Final results of the "Benchmark on computer simulation of radioactive nuclides production rate and heat generation rate in a spallation target"

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Abstract

Selection of the model or code most useful for the ADS target activation calculations and, if such a tool is not yet available, the indication of the main deficiencies of the existing tools was undertaken. Also the proper calculation of the spallation target heating, both during its exposition to the beam and after its switch off was analysed. Results of the measurement of radioactivity induced in massive Pb target were applied as reference to the presented benchmark calculations. For the heat generation in the target only the intercomparison of calculations was assumed. The specific goals of the benchmark were: calculation of the long lived residuals production rate and activity in a lead target after its irradiation with 660 MeV protons, comparison of the results with the measured ones for the isotopes: ^{46}Sc, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{75}Se, ^{83}Rb, ^{85}Sr, ^{88}Y, ^{88}Zr, ^{95}Nb, ^{95}Zr, ^{102m}Rh, ^{102}Rh, ^{110m}Ag, ^{121m}Te, ^{121}Te, ^{139}Ce, ^{172}Hf, ^{172}Lu, ^{173}Lu, ^{175}Hf, ^{183}Re, ^{185}Os, ^{194}Au, ^{194m2}Ir, ^{195}Au, ^{203}Hg, ^{207}Bi, *calculation of the heating rate in the target and comparison of the results of calculations.*

Introduction

Deployment of Accelerator Driven System (ADS) is regarded as an interesting option of improving safety of nuclear power by the spent fuel transmutation. Actinide recycling can reduce the need for the large-scale repository of waste. One of the essential parts of ADS is the spallation target serving as the external neutron source of the subcritical core. The study of ADS operation and maintenance includes also the analysis of the build-up and decay of the target radioactivity and heating. The heating resulting from the beam particles and decay of radioactive nuclides. Both, the radioactivity and heating, can be calculated with the use of standard computational tools such as MCNPX [1], FLUKA [2,3] and others. However, not always the calculation methodology is straightforward and unambiguous. In calculation of radioactivity for the intermediate and high-energy range of particles (~ 20 - 1500 MeV) a number of physical models of nuclear interactions between the incident particle and nucleus are at hand (MCNPX). The selection of the model or code most useful for the ADS calculations and, if such a tool is not yet available, the indication of the main deficiencies of the existing tools, is undertaken as a task in the NUDATRA domain of the Integrated Project EUROTRANS. It is also a part of the IAEA Coordinated Research Project on Analytical and Experimental Benchmark Analyses of Accelerator Driven Systems. The proper calculation of the spallation target heating, both during its exposition to the beam and after its switch off is of importance for the designing of the XT-ADS and EFIT systems [4]. The experiment devoted to the measurement of axial distributions of radionuclide activity induced in massive Pb target was conducted at the Dzhelepov Laboratory of Nuclear Problems in JINR Dubna (Russia) [5-7] and its results were applied as reference to the presented benchmark calculations. There was no experiment conducted within this research, resulting in the respective measurement of heat generation or temperature distribution in the target. Therefore only the intercomparison of results of calculations was assumed. Thus the specific goals of the benchmark were: calculation of: the long lived residuals production rate during the lead target irradiation and their activity at 219 days after the irradiation end, in 32 pieces of Pb samples, distributed inside the target, and in the whole target; comparison of these results with the measured ones for the following isotopes: ^{46}Sc, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{75}Se, ^{83}Rb, ^{85}Sr, ^{88}Y, ^{88}Zr, ^{95}Nb, ^{95}Zr, ^{102m}Rh, ^{102}Rh, ^{110m}Ag, ^{121m}Te, ^{121}Te, ^{139}Ce, ^{172}Hf, ^{172}Lu, ^{173}Lu, ^{175}Hf, ^{183}Re, ^{185}Os, ^{194}Au, ^{194m2}Ir, ^{195}Au, ^{203}Hg, ^{207}Bi; calculation of the heating rate and its distribution in the target both, during exposition to the proton beam and after its switch-off, comparison between the results of calculations of heating rate. The benchmark was presented in Nice during the International Conference on Nuclear Data for Science and Technology 2007 [8]. Also partial results were presented on the Physor 2008 conference [9].

