THE CHERNOBYL ACCIDENT: RADIOACTIVE INVENTORY AND RELEASES

by
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A Thesis submitted to the Faculty of Graduate Studies of the University of Manitoba in partial fulfillment of the requirements of the degree of

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Abstract

The Chernobyl accident that happened at the fourth unit of the Chernobyl nuclear power plant (ChNPP-4) almost ten years ago is the biggest accident in the history of the nuclear industry. However, many questions about the radioactivity produced in the reactor and released into the atmosphere still remain unanswered.

Several calculations of the radioactive inventory in the Chernobyl-4 reactor have been made so far, but none of them has calculated the short-term activity. In this thesis an attempt is made to review the available data and calculate the radioactive inventory in the ChNPP-4 reactor due to both, short-term and long-term activities for two different reactor power histories. These are the realistic power history that lead to the explosion while the reactor was operating at low power levels for certain length of time, and an assumed full power level of 32,000 MW (thermal).

The ratio of activities due to constant and realistic power histories was calculated to be about two at the time the prompt critical reaction stopped. This result means that firemen and other emergency workers would have received twice as much dose if the reactor exploded while operating at full power.

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1 Introduction

On April 26, 1986, at about 1:24:00 a.m. an accident occurred at the fourth unit of the Chernobyl Nuclear Power Plant (ChNPP-4) in the Soviet Union. During the accident the reactor core was completely destroyed, and large amounts of radioactive material were released into the atmosphere over the next 10 days.

In August, 1986, at the International Atomic Energy Agency Experts' Meeting in Vienna, the USSR State Committee on the Utilization of Atomic Energy submitted a report *The Accident at the Chernobyl Nuclear Power Plant and Its Consequences* [USSR 86][1]. This report presented information on the Chernobyl type of reactor (RBMK-1000), the chronology of the accident and estimates of releases of various radioactive isotopes (decay corrected to May 6), but it did not report the details of power and fuel history needed to compute radioactive inventory. The accident scenario was not at that time (and still is not) fully understood at the level that would allow reconstruction in detail of the intensity of power excursion leading to the explosion, and accurate calculations of the short-lived reactor inventory.

Three questions have to be answered in order to understand the reactor accident consequences: what was the reactor's radioactive inventory at the time of the accident; what fraction of this inventory had been released; and what are (and/or will be) the effects of these releases?

To get a reliable answer to the question about the reactor inventory at the time of accident we must have detailed information about the reactor power history, fuel content, as well as the reactor design characteristics. However, an estimate of the radioactive inventory of a reactor due to long-lived isotopes can be performed using only a few integral parameters of the reactor[2]. Those calculations were performed before or shortly after the Vienna meeting - [Deve 86], [Kirc 88]. A later Russian publication [Boro 89] provides more detailed information about the ChNPP-4 reactor

[1] After the meeting the International Nuclear Safety Advisory Group summarized the results of the meeting and published its own report [INSA 86]. Part I of the Soviet Report [USSR 86] was published two months later in USSR [Abag 86] which we will use in this text. Annexes from Part II were updated and published later in various Soviet journals.

[2] See below Section 5. "Physics of the Activity Calculations".

history, and makes it possible to obtain better estimates of the reactor inventory.

The estimate of radioactive release is heavily dependent on the experimental data taken during the ten days following the accident. Some of this data, as well as calculations of the release, were presented in Russian scientific literature – [Izra 87,90], [Alek 92] – although discrepancies in published results remain high (see Fig. 18)

The effects of radioactive release (and most important – health effects) depend on the radioactive fallout in the area under consideration. This in turn, depends on the meteorological conditions during the release period, the ability of various radioisotopes to penetrate into different types of soil, etc. The study of these effects requires much additional information.

In this thesis we will consider the first problem, detailed ChNPP-4 reactor inventory, and the second one, radioactive release. Unlike previously published papers, both short-term and long-term activity will be calculated[3].

The long lived isotopes ($T_{1/2} \geq 0.5$ hours) are of main concern while analyzing the effects of the ChNPP-4 radioactive releases on the public. However, at the time of the accident the short-term radioactivity ($T_{1/2} \leq 0.5$ hours) was the major contributor to the total radioactivity released from the reactor. This short-term activity could have affected people working at the four nuclear plants (ChNPP-1, -2, -3 and -4); firemen that arrived after the explosion occurred (three teams, 5, 10 and 20 minutes after the accident); night shift construction workers building the ChNPP-5 and ChNPP-6 reactors; civilians that happened to be in the neighborhood of the ChNPP-4 reactor the night of the accident, and inhabitants of nearby Pripyat before the town was evacuated.

Another important aspect of the ChNPP-4 accident is the fact that the explosion occurred while the reactor was working at a low power level. If the reactor exploded while working continuously at full power, the short term activity accumulated in the reactor would have been larger. How much larger? Would it have affected firefighters? If radiation levels were significantly higher, perhaps firemen wouldn't have been able

[3]We consider only the activity accumulated in the reactor fuel.

to extinguish the fires.

In order to calculate both the short-term and long-term radioactive inventory and to answer the questions raised above, it is necessary to have fairly detailed information regarding the operating regime of the RBMK reactor, as well as the detailed power history of the ChNPP-4 reactor. Short-term activity is very sensitive to these parameters. Much of that information is available in some Russian publications: [Doll 80] and [Spra 83] – about the RBMK design, and [Boro 89] – about the ChNPP-4 reactor history.

2 The RBMK Reactor: Design and Operating Regime

2.1 Some Design Details

The RBMK[4] reactor is a **channel type**, water-cooled and graphite-**moderated**[5] reactor. It has a uniquely Soviet design which is not utilized anywhere else in the world. As of January 1, 1987, 14 RBMK reactors were operating in USSR and 6 were under construction [Lega 87]. The first number grows to 16 units as at December 1, 1990 [Abra 92].

In choosing the first type of reactor for electrical power generation in late 1960's, the RBMK design was chosen as the most effective one[6]. The first RBMK reactor was put into operation in 1973 at Sosnovyi Bor, about 80 km west from St.Petersburg.

The reactor core consists of about 1700 zirconium vertical pressure tubes (channels) that can be filled with fuel or **absorbers**. The core has a cylindrical form with a diameter of 11.8 meters and height of 7 meters.

The RMBK's fuel rods consist of uranium dioxide pellets (2% **enriched**[7]) in a tubular metal (zircaloy) cladding. A cluster of 18 such tubular fuel rods forms a **fuel assembly** – a fuel unit that is inserted into or removed from the reactor. The cross section of such an assembly is shown in Fig. 1a. Under normal operation during the online refueling regime there are about 1660 fuel assemblies in the RBMK reactor.

The core contains enough fuel to constitute as much as 50 critical masses [Kres 87]. Several parameters influence the reactor's reactivity: the coolant, graphite and fuel temperature, the void fraction, the **burnup**, the fuel enrichment, and the presence of absorbing material (including xenon). Such a geometry and design require a large

[4]All four units (two more were under construction) of the Chernobyl complex were RBMK-1000 type of reactors where RBMK stands for large power channel reactor, 1000 stands for 1000 MW electric power.

[5]Reactor terms printed in **bold** are explained in the Glossary.

[6]Although two other types of reactors under consideration have had more safety features, the RBMK was chosen mainly due to experience the Soviet industry had in running graphite reactors: a reactor of similar design had been used in the USSR for weapon plutonium production [Amer 86].

[7]The first RBMK reactors were designed to use 1.8% enriched fuel. In a later design by 2% enrichment was used.

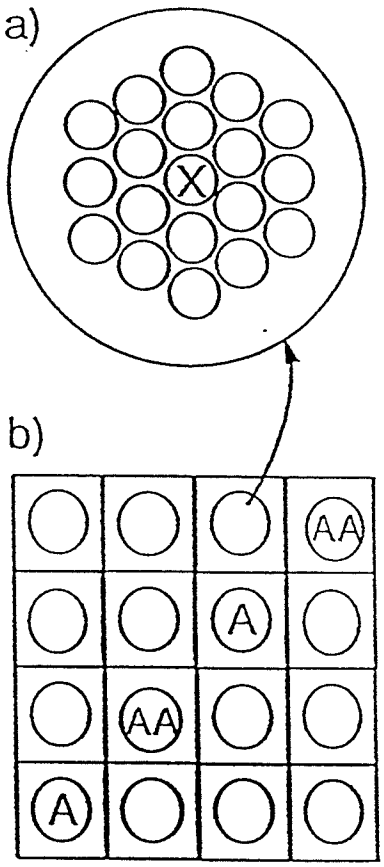

Figure 1: A schematic representation of the distribution of the RBMK fuel in the reactor core. The assembly, part a), consists of 18 tubes of fuel pellets in zircaloy cladding and one channel (marked with X) that under normal operation is filled with water. The assembly is 7 meters long and has two separate 3.5 meter long parts. Initial loading of the RBMK reactor, part b), consist of 12 fresh fuel assemblies, 2 regular absorbers (A) and 2 additional absorbers (AA). Absorbers in AA position will be removed and replaced by fuel during the intermediate period (see Section 2.2. Operating Regime) of the RBMK operating regime.

number of control rods, making the reactor difficult to operate.

The most comprehensive information about the RBMK-type reactor using 1.8% enriched fuel is given in the book [Doll 80] by Dolleshal' (scientific supervisor of the RBMK Project) and Emel'janov. This book is extensively used in this thesis as a source of factual information about RBMK's design.

An analysis of the radioactive content of two types of reactors – WWER (a Pressurized Water Reactor (PWR) type) and RBMK – for different **campaign** times is given in [Spra 83]. Unfortunately, the calculations regarding the RBMK reactor in [Spra 83] were performed under the assumption that the neutron flux is a constant during the campaign. Therefore, the results obtained are applicable only to the online refueling phase of the campaign (see Section 2.2, Operating Regime). This book also contains some useful information regarding the radioactive content of short-term irradiated fresh RBMK's fuel.

Both books – [Doll 80] and [Spra 83] – use the same calculation models, but the latter considers 2% enriched fuel. Although the data given in [Spra 83] were calculated for online refueling regime, and ChNPP-4 was not yet in that regime, we will make use of those data (see Section 8, Results of Calculations: Short Term Activity in ChNPP-4 Reactor).

2.2 Operating Regime

The operating regime of the RBMK reactor depends on the time the reactor has been producing energy. The RBMK campaign time can be divided into three periods [Doll 80]:

- *Start-up period.* Initially the reactor is loaded with fresh fuel. Since fresh fuel has a low neutron absorption rate, special measures have to be taken in order to keep the neutron flux (i.e. reactor power) below a certain level determined by the reactor design. There are three principal methods to solve this problem: first, to use low enriched fuel with a lower neutron production rate; second, to use additional absorbers and third, to use fewer fuel assemblies. In the RBMK

design the last two methods were implemented [Doll 80]. During the start-up period the reactor has only about 80% of its fuel loaded and twice as many absorbers as during steady state operation (see Fig. 1b). As the campaign goes on, **fission products** are accumulated and the non-fission neutron absorption rate increases. In order to compensate for the loss of neutrons due to absorption in these fission products, additional absorbers are eventually removed and replaced by fuel assemblies.

- *Intermediate period.* The intermediate period starts when the first additional absorber is replaced by fuel. This occurs when the average burnup is about 5 **MWd**/kg (megawatt-days per kilogram of fuel) [Doll 80]. While the reactor fuel is further burning out, the second additional absorber is also replaced by fuel. At the end of this period the average fuel burnup has reached half the level (10-12 MWd/kg) of the maximum burnup that can be achieved when both additional absorbers are removed.

- *Steady State or Online Refueling regime.* During this period the fuel with burnup 20-24 MWd/kg is removed from the core and replaced with fresh fuel. The reactor fuel then is a mixture of old and fresh fuel and the average burnup during this period is roughly constant[8]. The refueling procedure is performed while the reactor is producing energy. During this operating phase the RBMK reactor has 14 fuel assemblies and 2 absorbers in every 16 (4x4) channels.

According to [Doll 80], the first two periods in total last for about 1500 days for RBMKs with 1.8% enriched fuel[9] while the design life of an RBMK reactor is considered to be about 30 years [Doll 80].

The main integral parameter of a power reactor is the total amount of energy produced. Often it is expressed in a specific form: average energy per fuel assembly or per kilogram of fuel. This parameter is called **fuel burnup** or **fuel exposure**.

[8]This fact – constant burnup – greatly simplifies the calculations and is often used in RBMK inventory estimates [Spra 83].

[9]It is not stated explicitly what is the length of this period for RBMKs with 2% enriched fuel.

Another important parameter is the **neutron fluence**, neutron flux multiplied by time: ΦT. The long-lived isotope activity is directly proportional to the fluence (see section 5.2 Fission Products Activity). In addition, the fragility of the reactor vessel as well as the metal constructions in the reactor depend on the neutron fluence.

3 Power History of the ChNPP-4 Reactor

The ChNPP-4 reactor was started – "turned on" – in December, 1983 and thus had been operating for over 800 days[10]. Taking into account the routine testing procedures [Doll 80] during the first few months after starting an RBMK reactor, ChNPP-4 was producing electricity for 735 days [Boro 89][11].

On April 26, the reactor was planned to be shut down for scheduled maintenance, but a decision was made to run a special test before the shutdown. The goal of this test was to determine the ability of a turbogenerator to provide in-house power after shutting off its steam supply for the short time (nominally 40 to 50 seconds) needed for the emergency diesel generators to start and come online. The accident occurred while performing this test.

For the purpose of calculating the radioactive inventory, we split the ChNPP-4 power history into three intervals which we will consider separately.

3.1 *A-Period*

This period covers the time between connecting the ChNPP-4 reactor to the Soviet electricity grid in December 1983 and 01:06 a.m. April 25, 1986, when preparations to perform the special test were started. During this time (about 25 months) the reactor was operating under normal conditions delivering thermal power of 3200 MW (1000 MW electric) except when it was shut down for maintenance or repair. Information given by the Soviet team at the Vienna meeting [Abag 86] regarding the reactor characteristics (burnup, number of fuel assemblies in the reactor) corresponds to the end of this period, but does not provide enough data to reproduce in detail the power history until that time. More detailed information about the fuel history in the ChNPP-4 reactor was given in a preprint [Boro 89] published a few years after the

[10]The Chernobyl Complex (4 RBMK type reactors) was the third one built in the Soviet Union, after the Leningrad and Kursk Complexes.

[11]There are some discrepancies in different publications regarding this figure. For example, authors in [Alek 92] state that the reactor was operating for "715 effective days" - presumably full-power days. We use the number from [Boro 89] since this preprint gives detailed history for different fuel assemblies in the ChNPP-4 reactor.

accident and is reproduced in Table 1. Fig. 2 shows the assumed power history of the ChNPP-4 reactor used in our calculations. In this preprint the refueling timetable is given. All 1659 fuel assemblies that were present in the reactor at the time of the accident are divided into 18 groups using the exposure level and time the fuel from each group was present in the reactor. These data give us two important integral characteristics of the reactor fuel at the time of the accident:

- Total fuel burnup: 2069 GWd – see last line in column 4, Table 1. This gives an average power level of 2815 **MWt (thermal)** over 735 days of operation (nominal power - 3200 MWt[12]) and specific average burnup per kilogram of fuel – 10.9 MWd/kg (last line in column 6, Table 1): this number varies from 0.4 MWd/kg to 14.4 MWd/kg for different groups of fuel assemblies. In [Abag 86] the average burnup was reported to be 10.3 MWd/kg.

- Total number of fuel assemblies integrated over time. This fuel was present in the reactor: $1.06 * 10^6$ Assd (**assembly-days**) – see column 3, Table 1.

The information given in [Boro 89] also shows that no fuel had been removed from the reactor during the campaign. According to [Boro 89], 1383 fuel assemblies (83% of all assemblies at the time of the explosion) had been present in the reactor during the whole campaign and 276 fuel assemblies were added at different times starting from the 295th day of the campaign (see Table 1. and Fig. 2.). According to [Doll 80], RBMK reactors during the start-up period have only about 83% (160 tons of 192 tons) of their fuel loaded (see Section 2.2, Operating Regime). Although this information applies to RBMK's with 1.8% enriched fuel, we assume this number should be about the same for ChNPP-4 fuel with 2.0% enrichment. Therefore, if some fuel in the ChNPP-4 reactor would have been removed (replaced by fresh fuel), the number of fuel assemblies present in the reactor from starting the campaign would be under 83%, what apparently is not our case. Moreover, we notice that ChNPP-4 was

[12]This gives us the **capacity** factor of the ChNPP-4 reactor: $K = 2815 : 3200 = 0.88$. This number is in good agreement with the capacity factors of other RBMK reactors that are typically in the range of 0.8 to 0.9 [Abag 91].

Table 1: History of the ChNPP-4 reactor fuel. Data in columns 1,2 and 5 were taken from [Boro 89], in columns 3,4 and 6 - calculated: column 3 - multiplying the corresponding figures in column 1 and 2 - M*T, this gives us the total number of *assembly-days* (Ass d) for each group of fuel; column 4 - multiplying the corresponding figures in column 5 and 2 - B2=B1*M; column 6: dividing the figure in column 5 by the amount of fuel in one assembly - B3=B2/114.7 [Abag 86]. The average fuel burnup can be calculated by dividing the total energy (2,068.95 GWd - gigawatt days) by the total amount of fuel in the reactor at the time of explosion: 1659*114.7=190.29 tons. This gives us an average burnup of 10.9 MWd/kg.

Time in the core, T (days)	**Number of assemblies, M**	**Total Fuel, M*T (Ass d)**	**Burnup, B1 (GWd)**	**B2 (MWd/Ass)**	**B3 (MWd/kg)**
735	146	107310	240.9	1650	14.39
735	575	422625	891.25	1550	13.51
735	261	191835	378.45	1450	12.64
735	131	96285	176.85	1350	11.77
735	79	58065	98.75	1250	10.9
735	75	55125	86.25	1150	10.03
735	66	48510	69.3	1050	9.15
735	35	25725	33.25	950	8.28
735	15	11025	12.75	850	7.41
440	3	1320	2.55	850	7.41
388	17	6596	12.75	750	6.54
336	28	9408	18.2	650	5.67
285	15	4275	8.25	550	4.8
233	23	5359	10.35	450	3.92
181	18	3258	6.3	350	3.05
129	34	4386	8.5	250	2.18
78	74	5772	11.1	150	1.31
26	64	1664	3.2	50	0.44
TOTAL	**1659**	**1,058,543**	**2,068.95**		

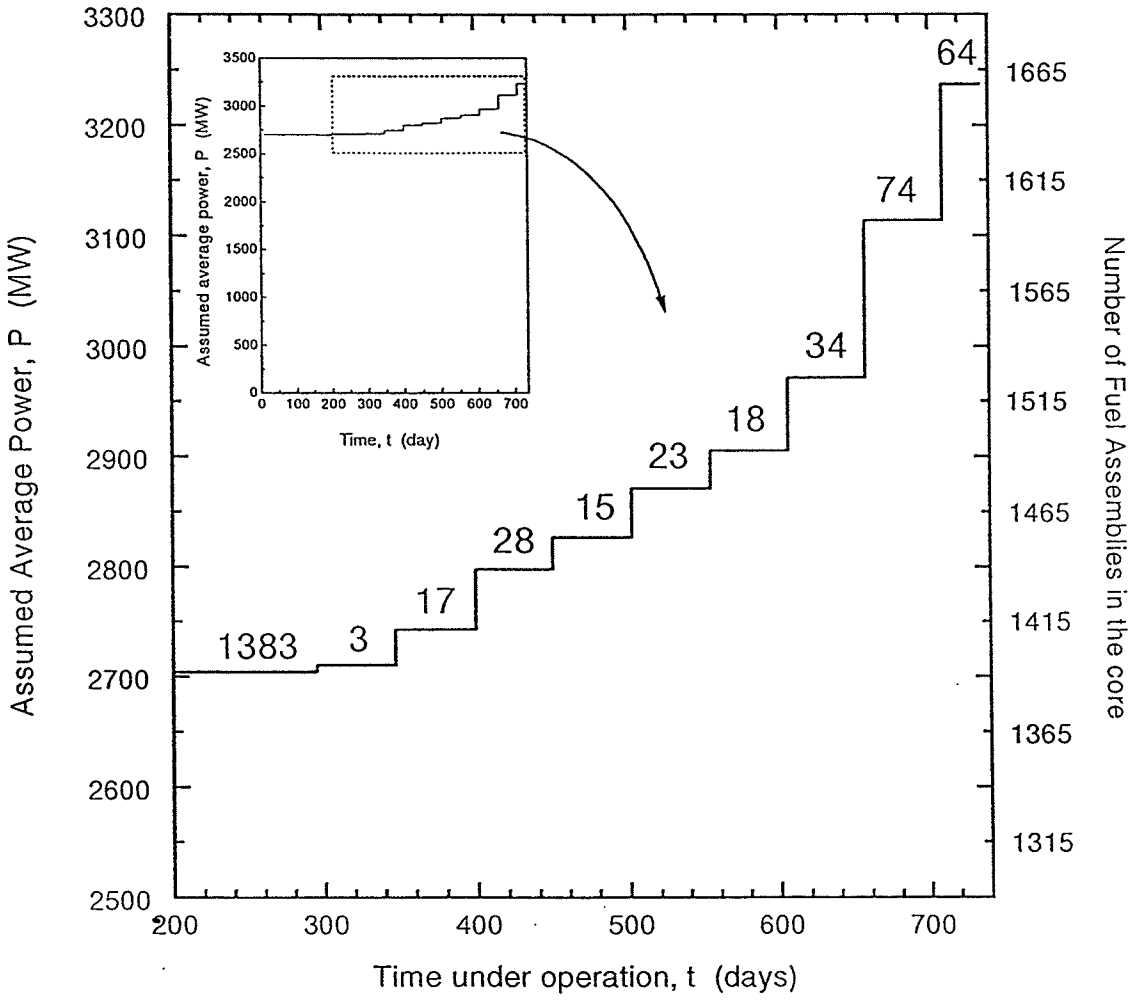

Figure 2: Power history of the ChNPP-4 reactor used in calculations. It is assumed that each of 18 groups of fuel assemblies (specified in Table 1) had the same energy production rate (i.e. constant power) during the time the fuel was in the core. For the last 26 days the ChNPP-4 reactor was working at about a nominal power level - 3200 MW. The figures on the "stairs" show the number of fuel assemblies added at different times.

just about to enter the *online refueling regime* (during which the refueling process takes place; see Section 2.2, Operating Regime), because 190.2 tons of fuel were in the reactor, whereas some 192 tons is the normal amount of fuel for this regime [Spra 83]. Therefore, we conclude that no fuel had been removed from ChNPP-4, hence all the radioactivity produced in the reactor was inside the core at the time of explosion.

3.2 *B-Period*

This period covers the next 24 hours, from the time preparations for the test started (reduction in power level to some 700-1000 MWt) to pressing the AZ-5[13] button. A few hours into the preparations, when the reactor power was half the nominal level – 1600 MWt, further power reduction was stopped. The order to keep the reactor running at this power level came from the Kiev energy dispatcher [Abag 86, Alek 92]. At 23:00 p.m., after 20 hours working at half nominal power level, the dispatcher gave permission to continue the test. After several power level changes during the next hour the test was started at 01:23 a.m. on April 26 with the reactor operating at a power level of 200MWt. A few minutes later an operator pressed the AZ-5 button to shut down the reactor, but the reactor went out of control and exploded.

