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Neutron spallation source and the Dubna Cascade Code

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Abstract. Neutron multiplicity per incident proton, n/p, in collision of high energy proton beam with voluminous Pb and W targets has been estimated from the Dubna Cascade Code and compared with the available experimental data for the purpose of benchmarking of the code. Contributions of various atomic and nuclear processes for heat production and isotopic yield of secondary nuclei are also estimated to assess the heat and radioactivity conditions of the targets. Results obtained from the code show excellent agreement with the experimental data at beam energy, $E < 1.2$ GeV and differ maximum up to 25% at higher energy.

Keywords. Neutron spallation source; inter- and intra-nuclear cascade.

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1. Introduction

Incineration of long life nuclear waste is one of the most important problem of the modern time and accelerator driven sub-critical system (ADSS) has been suggested [1–3] as one of the possible alternative. In ADSS not only thorium-like fuel can be burnt but it also has the ability to degrade plutonium-like highly dangerous waste by transmutation. In this way, developing new reactor technology based on ADSS seems highly promising. For designing ADSS, one of the most important requirement is to develop an intense neutron spallation source (INSS). This requires mathematical designing and modeling of INSS using a suitable computer code.

It is well known from the multiparticle phenomenology that at energies greater than several tens of MeV nuclear-cascade (both inter- and intra-nuclear) is the basic mechanism of particle production. In the early stage of an intra-nuclear cascade fast nucleons and pions are produced and at the second stage a highly excited 'after the cascade' nucleus relaxes into a stationary state sometimes emitting several nucleons. At the third and the last stage the excited residual nucleus decays by competitive successive evaporation of nucleons, fission and emission of γ-quanta.

Dubna Cascade Code [4] developed for the purpose of mathematical modeling of ADSS allows one to estimate ADSS parameters like probability of different kinds of interactions (elastic, inelastic, neutron capture and fission etc.) of primary projectiles in a composite medium, emission of various radiations and their transportation through complicated multi-layer medium, heat generation because of the stopping of both particles and other radiations. A detailed description of the code has been given in our recent publications [5,6]. It may be mentioned that the Dubna Code takes care of the transportation of all radiations up to thermal energy, 2.5×10^{-8} eV and uses 26-group calculations [7]. Many other cascade codes such as (i) Los Alamos High Energy Monte Carlo Transport Code (LAHET) [8], (ii) FLUKA [10], (iii) Monte Carlo Nuclear Particle Transport Code (MCNP-4b) [9], (iv) ISABEL [11], (v) CEN2K [12] and (vi) SHIELD [13] have also been developed with selective features. For example LAHET and FLUKA codes are developed for the transport of particles of high energy >20 MeV and MCNP for the low energy particles. Compared to them Dubna Code is a comprehensive code which may be used for the transport of both low and high energy particles as well as γ-radiation. Secondly, it has inbuilt designing features also.

During the last one decade, experiments related to spallation neutron sources have been conducted: (i) Berlin Neutron Ball (BNB) [14], (ii) Los Alamos National Laboratory (LANL) [15], (iii) Brookhaven National Laboratory (BNL) [16], (iv) SINQ [17], (v) Argonne National Laboratory (ANL) [18] etc. From these experiments it is revealed that the light mass spallation targets are less radioactive but suffer from the snag of tritium inventory. On the other hand, most of the very heavy targets although provide higher neutron-yield per incident proton but produce high radioactivity.

In this paper we have presented estimates of neutron multiplicity, isotopic distribution of the produced nuclei and heat contributions of different nuclear and atomic processes in collision of proton beam with heavy targets of different materials, shapes and sizes using current version of Dubna Cascade Code-2001. These data are also compared with the available experimental results for the purpose of benchmarking of the code.

2. Dubna Cascade Code

Dubna Cascade Code handles both inter- and intra-nuclear cascade and includes all those features of transport of radiations through the heterogeneous medium. For a brief introduction of the code some of the important features are given as follows:

1. In particular, ionization losses of charged hadrons are calculated using Sternheimer's method [19] and for the ions a more complicated method is used [20].
2. Properties of particles created in inelastic hadron $+$ nucleus collisions at high energy are calculated by means of the intra-nuclear cascade and evaporation model [21,22].
3. The non-stationary pre-compound processes of the after-cascade residual nucleus are taken into account.
   It may also be mentioned that up to now all the details of such complicated process have not been investigated comprehensively. For example, in some cases the fission occurs during the relaxation stage, before the formation of a stationary state and sometimes it turns out that in the case of light nuclei, excitation energy is so large that the model of successive evaporation becomes unapplicable and the decay process must be considered as an explosion. At present, we have no single satisfactory theory for the complete description of such a complicated phenomenon.
4. All the created fast cascade and decay neutrons from excited nuclei are moderated by numerous elastic nuclear collisions up to a very low energy. At the same time in

the case of fissionable targets, number of low-energy neutrons increases. Finally, the low-energy neutrons are captured by way of (n,γ) reactions, for example, producing Pu^{239} from U^{238} and U^{233} from Th^{232} target etc. The produced energy is taken away by a hard γ-quanta. Therefore, a neutron is considered to be lost either when it escapes the enclosure or it is absorbed in reactions like (n,γ), (n,xn'), (n,f) or it is slowed down up to the threshold energy.