Analysis of the benchmark results for activation

Results for the whole target

Instantaneous isotope production rate

First part of the benchmark analysis was devoted to testing the calculation results for consistency. Results of instantaneous production rate for 24 nuclides, obtained with the use of each physical model (Bertini, CEM, INCL4 and Isabel) were compared (Figure 1). Generally the received results are consistent (Table 1). In cases when the consistency is worse one can observe either the spread of results higher than the respective uncertainties or systematic difference for selected nuclides. Instantaneous production rate results in the whole target are consistent for majority of participants and for all models. Differences between models are significant.

Figure 1: Examples of the comparison of results of the instantaneous production rate of the radioactivity induced in the whole target

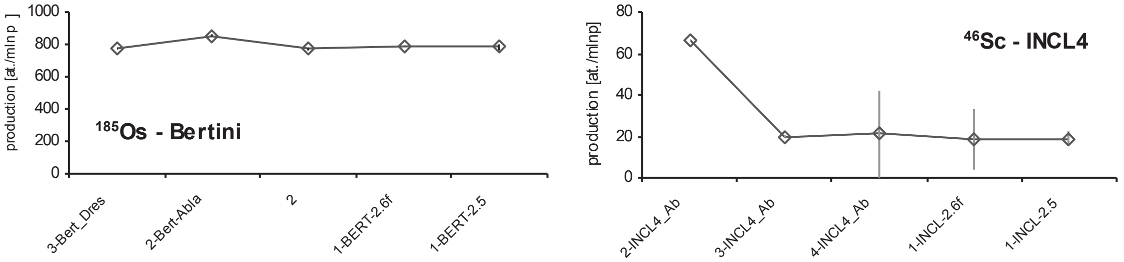


Table 1: Results of the comparison of instantaneous nuclide production rate

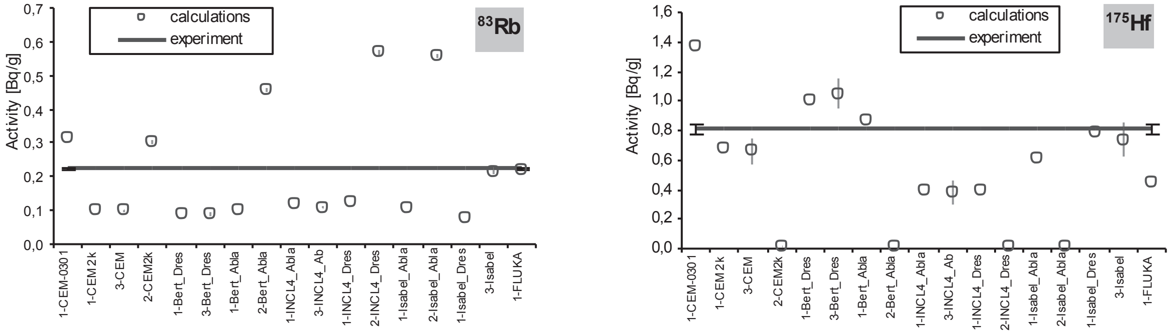
	Model:	Bertini	CEM	INCL4	Isabel
	Total number of results	Number of nuclides showing consistent results			
All nuclides	24	17	22	20	18
Light ($A \leq 60$)	3	2	2	3	3
Fission products ($60 < A < 140$)	11	10	11	10	9
Heavy spallation residues ($A \geq 140$)	9	5	9	7	6
^{207}Bi	1	-	-	-	-

Activity with account of the decay during and after activation

To compare the experimental results with the calculated ones one should recalculate the instantaneous production rates of the respective nuclide and its all predecessors for the moment of measurement. It should account both for the growth and decay during activation and the decay after activation end.