The Soviet report in Vienna did not provide detailed information for this period. In some later Russian publications – [Alek 92], [Abag 91] the missing information about the power level history was given and is summarized in Fig.3.

3.3 *C-Period*

This period[14] covers the time between pressing the AZ-5 button to the end of the explosion. Observers outside the plant heard two explosions reportedly about 2 seconds apart and saw hot fragments and sparks shoot into the sky above the reactor. A number of fires were started as these burning fragments fell onto the turbine hall and the reactor building roof.

[13]By pressing the AZ-5 button (known in the Western nuclear industry as *scram* button) all movable absorbers used for controlling the reactor power are sent into the reactor core to stop the chain reaction.

[14]For discussion about the length of this period see below Section 8.2.2, *C-period* Activity.

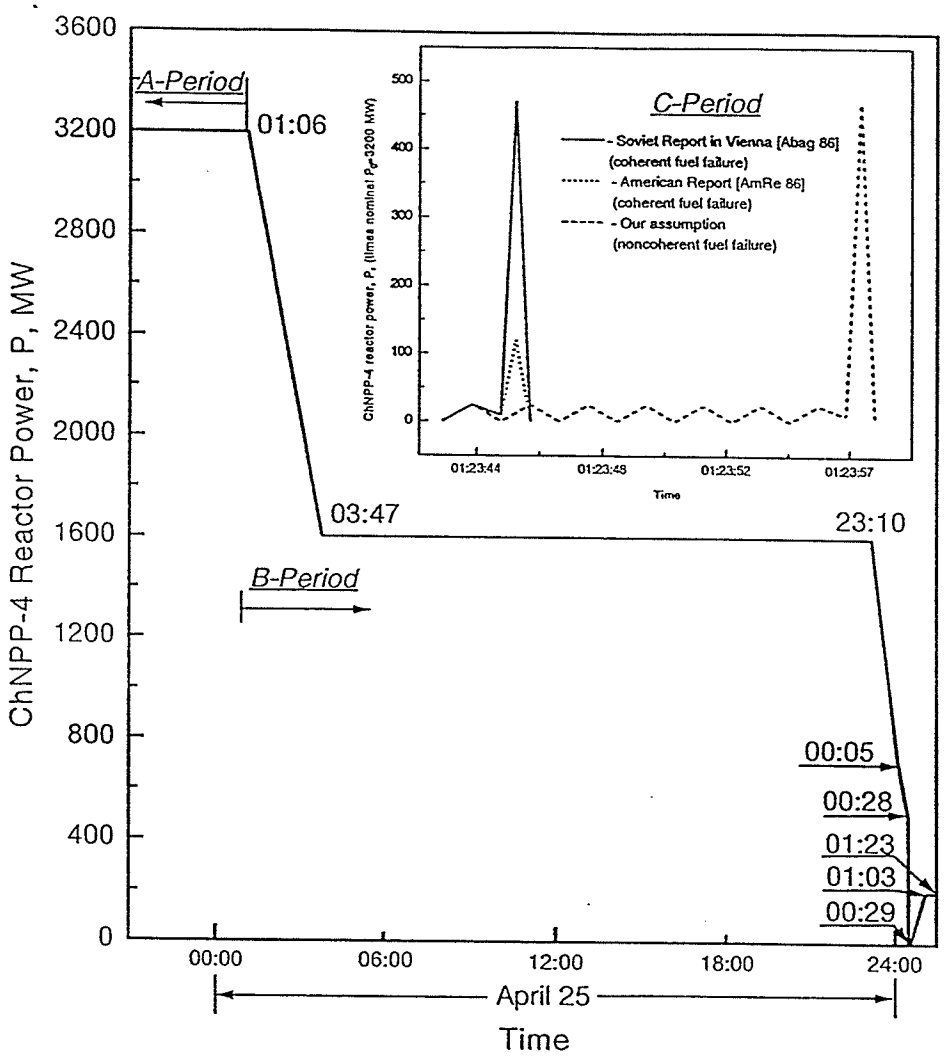

Figure 3: Power history of the ChNPP-4 reactor as presented in the Soviet Report in Vienna [Abag 86] and later corrected in some Russian publications ([Abag 91],[Alek 92]). The time for which the ChNPP-4 reactor was under operation is divided into three periods - A,B,C. Three different models describing C-Period are shown (see Section 8, Short-term Activity in the ChNPP-4 Reactor).

Although the Soviet team in Vienna presented their *calculations* of the reactor power during this time, as well as changes in reactivity and the temperature of the coolant and graphite, it is not quite clear under what assumptions those calculations were performed (see Section 8.2.2, *C-period*: Fission Product Activity)

4 Previous Calculations of the ChNPP-4 Reactor Inventory

4.1 Soviet Report in Vienna

The first information about the radioactive inventory of the Chernobyl reactor at the time of the explosion, as well as the amount of radioactivity released during the first 10 days after the accident, was given in the Soviet report in Vienna [Abag 86] and is reproduced in Table 2 and Table 3.

These Soviet data [Abag 86] were presented without much explanation and require some comments. It is stated that the activity presented in column 3 in both tables is decay corrected to May 6, 1986, 10 days after the accident. In addition, it is stated that the data in column 3, Table 2 give the releases "up to May 6, 1986" [Abag 86]. However, figures in this column add up to only 35 MCi instead of 50 MCi given in the Soviet Report. Thus, it is not clear if these data include the releases on the day of the accident, April 26 (column 2) and if these data are also decay corrected to May 6.

The activity level measurements were started immediately after the accident [Alek 92]. However, the Soviet report [Abag 86] gives only the activity due to the long lived isotopes (both, total amount and the fraction released) as of May 6, 1986. Since the activity due to nuclides with half-lives shorter than 1-2 days have decayed by that time, no information about them is given. Therefore, the data in [Abag 86] can be used only for estimates of the long term effects of the Chernobyl explosion. What were radioactivity levels during the first ten days after the explosion?

4.2 Other Inventory Calculations

Although detailed information regarding ChNPP-4 history became available only a few years after the accident, some attempts to estimate the Chernobyl inventory were performed outside the USSR, one of them right after the accident.

Ten days after the accident *Nature* received an article written by Devell et al. [Deve 86] where the ChNPP-4 radioactive inventory estimates based on the first fallout

Table 2: Radioactivity of the releases from the ChNNP-4 reactor as reported in the Soviet Report in Vienna (decay corrected to May 6) [Abag 86].

Nuclide	Activity of release, A (MCi) April 25, 1986	May 6, 1986	Fraction of activity released by May 6, 1986 (%)
Xe-133	5	45	up to 100
Kr-85m	0.15	-	up to 100
Kr-85	-	0.5	up to 100
I-131	4.5	7.3	20
Te-132	4	1.3	15
Cs-134	0.15	0.5	10
Cs-137	0.3	1	13
Mo-99	0.45	3	2.3
Zr-95	0.45	3.8	3.2
Ru-103	0.6	3.2	2.9
Ru-106	0.2	1.6	2.9
Ba-140	0.5	4.3	5.6
Ce-141	0.4	2.8	2.3
Ce-144	0.45	2.4	2.8
Sr-89	0.25	2.2	4
Sr-90	0.015	0.022	4
Np-239	2.7	1.2	3.2
Pu-236	0.0001	0.0008	3
Pu-239	0.0001	0.0007	3
Pu-240	0.0002	0.001	3
Pu-241	0.02	0.14	3
Pu-242	0.0000003	0.000002	3
Cm-242	0.0003	0.021	3
Total (no noble gases)	~15	~35	

Table 3: ChNPP-4 radioactivity releases (decay corrected to May 6) on a daily basis (**noble gases** excluded) as presented in the Soviet Report in Vienna [Abag 86]. For a graphical representation see Fig. 18.

Date	**Days after explosion**	**Activity, A (MCi)**
April 26	0	12
April 27	1	4.0
April 28	2	3.4
April 29	3	2.6
April 30	4	2.0
May 1	5	2.0
May 2	6	4.0
May 3	7	5.0
May 4	8	7.0
May 5	9	8.0
May 6	10	0.1
May 9	13	0.01
May 23	27	0.00002
Total		**50**

measurements done in Sweden were given. Not knowing any details about the reactor, the authors used the measured ratio of Cs^{134} to Cs^{137} activity to conclude that the fallout came from a nuclear reactor (no significant amount of Cs^{134} is produced in a nuclear bomb explosion) and that the reactor was under operation for about 400 days (assuming uninterrupted production of energy at a 1000 MWe **(electric)** power level). Using this information they then calculated the radioactive inventory of several isotopes in the reactor – see column 3, Table 4.

A similar approach was used by German scientists Kirchner and Noack [Kirc 88]. Using the measured ratios of Cs^{134} to Cs^{137} activity in the radioactive fallout in Munich, West Germany, the authors calculated that the average core burnup at the time of the accident was 12.9 MWd/kg, and concluded that the reactor was under operation for "722 equivalent-full-power-days". Based on these results, they then calculated the activity of several isotopes in the ChNPP-4 reactor (see column 5, Table 4).

According to [Boro 89], ChNPP-4 reactor had been operating for 735 days at an average power level of 2815 MWt (i.e. 880 MWe, since 3200 MWt corresponds to 1000 MWe). Therefore, we arrive at 646 "equivalent-full-power-days". Comparing this figure with the results of [Deve 86] and [Kirc 88], we can conclude that Devell et al. underestimated and [Kirc 88] overestimated the radioactive inventory (due to isotopes with half-lives longer that the campaign time – see Section 5, Physics of the Activity Calculations) in the ChNPP-4 reactor.

In addition, in both of these estimates the fallout in specific countries was used, and it was assumed that the releases were representative of the *average* core. Since the real burnup of different groups of fuel assemblies present in the ChNPP-4 reactor at the time of the accident changes in a wide range from 0.4 MWd/kg to 14.4 MWd/kg (see Table 1, column 6), it is difficult to obtain a correct result on the basis of these fallout measurements alone.

In 1990 a scientific team from the Kurchatov Institute (a leading nuclear research institution in Russia) reported the corrected values of the Chernobyl inventory on the

Table 4: Inventory activity (MCi) from different papers and calculated in this thesis for two power scenarios: constant power (ConstP column, it is assumed that during B-Period the ChNPP-4 reactor was working at full power level) – for comparisons with other results; real power (RealP column, the real power curve for B-Period from Fig. 3 was used) – for comparisons with ConsP results. For the listed long lived isotopes the difference in these results is not significant. The C-Period (explosion itself) was not taken into account.

Isotope	Halflife, hours	Different models						Present thesis	
		Deve 86	INSA86	KiNo 88	Gudi 89	Izra 90	Boro 89	ConsP	RealP
Fission Products									
Kr-85	93972.2		0.8		0.55			0.75	0.75
Sr-89	1211.94		62	90	80	52		97.5	96.5
Sr-90	255250.		5.4	6.2	4.2	5.2	5.9	5.6	5.6
Zr-95	1538.33		132	154	134	130		155	154
Mo-99	66.00		154		137	130		168	140
Ru-103	942.78	38	132	128	116	130		115	113
Ru-106	8836.11		54	33	29	52	23	24.9	24.9
I-131	192.97	27	83	66	82	90		86.4	81.7
Te-132	78.19	38	73	56	109	120		125	107
Xe-133	125.89		171		193			185	180
Cs-134	18075.0		5.1	4.2	3.7	4.0	4.1	4.1	4.1
Cs-137	264472.	1.4	7.8	8.2	5.6	7.2	7.0	6.8	6.8
Ba-140	306.94		135.	133.	140.	130.		163	157
Ce-141	780.00	49	147.			130.		149	148
Ce-144	6825.00		87.	113.	82.	90.	106.	101	101
Actinides									
Np-239	56.50	503.	720.	560.		1300.	1570.	1510	1410
Pu-239	0.21+11			0.024		0.023	0.03	0.022	0.022
Pu-240	0.574+8			0.05		0.033	0.04	0.038	0.038
Pu-241	128900.			5.7		4.6	5.0	4.4	4.4
Pu-242	0.33+10			6.7-E5			5.6-E5	7.2E-5	7.2E-5

basis of measurements performed in the fall of 1987 [Boro 89]. Again, these data take into account only the long lived isotopes and therefore don't answer the question of the inventory just after the accident (see column 8, Table 4).

5 Physics of the Activity Calculations

The time dependence of the number of nuclei (both, fission products and **actinides**) of a given isotope, $N_k(t)$ can be described using the following equation:

$$\frac{dN_k(t)}{dt} = -S_k N_k(t) + \sum_{(k')} S_{k' \to k} N_{k'}(t); \qquad N_k(0) = N_0, \tag{1}$$

where the first term on the right hand side describes the "disappearance channels" for $N_k(t)$ (decay, (n, γ), etc.) and the second term – the "source channels" for $N_k(t)$ production (direct production through fission (for fission products), decay of the parent isotope, etc.).

Fig. 4 shows the "disappearance" channels for the k-th isotope. The same figure can be used for the source channels k' that "feed" the k-th isotope i.e. k is the product

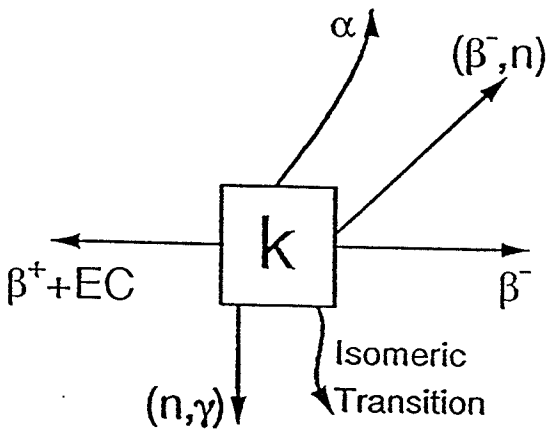

Figure 4: Schematic diagram of the isotope's "disappearance" channels. Each arrow represents a reaction that causes the "disappearance" of the k-isotope and creation of the k'-isotope (see equation (1)). The k-isotope, in turn, is produced through the same type of reactions from its parent isotopes.

of the reactions shown on Fig. 4. We can notice that in general the k-th isotope

can be "chained" to isotopes with different mass numbers A. The disappearance channels for such chains can be easily taken into account by putting the corresponding cross sections in S_k. But we can't do the same with the source term $S_{k'\to k}$: the corresponding cross sections apply to unknown $N_{k'}(t)$ that should also be calculated from a similar equation. The number of k' for which $S_{k'\to k}$ is non-zero gives the number of equations we have to solve simultaneously.

In our further calculations we will consider only two types of sources for $N_k(t)$ production: decay or (n,γ) reactions in the parent isotope. In addition, we assume that the given isotope $N_k(t)$ has only one parent isotope, i.e. only one of these terms is present in the equation.

By replacing the S_k and $S_{k'\to k}$[15] in equation (1) we get the equation for calculating the number of nuclei:

In case of decay of the parent isotope:

$$\frac{dN_k(t)}{dt} = -(\lambda+\sigma_{abs}\Phi)_k N_k(t)+\lambda_{k-1}N_{k-1}(t)+\Phi(t)\sum_{i=1}^{3} y_i\sigma_i^f U_i(t); \qquad N_k(0)=N_k^0 \quad (2)$$

In case of (n,γ) reaction in the parent isotope:

$$\frac{dN_k(t)}{dt} = -(\lambda+\sigma_{abs}\Phi)_k N_k(t)+\sigma_{(n,\gamma)}\Phi N_{k-1}(t)+\Phi(t)\sum_{i=1}^{3} y_i\sigma_i^f U_i(t); \qquad N_k(0)=N_k^0 \quad (3)$$

where $k=[1,L]$, L -- number of isotopes in the chain;

t - time;

$N_k(t)$ - number of fission product atoms per unit volume[16];

$(\lambda+\sigma_{abs}\Phi)_k$ - "disappearance" term: describes the disappearance channels for isotope nuclei - decay, (n,γ), etc.;

λ - radioactive constant;

σ_{abs} - total absorption cross section of the k-th isotope in the chain;

[15]Since we have only one parent isotope, we change the notation from k' to $k-1$ in the source term.

[16]Since we don't have any information about spatial distribution of the reactor parameters, in our calculations we will use the **point-kinetic** approximation and use averaged reactor parameters – reactor power, neutron flux, number of nuclei per unit volume of fuel, etc. Therefore, to get the total number of nuclei in the reactor we just multiply $N_k(t)$ by the total volume of the reactor fuel.

$\Phi(t)$ - neutron flux;

$N_{k-1}(t)$ - number of nuclei of the parent isotope per unit volume;

y_i - fission fraction;

σ_i^f - fission cross section;

$U_i(t)$ - number of fissionable atoms per unit volume, $i = 1, 2, 3$. The three terms correspond to U^{235}, Pu^{239} and Pu^{241}, respectively.

Furthermore, we will also use $F_k(t) = \Phi(t) \sum_{i=1}^{3} y_i \sigma_i^f U_i(t)$.

5.1 Fission Products: One Isotope per Chain

For the fission products $N_k^0 = 0$ since they are not presented initially in the core. In the case where we have one isotope per chain, the parent term is zero, and assuming $\lambda \gg \sigma_{abs}\Phi(t)$, only one fissionable isotope is present in the reactor (i=1, $F_k = \Phi y \sigma^f U$) and only $N_k(t)$ is time dependent, we have a simple differential equation (omitting the indexes)

$$\frac{dN(t)}{dt} = -\lambda N(t) + \Phi y \sigma^f U \quad ; \quad N(0) = 0. \tag{4}$$

Its solution after a campaign time of length T can be written in form

$$N(T) = y\sigma^f \Phi U (1 - e^{-\lambda T})/\lambda. \tag{5}$$

In the case $\lambda T \gg 1$ or $T \gg T_{1/2}$, this solution can be written in form

$$N(T) = y\sigma^f \Phi U/\lambda, \tag{6}$$

and therefore the activity

$$A(T) = y\sigma^f \Phi U. \tag{7}$$

In this case the radioactivity does not depend on the campaign time T (N for such isotopes has reached the saturation level) and is directly proportional to the neutron flux Φ.

For fission products with $\lambda T \ll 1$ or $T \ll T_{1/2}$, keeping two terms in a Taylor series for $e^{-\lambda T}$ gives the solution (5) in form

$$N(T) = y\sigma^f U\Phi T, \tag{8}$$

and the activity

$$A(T) = y\lambda\sigma^f U\Phi T. \tag{9}$$

The result is proportional to the neutron fluence, ΦT. In any reactor there are many fission products with $\lambda T \sim 1$ and therefore neither one of the approximations can produce correct results in calculations of the activity for those isotopes.

5.2 Fission Products: Two or More Isotopes per Chain

The result becomes more complicated when we use more than one isotope per chain and include fissionable nuclides that are produced in the reactor during the campaign, such as Pu^{239} and Pu^{241}. In this case, even for long lived isotopes with $T \ll T_{1/2}$ the neutron fluence ΦT is not anymore a universal parameter that completely defines $N(T)$, and therefore the radioactivity of the given isotope.

Nevertheless, these simple formulas help us to understand the difference in long-lived and short-lived radioactivity production. The activity of the short-lived isotopes will reach their saturation level which is directly proportional to the neutron flux. The activity of the long-lived isotopes is proportional to the neutron fluence or (since the reactor power is proportional to the thermal neutron flux) to the total energy produced in the reactor. This in turn justifies using the burnup (specific energy produced) as a parameter in activity calculations, but limits this approach only to long-lived isotopes that are not burned up and to cases with little amount of secondary fuel produced.

5.3 Actinide Activity

Unlike for the fission products, for actinides $F_k \equiv 0$ since they are not produced in fission. For the first isotope in chain -- U^{238} or U^{235} in our case – we again don't have the source term, and the exact solution for a campaign time of length T is

$$N(T) = N_k^0 exp(-\lambda T - \int_0^T \sigma_{abs}\Phi(t)dt). \tag{10}$$

For the rest of the actinides $N_k(0) = 0$, but we cannot write down an exact solution since the source term is not zero. If the actinide of interest is produced through a (n, γ) reaction in the parent actinide, we arrive at the same form of equation (3) as in the case of fission products by replacing $y\sigma^f$ with $\sigma_{(n,\gamma)}$, and U with N_{k-1}. We can again use the results from previous section.

If an actinide is produced by decay of its parent, equation (10) becomes as follows (σ_k=$(\sigma_{abs}\Phi)_k$):

$$\frac{dN_k}{dt} = -\sigma_k N_k + \lambda_{k-1} N_{k-1} \quad ; \quad N_k(0) = 0. \tag{11}$$

The solution can be written in the form[17]

$$N_k(T) = \lambda_{k-1} e^{-\sigma_k T} \int_0^T N_{k-1}(x) e^{\sigma_k x} dx. \tag{12}$$

Unlike the fission products, most of the actinides fall into two groups -- short-lived ($T_{1/2} \leq 30d$) and long-lived ($T_{1/2} \geq 15y$), with few exceptions that do not contribute significantly to the activity produced (Pu^{236} with $T_{1/2} = 2.9y$ and Cm^{242} with $T_{1/2} = 163d$). Therefore, with some caution we can use the above formulas (10) and (12) for evaluating of the amount of long-lived actinides produced in the reactor during the campaign.

5.4 Formula for Numerical Calculations

Now we can write down an equation that we will use in our calculations (it covers both cases -- fission products and actinides):

$$\frac{dN_k}{dt} = F_k(t) - \sigma_k N_k + \sigma^k_{k-1} N_{k-1} \quad ; \quad N_k(0) = N^0_k. \tag{13}$$

where σ_k=$\lambda + \sigma_{abs}\Phi$ and σ^k_{k-1} =λ_{k-1} in the case the of decay of the parent, and σ^k_{k-1}= $\sigma_{(n,\gamma)}\Phi$ in the case of the (n, γ) reaction.

[17]Here we assume that the neutron flux is constant in time. Although this is not always the case, we can achieve this by dividing the campaign time T into small intervals where this assumption will be reasonable.

To treat this set of equations as a set of simple first order differential equations in our further calculations we assume that both σ_k and σ_{k-1}^k are time-independent. Apparently this is not our case, since the neutron flux Φ can change significantly in time. To make this assumption acceptable, we divide the reactor campaign time into intervals during which the neutron flux *can be* considered as constant. So, we will use a 1 day time interval while calculating the activity produced in the ChNPP-4 reactor during the *A-period* [18]. While calculating the activity produced during *B-Period* we will use, where necessary, the average neutron flux for each of its intervals with different neutron flux levels[19] (see Fig.3).

The general solution of equations (2) and (3) that we will use in our further calculations is given in Appendix A[20].

