# ISTC PROJECT #2578

# "TRANSMUTATION OF RADIOACTIVE NUCLEAR WASTE – PRESENT STATUS AND REQUIREMENT FOR THE PROBLEM-ORIENTED NUCLEAR DATA BASE"

**Project Manager Yu. Korovin**

# Contents

# Introduction

**The transmutation of long lived actinides and, to a lesser extent, of fission products is becoming an important aspect of the overall nuclear fuel cycle assessment.**

**Reactors dedicated to transmutation are now being considered for introduction in the nuclear fuel cycle for burning radwaste: critical (fast reactors) and subcritical (of the ADS type) reactors.**

**WE NEED:**

- **reliable nuclear data are required for calculating the neutronics characteristics of critical (fast reactors) and subcritical (of the ADS type) reactors**
- **improved accuracy in cross-sections for minor actinides**
- **reliable data for estimating radiation damage of ADS**

# Introduction (2)

During the last decade there have been launched a number of **ISTC (International Science and Technology Center)** projects in Russia associated with analysis of nuclear data used in analysis of nuclear waste transmutation.

# The project #2578 (19 months, 2004 - 2005)

The project goal is to elaborate the essential requirements for the theoretical and experimental studies supported by ISTC in addressing the radwaste transmutation problem

**Project Tasks:**

1. Assessment of the present-day demand for nuclear data (on actinide and FP transmutation).
2. Analysis and expert evaluation of the projects completed in accordance with Task 1.
3. Development of ideas on the transmutation potential of FB, ADS and specialized hybrid fusion reactor blankets.

# The project #2578

## Foreign Collaborators

1. International Atomic Energy Agency -
   **Alexander Stanculescu**
2. CIEMAT (Spain) –
   **Enrique M. Gonzalez-Romero**
3. Forschungszemntrum Karlsruhe Institut für Kern und Energiete (Germany) -
   **Cornelis H.M. Broeders**
4. Japan Atomic Energy Research Institute (Japan) -
   **Tokio Fukahori**
5. Royal Institute of Technology (Sweden) -
   **Waclaw Gudowski**

# The project #2578

## Personnel Commitments

*Category I (weapon scientific and technical personnel)*

| Name | Organization |
| --- | --- |
| KOROVIN Yuriy Alexandrovich | Obninsk State University for Nuclear Power Engineering (INPE) |
| BLOKHIN Anatolii Ivanovich | Institute for Physics and Power Engineering (IPPE) |
| IGNATYUK Anatoliy Vladimirovich | Institute for Physics and Power Engineering (IPPE) |
| LOPATKIN Alexandr Victorovich | Research and Development Institute for Power Engineering |
| MATVIENKO Igor Pavlovich | Institute for Physics and Power Engineering (IPPE) |
| TITARENKO Yuriy Efimovich | Institute for Theoretical and Experimental Physics (ITEP) |
| FOMUSHKIN Edward Feodorovich | RENC-VNIIEF |
| SHMELEV Anatolii Nikolaevich | Moscow Engineering Physics Institute (State University) |
| EISMONT Vilen Pavlovich | Khlopin Radium Institute |
| YAVSHITS Sergey Georgievich | Khlopin Radium Institute |

# The project #2578

## Personnel Commitments

*Category II (other scientific and technical personnel)*

| Name | Organization |
|---|---|
| STANKOVSKIY Alexei Yurievich | Obninsk State University for Nuclear Power Engineering (INPE) |
| PILNOV Gennady Borisovich | Obninsk State University for Nuclear Power Engineering (INPE) |
| CHUKREEV Felix Evgenievich | RRC Kurchatov Institute |
| ARTISUK Vladimir Vasilevich | Obninsk State University for Nuclear Power Engineering (INPE) |
| LUKASEVICH Ivan Borisovich | Research and Development Institute for Power Engineering |
| BATYAEV Vyacheslav Felixovich | Institute for Theoretical and Experimental Physics (ITEP) |

# Assessment of the Present-day Demand for Nuclear Data on Transmutation Nuclear Waste

• During the last few years, there have been a number of attempts to estimate nuclear data needs as related to problem of RW transmutation and to highlight priorities for the future experimental and evaluation work.

•To evaluate uncertainties in both the available and required data on minor actinides, their influence on the neutronics characteristics of the fast reactor core were investigated.

# Assessment of the Present-day Demand for Nuclear Data on Transmutation Nuclear Waste

**Existing and required (in brackets) uncertainties of actinide cross-sections (the calculations were made at IPPE for the BN-800 reactor)**

| Nuclide | Capture cross-section, % | Fission cross-section, % | Inelastic scattering cross-section, % |
|---|---|---|---|
| Np-237 | 15 (5) | 7 (3) | 30 (10) |
| Pu-238 | 25 (10) | 10 (5) | 40 (30) |
| Pu-239 | 6 (4) | 3 (5) | 20 (15) |
| Pu-240 | 10 (5) | 5 (5) | 20 (15) |
| Pu-241 | 15 (5) | 5 (3) | 20 (20) |
| Am-241 | 10 (5) | 10 (5) | 30 (10) |
| Am-242m | 30 (10) | 15 (5) | 40 (30) |
| Am-243 | 30 (10) | 10 (5) | 30 (30) |
| Cm-242 | 50 (10) | 15 (5) | 30 (30) |
| Cm-243 | 50 (10) | 15 (5) | 30 (30) |
| Cm-244 | 30 (20) | 10 (5) | 30 (30) |

# Assessment of the Present-day Demand for Nuclear Data on Transmutation Nuclear Waste

- The analogous analysis has been performed recently for the subcritical reactor with Pu/MA nitride fuel (MA/Pu ratio equal to 2), lead-bismuth cooled and driven by an accelerator. This burner is representative of most current proposals, as indicated in different international programs.
- The greatest influence on the total uncertainty is exerted by the cross-sections in the energy range below 20 MeV. High-energy (above 20 MeV) data do not have a significant impact on most neutronics reactor parameters, except for the values related to radiation damage of materials.

# Assessment of the Present-day Demand for Nuclear Data on Transmutation Nuclear Waste

- The most of the ADS projects deal with lead-bismuth eutectic for the target material and blanket cooling. It necessitates possessing accurate nuclear data at high neutron energies. **The associated data presented in ENDF/B-VI, JENDL-3.2, BROND-2 are not consistent. The difference achieves 40-50%.**
- The contributions of minor actinides and fission products to the total transmutation rate in ADS-based transmuters are different from those in conventional reactors, which shows how important the influence of MA data uncertainties is. And this, in its turn, determines waste repository parameters.

# Assessment of the Present-day Demand for Nuclear Data on Transmutation Nuclear Waste

Decay heat uncertainty. It shows the component breakdown and their evolution with time.

| Contribution to decay heat (%) | t = 0 | t[(a)] = 10 days | t = 1 month | t = 1 year | t = 10 years |
|---|---|---|---|---|---|
| Actinides:<br>Fission products: | 23%<br>77% | 86%<br>14% | 90%[(b)]<br>10% | 94%[(b)]<br>6% | 100%[(c)]<br>- |
| Uncertainty in decay heat due to actinides: | ±10% | ±11% | ±11% | ±14% | ±65% |

(a) time after shut-down ;
(b) mostly due to Cm-242 and Cm-244 ;
(c) mostly due to Cm-242, 245 and 246.

