VALIDATION OF MINOR ACTINIDES FISSION NEUTRON CROSS-SECTIONS

by

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Verification of neutron fission cross-sections of minor actinides from some recently available evaluated nuclear data libraries was carried out by comparison of the reaction rates calculated by the MCNP6.1 computer code to the experimental values. The experimental samples, containing thin layers of ^{235}U, ^{237}Np, ^{238,239,240,241}Pu, ^{242m}Am, ^{243}Cm, ^{245}Cm, and ^{247}Cm, deposited on metal support and foils of ^{235}U (pseudo-alloy ^{27}Al + ^{235}U), ^{238}U, ^{nat}In, ^{64}Zn, ^{27}Al, and multi-component sample alloy ^{27}Al + ^{55}Mn + ^{nat}Cu + ^{nat}Lu + ^{197}Au, were irradiated in the channels of the tank containing fluorine salts 0.52NaF + 0.48ZrF$_4$, labelled as the Micromodel Salt Blanket, inserted in the lattice centre of the MAKET heavy water critical assembly at the Institute for Theoretical and Experimental Physics, Moscow. This paper is a continuation of earlier initiated scientific-research activities carried out for validation of the evaluated fission cross-sections of actinides that were supposed to be used for the quality examination of the fuel design of the accelerator driven systems or fast reactors, and consequently, determination of transmutation rates of actinides, and therefore, determination of operation parameters of these reactor facilities. These scientific-research activities were carried out within a frame of scientific projects supported by the International Science and Technology Center and the International Atomic Energy Agency co-ordinated research activities, from 1999 to 2010. Obtained results confirm that further research is needed in evaluations in order to establish better neutron cross-section data for the minor actinides and selected nuclides which could be used in the accelerator driven systems or fast reactors.

Key words: nuclear data library, accelerator driven system, fast reactor, actinide, reaction rate, MAKET, MCNP6.1

INTRODUCTION

Accelerator driven subcritical systems (ADS) and fast reactors (FR) are studied extensively in last decades as a promising option for nuclear power production and nuclear waste reduction by transmutation. Use of minor actinides (MA) in fuel design for the ADS and FR, or for nuclear waste elimination through nuclides transmutation in these nuclear reactors, requires knowledge of the MA neutron cross-sections with different accuracy, depending on the MA usage. If the MA are used for the fuel design (*i. e.*, added to the ADS or FR fuel elements in a large percent), their neutron cross-sections must be known with great accuracy, as they are known for the fuel elements' material (^{235}U or ^{239}Pu), *i. e.*, the uncertainty must be very low, less than 0.5%. If the MA are used for the waste side transmutation in (blankets of) the ADS or FR, the MA were supposed to be added to the ADS or FR fuel elements only in few percents, and hence the uncertainty of the MA neutron cross-sections is acceptable in a range of 5% to 10%. The higher uncertainty of the MA neutron cross-sections will be acceptable for studies of the irradiation properties of the nuclear spent fuel of the thermal nuclear reactors, fast nuclear reactors or ADS. In such cases, the uncertainty of neutron cross-sections up to 10% is acceptable for ^{237}Np nuclide, from 15% to 20% for Am isotopes and from 30% to 50% for Cm isotopes. These requirements [1] for the uncertainties of the MA neutron cross-sections initiated various researches on experimental activities and evaluation studies on neutron cross-sections of the MA and some other isotopes used for different purposes in nuclear reactors and ADS. A recent study of the MA cross-sections status and requirements for better nuclear data is given in [2].

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One of the experimental researches of the MA neutron cross-sections was carried out at the MAKET thermal critical assembly [3] at the Institute for Theoretical and Experimental Physics (ITEP) within frame of the International Science and Technology Center (ISTC) project #1145 [1]. This experimental research and numerical studies of the MA and selected nuclides (SN) reaction rates were the basic for a portion devoted to nuclear data of the International Atomic Energy Agency (IAEA) coordinated research project (CRP) on the Analytical and Benchmark Analyses of Accelerator Driven Systems. In this IAEA CRP [4], the reaction rates (RR) of the MA and some SN were calculated using older versions (4B2 and 5.1-6) of the MCNP computer code [5] and the calculation results of the MA and SN reaction rates were compared to the experimentally determined reaction rates of the MA and SN.

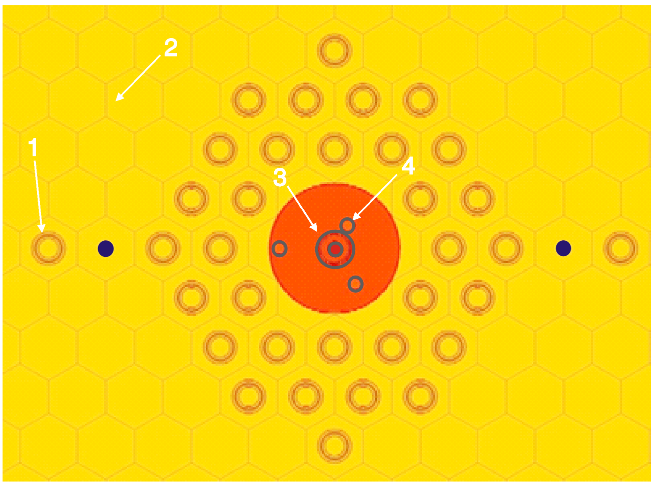
For the calculations of the MA and SN reaction rates in this IAEA CRP, the MCNP computer code was used with neutron cross-sections of the MA and SN prepared in the ACE format from the older evaluated nuclear data libraries: JENDL-3.2 [6], ENDF/B-VI (releases 0/1 and 7, [7]) and LLLDOS [8]. The ACE formatted nuclear data from some newer available evaluated nuclear data libraries were used in the IAEA CRP in 2014 as well as for: ENDF/B-VII.0 [9], ENDF/B-VII.1 [10], JEFF-3.1.1 [11], and JEFF- 3.2 [12].

