THE IGNALINA NUCLEAR POWER PLANT AND THE ENVIRONMENT Ignalinas atomelektrostacija un vide

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Abstract

Lithuania has to solve a lot of problems when closing Ignalina nuclear power plant (INPP). One of them is predicting, monitoring and managing of radiation situation and its impact to environment during this period. Calculated data files for nuclides in nuclear fuel could be used for this aim.

This work presents our calculations of concentrations and activities of actinides and fission products in RBMK-1500 reactor fuel in dependence on fuel burn up and cooling time.

Calculations were made using French computer code Apollo1 [1].

Keywords: Nuclear Power Plant, environment, radiation protection

Introduction

All the combustion of fossil fuel (coal, oil, natural gas) results in emissions harmful to the environment and to health.

The greatest environmental problem is emission of lot of carbon dioxide, dinitrogen oxide and methane into the atmosphere. These gases add to what is known as the greenhouse effect, they slow down the radiation of heat from the earth and raise the average temperature.

Under the Kyoto Protocol, the EU countries have agreed to reduce CO_2 emissions to 8 % below 1990 levels in the period from 2008 to 2012. The protocol calls for the promotion of CO_2 avoidance technology and of advanced, innovative and environmentally sound technologies. There is some agreement today concerning the fact that nuclear power offers a valuable contribution to the fight against global environment problems.

The environmental effects from well operating nuclear power plants are minor, and mainly connected to warm water releases. Nuclear power plants do not produce flue gases, carbon dioxide or nitric and sulphur oxides. Releases of radioactive substances are very small, closely measured and controlled.

Nuclear energy is one of technologies, which avoid each year the release of some 1,8 billion tones of CO_2 worldwide. In Europe alone, climate-friendly nuclear electricity saves the emission of about 800 million tonnes of CO_2 a year.

At the end of year 2000, 438 nuclear power plant units were in use in 30 countries worldwide, 31 new units were under construction and about the same number were under design. 16 % of the overall demands for electricity worldwide, 35 % the European Union, is supplied by nuclear power. The highest nuclear power contribution of the electricity generated is in Lithuania, France, Belgium, the Ukraine, Sweden and Bulgaria.

The Ignalina NPP produces about 85 % of electricity used in Lithuania. It supplies with some amount of electricity Latvia, Estonia, Belarus and Kaliningrad region.

The Ignalina nuclear power plant contains two RBMK-1500 water-cooled graphitemoderated channel-type power reactors. The first Unit of INPP went into service at the end of 1983, the second Unit - in August 1987. Their design lifetime is projected out to 2010 - 2015.

No industry is completely harmless to the environment. Ignalina NPP has implemented a lot of measures to improve the radiation safety of operating plant:

<u>The barriers of radioactive emissions.</u> Radioactive emissions from a nuclear power plant into surrounding area are prevented by means of a number of barriers.

The nuclear fuel has a form of ceramic pellets that retain most of the radioactive substances generated during the nuclear fission.

The fuel pellets are packed into gas-tight tubes made of a zirconium. The fuel element remains undamaged at high temperatures.

Piping of the reactor main coolant circuit

Reactor biological shielding

Particular containment system

The resulting solid and liquid activation products initially remain in the coolant. They are continuously removed from coolant by the reactor cleanup system.

The nuclear power plants with the RBMK-1500 reactor use a closed-circuit water supply system. Liquid radioactive effluents undergo special treatment. The liquid wastes are processed by filtration to remove suspended solids, by ion exchange in mixed-bed demineralises or by evaporation. To reduce the emissions of radioactive gases from the plant a two-stage purification process is used. The first stage consists of a holding chamber. There activity of the gases reduces by natural radioactive decay. The second stage is based on separation of radioactive substances by filtering and absorbing to an as low as possible level. The gaseous effluents are released in to the atmosphere via the 150 m height ventilation stack.

<u>Radwaste systems</u>. A nuclear power plant produces low- and medium- level radioactive waste. They are packed into drums. Wet waste is solidified prior to packing. The spent nuclear fuel represents about 95 % of all activity sources in the nuclear power plant and it is defined as high-level radioactive waste. Solid wastes are not released into environment. They are fragmented, compacted and prepared for storage.