# Assessment of the Present-day Demand for Nuclear Data on Transmutation Nuclear Waste

The uncertainty due to the actinide contribution to full decay heat results in the total uncertainty on this parameter for ADS being 2-3 times higher than the value currently achieved for conventional reactors

*Short list of the cross-sections uncertainties*

| Isotope | Cross-section | Bound. [a] | Accuracy achieved (%) | Accuracy required (%) |
|---|---|---|---|---|
| $Pu^{239}$ | $\sigma_{fiss}$ | 4 | 6.5 | 3.4 |
| | | 5 | 4 | 3.1 |
| $Np^{237}$ | $\sigma_{fiss}$ | 3 | 25 | 8.0 |
| | | 4 | 25 | 5.1 |
| | $\nu$ | 4 | 5 | 4.1 |
| $Am^{243}$ | $\sigma_{cap}$ | 4 | 40 | 10.4 |
| | | 5 | 40 | 5.5 |
| | | 6 | 40 | 5.1 |
| | | 7 | 20 | 5.9 |
| | | 8 | 20 | 6.3 |
| | $\sigma_{fiss}$ | 2 | 20 | 7.6 |
| | | 3 | 20 | 6.2 |
| | | 4 | 20 | 5.4 |

# Assessment of the Present-day Demand for Nuclear Data on Transmutation Nuclear Waste

| Isotope | Cross-section | Bound. [a] | Accuracy achieved (%) | Accuracy required (%) |
|---|---|---|---|---|
| $Cm^{241}$ | $\sigma_{fiss}$ | 2 | 40 | 10.0 |
| | | 3 | 40 | 8.5 |
| | | 4 | 40 | 5.0 |
| $Cm^{245}$ | $\sigma_{fiss}$ | 5 | 30 | 9.7 |
| | | 6 | 30 | 9.6 |
| $Fe^{56}$ | $\sigma_{inel}$ | 4 | 20 | 4.9 |

(a)

| Group | Upper boundary (MeV) | Group | Upper boundary (MeV) |
|---|---|---|---|
| 1 | 19.6 | 9 | $9.12 \cdot 10^{-3}$ |
| 2 | 6.07 | 10 | $2.04 \cdot 10^{-3}$ |
| 3 | 2.23 | 11 | $4.54 \cdot 10^{-4}$ |
| 4 | 1.35 | 12 | $2.26 \cdot 10^{-5}$ |
| 5 | 0.498 | 13 | $4.00 \cdot 10^{-6}$ |
| 6 | 0.183 | 14 | $5.40 \cdot 10^{-7}$ |
| 7 | $6.74 \cdot 10^{-2}$ | 15 | $1.00 \cdot 10^{-7}$ |
| 8 | $2.48 \cdot 10^{-2}$ | | |

# Assessment of the Present-day Demand for Nuclear Data on Transmutation Nuclear Waste

To sum up, it should be mentioned that the following areas cause much concern in terms of uncertainties:

- Data on the following microscopic MA cross-sections: $\sigma_f$, $\sigma_c$, $\sigma_{in}$. It is imperative to carry out experiments and to obtain evaluated data in the energy range of 10 MeV to 0.1 – 1 keV.
- Data in the energy range above 20 MeV which have a significant influence on certain reactor parameters, in particular, on the radiation damage evaluation.
- Data on decay heat which basically depend on the data for minor actinides (Cm, in particular).

# Nuclear Data Evaluation and Development of Nuclear Models for Heavy Nuclei

**Energy region below 20 MeV:**

The statistical models work well and major problems here are related to correct choice of model parameters (fission barriers and level density parameters).

**Energy region from 20 to 200 MeV and above 200 MeV:**

significantly rises the contribution of preequilibrium processes both in the form of nucleon-nucleon interactions (intranuclear cascades) and processes of system relaxation into equilibrium state (Griffin exciton models, etc.) Various versions of statistical model with preequilibrium emission are applied.

It is widely accepted to use intranuclear cascade models only above 200 MeV, where various versions of statistical model with preequilibrium emission can be applied.

Such approach leads to inconsistency of models at 200 MeV. European colleagues who use TALYS code below 200 MeV and INCL intranuclear cascade model above 200 MeV will face the same problem.

# Nuclear Data Evaluation and Development of Nuclear Models for Heavy Nuclei

## Problem:

- Neglecting the cascade part of reaction with nucleons below 200 MeV leads to wrong value of compound nuclei formation cross-section, which is initial for the models preequilibrium emission. At the same time, taking into account the cascade processes in the whole energy region above 20 MeV eliminate the necessity of model joining at 200 MeV and leads to consistent description of reaction mechanism in the whole energy range above 20 MeV. At the same time, it is necessary to keep all the advantages of statistical models obtained at describing the cross-sections of reactions with nucleons below 20 MeV.

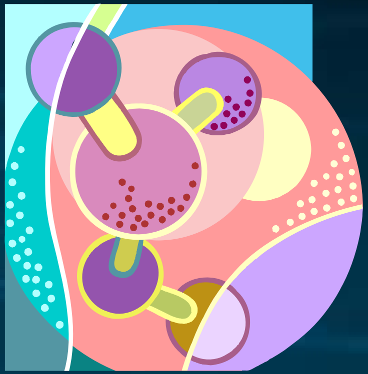
# Radionuclide Production for p+184W at the Proton Energy 1.6 GeV. Number of residuals is 91

| Factor | Bertini/ Dresner | Bertini/ ABLA | ISABEL/ Dresner | ISABEL / ABLA | INCL4/ Dresner | INCL4/ ABLA | CEM2k | CASCADE (original) | CASCADE/ ASF (present result) |
|---|---|---|---|---|---|---|---|---|---|
| H | 6.89 | 5.67 | 5.45 | 5.91 | 5.25 | 6.08 | 5.88 | 4.90 | 4.51 |
| D | 0.48 | 0.44 | 0.44 | 0.44 | 0.38 | 0.40 | 0.45 | 0.35 | 0.33 |
| F | 1.87 | 2.63 | 2.60 | 2.83 | 2.73 | 2.57 | 3.60 | 2.85 | 1.69 |

$$H=\left(\frac{1}{N}\sum_{i=1}^{N}\left(\frac{\sigma_i^{exp}-\sigma_i^{calc}}{\Delta\sigma_i^{exp}}\right)^2\right)^{1/2},\quad D=\frac{1}{N}\sum_{i=1}^{N}\left|\frac{\sigma_i^{exp}-\sigma_i^{calc}}{\sigma_i^{exp}}\right|,\quad F=10^{\left(\frac{1}{N}\sum_{i=1}^{N}\left[\log(\sigma_i^{exp})-\log(\sigma_i^{calc})\right]^2\right)^{1/2}}$$

# Cascade models

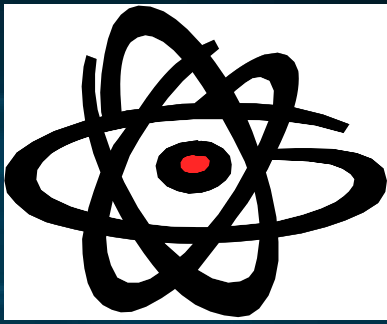
- were primary developed to describe high-energy processes with branched interactions and practically in all the codes the description of the statistical part of the reaction is considered very simplistically.



- Modern cascade models have to describe fast reaction part, preequlibrium model with multiple particle emission from excited nuclei, to describe the process of establishing equlibrium and Huser-Feshbach formalism and statistical part.

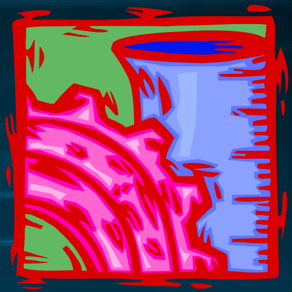
# Cascade models

It is necessary:



- to improve the model for calculation of fission barriers with accuracy better than 1 MeV
- develop a model for correct accounting the dependence of barrier from excitation energy
- develop models and perform calculations of fragment yields from excited nuclei both for evaluation of ADS target activation and for estimation of their influence on the ADS performance