A list of important features of Dubna Code is given as follows:

(i) The kind of reactions taking place inside any target or ADSS for which predictions are made for each incident beam particle are: (a) fissions at $E < 10.5$ MeV, (b) fissions at $E > 10.5$ MeV, (c) (n,γ) capture at $E < 10.5$ MeV, (d) $(n,n)+(n,2n)$ channels at $E < 10.5$ MeV, (e) inelastic collisions at $E > 10.5$ MeV (excluding high-energy fission), (f) elastic scattering at $E < 10.5$ MeV, (g) elastic scattering at $E > 10.5$ MeV,

(ii) average heat contributions, Q_i, of the following processes $(i = 1$ to $14)$ in a target or ADSS are given for each bombarded proton: (1) ionization losses, (2) stopped hadrons and nuclei, (3) low-energy fission ($E < 10.5$ MeV), (4) high-energy fission ($E > 10.5$ MeV), (5) γ-ray energy from (n,γ) reactions, (6) energy of the residual nuclei, γ-ray de-excitation by fission and all inelastic high-energy collisions, (7) γ-ray energy from pion decays, (8) recoil nuclei energy in (n,γ) reactions, (9) recoil nuclei energy in (n,n') and $(n,2n)$ reactions, (10) recoil nuclei energy in low-energy elastic scattering, (11) recoil nuclei energy in high-energy elastic scattering, (12) energy of escaped neutrons, (13) energy of escaped charge particles and (14) recoil energy of the residual cascade nucleus.

3. Comparison of experimental data with the Dubna Cascade Code

3.1 *Average number of spallation neutrons per beam proton* (n/p)

In BNL experiment [23,24], targets in cylindrical shapes, i.e., $2R \times L = 10.2 \times 61\ \mathrm{cm}^2$ for lead and $10.2 \times 40\ \mathrm{cm}^2$ for tungsten have been exposed to proton beam of energy ranging from 0.8 to 1.4 GeV for the purpose of study of INSS. Results of average neutron per beam proton obtained from this experiment have been compared with the calculated results of Dubna Cascade (April 2001 version) in table 1. Also, these results are displayed separately for the Pb and W targets as function of the beam energy in figures 1a and b respectively. For the calculations using Dubna Code, values of average binding energies of neutron and proton for Pb target are taken to be $En = 7.37$ MeV and $Ep = 3.717$ MeV respectively. Similarly, for the W target these values are $En = 5.749$ MeV and $Ep = 5.396$ MeV. When we compare total neutron yield per incident proton, it may be said that the results of Dubna Cascade Code are in agreement within less than 10% with the experimental results of Pb target up to 1.2 GeV energy. In this case, total neutron yield is almost the same as the number of the escaped neutrons from the target boundaries. In the case of W target, total neutron yield differs up to 25% and the multiplicity of escaped neutrons differs up to ∼20%. At $E > 1.2$ GeV, in the case of both Pb and W target, main reason of disagreement of the cascade results is the lack of a physical model for handling decay of highly excited neuclei. Chigrinov *et al* [24] have compared results of calculations of LAHET and SONET

Table 1. Comparison of results of average neutron multiplicity per beam proton (n/p) of the BNL experiment with lead (Pb 207.2) and tungsten (W 184) targets with the results of Dubna Cascade Code.

		0.8		1.0		1.2		1.4	
E (GeV)		Pb	W	Pb	W	Pb	W	Pb	W
BNL (Exp.)		13.5	15	17.5	20.5	22.3	–	26.3	28.5
Dubna Cascade	$(n/p)_{\text{esc}}$	13.51	18.2	16.41	23.8	20.31	28.9	22.82	33.8
	$(n/p)_{\text{cap}}$	0.01	1.2	0.01	1.3	0.01	1.6	0.02	1.8
LAHET		15	17.5	19.9	23.3	24.3	28.9	28.3	33.8
SONET		12.8	14.5	17.3	19.2	21.8	24.5	26	29.5