Ignalina NPP has decided for long-term dry storage of spent nuclear fuel in CASTOR and CONSTOR type casks. One cask obtains 102 fuel assemblies.

<u>Radiation monitoring system</u>. Radiation Monitoring system includes a system for monitoring the state of protective barriers, a process radiation monitoring system, a radiation dosimetry monitoring system, an environmental monitoring system, a system for monitoring the spread of radioactive contamination. The releases into the atmosphere are continuously monitored into order to prevent endangering public health and the environment. The monitoring is carried out in compliance with international standards, in particularly aerosols, iodine isotopes, noble gases, tritium and carbon 14 are monitored.

The Radiation Monitoring System at Ignalina NPP is designed for continuous monitoring of the main radiation parameters, which characterize plant performance. It reflected actual radiation levels in the main plant areas, on site and off site under all operating conditions.

However under EU political pressure well-operating and safe INPP must be closed: Unit 1 – until 2004 and Unit 2 – until 2010.

The same type younger generation reactors RBMK-1000 at present time are operated in Russia: Sosnovy Bor (4 Units), Smolensk (3 Units) and Kursk (5 Units).

Calculations of concentration and activities of nuclides in nuclear fuel

The operation of a nuclear reactor creates a large amount of radioactive material, produced through the fission process, accumulation process and neutron activation process.

<u>The actinides</u>. The RBMK fuel is low enrichment uranium oxide UO₂. Initial enrichment was 2 %; since 1997 reload fuel is (2,4%-2,6%) enriched with ²³⁵U and contains burnable poison erbium. The uranium isotope ²³⁵U is fissile and is only the source of thermal energy at the first stages of reactor operation. ²³⁸U is a fertile isotope.

The Fuel content changes during operation of reactor. The new nuclei occur, which were not observed in initial fuel: ²³⁹Np, ²³⁹Pu, ²⁴¹Am and ²⁴⁴Cm. The actinides are formed

through successive neutron capture. They emit alpha particles and low energy gamma radiation.

Formation diagram of actinides is presented in Fig. 1. Uranium chain includes new created fissile nuclides during neutron capture and α and β decay.

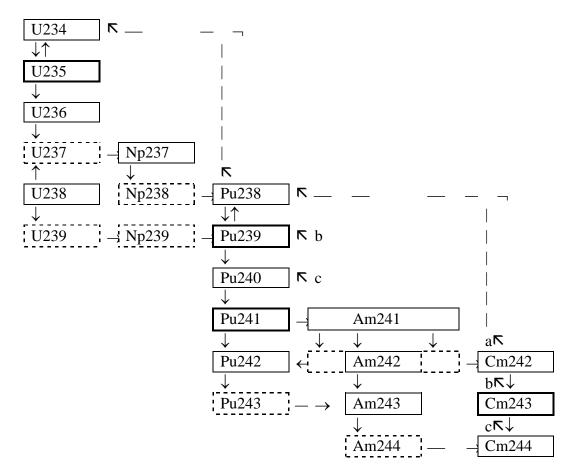


Fig. 1. Actinides formation diagram

Fissile nuclei are presented in bold-lined boxes, short-lived nuclei – in dash-lined boxes. Nuclei, which do not split after interaction with slow neutrons, are drawn normal.

Long-lived actinides can contribute to long- term environmental effects, associated with the final disposal of spent nuclear fuel.

Concentrations of nuclides produced in nuclear fuel depend on neutron flux $\Phi(\vec{r}, E, t)$, initial concentration and on radiation time (fuel burn up). Time dependent changes in concentration of j+1 isotope could be calculated using equation

$$\frac{\delta}{\delta t} n^{j+1}(\vec{r},t) = n^{j}(\vec{r},t) \int_{0}^{\infty} \sigma_{\gamma}^{j}(E) \Phi(\vec{r},E,t) dE - - n^{j+1}(\vec{r},t) \left[\int_{0}^{\infty} \sigma_{a}^{j+1}(E) \Phi(\vec{r},E,t) dE + \lambda^{j+1} \right],$$
(1)

where $\sigma_{\gamma}^{j}(E)$ is reaction (n, γ) cross-section; $\sigma_{a}^{j+1}(E)$ - neutron absorption cross-section; λ^{j+1} decay constant of j+1 isotope.