Examples of the comparison of results for the whole target are presented in Figure 2 while all ratios C/E (calculation to experiment) in Table 2

Figure 2: Examples of comparison of calculation results of the whole target activity for selected nuclides with account of the decay during and after activation



For the statistical analysis of the results the D and H coefficients were applied (Eq. (1))

$$D = \frac{\sum_{i=1}^{n} \left| 1 - \frac{C}{E} \right|}{n}, \qquad H = \sqrt{\frac{1}{n} \sum_{i=1}^{n} \left(\frac{E_i - C_i}{S_{E,i} + S_{C,i}} \right)^2} \tag{1}$$

where *n* – number of compared results due to different isotopes or participants and models. In calculations of the coefficient for isotopes the outlying results were omitted while when comparing models and participants the results of participant 2 were not evaluated. Also the results for isomeric states and respective nuclides were not considered due to the lack of separated production rates for the ground and excited state formation in majority of supplied data. Results are presented in Table 2.

Table 2: Cumulative results of the C/E ratio for the radionuclides activity of the whole target with account of the decay during and after activation

Participant No	C/E															
and	Nuclide															
model/code	^{60}Co	^{65}Zn	^{83}Rb	^{85}Sr	^{88}Y	^{95}Nb	^{102}Rh	$^{102m+g}$Rh	$^{121m+g}$Te	^{173}Lu	^{175}Hf	^{183}Re	^{185}Os	^{194}Au/Hg	^{203}Hg	^{207}Bi
1-CEM-0301	0.65	1.67	1.40	1.19	1.31	0.57	-	-	-	1.58	1.70	1.68	1.06	0.82	0.55	1.42
1-CEM2k	0.40	1.43	0.45	0.45	0.47	0.23	0.40	0.21	0.37	0.72	0.84	1.34	1.10	1.12	0.49	1.66
3-CEM	0.41	1.45	0.44	0.46	0.50	0.24	0.40	0.20	-	0.76	0.82	1.34	1.11	1.64	0.77	1.77
2-CEM2k	**9.81**	1.42	1.35	**69.24**	0.96	**8.63**	0.25	**0.13**	**60.80**	**0.01**	**0.02**	**0.19**	**0.04**	**8307.88**	0.64	1.78
1-Bert_Dres	0.41	1.20	0.39	0.33	0.38	0.23	0.42	0.22	0.51	1.30	1.24	1.53	1.05	0.85	1.30	1.40
3-Bert_Dres	0.46	1.41	0.39	0.32	0.38	0.24	0.42	0.22	-	1.36	1.29	1.54	1.07	1.35	1.44	1.47
1-Bert_Abla	0.43	0.61	0.44	0.37	0.38	0.36	0.67	0.34	0.42	0.99	1.07	1.46	1.16	0.94	1.32	1.06
2-Bert_Abla	**7.94**	0.84	2.05	**68.82**	1.17	8.33	0.41	0.21	**65.80**	**0.01**	**0.02**	0.24	**0.06**	**8021.72**	1.20	1.09
1-INCL4_Abla	0.51	0.54	0.53	0.45	0.44	0.64	0.84	0.43	0.59	0.38	0.49	0.96	0.82	0.93	1.61	1.17
3-INCL4_Abla	0.55	0.62	0.49	0.48	0.48	0.67	0.94	0.48	-	0.42	0.47	0.96	0.83	1.38	1.70	1.22
1-INCL4_Dres	0.51	0.47	0.57	0.44	0.45	-	0.89	0.46	0.56	0.38	0.48	0.97	0.82	0.92	1.62	1.17
2-INCL4_Dres	**11.35**	0.99	2.53	**69.60**	1.55	**8.33**	0.53	0.27	**56.84**	**0.01**	**0.01**	**0.17**	**0.04**	**9185.42**	1.65	1.18
1-Isabel_Abla	0.39	0.58	0.48	0.42	0.45	0.51	0.85	0.44	0.51	0.65	0.75	1.29	1.08	1.00	1.02	1.85
2-Isabel_Abla	**11.47**	0.96	2.48	**66.71**	1.42	**8.44**	0.49	0.25	**57.76**	**0.01**	**0.01**	0.22	**0.06**	**6715.72**	1.22	1.98
1-Isabel_Dres	0.42	1.16	0.33	0.28	0.35	0.23	0.41	0.21	0.47	0.93	0.97	1.44	1.04	0.92	1.30	1.40
3-Isabel	1.00	1.25	0.96	0.79	0.84	0.80	1.38	0.71	-	0.94	0.91	1.26	1.02	1.55	0.83	1.15
1-FLUKA	0.72	1.00	0.98	0.85	0.96	0.55	-	-	-	0.48	0.56	0.90	0.75	1.02	0.62	1.01