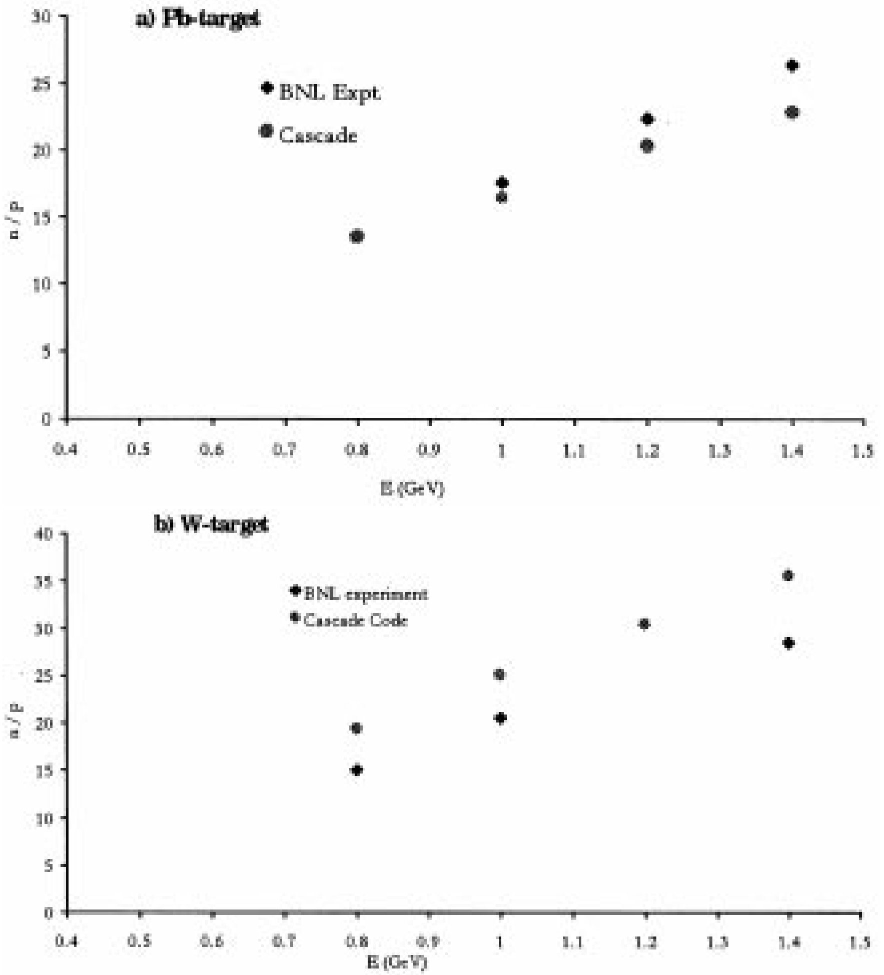

Figure 1. Variation of n/p with energy, E, in (**a**) Pb target and (**b**) W target. Results of Dubna Code calculations are compared with the given results of BNL experiment.

codes with the same experimental ones and these results are also given in table 1 for the sake of comparison. Here, it is important to point out that in the SONET Code, neutron histories are simulated until neutron kinetic energy becomes lower than 20 MeV while in Dubna Code this limit is 10^{-8} eV.

In table 2 results of Berlin Neutron Ball experiment [14] for the average neutron multiplicity per beam proton, n/p have been compared with the Dubna Code for the proton energy ranging from 1.22 to 4.2 GeV colliding with the Pb target. It may be seen that Dubna Code underestimates the neutron yield with 20–30% difference. It may be mentioned that although BNB results given in table 2 are after consideration of detector efficiency which is quoted to be 85% for low energy neutrons and that drops down to 15% at 100 MeV, no errors are quoted with the mean values. Results of both BNB experiment and Dubna Code are displayed as function of the beam energy in figure 2.

In table 3 results of n/p of the BNB experiment [14] and Dubna Code for the fixed proton beam energy, 1.22 GeV and diameter of the cylindrical Pb target being 15 cm have been given as a function of length of the target, L (cm). It may be seen from the table that n/p obtained from Dubna Code agrees with the experimental result within 5% difference. These results are displayed in figure 3 along with the following curves fitted to the data.

$$n/p = -8.76 + 8.44\ln(L) \text{ for the experimental results,}$$
$$n/p = -9.45 + 8.49\ln(L) \text{ for the Dubna Code results.}$$

It becomes clear that n/p rises slowly with the target length, L, after 35 cm length. Similarly, in table 4 results of BNB experiment and Dubna Code for the fixed beam energy $E = 1.22$ GeV and target length $L = 35$ cm are given as functions of the diameter of the cylindrical target. Experimental data are available for $2R = 8$ and 15 cm only.

Table 2. Comparison of results of average neutron multiplicity per beam proton of the BNB experiment [14] for Pb 207.2 target having dimensions $2R \star L = 15 \times 35$ cm^2 with the results of Dubna Cascade Code.

