostatky mamuta srstnatého, meteoritov a Vltavínov. Zároveň boli merané aj parametre neutrónového poľa na školskom jadrovom reaktore VR-1.

Abstract

The habilitation thesis is compiled in the form of commented set of scientific papers and deals with the applications of the neutron activation technique at the accelerator-driven fast neutron source NG-2 connected to the isochronous cyclotron U-120M of the NPI CAS in Rež and at the VR-1 training reactor operated by the FNSPE CTU in Prague. At the NG-2 neutron source, the neutron fields of the p + Be source reaction for the proton beams with energy of 35 MeV and 24 MeV and neutron fields of the d + Be reaction for 20 MeV and 15 MeV deuteron beams were investigated. New neutron energy spectra of the p(35) + Be, p(24) + Be, d(20) + Be and d(15) + Be reactions were determined. Newly developed neutron fields are important for experimental tasks of nuclear data validation, radiation hardness tests of electronics against fast neutrons, for material research, and neutron activation analysis. In addition, the method of neutron activation analysis was applied at the VR-1 training reactor. The utilization of nuclear reactor in combination with the neutron activation analysis as a tool for investigation of samples interesting for humanities and social sciences within the interdisciplinary research was considered. The composition of the European dietary supplements, historical Tibetan coins, and traditional Tibetan medicinal pills, meteorites and tektites, and remains of woolly mammoth was successfully analyzed. Moreover, the neutron field parameters of the VR-1 training reactor were measured as well.

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List of symbols

a_i	Coefficients of fitting function for detection efficiency $(-)$
A	Activity (Bq)
$A_{\rm sat}$	Saturated activity (Bq)
A_{sample}	Activity of investigated sample (Bq)
A_{standard}	Activity of standard sample (Bq)
A_0	Reference activity of calibration standard (Bq)
$B_{ m hf}$	Hyperfine magnetic field (T)
C/E	Calculated over experimental reaction rate ratio $(-)$
E_{γ}	γ -ray energy (keV)
E	Energy (MeV)
$E_{\rm d}$	Deuteron energy (MeV)
E_{\max}	Maximum neutron energy (MeV)
E_{\min}	Minimum neutron energy (MeV)
$E_{\rm n}$	Neutron energy (MeV)
$\overline{E}_{ m n}$	Mean energy of neutron spectrum (MeV)
$E_{\rm p}$	Proton energy (MeV)
$E_{ m th}$	Threshold energy (MeV)
$E_{\rm thresh}$	Threshold energy (MeV)
$f_{ m Cd}$	Cadmium correction factor $(-)$
g_0	We stcott's correction factor for thermal neutron region (-)
$I_{ m d}$	Deuteron beam current (μA)
I_i^{d}	Deuteron beam current in <i>i</i> -th irradiation interval (μA)
$I_{ m mean}^{ m d}$	Mean value of deuteron beam intensity at target in experiment (μA)
$I_{ m p}$	Proton beam current (μA)
$I_{ m total}^{ m p}$	Total number of charged particles $(-)$
I_{γ}	Intensity of γ -line (%)
$L_{\rm C}$	Critical level $(-)$

$L_{\rm D}$	Detection limit $(-)$
m	Mass (g)
$m_{\rm sample}$	Mass of element in investigated sample (g)
m_{standard}	Mass of element in standard sample (etalon) (g)
N(t)	Number of radioactive nuclei (–)
N_0	Number of target nuclei (–)
$N_{ m nom}$	Reactor nominal power (cps)
$N_{ m y}$	Reaction yield (-)
P	Production rate (s^{-1})
P_{R}	Reaction rate per one target nucleus (s^{-1})
$P_{ m R}^{ m Cd}$	Reaction rate for Cd-covered activation foil (s^{-1})
$P_{\mathrm{R}}^{\mathrm{ep}}$	Reaction rate for epithermal neutron (s^{-1})
$P_{ m R}^{ m th}$	Reaction rate for thermal neutron (s^{-1})
$P_{ m R}^{({ m n},\gamma)}$	Reaction rate for radiation capture (s^{-1})
$P_{ m R}^{({ m n,f})}$	Reaction rate for fission (s^{-1})
Q	Reaction energy (MeV)
Q_{p}	Charge at target (C)
R	Distance (mm)
$r_{ m f}$	Spectral index for fission $(-)$
$r_{\rm Cd}$	Cadmium ratio (-)
R_{R}	Reaction rate per one target nucleus (s^{-1})
$R_{ m R}^{ m b}$	Reaction rate for bare activation foil (s^{-1})
$R_{ m R}^{ m Cd}$	Reaction rate for Cd-covered activation foil (s^{-1})
$R_{ m R}^{ m th}$	Reaction rate for thermal neutron (s^{-1})
S	Peak area (imp)
S_{γ}	Photopeak area in γ -ray spectrum (imp)
$T_{1/2}$	Half-life period of radionuclide (s)
t	Time (s)
$t_{\rm cool}$	Cooling time γ -spectrometric measurement (s)
$t_{\rm cool}^{\rm etal}$	Cooling time of etalon (s)
$t_{ m irr}$	Irradiation time (s)
t_i^{act}	Irradiation (activation) time of i -th irradiation interval (s)
t_i^{e}	Time between end of i -th irradiation interval and end of experiment (s)
$t_{ m live}$	Live time of γ -spectrometric measurement with dead-time correction (s)
$T_{\rm n}$	Thermodynamic temperature (K)
$t_{\rm real}$	Real time of γ -spectrometric measurement (s)

v	Velocity $(m s^{-1})$
x	Effective thickness of material (mm)
$arepsilon_{\gamma}$	Absolute detection efficiency $(-)$
$arepsilon^{\gamma}_{\mathrm{FEP}}$	Absolute detection efficiency for full energy peak (FEP) $(-)$
$\eta_{ m B}$	Correction factor for beam fluctuation during irradiation $(-)$
η_{γ}	γ -ray self-shielding factor (-)
ϕ	Neutron flux $(cm^{-2}s^{-1})$
$\phi(E_{\rm n})$	Energy distribution of neutron flux $(cm^{-2}s^{-1}MeV^{-1})$
$\phi_{ m th}$	Thermal neutron flux $(cm^{-2}s^{-1})$
arphi	Neutron flux $(cm^{-2}s^{-1})$
$\varphi(E)$	Neutron spectrum ($\rm cm^{-2}s^{-1}MeV^{-1}$)
λ	Decay constant of radionuclide (s^{-1})
μ	Linear attenuation coefficient of γ -ray in material (cm ⁻¹)
$\mu_{ m p}$	Neutron fluence perturbation factor $(-)$
μ_{γ}	γ -ray self-shielding factor (-)
σ	Microscopic cross-section (cm^2)
$\sigma_{ m a}$	Microscopic activation cross-section (cm^2)
σ_0	Microscopic activation cross-section for 0.0253 eV neutrons $\rm (cm^2)$

List of acronyms

ADC	Analog to Digital Converter
ADTT	Accelerator-Driven Transmutation Technology
ARI	Applied Radiation and Isotopes
ASCR	Academy of Sciences of the Czech Republic
ATLAS	A Toroidal LHC Apparatus
AV ČR	The Academy of Sciences of the Czech Republic (Akademie věd České republiky)
BNCT	Boron Neutron Capture Therapy
CANAM	Center of Accelerators and Nuclear Analytical Methods
CAS	Czech Academy of Sciences (Akademie věd České republiky)
CE	Current Era
CEMS	Conversion Electron Mössbauer Spectrometry
CERN	European Organization for Nuclear Research
CMI	Czech Metrology Institute
CP-5	Chicago Pile-5
cps	Counts per seconds
CTU	Czech Technical University
CXMS	Conversion X-ray Mössbauer Spectrometry
ČVUT	Czech Technical University (České vysoké učení technické v Praze)
DEMO	Demonstration Reactor
DNA	Deoxyribonucleic acid
DOI	Digital Object Identifier
DONES	Demo Oriented NEutron Source
dpa	Displacements Per Atom
EAF	European Activation File
EDP Sciences	Édition Diffusion Presse Sciences
EERRI	Eastern European Research Reactor Initiative

ENDF	Evaluated Nuclear Data File
EPE	Electric Power Engineering
EPJ	European Physical Journal
EU	European Union
EXFOR	Experimental Nuclear Reaction Data
F4E	Fusion for Energy
FAFNIR	Facility for Fusion Neutron Irradiation Research
FEP	Full Energy Peak
FJFI	Faculty of Nuclear Sciences and Physical Engineering (Fakulta jaderná a fyzikálně inženýrská)
FNSPE	Faculty of Nuclear Sciences and Physical Engineering
$\mathrm{FRV}\check{\mathrm{S}}$	Higher Education Development Fund (Fond rozvoja vysokých škôl)
FWHM	Full Width at Half Maximum
HEP	High-energy physics
HPGe	High Purity Germanium
HPNS	High Power Neutron Source
HIFAR	High Flux Australian Reactor
HRCh	Horizontal radial channel
IAEA	International Atomic Energy Agency
ICRU	International Commision on Radiation Units and Measurements
IF	Impact Factor
IFMIF	International Fusion Material Irradiation Facility
INAA	Instrumental Neutron Activation Analysis
IS	Isomer Shift
ISBN	International Standard Book Number
ISSN	International Standard Serial Number
ITER	International Thermonuclear Experimental Reactor
JEFF	Joint Evaluated Fission and Fusion File
JRR	Japan Research Reactor
KISR	Kuwait Institute for Scientific Research
LA	Los Alamos
LHC	Large Hadron Collider
MCNP	Monte Carlo N-Particle transport code
MCNPX	Monte Carlo N-Particle eXtended transport code
MDA	Minimal Detectable Activity
MEYS	The Ministry of Education, Youth and Sports (of Czech Republic)

MERS	Ministry of Education, Science, Research and Sports (of Slovak Republic)
MIIS	Middlebury Institute of International Studies
MPI	Max Planck Institute in Munich
MS	Mössbauer Spectrometry
MŠMT	The Ministry of Education, Youth and Sports (of Czech Republic)
NAA	Neutron Activation Analysis
NAT	Neutron Activation Technique
nat	Natural
NISP	Number of Identified Specimens
NG-2	Neutron Generator no. 2
NPI	Nuclear Physics Institute
OP RDE	Operational Programme Research, Development and Education
p.r.i.	Public Research Institution
PRT	Proton Recoil Telescope
QM	Quasi-monoenergetic
QS	Quadrupole Splitting
RNA	Ribonucleic acid
RNAA	Radiochemical Neutron Activation Analysis
RPC	Radiation Physics and Chemistry
RPD	Radiation Protection and Dosimetry
RT	Room Temperature
R & D	Research and Development
SAND	Spectrum Analysis by Neutron Detectors
SARA	Safe Application of Radiation and Radionuclides
TOF	Time-of-Flight
TRIGA	Training, Research, Isotopes, General Atomics
ÚJF	Nuclear Physics Institute (Ústav jaderné fyziky)
UK	United Kingdom
USA	United States of America
WoS	Web of Science

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Introduction

Research nuclear reactors and compact accelerator-driven neutron sources are important facilities necessary, besides other things, for the isotope production, radiotherapy, nuclear data provisioning, nuclear analytical techniques, and material research. In addition to still current topics of measurement, validation, and refinement of nuclear data important for the contemporary fission and future fusion energetics, the importance of the interdisciplinary research increases nowadays as well. In this regard, the research nuclear reactors together with the accelerator-based neutron sources can be used for study of rare objects (such as historical, archaeological, geological, astronomical, medicinal samples, etc.), and they can provide information useful for scientists from other research branches. The interdisciplinary research and the study of the neutron fields of less explored neutron producing reactions at the accelerator-based sources appears to be of great interest.

In the dissertation thesis [1], the author dealt with the determination of broad neutron spectra generated by the $p(37) + D_2O$ source reaction used in the heavy-water target station with flowing target and by the p(35) + Be source reaction used in the original beryllium target station, that were marked as the neutron generator NG-2 and operated at the Nuclear Physics Institute of the CAS. Later, the heavy-water target station ceased to be used for irradiation experiments with intensive neutron fields due to its technically complicated operation and because of potential risk of accident, and it was replaced by the beryllium target station. However to meet the demands on the high intensity of the fast neutron yields at the irradiation positions close to the neutron target, that are dictated by the requirements on the experiments from the external scientists, the technical upgrade of the beryllium target station was subsequently performed. The technical modification consisted of reducing the amount of construction materials responsible for their parasitic effect on the neutron field. This modification influenced the source-to-sample geometry positions and enabled also to locate the investigated samples closer to the neutron target. Besides the necessity for performing new detailed measurement of the p(35) + Be neutron field for the upgraded version of the beryllium target station, the needs for determination of new neutron fields based on the d + Be source reactions for 20 MeV and 15 MeV deuteron beams as well as the p + Be source reaction for 24 MeV proton beam by means of the neutron activation technique appeared shortly.

In addition to these experimental activities, the author dealt intensively with the using of the neutron activation analysis at the VR-1 training reactor operated by the Czech Technical University in Prague. The focus was primarily put on the broadening of the applications of the neutron activation analysis at the VR-1 training reactor together with the aim to demonstrate its utilization for the purposes of the interdisciplinary research, such as in the humanities and social sciences. Several kinds of interesting and rare samples were investigated; among other things, the tradition Tibetan medicine, historical Tibetan coins, European dietary supplements, meteorites and tektites, and mammoth remains were analyzed at the VR-1 reactor in recent years. These new experiments focused on composition study of historical coins, dietary and medicinal pills, palaeontological and astronomical samples at the VR-1 training reactor as well as the neutron fields of the p + Be and d + Be source reactions at the accelerator-driven NG-2 neutron source are included in the habilitation thesis of the author.