The purpose of this study was to compare calculated results of the MA and SN reaction rates of several newer and dedicated evaluated neutron cross-sections data libraries to the experimental reaction rates results obtained for the same MA and SN irradiated at the MAKET assembly. To achieve this goal, the new (the last available production) version (6.1, released by the LANL in 2013) of the MCNP computer code [13] was used with the ACE type of neutron cross-sections created from the evaluated nuclear data libraries, issued by different nuclear data centres in a few last years: ENDF/B-VII.1 (LANL, 2012 [10]), JEFF-3.2 (OECD/NEA, 2014 [12]), TENDL-2014 (NRG, 2014 [14]), ADS-2.0 (IAEA, 2008 [15]), IRDF-2002 (IAEA, 2002 [16]), IRDFF-v-1.05 (IAEA, 2015 [17]), ROSFOND (CND, 2010 [18]), and JENDL-4.0up3 (JAEA, 2015 [19]). Some old evaluated data for the MA, like the ones known as "Maslov" (evaluated in 1994-6 by Maslov *et al.*, [20]) are not examined entirely in this study since these evaluations were being incomplete (in the number of isotopes) and that part of these evaluations was included in the JENDL-3.2 library.

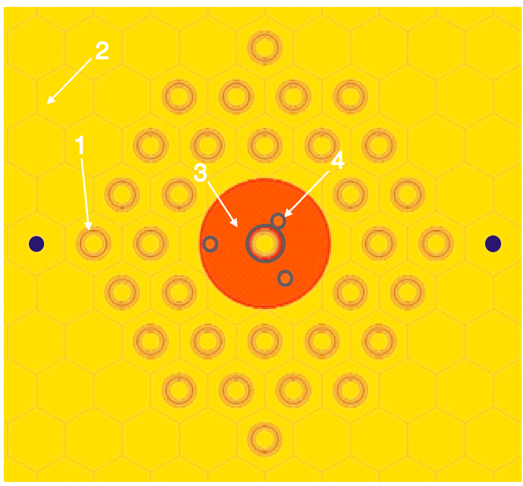
MAKET AND EXPERIMENTAL DATA

The detailed descriptions of the MAKET critical assembly and the MA and SN samples preparation, the samples characteristic, irradiation and the MA and SN reaction rates measurements were given in [1, 3]. The data provided in this article are shown only in order to provide a brief insight in the complexity of the MA and SN reaction rates experiments carried out at the MAKET assembly.

The MAKET heavy water critical assembly was designed [3, 21] to operate with the dispersed (in aluminum) tubular TVR-S type (90%) HEU (with aluminum clad) fuel elements placed in a hexagonal lattice of the core. Horizontal cross-sections of the MAKET core fuel lattices labelled as the 21-1-5(M2) and 21-2, used for irradiation of the samples (irradiation channels marked as small circles of dark colour, (4)) are extracted from the MAKET computational MCNP code three-dimensional (3-D) geometry and material (numerical) model and shown in fig. 1.



21-1-5(M2) lattice



21-2 lattice

Figure 1. A zoom in the horizontal cross-sections of the MAKET core fuel lattices; (1) – fuel elements, (2) – heavy water moderator, and (3) – SBM with samples positions (4)