Results of actinide accumulation in nuclear fuel for the different burn up calculated by means of French computer code Apollo1 are presented in Table1. The fuel is 2,4 % enrichment of 235 U with burnable absorber erbium. The sequence of nuclides is set in

accordance with their evolution chain. ²³⁸Pu and ²⁴⁰Pu isotopes are presented twice - they can occur during (n,γ) reaction or as a result of α decay of Cm isotope. Natural decay of actinides becomes very important after removing of nuclear fuel from reactor.

Table 1.

Actinides concentration n for different fuel burn up, MWd/kg

Nuclides			n,	$10^{24}/cm^3$			
	0	2	4	8	12	16	20
U-235	5,560-04	5,021-04	4,539-04	3,704-04	3,001-04	2,401-04	1,891-04
U-238	2.262-02	2,261-02	2,256-02	2,251-02	2,243-02	2,236-02	2,229-02
Np-237	0	3,435-07	7,631-07	1,783-06	2,981-06	4,285-06	5,636-06
Pu-236	0	1,018-13	4,380-13	1,679-10	3,711-12	6,513-12	1,001-11
Pu-238	0	9,197-09	3,897-08	1,719-07	4,173-07	7,861-07	1,281-06
Pu-239	0	2,459-05	4,219-05	6,456-05	7,655-05	8,247-05	8,471-05
Pu-240	0	1,551-06	4,939-06	1,362-05	2.266-05	3,116-05	3,875-05
Pu-241	0	1,668-07	9,927-07	4,566-06	9,384-06	1,421-05	1,839-05
Am 241	0	5,468-10	6,612-09	6,172-08	1,882-07	3,677-07	5,653-07
Pu-242	0	4,062-09	5,012-08	5,043-07	1,707-06	3,781-06	6,701-06
Am-242	0	1,941-12	2,903-11	5,420-10	1,951-09	4,136-09	6,637-09
Cm 242	0	1,744-11	4,042-10	7,098-09	3,105-08	7,830-08	1,473-07
Pu-238	0	1,286-12	6,177-11	2,309-09	1,595-08	5,595-08	1,361-07
Am-243	0	3,576-11	9,254-10	2,014-08	1,083-07	3,333-07	7,607-07
Cm-243	0	5,845-14	2,739-12	9,893-11	6,676-10	2,298-09	5,501-09
Cm-244	0	4,693-13	2,527-11	1,182-09	1,016-08	4,415-08	1,328-07
Pu-240	0	6,900-16	7,304-14	6,645-12	8,367-11	4,732-10	1,731-09

The largest variations of actinide concentration during long time cooling period could be observed for ²⁴¹Pu, ²⁴²Cm, ²⁴³Cm and ²⁴⁴Cm isotopes. Calculated variations of actinides during cooling time period of 500 years are presented in Table 2.

Table 2.

Nuclides	n ²						
1 (defides	0 a	40 a	80 a	120 a	160 a	200 a	500 a
Pu-241	1	1,46-01	2,23-02	6,20+01	6,22+01	7,94-05	6,04-11
Am-241	1	5,33+01	6,08+01	6,20+01	6,22+01	6,22+01	1,20+02
Cm-242	1	9,76-02	4,50-02	2,07-02	9,56-03	4,41-03	1.33-05
Pu-238	1	3,11+00	3,24+00	3,29+00	3,32+00	3,33+00	6,07+00
Cm-243	1	3,64-01	1,36-01	5,05-02	1,8802	7,02-03	4,28-06
Pu-239	1	1,65+00	1,88+00	1,97+00	2,00+00	2,01+00	3,17+00
Cm-244	1	2,04-01	4,32-02	9,16-03	1,94-03	4,11-04	3,64-09
Pu-240	1	8,20+01	9,84+01	1,02+02	1,03+02	1,03+02	2,21+02

Relative concentration n' changes of actinides for different cooling periods

Calculations were performed using programme CEFROI for fuel burn up 20 GWd/t.