5.5 Selecting Isotopes

Today we know about 1200 fission products, whereas only 877 of them have well known half-lives and neutron cross-sections and are listed in the ENDF/B-V nuclear data tables [EPRI 84]. While calculating the radioactive inventory of an RBMK reactor for campaign lengths of 800, 1100 and 1400 days, the authors of [Spra 83] used 520 radioactive fission products (A=72 – 166) and 58 actinides (A=231 – 257) with half-lives $T_{1/2} \geq 0.1$ sec [21]. The results show that the total activity in an RBMK reactor ten days after shutdown was only 10% of the activity one second after the shutdown. Although the model that has been used in these calculations cannot be directly applied to the Chernobyl reactor (see below Section 8.1 Constant Power Regime), it gives the order of magnitude of the difference. In addition, the huge power excursion that reflects the explosion in the ChNPP-4 reactor (see Fig. 3, *C-period*) will also produce short-term radioactivity and therefore requires special

[18] The results obtained for each such interval will be used as initial conditions for the next interval.

[19] For more details see Section 6, Calculational Model.

[20] Further in this text we will refer to the general equation (A.1) (Appendix A) that covers both cases – (2) and (3).

[21] About 25% of fission products (short lived with $T_{1/2} \leq 5$ min whose contribution is significant only during the first half an hour after the chain reaction stops) don't have accurate experimental data, and therefore some simple analytical formulas [Yosh 77] were used.

consideration.

To reduce the number of equations and simplify the calculation procedure, we can make the following approximations: we neglect the isotopes whose

- half-lives are smaller than some arbitraryly chosen half-live, $T^*_{1/2}$; and
- contributions to the total activity accumulated in the reactor are smaller than an arbitrary chosen value.

The first approximation limits the applicability of the results obtained: they can be used only for times 5 to 6 times the value of $T^*_{1/2}$, as by that time the neglected isotopes have almost completely decayed. The second approximation just introduces a correction factor to the final result and – if the cut-off level is chosen low enough – will not change significantly the final result[22].

5.5.1 Chosen Isotopes

In our calculations we will use both of these approximations and neglect isotopes

- with half-lives smaller than $T^*_{1/2} = 0.5$ hours. Therefore, the activity calculated using this cut-off level will be applicable for times $t > 3$ hours after the chain reaction stopped (i.e. the reactor exploded). To get the activity for times $t < 3$ hours we will use another approach (see Section 8, Short-term activity in ChNPP-4 Reactor);
- whose contribution to the total activity accumulated in the reactor is less than 3% of the activity of Cs^{137} accumulated in the reactor[23].

These assumptions reduce the number of isotopes under consideration to 97 (see Table 5) and allow us to use the formulas derived in the previous section. In most cases we will have only 1-2 isotopes of interest per chain, always in the parent-daughter relationship. In addition, few fission product chains (see Fig. 5) will require special

[22] In most cases this cut-off can easily be taken into account. For more details see Section 6, Calculational Model.

[23] To do so, we must *know* the activity due to Cs^{137}. In this case we will use the figure from [Spra 83] for a campaign time of 800 days.

Table 5: Fission fractions for fissionable isotopes used in calculations [EPRI 84]. Only isotopes with at least one non-zero fission fraction are presented. All isotopes are listed in Table 7.

Isotope	Halflife (hours)	Fission Fraction, %		
		U235	Pu239	Pu241
Ge-78	1.45	0.02	0.03	0.01
Br-83	2.39	0.53	0.30	0.20
Br-84	0.53	0.99	0.48	0.35
Kr-85m	4.48	1.17	0.50	0.35
Kr-85	10.7y	0.13	0.06	0.04
Kr-87	1.27	2.53	0.82	0.73
Kr-88	2.84	3.59	1.30	0.94
Sr-89	50.4d	4.75	1.72	1.21
Sr-90	29.1y	5.85	2.12	1.49
Sr-91	9.48	5.97	2.44	1.79
Sr-92	2.71	5.92	3.00	2.21
Y-92	3.54	0.05	0.03	0.02
Y-93	10.2	6.36	3.95	2.90
Zr-95	64.1d	6.51	4.98	4.02
Zr-97	16.9	6.00	5.54	4.81
Nb-98	0.86	5.81	5.87	5.66
Mo-99	66.0	6.14	6.19	6.17
Ru-103	39.3d	3.15	6.79	6.87
Ru-105	4.44	0.98	5.54	6.16
Ru-106	1.01y	0.39	4.29	6.10
Ag-110m	250d	0.02	0.54	1.21
Ag-111	7.5d	0.02	0.27	0.56
Pd-112	21.1	0.01	0.10	0.21
Ag-113	5.37	0.01	0.08	0.16
Cd-115	53.5	0.01	0.35	0.40
Cd-118	0.84	0.01	0.04	0.03
Sn-121	27.1	0.01	0.04	0.02
Sn-123m	0.668	0.02	0.07	0.03
Sn-125	9.6d	0.02	0.06	0.02
Sb-125	2.8y	0.01	0.06	0.02
Sn-127	2.10	0.11	0.40	0.20
Sb-127	92.4	0.02	0.05	0.02
Sn-128	0.99	0.03	0.08	0.04
Sb-128	9.01	0.02	0.05	0.04
Te-129m	33.6d	0.13	0.25	0.15

Isotope	Halflife (hours)	Fission Fraction, %		
		U235	Pu239	Pu241
Te-129	1.16	0.64	1.29	0.79
Sb-130m	0.105	1.57	2.61	2.50
Te-131m	30.0	0.38	0.54	0.46
Te-131	0.417	2.50	3.24	2.74
Te-132	78.2	4.30	5.15	4.58
I-132m	1.38	0.05	0.20	0.03
I-132	2.30	0.05	0.20	0.03
Te-133m	0.92	3.84	4.01	3.77
I-133	208	2.84	2.96	2.78
Te-134	0.697	6.38	4.24	6.58
I-134	0.877	1.13	3.07	1.45
I-135	6.61	5.98	5.92	7.17
Xe-135	9.09	0.56	1.26	0.30
Cs-136	13d	0.05	0.09	0.02
Cs-137	30.2y	6.22	6.64	6.50
Cs-138	0.537	6.91	5.82	6.49
Ba-139	1.39	6.65	5.73	6.48
Ba-140	307	6.29	5.50	5.82
La-140	40.3	0.01	0.08	0.00
La-141	3.93	5.81	5.34	4.79
La-142	1.54	5.89	5.00	4.79
Pr-142	19.1	5.91	5.00	4.79
Ce-143	33.0	5.94	4.47	4.42
Ce-144	284d	5.38	3.82	4.09
Pr-145	5.98	3.92	3.12	3.14
Pr-146	0.40	2.96	2.53	2.66
Nd-147	265	2.31	2.11	4.09
Nd-149	1.73	1.08	1.28	1.46
Pm-150	2.68	0.65	0.99	1.16
Pm-151	28.4	0.42	0.80	0.90
Sm-153	46.7	0.16	0.40	0.52
Sm-155	0.37	0.00	0.22	0.23
Sm-156	9.40	0.01	0.11	0.16
Eu-157	15.1	0.01	0.08	0.13
Eu-158	0.765	0.00	0.04	0.08

consideration. Each of these chains has an isotope in an isomeric state that should be taken into account since its contribution to the activity produced in the reactor is significant. In these cases we will divide the chains into two separate ones – as shown on the right side of Fig. 5. The fraction of fission fractions for each of these two chains will be calculated according to the branching ratios. So, for example, the $A = 133$ chain that includes Xe^{133m} will use 1.5% of the fission fractions of Te^{133m} and I^{133} – the only isotopes with non-zero fission fractions in the chain; the remaining 98.5% of the fission fractions will be used in the second chain (Xe^{133m} not present). In case of the $A = 129$ chain, 35% of the fission fraction of Te^{129m} will be used in the $Te^{129m} \rightarrow I^{129}$ chain, and the other 65% – in the $Te^{129m} \rightarrow Te^{129} \rightarrow I^{129}$ chain. To get the radioactivity of an isotope present in both chains (Te^{129m} in the last example), we just add up results from each chain.

In our calculations we consider only 7 isotopes from the actinide group (starting with U^{238}) since they contribute the most to the actinide activity produced in the RBMK reactor[24]. This actinide chain is

$$U^{238} \rightarrow U^{239} \rightarrow Np^{239} \rightarrow Pu^{239} \rightarrow Pu^{240} \rightarrow Pu^{241} \rightarrow Pu^{242} \qquad (14)$$

In this chain only Pu^{239} and Pu^{241} are fissionable with thermal neutrons thus, they represent secondary fuel produced during the campaign. Since fission product production rate is different for different fuel, we will take into account this fact and use the fission fractions that correspond to each fuel isotope (see Table 4).

5.6 Neutron Cross Sections

In neutron cross section calculations we will follow the 2-group model described in [Spra 83]. This model uses the thermal cross section and the resonance integral for actinide activity calculations. The reaction rate for the x-type of reaction ((n,α), (n,γ), $(n,2n)$ or (n,f)) in this model is written in the following form:

$$R = \Phi(\sigma^x + \alpha I^x), \qquad (15)$$

[24] We will also calculate the amount of U^{235}, since it is the main energy source in the reactor. However, since the activity due to U^{235} is negligibly small it won't be presented in the activity tables.

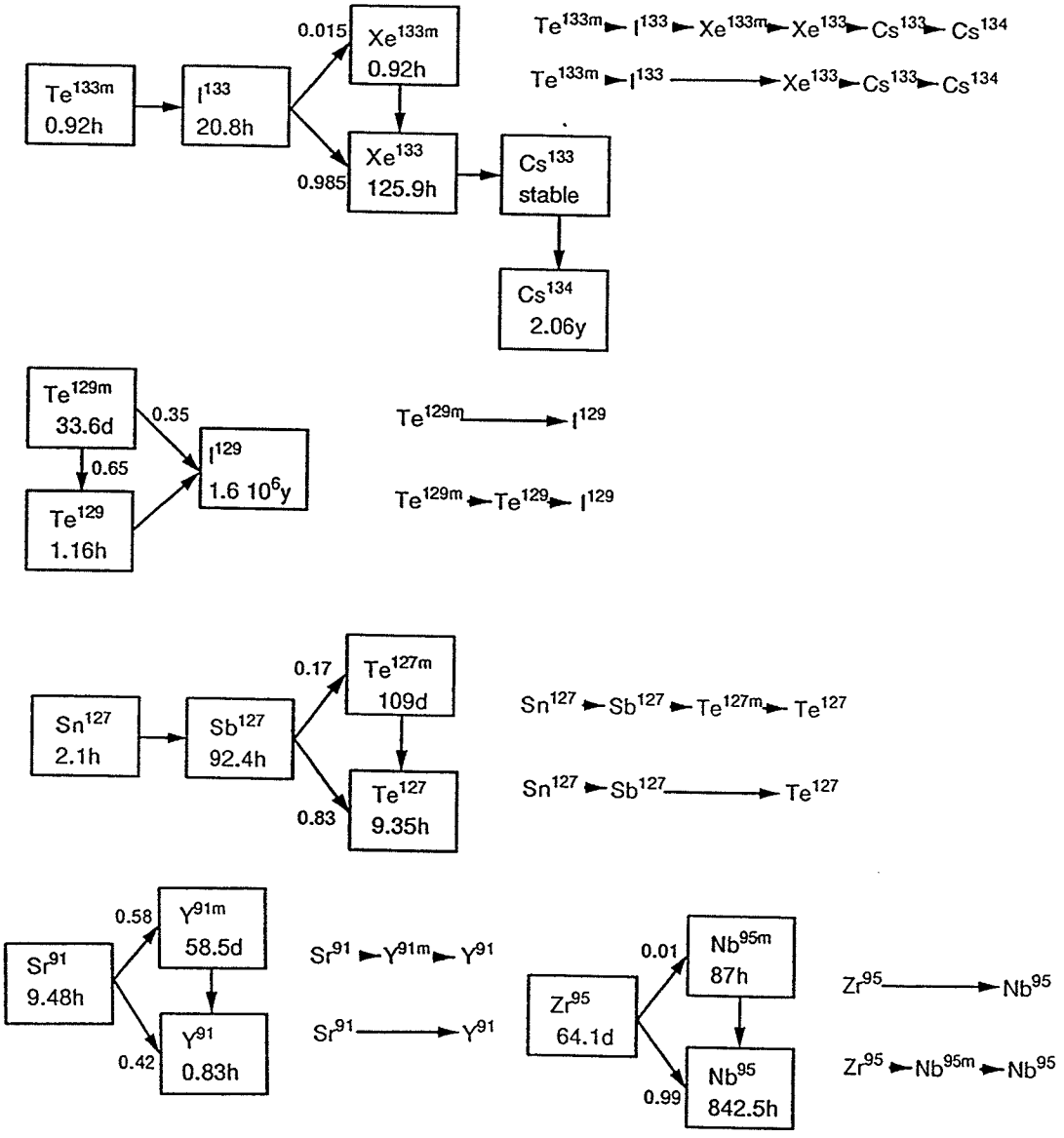

Figure 5: Chains with important isotopes in isomeric states. Two separate chains were used in calculations of the given isotopes - as shown on the right.

where R is the reaction rate, Φ the neutron flux, σ^x the reaction cross section, I^x reaction resonance integral, α neutron spectrum hardness[25]. The spectrum hardness as a function of burnup is shown in Fig. 6. The (n,γ), and (n,f) cross sections for actinides that are used in our calculations are given in Table 6 [Spra 83]. The resonance integral for U^{238} for the RBMK type of fuel assemblies was calculated to be 13.9 barns using formulas from [Hell 57]. In the case of Pu^{240}, the resonance integral was calculated using formula

$$I = fI^{\infty}, \tag{16}$$

where I^{∞} is the resonance integral for infinite dilution and f the self-screening coefficient that can be calculated as follows [Holm 60]:

$$f^{-1} = \sqrt{1 + \frac{\sigma_0 \Gamma_\gamma}{\sigma_m \Gamma}}; \qquad \sigma_m = \frac{1}{2rN_F}. \tag{17}$$

For the $E = 1.057$ eV resonance in Pu^{240} we have [Mugh 89] $\sigma_0 = 150000$ barns, $\Gamma_\gamma = 32.4$ meV, $\Gamma = 34.85$ meV. Thus, for the RBMK reactor which has fuel tablets with $r = 11.5$ mm we'll get

$$f^{-1} = \sqrt{1 + 3.2 \cdot 10^{-19} N_F}, \tag{18}$$

where N_F is the number of atoms of Pu^{240} per cm^3. This formula has been used in our calculations.

[25]This parameter shows how much the neutron spectrum deviates from the Maxwellian distribution. For an ideal Maxwellian spectrum α=0, and only the thermal cross section can be used.

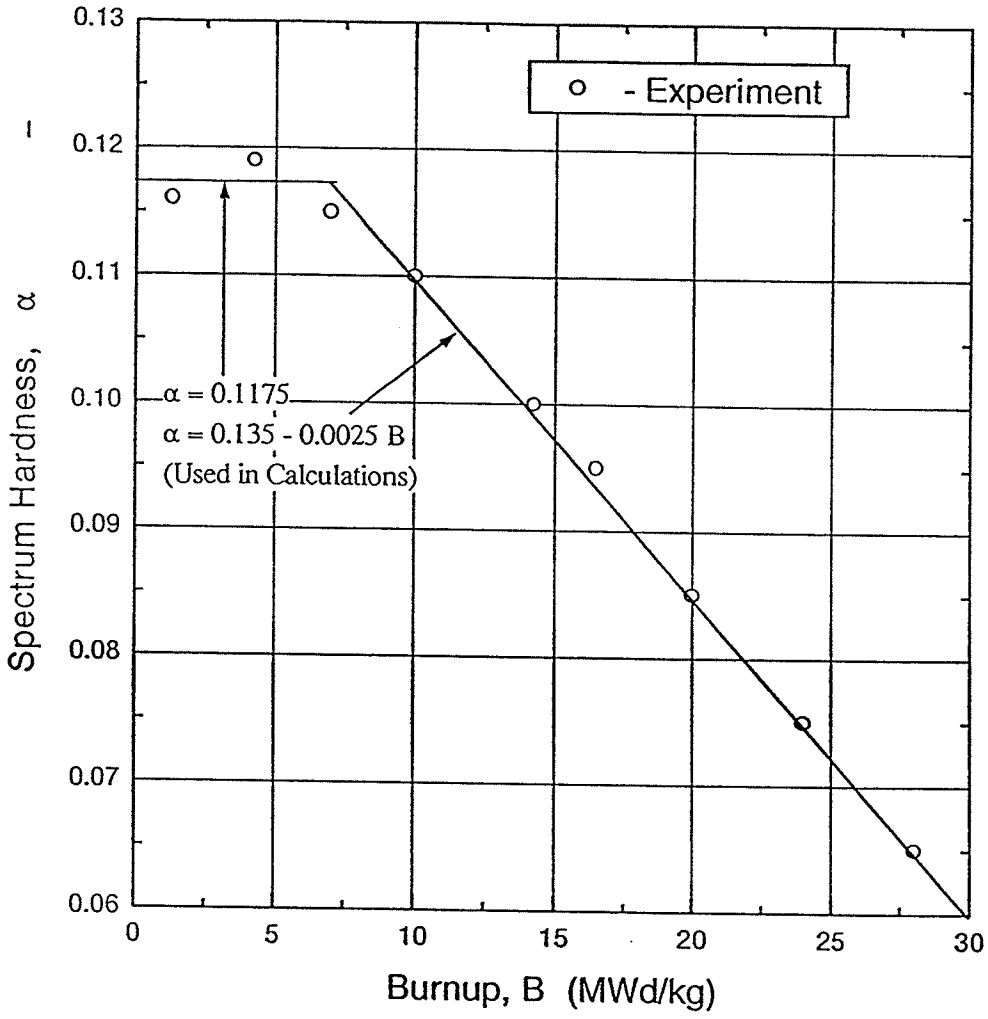

Figure 6: Neutron spectrum hardness vs. fuel burnup (taken from [Spra 83]).

Table 6: Neutron cross sections for the two group model used in calculations [Spra 83]. In a two group model, two parameters are used to describe the reaction rate: cross section σ and resonance integral RI.

Actinides *(n,f)* cross section, 2-group model		
	σ, barns	RI, barns
U-238	-	-
U-239	14	-
Np-239	1	-
Pu-239	742.5	301
Pu-240	0.03	-
Pu-241	1023	570
Pu-242	0.2	4.7
Actinides *(n,γ)* cross section, 2-group model		
U-238	2.71	278
U-239	22	-
Np-239	45	-
Pu-239	268.8	200
Pu-240	289.5	8620
Pu-241	355	162
Pu-242	18.5	1280

6 Calculational Model

In a "perfect" model we would calculate the activity of the actinides and fission products *simultaneously* using the equations we derived in the previous section. Only this approach is able to provide immediate (for each time interval we choose for solving these equations) feedback about the amount of fission products produced in the reactor, and therefore what the thermal neutron flux should be in the next time interval. However, such a "perfect" model would require detailed (in time) information about the position of the absorbers to account for their neutron absorption rate. This information is not available, and therefore we will use a simplified (and commonly used) approach that considers the actinides and fission products separately. Effectively this implies that the neutron absorption rate in fission products *and* absorbers is kept constant by adjusting the positioning of absorbers[26].

In this section we will describe the two-step procedure we will follow in our calculations and explain some averaging techniques widely used in activity calculations (that also will be used in this thesis).

6.1 Production of Actinides

Calculating the actinide radioactivity is the first step in our calculations, since it provides information about the production of secondary fuel. Unlike the fission products, actinides in the reactor are produced by decay and neutron capture in uranium initially in the reactor: in our case these are U^{235} and U^{238}. We will be only interested in the chain that starts from U^{238} because: first, most of the actinides with high α-activity have atomic numbers larger than 238; second, the two new fissionable isotopes we will take into account — Pu^{239} and Pu^{241} — are produced in this chain.

The algorithm we will use in our calculations is the following. Consider one of the 18 groups of fuel assemblies given in [Boro 89]. We divide the time this fuel has been in the core into small time intervals one day long[27] and solve equation (A.1) - using

[26]The neutron absorption due to actinides will be taken into account during calculations.

[27]We assume that during one day the parameters of the reactor can be considered as being constant and therefore, the solution from Appendix A can be used. In the calculations in [Spra 83] that time

the general solution given in Appendix A – for each of the time intervals as follows:

- We know the total energy produced E_{tot} by this group of assemblies and the number of days (T) this fuel had been in the core (see Table 1, column 1 and 4). Therefore, using our assumption of a constant energy production rate during the *A-Period*, we can calculate the amount of energy produced (by multiplying the number of fissions and the energy released per fission) during the given time interval of one day (let's denote it as E_1, $E_1 = E_{tot}/T$).

- By choosing an arbitrary neutron flux (that will be kept constant for this time interval), we can calculate the number of nuclei of each actinide in (A.1) as well as the number of fissions of each fissionable isotope (U^{235} and new produced Pu^{239} and Pu^{241}). This gives us the energy (E_2) produced in the reactor during this time interval (that corresponds to the chosen neutron flux).

- We adjust the neutron flux and repeat the last calculation until the calculated amount of energy matches the given amount ($E_1 = E_2$). At this point we calculate fractions ω_i ($\omega_1, \omega_2, \omega_3$ define the part of the total energy produced by fission of U^{235}, Pu^{239} and Pu^{241}, respectively) and the macroscopic fission cross section $\Sigma^f \equiv \sigma_1^f U_5 + \sigma_2^f U_9 + \sigma_3^f U_1$ that will be used in our further calculations.

As the campaign goes on, two factors will define the neutron flux change: first, U^{235} is burning out and the number of fissionable nuclei decreases; and second, actinides that absorb neutrons are produced in the reactor. This will require the neutron flux to increase to keep the energy production rate constant.

This procedure is performed for each of 18 groups of fuel assemblies and for each time interval. By adding up the results we will get the total number of nuclei of each isotope under consideration, and therefore the corresponding radioactivity. Data tables that include the neutron flux, macroscopic fission cross section, and ω_i for each fissionable isotope will be used in fission products activity calculations.

interval was chosen to be 100 days.

Unlike the *A-period*, the *B-period* will be treated without any flux adjustments. Both, *B-* and *C-periods* lasted together for about 24 hours, and therefore their contribution to the total energy produced in the ChNPP-4 reactor is not significant. Hence the burnup of the long-lived isotopes won't be affected during this period. However, the short-lived isotope activity is proportional to the neutron flux (see equation (7)) and therefore it is important to follow the real power curve during *B-period*. We assume that the neutron flux $\Phi(t)$ is proportional to the reactor power $P(t)$ that is given (see Fig. 3), and will calculate $\Phi(t)$ using the following formula:

$$\Phi(t) = \frac{P(t)}{P_0}\Phi_0, \tag{19}$$

where Φ_0 and P_0 denote the neutron flux (obtained using the procedure described above) and reactor power at the end of the *A-period*.

6.2 Production of Fission Products

For the fission products the source term F_k in equation (A.1) for the k-th isotope in the chain can be written in form:

$$F_k = (y_1\sigma_1^f U_1 + y_2\sigma_2^f U_2 + y_3\sigma_3^f U_3)_k\Phi, \tag{20}$$

where σ_i^f is the fission cross section, y_i the fission fraction, U_i the number of nuclei and the indices 1,2 and 3 correspond to U^{235}, Pu^{239} and Pu^{241}, respectively. In terms of the macroscopic fission cross section ($\Sigma^f \equiv \sigma_1^f U_1 + \sigma_2^f U_2 + \sigma_3^f U_3$), it can be rewritten (omitting the k index):

$$F = (y_1\omega_1 + y_2\omega_2 + y_3\omega_3)\Sigma^f\Phi, \tag{21}$$

where $\omega_i\Sigma^f = \sigma_i^f U_i$, $i = 1, 2, 3$, and obviously, $\omega_1 + \omega_2 + \omega_3 = 1$.