The submitted habilitation thesis is compiled in the form of commented set of twelve leadauthored publications that are indexed in the international databases Web of Science or Scopus. The commented set of publications includes eleven scientific papers that were published in international journals with impact factor. Eleven papers were successively created in the period after the defense of the author's Ph.D. dissertation [1]. The last paper included in the commented set of papers was produced during the author's Ph.D. studies at the FNSPE CTU in Prague, however it was not related to the topic of the Ph.D. thesis and was not part of it.

The habilitation thesis is divided into five chapters and twelve annexes in the form of individual scientific papers that are an integral part of the thesis. In the first chapter, the goals of the habilitation thesis are stated. On account of the fact that this thesis is written in the form of commented set of papers, the brief overview of the current state of the art described in individual papers is summarized in the second chapter. Because the topic is focused on the experimental research activities, the used experimental facilities such as the accelerator-driven NG-2 neutron source and VR-1 training reactor are presented in chapter three, and both of them are described with regard to their utilization for the purposes of the neutron activation technique. The principles of neutron activation technique are described in the fourth chapter. More details on current state of the art and methodology are provided in particular publications included in this commented set of papers. The fifth chapter together with all twelve annexes represent the core of the habilitation thesis, and they characterize the individual experiments, achieved results, and possibilities of their usage and benefits. Consequently, the main results of the thesis and their emphasized benefits and originality are summarized in the conclusions. The thesis included also the lifelong list of publications that contains the lead-authored as well as co-authored papers, and the list of all presentations of the author at the international and national conferences.

Chapter 1

Habilitation thesis objectives

Habilitation thesis is focused on the study of neutron fields generated by external NG-2 neutron source operated at the Nuclear Physics Institute (NPI) of the CAS in Řež and on applications of the radioanalytical technique, called the neutron activation analysis (NAA), at the VR-1 training reactor operated by the Faculty of Nuclear Sciences and Physical Engineering (FNSPE) of the CTU in Prague. The NG-2 source uses the charged particle beams delivered from the U-120M cyclotron which represents a unique experimental facility necessary for the research tasks of nuclear data measurement and validation within the fusion related IFMIF-DONES research program and radiation hardness tests of electronics and materials. The method of NAA at the VR-1 reactor shows the great potential of this nuclear facility as a supporting tool for investigation of objects in humanistic oriented research fields within the interdisciplinary approach. Interdisciplinary research as well as the task of neutron spectra measurement and refinement are highly topical. In this context, the objectives of the habilitation thesis are divided to:

- 1) Detailed study of the neutron field of the p(35) + Be reaction of modified NG-2 source at the NPI CAS and determination of the broad neutron spectra using the activation technique with the inclusion of the space integration effect of neutron yield in geometry arrangement where the source-to-sample distances and dimensions of samples and source are comparable.
- 2) Determination of new broad neutron energy spectra of the d(20) + Be, d(15) + Be and p(24) + Be source reactions of the NG-2 source in close distances from the neutron target by means of the multi-foil activation technique that were not earlier studied at the NPI CAS in Řež for the d + Be reaction at all and for the p + Be reaction with 24 MeV proton beam.
- 3) Utilization of the non-destructive neutron activation analysis at the VR-1 reactor within the interdisciplinary approach and study of historical, palaeontological, pharmaceutical samples, meteorites, tektites, and dietary supplements for determination of their composition.
- 4) Investigation of the neutron field parameters in the irradiation channel of the VR-1 training reactor utilizing the activation technique for purposes of the neutron activation analysis.

Chapter 2

State of the art

The habilitation thesis is focused on the topic that can be divided into two directions. On the one hand, it is the state of nuclear data, primarily the neutron fields and neutron yields and related research programs. On the other hand, it is the utilization of neutron activation analysis for investigation of valuable items at research reactors within the increasing collaboration with humanities and social sciences in the form of providing of nonconventional research tools for their research fields.

2.1 Source reactions for production of neutron fields

The interactions of charged particle beams with suitable target materials represent important sources of neutron fields and neutron beams necessary for wide spectrum of applications in neutron physics. Based on type of neutron field, they are used for irradiation experiments in tests of radiation hardness of electronics for dosimetry monitoring systems, in airplane industry and astronautic, for material research, in medicinal applications for irradiation of tumors, and for production of radiopharmaceuticals. They achieve the special attention from the point of view of an important tool for nuclear data measurement and validation as well. Neutron spectra of selected neutron sources serve as the reference neutron spectra in nuclear databases. The need for nuclear data measurement and obtaining of more accurate data is still current topic.

The type and energy of charged particle beam and type and thickness of target material dictate the characteristics of produced neutron field, i.e. intensity, shape, and energy range of neutron field. Based on the thickness of target material, the neutron field has quasi-monoenergetic (QM) spectrum or broad neutron spectrum. Thin targets with their quasi-monoenergetic neutron spectra are typically used for measurement of microscopic cross-sections and in applications that require only one particular energy or narrow energy range of neutrons. Thick targets with broad neutron spectra are important for intensive irradiation experiments and experimental tasks of integral validation of nuclear data.

Today, the accelerator-driven fast neutron sources all over the world primarily use beryllium and lithium as the target materials that are usually bombarded by proton or deuteron beams. As quasi-monoenergetic neutron sources, the p + Li and p + Be interactions at thin targets are used. The d + Be and p + Be source reaction at thick targets are primarily utilized in sources of neutron fields with broad spectra. The reaction energy Q dictates the position of monoenergetic neutron peak in the QM-spectrum or upper energy limit in broad neutron spectrum.

Author	Country	$E_{\rm p}~({\rm MeV})$	Target thickness	Method	Year
J.S. Levin [5]	USA	2.3-5.4	_	TOF	1959
R.J. Slobodrian [6]	USA	20.0	_	PRT	1967
J.D. Anderson [7]	USA	20.0	$10~{ m g/cm}^2$	PRT	1969
S.W. Johnsen [8]	USA	25 - 55	46 mm	NE-213	1976
M.A. Lone [9]	Canada	14.8 - 23.0	$0.82~{\rm g/cm}^2$	TOF	1977
S.W. Johnsen [10]	USA	30; 40	$0.82~{\rm g/cm^2}$	NE-213	1977
R. Madey [11]	USA	100.2	2.0 cm	TOF/PRT	1977
R.G. Graves [12]	USA	41	1.22 cm	TOF	1979
F.M. Waterman [13]	USA	35; 46	_	TOF	1979
J.L. Ullman [14]	USA	35	$1.16~\mathrm{cm}$	PRT	1981
R. Henneck [15]	Switzerland	55.0	$370~{\rm mg/cm}^2$	_	1988
Y. Uwamino [16]	Japan	40.0	$2 \mathrm{mm}$	NE-213	1988
H.J. Brede [17]	Germany	17.2 - 22.0	$5 \mathrm{mm}$	TOF	1989
D.T.L. Jones [18]	South Africa	66.0	$19.6 \mathrm{mm}$	NE-213	1992
W.B. Howard [19,20]	USA	3.0 – 5.0	$0.5 \mathrm{~mm}$	TOF	1997
A.A. Lychagin [21]	Russia	8.2 - 11.2	2.0 mm	Stilben	2003
S. Kamada [22]	Japan	$11.0;\ 70.0$	4.0 mm	NE-213	2011
S. Agosteo [23]	Italy	5.0	1.0 mm	PRT	2011
M. Osipenko [24]	Italy	62.0	3.0 cm	TOF	2013
J.W. Shin [25]	Korea	30; 35; 40	$1.05~\mathrm{cm}$	NAT	2015
A. Mattera [26]	Finland	30.0	$5.0 \mathrm{~mm}$	TOF	2017

Tab. 1 – Overview of studies of p + Be neutron fields [2,3,4]

From the point of view of topic solved in this habilitation thesis, the most interesting neutron producing reactions are the interactions of protons and deuterons with thick layers of beryllium. First of all, most of accelerator-based neutron sources are currently based on the beryllium targets and d + Be source reaction. The reaction energy of the main fast neutron producing interaction is about +4.36 MeV of the d + Be source and -1.85 MeV of p + Be source reaction [27]. The d + Be and p + Be source reactions, their neutron energy spectra and neutron yields, were intensively investigated by scientists from 1960s to 1980s. The measurements were mostly performed utilizing the scintillation probes and time-of-flight technique (TOF) at long distances from neutron source target in a point-like geometrical arrangement; however, there are some
discrepancies between results reported by various authors. The overview of studies of the d + Be and p + Be source reactions is provided in *Tab. 1* and *Tab. 2*. From experimentally determined neutron energy spectra, mainly the measurement carried out by M.A. Lone [9], the empirical formulas describing the fluence averaged neutron field energy in forward direction at an angle of 0° and valid above particular minimal energies of deuteron and proton beams were subsequently derived [9,2].

Author	Country	$E_{\rm d}~({\rm MeV})$	Target thickness	Method	Year
G.W. Schweimer [28]	Germany	54.0	_	NE-213	1967
J.P. Meulders [29]	Belgium	16.0 - 50.0	$1.85~{ m g/cm}^2$	NE-111	1975
M.J. Saltmarch [30]	USA	40.0	$6.3 \mathrm{mm}$	NE-213/NAA	1977
M.A. Lone [9]	Canada	14.8 - 23.0	$0.82~{\rm g/cm}^2$	TOF	1977
R. Madey [11]	USA	83.7	$2.0~\mathrm{cm}$	$\mathrm{TOF}/\mathrm{PRT}$	1977
R.G. Graves [12]	USA	49.0	$0.85~\mathrm{cm}$	TOF	1979
F.M. Waterman [13]	USA	16.0; 28.0	thick	NE-102	1979
D.L. Smith [3]	USA	7.0	$0.5 \mathrm{~mm}$	NE-102A	1985
H.J. Brede [17]	Germany	9.4 - 13.6	$5 \mathrm{mm}$	TOF	1989
J.V. Meadows [31]	USA	2.6 - 7.0	$0.75 \mathrm{~mm}$	TOF	1991
F. Maekawa [32]	Germany	19.0	_	NAA	1998
L. Oláh [33]	Hungary	9.72	3.0 mm	NAA	1998
N. Colonna [34]	USA	1.5	$0.8~{ m mg/cm}^2$	Scint. det.	1999
T. Aoki [35]	Japan	25.0	3.0 mm	TOF/NAT	2004
Y. Iwamoto [36]	Japan	10.0	$15 \ \mu m$	NE-213	2009
Y. Zuo [37]	China	0.2 - 3.0	1.0 mm	BF_3 counter	2014
J.J. Goodell [38]	USA	30.0	$2.54~\mathrm{cm}$	NAT	2018
S.J. Zhang [39]	China	0.25 - 0.30	1.0 mm	TOF	2021

Tab. 2 – Overview of studies of d + Be neutron fields [2,3,4]

2.2 Research programs focused on thermonuclear fusion

Due to the expected development of thermonuclear energetics, it is necessary to study the impact of intensive fast neutron fields with energy of 14.1 MeV on intended construction materials used in reactor vessels of future thermonuclear systems. Neutron field of planned international tokamak ITER, that represents the first stage for future fusion energetics and which commissioning with d-T reaction is expected in 2028 [40], will not reach the high values of neutron flux necessary for study of radiation and thermomechanics effects in construction steels of thermonuclear reactors. For that reason, the consequential project of pre-industrial demonstration thermonuclear reactor DEMO is the project aimed for final verification of conversion of thermonuclear energy to electricity. Besides the primary energy of fusion neutrons, that is 14 MeV, the energy range of 12 MeV to 20 MeV is important for the needs of ITER and DEMO

research programs. This energy range is in fusion research community called the region of fusion neutrons. An important supporting tool for both facilities should be the IFMIF project that should have preceded them.

Project of International Fusion Material Irradiation Facility (IFMIF) was already initiated in 1994 in the framework of cooperation between EU, USA, Russia, and Japan. It was aimed to build an advanced irradiation facility that would allow the completion of a new nuclear data library and testing the components of construction materials intended for use in future fusion reactors starting with the DEMO demonstration reactor. IFMIF was supposed to be a neutron source utilizing the d + Li source reaction at molten layer of lithium flowing at a velocity of 15 m s^{-1} whereas the deuteron beams with an energy of 40 MeV and an intensity of 125 mA were to be supplied by two accelerators [41]. In the first irradiation chamber close to the neutron target, the neutron field of order of $10^{14} \,\mathrm{cm}^{-2} \mathrm{s}^{-1}$ was to be available. The continuous neutron energy spectrum of the IFMIF was to have a mean energy of 14.1 MeV corresponding to a D +T fusion reaction with a main spectrum component up to 35 MeV and with a contribution of a high energy low intensity neutron component up to 55 MeV. The irradiation chambers were to be divided into three categories according to the achieved level of neutron intensity, and the workplace was also to be equipped with supporting hot chambers and laboratories. The IFMIF facility was intended to identify the suitable materials for fusion reactors, provide input data for the design of a DEMO fusion reactor, study gas-producing (n, p) and (n, α) reactions in construction materials, to assess the impact of radiation effects on these materials and to determine their safety limits, and to validate existing and supplement missing data for nuclear databases in the energy region of fusion applications [41,42]. The IFMIF project appeared to be costly, and it gradually delayed; according to the latest estimations, the installation is to be finally commissioned by 2028 [41], two years ahead of the expected start of the construction of the DEMO fusion demonstration reactor, which is extremely unsatisfactory.