E (GeV)	1.22	2.2	3.2	4.2
BNB Expt.	∼21	∼45	∼59	∼70
Dubna Cascade	20.40±0.04	33.10±0.06	47.10±0.07	57.70±0.08

Table 3. Results of BNB experiment [14] for Pb 207.2 target with $2R = 15$ cm and $Ep = 1.22$ GeV proton beam with the length of the spallation target compared with Dubna Cascade Code.

Length	5	15	20	28	35	40
BNB experiment	5	13.5	16.5	20	21	–
Dubna Cascade	4.57±0.02	13±0.03	15.6±0.04	18.9±0.04	21.3±0.05	21.7±0.05

Table 4. Results of BNB experiment and Dubna Cascade Code for the varying values of diameter of the spallation target cylinder of length $L = 35$ cm, $Ep = 1.22$ GeV energy.

$2R$ cm	8	12	15	20	25	30
BNB experiment	∼18	–	21	–	–	–
Dubna Casacde	16.4±0.04	18.5±0.04	19.1±0.04	20.1±0.04	22.2±0.05	22.4±0.05

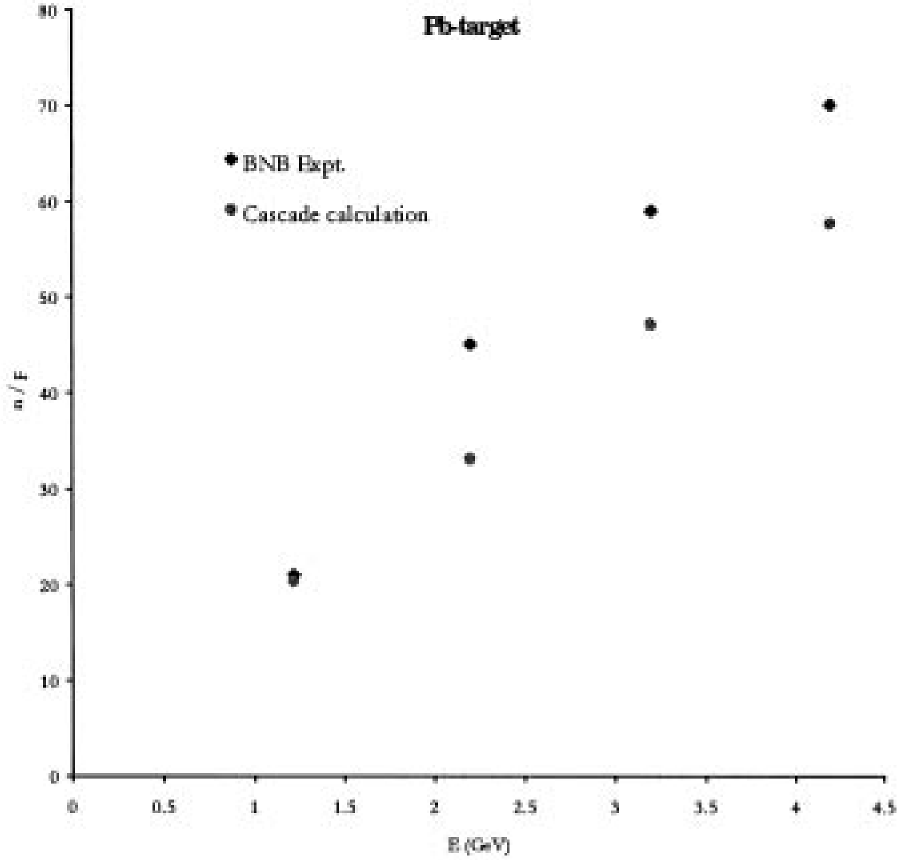

Figure 2. Variation of n/p with energy, E, in Pb target in BNB experiment. Results of Dubna Code calculations are compared with the experimental results.

On comparison, it may be pointed out that the results of the Dubna Code are in agreement with the experiment within 10% difference. These data are also displayed in figure 4. From the results given in figures 3 and 4 it may be pointed out that the neutron multiplicity changes faster with the length than the diameter of the target. This is because of the fact that the hadronic cascade has highly limited transverse momentum.

In table 5 more experimental results of neutron multiplicity obtained in p+Pb collision at energies up to 1.47 GeV [25–27] are compared with the corresponding results of Dubna Code. It may be seen that the results of these experiments are in very good agreement with results of Dubna Code within a difference of 10–15%. It is important to point out that in the case of the above mentioned data at energy $E = 1.47$ GeV, Cascade Code overestimates the experimental results only slightly. This observation is in contrast with the comparison presented in case of BNB experiment where the Code underestimates the neutron yield.

In another experiment by Chultem *et al* [28], average neutron multiplicity per beam proton of energy 3.65 GeV colliding with thicker Pb target of size $50 \times 50 \times 80$ cm^3 has been measured to be 101±20 which is quite comparable with the value 91±0.1 estimated from the Dubna Code.