All data necessary to calculate F were obtained during the first stage of our calculational procedure – actinide activity calculations. Along with the cross sections for each isotope, we are able to perform all calculations using the equation (A.1).

The procedure described will be used in activity calculations of isotopes with $T_{1/2} > 0.5$ hours produced in the reactor during *A-*, and *B-period*. To get the short-

term activity we will use another method (see Section 8, Results of Calculations: Short-term Activity in the ChNPP-4 Reactor).

6.3 Averaging Procedures

At this point we can also calculate (and compare results with the 18 groups calculations) the activity in the ChNPP-4 reactor by using averaging techniques – considering the whole fuel in the reactor as one group of fuel (instead of 18 groups in the sections above). In this case we need only the total burnup (i.e. total energy produced in the reactor during the campaign) and the total amount of fuel. These average procedures are exactly the same as [Deve 86], [Kirc 88] and others have used in their calculations.

As we discussed above, the total energy produced in the reactor during a given time interval can easily be adjusted by varying the neutron flux. Therefore, it won't be a problem to implement this energy "conservation law" in the averaging procedure.

According to [Boro 89], different groups of fuel assemblies had been present in the core for different times. Therefore, the term "total amount of fuel in the reactor" can be interpreted in a few different ways.

To overcome this problem, we will make use of the second integral parameter from [Boro 89] – total number of assemblies integrated over time it was in the core: $1.06 * 10^6$ Assd (see Section 3.1, *A-Period*).

By adjusting the number of fuel assemblies in the reactor or the time ChNPP-4 was under operation we can build three different average models (in all of them it is assumed that no fuel had been added or removed from the reactor during the campaign time):

- *Av1.* No adjustments.

  1659 assemblies stayed in the core for 735 days.

- *Av2.* Time adjustment.

  1659 assemblies stayed in the core for 638 days

  (total $1.06 \cdot 10^6$ Assembly*days).

- *Av3.* Fuel adjustment.

  1440 assemblies stayed in the core for 735 days

  (total $1.06 \cdot 10^6$ Assembly*days)

Both, *Av*2 and *Av*3 have the same amount of fuel integrated over the time it was in the core – $1.06 \cdot 10^6$ Assd [Boro 89]. In *Av*2 the length of the campaign and in *Av*3 the number of fuel assemblies were changed in order to meet this figure. The *Av*1 model does not obey this rule, and uses the total number of fuel assemblies in the ChNPP-4 at the time of the accident and the total length of the campaign time.

Now we can apply the same calculational model we used for each of the 18 groups of fuel to each of the average models. The averaging procedure that will give the closest results to the 18 group calculations can be considered as the most accurate one[28].

[28]We should point out that all models used the "energy conservation law" – the total energy produced in the ChNPP-4 reactor was the same for all models.

7 Results of Calculations: Long Term Activity in the ChNPP-4 Reactor

As we discussed before, isotopes we have chosen for our calculations (Table 6) will give correct values for the long term radioactive inventory in the ChNPP-4 reactor – some 3 hours after the accident and later. For the short term activity we must use either other methods or an extended list of isotopes.

Using the computational model and procedure described in the previous section, we calculated the total amount of radioactivity produced in the ChNPP-4 reactor during the *A*- and *B-period* for the "18 groups" data [Boro 89] as well as for each of the average models. A computer program that performs this calculational procedure was written in FORTRAN and run on an IBM PC computer. The results are shown in Table 7 and Table 8.

For each of these models two different power scenarios during the *B-period* were used: constant power (ConstP column in Table 7 and 8) and real power (RealP column). The "RealP" scenario uses the power history as given in Fig. 3. In the "ConstP" scenario the power during *B-period* remains the same as at the end of the *A-period*. This would be the activity in the ChNPP-4 reactor if the explosion occurred while the reactor was at full power.

In comparing the reference "18-groups" and average models results, we note that the *Av2* model gives the most accurate results (comparing to the "18-groups" calculations) for both fission products and actinide activity: the difference in calculated activity of fission products and actinides is about 1% for both "RealP" and "ConstP" scenarios[29]. Therefore we can conclude that keeping the number of fuel assemblies present in the reactor (rather than the campaign time) gives the most accurate "average" model.

The time dependence of the activity produced in the ChNPP-4 reactor is impor-

[29] At this point we do not pretend to calculate the activity in the ChNPP-4 with an accuracy of about 1% since the uncertainties in the initial data, models used, and assumptions will be much higher. The stated result only tells us which averaging procedure gives the closest to the reference 18-group model results.

Table 7: Calculated inventory activity in the ChNPP-4 reactor (A+B Periods) using the data for 18 groups of fuel assemblies (see Table 1). The calculations were performed using two models: real power (RealP column) – takes into account the power history from Fig.3; and constant power (ConstP column) – the power was kept constant during the B-Period. This gives the activity in the reactor if the explosion would occur at full power. The C-Period is not taken into account.

Isotopes	Halflife, hours	RealP (MCi)	ConstP (MCi)	ConstP / RealP
Fission Products				
Ge-78	1.45	0.07	0.63	8.70
As-78	1.51	0.13	0.63	4.94
Br-83	2.39	1.91	12.2	6.39
Kr-83m	1.86	2.76	12.2	4.42
Br-84	0.53	0.77	22.2	28.76
Kr-85m	4.48	5.46	25.5	4.66
Kr-85	10.7 y	0.75	0.75	1.00
Kr-87	1.27	5.47	52.7	9.63
Kr-88	2.84	12.9	75.9	5.88
Sr-89	50.4 d	96.5	97.5	1.01
Sr-90	29.1 y	5.64	5.64	1.00
Y-90	64.00	5.61	5.61	1.00
Sr-91	9.48	46.4	129.	2.78
Y-91m	58.5 d	124	125.	1.01
Y-91	0.83	29	74.8	2.58
Sr-92	2.71	22.1	133.	6.02
Y-92	3.54	35.4	134.	3.79
Y-93	10.20	56.8	150.	2.64
Zr-95	64.1 d	154.	155.	1.01
Nb-95	35.1 d	151.	151.	1.00
Nb-95m	87.00	1.63	1.63	1.00
Zr-97	16.90	83.3	159.	1.91
Nb-97	1.20	88.4	159.	1.80
Nb-98	0.85	11.1	159.	14.32
Mo-99	66.00	140.	168.	1.20
Tc-99m	6.02	150.	168.	1.12
Ru-103	39.3 d	113.	115.	1.02
Rh-103m	0.94	113.	115.	1.02
Ru-105	4.44	15.1	70.9	4.70
Rh-105	35.40	56.	70.8	1.26
Ru-106	1.01 y	24.9	24.9	1.00
Ag-110m	250 d	3.54.	3.55	1.00
Ag-111	7.5 d	2.89	3.09	1.07
Pd-112	21.10	0.71	1.21	1.70
Ag-112	3.12	0.80	1.21	1.52
Ag-113	5.37	0.23	0.97	4.17
Cd-115	53.5	2.79	3.56	1.28

Table 7: continued

Isotopes	Halflife, hours	RealP (MCi)	ConstP (MCi)	ConstP / RealP
In-115m	4.30	1.29	3.61	2.80
Cd-118	0.84	0.04	0.55	14.75
Sn-121	27.10	0.40	0.61	1.53
Sn-123m	0.67	0.05	0.92	19.94
Sn-125	9.6 d	0.76	0.80	1.06
Sb-125	2.8 y	0.46	0.46	1.00
Sn-127	2.10	0.72	5.64	7.80
Sb-127	92.40	5.64	6.32	1.12
Te-127	9.35	5.88	6.16	1.05
Te-127m	109 d	0.94	0.94	1.00
Sn-128	0.98	0.11	1.37	12.23
Sb-128	9.01	0.30	0.86	2.88
Te-129m	33.6 d	4.39	4.46	1.02
Te-129	1.16	5.1	26.30	5.16
Te-131m	30.00	8.0	11.80	1.48
I-131	193.00	81.7	86.40	1.06
Te-131	0.42	1.68	75.	44.6
Te-132	78.20	107.	125.	1.17
I-132	2.30	111.	133.4	1.16
I-132m	1.38	0.3	2.68	8.99
Te-133m	0.92	8.08	107.	13.24
I-133	20.80	111.	185.	1.67
Xe-133	5.3 d	180.	185.	1.03
Cs-134	2.06 y	4.05	4.06	1.00
Xe-133m	52.50	2.57	2.73	1.06
Te-134	0.70	8.29	155.	18.70
I-134	0.88	25.6	204.	7.97
I-135	6.61	45.2	164.	3.63
Xe-135	9.09	96.0	185.	1.93
Cs-136	13 d	1.61	1.68	1.04
Cs-137	30.2 y	6.78	6.79	1.00
Cs-138	0.54	6.36	179.	28.14
Ba-139	1.39	19.3	173.	8.96
Ba-140	307.00	157.	163.	1.04
La-140	40.30	162.	164.	1.01
La-141	3.93	30.7	154.	5.02
Ce-141	780.00	148.	149.	1.01
La-142	1.54	18.3	152.	8.31
Pr-142	19.10	85.4	153.	1.79
Ce-143	33.00	104.	148.	1.42
Pr-143	326.00	146.	147.	1.01

Table 7: continued

Isotopes	Halflife, hours	RealP (MCi)	ConstP (MCi)	ConstP / RealP
Ce-144	284.1 d	101.	101.	1.00
Pr-145	5.98	25.6	99.5	3.89
Pr-146	0.40	1.59	76.9	48.36
Nd-147	265.00	59.3	62.1	1.05
Pm-147	2.6 y	22.1	22.1	1.00
Nd-149	1.73	4.1	31.6	7.71
Pm-149	53.10	25.9	31.5	1.22
Pm-150	2.68	3.47	21.1	6.08
Pm-151	28.40	10.1	15.2	1.50
Sm-151	87 y	0.18	0.18	1.00
Sm-153	46.70	5.20	6.71	1.29
Sm-155	0.37	0.04	2.17	56.96
Eu-155	4.9 y	0.29	0.29	1.00
Sm-156	9.40	0.48	1.33	2.79
Eu-156	364.00	1.27	1.30	1.02
Eu-157	15.20	0.43	0.87	2.04
Eu-158	0.77	0.03	0.46	16.51
Total fission products		**3520.1**	**6366.1**	**1.81**
Actinides				
U-238	0.39+14	6.23-5	6.23-5	1.00
U-239	0.39	161.	1510.	14.5
Np-239	56.50	1410.	1510.	1.07
Pu-239	0.21+11	0.0215	0.0215	1.00
Pu-240	0.574+8	0.0376	0.0376	1.00
Pu-241	128900.	4.38	4.38	1.00
Pu-242	0.33+10	7.2-5	7.2-5	1.00
Total actinides		**1575.4**	**3024.4**	**1.91**
Total Activity		**5095.5**	**9390.5**	**1.85**

Table 8: Calculated inventory activity in the ChNPP-4 reactor (A+B Periods) using the different average models. The calculations for each model were performed using two models: real power (RealP column) – takes into account the power history from Fig.3; and constant power (ConstP column) – the power was kept constant during the B-Period. This gives the activity in the reactor if the explosion would occur at full power. The C-Period is not taken into account.

Isotope	Halflife, hours	Average 1 (MCi)		Average 2 (MCi)		Average 3 (MCi)		Average/18 groups		
		RealP	ConstP	RealP	ConstP	RealP	ConstP	Av1	Av2	Av3
Fission Products										
Ge-78	1.45	0.06	0.55	0.07	0.63	0.06	0.55	0.88	1.01	0.87
As-78	1.51	0.11	0.55	0.13	0.63	0.11	0.55	0.88	1.01	0.87
Br-83	2.39	1.62	10.40	1.92	12.30	1.66	10.70	0.85	1.01	0.87
Kr-83m	1.86	2.35	10.40	2.78	12.30	2.41	10.70	0.85	1.01	0.87
Br-84	0.53	0.65	18.70	0.78	22.30	0.67	19.40	0.85	1.01	0.87
Kr-85m	4.48	4.76	22.2	5.50	25.7	4.59	21.4	0.84	1.01	0.88
Kr-85	10.7 y	0.75	0.75	0.78	0.78	0.77	0.77	1.00	1.03	1.02
Kr-87	1.27	4.57	44.10	5.51	53.20	4.77	46.00	0.84	1.01	0.87
Kr-88	2.84	10.8	63.70	13.	76.6	11.3	66.3	0.84	1.01	0.88
Sr-89	50.4 d	86.1	86.90	103.	104.	89.2	90.1	0.89	1.07	0.92
Sr-90	29.1 y	5.62	5.63	5.74	5.75	5.72	5.73	1.00	1.02	1.01
Y-90	64.00	5.6	5.60	5.72	5.72	5.7	5.7	1.00	1.02	1.02
Sr-91	9.48	39.0	108.	46.7	130.	40.5	113.	0.84	1.01	0.87
Y-91m	58.5 d	112.	112.	133.	134.	115.	116.	0.90	1.07	0.93
Y-91	0.83	24.4	62.9	29.2	75.4	25.3	65.3	0.84	1.01	0.87
Sr-92	2.71	18.7	113.	22.3	134.	19.3	116.	0.85	1.01	0.87
Y-92	3.54	29.9	114.	35.6	135.	30.9	117.	0.84	1.01	0.87
Y-93	10.20	48.4	128.	57.2	151.	49.5	131.	0.85	1.01	0.87
Zr-95	64.1 d	141.	142.	164.	165.	143.	144.	0.92	1.06	0.93
Nb-95	35.1 d	143.	143.	166.	166.	144.	144.	0.95	1.10	0.95
Nb-95m	87.00	1.49	1.5	1.75	1.75	1.51	1.52	0.91	1.07	0.93
Zr-97	16.90	72.10	138.	83.6	160.	72.4	138.	0.87	1.00	0.87
Nb-97	1.20	76.60	138.	88.8	160.	76.9	138.	0.87	1.00	0.87
Nb-98	0.85	9.62	138.	11.1	160.	9.61	138.	0.87	1.00	0.87
Mo-99	66.00	122.	146.	141.	169.	122.	146.	0.87	1.01	0.87
Tc-99m	6.02	130.	146.	150.	169.	130.	146.	0.87	1.00	0.87
Ru-103	39.3 d	105.	107.	116.	118.	101.	103.	0.93	1.03	0.89
Rh103m	0.94	105.	107.	116.	118.	101.	103.	0.93	1.03	0.89
Ru-105	4.44	14.1	66.3	15.0	70.1	12.9	60.7	0.93	0.99	0.85
Rh-105	35.40	52.3	66.2	55.4	70.1	48.0	60.7	0.93	0.99	0.86
Ru-106	1.01 y	24.8	24.9	24.00	24.1	22.7	22.7	1.00	0.96	0.91
Ag110m	250 d	3.53	3.54	3.37	3.39	3.14	3.15	1.00	0.95	0.89
Ag-111	7.5 d	2.76	2.96	2.81	3.01	2.44	2.62	0.96	0.97	0.84
Pd-112	21.10	0.68	1.15	0.69	1.18	0.60	1.02	0.95	0.97	0.84
Ag-112	3.12	0.76	1.15	0.77	1.18	0.67	1.02	0.95	0.97	0.84
Ag-113	5.37	0.22	0.91	0.23	0.94	0.20	0.82	0.94	0.97	0.84
Cd-115	53.5	2.72	3.44	2.79	3.50	2.34	3.03	0.97	0.98	0.85
In-115m	4.30	1.25	3.49	1.27	3.55	1.10	3.07	0.97	0.98	0.85
Cd-118	0.84	0.03	0.51	0.04	0.55	0.03	0.47	0.92	1.00	0.87
Sn-121	27.10	0.37	0.56	0.40	0.61	0.35	0.53	0.92	1.00	0.87

Table 8: continued

Isotope	Halflife, hours	Average 1 (MCi)		Average 2 (MCi)		Average 3 (MCi)		Average/18 groups		
		RealP	ConstP	RealP	ConstP	RealP	ConstP	Av1	Av2	Av3
Sn-123m	0.67	0.04	0.85	0.05	0.92	0.04	0.8	0.93	1.00	0.87
Sn-125	9.6 d	0.71	0.74	0.77	0.81	0.67	0.7	0.93	1.01	0.87
Sb-125	2.8 y	0.46	0.46	0.45	0.45	0.44	0.44	1.00	0.98	0.96
Sn-127	2.10	0.76	5.18	0.82	5.64	0.71	4.88	1.05	1.14	0.98
Sb-127	92.40	5.18	5.81	5.64	6.33	4.89	5.49	0.92	1.00	0.87
Te-127	9.35	5.45	5.71	5.93	6.21	5.16	5.4	0.93	1.01	0.88
Te-127m	109 d	0.91	0.91	0.98	0.98	0.86	0.86	0.97	1.04	0.92
Sn-128	0.98	0.10	1.24	0.11	1.37	0.10	1.19	0.90	1.00	0.87
Sb-128	9.01	0.27	0.78	0.30	0.86	0.26	0.75	0.90	1.00	0.86
Te-129m	33.6 d	4.04	4.10	4.51	4.59	3.93	3.99	0.92	1.03	0.90
Te-129	1.16	4.63	23.6	5.18	26.4	4.50	22.9	0.91	1.02	0.88
Te-131m	30.00	7.06	10.4	8.01	11.8	6.94	10.2	0.88	1.00	0.87
I-131	193.00	72.20	76.3	82.20	86.9	71.1	75.2	0.89	1.01	0.88
Te-131	0.42	1.47	65.9	1.68	75.2	1.45	65.1	0.88	1.00	0.86
Te-132	78.20	93.80	109.	107.	125.	93.0	109.	0.88	1.00	0.87
I-132	2.30	97.41	116.95	111.5	132.	96.56	115.67	0.88	1.00	0.87
I-132m	1.38	0.28	2.47	0.30	2.7	0.26	2.33	0.92	1.00	0.87
Te-133m	0.92	7.04	92.6	8.25	107.	7.02	92.4	0.87	1.02	0.87
I-133	20.80	96.1	161.	111.	186.	95.90	160.	0.87	1.00	0.86
Xe133m	52.50	2.24	2.38	2.58	2.74	2.24	2.38	0.87	1.00	0.87
Xe-133	5.3 d	157.	161.	181.	189.	156.	163.	0.87	1.01	0.87
Cs-134	2.06 y	4.08	4.09	3.57	3.58	3.51	3.52	1.01	0.88	0.87
Te-134	0.70	7.1	133.	8.31	156.	7.19	135.	0.86	1.00	0.87
I-134	0.88	22.1	177.	25.6	204.	22.2	177.	0.86	1.00	0.87
I-135	6.61	39.3	142.	45.3	164.	39.2	142.	0.87	1.00	0.87
Xe-135	9.09	83.7	161.	96.2	185.	83.3	161.	0.87	1.00	0.87
Cs-136	13 d	1.45	1.51	1.64	1.70	1.42	1.48	0.90	1.02	0.88
Cs-137	30.2 y	6.78	6.79	6.79	6.79	6.77	6.77	1.00	1.00	1.00
Cs-138	0.54	5.49	154.	6.38	179.	5.53	155.	0.86	1.00	0.87
Ba-139	1.39	16.7	150.	19.3	174.	16.7	151.	0.87	1.00	0.87
Ba-140	307.	137.	143.	159.	165.	138.	143.	0.87	1.01	0.88
La-140	40.3	142.	143.	165.	166.	143.	144.	0.88	1.02	0.88
La-141	3.93	26.6	133.	30.8	154.	26.7	134.	0.87	1.00	0.87
Ce-141	780.	132.	134.	153.	155.	133.	134.	0.89	1.03	0.90
La-142	1.54	15.8	131.	18.4	153.	15.9	132.	0.86	1.01	0.87
Pr-142	19.1	73.7	132.	85.7	153.	74.3	133.	0.86	1.00	0.87
Ce-143	33.	89.5	127.	105.	149.	90.7	129.	0.86	1.01	0.87
Pr-143	326	127.	128.	148.	149.	129.	129.	0.87	1.01	0.88
Ce-144	284.1 d	98.3	98.5	108.	109.	99.2	99.4	0.97	1.07	0.98
Pr-145	5.98	22.1	85.6	25.7	99.9	22.3	86.5	0.86	1.00	0.87
Pr-146	0.4	1.37	66.4	1.6	77.1	1.38	66.8	0.86	1.01	0.87
Nd-147	265.	51.9	54.3	59.5	62.4	51.7	54.1	0.88	1.00	0.87
Pm-147	2.6 y	22.	22.0	22.7	22.7	22.0	22.0	1.00	1.03	1.00
Nd-149	1.73	3.59	27.6	4.1	31.6	3.55	27.3	0.88	1.00	0.87
Pm-149	53.1	22.7	27.6	25.9	31.6	22.5	27.4	0.88	1.00	0.87

Table 8: continued

Isotope	Halflife, hours	Average 1 (MCi)		Average 2 (MCi)		Average 3 (MCi)		Average/18 groups		
		RealP	ConstP	RealP	ConstP	RealP	ConstP	Av1	Av2	Av3
Pm-150	2.68	3.08	18.70	3.47	21.00	3.00	18.20	0.89	1.00	0.86
Pm-151	28.40	9.05	13.60	10.10	15.10	8.73	13.10	0.90	1.00	0.86
Sm-151	87 y	0.18	0.18	0.18	0.18	0.18	0.18	1.00	0.98	0.98
Sm-153	46.70	4.72	6.09	5.17	6.67	4.49	5.79	0.91	0.99	0.86
Sm-155	0.37	0.04	2.10	0.04	2.13	0.03	1.85	0.97	0.98	0.85
Eu-155	4.9 y	0.30	0.30	0.27	0.27	0.26	0.27	1.01	0.91	0.90
Sm-156	9.40	0.45	1.26	0.47	1.31	0.41	1.13	0.95	0.98	0.85
Eu-156	364.00	1.21	1.23	1.26	1.28	1.10	1.12	0.95	0.99	0.87
Eu-157	15.20	0.41	0.83	0.42	0.85	0.36	0.74	0.96	0.98	0.85
Eu-158	0.77	0.03	0.44	0.03	0.45	0.02	0.39	0.95	0.97	0.84
Total Fission Products		**3127.3**	**5589.0**	**3591.5**	**6452.1**	**3127.4**	**5602.5**	**0.89**	**0.99**	**0.89**
Actinides										
U-238	0.39+14	6.23-5	6.23-5	6.23-5	6.23-5	5.14-5	5.14-5	1.0	1.0	0.91
U-239	0.39	138.2	1300.	159.	1500.	110.	1130.	0.83	1.0	0.77
Np-239	56.50	1210.	1300.	1400.	1499.	1050.	1130.	0.83	1.0	0.77
Pu-239	0.21+11	0.0215	0.0215	0.0215	0.0215	0.0187	0.0187	1.0	1.0	0.91
Pu-240	0.574+8	0.0432	0.0432	0.0423	0.0423	0.0376	0.0376	1.1	1.1	0.91
Pu-241	128900.	3.72	3.72	3.72	3.72	3.22	3.22	0.83	0.83	0.71
Pu-242	0.33+10	1.48-4	1.48-4	1.48-4	1.48-4	1.28-4	1.28-4	0.91	0.91	0.77
Total actinides		**1351.9**	**2603.7**	**1562.7**	**3002.7**	**1163.3**	**2263.2**	**0.83**	**1.0**	**0.77**
Total Activity		**4478.7**	**8192.7**	**5153.2**	**9454.8**	**4290.7**	**7865.7**	**0.88**	**0.99**	**0.84**

tant while analyzing the impact of those activity in the future. The same computer program was used for those calculations with one modification: the neutron flux in equation (A.1) was set to zero. Figure 7 shows the dynamics of the sum of all fission products activity calculated by using different models. Again, the given results are valid only for some times $t > 3$ hours due to half-lives of isotopes used in our calculations. We notice that there is not much difference between the results for different models – less than 15% at 1 day after explosion to almost none at 1 month after explosion. Therefore, either of these models can be used for activity calculations for times more than few months after the explosion.