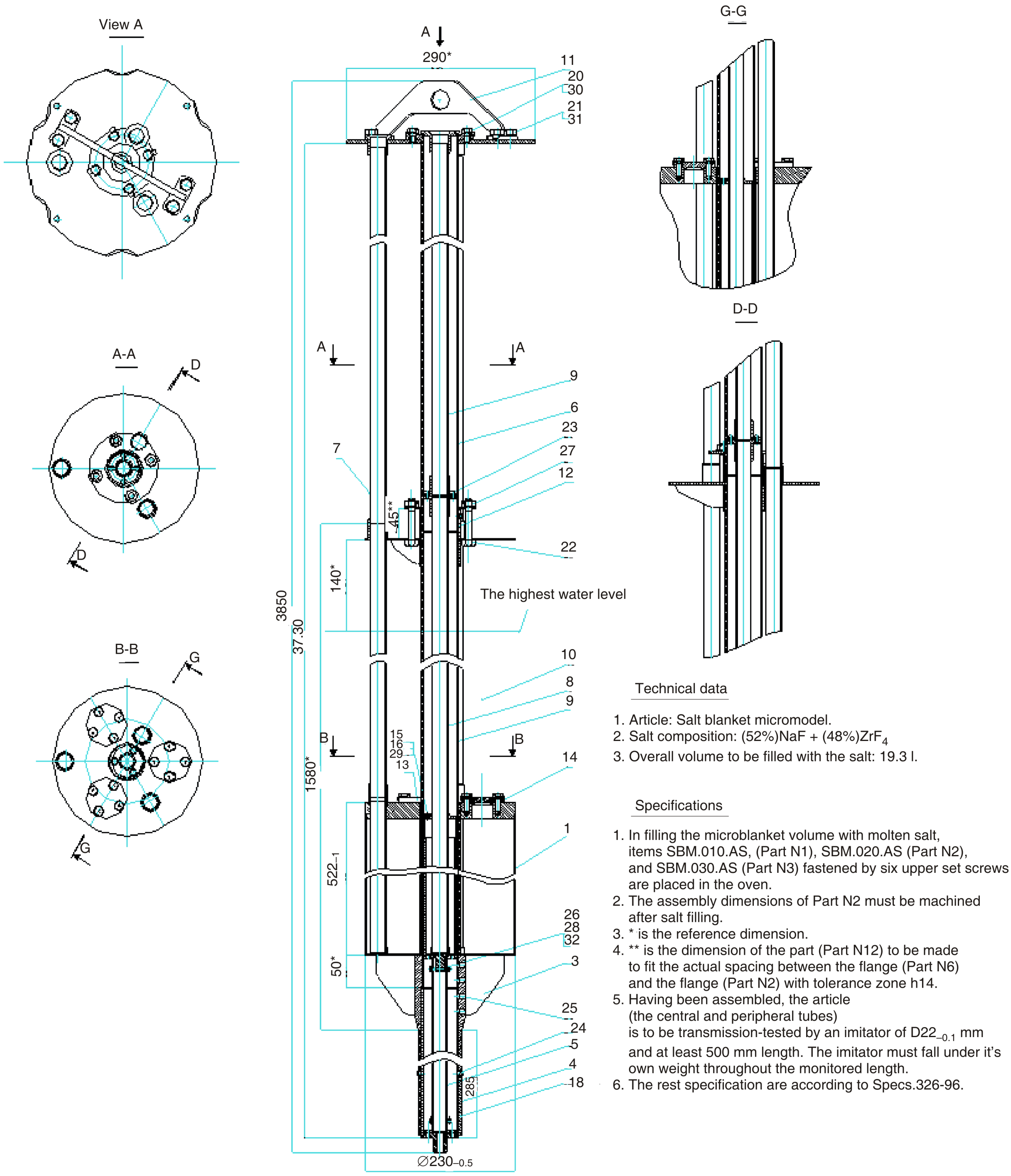


Figure 2. The SBM cross-sections

The MA experimental samples, containing thin layers of the ^{235}U, ^{237}Np, ^{238,239,240,241}Pu, ^{242m}Am, ^{243}Cm, ^{245}Cm, ^{247}Cm, deposited on metal support and SN foils of ^{235}U (pseudo-alloy ^{27}Al + ^{235}U), ^{238}U, ^{nat}In, ^{64}Zn, ^{27}Al, MCS (alloy ^{27}Al + ^{55}Mn + ^{nat}Cu + ^{nat}Lu + ^{197}Au) [1] were irradiated in dry channels (small dark circles (4) in fig. 1) of the micromodel salt blanket (SBM) inserted in the lattice centre of the MAKET critical facility. Cross-sections of the SBM are shown in fig. 2. The method of the solid state nuclear track detector (SSNTD) was used for measurements of the MA fission reaction rates. The gamma-spectroscopy method was used for measurements of the reaction rates of other SN: ^{235}U(n,f), ^{238}U(n,γ), ^{55}Mn(n,γ), ^{63}Cu(n,γ), ^{197}Au(n,γ), ^{176}Lu(n,γ), ^{115}In(n,n'), ^{27}Al(n,α), ^{64}Zn(n,p), and ^{64}Zn$(n,2n)$.

The MAKET critical assembly has operated for 2 hours, during the SN samples irradiation, or for 40 minutes during the MA samples irradiation. The MAKET steady fission power, during samples irradia-

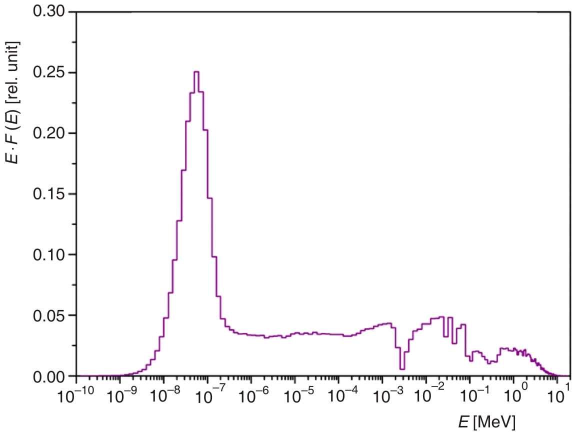


Figure 3. Normalized neutron spectrum per lethargy unit at the sample '667' position (the central channel) at the MAKET core lattice 21-1-5(M2)

tion, was (81.4   1.7) W and (78.8   1.6) W, for the core lattices 21-1-5(M2) and 21-2, respectively [1]. The MCNP calculations have shown that the neutron flux density energy distribution (neutron spectrum) had very similar shape for different positions at the MAKET core lattices used for irradiation of the samples [1]. The normalized neutron spectrum per unit of lethargy, $E{\cdot}F(E)$, determined in 176 neutron energy groups in range of 10   eV to 18 MeV, calculated by the MCNP6.1 code for the MAKET core lattice 21-1-5(M2) and fission power of 81.4 W, is given in fig. 3. The neutron spectrum was obtained after the MCNP6.1 code run for 150 million neutron histories, at the sample position in the central channel at the MAKET core (labelled also the '667') in the 21-1-5(M2) lattice. Such number of neutron histories produced calculated group neutron spectrum (MCNP F4 tally) with the statistical relative 1   uncertainty below 5% in all energy groups between 2.5 meV and 2 MeV. This neutron spectrum is in well agreement with the neutron spectrum determined by the MCNP4B2 code using ACE type older neutron cross-section data of the endf60 (*.60c) and the tmccs (*.01t) libraries [1].

The measured (experimental) values of the reaction rates of the MA and SN of the irradiated samples are given in [1] and [3] and normalized to the MAKET fission energy unit [Ws] and the nuclide sample unit of mass [g], before the comparison to the calculated values of the reaction rates of the same nuclide. Table 1 shows the normalized values of the samples nuclide reaction rates determined by the SSNTD (silicate glass and polycarbonate thin film) method [1] after the samples of the MA and SN irradiation at the various positions at the MAKET core lattices. The irradiation positions were labelled as the '667' = the central channel (0 mm), '327' = 1st channel (46.6 mm), '337' = 2nd channel (72.0 mm), and '317' = 3rd channel (96.5 mm). The dimensions associated to the channels above show samples positions at the corresponding radii of the SBM devices (containers) inserted in the MAKET core lattice [1]. The normalization formula, used for the reaction rate of the sample isotope k, $RR(k)$, is given in [1] as

$$RR(k)^{\rm exp}_{\rm norm} \quad RR(k)^{\rm exp}_{\rm meas} \frac{N_{\rm Av}}{A_k} \frac{1}{P}$$

In the normalization process the following values, beside the MAKET assembly fission generated power P (given in W), were used: Avogadro's number $N_{\rm Av}$ = 6.023·10^{23} mol^{-1}; the measured reaction rate $RR(k)_{\rm meas}$ (given in s^{-1}, [1]) and the sample isotope k atomic mass A_k (given in g mol^{-1}). The values of Avogadro number and the atomic masses of the isotopes were taken from [22]. The isotopic composition of the samples was determined by the mass spectrometry (before the samples irradiation) and shown in the atom fractions a_k [1].

CALCULATIONS

Various versions of the MCNP computer code, with different neutron ACE type cross-section libraries, were extensively used at the Vinča Institute of Nuclear Sciences for validation of diverse models of complex heavy water systems, *e. g.* [23], since late 1970s. The calculation of the MA and SN reaction rates in this study were carried out by the 6.1 version of the MCNP computer code, released by the LANL in 2013. A detailed 3-D geometry and materials model of the MAKET assembly and the SBM samples is created as credible as possible [1] and the assembly materials are described in the MCNP6.1 using endf60 (*.60c) and Sab2002 (*.60t) ACE type neutron cross-section data. This MAKET 3-D model was verified in the extensive validation calculations against different MAKET core configurations [1].