10%>C/E>0%

20%>C/E>10%

0.2 > C/E > 5

Examination of the Table 2 and qualitative comparison of calculated activities with the experimental ones show for the whole target, for different models:

- the worst performance of Bertini-Dresner (~ 12% acceptable results, below 20% difference)
- better of CEM, INCL4, ISABEL-Dresner and Bertini-Abla (~20-30%)
- the best but still unsatisfactory of ISABEL-Abla and FLUKA (~45 %)

Analysis of the quality of 13 calculated results for different nuclides (excluding these strongly outlying from unity - marked in the Table 2) show:

- the best performance for ^{185}Os and ^{194}Au/^{194}Hg, (12 and 9 results within 20%, respectively)
- for ^{183}Re, ^{175}Hf and ^{207}Bi only 4 – 6 results within this limit
- for nuclides from ^{60}Co to ^{121}Te almost all calculations underestimated
- for heavier mainly overestimated.

The quantitative analysis with the use of coefficient D, reflecting the absolute deviation of the C/E ratio from unity (Table 3), confirms the best performance of Isabel model and FLUKA code and worst of Bertini-Dresner and CEM 2k models. Among isotopes the D values for ^{185}Os and ^{194}Au/^{194}Hg are the lowest

Table 3: Values of the D coefficient for models and nuclides

Participant code - model	D	Nuclide	D
1-CEM-0301	0.42	^{60}Co	0.47
1-CEM 2k	0.48	^{65}Zn	0.32
3-CEM	0.49	^{83}Rb	0.61
1-Bert_Dres	0.46	^{85}Sr	0.50
3-Bert_Dres	0.50	^{88}Y	0.44
1-Bert_Abla	0.38	^{95}Nb	0.56
1-INCL4_Abla	0.39	^{173}Lu	0.35
3-INCL4_Ab	0.39	^{175}Hf	0.31
1-INCL4_Dres	0.39	^{183}Re	0.38
1-Isabel_Abla	0.39	^{185}Os	0.11
1-Isabel_Dres	0.43	^{194}Au/Hg	0.21
3-Isabel	0.19	^{203}Hg	0.38

Results for the distribution along the target

Example of the comparison of results for the distribution along the target is presented in Figure 3 while the ratios C/E (calculation to experiment) in Figure 4.

The quantitative evaluation, of results for all 22 analysed nuclides and 4 physical models of the spallation reaction, was done with the use of the D and H coefficients (Eq. (1)). The coefficient H represents the weighed quadratic average of absolute distance of points from the line representing the equality of measured and calculated results (as can be seen on the right side graph in Figure 4). As the weight the reciprocal of the sum of standard deviations of experimental and calculated result was applied. The calculated values of coefficients D and H are presented in Table 4.

Analysis of the values of coefficient D allows for the following observations:

- average values for all nuclides differ only slightly;
- the Isabel model seems to be better (D = 0.40) and the Bertini-Dresner worse (D = 0.56) then the two others,
- only ~12 % of calculated results from all models differ from experimental ones by less than 20 %.

Figure 3: Cumulative comparison of calculated (weighed mean) and measured specific activity distribution along the target with the account of decay during and after activation for different physical models applied in calculations