Now we can compare our results with the ones from other calculations, since the last correspond only to the long-lived isotopes. Those data are shown in Table 4, column 9.

As we discussed above, Borovoy et al. [Boro 89] used the correct value of the total energy produced in the ChNPP-4 during the campaign and therefore obtained the most accurate results. We notice that our results are in good agreement with the ones reported in [Boro 89].

As we discussed earlier in this thesis (see Section 4.2, Other Inventory Calculations), the ChNPP-4 reactor was operating 646 "equivalent-full-power-days". As expected, the results of [Deve 86] underestimated and [Kirc 88] overestimated the radioactive inventory (due to the isotopes with half-lives longer than the campaign time which activity is proportional to the campaign time, see formula (9)) in the ChNPP-4 reactor since they used 400 and 722 "equivalent-full-power-days", respectively. The data in Table 4 support this statement.

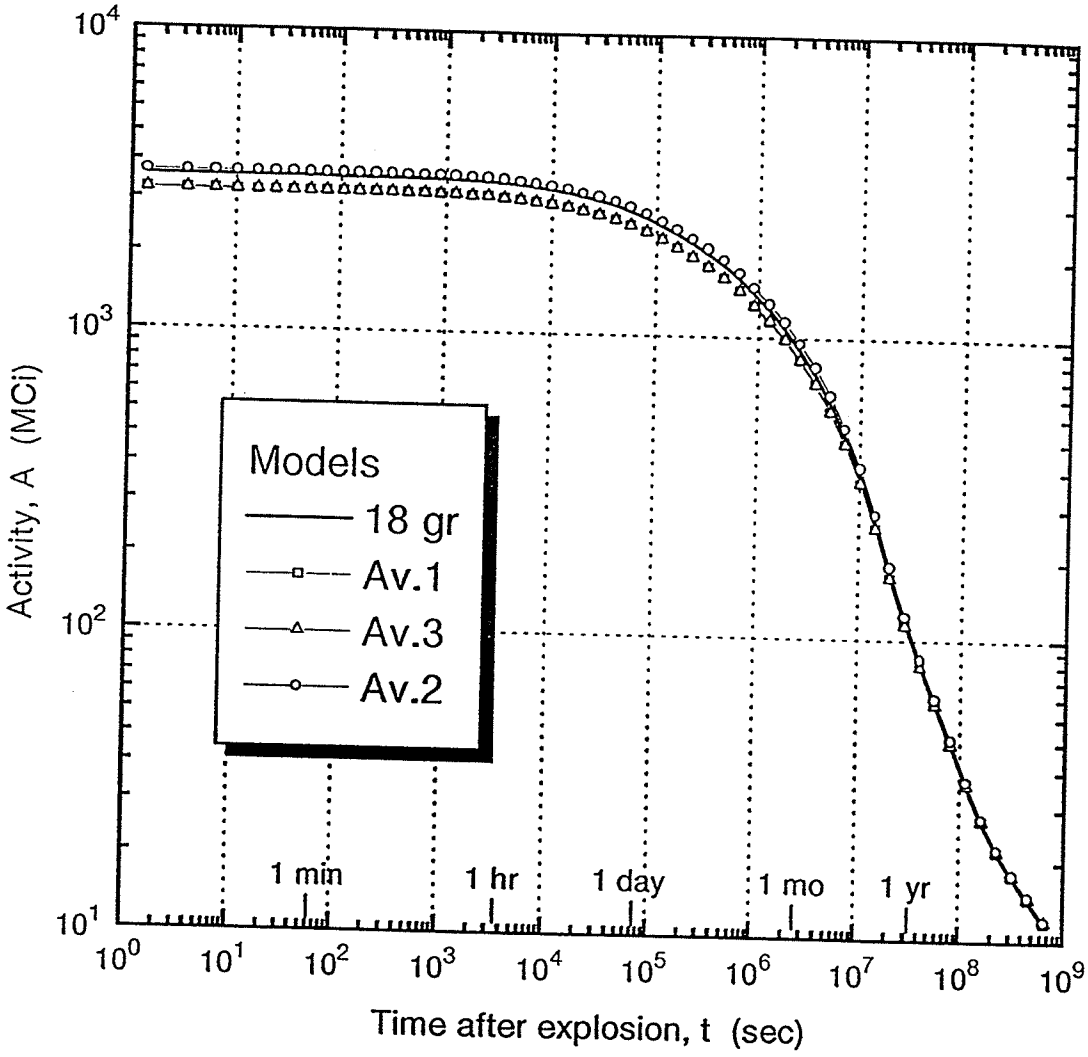

Figure 7: Calculated Fission Product Activity: A+B Periods. Long-term fission products activity calculated using the 3 average and 18-group models. The Av.2 model gives the closest to 18-group results. For decay times of order a few months and longer there is no difference between the results obtained using either of the models.

8 Results of Calculations: Short Term Activity in the ChNPP-4 Reactor

The method we used in the previous section for calculation of long-term radioactivity produced in the ChNPP-4 reactor cannot be applied to short-term activity calculations due to the absence of short-lived isotopes in our list (Table 4.). To obtain results for short term activity we will make use of some additional information provided in [Spra 83].

8.1 Constant Power Regime

Data given in [Spra 83] give the total radioactivity due to fission products as well as actinides produced in an RBMK reactor during three different campaign lengths – 800, 1100 and 1400 days. All these calculations were performed using an assumption that the average neutron flux over this time is constant and equal to $0.5*10^{14} cm^{-2} s^{-1}$. The authors mention that the results are applicable only to the *online refueling* regime of the RBMK operation when the average fuel burnup remains constant, and therefore constant neutron flux leads to constant power. Since ChNPP-4 was still not operating under this regime, we cannot apply directly the results from [Spra 83] to the ChNPP-4 reactor.

In general, using a constant in time neutron flux while calculating the radioactive inventory of RBMK reactor with fresh (zero burnup) fuel, creates a disproportion in energy production – the amount of energy produced at given period of time at the beginning and at the end of this period will differ significantly. This can easily be explained. The fraction of neutrons that initiate fission is much higher at the beginning of the campaign, when there are very few neutron absorbing isotopes, than at the end when a certain amount of such absorbers is produced. This, in turn, will lower the production rate of radioactive isotopes at the end of the campaign and consequently, the total activity of the ChNPP-4 inventory.

Therefore, we cannot use the data from [Spra 83] in our calculations. However, we consider the decay of the radioactive isotopes as a function of time after the reactor is

shut down. Three curves for different campaign lengths are shown on Fig. 8. Being present in a normalized form (equal to each other at a given point, in this case at $t = 1$ sec), the *shape* of these three curves is exactly the same up to some 50 days after the chain reaction stops, although the campaign time has almost doubled from 800 to 1400 days. We will use this *shape* in our calculations in the following way:

- Since our data for the total activity (fission products and actinides) are correct for $t > 3$ hours, we will normalize both curves – ours and that from [Spra 83] (Fig. 8) – at $t = 3$ hours: see Fig.9.

- We take the shape from the [Spra 83] curve (for times $t \leq 3h$) and transfer it to our data. By returning to the non-normalized form we get the total activity in the ChNPP-4 reactor valid for times starting as low as $0.1sec$.

Since the data we used from [Spra 83] imply constant nominal power during the whole campaign, the final result (Fig. 9) gives us the total activity accumulated in the ChNPP-4 reactor after the reactor is shut down at full power ("ConstP" scenario in Table 7, no C-Period).

8.2 Real Power Regime - Integral Approach

The method used in the previous section still does not allow us to calculate the short-term activity due to *B-* and *C-period* of the ChNPP-4 reactor campaign. To solve this problem we will use an *integral* approach.

The *integral* approach is based on the data given in [Spra 83] that gives the fission products activity of fresh RBMK fuel irradiated for a short period of time: from 1 sec to 100,000 sec [30] - some 28 hours. The activity of this fuel is given in Fig. 10. These data can't be used in calculating the activity of each particular isotope of interest, but will provide a method for total short-term fission products activity calculations. In addition, since the *B-period* lasted for about 24 hours, we can also calculate the

[30] In these calculations the length of irradiation time is limited by the assumption that the burnup of RBMK fuel remains negligible and no additional fissionable products are produced. Therefore, radioactivity of the actinides is not included and the data present only fission product activity.

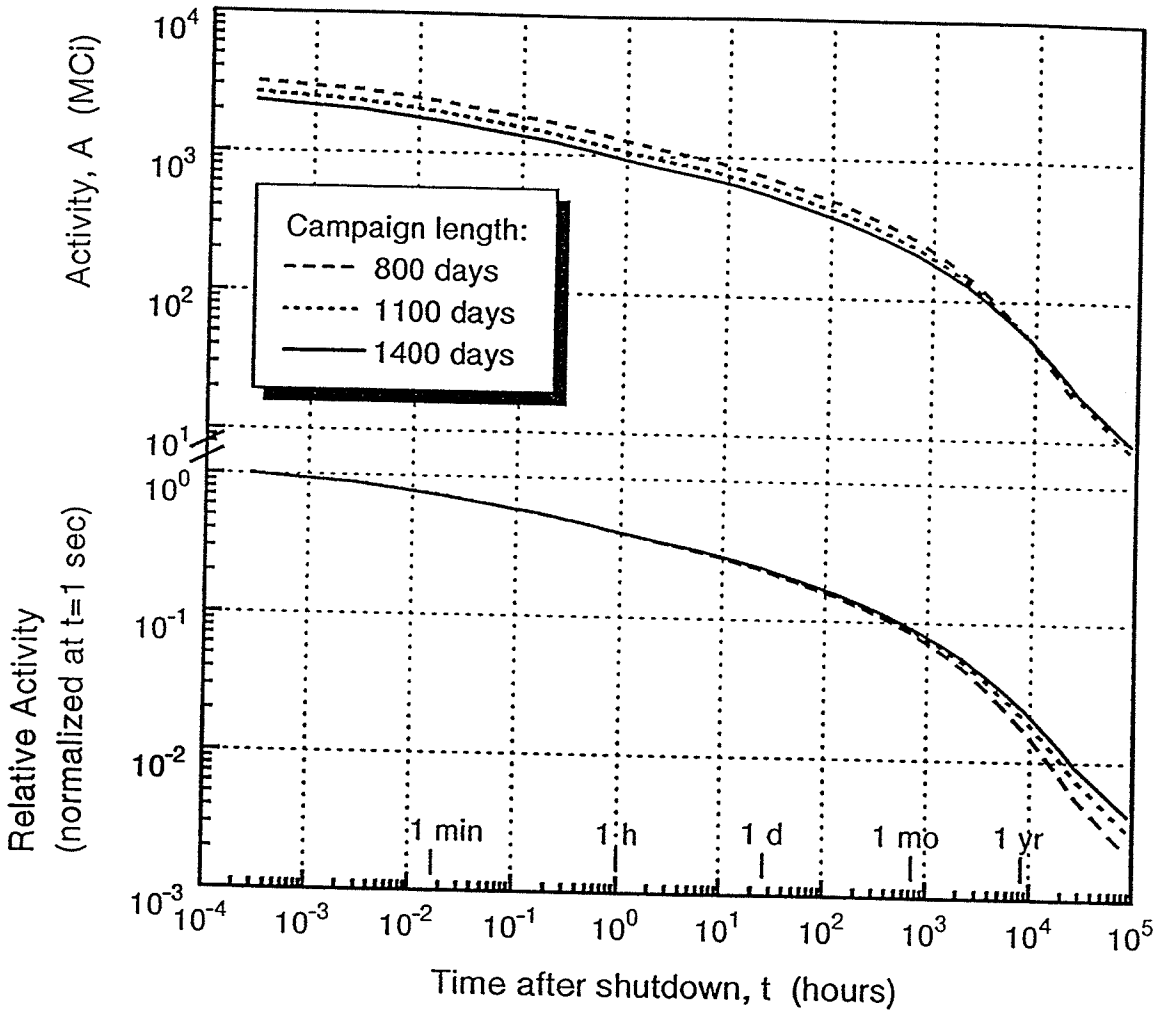

Figure 8: Activity produced in a RBMK reactor during different campaign times (taken from [Spra 83]). Although the amount of activity produced in the reactor depends on the campaign length (see top curves), being presented in a normalized form (see bottom graph, normalization point = 1 sec) the shape of the decay curve is the same for times up to one month. The long-term activity differs due to the different amount of long-lived isotopes produced in the reactor (the last is proportional to the campaign time).

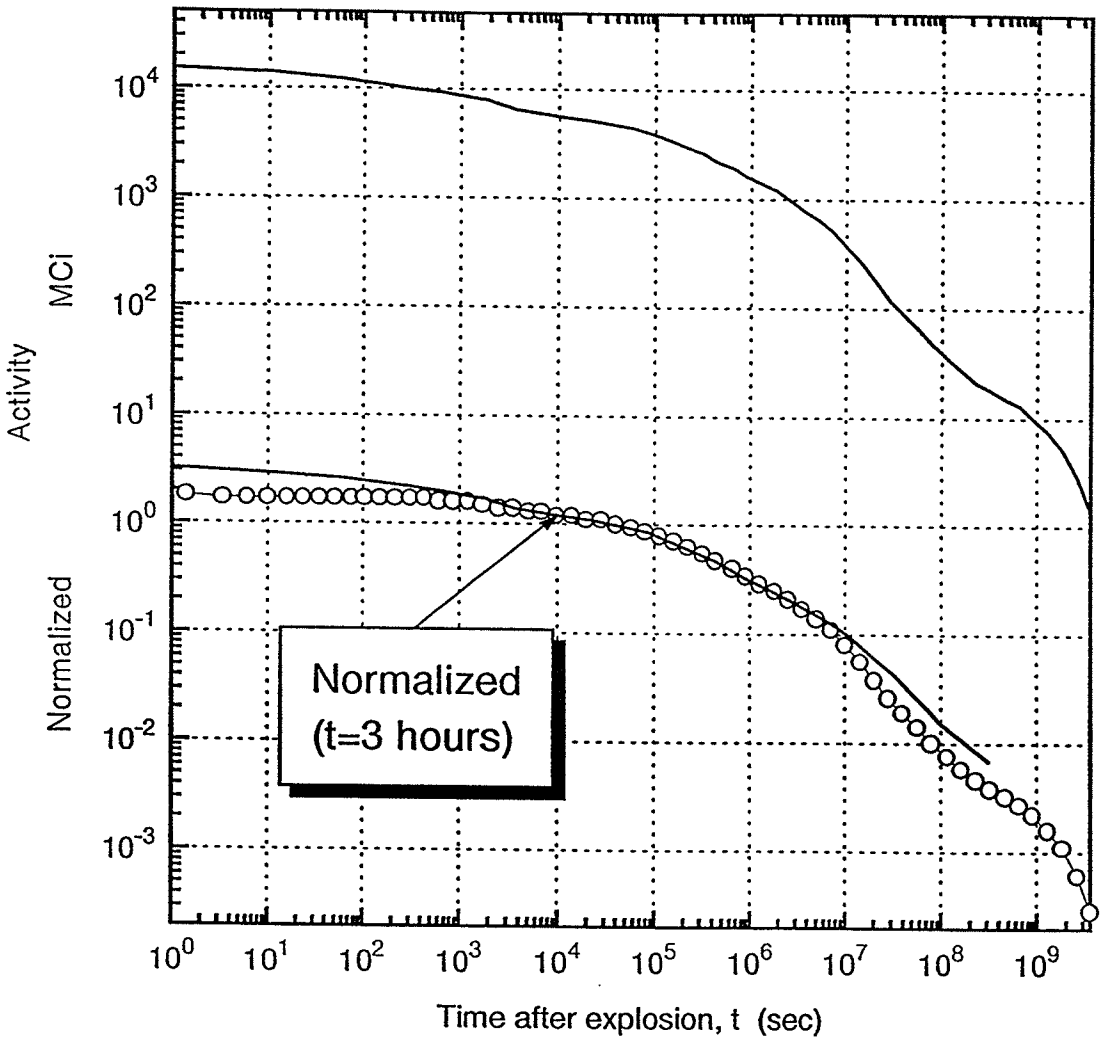

Figure 9: Total Activity of the ChNPP-4 Reactor (Constant Power). This figure demonstrates the method for obtaining short-term activity in the ChNPP-4 reactor. Since the normalized decay curve does not depend on the campaign time (see Fig. 8), we use its shape to get the short-term activity in the reactor. After normalizing at $t = 3$ hours (empty circles - our results, line - Fig. 8), we apply the short-term ($t < 3$ hours) RBMK curve to the calculated activity for the ChNPP-4 reactor.

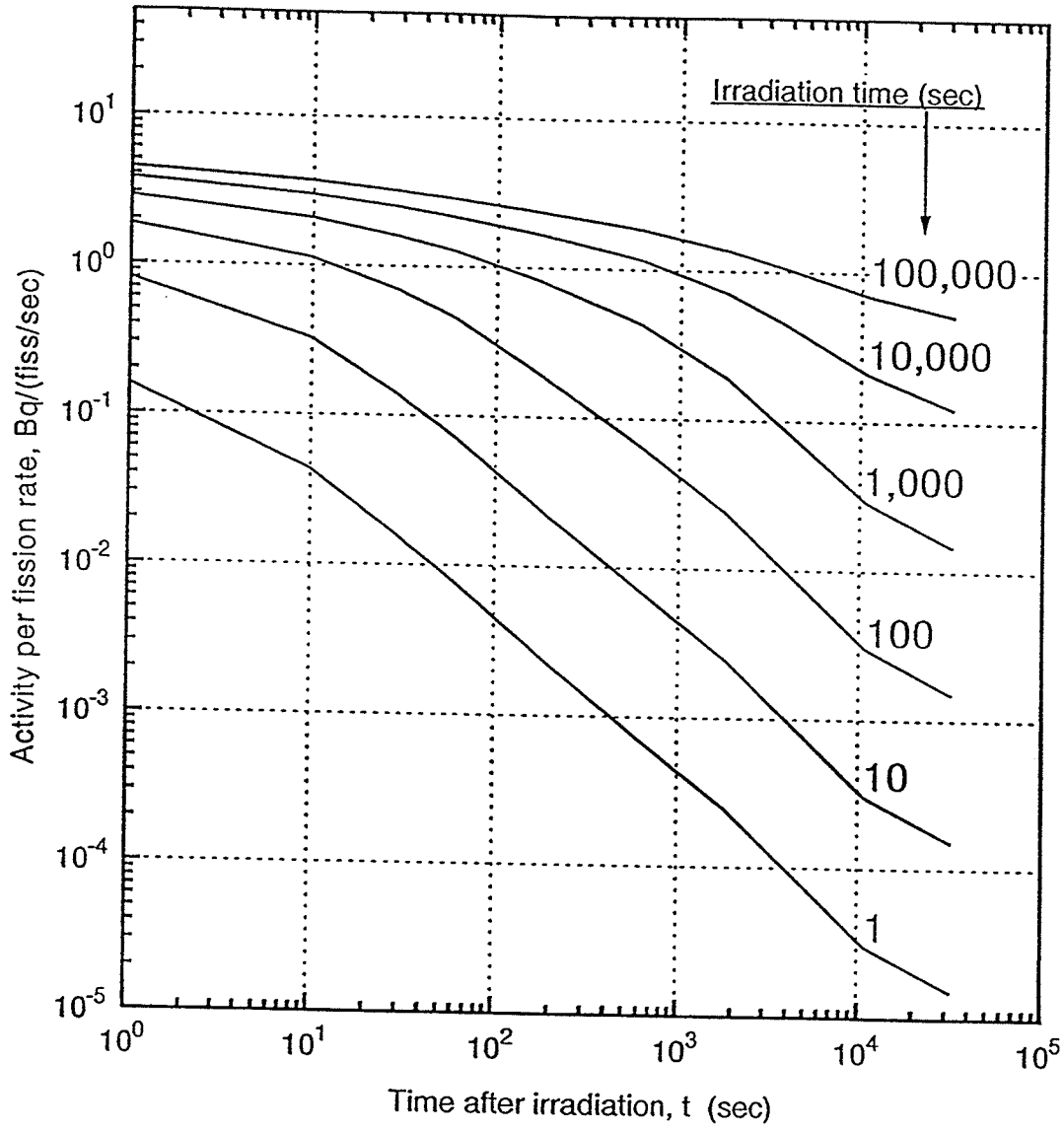

Figure 10: Fission product activity produced in short-term irradiated fresh RBMK fuel (plotted using data from [Spra 83]). The irradiation time is limited by the assumption that no secondary fuel is produced in the reactor during irradiation.

fission products activity produced during this period using the *integral* method and compare the result with the one obtained using equation (A.1). Hence, these data from [Spra 83] give us the opportunity to check our previously obtained results.

Data in Fig. 10 do not take into account the activity of fission products produced in the reactor by fission of Pu^{239} and Pu^{241}, these being produced during the *A-period*. However, since the data presented in Fig. 10 are normalized to the fission rate g,

$$g = \sigma_5^f \Phi U_5, \tag{22}$$

we will modify this normalization factor and use it in the form that will account for additional fuel produced in the reactor,

$$g = \Phi(\sigma_5^f U_5 + \sigma_9^f U_9 + \sigma_1^f U_1) = \Phi \Sigma_f. \tag{23}$$

This simplified formula does not take into account the difference in fission fractions corresponding to different fuel for a given isotope, and implies the same activity production rate for any fuel (corrected for fission cross section). Although this approach is not absolutely correct, the small amount of Pu isotopes as secondary fuel[31] produced in the ChNPP-4 makes this assumption reasonable.

8.2.1 *B-period*: Fission Product Activity

The *B-period* of the ChNPP-4 reactor lasted for 24 hours and 17 minutes. Therefore, we can use the *integral* approach to get the activity due to fission products produced in the reactor during this time. The results of these calculations are given in Fig. 9. At this point we are able to compare those results with the ones discussed in Section 6.

In order to make such a comparison let's calculate the fission products activity at the end of *B-period*. To do so, we will perform the following procedure:

- Step 1. *A-period* activity – solution of the equation (A.1).