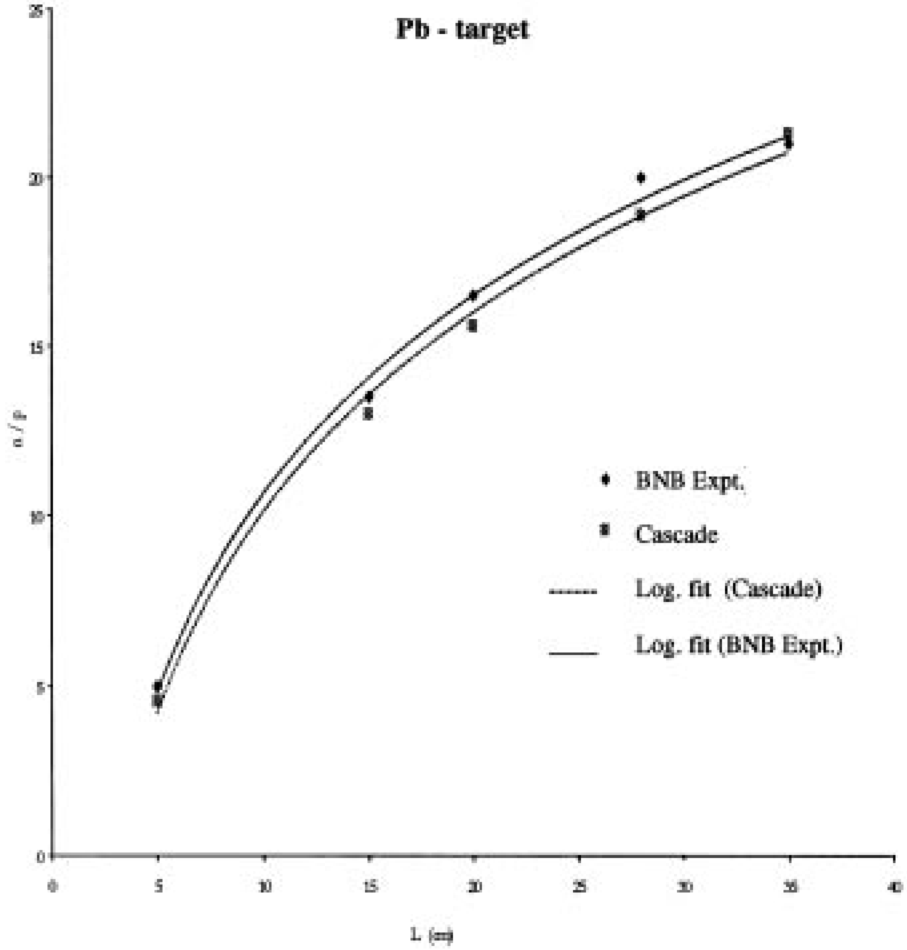

Figure 3. Variation of n/p with the length of the Pb target in BNB experiment. Results of Dubna Code calculation are compared with the given experimental results.

Table 5. Some more Cascade Code calculations of n/p (produced average number of neutrons per incident proton) for Pb are compared with the experimental results given below.

Ep (GeV)	Target dimensions $2R\star L$ cm^2	n/p (Expt.)	n/p (Dubna Code)	Reference for expt. data
0.317	20.0 × 60	3.13 ± 0.06	4.03 ± 0.02	[25]
0.47	20.4 × 64	8.00 ± 0.40	8.17 ± 0.03	[26]
0.72	20.4 × 64	11.8 ± 0.60	13.5 ± 0.37	[26]
0.86	20.4 × 64	16.6 ± 0.80	19.5 ± 0.04	[26]
1.47	20.4 × 64	24.6 ± 1.40	28.4 ± 0.05	[26]
0.47	40.8 × 61	8.70 ± 0.40	9.20 ± 0.03	[27]
1.47	40.8 × 61	31.5 ± 1.60	35.5 ± 0.06	[27]