The unfavorable development of the situation around the planned IFMIF facility led to design of an alternative concept of FAFNIR (Facility for Fusion Neutron Irradiation Research), which was based on existing technologies without the need to develop the high-performance charged particle accelerators and a complicated target system [43]. The FAFNIR concept was not intended as a replacement for the IFMIF facility, but it was intended to be an initial stage to the IFMIF facility and to provide a part of the experimental data and experimental background until the IFMIF was to be built. The FAFNIR was to use a 40 MeV deuteron beam with an intensity of 5 mA with the possibility of a later upgrade to 30 mA which was to bombard a rotating carbon target [44]. From the point of view of the material damage study (achieved dpa), the performance of the FAFNIR should have been approximately 12 times lower compared to the IFMIF facility [45,43]. In the end, the concept of the FAFNIR facility remained only on paper.

The IFMIF-DONES project (DEMO Oriented Neutron Source) is based on the original IFMIF concept, but it introduced several simplifications that should reduce the financial costs

of the entire project and reduce the time required for development of equipment, design, and construction of the facility as well. The IFMIF-DONES will use only one accelerator, and it will include only the irradiation modules necessary for structural tests of materials for fusion applications. The accelerator will provide a deuteron beam with energy of 40 MeV and intensity of 125 mA that will bombard a liquid lithium target flowing at a velocity of 15 m s^{-1} . The d(40) + Li source reaction will provide a neutron flux of $5 \times 10^{18} \text{ m}^{-2} \text{s}^{-1}$ at the sample irradiation position, the neutron spectrum will contain a wide peak at energy of 14 MeV [46]. The damage (dpa – displacements per atom) of materials irradiated close to neutron source is expected to be 15 dpa. For financial reasons, the supporting laboratories for the analysis of irradiated samples are not included in this project by default. However, the entire device will be designed so that it can be upgraded to the full specifications of the original IFMIF device in the future. The start of construction of the IFMIF-DONES facility is planned for 2025 [47,46].

The Nuclear Physics Institute of the CAS with its fast neutron sources NG-2 connected to the cyclotron U-120M is involved in research activities of measurement and validation of activation cross-sections relevant to the IFMIF and IFMIF-DONES research programs, mainly in terms of the energy range (i.e., up to 35 MeV) that corresponds to the main energy range of the IFMIF neutron source. Currently, the NPI is developing a powerful neutron source HPNS (High Power Neutron Source) with a thick beryllium target operated at the new TR-24 cyclotron and employing the p(24) + Be source reaction. This neutron source will use a 300 μ A proton beam with energy up to 24 MeV, and it will produce intensive neutron field with a broad spectrum up to 22 MeV. It covers the energy range important for thermonuclear fusion and ITER tokamak, i.e., the "fusion neutrons" with the range of 12 to 20 MeV, where the most of gas-producing (n, p) and (n, α) reactions in construction materials of fusion applications reach the maximum values of microscopic cross-sections.

2.3 Activation analysis by means of nuclear reactors

There are currently 223 research nuclear reactors in operation worldwide, of which 133 units are operated in developed countries and 90 units in developing countries, and 11 nuclear reactors are under construction. In fact, 78 nuclear reactors are utilizing for isotope production, 67 units are used for neutron radiography, 62 facilities are employed for material research, 14 research reactors are utilized for neutron therapy and last but not least 14 research reactors are used for nuclear data measurement and validation. Other important applications of nuclear research reactors include the training of students and operating staff for nuclear power plants, geochronology, silicon doping, gem coloring, etc. It is noteworthy that 111 research reactors are utilized worldwide for neutron activation analysis. Data were adopted from [48].

Activation analysis is a radioanalytical technique based on nuclear activation of elements presented in analyzed samples. It is one of the most sensitive methods of chemical analysis. Activation analysis was used for the first time for elemental analysis in 1936 by George de Hevesy and Hilde Levi, and neutrons were utilized as activating particles [49]. According to the type of used activating particles, activation analyses is divided to photon activation analysis, (prompt) activation analysis with charged particles, and neutron activation analysis. The latter is further subdivided into thermal, epithermal, and fast neutron activation analysis. Apart from research nuclear reactors, accelerator-driven fast neutron sources and 14 MeV neutron generators can be advantageously utilized for fast neutron activation analysis as well.

The most frequently used technique in international research laboratories is neutron activation analysis with thermal neutrons delivered by research nuclear reactors. The method is characterized by high sensitivity, and analysis can be performed in nondestructive way as the instrumental neutron activation analysis or in destructive way with radiochemical separation. The most commonly used techniques of NAA are k_0 -standardization method and comparative (relative) method. The absolute NAA, which requires precise knowledge of neutron spectrum in irradiation position and excitation functions, is used rarely.

The practical utilization of neutron activation analysis is rather broad, as this experimental method is widely used by various scientific and industrial branches, life sciences, humanities, and social sciences. Applications of neutron activation analysis include:

- Analysis of archaeological and paleontological samples (e.g., elemental compositions of archaeological artefacts, ancient and medieval coins, elemental mass ratio in fossil bones and uranium uptake, provenance research in archaeology, [50,51,52,53,54]),
- Study of cultural heritage and analysis of historical samples (e.g., authentication studies, analysis of pottery objects to establish their provenance, provenance of artistic objects and state of conservation, composition of historical coins, [55,56,57,58,59,60]),
- Analysis in geology and geochemistry (e.g., rare-Earth elements in geological materials, study of cosmochemical samples, [61,62,63,64]),
- In Vivo analysis and health projects (e.g., nutritional projects, therapy evaluation, clinical diagnostic, bone analysis, calcium-to-phosphorus ratio in rib bone, [65,66,67,68]),
- Analysis of biological samples (e.g., measurement of toxic elements (Hg, As) in biological samples, elemental investigation of animal samples, hair samples, whole-body neutron activation analysis, [69,70,71,72,73,74]),
- Analysis in botany and agriculture (e.g., tea leaves, Arabica and Robusta coffee beans, kola-nits, medicinal plants, shea butter and nuts, agricultural crops, [75,76,77,78,79,80]),
- Analysis of environmental samples (e.g., essential trace elements and toxic elements in mosses, mushrooms, and other biological monitors, analysis of human hair for environmental purposes [81,82,83,84,85,86]),
- Analysis of water samples, coal, and coal effluents (e.g., trace elements in fly ash and effluent waters, toxic element concentration in mined coal samples, [87,88,89,90]),

- Analysis of air particulate matter (e.g., air suspended particles in city, ambient air dust particulates, particular matter from a biomonitoring, [91,92,93,94,95]),
- Analysis in forensic studies (e.g., toxicology and falsification, identification of evidence and trace elements determination, whether a person fired a gun based on gunpowder analysis, [96,97,98,99]),
- Analysis of semiconductor materials and industrial products (e.g., determination of impurities and trace elements in silicon, characterization of semiconductor materials, analysis of osmium in industrial products, [100,101,102,103,104]).

In last decades, the idea of interdisciplinary research has been still increasing all over the world, and many research laboratories were looking for interconnection with soft sciences, as it can be seen from some items in the list of various applications of NAA mentioned above. Faculty of Nuclear Sciences and Physical Engineering of the CTU in Prague thank to its Training Reactor VR-1 has great capabilities in nuclear analytical technique NAA and potential for collaboration on interdisciplinary research between hard sciences and soft sciences.

Chapter 3

Neutron source NG-2 and Training Reactor VR-1

The research described in the presented habilitation thesis is based on experimental measurements in specific research facilities providing intensive neutron fields. The two used neutron sources including a brief description of the applications of the neutron activation technique are therefore described below.

3.1 Accelerator-driven neutron source NG-2

Nuclear Physics Institute of the CAS, p.r.i., operates the isochronous cyclotron U-120M, which is one of the main experimental facilities of this research institution. The accelerator was put into operation in 1977, and since then it has been innovated several times. At present, the accelerator provides monoenergetic beams of protons, deuterons, and helium ³He and ⁴He nuclei in the positive ion mode of acceleration. In the negative ion mode of acceleration, the intensive proton and deuteron beams are extracted from the cyclotron. Parameters of charged particle beams delivered by the accelerator are summarized in *Tab. 3* and the body of the cyclotron together with ion beam tube are displayed in *Fig. 1* [105]. The cyclotron is primarily used for the production of radiopharmaceuticals, materials research, study of the reactions of charged particles with materials, measurement of excitation functions, and in combination with suitable target materials it is also used for neutron applications.

The energy range and high intensity of the charged particle beams were the motivation for the design and construction of external neutron sources connected to the U-120M cyclotron. Significant development of neutron sources occurred mainly after the year 2000. Target stations were developed for the purposes of production of continuous and quasi-monoenergetic (QM) neutron spectra, and they were focused first on the ADTT (Accelerator Driven Transmutation Technology) research program and later mainly on the IFMIF fusion program [106,107]. At present, a target station with a thin lithium target and with the p + Li(C) source reaction is used as a source of quasi-monoenergetic neutrons in energy range of 18 to 33 MeV. A target station with a thick beryllium target utilizing the p + Be source reaction is a neutron source with a broad spectrum. Both these target stations connected to the cyclotron system are referred to as the NG-2 neutron source.

Ion mode of acceleration	Accelerated ions	Energy range (MeV)	Maximum current (µA)
Positive	H^+	6-25	5
	D^+	12-20	5
	$^{3}\mathrm{He}^{\mathrm{+II}}$	18 - 52	2
	${}^{4}\mathrm{He}^{\mathrm{+II}}$	24-38	5
Negative	H^{-}	6 - 35	30
	D^-	11 - 20	20

Tab. 3 – Parameters of charged particle beams extracted from U-120M cyclotron [108]



Fig. 1 – Body of U-120M isochronous cyclotron at NPI in Řež

Fig. 2 – Beryllium target station of NG-2 accelerator-driven neutron source

The beryllium target station (see Fig. 2) is used as a neutron source with adjustable maximum energy and a broad spectrum of fast neutrons. In the standard mode, it is operated as a source with the neutron field of the maximum possible energy range in the conditions at the NPI, i.e. with a spectrum up to 33 MeV and with the p(35) + Be source reaction. The target station is equipped with the thick beryllium target¹ with a thickness of 8 mm and diameter of 50 mm, that is actively cooled on the backside by a 4 mm layer of flowing alcohol. The neutron field from the proton-induced reactions on the thick beryllium layer has a forward-oriented direction and the fast neutrons spectral yield is $10^{11} \text{ cm}^{-2} \text{s}^{-1}$. The neutron source is primarily

¹ Owing to parameters of charged particle beams delivered by a cyclotron, it is energetic thick target.

used in experimental tasks of integral validation of nuclear data, in tests of destruction and radiation hardness of electronics, in materials research, and fast neutron activation analysis experiments. During operation, the technological parameters of the neutron source are monitored online, namely the intensity of charged particle beam on the target and collimators, inlet and outlet temperatures and pressure of cooling alcohol in target station head, its flow rate, flow rate of cooling water on collimators, and vacuum in the target station chamber.

3.2 Use of neutron activation technique at NG-2 neutron source

In addition to the time-of-flight (TOF) spectrometry, the neutron activation technique is one of the main experimental methods used in laboratories with the accelerator-driven neutron sources, and it has its indisputable importance for irradiation in close source-to-sample geometries. Other commonly available experimental techniques are practically unusable if the irradiation system has a non-point-like geometrical arrangement where the source-to-sample distance² and the dimensions of neutron source and irradiated samples are comparable.

The activation technique utilizing a set of dosimetry foils was used at the workplace of the NG-2 neutron source for the first time to determine the white neutron spectrum of a heavy water target station with the $p(37) + D_2O$ source reaction [109,110] and later for the neutron spectrum determination of the original version of beryllium target station with the p(35) + Besource reaction [111,112]. After the technological upgrade of the beryllium target station, which happened after the decommissioning of the heavy-water target station, the neutron field of the p(35) + Be reaction was preferentially measured in detail [113,114,115], because it is primarily used neutron field in most experimental tasks of nuclear-data validation and destruction tests of materials and electronics at the NPI. The neutron field of the p(35) + Be source reaction is used in irradiation experiments of photodiodes [116,117] and silicon photomultipliers [118,119], radiation hardness tests of microelectronics developed by MPI (Max Planck Institute) research group for ATLAS-CERN experiments [120] and radiation hardness tests of semiconductor detectors for experiments HEP-CERN [121,122] and ceramics, tests in the development of new detection systems [123], integral validation of neutron cross-sections in the energy range up to 33 MeV [124,125,126], and it was also used for measurements of gamma-ray intensities of ^{196m2}Au [127]. In addition to the p(35) + Be source reaction, the neutron energy spectra of the d + Be source reaction for 15 MeV and 20 MeV deuteron beams [128,129] and neutron spectra of the p(24) + Be source reaction [130] were also studied with respect to the energy range of interest of the ITER and DEMO programs and for the needs of selected applications of fast neutron activation analysis.