  Calculate the activity at the end of *A-period* and let the fission products decay

[31] For fuel with burnup of 10.3 MWd/kg the amount of Pu^{239} and Pu^{241} together is 3 times less than U^{235}: 2.7 and 8.6 kg per ton of fuel, respectively [Doll 80].

during the *B-period* – (let's denote the result as A^1_{FP}). Since the *B-period* is about 24 hours long, all the short-lived isotopes not taken into account explicitly ($T_{1/2} \leq 0.5h$) will decay and our result will be valid despite the fact, that we used only isotopes with $T_{1/2} \geq 0.5h$.

- Step 2. *B-period* activity – *integral* approach.
  Calculate the activity (A^2_{FP}) produced during *B-period* (Fig. 11). In these calculations we used average neutron flux, corresponding to average power level equation (19), for different intervals that are shown on Fig. 3 with time marks. The sum $A^{12}_{FP} = A^1_{FP} + A^2_{FP}$ will give us the fission product activity at the end of the *B-period.*

- Step 3 *A-* and *B-period* activity – solution of the equation (A.1). Calculate the activity (A^3_{FP}) at the end of *B-period* using the power history (Fig. 3) and formulas from Appendix A (data from Table 7 for real power regime).

Since the last result will be valid only some 3 hours after the end of *B-period*, we will let the activity A^{12}_{FP} calculated at Step 2 decay for 3 hours and after that compare it with the result from Step 3.

Calculation of Step 3. gives us $A^3_{FP} = 2900MCi$ whereas after 3 hours decay A^{12}_{FP} $= 2570 + 470 = 3040MCi$. There is only a 5% difference between these two figures and we consider this result as acceptable to justify the procedure we followed in our calculations.

8.2.2 *C-period*: Fission Product Activity

As we mentioned before, power pulses that correspond to this "run away" period (see Fig. 3) were *calculated* by the Soviet team and reported in Vienna. No detailed information about assumptions and models used in these calculations was given. An analysis of this period was made by an American team [AmRe 86]. Based on their analysis, they state that the Soviet power history during the *C-period* was obtained under the assumption of coherent fuel failures, i.e. a point-kinetic model has been

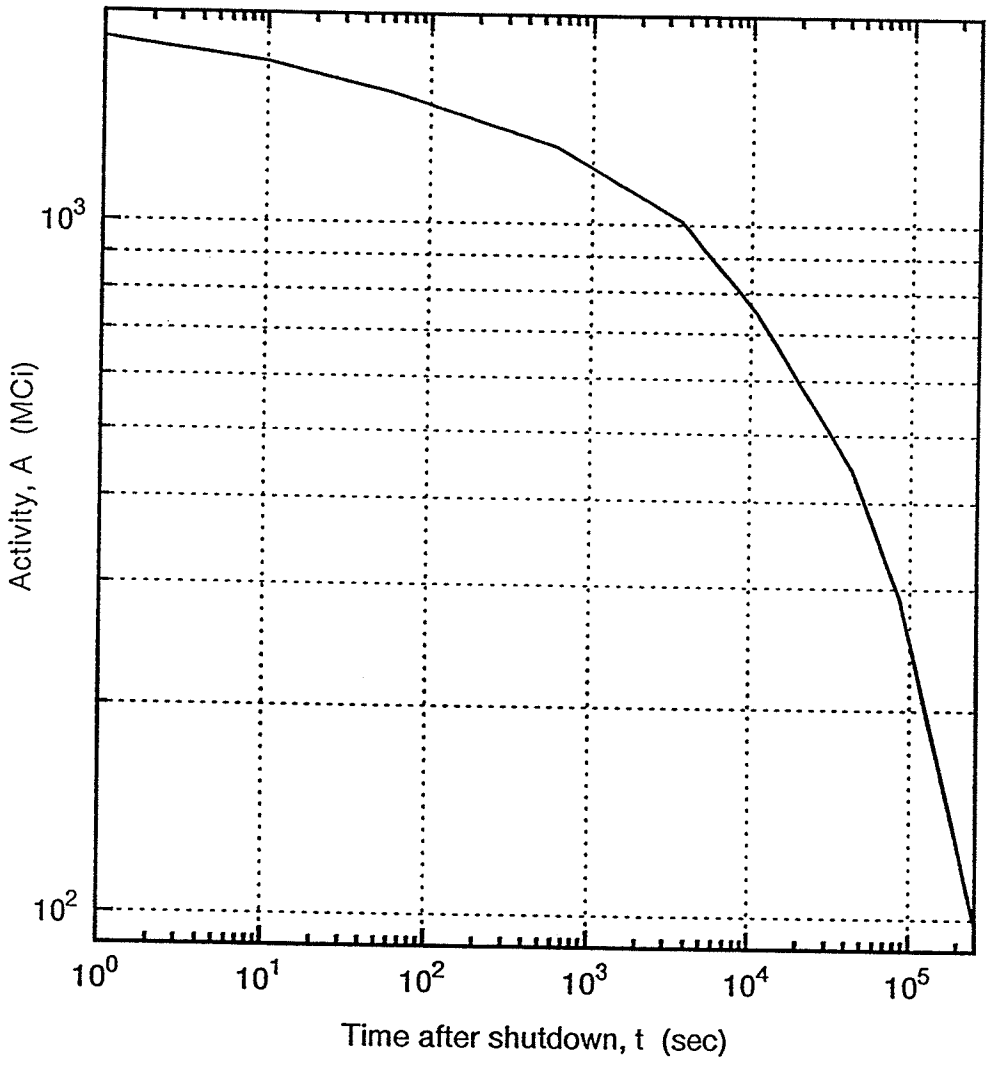

Figure 11: Calculated fission products activity produced in the ChNPP-4 reactor during B-Period, calculated using the data for short-term irradiated fresh RBMK fuel (see Fig. 7). Since the B-Period lasted for about 24 hours, we can use this approach: maximum irradiation time allowed is about 28 hours.

used to produce this result. An account for noncoherence in fuel failures (using the spatial distribution of the reactor parameters) can "stretch out" the *C-period* and therefore, delay the time of explosion) [AmRe 86]. A similar correction to the time of explosion reported in Vienna was made by Soviet scientists [Veli 90], which is also in agreement with G.Medvedev's [Medv 88] statement (without any references or explanations) that the explosion occurred at 1.23.58 a.m. – 12 seconds later than stated in the Vienna report.

In addition, even using a point-kinetic model the American team was unable to reproduce the *C-period* power history from the Soviet Vienna report. According to their best estimates, the power excursion in the last peak (explosion) should be 4 times lower than stated in the Soviet report.

Again, using the *integral* approach we will calculate the activity using three different power scenarios during the *C-period*:

- As given by the Soviet team in the Vienna report (coherent fuel failure). In this case about 222 **MWh** energy was produced in the ChNPP-4 reactor during *C-period*.

- As given in the Vienna report with 4 times lower power peak (120 times the nominal power instead of 470 times) – coherent fuel failure in the American interpretation: 75 **MWh** energy produced.

- Additional 6 power peaks that mirror the first small peak before the explosion (last power excursion) – our approximation of the noncoherent fuel failure: 355 MWh energy produced. This scenario shifts the time of explosion to 01:23:58 a.m.

The results of those calculations are shown on Fig. 12.

8.2.3 Total Fission Product Activity

Now we are able to calculate the short- and long-term activity accumulated in the ChNPP-4 reactor due to fission products. We must simply add the results in Fig.

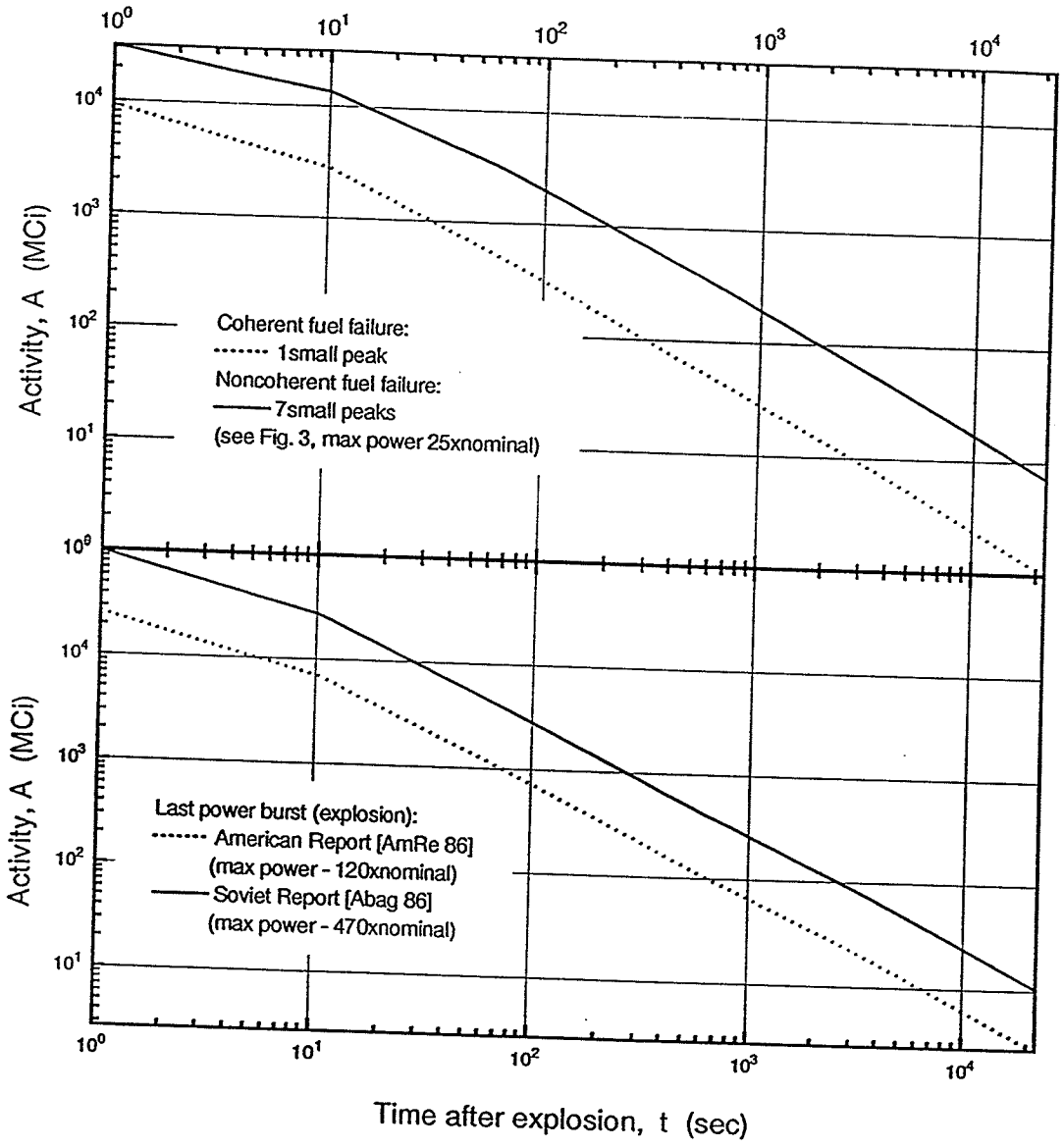

Figure 12: Radioactivity produced in the ChNPP-4 reactor during the explosion: first stage (top) and second stage (bottom) for different models used in our calculations.

11 (*B-period*), Fig. 12 (*C-period* for the Soviet and American scenarios) and Fig. 9 (after 24 hours decay time, i.e. shifting the time scale for 24 hours). This result is shown in Fig. 13.

8.2.4 *B-* and *C-period*: Actinide Activity

The integral approach does not allow us to calculate the short-term activity due to the actinides since it considers only fresh fuel and uses short irradiation times (up to 100,000 sec) for which the amount of actinides produced is negligible[32]. We calculated the actinides activity using our list of isotopes (Table 7), but this result is valid for times from about 3 hours and later.

To overcome this problem we turn again to the activity data given in [Spra 83] for different campaign times. In comparing the data for fission products and actinides radioactivity, we find that the actinide activity accounts for about 20% of the total activity during the first few minutes and up to 33% for times from few hours to 1 day. Therefore, we simply apply those ratios to the fission product activity given in Fig. 13. The final result of the total activity (fission products and actinides) for the Soviet and American estimates is given in Fig. 14.

We would like also to know the difference between the accumulated activity for "ConstP" and "RealP" scenarios. The curves in Fig. 14 provide information for those comparisons.

Now we can calculate the total activity in the ChNPP-4 reactor 10 days after the accident and compare with the Soviet figure given in Vienna. Our calculations, as shown on Fig. 14, give 1450 MCi. According to the Soviet Report [Abag 86], 50 MCi (decay corrected to May 6, 1986) had been released, which accounted for 3.5% of the total activity in the reactor (noble gases excluded). This gives us $50/0.035 = 1429$ MCi. If we include 50 MCi of activity due to noble gases (see Table 2 and also Fig. 19) we get that their estimate was 1479 MCi.Thus, there is only 2% difference between this figure and the one we have calculated (this difference is undistinguishable on Fig.

[32]The activity of U^{235} and U^{238} initially present in the RBMK reactor is negligible in comparison to the fission products activity.

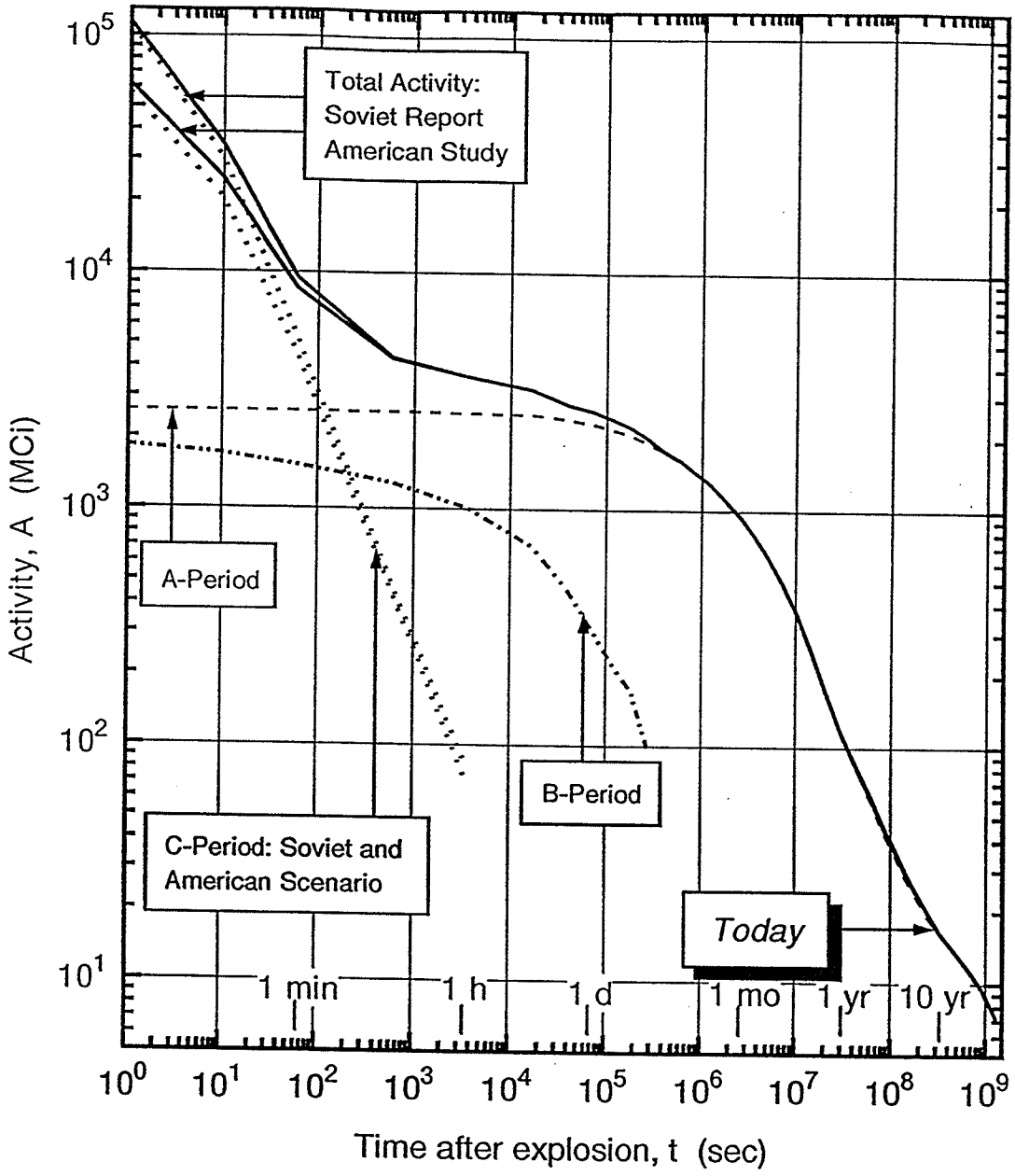

Figure 13: Total fission products activity produced in the ChNPP-4 reactor calculated for different scenarios: activity produced during the B- and C-Periods was calculated using the integral approach (see chapter 8.2 Real Power Regime - Integral Approach); A-Period activity calculated using equation (A.1) - see Appendix A.

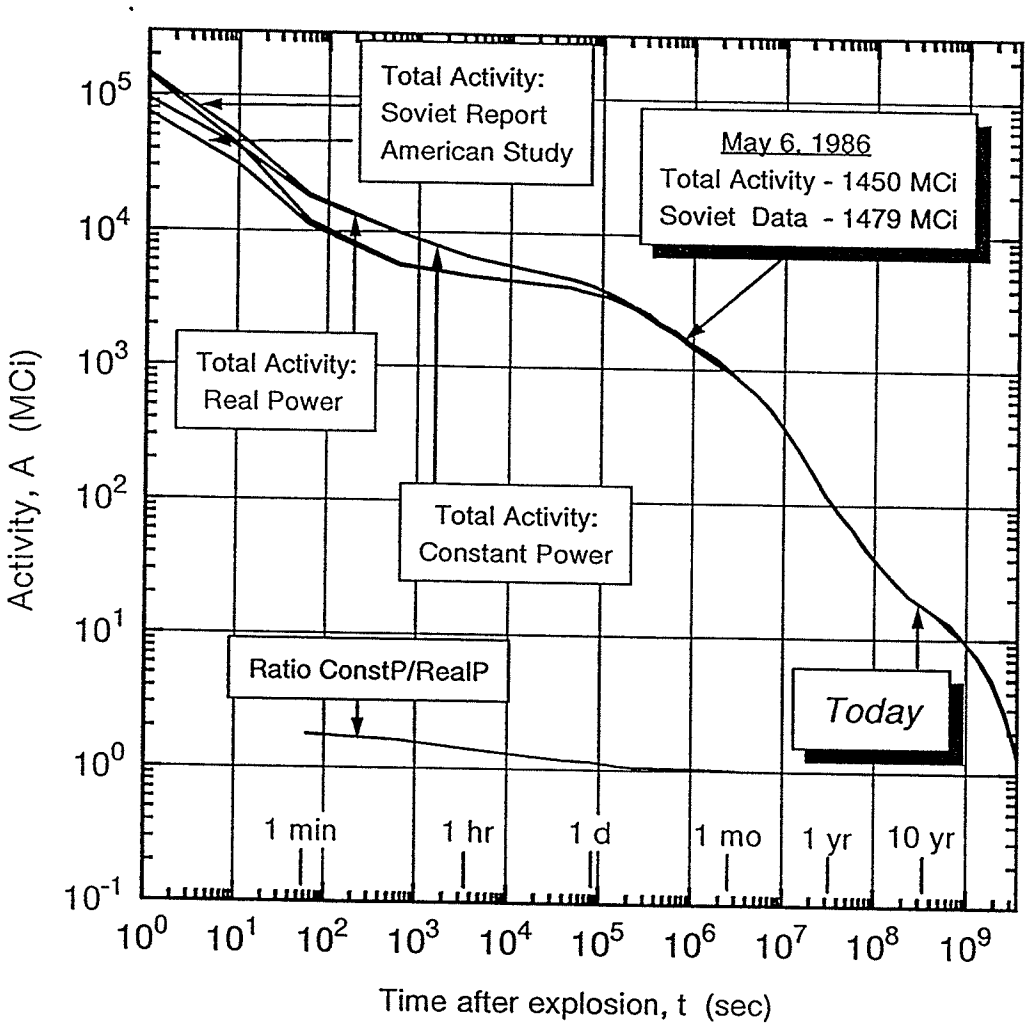

Figure 14: Total activity (fission products and actinides) produced in the ChNPP-4 reactor for different scenarios: fission products (short- and long-term) activity is taken from Fig. 13; long-term actinide activity is calculated using the formulas from Appendix A; short-term actinide activity is calculated using the ratio for actinide/fission product activity taken from [Spra 83].

14).

In addition, we calculated the radioactivity of the iodine isotopes after the explosion. Those data are shown in Fig. 15.

To show the importance of taking into account the burnup of the reactor fuel, we calculated the energy producing ratios (ω_i, see Section 6.1, Production of Actinides) for two groups of fuel assemblies with different burnup (see Fig. 16). It can be noticed, that in fuel with highest burnup – 14.4 MWd/kg – almost half of the energy is produced in fission of Pu^{239}. Therefore, it is important to include Pu^{239} that is produced in the reactor.

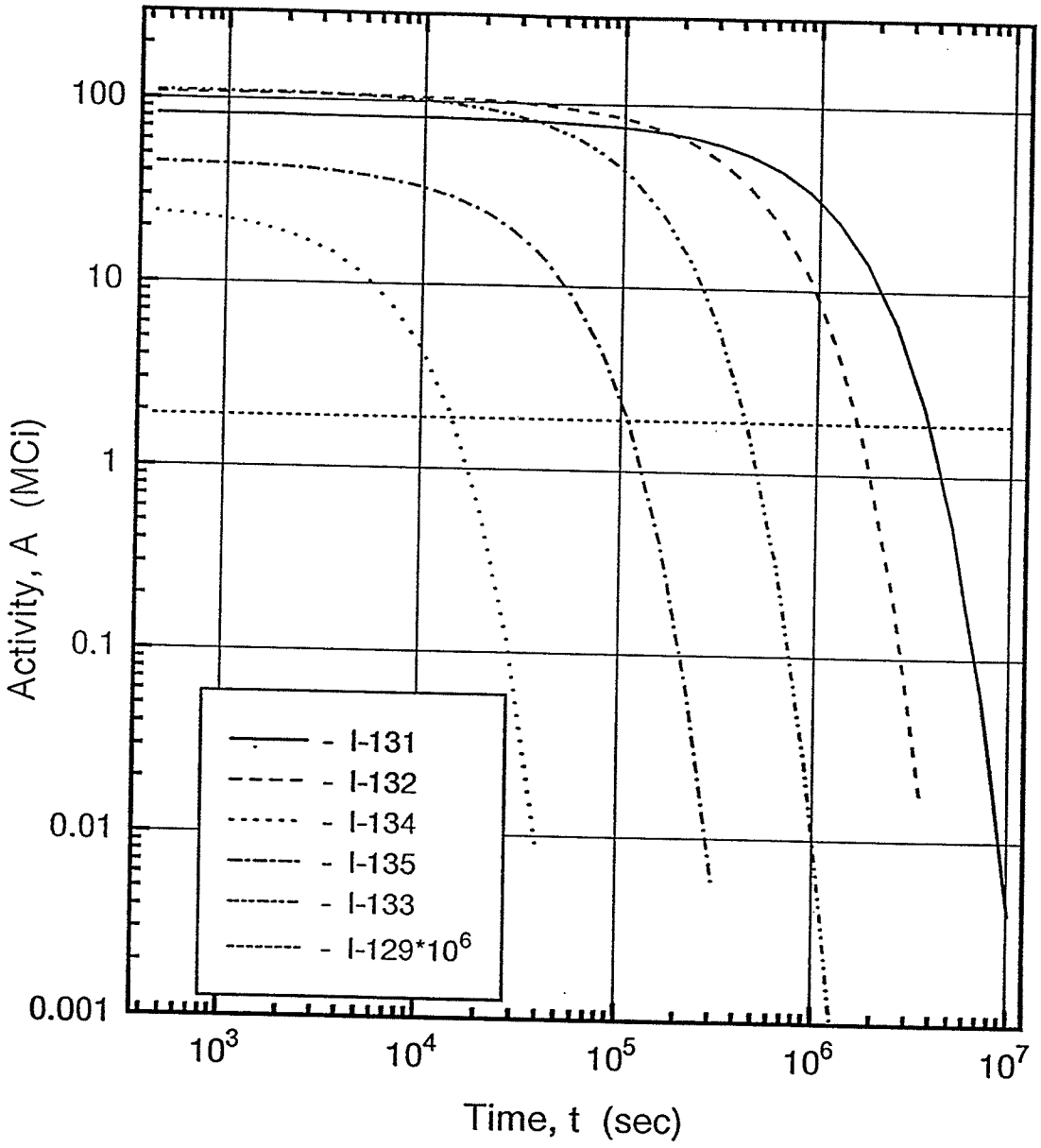

Figure 15: Calculated time dependence of the iodine radioactive inventory in the ChNPP-4 reactor after explosion (18 groups of fuel assemblies, real power scenario, no C-Period).