<u>Fission products</u>. During fission the nucleus split into two separate nuclei. Fission fragments have redundant number of neutrons and are β unstable. Nuclides are presented in genetic decay chains. Build up and accumulation of them depends on the irradiation and cooling time.

Variations of concentration of i-th fission product in decay chain during irradiation period could be described with radiation equation:

$$\frac{dN_i}{dt} = -(\lambda_i + \sigma_i)N_i + \sum_i f_{j \to i}(\lambda_i N_i) + \sum_k g_{k \to i}(\sigma_k \phi_k N_k) + y_i F, \qquad (2)$$

where N_i - is the number of nuclei of i-th product at the time t; F – fission rate; y – fission yield of nuclide i; $f_{j\rightarrow i}$ – build up of nuclide i by the unit density of nuclide j; $g_{k\rightarrow i}$ – build up of nuclide i by the unit neutron reaction of nuclide k; σ_k – average microscopic neutron reaction cross section of nuclide k; ϕ - neutron flux, λ_k – decay constant.

During cooling time the mother nuclei are split and daughter products in the mixture with mother nuclei is equal:

$$N_{2}(t) = N_{1} \frac{\lambda_{1}}{\lambda_{2} - \lambda_{1}} (e^{-\lambda_{1} t} - e^{-\lambda_{2} t}), \qquad (3)$$

where N₂(t) – is the number of daughter nuclei at the time t; N₁ – the number of mother nuclei at the time t = 0; λ_1 and λ_2 – are the decay constants for mother and daughter nuclei respectively. Transformations in the chain with n members follow the scheme: $1 \xrightarrow{\lambda_1} 2 \xrightarrow{\lambda_2} 3 \xrightarrow{\lambda_3} \dots (n-2) \xrightarrow{\lambda_{n-2}} (n-1) \xrightarrow{\lambda_{n-1}} n \xrightarrow{\lambda_n}$

Time dependent variations of concentration and activity for different nuclides could be calculated using equation of Bateman:

$$A_{n} = \lambda_{1}\lambda_{2}...\lambda_{n}N_{1}(0) \begin{bmatrix} \frac{e^{-\lambda_{1}t}}{(\lambda_{2} - \lambda_{1})(\lambda_{3} - \lambda_{1})...(\lambda_{n} - \lambda_{1})} + \\ + \frac{e^{-\lambda_{2}t}}{(\lambda_{1} - \lambda_{2})(\lambda_{3} - \lambda_{2})...(\lambda_{n} - \lambda_{2})} + \\ + \frac{e^{-\lambda_{n}t}}{(\lambda_{1} - \lambda_{n})(\lambda_{2} - \lambda_{n})...(\lambda_{n-1} - \lambda_{n})} \end{bmatrix}$$

$$(4)$$

Using these equations and renewed data files of nuclides new programme were created. We have used PEPIN code [2] for the evaluation of time dependent variations of activity of nuclides. Significant number of short-lived fission products decay during cooling time. This leads to decreasing of the number of nuclides with a high activity. Only a small number of decay chains contribute to the total activity of spent nuclear fuel.

The most important radionuclides, created in the course of nuclear power plant operation are noble gases (⁴¹Ar, ⁸⁵Kr and ¹³³Xe), iodine isotopes ¹³¹I and ¹³³I,⁹⁰Sr, ¹³⁴Cs and ¹³⁷Cs.

Table 3 shows activity changes of most environmentally important fission products during long-term storage of spent nuclear fuel. The fission products, which can be released into the environment, are of particularly interest. For a release to occur, the fuel cladding, the primary system boundary must be penetrated.

Table 3.