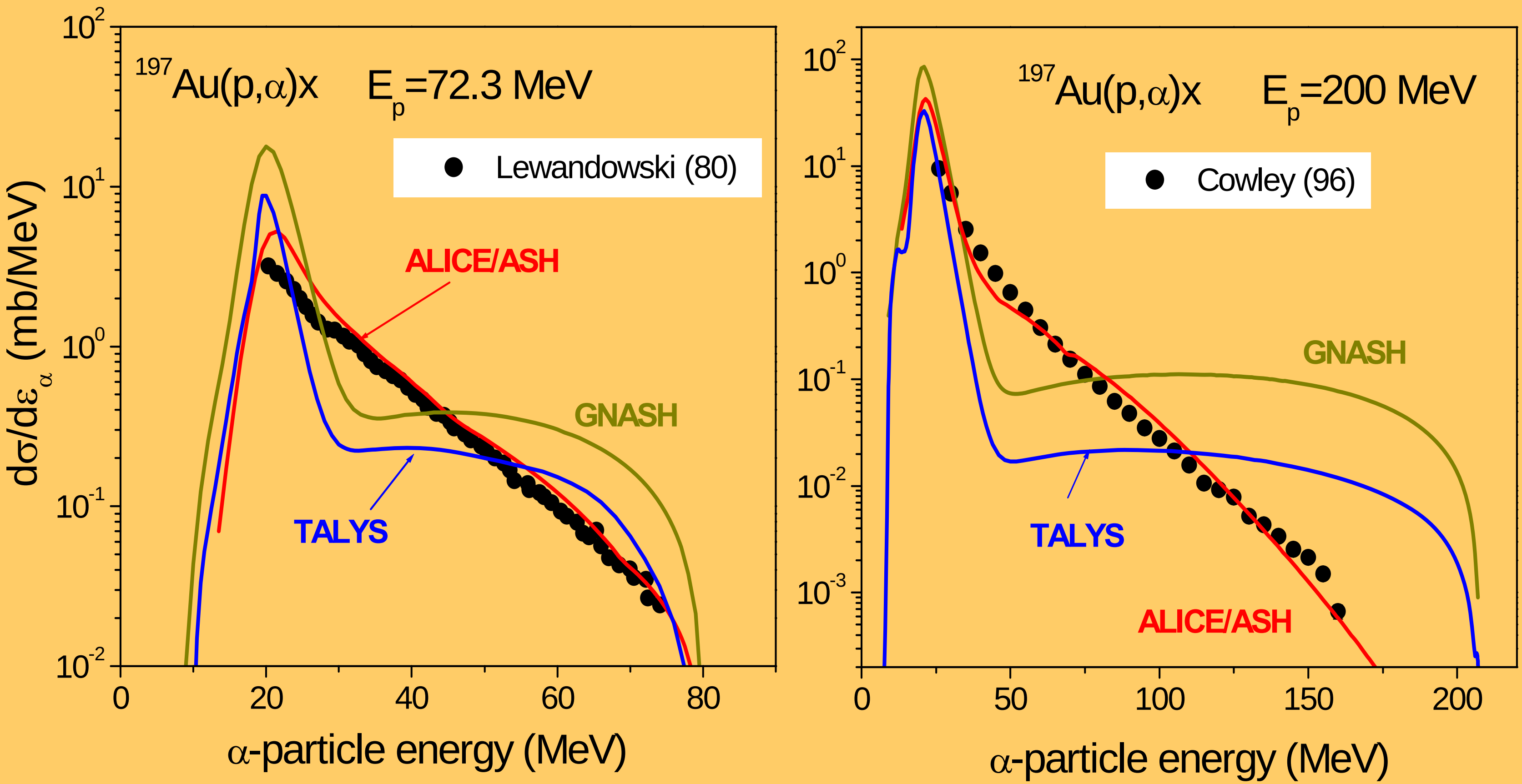
# Effect of Nuclear Data Uncertainties on Radiation Damage of Structural Materials



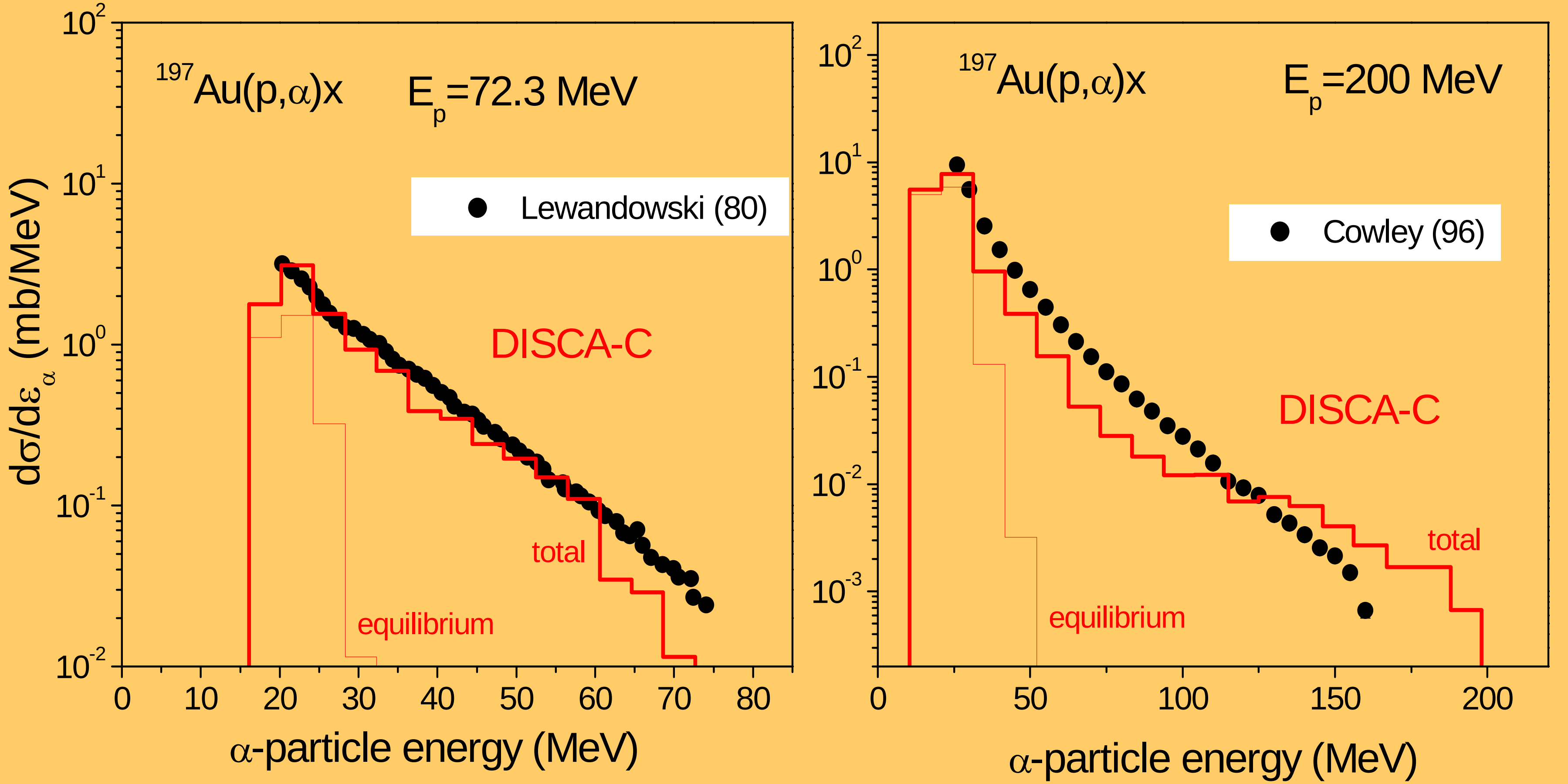
The gas production rates due to proton interactions with matter exceeds the gas generation rate due to neutron by more than one order of magnitude.