Besides the applications for neutron field spectrometry, the activation method is used at the NG-2 neutron source also for the tasks of activation cross-sections measurement in the energy range of 18 to 33 MeV using the quasi-monoenergetic neutrons from the p + Li(C) source. As

² Complications with a large load of measuring devices.

examples can be mentioned the measurement of activation cross-sections on cobalt [131], tantalum and niobium [132,133], chromium [134], gold, bismuth and thulium [126,124], yttrium [135,136], copper and vanadium [125]. Utilizing the activation method, an integral validation of EUROFER steel components using the broad neutron spectrum of the neutron source $p(37) + D_2O$ was performed as well [137,138].

3.3 Training reactor VR-1

The VR-1 training reactor is a pool-type research nuclear reactor operated by the FNSPE CTU in Prague. The first criticality state of the reactor was reached in December 1990, and since January 1992, the reactor has been in permanent operation. It is a low-power reactor with power of 80 W. It utilizes the tube-type nuclear fuel elements (IRT-4M fuel) with enrichment slightly below 20 %, obtained from Russia. Reactor uses light demineralized water as a neutron moderator, biological shielding, and coolant using the natural convection. The main part of the reactor is represented by reactor core that is composed by three types of the IRT-4M fuel assemblies, located at the bottom of the reactor vessel filled up by 17 m^3 of demineralized water. The pool-type arrangement of the reactor with free water level enables easy access to the reactor core if some manipulations are necessary or if some equipment need to be inserted into reactor core. There are also several dry vertical experimental channels of various diameters (25, 32, 56, and 90 mm) that make it possible to insert the detectors or investigated samples to selected irradiation positions in the reactor core. Other experimental equipment includes the horizontal channels and rabbit system enabling the transport of samples between the reactor core and gamma-spectrometry laboratory with an HPGe detector. The VR-1 reactor workplace includes also adjacent laboratories with gamma-ray spectrometers, neutron physics laboratory, and laboratory of physical protection. Reactor hall and reactor core are shown in Fig. 3 and Fig. 4.



Fig. 3 – Reactor hall of VR-1 reactor

Fig. 4 – Reactor core of VR-1 reactor

The VR-1 reactor is primarily intended to train the students from the FNSPE CTU and other faculties with nuclear engineering related branches of study both in the Czech Republic and abroad. The staff of nuclear power plants is the regular user of training reactor as well. The teaching program at the VR-1 reactor comprises primarily the experimental tasks on neutron detection, neutron flux distribution measurement, delayed neutron measurement, reactivity measurement and control rod calibration, reactor dynamics study, and criticality experiment. For past several years, the training program of several courses includes also the tasks of neutron activation analysis.

Besides the teaching of students, the reactor is also intensively used for experimental purposes. As the examples of research utilization, the pilot application of neutron radiography, material research by means of Mössbauer spectroscopy, radiation hardness tests of electronics, and mainly the neutron activation analysis can be mentioned.

3.4 Utilization of activation analysis at the VR-1 training reactor

Since the reactor commissioning, the neutron activation analysis was sporadically used at the VR-1 reactor in education of students and for research purposes. The increasing emphasis has been gradually placed on this experimental technique since 2010. The author of the submitted habilitation thesis has been utilizing the neutron activation technique for neutron field parameters measurement at selected irradiation positions in the VR-1 reactor core and latter was primarily focused on investigation of samples from every-day life as a content of his research project during his doctoral study. The experiments aimed at determination of aluminium and copper content in contemporary Central European coins and in several foreign coins (see *Fig. 5*) have shown the great potential of this experimental method and the nuclear reactor itself, as a supporting radioanalytical tool, in unconventional approach to study samples attractive for hard sciences as well as humanities and social sciences. Since then, the NAA has been more intensively used for teaching of domestic and foreign students and in bachelor's and master's thesis projects of students at the Department of nuclear reactors of the CTU.



Fig. 5 – Holder with activation foils and coins from pilot NAA experiments

Neutron activation analysis was incorporated into the training of several courses for foreigners, such as KISR training course since 2012, EERRI course since 2013, SARA course since 2013, and MISS course since 2017. Beside the qualitative analysis of investigated objects, the concentrations of copper, nickel, and zinc in various coins, amount of zinc in dietary supplements, and amount of uranium in uraninite are determined. In parallel, the qualitative analysis of Moldavite was demonstrated to students too.

Within the research activities, the dietary mineral supplements containing zinc [139], historical Tibetan coins containing silver and copper [140], traditional Tibetan medicinal pills [141], iron-nickel meteorites and tektites [142], and remains of woolly mammoth [143] have been studied at the VR-1 reactor recently.

Chapter 4

Neutron activation technique

The interactions of neutrons with matter are successfully used by a wide range of applications including the detection systems of some experimental techniques, material science, dosimetry, radioanalytical methods as well as the medical applications with tumors exposure to neutrons, etc. The neutron interactions are essential for detection and spectrometry in several online and offline experimental techniques that are used at the nuclear reactors and workplaces with the accelerator-based neutron sources. They are utilized together with the gas-filed detectors for neutron detection, thermoluminescence dosimetry, scintillation and Bonner spectrometry, and activation measurements. And just the activation technique is based on the nuclear reactions induced by neutrons.

4.1 **Principles of activation measurements**

Neutron activation method uses a fact, that in the neutron interactions with atomic nuclei, the new radionuclides are produced. These radionuclides are usually unstable, and their decay is accompanied by a gamma-ray emission that is typical for particular produced nucleus. The emitted ray is measured by a suitable detector. The gamma-ray identification is matter of the gamma-ray spectrometry that is an integral part of the activation measurements.

Based on the specifications of the neutron source, various kinds of nuclear reactions can be used for activation measurements, such as the non-threshold radiative capture reaction (n, γ) , charged particle producing reactions (n, α) and (n, p), and inelastic scattering reaction (n, n')or neutron producing reactions (n, xn) that depend on the upper energy range limit of neutron field and particular material. The energy range of neutron spectrum, its intensity, properties of target nucleus given by an activation cross-section and isotopic abundance in a mixture, properties of produced radionuclides such as the half-life and energy and intensity of emitted γ -ray are the most important factors that influence the measurability of individual radionuclides in investigated sample. Moreover, the gamma-ray self-shielding effects in sample as well as the influence of radioactive products generated from other reaction channels at the same nuclide or presence of another major or minor elements in sample together with their activation products are not always negligible.

In principle, the procedure of neutron activation experiments consists of several steps. Based on type of analysed object, the object is encapsulated before the irradiation experiments in order to prevent its damage during manipulation or contamination of the irradiation position by the abrasive materials, or sample with smaller dimension is prepared from the original object if needed. In the next step, the prepared sample is inserted into the irradiation position where it is exposed to the neutron field for requested time period. After irradiation, the sample is unloaded from the irradiation position, and it is transported to the gamma-spectrometric laboratory usually equipped with the HPGe detector. Then the phase of gamma-spectrometric measurements is started. Finally, the measured gamma-ray spectra are analysed, and based on observed gamma-lines, the radionuclides produced in the activation reactions are identified. From the measured data, the relevant quantities such as the reaction rate $R_{\rm R}$, production rate P, or saturated activity $A_{\rm sat}$ are extracted. Obtained quantity is subsequently utilized based the on character of relevant irradiation experiment.

In practise, the activation technique can be used in several ways. It can be applied as a spectrometric technique for measurement of the neutron fluence distribution (neutron spectrum) or neutron field parameters (reaction rates or spectral indexes) at the selected positions of the neutron field. Other useful application of activation technique is the radioanalytical method called the neutron activation analysis that is used for investigation of unknown samples. The activation method can by also used as an experimental tool for measurement of the activation cross-sections $\sigma_{\rm a}$ of nuclear reactions or determination of the fission yields $N_{\rm v}$.

4.2 Neutron field spectrometry using the activation technique

Several experimental techniques can be used for the tasks of neutron field spectrometry at the accelerator-driven fast neutron sources or research nuclear reactors. Primarily, the organic scintillation probes, Bonner-sphere spectrometry, time-of-flight technique, proton-recoiled spectrometry, or activation technique can be applied for the neutron spectrum determination.

The activation measurement of neutron fields is the primary spectrometric method intended for geometrical arrangement where the dimensions of irradiated samples and neutron source target and the source-to-sample distance are comparable. Under these conditions, the above mentioned spectrometric methods, except the neutron activation technique, are not satisfactory usable due to the non-point-like geometrical arrangement of investigated system.³ The activation technique uses a set of spectroscopic thin activation foils, called the activation detectors, and they finds their important applications for determination of neutron energy spectra of

³ Another complication resulting from the close source-to-detector arrangement is the overloading of the measuring apparatus in terms of the number of registered events (dead time) due to the more intensive neutron flux.

intensive neutron fields nearby the neutron sources based on charged particle accelerators or in space constrained conditions in nuclear reactors. For this reason, the neutron activation technique was chosen for neutron spectra determination of the NG-2 neutron source operated at the NPI of the CAS in Řež near Prague.

If the neutron activation technique is utilized for neutron spectrum measurement, the responses on neutron field in given irradiation position close to neutron target are measured using the appropriate activation foils. A set of dosimetry foils usually consists of several activation materials that are sensitive to certain energy region of neutron field according to particular activation cross-sections. The responses to neutron field in investigated source-to-sample distance are presented by the above-mentioned reaction rates related to one target nucleus. From the set of experimentally determined reaction rates, neutron cross-sections of corresponding nuclear reactions, and *a priori* information on neutron spectrum (so called guess neutron spectrum) obtained usually from a simulation of irradiation system, the neutron energy spectrum is reconstructed utilizing a suitable unfolding code. For neutron spectra reconstruction of the NG-2 fast neutron source, a modified version of SAND-II [144] unfolding code with extended support for experimental data up to 60 MeV was used, and the necessary predictions of neutron spectra were obtained using the Monte Carlo MCNPX transport code [145].

The utilization of Westcott formalism [146] for measurement of integral value of thermal neutron flux in well moderated environment of thermal nuclear reactors represents other useful application of activation method. In this method, the reaction rates related to thermal neutron field using appropriate activation material sensitive to thermal neutrons and corresponding activation cross-section are used, and correction for deviation from 1/v law is considered.

4.3 Neutron activation analysis

In contrary to activation measurement of neutron spectrum when the response to unknown neutron field is measured by a set of samples (activation foils) with exactly known mass and composition, the neutron activation analysis serves for determination of composition of unknown investigated object that was irradiated in suitable neutron field.

Neutron activation analysis is a sensitive radioanalytical method that makes it possible to determine qualitative as well as quantitative composition of unknown samples using the neutron activation technique. In general, neutron activation analysis can be carried out in a non-destructive or destructive way. If a destructive non-instrumental neutron activation analysis is applied, the investigated sample is dissolved by appropriate acids or alkalis, and the investigated component is selectively separated from the disruptive elements using radiochemical separation methods. Separated component is subsequently investigated by the gamma-ray spectrometry technique. By contrast, the instrumental neutron activation analysis does not use radiochemical separation processes, and the irradiated sample is directly measured at the HPGe detector. The advantage of instrumental activation analysis is the fact that the investigation of the sample is performed without the sample consumption or its damage, and therefore it is very suitable for study of historical objects and other valuable samples. Under certain circumstances, the sensitivity of the instrumental NAA can be influenced by the presence of an interfering component which may affect the gamma-spectrometric measurements⁴.

The activation analysis can be further divided into a comparative and absolute method. In the case of a comparative method, the saturated activity or production rate of investigated sample is compared with the activity or production rate of comparative standard that was irradiated together with studied sample under the same irradiation conditions, however the precise knowledge of neutron energy spectrum at position of irradiated samples is not required. Using the activation etalon with exactly known mass and composition, it is possible to determine the mass of particular element in analysed sample by means of instrumental NAA. In absolute NAA, the comparative standard is not used, and the mass of studied nuclide is determined based on gamma-spectrometrically determined production rate, exact knowledge of the neutron spectrum at the position of the irradiated sample and the corresponding activation cross-section. In the practical utilization of NAA, the comparative method is preferred.

At the training reactor VR-1, the comparative method of NAA is used for study of any kinds of samples such as samples of historical coins, dietary supplements and pharmaceutical pills, meteorites, organic, biological, archaeological, or food samples.

4.4 Supporting tools

The neutron activation technique is closely connected with nuclear gamma-ray spectrometry using the HPGe semiconductor detector made of high-purity germanium. The main role of gamma-spectrometry is the measurement of energy and intensity of gamma-ray emitted by radionuclides produced in samples by activation reactions during irradiation in neutron field. The radionuclides are identified according to energy and intensity of gamma-ray and with respect to half-life period. Other supporting tool necessary for neutron field spectrometry by activation technique is the unfolding code, e.g., SAND-II [144], that enables the reconstruction of neutron spectrum based on measured reaction rates and using the relevant activation crosssections. The nuclear databases represent another important tool for neutron field spectrometry of the NG-2 neutron source. Taking into consideration the energy range of the NG-2 neutron source (energy region above 20 MeV), primarily the EAF-2010 [147], LA-150 [148], and ENDF/B-VII.1 [149] nuclear libraries are available, and they are essential for deconvolution process of the SAND-II unfolding code and Monte Carlo MCNPX [145,150] transport code, that is used for detailed simulations of irradiation systems and for obtaining the preliminary information on irradiation conditions and expected neutron spectra necessary as the input guess neutron spectra for the SAND-II deconvolution process.

⁴ The interfering component may e.g. increase the induced activity of the sample and thus affect the dead time of the HPGe detector or the Compton scattering background of the gamma-ray spectrum.