The irradiated samples were modelled in their containers. For all the samples it was possible only to model and simulate (*i. e.*, calculate) the MA and the SN reaction rates in a homogeneous region, describing the sample position by the MCNP F4 neutron flux tally modified (folded) by the response function (*i. e.*, neutron cross-sections) of the required reaction rate.

In this study, the neutron cross-sections of the MA and the SN in the irradiated samples were taken using the following ACE type nuclear data libraries:

– endf71 (*.80c), based on the LANL evaluation of the ENDF/B-VII.1 file, released in 2013,
– jeff3.2 (*.03c), based on the OECD/NEA evaluation of the JEFF-3.2 file, released in 2014,
– tendl2014 (*.00c), based on the NRG evaluation of the TALYS-1.6, released in 2014,
– ads20 (*.70c), the IAEA processed and dedicated library for the ADS, ADS-2.0, released in 2008,
– irdf2002 (*.34y), the IAEA processed and dedicated reactor dosimetry library, IRDF-2002, released in 2002, and

Table 1. Normalized measured nuclides reaction rates (RR) values (relative uncertainty of the RR values is in range of 2% to 5%)

Lattice	21-1-5(M2)				21-2		
Power	(81.4 1.7) W				(78.8 1.6) W		
Position	'667'	'327'	'337'	'317'	'327'	'337'	'317'
Nuclide RR	Normalized measured nuclides reaction rates RR [$W^{-1}s^{-1}g^{-1}$]						
$^{235}U(n, f)$	$1.608 \cdot 10^7$	$1.690 \cdot 10^7$	$1.610 \cdot 10^7$	$1.569 \cdot 10^7$	$1.501 \cdot 10^7$	$1.877 \cdot 10^7$	$2.010 \cdot 10^7$
$^{237}Np(n, f)$	$6.611 \cdot 10^3$				$1.202 \cdot 10^4$		
$^{238}Pu(n, f)$	$6.043 \cdot 10^5$				$6.001 \cdot 10^5$		
$^{239}Pu(n, f)$	$2.369 \cdot 10^7$	$2.584 \cdot 10^7$	$2.235 \cdot 10^7$	$2.442 \cdot 10^7$	$2.733 \cdot 10^7$	$2.363 \cdot 10^7$	$2.906 \cdot 10^7$
$^{240}Pu(n, f)$	$1.221 \cdot 10^5$				$1.254 \cdot 10^5$		
$^{241}Pu(n, f)$	$2.718 \cdot 10^7$				$2.648 \cdot 10^7$		
$^{242m}Am(n, f)$	$1.962 \cdot 10^8$				$2.067 \cdot 10^8$		
$^{243}Cm(n, f)$	$1.991 \cdot 10^7$				$2.201 \cdot 10^7$		
$^{245}Cm(n, f)$	$4.369 \cdot 10^7$				$4.357 \cdot 10^7$		
$^{247}Cm(n, f)$	$7.319 \cdot 10^6$				$8.396 \cdot 10^6$		
$^{238}U(n, \gamma)$	$2.769 \cdot 10^5$	$2.837 \cdot 10^5$	$2.906 \cdot 10^5$	$2.943 \cdot 10^5$	$2.915 \cdot 10^5$	$3.243 \cdot 10^5$	$3.188 \cdot 10^5$
$^{55}Mn(n, \gamma)$	$1.697 \cdot 10^6$	$1.724 \cdot 10^6$	$1.764 \cdot 10^6$	$1.845 \cdot 10^6$	$1.642 \cdot 10^6$	$1.697 \cdot 10^6$	$1.878 \cdot 10^6$
$^{63}Cu(n, \gamma)$	$5.291 \cdot 10^5$	$5.209 \cdot 10^5$	$5.526 \cdot 10^5$	$5.420 \cdot 10^5$	$4.737 \cdot 10^5$	$4.822 \cdot 10^5$	$5.393 \cdot 10^5$
$^{197}Au(n, \gamma)$	$7.363 \cdot 10^6$	$7.438 \cdot 10^6$	$7.551 \cdot 10^6$	$7.739 \cdot 10^6$	$7.489 \cdot 10^6$	$7.567 \cdot 10^6$	$8.382 \cdot 10^6$
$^{176}Lu(n, \gamma)$	$1.741 \cdot 10^8$	$1.758 \cdot 10^8$	$1.771 \cdot 10^8$	$1.842 \cdot 10^8$	$1.677 \cdot 10^8$	$1.720 \cdot 10^8$	$1.894 \cdot 10^8$
$^{68}Zn(n, \gamma)$	$1.020 \cdot 10^4$	$1.176 \cdot 10^4$	$1.100 \cdot 10^4$	$1.155 \cdot 10^4$	$1.089 \cdot 10^4$	$1.159 \cdot 10^4$	$1.294 \cdot 10^4$
$^{27}Al(n, \alpha)$	$1.453 \cdot 10^1$	$2.155 \cdot 10^1$	$1.804 \cdot 10^1$	$2.304 \cdot 10^1$	$3.059 \cdot 10^1$	$2.368 \cdot 10^1$	$2.742 \cdot 10^1$
$^{64}Zn(n, p)$	$2.789 \cdot 10^2$	$2.917 \cdot 10^2$	$3.380 \cdot 10^2$	$3.889 \cdot 10^2$	$6.660 \cdot 10^2$	$5.189 \cdot 10^2$	$5.189 \cdot 10^2$
$^{64}Zn(n, \gamma)$	$8.403 \cdot 10^4$	$8.507 \cdot 10^4$	$8.704 \cdot 10^4$	$9.282 \cdot 10^4$	$8.537 \cdot 10^4$	$8.847 \cdot 10^4$	$9.517 \cdot 10^4$
$^{115}In(n, n')$	$9.852 \cdot 10^2$	$1.082 \cdot 10^3$	$1.159 \cdot 10^3$	$1.146 \cdot 10^3$	$2.242 \cdot 10^3$	$1.610 \cdot 10^3$	$1.849 \cdot 10^3$
$^{115}In(n, \gamma)$	$8.756 \cdot 10^6$	$9.080 \cdot 10^6$	$9.466 \cdot 10^6$	$9.981 \cdot 10^6$	$8.382 \cdot 10^6$	$8.980 \cdot 10^6$	$9.645 \cdot 10^6$
$^{115}In(n, 2n)$	$3.278 \cdot 10^4$	$3.336 \cdot 10^4$	$3.413 \cdot 10^4$	$3.419 \cdot 10^4$	$5.947 \cdot 10^4$	$5.947 \cdot 10^4$	$6.100 \cdot 10^4$

Note: Empty cells in the table mean that measurements of these RR were not done

– irdff-v-1.05 (*.10y), the IAEA processed and dedicated reactor dosimetry and fusion library, IRDFF-v-1.05, released in 2015.