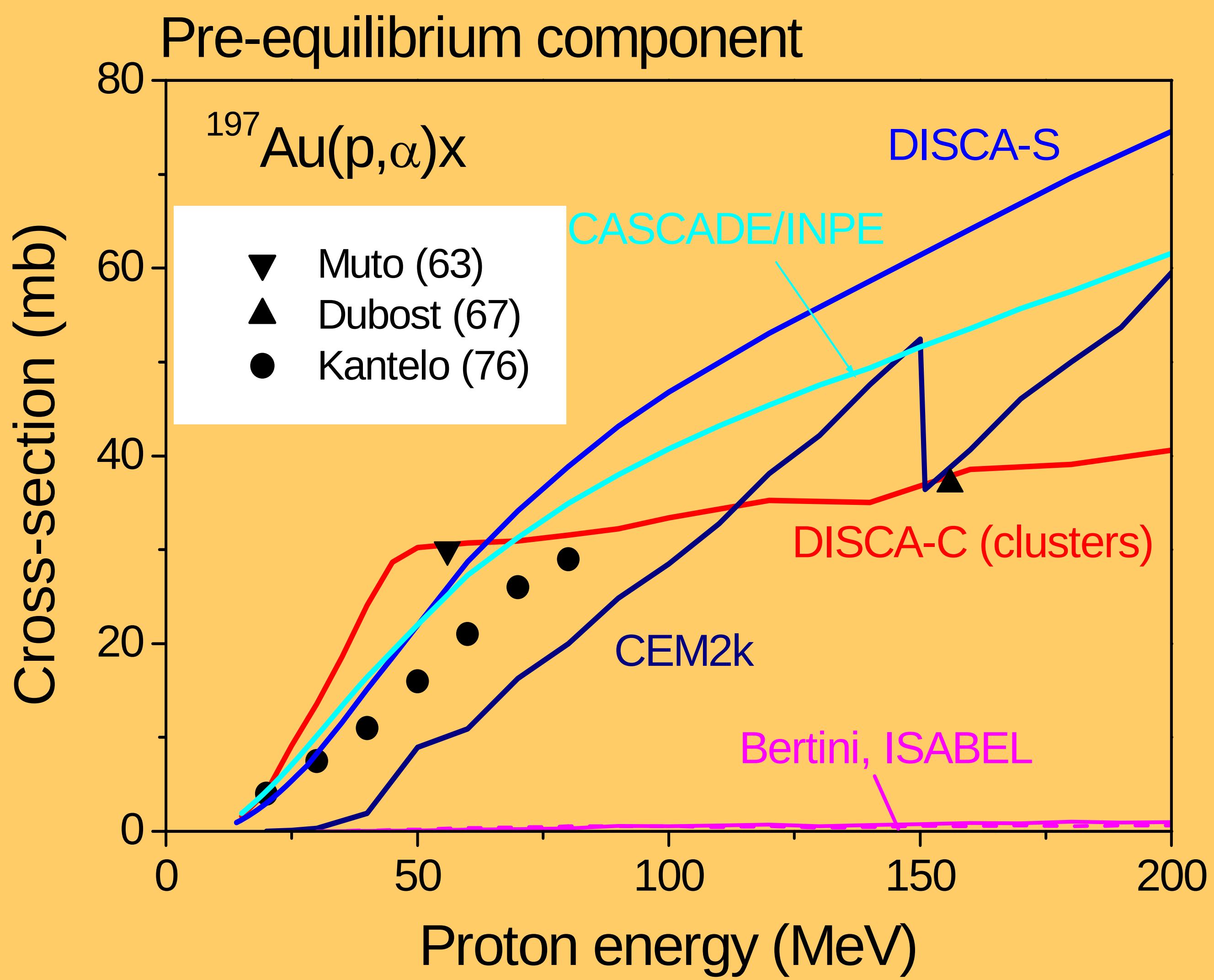
This is significant feature of irradiation conditions compared to standard fission reactor spectrum. The concentrations of helium and hydrogen differ in almost one order between each other, but owing to high mobility of hydrogen because of diffusion at high temperatures its concentration reduces.

# GNASH, TALYS and ALICE/ASH

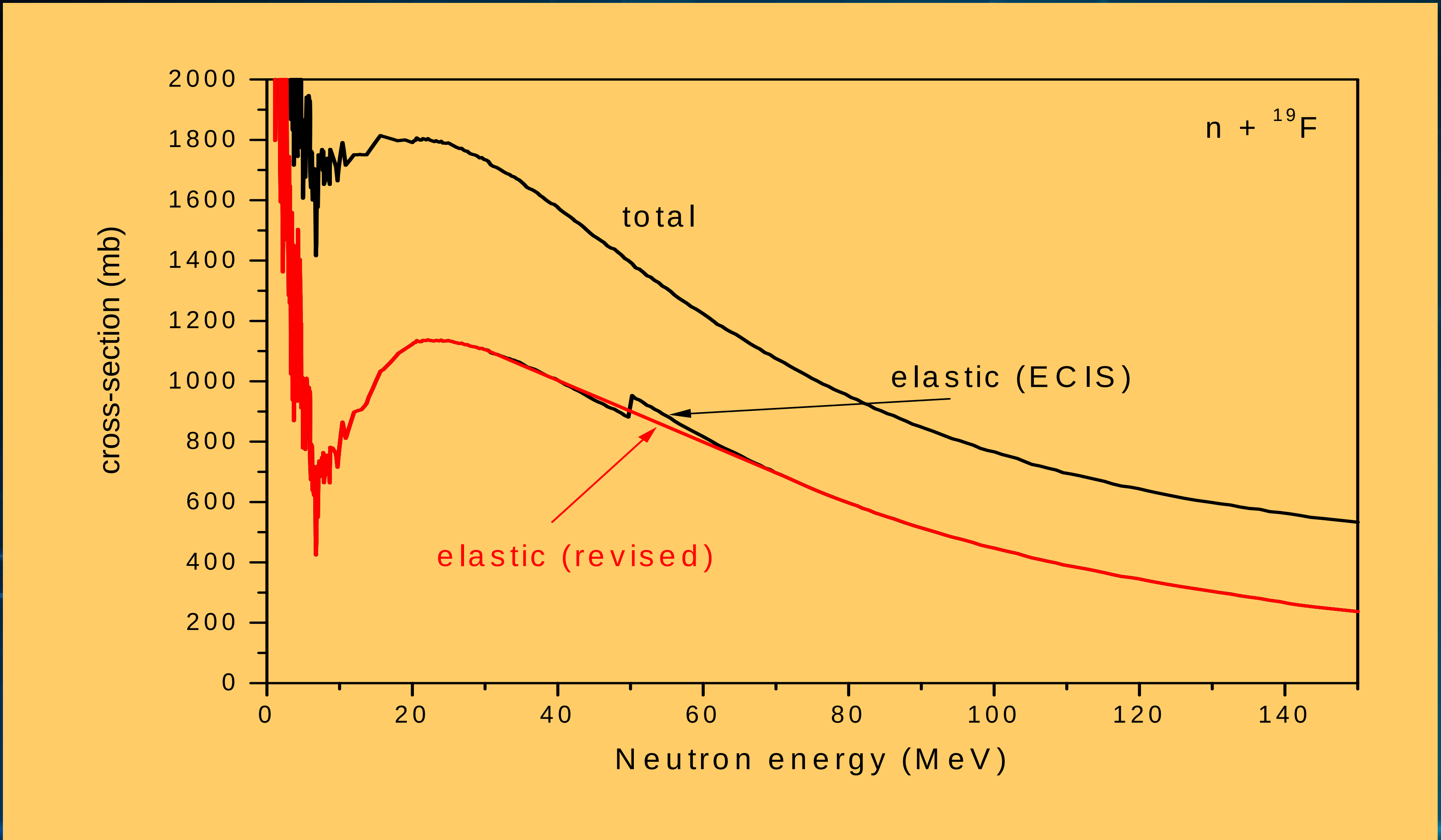


# INC (clusters) +EQ model. DISCA-C




Pre-equilibrium component
$^{197}$Au(p,α)x
Muto (63)
Dubost (67)
Kantelo (76)
DISCA-S
CASCADE/INPE
DISCA-C (clusters)
CEM2k
Bertini, ISABEL
Cross-section (mb)
Proton energy (MeV)
0
20
40
60
80
0
50
100
150
200

# Difficulties at Cross-section Evaluation for Light Nuclei



# Models for Radiation Damage Calculations

To obtain realistic estimation of lifetimes of the materials in such conditions, the experimental data are necessary

*Data on dpa and gas production rate uncertainties*

| | Max. dpa | Max. Helium production rate | Max. hydrogen production rate | Max (He / dpa) |
|---|---|---|---|---|
| $\Delta I_{no\_correlation}$ | ±29.9 | ±43.6 | ±28.5 | ±45.5 |
| $\Delta I_{PEC}$ (a) | ±48.9 | ±59.1 | ±53.1 | ±67.4 |

*(a) Partial correlation in energy*

# Models for Radiation Damage Calculations

**The radiation damage characteristics might be calculated by using of NJOY code. It uses the NRT formulas for the calculation of the damage energy.**

**But such calculation is not correct for:**

- **the light secondary ions at the energies, where electronic loss is not proportional to $E^{1/2}$**
- **the case of noticeable difference between Z and A for the secondary ion and material**
- **compounds**

***The IOTA (Ion TrAnsport in Materials) has been developed***

# Nuclear Data for Radiation Damage Issues

The code IOTA was developed to obtain the total number of primary defects created in materials, the displacement cross-section and the space defect distribution. The simulation of the ion movement in the media is performed with the help of the different approaches:

- *binary collision approximation (BCA)*
- *Monte Carlo method.*

The experimental data for ion stopping power are used for the calculation. Besides that, the special edition of the IOTA code allows to consider the inelastic nuclear interactions of the primary protons with materials. It uses the inelastic displacement cross-sections calculated with the help of the intranuclear cascade evaporation model for a wide number of materials

# Project Analysis

The projects analyzed can be roughly subdivided into 3 groups:

1. **Projects related to the measurement of microscopic nuclear data (cross-sections, spectra, multiplicities, etc.);**
2. **Projects related to studying the interaction of charged particles with different materials;**
3. **Integral experiments carried out by using blanket mockups to investigate the integral neutron physics characteristics of the facilities.**

# Project Analysis

**16 ISTC projects of status 6 (Project underway) and status 8 (Project completed) related to evaluating and measuring micro-data were analyzed.**

# Project Analysis

| ## | TITLE | Status | Dates From / To | Manager | Leading institute / Collaborators |
|---|---|---|---|---|---|
| *Nuclear data measurements* | | | | | |
| 183.1, 183.2 | **Fission-Neutron Spectra for Minor Actinides** | 8 | 1995 / 1999 | L.Drapchinsky | KRI /FZK, CEA, JAERI, ENEA, Belgonucleaire |
| 304 | **Measurements and Analysis of Basic Nuclear Data for Minor Actinides** | 8 | 1995 / 1997 | N.Rabotnov | IPPE /JAERI, LANL |
| 471 | **Neutron Fission and Capture Data** | 8 | 1996 / 1998 | V.Furman | JINR /EC |
| 540 | **Measurement of cross-sections for nuclear fission by 15<En<160 MeV neutrons for fundamental and applied purposes** | 8 | 1997 / 1999 | V.Eismont | KRI / Uppsala Univ., FZK |

# Project Analysis

| | | | | | |
|---|---|---|---|---|---|
| 554 | **Fission-Neutron Multiplicity Distributions in the Spontaneous Fission of Cm-244, Cm-248 and in the Neutron Induced Fission of U-233 and Pu-239** | **8** | 1997 / 1999<br>2001 / 2003 | V.Shpakov | KRI / EC |
| 609 | **Neutron Induced Fission Cross-sections of Some Actinides and other Heavy Nuclei in Energy Region 1-200 MeV** | **8** | | O.Scherbakov | PNPI / JAERI |
| 731 | **Benchmark Data on Gamma-ray Production for Fusion Application** | **8** | 1997 / 2000 | A.Ignatyuk | IPPE / JAERI |
| 1309 | **Comparison of Proton- and Neutron-Induced Fission Cross Sections of Lead and Neighboring Nuclei in the 20-200 MeV Energy Region** | **8** | 1999 / 2002 | V.Eismont | KRI / EC, FZK, Uppsala Univ. |

# Project Analysis

| | | | | | |
|---|---|---|---|---|---|
| 1828 | **Measurements of the Prompt Neutron Spectra of Minor Actinides. Fast Neutron Induced Fission of 241Am and 243Am, Thermal Neutron Induced Fission of 243Cm** | **8** | 2001 / 2004 | L.Drapchinsky | KRI / FZK, Belgonucleaire , CEA |
| 1971 | **Neutron Induced Fission Cross-Sections of Pu-240, Am-243 and W in the Energy Range 1–200 MeV** | **8** | 2001 / 2004 | A.Laptev | PNPI / LANL |
| 2253 | **Investigation of the Delayed Neutron Characteristics from the Fission of Compound Nuclei Th-233, U-234, U-235, Am-244, Np-238, Cm-246, Pa-233, Pa-234, Np-239, Np-240 at the Excitation Energies from 5 to 20 MeV** | **6** | 2004 / | W.Furman | JINR / LANL, CEA, ENEA, JAERI |

# Project Analysis

| | | | | | |
|---|---|---|---|---|---|
| | *Nuclear data evaluations* | | | | |
| B-003 | **Evaluation of Actinide Nuclear Data** | **8** | 1996/ 1998 | V.Maslov | IRPCP / JAERI |
| B-404 | **Evaluation of Nuclear Data for Th-U Fuel Cycle** | **8** | 2001 / 2003 | V.Maslov | IRPCP / JAERI |
| 964 | **Development of Model Code and Theoretical Data Library on Fission Cross Sections for Wide Energy Range** | **8** | 1999 / 2001 | S.Yavshits | KRI / FZK |
| 2524 | **Development of Nuclear Data Library for Nucleon-Induced Reactions on Heavy Nuclei in Wide Energy Region** | **6** | 2003 / | S.Yavshits | KRI / Kyungpook Univ., Texas A&M Univ., FZK, IAEA |

# Project Analysis

| *Proposed projects* | | | | |
|---|---|---|---|---|
| ## | TITLE | Status | Manager | Leading institute |
| 217 | Capture Cross-Section Measurements of Short-Lived Nuclei with the Pulse Neutron Source | 3 | L.Drapchinsky | VNIIEF |
| 969 | Measurement and Evaluation of the Thorium Fuel Cycle Neutron Data | 3 | N.Kornilov | IPPE |
| 1049 | Level Density Studies Based on Measurements of Evaporation Spectra and Cross-Section Fluctuations in Nuclear Reactions | 3 | B.Zhuravlev | IPPE |
| 1069 | Double-Differential Prompt Neutron Spectra Measurements in 248cm Spontaneous Fission. Calculation and Evaluation of Prompt Neutron Spectra in Fission of Neptunium, Americium and Curium Isotopes | 3 | O.Batenkov | Khlopin Radium Institute |

# Project Analysis

| | | | | |
|---|---|---|---|---|
| 1191 | Measurement of Reaction Cross Sections Leading to Production and Destruction of Radioactive Material | 3 | A.Filatenkov | Khlopin Radium Institute |
| 1227 | Transuranium Radionuclides: Producing Highly Enriched Isotope Samples, Measuring Emission Probabilities of Radiations and Decay Data Evaluation | 3 | V.Chechev | Khlopin Radium Institute |
| 1749 | Measurements of the Cross Sections of Fast and Resonance Neutrons Induced Fission of Minor Actinides for Their Transmutation with Accelerator-Driven Systems | 3 | B.Fursov | IPPE |
| 2199 | Neutron Cross-Sections in Resonance Energy Range and Nuclear Level Density for Fission-Products, Th, 233-U | 3 | B.Zhuravlev | IPPE |
| B-379 | Actinide Nucleon-Indused Fission Cross Sections up to 150 MeV | 3 | V.Maslov | IRPCP |

# Project Analysis

12 ISTC project of status 6 (Project underway) and status 8 (Project completed) related to studying the interaction of charged particles with different materials were analyzed.

# Project Analysis

| ## | TITLE | Status | Dates From / To | Manager | Leading institute / Collaborators |
|---|---|---|---|---|---|
| 157 | Spallation Experiment with Tungsten Target | 8 | 1995 / 1996 | V.Belyakov-Bodin | ITEP / JAERI |
| 187 | Development of a Nuclear Data System for the Radiation Problems of High and Intermediate Energy Physics | 8 | 1995 / 1996 | E.Gelfand | MRTI RAS / IAEA |
| 477 | Nuclear-physical experiment with chloride target | 8 | 1997 / 1998 | V.Belyakov-Bodin | ITEP / JAERI |
| 839 | Experimental and Theoretical Study of the Yields of Residual Product Nuclei Produced in thin Targets Irradiated by 100-2600 MeV Protons | 8 | 1997 / 1998 | Yu.Titarenko | ITEP / CEA, JAERI |