Chapter 5

Activation experiments using NG-2 source and VR-1 reactor

The presented habilitation thesis is focused on two main approaches in the application of neutron activation technique, and it discusses their practical use at the NG-2 accelerator-driven fast neutron source with a beryllium target station operated by the Nuclear Physics Institute of the CAS in Řež and at the VR-1 training reactor operated by the Faculty of Nuclear Sciences and Physical Engineering of the CTU in Prague.

In the case of the NG-2 neutron source, the issue is the study of neutron fields, with intensive components of fast neutron spectra, generated by the p + Be and d + Be source reactions that are required by the scientific community, e.g. for integral validations of activation cross-sections for nuclear data libraries; they also represent a powerful tool for the study of radiation hardness of materials and electronic components used e.g. in research facilities or in the aerospace industry. The importance of the application of the activation technique at the NG-2 neutron source is also given by the fact that new more accurate results on spectral data for energy regions with the lack of experimental measurements are obtained; moreover in the case of the d(20) + Be, d(15) + Be, and p(24) + Be source reactions, the neutron energy spectra are determined for the first time ever at this experimental facility. In the same way, a detailed study of the neutron field of the p(35) + Be source reaction after a technological hardware upgrade of the Be-target station aimed at reducing the amount of construction materials has its own important reason regarding the above mentioned facts.

On the other hand, the use of activation technique at the VR-1 training reactor clearly shows the wide possibilities of utilizing the nuclear reactor as a neutron source for analytical applications that can provide useful data or results important for other scientific branches within an interdisciplinary approach for solving specific problematics. In particular, it points to the utilization of neutron fields for the study of historical objects of cultural heritage and palaeontological samples as well as possible applications in the pharmaceutical and food industry or in geology. As part of the habilitation thesis, historical coins from the Tibetan region are studied in order to show historians how the composition of the investigated objects can be determined using radioanalytical methods, and that can be used e.g. to identify the site or as the economic criteria in particular time period. The study of the dietary supplements composition can be used for instance to assess the content of the active substance or additives in the pills and thus to validate their composition. Investigation of mammoth remains can provide data utilizable e.g. for interpretation of migration of individuals or level of sample taphonomy. Similarly based on results of the NAA experiments and obtained sample composition, the meteorites can also be classified and their authenticity can be verified. Moreover, the neutron field parameters of the VR-1 training reactor are studied in the thesis in a similar way as in the case of the NG-2 neutron field determination at the accelerator-driven neutron source.

Because the NG-2 accelerator-driven neutron source at the NPI of the CAS as well as the VR-1 training reactor at the FNSPE of the CTU are both the unique experimental facilities with the specific working procedures during their operation, the experimental procedures and activities are briefly described in the following subchapters.

5.1 Execution of irradiation experiments at NG-2 neutron source

Irradiation experiments utilizing the NG-2 neutron source connected to the U-120M isochronous cyclotron at the Nuclear Physics Institute of the CAS are time and procedurally demanding activities that are given by the nature of the experiments themselves, used equipment, and operational capabilities of the U-120M cyclotron. Apart from the applied research, the cyclotron is intensively used for the production of radiopharmaceuticals for hospitals that are produced several times a week. Therefore, the maximum operating time of cyclotron available for neutron experiments is usually limited to 48 hours⁵. The approval of the experiments is subject to the head of Department of accelerators NPI and cyclotron manager. The cyclotron committee, which meets twice a year, decides on the allocation of irradiation time for all users.

Approved experiments with the NG-2 neutron sources consist of several steps, namely the preparation of the production target for the target station, setting up a charged particle beam, recording the beam axis, installation of the neutron source on the cyclotron beam-line, the irradiation experiment itself, and finally a post-experimental phase involving the neutron source uninstallation and possible manipulations with the irradiated production target.

In the preparatory stage, the inspection and cleaning of the beryllium target in the NG-2 neutron source are performed, if it is an experiment with a continuous neutron spectrum produced in beryllium target station. Or the installation of a new thin lithium target in the lithium

⁵ Some exceptions are possible under certain circumstances for experiments that are pre-negotiated in advance.

station is performed, if it is an experiment with quasi-monoenergetic neutrons. In the following stage, the technicians check the vacuum and insulation resistances of the used target station.

In the next step, the cyclotron operators set the desired beam of charged particles, either proton or deuteron beam, and then they measure the energy of the beam extracted from the acceleration chamber of the cyclotron. For irradiation experiments with neutron sources, a negative ion mode of acceleration is used, and the prepared charged particle beam is extracted from the cyclotron through a carbon stripping foil.

Using the special foils with a willemite layer, screen, and cameras, two points are obtained on the axis of the charged particle beam. After shutting-down the beam, a laser sight is adjusted to these two points, and thus the geometric axis of the beam is obtained. Subsequently, a neutron source target station is installed to the end of the beam-line, and it is adjusted to the laser beam so that the geometric axis of the beam passes through the center of the target in the target station. After reaching the required value of vacuum in the beam-line and inside target station chamber, the target chamber is connected to the cyclotron beam-tube. At the same time, the control and measuring units of vacuum, high voltage, beam-currents on the target and carbon collimators of the station as well as the cooling circuits of water and alcohol are connected to the target station. To verify the functionality and accuracy of the neutron source setting, a cyclotron is turned on at the end of the installation phase, and the charged particle beam travels through a set of collimators are measured, and their ratios are used for assessing the setting accuracy of the neutron source axis.

Subsequently, the main part of the experiment is started. It consists of irradiation stages with fast neutrons, while the operating parameters of the neutron source are monitored. When the experiment is finished and after a certain time interval depending on the radiation situation in the cyclotron hall, the NG-2 neutron source is uninstalled and transported from the cyclotron hall to the target preparation room. If a lithium target station was used for the experiment, the lithium target is removed from the target chamber in an argon protective atmosphere, it is inserted in a bottle with kerosene, and then it is placed behind a lead shielding for later gammaspectrometric measurements. In the case of irradiation experiment with use of the beryllium target station, the Be-target is not manipulated after the experiment.

In the case of realization of an activation experiment, the activation foils are also prepared during the preparatory phase of the experiment, they are weighed by analytical scale, and after marking of each foil, the corresponding foil kits are enclosed in thin plastic cases. Furthermore, a laboratory with the HPGe detectors is prepared as well.

5.2 Execution of activation experiments at VR-1 training reactor

The VR-1 training reactor was built for the purpose of teaching students in fields of study focused on nuclear energetic, and therefore the most of the activities carried out at the reactor,

especially in education, are related to neutron detection, reactivity measurements, delayed neutrons measurements, reactor dynamics studies, and operating the reactor. As mentioned in the previous chapters, experimental tasks on neutron activation analysis and activation measurement of thermal neutrons have been gradually included in teaching in recent years. Besides the education of students, research activities run on the reactor as well. From the point of view of the topics addressed in this habilitation thesis, the following reactor characteristics will be focused on the matters of irradiation experiments related to radioanalytical applications.

For the purposes of activation experiments, the VR-1 reactor is equipped with dry vertical channels with an outer diameter of 25 mm, 32 mm, and 56 mm which are located in the reactor core or on its edge. For the purposes of study of samples in which the production of short-lived radionuclides is expected, a rabbit system is also available, and it enables the transport of activation samples between the reactor core and the gamma-spectrometry laboratory in a time of approx. six seconds. The ending of the rabbit system and the dry vertical channels offer the irradiation position of high thermal neutron fluxes. Moreover, the reactor is equipped with a horizontal radial channel with a maximum diameter of 25 cm, and it extends to the edge of the core, and it thus allows the irradiation and study of larger samples. The workplace of the VR-1 nuclear reactor is also equipped with a gamma-spectrometric laboratory with a semiconductor HPGe detector that is primarily intended for measurements of the radioactivity of samples irradiated in the reactor. Adjacent laboratories are used for preparation of samples before the experiment as well as to store them in a lead-shielded safe after irradiation experiment.

The workplace of the VR-1 reactor and its adjacent laboratories provides the irradiation time and background for research activities of external users in free *Open Access* mode [151]. The external scientists interested in the experiment have to prepare a proposal for experiment with required parameters. This proposal is assessed by the reactor manager and, if the experiment is feasible, the corresponding irradiation time is allocated for the irradiation experiment.

For investigation of samples using the neutron activation analysis, the analyzed samples are required to be in solid state and non-abrasive and non-volatile form. Samples are usually encapsulated in paper capsule prior to the experiment or are placed in small plastic bags. If the studied sample is available only in liquid form, it can be placed in the reactor after suitable encapsulation in ampoules⁶ and with the consent of the reactor manager. In the experiments, the half-life of the produced radionuclides is also taken into account, and therefore measures are introduced to prevent the production of a radionuclide with a longer half-life in larger quantities, e.g. ⁶⁰Co with a half-life of 5.2 years. For that reason, the irradiation of unknown samples that are studied using the neutron activation analysis consists of two steps, namely (1) short-term pre-irradiation at lower power, and unless the following gamma-spectrometric analysis confirms the possibility of long-lived radionuclide production, (2) then the studied sample

⁶ Usually in a double encapsulation of a smaller ampoule placed in a larger one.

is irradiated at high (maximum) power. In the case of two-stage irradiation of unknown samples, the requirements for the reactor power and duration of the main irradiation are subsequently derived on the basis of the results obtained from a short pre-irradiation at lower power.

5.3 Neutron field study of NG-2 source with p(35)+Be reaction

The accelerator-driven NG-2 neutron source with a beryllium target station is standardly operated with the p + Be source reaction for a 35 MeV proton beam provided by the U-120M isochronous cyclotron. The beryllium target station is a source of the fast neutron field with broad spectrum up to 33 MeV. This energy range predestinates it primarily for the experimental tasks of validation of microscopic cross-sections of threshold reactions induced by fast neutrons for needs of nuclear data libraries and for tasks of intensive irradiation experiments in materials research, studies of radiation hardness of electronics against fast neutrons, development of new detection systems, and for applications of neutron activation analysis. Improving of the beryllium target station in combination with a detailed study of the fast neutron field is therefore a highly topical and desirable task out of consideration for the frequent use of this neutron source for experimental measurements related to the IFMIF (DONES) fusion program as well as the increasing use by external users.

Due to the utilization of the NG-2 neutron source to study the radiation hardness of electronics against intensive fast neutron field, a hardware upgrade of the beryllium target station was performed in order to reduce the layer of construction materials (target station shell) in the space between the production target and the studied samples. The change in the thickness of shell of the target station head positively affected the irradiation positions, and it allowed the investigated samples to be placed at a smaller distance from the production target. Thus, it contributed to the increase of the neutron flux density in the nearby geometries from the neutron target. This technological modification required subsequently a detailed mapping of the neutron field on the neutron beam axis, where the studied samples and instruments are standardly placed.

After the modification of the Be-target station head, a series of extensive experiments were performed in order to measure the neutron energy spectra at close distances where the samples and electronic components are usually irradiated. Because it was a neutron field measurement in a geometric arrangement where the target-to-sample distances as well as their dimensions are comparable, the neutron activation technique with a set of high purity spectroscopically thin dosimetry foils was used to determine the neutron field. Neutron energy spectra were measured at four positions on the beam axis and at two positions at selected distances from the geometrical axis of the fast neutron beam. In total, three extensive activation experiments were performed to study the neutron field of the p(35) + Be source reaction. Activation foil kits used in the irradiation experiments included dosimetry foils of Al, Au, In, Ti, Fe, Y, Lu, Co, Ni, Nb, and Bi. The results of neutron spectra measurements at positions P0, P2, and P14, i.e. at distances of 14 mm, 34 mm, and 154 mm from the neutron production target, were described and presented in the publication [113], which is included in this commented set of scientific papers, and it is located in Annex P.1. The neutron spectra investigated at positions P6-A and P6-B, which correspond to a distance of 74 mm on the beam axis from target, and 0 mm and 34 mm from the beam axis, were reported in the publication [114], which is located in Annex P.2 of this habilitation thesis. Annex P.3 consists of a publication [115], which closes the topic of the p(35) + Be neutron field measurement, summarizes the previous results of extensive irradiation experiments and adds the obtained result of neutron spectrum measurement at position P6-C at a distance of 74 mm from the target and 16 mm from the beam axis, and it brings the results in line with the measurements of other authors from foreign scientific laboratories. At this point, it is worth noting the benefit of the neutron field determination of the NG-2 neutron source with the p(35) + Be source reaction for the 35 MeV proton beam energy due to the lack of experimentally measured data in relevant energy region. In the EXFOR database, only data for lower energies of proton beams on thick beryllium targets are available in most cases.

The fast neutron flux for a proton beam current of 10 µA on the thick beryllium target reaches values of the order of 10^{11} cm⁻²s⁻¹ at the irradiation position of 14 mm from the production target and the order of 10^9 cm⁻²s⁻¹ at the irradiation position of 154 mm from the target. Specific values of neutron spectral yields are summarized in Table 3 [115] in Annex P.3. The mean energy of the fast neutron spectra reaches a value of 14.2 MeV and it corresponds to the value expected according to the empirical formula derived from the measurements of M.A. Lone [9,2,4]. The accuracy and correctness of the unfolded energy spectra at corresponding positions were further assessed through the C/E ratios, ratios of neutron spectra, and ratios of the experimental reaction rates at position P0, as a fixed position, to the particular investigated position. All these tests are mentioned in the publications in Annexes P.1, P.2 and P.3. Main contributions of the habilitation thesis author to the p(35)+Be neutron field research are as follows: organizing and performing the irradiation experiments at the U-120M cyclotron, gamma-spectrometric measurements, data analysis, MCNPX calculations, neutron spectra reconstruction, and preparation and finalization of scientific papers.