Additionally, ACE type neutron cross-section libraries, at the neutron temperature of 293.6 K, only for the MA isotopes analysed in this study, were prepared from:

– Japanese JENDL-4.0 (update 3, 2015, *.88c),
– Maslov (rev. 2003, *.20c), and
– Russian ROSFOND-2010 (2010, *.99c),

using the NJOY99 (version 396) computer code [24]. The ACE cross-sections were generated in resolved resonance range using fractional tolerance of 0.001 for thinning, fractional integral tolerance of 0.003 and probability tables in unresolved resonance range.

These ACE type nuclear cross-section libraries are processed from the evaluated nuclear data at various neutron temperatures. The ACE type libraries used in this validation study were selected at almost the same temperature values: 293.6 K (= 25.301 meV) for the endf71, jeff3.2, and ads2.0; 293.1 K (= 25.263 meV) for the tendl2014 or 300.0 K (= 25.852 meV) for the irdff-v1.05. Only the ACE type nuclear data library irdf2002 was released at 0 K neutron temperature. Additionally, the MCNP6.1 code input was modified by the cell free-gas thermal temperature TMP card (affects only the neutron elastic scattering cross-sections) with the value of a 24.46 meV (= 10.71 ºC) for the both MAKET lattices 21-1-5(M2) and 21-2, which correspond to the experimental temperatures measured within 1.5 ºC. It is judged that a small difference in neutron temperatures of ACE type nuclear cross-section libraries does not influence significantly the obtained calculation results.

All reaction cross-sections of the MA and the SN, which were examined, have been found in the endf71, jeff32, and tendl2014 ACE type nuclear libraries, as well in the ROSFOND-2010 and JENDL-4.0up3 libraries. However, some reaction cross-sections of the MA and the SN are missing in the ads20, irdf2002, irdff-v-1.05, and Maslov ACE type nuclear data libraries and no alternatives were used in the simulation. This information is shown as the '–' character in tab. 2 (and tab. 4), while the MA and the SN reaction cross-sections existence in these ACE type libraries is marked with the '+' character in tab. 2. Obviously, the calculation results for the selected reaction rates in the cases of the missing nuclide reaction cross-sections were zero.

The MCNP6.1 computer code was run in the MODE N for 50 million neutron histories (KCODE option with 50 000 neutron histories in 1000 active cycles, after 100 initial cycles). Such MCNP6.1 code run enabled that the statistical relative 1 uncertainties of

Table 2. Missing reactions cross sections of the MA and SN in the ads20, irdf2002, irdff-v-1.05 and Maslov ACE type libraries

ACE library / Nuclide reaction	ads20	irdf2002	irdff-v1.05	Maslov
^{238}Pu(n, f)	–	–	–	+
^{240}Pu(n, f)	–	–	–	–
^{241}Pu(n, f)	+	–	–	–
^{242}Pu(n, f)	+	–	–	+
^{244}Pu(n, f)	–	–	–	–
^{242m}Am(n, f)	+	–	–	+
^{243}Am(n, f)	+	–	–	+
^{243}Cm(n, f)	–	–	–	+
^{244}Cm(n, f)	+	–	–	–(*)
^{245}Cm(n, f)	+	–	–	+
^{246}Cm(n, f)	–	–	–	+
^{247}Cm(n, f)	+	–	–	–
^{248}Cm(n, f)	–	–	–	–
^{176}Lu(n, γ)	+	–	–	–
^{68}Zn(n, γ)	–	–	–	–
^{64}Zn(n, p)	–	+	+	–
^{64}Zn(n, γ)	–	–	–	–
^{115}In(n, n')	+	–	–	–
^{115}In(n, γ)	+	–	+	–
^{115}In($n, 2n$)	+	–	–	–
^{113}In(n, γ)	–	–	+	–

Note: (*) NJOY99.396 code failed to process ^{244}Cm Maslov endf/b file

the (MCNP F4 tally) determined reaction rates (RR) of the MA and the SN at all the irradiation positions of the samples at the MAKET assembly lattices were in the range from 0.5% to 5%, except for the ^{27}Al(n, α) reaction rates, for which the statistical relative 1σ uncertainties were in the range from 14% to 19%.

The MCNP6.1 code calculated values of the reaction rates of the MA and the SN were normalized to the MAKET fission energy unit [Ws] and the nuclide sample unit of mass [g], before the comparison to the experimental normalized reaction rate values (tab. 1) was done. In this normalization process, the following values were used:

– Avogadro's number $N_{Av} = 6.023 \cdot 10^{23}$ mol^{-1},
– the energy per fission E_f = 194.0833 MeV = = $3.10956 \cdot 10^{-11}$ Ws [1],
– the number of neutrons lost per fission (*i. e.*, the total number of fission events normalized per a single fission neutron) in the MAKET core (taken from the MCNP6.1 code output) lpf = 0.41634 ($1/\upsilon = 1/2.433$) for the lattice 21-1-5(M2) or lpf = 0.41591($1/\upsilon = 1/2.432$) for the lattice 21-2, and
– the atomic mass A_k (given in g mol^{-1}) of the isotope k.

The values of Avogadro number and the atomic masses of the isotopes were taken from [22]. Since the reaction rates calculated in the MCNP6.1 code runs were obtained normalized per fission neutron and per atom, the post-calculation normalization process was included, also the corrections to the isotopic composition of the real MA and nuclide samples (given as the atomic fraction a_k, [1]) and the MCNP microscopic cross-section units (1 barn = 10^{-24} cm^2). The normalization formula, for the reaction rate of the sample isotope k, $RR(k)$, is given in [1] as

$$RR(k)^{\rm calc}_{\rm norm} = \sum_{j=1}^{n} RR(k)^{\rm calc}_{\rm MCNP}\, a_{\rm kj} \frac{N_{\rm Av}}{A_{\rm kj}} \frac{10^{24}}{E_f\, lpf}$$

where j = 1... n is the number of isotopes mixed with isotope k, and $\sum_{j=1}^{n} a_{\rm kj} = 1$.