# Project Analysis

| | | | | | |
|---|---|---|---|---|---|
| 839 (2) | Experimental and Theoretical Study of the Yields of Residual Product Nuclei Produced in thin Targets Irradiated by 100-2600 MeV Protons | 8 | 1999 / 2001 | Yu.Titarenko | ITEP / JAERI, CEA, Royal Inst., Universitat Hannover (ZSR), ENEA |
| 1145 | Nuclear-Physics Investigations Aimed at the Solution of Weapon Plutonium Conversion and Long-Lived Radioactive Wastes Transmutation Problems | 8 | 1999 / 2002 | E.Fomushkin | VNIIEF / JAERI |
| 1309 | Measurements and Comparison of Proton- and Neutron-Induced Fission Cross Sections of Lead and Neighboring Nuclei in the 20-200 MeV Energy Region | 8 | 1999 / 2002 | V.Eismont | KRI / EC, FZK, Uppsala Univ. |
| 1405 | Proton Induced Fission Cross Sections for Heavy Nuclei in the Energy Range 200-1000 MeV | 8 | 1999 / 2001 | V.Vovchenko | PNPI / JAERI |

# Project Analysis

| | | | | | |
|---|---|---|---|---|---|
| 2002 | Proton-Pb and Proton-Bi Reaction Yields | 8 | 2002 / 2004 | Yu.Titarenko | ITEP / Royal Inst., LANL, CEA, Gutenberg-Universitat, Tokyo Inst. |
| 2213 | Fission Cross Sections of Tungsten Isotopes | 6 | 2002 / | V. Eismont | KRI / Uppsala Univ., EC, FZK |
| 2267 | Construction of the Subcritical Assembly with Combined Neutron Spectra Driven by Proton Accelerator at Proton's Energy 660 MeV for Experiments on Long Lived Fission Products and Minor Actinides transmutation" | 6 | 2005 / | V.Shvetsov | JINR / CEA, Royal Inst.; FZK, EURATOM-Ciemat |
| 2405 | Experimental Nuclear-Physics Data for Transmutation | 6 | 2005 / | E.Fomushkin | VNIIEF / LANL |

# Project Analysis

*Proposed projects*

| # | ## | TITLE | Status | Dates From / To | Manager | Leading institute |
|---|---|---|---|---|---|---|
| | 1251 | Neutron- and Heat- Physical Experiments with Cylindrical Targets Bombarded with Medium-Energy Protons | 3 | | V.Belyakov-Bodin | ITEP |
| | 1314 | Measurement and Analysis of Fission Cross Sections of Heavy Targets Induced by 30-3000 MeV Protons | 3 | | V.Eismont | KRI |
| | 3266 | Experimental and Theoretical Study of the Residual Nuclide Production in 40-2600 MeV Proton-Irradiated Thin Targets of ADS Structure Materials | 2 | | Yu.Titarenko | ITEP |

# Project Analysis

**Along with new micro-data, the analysis of the integral experiments provides valuable ( and in some cases crucial) information.**
**11 ISTC projects (Status 6-8) of this kind were analyzed**

# Project Analysis

- Table

| ## | TITLE | Status | Dates From / To | Manager | Leading institute / Collaborators |
|---|---|---|---|---|---|
| 017 | Feasibility study of technologies for accelerator based conversion of military plutonium and long-lived radioactive waste | 8 | 1994 / 1996 | V.Kazaritsky | ITEP / LANL, CERN |
| 304 | Measurements and Analysis of Basic Nuclear Data for Minor Actinides | 8 | 1995 / 1997 | N.Rabotnov | IPPE / JAERI, LANL |
| 559 | Pilot Flow Lead-bismuth Target of MW Power for Accelerator-Driven Systems | 8 | 1996 / 1998 | E.Yefimov | IPPE / FZK, Royal Inst. of Technology, LANL, CEA |

# Project Analysis

| | | | | | |
|---|---|---|---|---|---|
| 910 | Execution of the Complex of Benchmark Experiments for Testing the Nuclear Data of Vanadium - Main Component of Low-Activation Structural Materials for Perspective Nuclear Energetics | 8 | 1997 / 2000 | D.Markovskiy | RRC KI / IAEA, FZK |
| 910 (2) | Execution of the Complex of Benchmark Experiments for Testing the Nuclear Data of Vanadium - Main Component of Low-Activation Structural Materials for Perspective Nuclear Energetics | 8 | 2001 / 2002 | D.Markovskiy | RRC KI / FZK, JAERI, IAEA |
| 1145 | Nuclear-Physics Investigations Aimed at the Solution of Weapon Plutonium Conversion and Long-Lived Radioactive Wastes Transmutation Problems | 8 | 1999 / 2002 | E.Fomushkin | VNIIEF / JAERI |

# Project Analysis

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| 1372 | **Analysis of Long-Lived Nuclear Waste Transmutation in Fast Reactors and High Energy Accelerators** | **8** | **2002 / 2005** | **E.Smetanin** | IPPE / EC |
| 1486 | **Experimental and Theoretical Justification of the Cascade Scheme of the Subcritical Molten-Salt Reactor for Transmutation of Long-Lived Radioactive Wastes of the Nuclear Fuel Cycle** | **8** | **2001 / 2003** | **L.Ponomarev** | MUCATEX/ JAERI, Tokyo Inst., LANL |
| 2267 | **Construction of the Subcritical Assembly with Combined Neutron Spectra Driven by Proton Accelerator at Proton's Energy 660 MeV for Experiments on Long Lived Fission Products and Minor Actinides transmutation" (Phase I: Design, Design Documentation and Safety Substantiation)** | **6** | **2005 /** | **V.Shvetsov** | JINR / CEA, Royal Inst.; FZK, EURATOM -Ciemat |

# Project Analysis

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|---|---|---|---|---|---|
| 2582 | Experimental Study of Minor Actinides Transmutation Problem at BFS-73-1 Fast Critical Assembly | 6 | 2005 / | A.Kochetkov | IPPE / KAERI, Idaho Natl. Eng. and Envir. Lab. |
| 2680 | Study of Minor Actinide Transmutation in Nitrides: Modelling and Measurements of Out-of-pile Properties | 6 | 2005 / | L.Zabud’ko | IPPE / Kungl Tekniska Hogskolan, CEA |

# Project Analysis

*Proposed projects*

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| 735 | Transmutation of Radioactive and Transuranium Isotopes by High-Energy Neutrons of the Thermonuclear Reactor | 3 | I.Kuzmitsky | VNIIEF / SKB (Swedish Nucl. Fuel and Waste Manadgement Co.) |
| 1755 | Experimental Study of Fast and Fast-Thermal Accelerator Driven Systems on the Basis of BFS-1 – Microtron Complex | 3 | B.Kochurov | ITEP |
| 2661 | Analytical and Experimental Substantiation of Neutron-Physical Characteristics of Fast Reactors with Lead Coolant | 3 | I.Matveenko | IPPE |

# Project Analysis

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| 2884 | **Integral Experiments at BFS Critical Facilities for Justification of Minor Actinides Transmutation and Their Analysis** | **3** | **Yu.Khomiakov** | IPPE |
| 2925 | **Measurement of Transmutation Properties of Minor Actinides Irradiated in Intermediate Reactor Neutron Spectrum** | **3** | **M.Melnik** | RIAR |

# Main Results

The main results of the completed ISTC projects can be summarized as follows.

1. The spectra of prompt spontaneous fission neutrons from $^{240}Pu$ and $^{242}Pu$ as well as fission neutrons from thermal neutron fission of $^{243}Cm$ and $^{245}Cm$ were measured. Also the prompt neutron multiplicity distributions were measured for spontaneous fission of $^{244}Cm$ and 248Cm and for fission of $^{235}U$ and $^{239}Pu$ by thermal neutrons.
2. There were performed for $^{237}Np$ the measurements of integral neutron inelastic scattering cross-sections, fission neutron spectra and mass-energy distributions of fission fragments for energies of emitted neutrons corresponding to first plateau of fission cross-section.
3. In the energy range from 1 to 7 MeV the detailed measurements of fision cross-sections for $^{238}Pu$, $^{242m}Am$, 243, 244, 245, 247Cm were done.