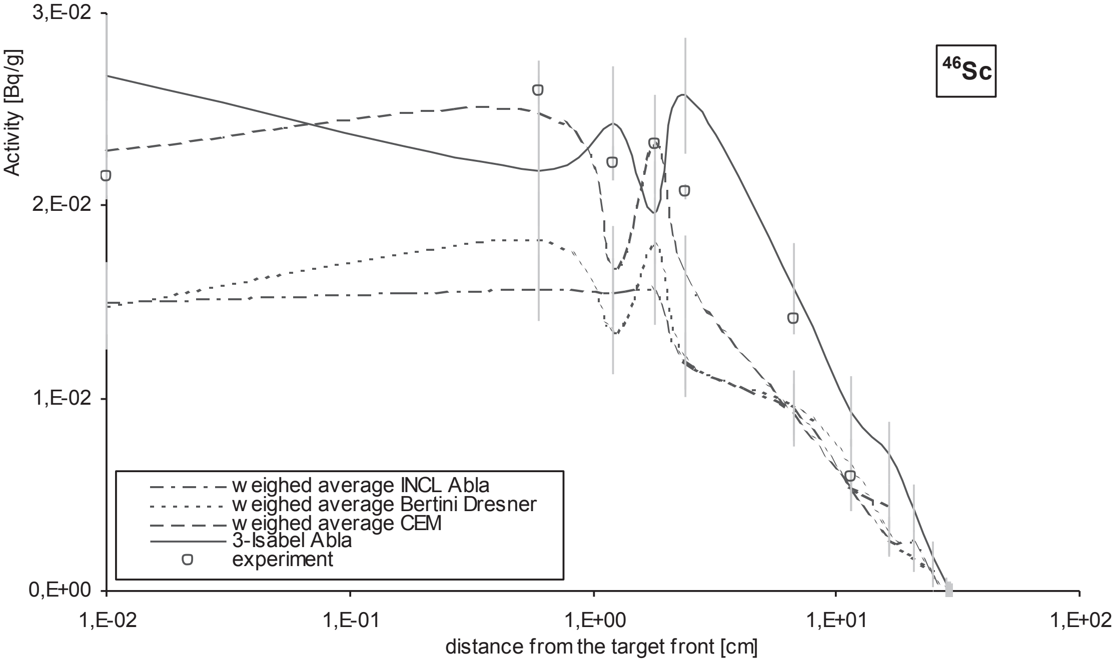


Figure 4: Comparison of the experimental and calculated distributions of 95Nb (specific activity in Bq/g) along the target. The calculated values are weighed averages of results from participants.