Due to a large number of the calculations data, the normalized results of the MCNP6.1 calculations of the reaction rates (of the MA and the SN) are given in tab. 3 only for the neutron cross-sections of the endf71 ACE type nuclear data library. Therefore, the normalized results of the MCNP6.1 calculation reaction rates (of the MA and the SN) for neutron cross-sections of the jeff32, tendl2014, irdf2002, irdff-v-1.05, jendl40, Maslov, and rosfond2010 ACE type nuclear data libraries are not given in this article.

RESULTS AND CONCLUSIONS

In aim to compare the normalized MCNP6.1 code calculated reaction rates to the normalized measured reaction rates for the isotope k, the simulation-to-experiment (*i. e.*, the calculations-to-measurements) mean squared deviation factor, labelled as the $\langle F_k \rangle$, was calculated according to the formula (as given in [1])

$$\langle F_{\rm k} \rangle = 10^{\sqrt{\left\langle \left(\log \frac{RR(k)^{\rm calc}_{\rm norm}}{RR(k)^{\rm exp}_{\rm norm}} \right)^2 \right\rangle}}$$

where < > represents averaging over all measured (experimental) and calculated normalized reaction rate data values used in the comparison. Obviously, the missing correspondent reaction rates data (either the experimental or calculated) of the nuclides were not included in the comparison. The calculated values of the mean squared deviation factor $\langle F_k \rangle$, according to the formula above, are given in tabs. 4 and 5.

The calculated mean squared deviation normalization factors $\langle F_k \rangle$ with values within 5% to 7% difference are considered practically equal [1, 4], due to values of the uncertainties of the measured (experimental) and simulated (calculated) reaction rates of the MA and the SN. The values of the $\langle F_k \rangle$ factor, given in tabs. 4 and 5, show that the difference of the experimental and the simulated reaction rates of MA are in the range of 1% to 63%. The reaction rates difference for the SN with well known cross-sections are in the range of 0% to 66%, except for the reaction rates of ^{115}In(n, n'), ^{68}Zn(n, γ), and ^{115}In($n, 2n$) plus ^{113}In(n, γ).

Therefore, the following conclusions about neutron cross-sections data of the MA and the SN, beside a

Table 3. Normalized calculated values[(1)] of the reaction rates (RR) of the MA and SN for the endf71 ACE type neutron cross-section data library

Lattice	21-1-5(M2)				21-2		
Position	'667'	'327'	'337'	'317'	'327'	'337'	'317'
Nuclide RR	Normalized calculated nuclides reaction rates RR [$W^{-1}s^{-1}g^{-1}$]						
^{235}U(n, f)	1.406E+07	1.458E+07	1.518E+07	1.600E+07	1.408E+07	1.508E+07	1.626E+07
^{237}Np(n, f)	5.450E+03	5.889E+03	6.133E+03	6.695E+03	1.066E+04	8.689E+03	8.243E+03
^{238}Pu(n, f)	5.957E+05	6.118E+05	6.377E+05	6.705E+05	6.103E+05	6.460E+05	6.896E+05
^{239}Pu(n, f)	2.276E+07	2.325E+07	2.428E+07	2.528E+07	2.306E+07	2.426E+07	2.615E+07
^{240}Pu(n, f)	1.245E+05	1.281E+05	1.331E+05	1.391E+05	1.315E+05	1.361E+05	1.455E+05
^{241}Pu(n, f)	2.768E+07	2.846E+07	2.973E+07	3.111E+07	2.789E+07	2.958E+07	3.191E+07
^{242}Pu(n, f)	3.780E+03	4.071E+03	4.436E+03	4.747E+03	8.043E+03	6.474E+03	5.937E+03
^{244}Pu(n, f)	2.835E+03	3.101E+03	3.333E+03	3.646E+03	6.523E+03	5.107E+03	4.651E+03
^{241}Am(n, f)	1.140E+05	1.154E+05	1.204E+05	1.252E+05	1.217E+05	1.238E+05	1.311E+05
^{242m}Am(n, f)	1.469E+08	1.518E+08	1.583E+08	1.661E+08	1.472E+08	1.573E+08	1.694E+08
^{243}Am(n, f)	8.297E+03	8.722E+03	9.079E+03	1.661E+08	1.256E+04	1.134E+04	1.091E+04
^{243}Cm(n, f)	1.668E+07	1.711E+07	1.776E+07	1.857E+07	1.691E+07	1.788E+07	1.910E+07
^{244}Cm(n, f)	4.195E+04	4.430E+04	4.496E+04	4.779E+04	5.106E+04	4.970E+04	5.065E+04
^{245}Cm(n, f)	4.335E+07	4.508E+07	4.698E+07	4.965E+07	4.329E+07	4.657E+07	5.029E+07
^{246}Cm(n, f)	1.014E+04	1.057E+04	1.101E+04	1.160E+04	1.538E+04	1.355E+04	1.340E+04
^{247}Cm(n, f)	5.622E+06	5.684E+06	5.920E+06	6.204E+06	5.746E+06	6.047E+06	6.409E+06
^{248}Cm(n, f)	1.406E+07	1.458E+07	1.518E+07	1.600E+07	2.338E+04	2.180E+04	2.042E+04
^{238}U(n, f)	1.722E+04	1.766E+04	2.004E+04	1.976E+04	1.484E+03	1.150E+03	1.082E+03
^{238}U(n, γ)	5.968E+02	6.933E+02	7.754E+02	8.581E+02	6.488E+05	6.610E+05	7.055E+05
^{55}Mn(n, γ)	6.325E+05	6.794E+05	5.819E+05	6.525E+05	1.502E+06	1.600E+06	1.716E+06
^{63}Cu(n, γ)	1.490E+06	1.536E+06	1.597E+06	1.674E+06	4.472E+05	4.698E+05	5.096E+05
^{197}Au(n, γ)	4.357E+05	4.538E+05	4.744E+05	4.922E+05	7.000E+06	7.591E+06	7.839E+06
^{176}Lu(n, γ)	6.834E+06	6.853E+06	7.038E+06	7.073E+06	1.435E+08	1.519E+08	1.622E+08
^{68}Zn(n, γ)	1.419E+08	1.453E+08	1.518E+08	1.576E+08	1.199E+05	1.244E+05	1.310E+05
^{32}S(n, p)	1.129E+05	1.160E+05	1.203E+05	1.268E+05	2.069E+03	1.572E+03	1.609E+03
^{58}Ni(n, p)	8.215E+02	9.779E+02	1.116E+03	1.244E+03	1.775E+03	1.348E+03	1.370E+03
^{27}Al(n, α)	7.074E+02	8.396E+02	9.567E+02	1.063E+03	2.019E+01	1.649E+01	1.721E+01
^{19}F(n, 2n)	9.044E+00	1.166E+01	1.395E+01	1.348E+01	5.552E-03	8.346E-02	3.041E-01
^{64}Zn(n, p)	8.907E-02	1.767E-01	1.713E-01	8.555E-01	5.164E+02	3.930E+02	4.043E+02
^{64}Zn(n, γ)	2.058E+02	2.449E+02	2.809E+02	3.119E+02	8.111E+04	8.603E+04	9.171E+04
^{115}In(n, n')	7.854E+04	8.182E+04	8.388E+04	8.878E+04	1.681E+02	1.311E+02	1.153E+02
^{115}In(n, γ)	7.173E+01	7.662E+01	8.205E+01	8.907E+01	2.555E+07	2.582E+07	2.591E+07
^{115}In(n, 2n)[(2)]	2.373E+07	2.377E+07	2.387E+07	2.504E+07	9.742E+04	9.975E+04	1.002E+05
^{113}In(n, γ)	8.968E+04	9.004E+04	9.180E+04	9.323E+04	2.265E+06	2.320E+06	2.331E+06