# Main Results (2)

4. The fission cross-sections for neutron induced fission of $^{204, 206, 207, 208}Pb$, $^{209}Bi$ $^{232}Th$, $^{233}U$, $^{237}Np$, $^{239}Pu$, $^{243}Am$ at the energies from reaction threshold up to 200 MeV were measured.
5. The energy dependence change of delayed neutron yields for $^{237}Np$ in the energy range corresponding to first plateau was measured. The analogous experiments are carried out at present for $^{233}U$, $^{232}Th$, $^{239}Pu$, and $^{241}Am$, for $^{239}Pu$ the incident neutron energy range will be expanded up to 18.0 MeV.
6. The systematic measurements and analysis for the basic structural materials of spectra and integral gamma-ray production yields from the interactions with 14-MeV neutrons were performed.
7. The evaluations were done and the general-purpose files were created for isotopes $^{232}Th$, $^{231, 233}Pa$, $^{232, 233, 234, 238}U$, $^{239}Np$, $^{238, 242}Pu$, $^{241, 242g\&m}$, $^{243}Am$, $^{243, 245, 246}Cm$ in the incident neutron energy range up to 20 MeV and for isotopes $^{237}Np$, $^{240}Pu$, $^{241}Am$ at the neutron energies up to 150 MeV.

# Main Results (3)

A large amount of experiments with charged particles was performed:

8. A comparison and verification of the methods for determining reaction product yields resulting from proton-nuclei interaction were made. An experiment was designed and performed whose aim was to irradiate targets made of $^{63}Cu$, $^{65}Cu$ by 1.2 GeV protons with subsequent processing of results independently in ITEP and JAERI. To determine the yields of radioactive product nuclei formed in target and structural materials, various thin targets were irradiated 47 times ($^{182,183,184,186}W$ at the proton energy of 0.2, 0.8 and 1.6 GeV; $^{nat}W$, $^{56}Fe$, $^{58}Ni$, and $^{93}Nb$ at 2.6 GeV; $^{232}Th$, $^{nat}U$, $^{99}Tc$ at 0.1, 0.2, 0.8, 1.2 and 1.6 GeV; $^{59}Co$ and $^{63,65}Cu$ at 0.2, 1.2, 1.6 and 2.6 GeV; $^{nat}Hg$ at 0.1, 0.2, 0.8 and 2.6 GeV and, in addition, $^{208}Pb$ at 1.0 GeV) and 4050 values of cumulative and independent yields of residual radioactive product nuclei with half-lives from 8 minutes to 32 years. The experimental values of nuclear reaction product yields were compared with the values calculated by LAHET, CEM95, CEM2k, CASCADE, CASCADE/INPE, YIELDX and INUCL which model hadron-nuclei interactions.

# Main Results (4)

9. The independent and cumulative yields of radioactive residual product nuclei in thin targets made only of the isotopes of Pb ($^{206,207,208,nat}Pb$) and bismuth ($^{209}Bi$) were determined experimentally and modeled theoretically. Thin targets made of $^{206,207,208,nat}Pb$ and $^{209}Bi$ were irradiated 55 times (at the proton energy of 04, 0.07, 0.10, 0.15, 0.25, 0.4, 0.6, 0.8, 1.2, 1.4, 1.6, 2.6 GeV) and more than 5900 cumulative and independent yields of residual radioactive product nuclei with half-lives from 8 minutes to 32 years were determined. Besides, apart from standard modeling of experimental results by using various codes, some work was done on LAHET and CASCADE modification and new versions LAHETO and CASCADO were developed, respectively.

# Main Results (5)

10. Measurements were made of the secondary neutron spectra from “thin” W – Na targets irradiated by 0.8 and 1.6 GeV protons. Prompt neutron spectra were measured of the fission induced by bombarding isotopes of thorium, uranium and neptunium by 50 and 100 MeV protons. The double differential cross-sections of neutron generation, with due account for errors, are given for 22 groups in the energy interval from 3.10 to 330 MeV. Fission cross—sections were measured for $^{204, 206, 207, 208}Pb$ and for $^{209}Bi$ and $^{205}Tl$. The fission cross-sections were determined for natPb and $^{209}Bi$ (mono-isotope). Absolute neutron- and proton-induced fission cross-sections for natPb and 209Bi have uncertainties about 10%. The data on neutron-induced fission cross-sections for the above nuclides are consistent with those found in other literature, the maximum discrepancy (for 209Bi at 97MeV neutron energy) is about 20 %. The data on proton-induced fission cross-sections are also consistent with the data available in literature.

# Main Results (6)

Among the results achieved in the frameworks of ISTC projects related to integral experiments the following might be highlighted:

11. The effective fission cross-sections and reactivity coefficients for Np-237 and Am-241 at the fast critical assemblies BFS-67, -69 (IPPE) spectra were measured.
12. The effective fission cross-sections for 235U, 237Np, 238Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu, $^{244}$Pu, $^{241}$Am, $^{242m}$Am, $^{243}$Am, $^{243}$Cm, $^{244}$Cm, $^{245}$Cm, $^{246}$Cm, $^{247}$Cm, $^{248}$Cm were measured on the spectra of BIGR reactor, molten salt blankets MSB-1, -2 (VNIIEF) and thermal reactor MAKET (ITEP).
13. The isotopic composition change of U-235, Np-237, Pu-238, -240, Am-241 and Cm-244 samples irradiated in the commercial fast reactor BN-350 with integral fluence ~ 2.2 1023 n/cm$^2$ was obtained.

# Main Results (7)

14. Interesting data were obtained while examining the power density distribution and threshold reaction rates in thick W- and NaCl+$PbCl_2$ targets irradiated by 0.8, 1.0 and 1.2 GeV protons. The investigated parameters are of great importance to the analysis and implementation of ADS projects. The measurement results of the threshold reaction rates for $^{209}$Bi, $^{197}$Au, $^{181}$Ta, $^{169}$Tm, $^{115}$In, $^{nat}$In, $^{93}$Nb, $^{65}$Cu, $^{63}$Cu, $^{64}$Zn, $^{59}$Co, $^{27}$Al, $^{19}$F, $^{12}$C and absorbed doses inside and on the external surface of “thick” W-Na target irradiated by 0.8 GeV protons are presented. The calculation modeling of the measured reaction rates was made by means of the LAHET code used to calculate a stream of particles as well as excitation functions of the nuclides formed available in the MENDL, MENDL2p and IEAF libraries ( in the range of ~100 MeV) and in the LAHET calculations at the energies above 100 MeV. The experimental and calculation results were compared and the most essential discrepancies were analyzed, a further study will be made into the neutron field characteristics on the external surface of the “thick” Pb target irradiated by 0.8 GeV protons.

# Main Results (8)

15. The completed ISTC Project #1486 devoted to investigation of feasibility to create MA burner on the basis of molten-salt cascade subcritical reactor revealed the necessity to improve the MA microscopic data, especially for $^{237}Np$, $^{242m}Am$.

16. The started ISTC Project #2267 (SAD) might identify new requirements for future experiments and data evaluations in the course of Project implementation

# Recommendations for Differential Experiments

From the analysis of completed ISTC projects and present-day requirements to accuracy of nuclear data related to MA transmutation in ADS, it might be concluded that the first priority list of reactions to be studied includes:

- Neutron inelastic scattering on $^{243}$Am;
- Neutron induced fission of $^{244}$Cm above ~200 keV;
- Neutron capture on $^{238}$Pu, $^{237,238}$Np;
- (n,2n) reaction on $^{235}$U, $^{238}$U, $^{239}$Pu;
- (n,xn) reactions on Pb and Bi isotopes in the wide energy range (up to proton beam energies if lead, bismuth or lead-bismuth is assumed to be a spallation target material and coolant).

# Recommendations for Differential Experiments (2)

The ultimate goal of obtaining reliable evaluated data files covering all the needs of ADS-related pending and forthcoming projects would require performing the following experiments:

1. Measurement of the total neutron yield, total neutron spectra and fission neutron spectra for isotopes of Th, Pa, U, Np, Pu, Am, as well as for spallation target materials Ta, W, Pb, Bi, irradiated by neutrons and protons of energies from 20 MeV up to 1 GeV.
2. Measurement of the total gamma yield and emission spectra for isotopes of Th, Pa, U, Np, Pu, Am in the energy region 20 MeV – 1 GeV.
3. Measurement of fission product yields for transuranics at the energies 20 - 200 MeV.

# Recommendations for Differential Experiments (3)

4. Obtaining the excitation functions for reactions (n,xn), (n,pxn), (n,2pxn) etc for isotopes of Th, Pa, U, Np, Pu, Am at the primary neutron energies 20 – 200 MeV as well as for analogous reactions initiated by protons.
5. Measurements of the resistivity damage rates necessary for evaluation of damage energy cross-sections for iron, chromium, nickel and other components of steels at the energies 20 MeV – 1 GeV.