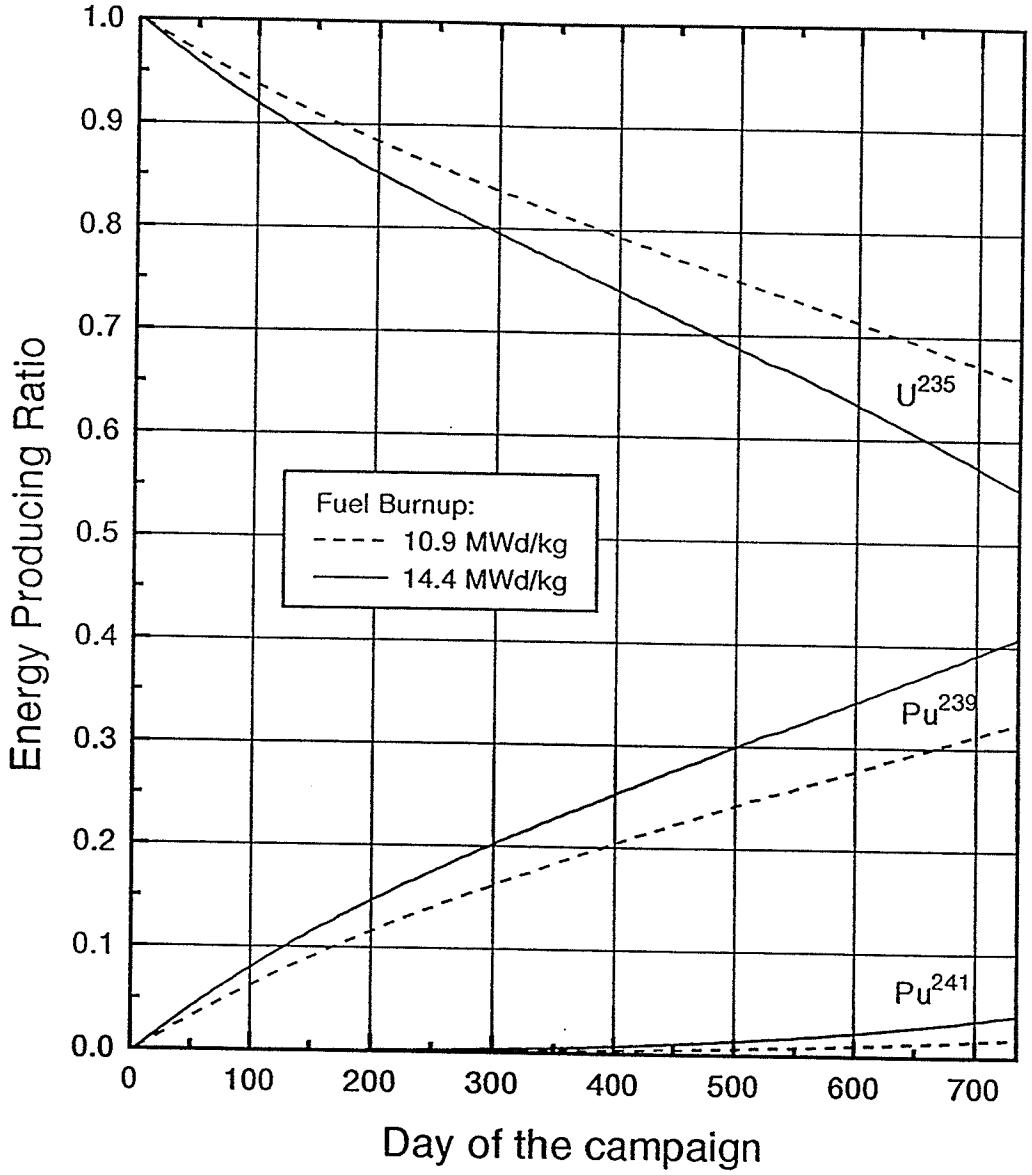

Figure 16: Energy producing ratios for different fuel assemblies. In fuel with burnup of 14.4 MWd/kg almost half of the energy is produced in fission of Pu^{239}.

9 ChNPP-4 Radioactivity Release

In previous sections we calculated the ChNPP-4 long-term and short-term radioactive inventory as well as the time dependence (due to the radioactive isotope decay) of this activity. In order to estimate the impact of this activity (and primarily - the health effects this activity can cause), we need information about the amount of this activity that was released into the atmosphere.

These data – total amount of radioactivity released on a daily basis as well as fractions for long-lived isotopes, both decay corrected to May 6 – were given in the Soviet Report in Vienna [Abag 86]. These data are claimed to be of experimental origin and checking this information goes beyond this thesis. However, by comparing the reported results [Abag 86] with the ones from some American reactor research publications, we will be able to estimate the plausibility of the Soviet data.

9.1 WASH Reports and APS Light-Water Reactor Safety Study

Studies of potential consequences of a major nuclear reactor accident have been undertaken by the Atomic Energy Commission (AEC) of the US in 1957 [WASH 57], in 1965 [WASH 65] and in 1974 [WASH 74][33]. The preliminary draft of the last report (well known as WASH-1400 Report) was reviewed in an independent light-water reactor safety study [AECR 75] sponsored by the American Physical Society (APS). This study used some more simplified calculational models, but nevertheless was able to confirm the information provided in the WASH-1400 report by producing comparable results.

The purpose of these two studies (WASH-1400 and [AECR-75]) was to make a quantitative estimate of the likelihood of an accident of given severity. The WASH-1400 Report is a detailed analysis of such consequences, whereas the APS Study deals only with releases-contamination-doses parts of the WASH-1400 Report.

[33]The [WASH 65] report was planned to use more realistic assumptions than [WASH 57]. However, after drafts of the new report had been circulated for review within AEC and the nuclear industry, a decision was made to abort the study [WASH 74].

The American nuclear power reactor design differs significantly from the RBMK reactor design. However, as will be shown below, most of these two reactors' parameters relevant to the problem of radioactivity release – power, fuel type, different isotope contribution factors to the radioactive inventory – are comparable, and we can use the results obtained in those American reports in our estimates of the ChNPP-4 radioactivity release.

9.2 ChNPP-4 and PWR Reactors

Both the APS Study and the WASH-1400 Report consider a "Reference Accident" on a Pressurized Water Reactor (PWR) a meltdown of a 1000 MWe PWR with essentially complete containment failure.

To estimate the activity release in the "Reference Accident" in a PWR, the radioactive nuclides were divided into seven principal groups based on chemical properties, boiling and melting temperatures, ability to form oxides, volatility of those oxides, etc. The fraction of the accumulated radioactivity that was released into the atmosphere was estimated for some isotopes in these principal groups and was assigned to all isotopes in the corresponding group. The per-group releases in the PWR "Reference Accident" are given in Table 9. The ChNPP-4 column in this table contains information taken from the [Abag 86]. We also assigned the fraction of the given isotope releases [Abag 86] to all isotopes in the corresponding principal group[34]. The main parameters of those two reactors are also given in Table 9. Excluding the non-volatile oxides, there is a quite good agreement in the ratios of the activity released. Data in Table 10 and Table 11 give the estimated amount of activity present in the reactors on the per-isotope basis as well as released for each principal group (only those isotopes under consideration in WASH-1400 are shown is Table 10). Again, there is a good agreement between these data[35].

[34]In this section we use the calculated data for ChNPP-4 constant power scenario since in the "Reference Accident" it is assumed that the reactor exploded at full power.

[35]The 20% release of the total inventory activity in the ChNPP-4 reactor (Table 11) should not be compared with the Soviet figure 3.5% [Abag 86] since the last is valid only 10 days after the explosion (see Fig. 14.)

Table 9: PWR vs. ChNPP: parameters and release fractions.

Principal chemical groups	**PWR Reference Accident WASH-1400**	**ChNPP-4**
Noble gases	0.9	up to 1.0
Iodines	0.7	0.2
Cesiums	0.5	0.1-0.13(1)
Telluriums	0.3	0.15
Alkaline Earth	0.06	0.04-0.06
Volatile Oxides	0.02	0.023-0.029
Nonvolatile Oxides	0.004	0.023-0.032
Reactor Parameters		
Power, MWe	1000	1000
Fuel enrichment, %	3.3	2
Fuel exposure, MWD/kg	17.6	10.9

(1)The amount of cesium released has been corrected to be 0.25-0.30 [Izra 87].

Table 10: PWR vs. ChNPP: inventory activity. The WASH-1400 data were taken from [AECR 75], the ChNPP-4 data - from Table 7, 18-group model, constant power regime. The per chemical group radioactive inventory, as well as the released activity is given in Table 11.

Chemical group Isotope	Halflife (hours)	Present Paper (MCi)	WASH -1400 (MCi)
Noble Gases			
Kr-85	93972.2	0.75	0.6
Kr-85m	4.48	25.5	26
Kr-87	1.27	52.7	52
Kr-88	2.84	75.9	76
Xe-133	125.89	185	170
Xe-135	9.09	185	26
Iodines			
I-131	192.97	86.4	85
I-132	2.3	133	120
I-133	20.80	185	170
I-134	0.88	204	200
I-135	6.61	164	150
Telluriums			
Te-129m	806.39	26.3	28
Te-129	1.16	8.8	10
Te-131m	30.00	11.8	15
Te-132	78.19	133	120
Cesiums			
Cs-134	18075.0	4.1	1.7
Cs-136	311.53	1.7	6
Cs-137	264472.	6.8	5.8
Alkaline Earth			
Sr-89	1211.94	97.5	110
Sr-90	255250.	5.6	5.2
Sr-91	9.48	129	130
Ba-140	306.94	163	160

(Table 10 continued)

Chemical group Isotope	Halflife (hours)	Present Paper (MCi)	WASH -1400 (MCi)
Volatile Oxides			
Mo-99	66.00	168	160
Tc-99m	6.02	168	140
Ru-103	942.78	115	100
Ru-105	4.44	70.9	58
Rh-105	35.36	70.8	58
Ru-106	8836.11	24.9	19
Nonvolatile Oxides			
Y-90	64.00	5.61	5.2
Y-91m	1404.17	125	140
Zr-95	1538.33	155	160
Zr-97	16.90	159	160
Nb-95	842.50	151	160
La-140	40.25	164	160
Ce-141	780.00	149	160
Ce-143	33.00	148	150
Ce-144	6825.00	101	110
Pr-143	325.56	147	150
Nd-147	265.44	62.1	60
Pm-147	22997.2	22.1	17
Pm-149	53.08	31.5	40
Pu-239	0.21+11	0.022	0.01

Table 11: PWR vs. ChNPP: Total Activity and Release. The activity in the ChNPP-4 reactor (per chemical group and total) was calculated on basis of the results from Table 7 and 8 and Table 10. The released activity was calculated on basis of the release fractions given in Table 9 (average values were used for cesium, alkaline earths, volatile and nonvolatile oxides).

Chemical Group	*PWR Activity (MCi)*		*ChNPP-4 Activity Const P (MCi)*		*ChNPP-4 Activity Real P (MCi)*	
	In core	Released	In core	Released	In core	Released
Noble gases	350	315	525	525	300	300
Iodines	725	508	772	154	374	75
Cesiums	13.5	6.8	12.6	3.5	12.5	3.5
Telluriums	173	52.	180	27	125	18.8
Alkaline earth	405	24	395	20	306	15
Volatile oxides	535	11	618	16	499	13
Nonvolatile oxides	1472	6	1420	39	1286	35
Total	**3675**	**923**	**3922**	**785**	**2903**	**460**
(% released)		**25**		**20**		**16**

Finally, we can compare the time dependence of the total activity in the PWR [AECR 75] and ChNPP-4 reactors after shutdown[36]. Those data are shown in Fig. 17. We can notice that there is no significant difference between the data for these two reactors.

Thus we can conclude that although these two reactors have a completely different design, and use different fuel elements and enrichment, the total amount of radioactivity present in these reactors is comparable. Finally, the release fractions reported in [Abag 86] and WASH-1400 are also comparable. Therefore, we will assume that the same methods are applicable – applying the release fraction of a given isotope (that is known) to all the isotopes in its chemical group (this time all the isotopes from Table 7) – as in the PWR accident analysis by the American researchers. In our further calculations we will use the published release fractions [Abag 86].

The melting and/or boiling points of the isotopes used in our calculations and their oxides are given in Table 12[37]. Separating the isotopes into volatile and non-volatile oxides is somewhat arbitrary since this chemical behavior of a given isotope depends on the temperature of the core. It is not clear how this separation was done in the WASH-1400 Report. But since we use the released fractions reported in the Vienna report, this will not make any difference – the reported fractions are the same for both chemical groups. Moreover, these fractions are higher than considered in the "Reference Accident" in WASH-1400 and APS reports.

9.3 Short-term Activity Releases

Short-term activity produced mainly during the *C-period* in the ChNPP-4 reactor campaign requires special consideration. Due to its nature, this activity will have an impact very limited in time – some few hours after the explosion. Since the accident happened at 01:24 in the morning this means that only people in close vicinity to the ChNPP-4 were affected. Therefore, an analysis of the impact of short-term activity

[36]For the ChNPP-4 reactor those data were taken from Fig. 9.

[37]Only oxides [Hand 94] with known at least one critical temperature – boiling or melting point – are given in the table.

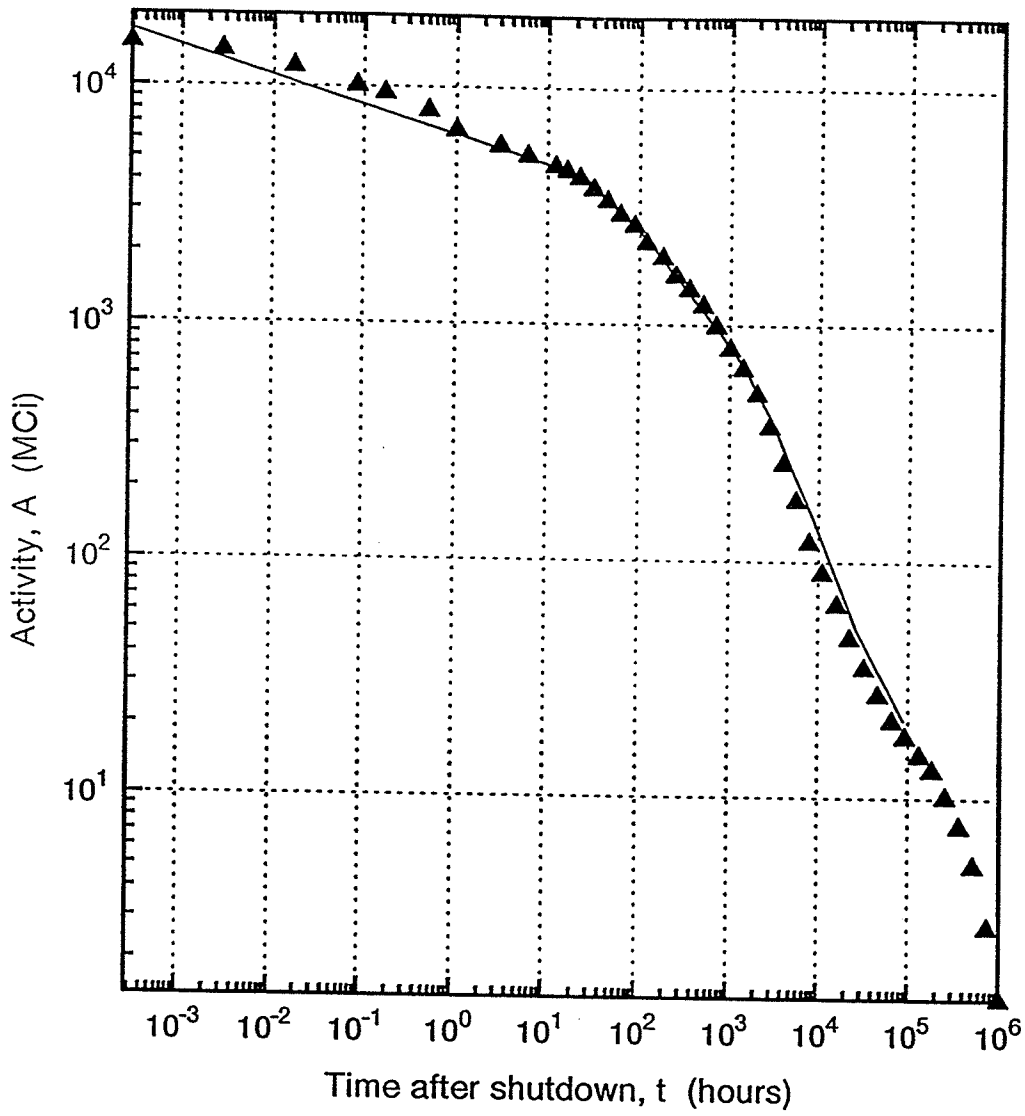

Figure 17: Total activity in the PWR and ChNPP-4 reactors after shutdown: ChNPP-4 - filled triangles, PWR - line. The PWR data were taken from [AECP 75] and correspond to a 1100 MWe PWR similar to the one considered in the "Reference Accident" in the WASH-1400 Report. The ChNPP-4 data were taken for constant power regime, A+B-Periods scenario (see Fig. 9).

Table 12: Melting and boiling temperatures (where known) for isotopes used in calculations [Hand 94]. Isotopes given in italics were not used in WASH-1400 Report, but are used in this thesis (see Table 7) [(d) - decomposes, (v) - volatizes].

Isotope	Melting temp. °C	Boiling temp. °C
Br	*-7.2*	*58.78*
Br_2O	*-17*	
BrO_2	*0(d)*	
$Br_2 10H_2O$	*6.8(d)*	
Kr	-156.6	-152.3
Xe	-111.9	-107.1
Sr	769	1334
Ba	725	1640
BaO	1918	2000
BaO_2	450	800
Cs	28.4	669.3
Cs_2O_2	400	650
Cs_2O_3	400	
I	113.5	184.4
IO_2	75(d)	
I_2O_5	300-350	
I_4O_9		75(d)
Te	452	1390
TeO_2	733	1245
TeO	370(d)	
TeO_3	395(d)	
Mo	2610	5560
Ru	2310	3900
RuO_4	25.5	108(d)
Rh	1966	3727
Rh_2O_3	1100	
Pd	*1554*	*2970*
PdO	*870*	
In	*1566*	*2050*
In_2O_3		*850(v)*
Y	1522	3338
Y_2O_3	2410	
Nb	2468	5127
Nb_2O_5	1520	
Nb_2O_3	1780	

(Table 12 continued)

Isotope	Melting temp. °C	Boiling temp. °C
Zr	1852	4377
ZrO_2	2700	5000
La	918	3464
La_2O_3	2307	4200
Ce	798	3443
Ce_2O_3	1692	
Pr	931	3520
PrO_2	>350	
Nd	1021	3074
Nd_2O_3	1900	
Ag	*961.33*	*2212*
Ag_2O	*230(d)*	
Ag_2O_2	*>100(d)*	
Cd	*320.9*	*765*
CdO	*>1500*	
Sn	*232*	*2270*
SnO_2	*1630*	*1800*
SnO	*1080(d)*	
Sb	*630.5*	*1750*
Sb_2O_5	*380*	
Sb_2O_4	*930*	
Sb_2O_3	*656*	*1550*
Ge	*937.4*	*2830*
GeO_2	*1086*	
Pm	1042	3000
Pu	641	3232
Np	*630±1278*	
Np_3O_3	*500 (d)*	
Sm	*1074*	*1794*
Eu	*822*	*1527*
As	*817 (28a)*	*613*
U	*1132*	*3818*

on those people would heavily depend on the location at any given time of each person that happened to be close to the reactor. For those people (e.g. operators, firemen) the activity that remained in the reactor might add significantly to the total effect of the radiation. Therefore, the average approach that is often used in calculation of the impact of the radioactivity released (and in which the results of our calculations could be used) is not applicable, and we will not consider the short term activity releases (up to few hours) in this thesis.

9.4 Releases on a Daily Basis

The figures of activity released depend on the "point of view" – how much time passed from the accident until the measurements were performed. In the PWR "Reference Accident" it is assumed that the activity was released in one blast, i.e. the released fraction of the radioactive inventory was decaying while being outside the reactor. This was not the case in the ChNPP-4 accident. According to [Abag 86] the ChNPP-4 reactor had been releasing activity for 10 days. This in particular was the reason why in the Soviet report in Vienna [Abag 86] the activity released was decay corrected to May 6, 1986, 10 days after the accident. The same approach was used in [Izra 87] that reported different activity release dynamics (another model and some additional experimental data were used), but no information about how those results were obtained was given. Both of these results are shown in Fig. 18.

We will be interested in the activity *as of the day of release*, since just after the release the activity presents a potential danger to the population. These figures are more important than the data 10 days after the accident.