Notes: (1) Read 1.2345E+07 as 1.2345 10^{+7}; (2) Values include RR of ^{115}In(n, 2n) plus RR of ^{113}In(n,γ) in aim to be comparable to the measured RR attributed to the ^{115}In(n, 2n)

general statement that the improving quality of neutron cross-section data in newer evaluated ENDF/B data is a continuous progress, which can be also seen from this comparison, can be drown:

- the fission neutron cross-sections of ^{237}Np in jeff3.2 show some degradation corresponding to other libraries, which has been concluded also in [4],
- the fission neutron cross-sections of isotopes of ^{243}Cm, ^{247}Cm, and ^{242m}Am still require improvements,
- evaluated cross-sections for Zn isotopes were given in endf71 and jeff32 libraries for the first time and obviously require further attention, as well as the In isotopes reaction cross-sections,
- the tendl2014 library in case of ^{242m}Am neutron fission cross-section shows the biggest deviation relative to the other libraries and to the experimental reaction rates,
- new created jendl40 and rosfond2010 ACE type libraries show similar RR deviations for all MA as the other available ACE type libraries,
- factor $\langle F_k \rangle$ shows larger differences among the simulated and the experimental reaction rates of ^{238}U(n, γ), ^{176}Lu(n, γ) and In isotopes since no self-shielding effect is accounted due to samples real size,
- the new irdff-v-1.05 ACE type nuclear library shows no big advantage over the older irdf2002 library except that the ^{115}In(n, γ) cross-sections are available, and
- almost all libraries show relative large deviation of the calculated to the experimental reaction rates

Table 4. Values of the mean squared deviation factor $<F_k>$ for MAKET lattice 21-1-5(M2)

ACE library	endf71	jeff32	tendl2014	ads20	irdf2002	irdff-1.05	jendl40	rosfond	Maslov
^{235}U(n, f)	1.11	1.11	1.11	1.11	1.11	1.11	1.11	1.11	–$^{(1)}$
^{237}Np(n, f)	1.21	1.49	1.25	1.23	1.20	1.19	1.25	1.19	–
^{238}Pu(n, f)	1.01	1.04	1.00	–	–	–	1.02	1.02	1.49
^{239}Pu(n, f)	1.07	1.08	1.07	1.07	1.07	1.07	1.07	1.07	–
^{240}Pu(n, f)	1.02	1.03	1.03	1.07	–	–	1.03	1.03	–
^{241}Pu(n, f)	1.02	1.02	1.02	1.02	–	–	1.02	1.02	–
^{242m}Am(n, f)	1.34	1.34	1.55	1.34	–	–	1.33	1.33	1.32
^{243}Cm(n, f)	1.19	1.19	1.15	–	–	–	1.20	1.16	–
^{245}Cm(n, f)	1.01	1.01	1.06	1.04	–	–	1.01	1.05	–
^{247}Cm(n, f)	1.30	1.30	1.44	1.28	–	–	1.30	1.18	–
^{238}U(n, γ)$^{(2)}$	2.23	2.22	2.23	2.23	2.25	2.19	2.21	2.17	2.16
^{55}Mn(n, γ)	1.12	1.11	1.12	1.12	1.12	1.12	–	–	–
^{63}Cu(n, γ)	1.16	1.16	1.15	1.16	1.16	1.16	–	–	–
^{197}Au(n, γ)	1.08	1.09	1.08	1.08	1.09	1.09	–	–	–
^{176}Lu(n, γ)$^{(2)}$	1.19	1.21	1.21	1.19	–	–	–	–	–
^{68}Zn(n, γ)	10.70	10.78	10.78	–	–	–	–	–	–
^{27}Al(n, α)	1.63	1.63	1.66	1.63	1.63	1.64	–	–	–
^{64}Zn(n, p)	1.25	1.14	1.14	–	1.15	1.13	–	–	–
^{64}Zn(n, γ)	1.05	1.04	1.04	–	–	–	–	–	–
^{115}In(n, n')$^{(2)}$	14.27	14.27	18.99	14.27	–	–	–	–	–
^{115}In(n, γ)$^{(2)}$	2.59	2.58	2.59	2.59	–	2.68	–	–	–
^{115}In($n, 2n$)$^{(2, 3)}$	2.71	2.67	2.72	2.71	–	–	–	–	–