# Recommendations for Differential Experiments (4)

6. Measurements and analysis of the total yields, time and energy dependencies of characteristics of delayed neutrons from fission of Np-237, Am-241, Am-242m by fast neutrons.
7. Carrying out the sensitive measurements of neutron capture and fission cross-sections for minor actinides at resonance and fast neutron energies to define the accuracy of nuclear data.
8. Measurements of excitation functions to obtain secondary reaction alpha- and beta-active product yields from spallation target unit structural materials irradiated by protons and neutrons with energies up to ~ 1 GeV.

# Recommendations for Integral Experiments

Integral experiments as applied to physics of innovative nuclear reactors and ADS is reasonable if the following conditions necessarily satisfied:

- The principal choice of the transmutor type is made, i.e. materials and zone dimensions are specified.
- Material composition and geometry dimensions of characteristic physical zones of an experimental prototype and designed installation should be comparable, i.e. space and energy distribution of neutron fields should be close.

# Recommendations for Integral Experiments (2)

- The verification of nuclear data should be performed on the basis of experimental results and their evaluated version has to be included in the open nuclear data library; or the experimental results with estimated uncertainty and the detailed description of the experimental installation (geometries, material compositions, temperatures) are described (or will be described at the final stage of experiments), which will be used by the developer of the transmutor to verify the calculation methods.

# Recommendations for Integral Experiments (3)

Unfortunately, these conditions were not satisfied in the experimental works performed in the framework of ISTC projects.

It is necessary to perform additional works (if it is possible after several years passed from the experiment completion) on analysis and representation of experimental results. It would be useful to create data bank of evaluated or described in details completed experiments open for general use, at the condition of mutual consent of customers and performers.

# Recommendations for Integral Experiments (4)

- To assure that the future experimental works will not follow the experiments performed, the serious co-ordination of working plans of Projects in the framework of unified scientific and technical policy is necessary, if the latter is a standard practice of ISTC. But before that, it is necessary to formulate such policy, that is to take a decision on the transmuter type) - to formulate the ultimate definite goal. In Russia, the great experience of successful carrying out the huge experimental programs with clear ultimate goal is accumulated.

# Recommendations for Integral Experiments (5)

- If the installation with fast neutron spectrum and heavy metal coolant will be accepted for the purpose of MA transmutation, it is obvious (based on results described in Chapter 3.2 of the Project Final Report) that integral experiments should be accentuated on verification of nuclear data for $^{237}Np$, $^{238,242}Pu$, Am, Cm, new structural materials, lead and bismuth.

# Recommendations for Integral Experiments (6)

These tasks are partially solved in the already formulated Projects:

- creation of special benchmarks to test neutron data for Pb and Bi in the spectrum close to ADS one (ISTC Projects 2661 and 3239);
- creation of benchmarks to test and refine neutron data for MA in the various fast spectra characteristic to ADS with different kinds of coolant and fuel (ISTC Project 2884).

# Recommendations for Integral Experiments (7)

- It is expedient also to realize the integral experiment on the designing of ADS model most close to the commercial installation design, and to study its neutronics characteristics. Experiments should be evaluated, should have detailed description and accordingly on their basis the international data comparison should be done.
- Besides the stated above, the experiments on the MA and Pu samples irradiation in the spectrum close to ADS one might be useful, as well as the analysis of isotopic composition change.

# Recommendations on the Evaluated Data Preparation

The following works on the preparation of new nuclear data files, which are absent at present, or contain the insufficient information on nuclear data, necessary for calculation of transmutation installation:

# Recommendations on the Evaluated Data Preparation (2)

1. Forming the files of recommended neutron and proton data for the total actinide chain from Th to Cm at the energies up to 150-200 MeV with inclusion the information on the secondary gamma-quanta production and complete information on data uncertainties and corresponding correlation matrixes. It is necessary to perform the following tasks:

# Recommendations on the Evaluated Data Preparation (3)

- The estimation of neutron and proton integral and differential cross-sections on the basis of analysis of available experimental data and developed in the framework of ISTC projects new theoretical methods of cross-section description at the intermediate (up to 200 MeV) energies;
- On the basis of energy release balance analysis in the fissionable nuclei and with the use of theoretical models to prepare the consistent sets of the evaluated nuclear data of secondary gamma production for the total number of actinides, relied on available experiments on energy release for the basic fuel elements.
- Creation of consistent correlation uncertainty matrixes of recommended cross-section data for the basic fuel elements;

# Recommendations on the Evaluated Data Preparation (4)

2. Forming the files of recommended data on fission products yields for actinides from Th to Cm at the neutron and proton energies up to 150 – 200 MeV.
3. Specification of radioactive nuclei characteristics in the fission products region on the basis of experimental works performed in Russia during last 15 years, and preparation of corrected files containing the data on radioactive decay modes for essential nuclides.

# CONCLUSION

The analysis of the ISTC Projects associated with accelerator-driven waste transmutation reveals the following important issues to be outlined

- The main bulk of the ISTC supported activity deals with improvement of nuclear data for transuranics isotopes (differential experiments: cross-sections in a broad energy interval, neutron yields, etc) and their effect on the integral characteristics of the facilities (integral experiments).

# CONCLUSION (2)

- The nuclear data obtained are significant only at the early stages of transmutation i.e. for the short irradiation time.
- In the foreseen Accelerator-Driven Systems concepts, the equilibrium transmutation cycle is characterized by dominant role of Pu-238 (about 25% in the isotopic mixture of all the heavy isotopes in the blanket). Uncertainty in its capture cross-section is 20% while the required accuracy is 10%.

# CONCLUSION (3)

- The similar characteristics for Am-241 are 10 and 5%, correspondingly.
- It seems to be important to improve nuclear data particularly for Pu-238 and adjacent isotopes Np-237 and Np-238.
- Experimental data on the yield of residual nuclides accumulated in spallation target (spallation products) were obtained mainly for the short-lived nuclides characteristic to the small proton fluence.

# CONCLUSION (4)

- Accumulation of long-lived rare earth nuclides was out of consideration. Nevertheless some of these nuclides (**like for example $^{146}Sm$ ($T_{1/2}=1\times10^{8}$ yrs), $^{148}Gd$ ($T_{1/2}=74.6$ yrs), $^{150}Gd$ ($T_{1/2}=1.8\times10^{6}$ yrs) , $^{154}Dy$ ($T_{1/2}=3\times10^{6}$ yrs)** ) appear to be of high toxicity due to their alpha decay mode.
- In addition to that some of them are known to be neutron poisons with anomalously large neutron capture cross-sections. It seems to be of importance to identify the domains of proton energies and neutron spectra in which their accumulation could significantly affect the safety of accelerator-driven transmuters.

# CONCLUSION (5)

- There has been observed a large discrepancy in cross-sections between neutron induced ((n,xn), (n,pxn), (n,2pxn), etc) and similar proton-induced reactions for transuranics , estimated by various models in the energy interval 20 – 150 MeV.  It seems to be instructive to conduct the relevant experimental studies for isotopes of Th, U, Np, Pu, Am to validate theoretical models and computer codes. For the first priority one could reasonably put the reactions producing long-lived toxic nuclides like $^{238}U(n,7n)^{232}U$, for example.

# CONCLUSION (6)

- The endurance of structural materials under irradiation might happen to be of crucial importance in justification of the transmutation strategy. Currently, the accumulated damage dose (in terms of displacements-per-atom), characteristics of gas accumulation is predicted with the accuracy of 15%. This accuracy was well assumed for the conventional reactors at the early stages of their development. At the energies of about 1 GeV uncertainties in these characteristics approach to 25 – 50%.  Needless to say, this gives a rather shaky estimation of the lifetime of structural components in the spallation target region. In this connection, it seems to be instructive to improve the data base for prediction of damage dose and gas accumulation.

**Obninsk State Technology University for Nuclear Power Engineering has a number of codes and libraries for calculations and evaluations of nuclear data for transport, heating, shielding and medical applications.**

**Tel: +7 (48439) 3-83-20**

**Fax: +7 (48439) 3-83-20**

**E-mail: korovin@iate.obninsk.ru**