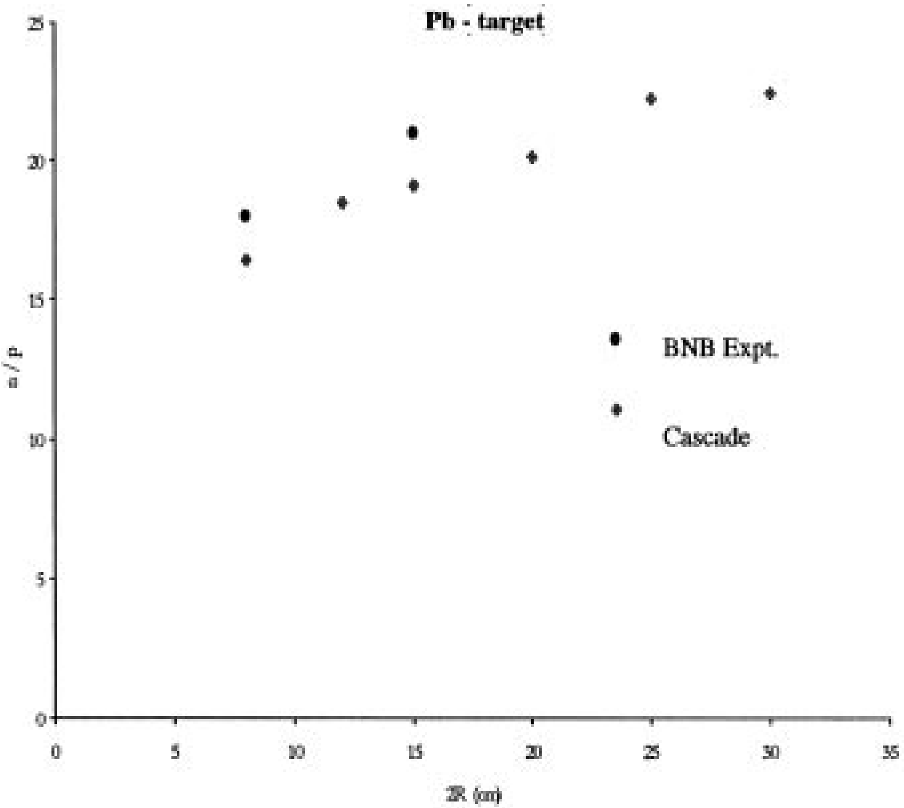

Figure 4. Variation of n/p with diameter of the Pb target. Results of Dubna Code are compared with that of BNB experiment.

3.2 *Heat contributions of various processes*

In §2, heat contributions of various nuclear and sub-nuclear processes taking place inside thick target or ADSS have been given. In table 6 results of calculations from Dubna Code for the heat contributions of these processes in the case of p+Pb collision system at beam energies ranging from 0.3 to 2 GeV are given. For these calculations, we have considered arbitrary dimension of the target to be $2R \times L = 20 \times 35\ \mathrm{cm}^2$. It may be seen from the data in table 6 that at low incident energy, major heat contribution comes from the ionization process. For example, it is 81% at 0.3 GeV decreasing to 33% at 2 GeV. A very small fraction of heat contribution, ~0.5% is from the high energy fission of Pb target. It is important to point out that the percentage heat contribution of the escaped neutrons increases with the beam energy, i.e., ~4% at $E = 0.3$ GeV and ~15% at 2 GeV. On avoiding energy of escaped γ-rays being less than 0.1%, it may be said that nearly 95% of the beam energy at $E = 0.3$ GeV and 85% at 2 GeV remains inside the block of Pb target. It is evident from the comparison of total heat contributions with beam energy E that in the case of Pb, multiplication of energy is insignificant. In column 8 of table 6, heat contributions are given for the block of tungsten for 1 GeV beam energy for comparison. It is important to point out that in W block heat contribution from (n,γ) reactions is about 24 times, recoil nuclei in (n,n') reactions is 50%, and due to ionization processes is 15% higher than the Pb block. On the other hand, heat contributions by escaped neutrons is 25% and high energy fission is 7 times lesser than the Pb block. Cross-section of (n,γ) reaction being higher in W reaction affects the net neutron yield adversely and such neutrons increase the heat contribution.

3.3 *Isotopic distribution*

It is important to study isotopic distribution of the produced nuclear fragments from the point of study of radioactivity. Results of calculations depend largely on parameterization and 'number of groups' used in calculations specially in the resonance region. Other practical difficulty in comparing the calculated result with the experimental one

Table 6. Average heat contributions of prominent processes, Q_i (see §2) inside the Pb target ($2R \times L = 20 \times 35$ cm^2) on collision of proton at different energies are listed. Relative heat contribution from the ionization process, $q = Q_{\text{ion}}/E$ (%) and total heat produced by each colliding proton is also given in the third row. In the column of 1 GeV energy Q_i for the block of same dimensions of W are also given in the brackets.