Notes: (1) Character '–' indicates that no RR cross-section data of the nuclide exists in the ACE library; (2) Not accounted for the self-shielding effect due to sample real size; (3) Reaction rate value shows ^{115}In(n, 2n) plus ^{113}In(n, γ)

Table 5. Values of the mean squared deviation factor $<F_k>$ for MAKET lattice 21-2

ACE library	endf71	jeff32	tendl2014	ads20	irdf2002	irdff-1.05	jendl40	rosfond	Maslov
^{235}U(n, f)	1.20	1.20	1.20	1.20	1.20	1.19	1.20	1.19	–$^{(1)}$
^{237}Np(n, f)	1.13	1.28	1.16	1.14	1.12	1.12	1.14	1.14	–
^{238}Pu(n, f)	1.02	1.08	1.04	–	–	–	1.01	1.05	1.44
^{239}Pu(n, f)	1.12	1.13	1.12	1.12	1.12	1.13	1.13	1.13	–
^{240}Pu(n, f)	1.05	1.06	1.06	1.09	–	–	1.04	1.04	–
^{241}Pu(n, f)	1.05	1.05	1.05	1.05	–	–	1.04	1.04	–
^{242m}Am(n, f)	1.40	1.40	1.63	1.40	–	–	1.41	1.42	1.42
^{243}Cm(n, f)	1.30	1.30	1.25	–	–	–	1.31	1.27	–
^{245}Cm(n, f)	1.01	1.01	1.07	1.04	–	–	1.01	1.03	–
^{247}Cm(n, f)	1.46	1.46	1.63	1.43	–	–	1.47	1.30	–
^{238}U(n, γ)$^{(2)}$	2.16	2.16	2.16	2.16	2.18	2.10	2.30	2.36	2.17
^{55}Mn(n, γ)	1.08	1.08	1.08	1.09	1.09	1.09	–	–	–
^{63}Cu(n, γ)	1.05	1.05	1.04	1.05	1.05	1.05	–	–	–
^{197}Au(n, γ)	1.06	1.07	1.06	1.06	1.06	1.07	–	–	–
^{176}Lu(n, γ)$^{(2)}$	1.16	1.17	1.17	1.16	–	–	–	–	–
^{68}Zn(n, γ)	10.62	10.68	10.69	–	–	–	–	–	–
^{27}Al(n, α)	1.52	1.52	1.54	1.52	1.52	1.46	–	–	–
^{64}Zn(n, p)	1.30	1.17	1.17	–	1.18	1.15	–	–	–
^{64}Zn(n, γ)	1.04	1.04	1.04	–	–	–	–	–	–
^{115}In(n, n')$^{(2)}$	13.83	13.83	18.41	13.83	–	–	–	–	–
^{115}In(n,)$^{(2)}$	2.87	2.86	2.87	2.87	–	2.87	–	–	–
^{115}In($n, 2n$)$^{(2, 3)}$	1.65	1.62	1.66	1.65	–	–	–	–	–

Notes: (1) Character '–' indicates that no RR cross-section data of the nuclide exists in the ACE library; (2) Not accounted for the self-shielding effect due to sample real size; (3) Reaction rate value shows ^{115}In(n, 2n) plus ^{113}In(n, γ)

of SN as ^{27}Al(n, α), ^{115}In(n, γ), ^{64}Zn(n, p), and ^{115}In(n, 2n), which (deviation) reach an order of magnitude in case of ^{115}In(n, n') and ^{68}Zn(n, γ) reaction rates. It confirms that further work on the particular neutron reaction cross-sections data of these isotopes is necessary.

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AUTHOR CONTRIBUTIONS

The experimental work and the evaluation of the experimental results were carried out at the MAKET critical assembly by the ITEP team leading by Yu. E. Titarenko, M. P. Pešić has created some new MA ACE type libraries using NJOY99.396 code and has used also the recent available ACE type nuclear data libraries to calculate, by the MCNP6.1 code, the nuclides reaction rates at the Vinča Institute of Nuclear Sciences. All the authors participated in the nuclides reaction rates comparison study, analysis and discussion of the results presented, and in preparation of figures, tables, and text of the manuscript.

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ВАЛИДАЦИЈА ФИСИОНИХ НЕУТРОНСКИХ ПРЕСЕКА МИНОР АКТИНИДА

Валидација фисионих нуклеарних пресека минор актинида из недавно доступних евалуираних библиотека нуклеарних података извршена је поређењем брзина нуклеарних реакција израчунатих програмским пакетом MCNP6.1 са експерименталним вредностима. Експериментални узорци, који су садржавали танке слојеве ^{235}U, ^{237}Np, ^{238,239,240,241}Pu, ^{242m}Am, ^{243}Cm, ^{245}Cm, и ^{247}Cm, напарене на металну подлогу, фолије од ^{235}U (псеудо-легуре ^{27}Al + ^{235}U), ^{238}U, ^{nat}In, ^{64}Zn, ^{27}Al и вишекомпонентне узорке легуре ^{27}Al + ^{55}Mn + ^{nat}Cu + ^{nat}Lu + ^{197}Au, озрачени су у каналима у танку који су садржали соли флуора (0.52NaF + 0.48ZrF_4). Ови канали, означени као Micromodel Salt Blanket, постављени су у центру решетке на критичном систему са тешком водом MAKET у Институту теоријске и експерименталне физике у Москви. Овај рад је наставак научноистраживачких активности започетих на валидацији фисиони неутронских пресека за актиниде за које се претпоставља да ће се користити за испитивање квалитета нуклеарног горива за поткритичне реакторе погоњене акцелератором и фисионе брзе нуклеарне реакторе, одређивање брзина трансмутације, као и одређивање параметара рада ових фисионих система. Ове научноистраживачке активности су рађене у оквиру пројеката које су подржали Међународни научни и технолошки центар и Међународна агенција за атомску енергију кроз координиране истраживачке пројекте, у периоду од 2005–2010. године. Добијени резултати потврђују да су даља истраживања у евалуцијама потребна да би се остварили бољи подаци за неутронске пресеке за минор актиниде и изабране нуклиде који могу бити коришћени у поткритичним реакторима погоњеним акцелератором и фисионим брзим нуклеарним реакторима.

Кључне речи: библиотека нуклеарних података, ADS, брзи реактор, актинид, брзина реакције, MAKET, MCNP6.1