The neutron field of the NG-2 neutron generator with upgraded target station head and utilizing the p(35) + Be source reaction has been described in detail at selected irradiation positions in close geometries from the target that are standardly used in several research applications. In addition, the neutron spectrum with its mean energy of 14 MeV is compatible with the main energy range of expected neutron spectrum at the IFMIF-DONES facility.

5.4 Development of neutron fields of NG-2 source with d(20) + Beand d(15) + Be reactions

To provide a neutron spectrum with a maximum energy range with respect to the parameters of the charged particle beams provided by the U-120M isochronous cyclotron, the NG-2 neutron source with the beryllium target station is operated by default with the p + Be source reaction induced by the 35 MeV proton beam on the thick target. This source reaction provides neutron field with a broad spectrum up to 33 MeV, and thus it has its main importance for research activities related to the IFMIF-DONES program. However, the international research laboratories, that operate the accelerator-driven neutron sources based on thick beryllium targets, primarily utilize the deuteron bombardment of beryllium as a source reaction, i.e. the d + Be reaction, which provides a neutron spectrum of a similar energy range as the proton induced reactions for the same energy of charged particle beam, but the obtained neutron field has a higher intensity.

The neutron spectra based on the d + Be source reaction are preferred as the reference spectra in nuclear databases by their authors. And also thanks to the higher value of neutron spectral yield of the d + Be reaction in comparison with the p + Be reaction, it is desirable to know the neutron field produced by the d + Be source reaction for maximum deuteron beam energy ($E_d \leq 20 \text{ MeV}$) extracted from the isochronous cyclotron U-120M at the NPI in Řež. Such neutron field would be utilizable in intensive irradiation experiments within the applications of fast neutron activation analysis or radiation hardness studies of materials and electronics. Moreover from the point of view of the operation of the NG-2 neutron source, it is also necessary to know the neutron spectra based on the d + Be reaction at selected short distances from the production target, because the neutron field of the d + Be reaction has not been studied at NPI before.

On account of the above-mentioned facts, an activation experiment using a Be-target station and a deuteron beam with an energy of 20 MeV was performed first. It was a brand new experiment to obtain new experimental data for the operated equipment and thus to expand the operational capabilities of the NG-2 neutron source.

For the purpose of determination of the neutron field in close distance to neutron target, the activation method of dosimetry foils was again used for the same reasons as in the case of the study of the neutron field based on the p(35) + Be source reaction. The sets of activation foils for the neutron field spectrometry of the d(20) + Be source reaction included eight materials. Specifically, Al, Au, In, Ti, Fe, Y, Lu, and Co were used. These materials were selected to cover the whole assumed energy range of the investigated neutron field by their expected threshold reactions. Neutron spectra were directly measured by complete sets of activation foils at positions P0 and P14. At positions P2 and P6, the neutron field was monitored by aluminum activation detectors. The designation of the irradiation positions and the associated specific distances corresponds to the designation that was introduced in the study of the p(35) + Besource reaction field from subchapter 5.3.

Experiments and results of measurements of fast neutron spectra from the d(20)+Be source reaction induced by 20 MeV deuteron beam on thick beryllium target are analyzed and presented in paper [129] that is included in Annex P.4 of this commented set of papers. For a deuteron beam with an intensity of 7.25 μ A on a beryllium target, the integral neutron flux at position P0 reached a value of $5.5 \times 10^{10} \text{ cm}^{-2} \text{s}^{-1}$ and at position P14 a value of $1.7 \times 10^9 \text{ cm}^{-2} \text{s}^{-1}$. The neutron fluxes recalculated for the current of 10 μ A reach the values of $7.6 \times 10^{10} \text{ cm}^{-2} \text{s}^{-1}$ at position P0 and $2.3 \times 10^9 \text{ cm}^{-2} \text{s}^{-1}$ at position P14. Assuming the same intensity of the charged particle beam at the Be-target, the neutron flux at the irradiation position P0 is 37% lower for the d(20) + Be source action in comparison with the p(35) + Be source reaction. However at position P14, the fast neutron flux is about 15% higher for the d(20) + Be reaction compared to the p(35) + Be reaction, and it indicates a noticeable forward oriented direction of the d + Be fast neutron field. Even in the case of the neutron spectra measurement of the d + Be reaction by the multi-foil activation technique, the mean energy of the experimentally determined spectra reaches the value predicted according to the empirical equation derived from the measurements of M.A. Lone [9,2,4], i.e. fluence averaged neutron energy for 20 MeV deuteron beam is about 7.7 MeV.

After successful investigation of neutron spectra emitted from the the d(20) + Be source reaction, the d(15) + Be source reaction was studied afterwards. Similarly to the measurements of the d(20) + Be reaction, a new experiment with 15 MeV deuteron beam on a thick beryllium layer was arranged and carried out. The neutron spectra were measured utilizing two sets of nine activation materials (Au, Co, Ti, In, Al, Y, Fe, Ni, Nb) at two standard irradiation positions P0 and P14, and neutron field was also monitored in the longitudinal direction using aluminum activation detectors. From measured responses to the neutron field, neutron spectra with the energy range approx. up to 20 MeV at two source-to-sample distances were reconstructed. For 15 MeV deuteron beam with an intensity of 6.1 µA, the spectral yield of fast neutrons reached a value of 3.1×10^{10} cm⁻² s⁻¹ at position P0 and 8.6×10^8 cm⁻² s⁻¹ at position P14. Details on development of the d(15) + Be neutron field are provided in publication [128] in Annex P.5, where Figures 5 and 6 show the determined neutron energy spectra and Table 3 together with Figure 7 show the values of the observed C/E ratios.

The developed neutron fields based on the d(20) + Be and d(15) + Be source reactions extend the operational and experimental capabilities of the NG-2 neutron source. In both cases, new neutron energy spectra were obtained in two geometries nearby the source target that were measured for the first time at this neutron source, and at the same time, useful data were obtained for the energy region with a lack of experimental data for the d + Be interaction. The neutron fields of the d(20) + Be and d(15) + Be reactions extend the area of utilization of the NG-2 source for intensive irradiation experiments, and they enable the choice of appropriate neutron field depending on the requirements for particular threshold reactions of materials in relevant applications, such as the fast neutron activation analysis. Main contributions of the habilitation thesis author to the d(20) + Be and d(15) + Be neutron fields research are as follows: organizing and performing the irradiation experiments at the U-120M cyclotron, gammaspectrometric measurements, data analysis, MCNPX calculations, neutron spectra reconstruction, and preparation and finalization of scientific papers.

5.5 Development of the p(24)+Be source neutron field

In addition to the research activities related to the IFMIF-DONES program, the NPI has recently started to develop the experimental background for supporting activities focused on the fusion programs ITER and DEMO. From the point of view of ITER and DEMO applications, the neutronics of irradiation systems in energy range of 12 MeV to 20 MeV is interesting as well. In connection to this fact, the NPI is actively developing an HPNS neutron source (High Power Neutron Source) that will be connected to the TR-24 cyclotron, and it will be based on the p + Be source reaction utilizing intensive 300 µA proton beam from the TR-24 cyclotron and energy of 24 MeV. It will create the experimental conditions for material research relevant to the ITER program in the energy region up to 22 MeV.

The already existing experimental facility at the NPI allows to study in advance the p(24) + p(24) +Be source reaction using 24 MeV proton beam delivered by the U-120M cyclotron and the beryllium target station of the NG-2 neutron source. The matters of measurements of new neutron spectra based on the p(24) + Be reaction at the neutron source NG-2 using the multifoil activation technique are discussed in publication [130] in Annex P.6. Neutron spectra were again determined at positions P0 and P14 using a set of ten activation materials (Al, Au, Fe, Ni, Y, Ti, In, Co, Nb, Lu) and employing the SAND-II deconvolution tool. The determined fast neutron spectral yields reached values of $5.4 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$ resp. $1.1 \times 10^9 \text{ cm}^{-2} \text{ s}^{-1}$ at geometrical positions P0 resp. P14 and for 24 MeV proton beam with 10.3 µA intensity. The fluence averaged neutron energy of the determined neutron spectrum corresponds well to the prediction according to the equation by M.A. Lone [9]. The obtained neutron spectra are suitable for validation of activation cross-sections within the ITER program in the energy region above 12 MeV, i.e. determination of cross-sections of gas-producing reactions (n, p) and (n, α) in materials to be used in fusion technologies. At the same time, the performed experiment provided important data necessary for pilot experiments at the HPNS source, in particular for experimental tasks of neutron field spectrometry in a geometrical arrangement nearby target station head. Main contributions of the habilitation thesis author to the p(24) + Be neutron field research are as follows: organizing and performing the irradiation experiment at the U-120M cyclotron, gamma-spectrometric measurements, data analysis, MCNPX calculations, neutron spectra reconstruction, and preparation and finalization of scientific paper.

5.6 Neutron activation analysis in interdisciplinary approach at the VR-1 reactor

In previous sections, the neutron activation method was utilized as a spectrometry technique for a new neutron field measurement of the NG-2 source. However, the experimental approach based on the method of activation of nuclei in irradiated samples can also be successfully used to study the composition of samples. Parallel to neutron field spectrometry of the NG-2 source, neutron activation analysis as a useful radioanalytical method was applied by author of this thesis at the VR-1 training reactor, which is an intensive source of neutrons. The aim was to use the activation analysis at the "edge" of scientific disciplines as a method enabling the overhanging from neutron applications and reactor dosimetry to fields focused on humanities and social sciences.

As part of an interdisciplinary research, dietary supplements composition, historical coins from Tibet and Tibetan traditional medicinal pills, meteorites and tektites, and woolly mammoth remains were investigated at the VR-1 training reactor. The research of samples from everyday life shows the possible ways of interconnection between physical branches and humanities, and it suggests how this kind of specific research at nuclear facilities can help in solving problems in the fields of history, archeology, or industry, etc.

The study of dietary supplements compositions using the neutron activation analysis was published in paper [139] which is incorporated in Annex P.7. Dietary supplements containing zinc, which are over-the-counter drugs in pharmacies on the Czech and Slovak markets, were investigated. Zinc is an essential element for animals as well as plants. It is part of a large number of enzymes in the human body and plays an important role in the metabolism of fats, sugars, and proteins, in the synthesis of the DNA and RNA nucleic acids and positively affects the state of bones, skin and immune system. As a dietary supplement, zinc is preferred by people in the prevention of colds⁷, and so it is taken by many people on regular basis, especially in the winter season. Zinc pills supplied by several manufacturers are available on the market; they differ each other by different contents of active substance, and usual doses change from 10 mg to Forte doses up to 25 mg of zinc per pill. In paper [139], zinc contents in dietary supplements by five manufacturers [152,153,154,155,156] were analyzed, and two to three batches from each manufacturer were monitored. European Union legislation allows certain deviations in the amount of active substance declared by manufacturers on the packaging of products, and in case of minerals, a positive deviation of +45% and a negative deviation of -20% are allowed [157]. The results of NAA experiments with zinc pills at the VR-1 reactor confirmed the declared contents of the active substance in the individual preparations; in the vast majority of cases, these experiments reveal even slightly higher values of zinc per pill than those declared by the manufacturers but still within the permitted tolerances. Based on the results, it can be further stated that the activation analysis at a low-power reactor represents a useful tool in the investigation or verifying the composition of selected dietary supplements, and it has the potential for use in the pharmaceutical and food industries for the investigation of certain substances⁸ or contaminants. Main contributions of the habilitation thesis author to the investigation of zinc pills using NAA are as follows: obtaining the samples, organizing and

 $^{^7}$ Most often in combination with vitamin C.

⁸ With respect to the activation cross-sections and specific properties of the products from the point of view of nuclear gamma-ray spectrometry.

performing the NAA experiments at the VR-1 reactor, gamma-spectrometric measurements, data analysis, and preparation and finalization of scientific paper.

Another type of objects analyzed at the VR-1 reactor were historical Tibetan coins, the results of which were published in paper [140] that is included in Annex P.8. Four pieces of silver and four pieces of copper coins were studied, of which seven coins were real historical coins and the last one was the counterfeit modern coin. In addition to copper and silver, qualitative analysis of coins revealed the presence of silver, gold, indium, zinc, manganese, antimony, and sodium as well. After that, the amount of copper, silver, gold, and zinc were determined by quantitative activation analysis. The results are provided in Table 2 and Figures 7 to 10 in Annex P.8. In addition to determination of the coin composition using the NAA method, the aim was to demonstrate to scientific communities dealing with historical and cultural heritage the possibility of using radioanalytical NAA technique, which can provide them useful data utilizable for interpretation e.g. of origin of their research objects and their movement. For example, data on composition of coins can help Tibetologists with interpretation of the origin of coins, and they allow the identification of a mint in a given location and time, and at the same time they can be economically evident in a given region in corresponding historical age. Main contributions of the habilitation thesis author to the investigation of Tibetan coins using NAA are as follows: organizing and performing the NAA experiments at the VR-1 reactor, gamma-spectrometric measurements, data analysis, thermal neutron flux measurement, and preparation and finalization of scientific paper.