E (GeV)	0.3	0.4	0.5	0.6	0.66	0.8	1.0	1.1	1.2	1.5	2.0
Q_1 (MeV)	242	303	341	376	387	412	438	458	485	549	669
$q = Q_{\text{ion}}/E$ (%)	81	76	68	62	59	52	44 (500)	42	40	37	33
Q_2 (MeV)	0.604	1.29	2.91	4.3	6.08	9.82	15.7 (17.7)	18.3	21.3	31.4	46.3
Q_4 (MeV)	1.65	6.6	9.9	14.3	18.7	20.6	28.3 (3.88)	34	27	32.7	33.6
Q_5 (MeV)	0.158	0.207	0.275	0.397	0.58	0.519	0.977 (25.3)	0.971	0.938	1.22	1.55
Q_6 (MeV)	3.56	5.19	8.17	10.5	12.2	14.1	18.8 (19.9)	21.2	22	28.4	35.8
Q_7 (MeV)	0.172	0.18	1.73	3.95	7.46	12.8	25.6 (31.8)	37.1	49.7	69.9	119
Q_8 (MeV)	0.018	0.022	0.025	0.036	0.047	0.049	0.08 (2.08)	0.096	0.088	0.117	0.138
Q_9 (MeV)	4.22	6.96	11.3	16.1	19	24	30.5 (45.5)	34.6	36.5	45.6	57.9
Q_{10} (MeV)	0.214	0.353	0.574	0.776	0.916	1.09	1.4 (0.87)	1.59	1.62	2.05	2.6
Q_{11} (MeV)	0.004	0.006	0.01	0.011	0.014	0.017	0.022 (0.03)	0.024	0.027	0.034	0.05
Q_{12} (MeV)	13	26	41	58	75	101	136	155	163	222	298
$q_n = Q_{12}/E(\%)$	4.33	6.5	8.2	9.6	11.4	12.6	13.6 (113)	14.1	13.6	14.8	14.9
Q_{13} (MeV)	0	0.1	0.5	1.46	2.21	39.5	87.2 (3.43)	102	125	172	309
Q_{14} (MeV)	40	63.5	103	145	171	215	291 (313)	335	359	465	604
Q_{total} (MeV)	305.6	413.4	520.4	630.8	700.2	852.5	1073.6 (1079.4)	1198	1291	1619	2177

is the availability of the data corresponding to heavy mass fragments products in $p + A$ collision around 1 GeV. In this section, results of isotopic yield in thick Pb target ($d \times L = 1 \times 1$ m^2) bombarded by 1 GeV proton corresponding to the neutron spallation experiment by Bakhmutkin *et al* [29] are discussed. In figure 5, results of Dubna Code are displayed. According to the Dubna Code, the combined yield of Bi^{203}, Bi^{204} and Bi^{206} is estimated to be 0.59×10^{-4} reactions/(g·cm^{-2}·p) which is comparable with the experimental value $(0.734 \pm 0.047) \times 10^{-4}$ reactions/(g·cm^{-2}·p) [29]. In the figure one can see the presence of three distinctly separated groups of nuclear products, i.e., nuclear fragments with mass numbers $A < 5$ evaporated from excited residual (after Cascade) nuclei, fission products with $A = 50$–150 and residual nuclei after evaporation processes with mass close to the target mass. Also on comparing detailed results of the yield of heavy nuclei with $A > 150$ and very light nuclei, $A < 5$ of our calculations with the results of Cascade-INPE Code [30] it can be mentioned that the two are comparable. On the other hand, the yield of middle mass nuclei ranging from $A \sim 50$ to 150 given by Dubna Code and Cascade-INPE have large difference which is because of the underestimation of high energy fission of Pb by the Dubna Code. In the same figure isotopic yield in the case of 1 GeV proton colliding with the 1×1 m^2 block of natural uranium are also given for the sake of comparison. It may be seen that the yield of middle mass nuclei is much higher in uranium than in lead. This is because of higher fission cross-section of uranium than lead.