9.4.1 Releases on the Day of the Accident

The releases during the first day after the accident were given in the Soviet Report in Vienna [Abag 86] (see Table 2). As we discussed earlier (see Section 3.2, ChNPP-4 Reactor Inventory – Vienna Report), it is not clear what the data for the releases on the day of the accident mean (Table 2, column 2), and therefore we cannot make use

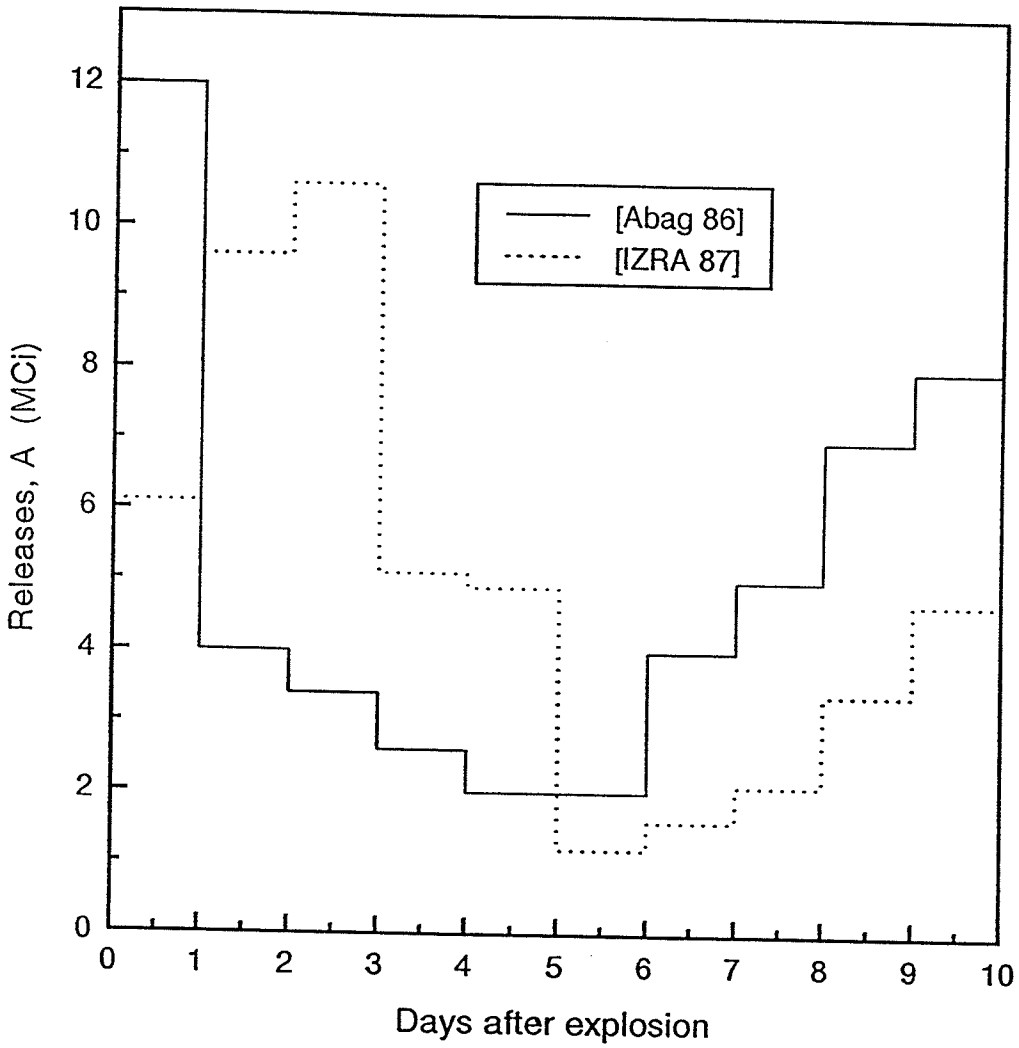

Figure 18: Daily ChNPP-4 radioactivity releases during the 10 days after the explosion from different papers. Unlike the Soviet report in Vienna [Abag 86] (see also Table 3), [Izra 87] claims the releases were increasing during the first three days.

of this information. However, we can use the data from Table 3 about the amount of activity released as of May 6. According to these data, 12 MCi were released on the day of explosion (noble gases excluded). Since only 10% of the initial activity is left in the ChNPP-4 reactor 10 days after the chain reaction stopped (see Section 3.1, Reactor Power History), the released activity *as of the day of release* was 120 MCi [38]. This figure is in agreement with that stated in [Izra 90]: 120-150 MCi. If we use the assumption that noble gas releases had the fractions of the average [Izra 90] releases (in our case 40% of the total releases 300 MCi for noble gases (see Table 11) happened during the first day[39]), we arrive at the total number of 240 MCi released including noble gases.

9.4.2 Releases During the Following 10 Days

A similar approach can be used to obtain the releases during the following 9 days after explosion. In this case we must convert the reported in Vienna releases 10 days after the explosion (see Table 11) to the day of release. This can be done by using the time dependence of the activity from in Fig. 13 for the "RealP" scenario[40]. To include the noble gases in the total releases, let's consider the decay curve of the noble gas activity, that is presented on Fig. 19. Since we know the fractions of release during each day (data from Table 3 converted to releases as of the day of release), we apply them to the noble gas activity and correct for decay using Fig. 19. Adding this to the rest of the releases, we get the total activity released during each of the ten days after the accident (as of the day of release). The results of these calculations are shown on Fig. 20. The same approach was used in the [Izra 90] calculations (the data

[38] At this point we can get back to the controversial Table 2, column 2. Since the activity of the given isotopes adds up to about 15 MCi (noble gases excluded) - which is close to 12 MCi from Table 3 - we come back to the assumption that the presented data are as of May 6 and are not included in column 3. Therefore, the total activity released *as of the day of release* will be about 150 MCi.

[39] 300 MCi of noble gases have been released (see Table 11), hence 120 MCi - during the day of explosion.

[40] Here we assume that the decay curve of the releases will follow the same shape, i.e. the activity of different groups has the same release fraction during a given day. More accurate calculations could be performed if more data for per-group releases were available.

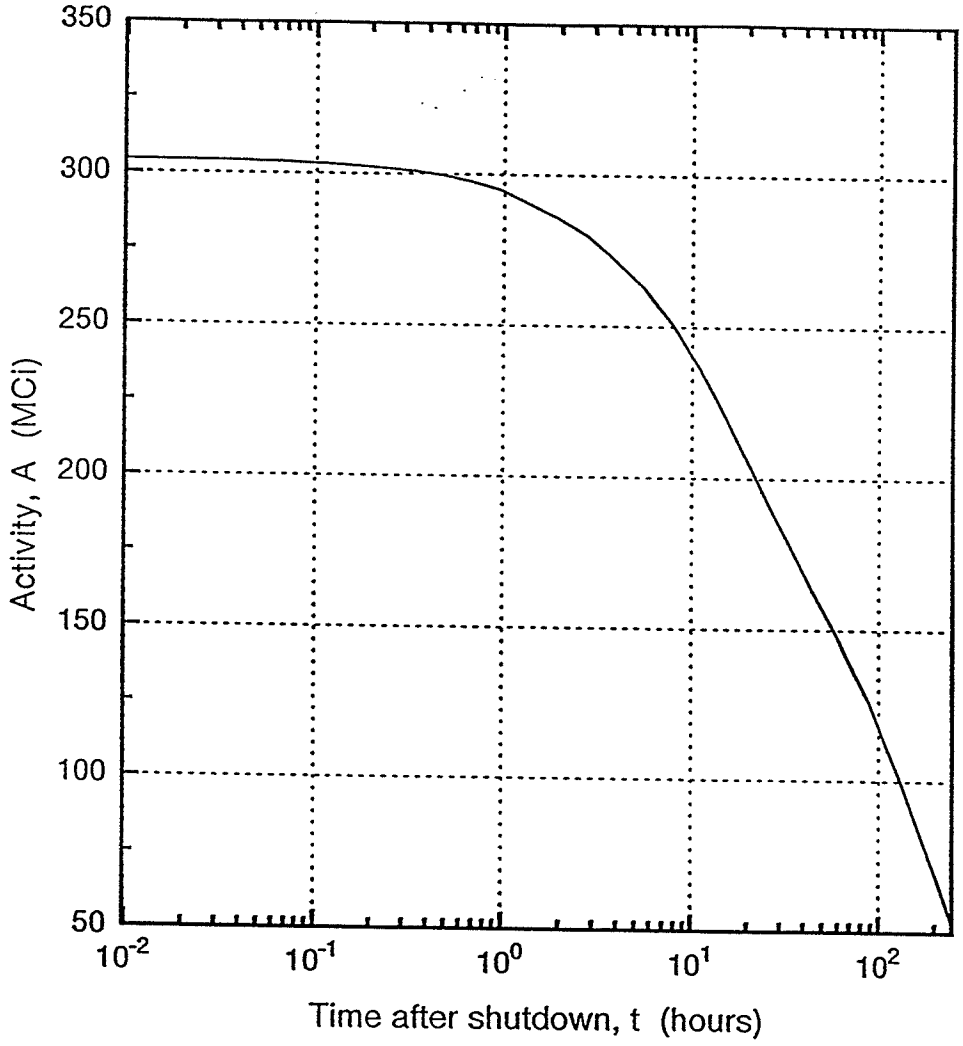

Figure 19: Time dependence of the noble gas activity after explosion.

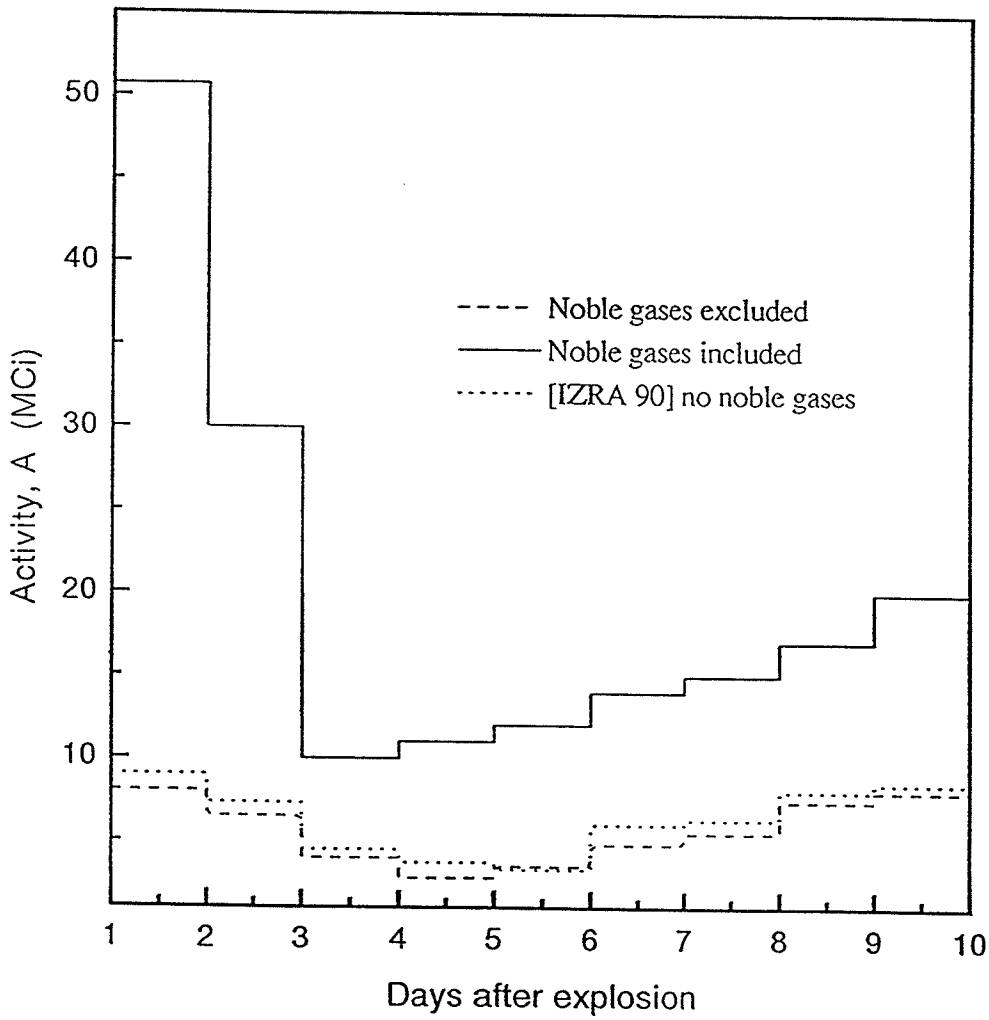

Figure 20: Calculated radioactivity releases (day of explosion excluded) "as of the day of release" and data from [Izra 90] for comparison.

corresponding to the 800 days campaign from [Spra 83] were used). As was shown (see Section 7.1, Constant Power Regime), all three curves (800, 1100 and 1400 days, normalized) agree for times from 1 to 1,000 hours (see Fig. 8), and therefore the results are very close for releases excluding noble gases.

10 Conclusions

In the present thesis the radioactive inventory due to fission products and actinides as well as the activity released into the atmosphere during the ChNPP-4 reactor accident were studied.

Using a simple computer model, we calculated the activity in the ChNPP-4 reactor. Two different models were used for these calculations. For isotopes with $T_{1/2} \geq 0.5$ hour the activity due to each such isotope was calculated. The obtained results are valid for any time $t > 3$ hours after the explosion.

An *integral* approach based on information given in [Spra 83] was used for short term radioactivity calculations – due to isotopes with $T_{1/2} \leq 0.5$ hour. This method gives only the total activity from all isotopes (fission products and/or actinides group) present in the reactor.

A special method was used to calculate the activity produced in the reactor during the last few seconds and during the explosion itself. Data about the radioactivity of short term irradiated fresh RBMK fuel from [Spra 83] were used. These calculations were performed for different models (Soviet and American) describing this period.

By comparing the total activity in the reactor for two different scenarios, both constant and real power (see Fig. 14), we can conclude that this activity would be twice as high if the reactor exploded at full power. Implication of this result is that firemen would have been getting radiation sickness in about 15 minutes after arriving at the site, rather than after about half an hour, had the reactor exploded at full power. This could have made impossible for the firemen to extinguish the fires. For times some 10 hours and more after the accident the difference in activity produced in these two scenarios is negligible.

The calculated total radioactive inventory in the ChNPP-4 reactor and the data provided by the Soviet team in Vienna allowed us to calculate the activity releases on a daily basis.

A Appendix A

A.1 Solution of the General Equation

The number of nuclei of each isotope in the chain $N_k(t)$, can be calculated by solving the following set of differential equations:

$$\frac{dN_k}{dt} = F_k(t) - \sigma_k N_k + \sigma_{k-1}^k N_{k-1}; \qquad N_k(0) = N_k^0, \tag{A.1}$$

where $k = [1, L]$, L – number of isotopes in the chain and N_k^0 - the initial amount of a given isotope in the reactor.

To solve the set of differential equations (A.1), let us express the solution in form[41]

$$N_k(t) = N_k^0 e^{-\sigma_k t} + \sum_{i=1}^{k} a_i^k e^{-\sigma_i t} + \sum_{i=1}^{k} \int_0^t b_i^k e^{\sigma_i (y-t)} dy, \tag{A.2}$$

where a_i^k and $b_i^k(t)$ are some coefficients we must find[42].

Since $N_k(0) = N_k^0$, we require

$$\sum_{i=1}^{k} a_i^k = 0. \tag{A.3}$$

In addition, for the first isotope in the chain ($\sigma_{k-1}^k = 0$) the solution of (A.1) is

$$N_1(t) = N_1^0 e^{-\sigma_1 t} + \int_0^t F_1 e^{\sigma_1 (y-t)} dy. \tag{A.4}$$

and therefore

$$a_1^1 = 0; \qquad b_1^1 = F_1. \tag{A.5}$$

Now we can express $\frac{dN_k}{dt}$ in two different ways: first, by taking the derivative directly in (A.2); second, by substituting in the right hand side of (A.1) $N_k(t)$ from (A.2). By equating these two expressions we'll obtain

$$-\sigma_k N_k^0 e^{-\sigma_k t} - \sum_{i=1}^{k} \sigma_i a_i^k e^{-\sigma_i t} + \sum_{i=1}^{k} b_i^k - \sum_{i=1}^{k} \sigma_i \int_0^t b_i^k e^{\sigma_i (y-t)} dy =$$

$$F_k(t) - \sigma_k (N_k^0 e^{-\sigma_k t} + \sum_{i=1}^{k} a_i^k e^{-\sigma_i t} + \sum_{i=1}^{k} \int_0^t b_i^k e^{\sigma_i (y-t)} dy) \tag{A.6}$$

[41] The easiest way to arrive at this form of the solution is to solve the equations (A.1) for k=1,2,3... explicitly. After that the choice (A.2) becomes obvious.

[42] The a_i^k coefficients are time independent, whereas the $b_i^k(t)$ coefficients are time dependent.

$$+\sigma_{k-1}^k(N_{k-1}^0 e^{-\sigma_{k-1}t} + \sum_{i=1}^{k-1} a_i^{k-1} e^{-\sigma_i t} + \sum_{i=1}^{k-1} \int_0^t b_i^{k-1} e^{\sigma_i(y-t)} dy).$$

This expression gives us the two desired equations for finding a_i^k and b_i^k. For the a_i^k coefficients we obtain:

$$-\sum_{i=1}^{k} \sigma_k a_i^k e^{-\sigma_i t} = -\sigma_k \sum_{i=1}^{k} a_i^k e^{-\sigma_i t} + \sigma_{k-1}^k (N_{k-1}^0 e^{-\sigma_{k-1}t} + \sum_{i=1}^{k-1} a_i^{k-1} e^{-\sigma_i t}), \tag{A.7}$$

or in a simpler form

$$\sum_{i=1}^{k} (\sigma_k - \sigma_i) a_i^k e^{-\sigma_i t} = \sigma_{k-1}^k (N_{k-1}^0 e^{-\sigma_{k-1}t} + \sum_{i=1}^{k-1} a_i^{k-1} e^{-\sigma_i t}). \tag{A.8}$$

Since the last relation must hold for any time t, the corresponding coefficients must be equal:

$$(\sigma_k - \sigma_{k-1}) a_{k-1}^k = \sigma_{k-1}^k (N_{k-1}^0 + a_{k-1}^{k-1})$$

$$(\sigma_k - \sigma_j) a_j^k = \sigma_{k-1}^k a_j^{k-1}.$$

Thus we obtain the result for a_i^k:

$$a_j^k = \frac{\sigma_{k-1}^k}{\sigma_k - \sigma_j} a_j^{k-1}; \qquad j = [1, k-2]$$

$$a_{k-1}^k = \frac{\sigma_{k-1}^k}{\sigma_k - \sigma_{k-1}} (a_{k-1}^{k-1} + N_{k-1}^0). \tag{A.9}$$

Following the same procedure for b_i^k we'll get

$$\sum_{i=1}^{k} (\sigma_k - \sigma_i) \int_0^t b_i^k e^{\sigma_i(y-t)} = \sigma_{k-1}^k \int_0^t b_i^{k-1} e^{\sigma_i(y-t)}$$

$$\sum_{i=1}^{k} b_i^k = F_k. \tag{A.10}$$

This gives us

$$b_j^k = \frac{\sigma_{k-1}^k}{\sigma_k - \sigma_j} b_j^{k-1}; \qquad j = [1, k-1]$$

$$b_k^k = F_k - \sum_{i=1}^{k-1} b_i^k. \tag{A.11}$$

Together with (A.3) and (A.5), formulas (A.9) and (A.11) give us the coefficients a_i^k and b_i^k in (A.2).

Thus, summarizing the results, solution of the equation (A.1) can be written in form

$$N_k(t) = N_k^0 e^{-\sigma_k t} + \sum_{i=1}^{k} a_i^k e^{-\sigma_i t} + \sum_{i=1}^{k} \int_0^t b_i^k e^{\sigma_i (y-t)} dy, \tag{A.12}$$

where the coefficients a_i^k and b_i^k obey the following relations:

$$a_1^1 = 0$$

$$\sum_{i=1}^{k} a_i^k = 0$$

$$a_{k-1}^k = \frac{\sigma_{k-1}^k}{\sigma_k - \sigma_j}(a_{k-1}^{k-1} + N_{k-1}^0) \tag{A.13}$$

$$a_j^k = \frac{\sigma_{k-1}^k}{\sigma_k - \sigma_{k-1}} a_j^{k-1} \qquad j = [1, k-2]$$

and

$$b_1^1 = F_1$$

$$b_j^k = \frac{\sigma_{k-1}^k}{\sigma_k - \sigma_j} b_j^{k-1} \qquad j = [1, k-1] \tag{A.14}$$

$$b_k^k = F_k - \sum_{i=1}^{k-1} b_i^k.$$

The corresponding activity of each isotope $A_k(t)$ is calculated as

$$A_k(t) = \lambda_k N_k(t). \tag{A.15}$$

These formulas allow for time dependence of F. Assume that F is equal zero – than so are b_i^k in (A.12). Assuming in addition only $N_1^0 \neq 0$, the solution can be written in the form [Conn 78]:

$$N_k(t) = N_1^0 \prod_{j=2}^{k} \sigma_{j-1}^j \sum_{i=1}^{k} \frac{e^{-\sigma_i t}}{\prod_{l=1}^{k}(\sigma_l - \sigma_i)}. \tag{A.16}$$

This formula can be used for actinide activity calculations, since only for the first isotope in chain – U^{238}, $N^0 \neq 0$ and actinides are not produced in fission, i.e. $F \equiv 0$.

Glossary

Absorber - special rod made of neutron absorbing material (e.g. cadmium) and inserted into core in place of fuel assemblies. These absorbers are used for controlling the fission rate in the reactor.

Actinides - isotopes with atomic numbers Z=89-103.

Assembly-days - number of fuel assemblies integrated over time each assembly was in the core.

Campaign time - time during which the reactor has been under operation (producing energy).

Capacity factor - ratio of total energy produced in a reactor to the maximum energy this reactor is capable of producing. It is most often applied to one year time interval.

Channel type reactor - a reactor whose core consists of a large number of channels that can contain **fuel assemblies, absorbers,** and other special equipment.

Enrichment - a chemical process that increases the content of ^{235}U from 0.7% (natural uranium) to some 2%-4% used in power reactors.

Fission products - isotopes with mass numbers A=72-166. These isotopes are produced in the reactor through fission reaction in **actinides** present in the reactor.

Fuel assembly - unit containing reactor fuel. It is so constructed that it can be loaded into or removed from the reactor as one piece.

Fuel burnup (or fuel exposure) - cumulative energy produced by the fuel. Usually expressed in **MWd** or **GWd** per kilogram of fuel or per fuel assembly.

GWd (gigawatt-day) = $8.64*10^{13}$ J - unit of energy used in nuclear reactor industry (see also **MWd**).

Moderator - part of a reactor that consists of special material (e.g. heavy water, beryllium, graphite) with very low absorption cross section and the ability to slow down neutrons to thermal energy - 0.025eV.

MCi - megacurie, unit that is used for measuring the activity of the radioactive isotopes produced in the reactor.

MWd (megawatt-day) = $8.64*10^{10}$ J - unit of energy used in nuclear reactor industry (see also **GWd**).

MWt, MWe - thermal (**MWt**) and electric (**MWe**) power of a reactor. RBMK reactor under normal conditions is working at 3200 MWt or 1000 MWe power level.

Neutron fluence - thermal neutron flux in a reactor integrated over the **campaign time.**

Noble gases - volatile radioactive isotopes of Kr and Xe (see Table 10).

Point-kinetic approach - a calculational model in which the reactor is considered as one point, i.e. the spatial distribution of reactor parameters (fuel and coolant temperature, neutron flux, void fraction etc.) is not taken into account and averages over the whole reactor are used instead. This approach is often used in time-dependent calculations of the reactor parameters.

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