The third type of samples analyzed using the NAA technique were the traditional Tibetan medicinal pills. This research was characterized in paper [141], which is included in Annex P.9. Three types of pills manufactured in Dharamsala, India, were studied. The pills consist of 42 components, including plant, animal, and geological materials, gems, several metals, and the addition of mercury fixed on sulfur. In India, these pills are used to treat serious diseases such as the epilepsy. For these samples, the activation analysis at the VR-1 reactor was used to determine the amount of mercury, arsenic, gold, sodium, iron, potassium, and strontium. The results of the qualitative analysis are provided in Table 1, and the results of the quantitative analysis are summarized in Tables 2 to 4, which are listed in paper in Annex P.9. The aim of this research was to show to the community of Tibetologists and Tibetan physicians that the NAA can be a useful new tool for them to study the impact of Tibetan traditional medicine on human health and Tibetan society. At the same time, it was emphasized that the NAA experiments at the VR-1 reactor are not intended to assess the biological impact of medicine, dietary supplements, food, etc., because the NAA method cannot determine the chemical form of compounds that dictates its absorption in human body and its metabolism. Main contributions of the habilitation thesis author to the investigation of Tibetan medicinal pills using NAA are as follows: organizing and performing the NAA experiments at the VR-1 reactor, gammaspectrometric measurements, data analysis, and preparation and finalization of scientific paper.

In the next stage of the interdisciplinary research at the VR-1 reactor, a detailed analysis of the composition of fragments of two iron-nickel meteorites was carried out, in particular the Muonionalusta meteorite from Sweden, which impacted in the Arctic Circle more than million years ago [158] and the Sikhote Alin meteorite from Russia, which hit as a bolide in 1947 [159]. The samples of fragments of meteorites were moreover supplemented by samples of Moldavites (tektite glass) that were formed by the impact of a meteorite in Bavaria about 15 million years ago [160]. The research on meteorites and tektites composition is described in paper [142], which is included in Annex P.10. The NAA experiments confirm the presence of many elements such as iron, nickel, sodium, magnesium, aluminum, chlorine, potassium, titanium, vanadium, manganese, cobalt, copper, strontium, barium, and gold. For selected elements, the elemental concentration was determined. The results of qualitative analysis are summarized in Table 3, and results of quantitative analysis are presented in Tables 5 and 6 of scientific paper presented in Annex P.10 of this commented paper set. Major and trace elements as well as some contaminants were determined in analyzed samples. It is evident that the NAA technique at low-power VR-1 nuclear reactor can provide astronomers or geologists data utilizable for classification of meteorites, and based on determined sample composition, it can also help to identify the place where the meteoroid impacted on the Earth. Main contributions of the habilitation thesis author to the investigation of meteorites and tektites using NAA are as follows: obtaining the samples, organizing and performing the NAA experiments at the VR-1 reactor, gamma-spectrometric measurements, data analysis for NAA experiments, and preparation and finalization of scientific paper.

In parallel to research of meteorites and tektites, the investigation of composition of mammoth remains was carried out at the VR-1 reactor. Specifically, the fragments of mammoth tusk, molar, and bones, including burned bones were analyzed. The aim was to identify the isotopic vector, i.e., to determine the concentration of selected important radionuclides that make it possible to identify the region of motion of particular individual. The research was realized in collaboration with the Institute of Archaeology of the Czech Academy of Sciences in Brno, and it is presented in paper [143] included in Annex P.11. The qualitative analysis showed the presence of sodium, chlorine, potassium, arsenic, iron, strontium, manganese, bromine, iodine, barium, and uranium (see Table 2 in Annex P.11). The quantitative analysis of woolly mammoth samples for selected elements is summed up in Table 3 in Annex P.11. Based on this research at the VR-1 training reactor, it can be stated that the non-destructive NAA can provide palaeontologists and archaeologists data important e.g. for the inter-comparison of archaeological samples, identification of archaeological site, movement of individuals, or determination of the sample taphonomy (i.e. level of fossilization). Main contributions of the habilitation thesis author to the investigation of mammoth remains using NAA are as follows: organizing and performing the NAA experiments at the VR-1 reactor, gamma-spectrometric measurements, data analysis, and preparation and finalization of scientific paper.

5.7 Measurement of neutron field parameters at the VR-1 reactor

In addition to investigation of the VR-1 research reactor utilization for neutron activation analysis applications, the neutron field parameters in the selected irradiation channel were also measured using the activation technique. The aim was to determine the spectral indexes lengthwise the horizontal radial channel (HRCh) and to determine the neutron energy spectrum. The spectral indexes (in terms of cadmium ratio) are given by the ratio of the reaction rates of the activation foils with the response to the whole energy spectrum and to the epithermal and fast neutron spectrum, that makes it possible to assess the representation of the corresponding part of the neutron spectrum. The matters of determination of selected parameters of the neutron field at the VR-1 reactor for the C3-type core are published in paper [161] which is included in Annex P.12.

Usually, the vertical channels with outer diameter of 32 mm or 25 mm are used for irradiation experiments. They are placed in the reactor core or on its edge. However, the channel diameter is a limiting factor for the dimensions of the irradiated samples. And just for the needs of irradiation experiments of larger samples, the reactor is equipped with a horizontal radial channel with an inner diameter of 250 mm or 90 mm if a reducer is used. To use the radial channel for irradiation experiments, it is therefore appropriate to have some information on the neutron field along its length. Activation detectors of fourteen materials were used to measure the spectral indexes lengthwise the HRCh, in particular Al, NaCl, Sc, Mn, Fe, Ni, Cu, Mo, In, Lu, W, Au, Th, and U bare and cadmium-covered were utilized. Moreover, the activation method was used to determine the neutron spectrum in the HRCh with a set of ten activation foils (Fe, Mn, Cu, Mo, Au, Sc, Dy, In, Ti, and Ta) in a similar way as in the case of the neutron field measurements of the p + Be and d + Be source reactions of the NG-2 neutron source at the NPI CAS in Rež. The obtained results confirm the usability of the radial channel for the NAA irradiation experiments with thermal and epithermal neutrons. These experiments at the HRCh just preceded the wider deployment of neutron activation analysis at the VR-1 training reactor in research activities within the interdisciplinary approach as well as in the standard teaching of students and users of the VR-1 reactor. Main contributions of the habilitation thesis author to the investigation of neutron field parameters at the VR-1 reactor using the dosimetry foils method are as follows: performing the activation experiments at the VR-1 reactor, gamma-spectrometric measurements, data analysis, spectral indexes determination, and preparation and finalization of scientific paper.

Discussion and conclusions

The habilitation thesis was solved at the Faculty of Nuclear Sciences and Physical Engineering of the CTU in Prague and at the Nuclear Physics Institute of the CAS in Řež. It was prepared in the form of the commented set of twelve scientific papers that are an integral part of the thesis and represent the main part of the thesis, and they are supplemented by a unifying text. The research was focused on the use of neutron activation technique in two ways, in particular as the neutron spectrometry method and as the radioanalytical tool in the study of sample composition for a need of the interdisciplinary research.

The thesis dealt with the analysis of issue of neutron field production based on the interaction of charged particle beams delivered by accelerators with suitable target material. Owing to the properties of the accelerator-driven NG-2 neutron source at the NPI, the main focus was put on the matters of neutron fields induced by protons and deuterons on the thick beryllium layers. The energy range above 20 MeV is still relatively poor on experimental data, especially on empirical data on neutron spectral yields, so it certainly provides a space for research purposes. The thesis also briefly characterized the situation about supporting research programs focused on applications of thermonuclear fusion, in particular the status of the IFMIF, FAF-NIR, and IFMIF-DONES research programs that are derived from the ITER and DEMO fusion programs as well as the type of their neutron fields. In connection with them, the research program implemented on the neutron sources of the Nuclear Physics Institute of the CAS was introduced. From the point of view of the fusion applications, the energy range of 12 to 20 MeV is also interesting. Furthermore, the utilization of research nuclear reactors in the world was characterized with respect to the applications of radioanalytical method called the neutron activation analysis in the interdisciplinary approach for the study of historical and archaeological samples, and cultural heritage objects. Because this was an experimentally focused research work, the individual experimental devices on which the research tasks described in this thesis took place were also presented, namely the NG-2 neutron source connected to the U-120M cyclotron and the VR-1 training reactor. The use of both devices was described in a broader context, but primarily with regard to the different variations of irradiation experiments and activation techniques. The thesis also includes the brief methodological background of neutron activation technique supported by the gamma-ray spectroscopy and the method of neutron spectrometry based on them, neutron activation analysis procedures, nuclear data issues and supporting computational tools. Performed research activities were characterized in detail in individual research papers of this commented set of papers.

Extensive and time-consuming research activities were performed within the submitted habilitation thesis. Its merit can be divided into several aspects, particularly into the study of less investigated p + Be and d + Be source reactions, obtaining new experimental data important not only from the research point of view but also important for extending the operational and especially experimental capabilities of used research facilities, and thus the strengthening of the experimental background for running as well as new research programs at the given facilities, and most of these in an interdisciplinary approach with the overlap of applications into other scientific disciplines, which also represent the appropriate degree of innovation. The individual main benefits of the thesis are listed one by one in the following paragraphs.

The neutron field of the p(35) + Be source reaction after technological modification of the NG-2 neutron source was investigated in detail. Using the activation method, the neutron energy spectra were mapped at several important irradiation positions nearby the production target. Good knowledge of the p(35) + Be neutron field is essential for most of research activities currently carried out on this neutron source, specifically for nuclear data validation tasks related to the IFMIF and IFMIF-DONES programs and for newly developed nuclear databases, radiation hardness tests of materials, and destruction test of electronics primarily for ATLAS-CERN experiments.

Moreover, the d + Be source reaction at thick target was investigated at two different deuteron energies for the first time ever at the NPI. New neutron fields of the d(20) + Be and d(15) + Be reactions with continuous energy spectra up to 25 MeV and 20 MeV were developed. They further expand the usability of the NG-2 source for specific intensive irradiation experiments and selected applications of fast neutron activation analysis. Despite the fact that the energy of deuteron beams is about half of the maximum proton beam energy provided by the U-120M isochronous cyclotron, the neutron fields of d + Be reactions can successfully compete with the p(35) + Be neutron field in terms of neutron flux intensity. It should be also stressed that new experimental data of the d + Be source reaction were obtained.

Similarly, the p(24) + Be source reaction was studied for the first time at the NPI as well. The developed neutron field represents again new empirical data important from the point of view of the less known p + Be reaction in the range of high-energy neutrons as well as for extending operational capabilities of the NG-2 source. In should also be stressed that the newly developed neutron field up to 22 MeV covers the range of "fusion" neutrons, that are interesting for applications related to the ITER and DEMO research programs. However, the main benefit is given by the fact that the p + Be source reaction for 24 MeV proton beam, which will be in near future used by new powerful HPNS neutron source at the TR-24 cyclotron, was studied. This made it possible to identify the suitable dosimetry reactions necessary for monitoring such a kind of neutron field and to determine the suitable irradiation parameters. This form of transferability of the experimental conditions will contribute to streamlining the future pilot experiments of neutron spectrometry at the HPNS neutron source.

Neutron activation analysis method was intensively used at the VR-1 training reactor. The possibility of using this radioanalytical technique together with the VR-1 training nuclear reactor as an analytical tool providing valuable data useful for research in humanities, social and natural sciences was investigated. By studying the composition of selected types of samples, the intention was to demonstrate to the community of researchers from the humanities and social sciences the applicability of radioanalytical techniques for obtaining data suitable e.g. for identification the locality of the sample, the economic level of the region, the interpretation of the movement of the sample, verification of authenticity, the analysis of the declared content of the selected substance in the relevant sample, or the representation of contaminants, etc. With this research aim, the historical Tibetan coins containing silver, gold and copper were analyzed, the content of which is characteristic for particular region. Tibetan traditional medicine based on a content of plant, animal, and inorganic constituents with metal and mercury additives was also studied. The content of the active substance in selected dietary supplements available on the Central Europe market was also analyzed. The composition of fragments of two iron-nickel meteorites and Moldavite tektite glasses were investigated as well. Major and trace elements in meteorites and tektites were determined, such data can be useful for classification of meteorites or identifying the place of impact. The woolly mammoth remains were also analyzed at the VR-1 reactor in a non-destructive way, and data utilizable for selection of suitable elemental markers were obtained. They can be used e.g. for monitoring of migration of individuals or determination of the sample fossilization level. All obtained results successfully confirmed the usability of the VR-1 training reactor for neutron activation analysis, and they showed the great potential of this nuclear facility for the research based on interdisciplinary approach. To close the matters of irradiation experiments at the VR-1 reactor it should also be mentioned that the neutron activation technique provided useful data on neutron field parameters of this nuclear facility.

The achieved results were presented at the international scientific ground and published in impacted research and peer-reviewed journals. For the entire period of the research activities, the author of the submitted habilitation thesis published as the main author or as a co-author with active participation in total 47 papers in research scientific journals with the impact factor. In addition, he is the author or co-author of another 28 publications in scientific peer-reviewed journals or in peer-reviewed proceedings with a citation index in the WoS or SCOPUS databases. He presented the results achieved during the entire period of his research life at 31 presentations at prestigious international and domestic conferences.