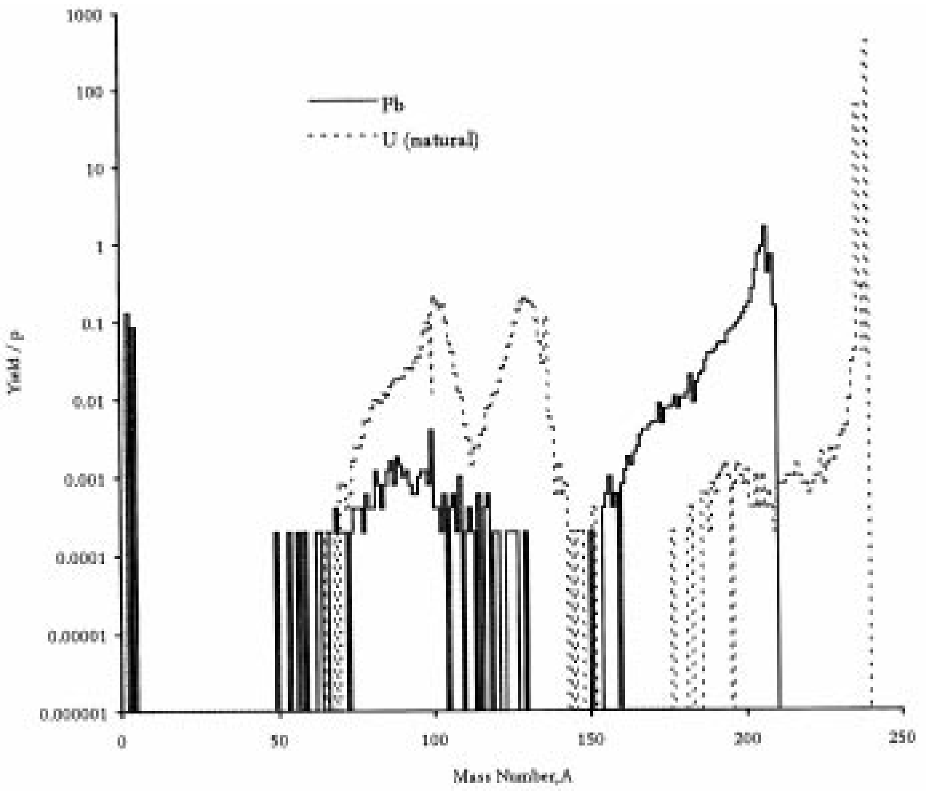

Figure 5. Isotopic distribution of the produced nuclei in collision of 1 GeV proton with (Pb 207.2) and natural uranium targets of dimensions $2R \times L = 1 \times 1$ m^2.

4. Discussions and conclusions

Success of a code depends largely on the reliability of the input data, may be from the experiment or from some physical model and in addition to this it is also desired that similar data is available from more than one experiments. Secondly, for the modeling of transportation of radiation through matter adequate number of the 'groups' and corresponding 'constants' are necessary. Compared to the experimental data of Pb target, W target data is limited. But, it is well-known that W provides a better neutron yield and hence stands a chance to be considered for INSS. From the detailed comparison of multiplicity of neutrons, heat contributions and isotopic yield in large blocks of Pb and U with the Dubna Code, following inferences may be drawn:

(i) Looking at n/p vs. E data of BNL and BNB experiments given in figures 1a and 2 respectively for the Pb target it may be said that the predictions of Dubna Code are well in agreement up to $E \sim 1.2$ GeV and at higher energies the Code underestimates the neutron yield up to $\sim 25\%$. In this energy region although predictions of SONET Code are better than the Dubna Code, it is well known that SONET Code depends on MCNP Code for calculations in the $E < 20$ MeV region [24]. At the same time, looking at Pb data at 1.47 GeV given in table 5, Code results are just in excess to the experimental results when we consider the errors. This observation is in contrast with our earlier observation made in the case of Pb data from the BNL and BNB experiments. This emphasizes that first of all physical models used for the decay of highly excited nuclei need improvement particularly in case of their decay by fission and secondly more detailed experimental data is required for a variety of targets.

(ii) Tungsten has larger capture cross-section for neutrons than Pb and this is duly considered by the Dubna Code. Neutron multiplicity of escaped neutrons given in table 1 is a better way of comparison with the experimental results. It may be seen that the Dubna Code overestimates n/p by nearly 20% in the case of block of W target and that is not small to be avoided. To say the last word about the disagreement, we must first of all test the code with some other experimental data of W target which quotes errors in experimental observations. Secondly, this may also be because of inadequate values of the available constants of tungsten and the code needs their upgradation.

(iii) Variation of n/p with length and diameter of the cylindrical Pb target shows that the Code calculations are in reasonably good agreement with the experimental data up to $\sim$1.2 GeV. This also reveals that the development of cascade along the beam axis and in transverse direction is satisfactory in the Dubna Code. Both the curves, i.e., n/p vs. L in figure 3 and n/p vs. $2R$ in figure 4 tend towards saturation and such information is very useful in designing INSS for ADSS.

(iv) From the data of heat contributions it is clear that in the case of Pb target at low beam energy about 85% of the beam energy is deposited in the target. For 300 MeV proton beam of 1 mA current, heat deposition in the cylindrical Pb target of size $d \times L = 20 \times 35\ \mathrm{cm}^2$ is nearly 8×10^5 J/s. Heat deposition from the non-fission processes such as ionization, γ-rays and recoil nuclei is more in W block than in the Pb block but it suffers from the high activity of γ-rays.

It is important to note from the total heat data in table 6 that there is insignificant energy amplification in the case of lead and tungsten blocks and the code accounts very well for the beam energy conversion into heat.

(v) Dubna Code provides reasonable estimates of very light $(A < 5)$ and very heavy $(A > 150)$ spallation products in the case of both fissionable and non-fissionable targets. In the case of Pb target, middle mass products are estimated poorly by the Code than in the case of uranium. This is because of the fact that reliable data of cross-sections corresponding to high energy fission of Pb is not available. Secondly, uranium has very high fission cross-section than lead and hence the middle mass range is swollen in the case of uranium.

(vi) It is also important to point out that revision of the Dubna Code is a regular feature and on availability of better physical model or the data of constants they are immeditaly incorporated into the code. We are also considering inclusion of change in nuclear characteristics on development of the intra-nuclear cascade in near future.

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