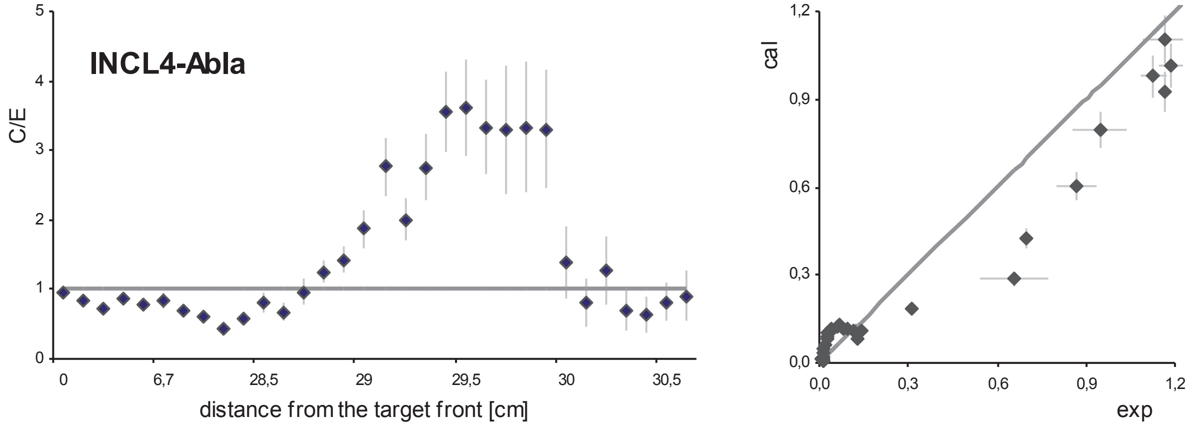


Table 4: Values of the D and H coefficients for models and nuclides*

Nuclide	CEM	BerDre	INCL4Abla	Isabel	CEM	BerDre	INCL4Abla	Isabel
	D				H			
Sc46	0.117	0.294	0.316	0.229	0.76	1.76	2.20	0.99
Fe59	0.257	0.276	0.179	0.304	2.55	2.02	1.97	2.02
Co60	0.587	0.588	0.530	0.223	4.64	4.71	4.72	1.73
Zn65	0.816	0.850	0.926	0.848	2.60	2.36	3.48	2.44
Se75	0.215	0.566	0.284	0.593	1.16	3.25	2.51	2.57
Rb83	0.565	0.584	0.443	0.148	4.99	5.71	4.41	1.14
Sr85	0.781	0.559	0.492	0.246	4.13	4.96	4.08	1.76
Y88	0.593	0.697	0.503	0.408	5.14	5.09	5.29	2.99
Zr88	0.137	0.313	0.576	0.210	0.97	2.55	5.24	1.65
Nb95	0.862	0.805	0.748	0.750	9.66	6.19	2.44	2.09
Zr95	0.846	0.755	0.900	0.852	9.78	6.77	4.44	3.58
Ce139	0.474	0.095	0.075	0.112	2.07	0.44	0.42	0.38
Hf172	0.627	0.906	0.404	0.480	4.26	5.85	3.24	2.76
Lu172	0.331	0.596	0.474	0.263	1.50	2.85	2.52	0.61
Lu173	0.350	0.473	0.559	0.204	3.30	1.92	3.07	1.03
Hf175	0.273	0.380	0.511	0.271	1.08	1.02	2.45	0.58
Re183	0.347	0.469	0.174	0.349	1.81	2.01	1.15	1.00
Os185	0.231	0.244	0.363	0.270	0.74	0.65	1.50	0.86
Au194	0.502	0.287	0.198	0.618	3.74	1.52	0.97	3.69
Au195	0.281	0.144	0.606	0.587	1.70	1.27	5.25	2.37
Hg203	0.424	1.487	0.571	0.178	4.03	6.45	5.05	1.25
Bi207	1.134	0.946	0.420	0.612	15.19	13.93	6.13	9.53
average	0.49	0.56	0.47	0.40	3.90	3.79	3.30	2.14

*Each value represents all points of one distribution along the target.

0.10 > D > 0 **0.2 > D ≥ 0.1** **1.0 > H > 0** **2.0 > H ≥ 1.0**

When the difference is related to the result standard deviation, both from calculation and experiment (in coefficient H) the differentiation between models and nuclides looks more distinct:

- the Isabel model yields best results – the average difference is slightly larger than 2 standard deviation while for the other 3 models it is between 3.3 (INCL4-Abla) and 3.9 (CEM),
- for Isabel model over 55 % of values is smaller than 2 and for the other models only ~33 %.

The distributions along the target contain also some information about the proton energy influence on the results of calculation. Samples placed in different distance from the target front were irradiated with protons (mainly) of different spectra. Based on the approximate dependence of the respective distribution peak energy on the sample position one can observe the following general regularities:

- best (but not always satisfactory) results of calculations for proton energy above ~120 MeV (on the average for all measured nuclides) are obtained for the Isabel model, while below this energy results from INCL4-Abla are the best but only slightly better than that from Bertini-Dresner,
- calculated results are in satisfactory agreement with the experimental ones only for proton energy above ~ 460 MeV.

This is more difficult to recognize clear regularities when looking on the results from the point of view of the measured radionuclide or group of them, for the whole proton energy range on average. What can be observed is:

- the quality of results calculated with the use of Isabel model prevails over the one from other models for: ^{83}Rb, ^{85}Sr, ^{139}Ce, ^{172}Hf, ^{172}Lu, ^{173}Lu, ^{175}Hf, ^{183}Re, ^{195}Au, ^{203}Hg and ^{207}Bi,
- no model is satisfactory, even approximately, for such nuclides as: ^{60}Co, ^{65}Zn, ^{88}Y, ^{88}Zr, ^{95}Nb, ^{95}Zr, ^{172}Hf, and ^{203}Hg,
- other models (but Isabel) produce relatively good results for certain nuclides, i.e.: INCL4-Abla for ^{59}Fe, ^{139}Ce and ^{183}Re, CEM for ^{185}Os, Bertini-Dresner for ^{194}Au.

Analysis of the benchmark results for heating

The only goal to be achieved from this part of the benchmark is the comparison of calculation results and analysis of their compatibility among themselves and with earlier expectations. For this a similar target was designed with slightly different structure of cells [9].