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List of publications

Publications in scientific journals with impact factor

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- M. Stefanik, E. Simeckova, P. Bem, M. Majerle, J. Novak, M. Ansorge, J. Mrazek, J. Stursa: Neutron spectrum determination of p+Be reaction for 30 MeV protons using the multi-foil activation technique (ND 2019: INTERNATIONAL CONFERENCE ON NUCLEAR DATA FOR SCIENCE AND TECHNOLOGY, Beijing (2019))
- 2. M. Štefánik, P. Bém, E. Šimečková, J. Štursa, M. Majerle, J. Mrázek: The p(20)+Be reaction as a source of fusion relevant neutrons (THE INTERNATIONAL SYMPOSIUM ON FUSION NUCLEAR TECHNOLOGY ISFNT-14, Budapest (2019))
- 3. M. Štefánik, M. Cesnek, Ľ. Sklenka, T. Kmječ, M. Miglierini: Neutron activation analysis of meteorites at the VR-1 training reactor (3RD INTERNATIONAL CONFERENCE ON DOSI-METRY AND ITS APPLICATIONS – ICDA 3, Lisbon (2019))
- 4. M. Štefánik, P. Bém, E. Šimečková, J. Štursa, V. Zach: Neutron spectrum determination of accelerator-driven d(10)+Be neutron source using the multi-foil activation technique (3RD INTERNATIONAL CONF. ON DOSIMETRY AND ITS APPLICATIONS – ICDA 3, Lisbon (2019))
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- M. Štefánik, Ľ. Sklenka, M. Cesnek, M. Miglierini, and J. Rataj: Neutron activation analysis of Tibetan traditional medicinal pills at the VR-1 training reactor Training Reactor (INTERNATIONAL SYMPOSIUM ON RADIATION PHYSICS 14 ISRP-14, Córdoba (2018))
- 7. M. Štefánik, P. Bém, E. Šimečková, J. Štursa, V. Zach, and M. Majerle: Neutron field study of p(24) + Be source reaction using the multi-foil activation technique (30TH SYMPO-SIUM ON FUSION TECHNOLOGY – SOFT-2018, Sicily (2018))
- 8. M. Štefánik, P. Bém, M. Majerle, J. Novák, E. Šimečková, and J. Štursa: Neutron field study of p(35)+Be source reaction at the NPI Rez (THE INTERNATIONAL TOPICAL MEETING ON INDUSTRIAL RADIATION AND RADIOISOTOPE MEASUREMENT APPLICATIONS – IRRMA-X, Chicago (2017))

- 9. M. Štefánik, Ľ. Sklenka, O. Huml, J. Rataj: Activation Analysis of Tibetan Coins and Thermal Neutron Flux Measurement at the VR-1 Training Reactor (THE INTERNATIONAL TOPICAL MEETING ON INDUSTRIAL RADIATION AND RADIOISOTOPE MEASUREMENT APPLI-CATIONS - IRRMA-X, Chicago (2017))
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- 12. M. Štefánik, P. Bém, M. Majerle, J. Novák, and E. Šimečková: Neutron field of accelerator-driven p(35MeV)+Be fast neutron source at NPI Rez (INTERNATIONAL CONFERENCE ON NUCLEAR DATA FOR SCIENCE AND TECHNOLOGY ND2016, Bruges (2016))
- 13. M. Štefánik, P. Bém, M. Majerle, J. Novák, and E. Šimečková: Neutron spectrum determination of d(20)+Be source reaction by the dosimetry foils method (2ND INTERNATIONAL CONFERENCE ON DOSIMETRY AND ITS APPLICATIONS – ICDA 2, Guildford (2016))
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- 18. M. Štefánik, P. Bém, M. Majerle, J. Novák, and E. Šimečková: Experimental validation of IRDFF cross-sections in quasi-monoenergetic neutron fluxes in 20 – 35 MeV energy range (FIRST RESEARCH COORDINATION MEETING ON TESTING AND IMPROVING THE IN-TERNATIONAL REACTOR DOSIMETRY AND FUSION FILE (IRDFF), IAEA, Vienna (2013))
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- 21. M. Štefánik, P. Bém, M. Götz, K. Katovský, M. Majerle, J. Novák, and E. Šimečková: Neutron Spectrum Determination of the p(35 MeV)-Be Source Reaction by the Dosimetry Foils Method (INTERNATIONAL NUCLEAR DATA CONFERENCE FOR SCIENCE AND TECHNO-LOGY ND2013, New York (2013))
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- 25. M. Štefánik, K. Katovský, M. Miletić, J. Šoltés, M. Vinš, and L. Závorka: Amount of Substance Determination by Neutron Activation Analysis at the CTU Training Reactor VR-1 (13TH INTERNATIONAL SCIENTIFIC CONFERENCE ON ELECTRIC POWER ENGINEERING, Liberec (2012))
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Annexes

P.1 Neutron field of accelerator-driven p(35MeV)+Be fast neutron source at NPI Rez

Scientific journal: Volume and year of publication: ISBN, ISSN: DOI: Paper ID in SCOPUS: Accession Number in WoS:

The European Physical Journal – Web of Conferences Volume 146, Article number: 03011, Year: 2017 978-275989020-0, 21016275 doi.org/10.1051/epjconf/201714603011 2-s2.0-85030479329 000426429500078

The undersigned co-authors of the paper "Neutron field of accelerator-driven p(35MeV)+Befast neutron source at NPI Rez" confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

RNDr. Pavel Bém, CSc. Mgr. Mitja Majerle, Ph.D.

Ing. Jan Novák, CSc.

Mgr. Eva Šimečková, CSc.

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P.2 Neutron field measurement of p(35) + Be source using the multi-foil activation method

Scientific journal: Volume and year of publication: ISSN: Impact factor: DOI: Paper ID in SCOPUS: Accession Number in WoS:

Radiation Protection Dosimetry Volume 180, pp. 377–381, Year: 2018 0144-8420 0.831 (year: 2018) doi.org/10.1093/rpd/ncx249 2-s2.0-85055133424 000440983000079

The undersigned co-authors of the paper "Neutron Field Measurement of p(35)+Be Source Using the Multi-Foil Activation Method' confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

RNDr. Pavel Bém, CSc.

Mgr. Mitja Majerle, Ph.D.

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P.3 Neutron field study of p(35)+Be source reaction at the NPI Rez

Scientific journal: Volume and year of publication: ISSN: Impact factor: DOI: Paper ID in SCOPUS: Accession Number in WoS: Radiation Physics and Chemistry Volume 155, pp. 294–298, Year: 2019 0969-806X 2.226 (year: 2019) doi.org/10.1016/j.radphyschem.2018.06.046 2-s2.0-85049526857 000454467400054

The undersigned co-authors of the paper "Neutron field study of p(35)+Be source reaction at the NPI Rez" confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

RNDr. Pavel Bém, CSc.

Mgr. Mitja Majerle, Ph.D.

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Ing. Jan Štursa

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P.4 Neutron spectrum determination of d(20)+Be source reaction by the dosimetry foils method

Scientific journal: Volume and year of publication: ISSN: Impact factor: DOI: Paper ID in SCOPUS: Accession Number in WoS:

Radiation Physics and Chemistry Volume 140, pp. 466–470, Year: 2017 0969806X 1.435 (year: 2017) doi.org/10.1016/j.radphyschem.2017.03.029 2-s2.0-85015628195 000411533500089

The undersigned co-authors of the paper "Neutron spectrum determination of d(20)+Be source reaction by the dosimetry foils method" confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

RNDr. Pavel Bém, CSc. Mgr. Mitja Majerle, Ph.D. Ing. Jan Novák, CSc. Mgr. Eva Šimečková, CSc.

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P.5 Neutron field determination of d+Be reaction for 15 MeV deuterons using the multi-foil activation technique

Scientific journal: Volume and year of publication: ISSN: Impact factor: DOI: Paper ID in SCOPUS: Accession Number in WoS:

Radiation Physics and Chemistry Volume 160, pp. 30–34, Year: 2019 0969-806X 2.226 (year: 2019) doi.org/10.1016/j.radphyschem.2019.03.022 2-s2.0-85063618352 000471733500005

The undersigned co-authors of the paper "Neutron field determination of d+Be reaction for $15 \, MeV$ deuterons using the multi-foil activation technique" confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

RNDr. Pavel Bém, CSc.

Mgr. Eva Šimečková, CSc.

Ing. Jan Štursa

Ing. Václav Zach

Mgr. Jaromír Mrázek, Ph.D.

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## P.6 Neutron field study of p(24)+Be source reaction using the multi-foil activation technique

Scientific journal: Volume and year of publication: ISSN: Impact factor: DOI: Paper ID in SCOPUS: Accession Number in WoS: Fusion Engineering and Design Volume 146, pp. 2053–2056, Year: 2019 0920-3796 1.692 (year: 2019) doi.org/10.1016/j.fusengdes.2019.03.100 2-s2.0-85063229398 000488313700143

The undersigned co-authors of the paper "Neutron field study of p(24)+Be source reaction using the multi-foil activation technique" confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

RNDr. Pavel Bém, CSc. Mgr. Eva Šimečková, CSc. Ing. Jan Štursa Ing. Václav Zach

Mgr. Mitja Majerle, Ph.D.

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#### P.7 Study of dietary supplements compositions by neutron activation analysis at the VR-1 training reactor

Scientific journal: Volume and year of publication: ISSN: Impact factor: DOI: Paper ID in SCOPUS: Accession Number in WoS: Radiation Physics and Chemistry Volume 140, pp. 471–474, Year: 2017 0969-806X 1.435 (year: 2017) doi.org/10.1016/j.radphyschem.2017.03.017 2-s2.0-85019667731 000411533500090

The undersigned co-authors of the paper "Study of dietary supplements compositions by neutron activation analysis at the VR-1 training reactor" confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

Ing. Jan Rataj, Ph.D.

Ing. Ondřej Huml, Ph.D.

doc. Ing. Ľubomír Sklenka, Ph.D.

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#### P.8 Activation analysis of Tibetan coins and thermal neutron flux measurement at the VR-1 training reactor

Scientific journal: Volume and year of publication: Impact factor: DOI: Paper ID in SCOPUS: Accession Number in WoS: Radiation Physics and Chemistry Volume 155, pp. 304–309, Year: 2019 2.226 (year: 2019) doi.org/10.1016/j.radphyschem.2018.06.032 2-s2.0-85049060607 000454467400056

The undersigned co-authors of the paper "Activation Analysis of Tibetan Coins and Thermal Neutron Flux Measurement at the VR-1 Training Reactor" confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

doc. Ing. Ľubomír Sklenka, Ph.D.

Ing. Ondřej Huml, Ph.D.

Ing. Jan Rataj, Ph.D.

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#### P.9 Neutron activation analysis of Tibetan traditional medicinal pills at the VR-1 training reactor

Scientific journal: Volume and year of publication: ISSN: Impact factor: DOI: Paper ID in SCOPUS: Accession Number in WoS: Radiation Physics and Chemistry Volume 160, pp. 96–99, Year: 2019 0969-806X 2.226 (year: 2019) doi.org/10.1016/j.radphyschem.2019.03.020 2-s2.0-85063647578 000471733500015

The undersigned co-authors of the paper "Neutron activation analysis of Tibetan traditional medicinal pills at the VR-1 training reactor" confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

doc. Ing. Ľubomír Sklenka, Ph.D.

Ing. Martin Cesnek, Ph.D.

prof. Ing. Marcel Miglierini, Dr.Sc.

Ing. Jan Rataj, Ph.D.
## P.10 Neutron activation analysis of meteorites at the VR-1 training reactor

Scientific journal: Volume and year of publication: ISSN: Impact factor: DOI: Paper ID in SCOPUS: Accession Number in WoS: Radiation Physics and Chemistry Volume 171, Article number: 108675, Year: 2020 0969-806X 2.858 (year: 2020) doi.org/10.1016/j.radphyschem.2019.108675 2-s2.0-85077698363 000525947600048

The undersigned co-authors of the paper "Neutron activation analysis of meteorites at the VR-1 training reactor" confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

Ing. Marin Cesnek, Ph.D.

doc. Ing. Ľubomír Sklenka, Ph.D.

Mgr. Tomáš Kmječ, Ph.D.

prof. Ing. Marcel Miglierini, Dr.Sc.

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## P.11 Investigation of mammoth remains using the neutron activation analysis at the Training Reactor VR-1

Scientific journal: Volume and year of publication ISSN: Impact factor: DOI: Paper ID in SCOPUS: Accession Number in WoS: Applied Radiation and Isotopes Volume: 166, Article number: 109292, Year: 2020 0969-8043 1.513 (year: 2020) doi.org/10.1016/j.apradiso.2020.109292 2-s2.0-85089540949 000589093400005

The undersigned co-authors of the paper "Investigation of mammoth remains using the neutron activa-tion analysis at the Training Reactor VR-1" confirm the essential contribution of Milan Štefánik to this work. At the same time, we agree that this paper will be included into his habilitation thesis.

doc. Mgr. Sandra Sázelová, Ph.D.

doc. Ing. Ľubomír Sklenka, Ph.D.

Contre Salut

## P.12 Neutron field for activation experiments in horizontal channel of training reactor VR-1

Scientific journal:	Radiation Physics and Chemistry
Volume and year of publication:	Volume 104, pp. 302–305, Year: 2014
ISSN:	0969-806X
Impact factor:	1.189 (year: 2013)
DOI:	doi.org/10.1016/j.radphyschem.2014.05.047
Paper ID in SCOPUS:	2-s2.0-84905907017
Accession Number in WoS:	000341463600063

The undersigned leading academic staff of the research team and supervisor of dissertation thesis [1] confirms the essential contribution of Milan Štefánik to the paper "Neutron field for activation experiments in horizontal channel of training reactor VR-1". At the same time, I agree that this paper will be included into his habilitation thesis.

doc. Ing. Karel Katovský, Ph.D.