Power released in the target during the beam operation

The calculation results for the whole target heating rate are presented in the Table 5. The assumed beam parameters were 1 mA continuous Gaussian beam of 660 MeV protons. The results from all participants are consistent. Around 400 kW (ca 60 % of the beam power) is released. Only small differences (ca 2 % spread) are observed between results from different models.

Table 5: Comparison of calculated results of the whole target heating power [kW] by a 1 mA beam of 660 MeV protons

Participant No	Physical model						Remark
	Bertini Abla	Bertini Dresner	INCL4 Abla	CEM	Isabel Abla	Isabel Dresner	
1	387.6	392.1	391.7	391.7	391.9	395.8	MCNPX 2.2.3
3	-	389.9	399.3	369.5	397.2	-	MCNPX 2.5.0
5	-	-	405.0	396.0	-	-	MCNPX 2.5.0

Figure 5. Distributions of the total heating power in the target calculated with the use of INCL4-Abla model. The numbers on axis x represent target cells of different thickness in the longitudinal direction.

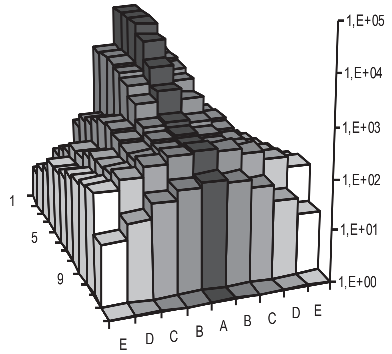
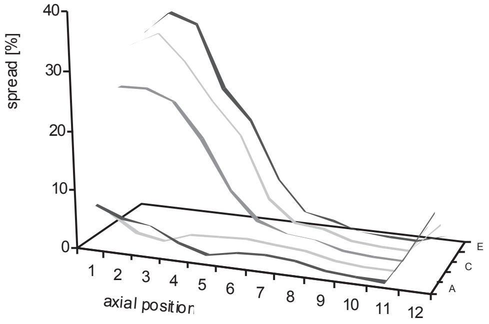


Figure 6. The distribution of relative total heating power spread between results obtained with models CEM, Bertini-Dresner, INCL4-Abla and Isabel. The numbers and letters on axes x and y represent target cells.



Example of the longitudinal and radial heating distribution is presented in Figure 5. Comparison of the distributions yielded by different spallation reaction models show serious differences. One example is presented in Figure 6. For the major part of the target the spread between the most differing models is smaller than 10 %. However there are regions where spread higher than 40 % is obtained. These are the most radially outside parts of the target front and at the target end behind the primary proton range. One can suspect in both cases the different "behaviour" of secondary particles in different simulation models. Difference in evaluation of such parameters of secondary particles as their energy and direction as well as their production cross-sections can cause the difference.

Conclusions

Results from the limited number of laboratories that took part in the exercise still present a clear picture of the situation regarding the thick target activation simulation. However, the relatively small amount of the used experimental data may restrict the general validity of the below conclusions.

The physical models applied for the *calculation of thick lead target activation do not produce satisfactory results* for the majority of analysed nuclides, however one can observe better or worse quantitative compliance with the experimental results. Analysis of the quality of calculated results show the best performance for heavy nuclides (A ≅ 170 – 190). For intermediate nuclides (A ≅ 60 – 130) almost all are underestimated while for A ≅ 130 – 170 mainly overestimated.

The *shape of the activity distribution* in the target *is well reproduced* in calculations by all models but the numerical comparison shows similar performance as for the whole target. The Isabel model yields best results. Analysing the distributions of C/E ratio along the target length as a function of the dominating proton energy one can observe the best (but not always satisfactory) results for proton energy above ~120 MeV for the Isabel model. Below this energy results from INCL4-Abla are the best but only slightly better than that from Bertini-Dresner. Calculated results are in satisfactory agreement with the experimental ones only for proton energy above ~ 460 MeV.

The situation is different for calculations of heating. For the *whole target heating rate the results* from all participants *are consistent*. Only small differences are observed between results from physical models.

For the *heating distribution in the target* it looks not quite similar. The quantitative comparison of the distributions yielded by different spallation reaction models shows for the major part of the target no serious differences – generally below 10%. However, in the most outside parts of the target front layers and the part of the target at its end behind the primary protons range spread higher than 40 % is obtained.

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