			01			
Nuclides		Activity,	Bq			
	5 a	10 a	20 a	30 a	40 a	50 a
Kr-85	1,591+11	1,131+11	6,031+10	3,159+10	1,655+10	8,669+09
Sr-90	1,474+12	1,308+12	1,031+12	8,128+11	6,406+11	5,049+11
Cs-134	3,434+11	6,394+10	2,218+09	7,691+07	2,481+07	8,281+04
Cs-137	2,026+12	1,806+12	1,436+12	1,141+12	9,068+11	7,206+11

Fission products activity for different cooling periods

Activation products. The nuclear fission probability depends on bombarding neutron energy. For moderation of neutrons in RBMK type reactor core the graphite is used.

The neutron radiation activates the graphite, the structure materials, the corrosion and erosion products of the primary circuit and coolant impurities and chemical additives. The activation can occur after neutron capture by nuclide $\frac{A_i}{Z}X$:

In many cases the new nuclide $\frac{A_i+1}{Z_i}X$ might be not stable (radioactive) and might emit β particles and γ rays. As a result of neutron absorption, the following processes take place: atoms of atomic mass number Ai are destroyed and atoms with atomic mass number Ai+1 are produced. They decay or can be destroyed by absorbing a neutron.

The production of the A_{i+1} is expressed by

$$N_{i+1}\left(t\right) = \frac{\sigma_{i} N_{i0} \Phi}{\lambda_{i+1} + \sigma_{i+1} \Phi - \rho_{i} \Phi} \left\{ \exp\left(-\sigma_{i} \Phi t\right) - \exp\left[-\left(\lambda_{i+1} + \sigma_{i+1} \Phi\right) t\right] \right\}, \quad (6)$$

 σ_i , σ_{i+1} - neutron absorption cross section of isotope A_i and A_{i+1} accordingly; λ_{i+1} - decay constant of isotope with atomic mass number A_{i+1} ; N_{i0} - number of atoms of nuclide with atomic mass number A_i at time t=0; Φ - neutron flux.

Activation level depends on the neutron flux, the irradiation time, the material composition, on cooling time. If half-life of radionuclide produced is long compared to the irradiation time, the activity increases linearly with time (¹³C, ³⁶Cl, ⁴¹Ca, ⁵⁹Ni, ⁹⁴Nb). For halflives many times shorter than irradiation time, the activity reaches a saturation value (⁴⁵Ca, ⁵⁴Mn, ⁵⁹Fe, ⁵⁸Co). The activation products are fixed in concrete, graphite and steel.

The most important activation products after shut down of reactor are ⁵⁵Fe, ⁶⁰Co, ⁵⁹Ni, ⁶³Ni, ³⁹Ar, ⁹⁴Nb in steel, ³H, ¹⁴C, ⁴¹Ca, ⁵⁵Fe, ⁶⁰Co, ¹⁵²Eu, ¹⁵⁴Eu in concrete and ³H, ¹⁴C, ¹⁵²Eu, ¹⁵⁴Eu in graphite.

In the time period from 10 years to 20 years most important radionuclides are ³H, ⁶⁰Co, ⁵⁵Fe and ¹³⁷Cs. In the time period from 20 years to 30 years – ⁶³Ni, ¹³⁷Cs, ⁶⁰Co and ⁹⁰Sr.

Corrosion products can be released into the reactor coolant in dissolved or suspended form and are activated when the coolant passes trough the reactor core. The steam generated in reactor contains activation products, particularly those originated from the water itself. The most important of these is nitrogen ¹⁶N. It has a short half-life and its activity decreases rapidly when the reactor is shut down. The environmental effects of ¹⁶N is negligible.

The part of activated coolant products (³H and ¹⁴C) releases to the environment and contributes to the global collective dose. Activated corrosion products (⁵¹Cr, ⁵⁵Fe, ⁵⁷Mn, ⁵⁹Ni, ⁶⁰Co, ⁶⁵Zn) are removed from the coolant and radioactive wastes within nuclear power plant are formed.

Conclusions

Calculation results describe the time-dependent behaviour of radionuclides in nuclear fuel and variations of environmentally important their characteristics.

Calculation results allow us to predict the radiation situation arround Ignalina NPP for the long time period. They could be used for limitation of public exposure in the environment.

References

APOLLO1. Notice d'utilisation, Rapport DMT/SERMA/LENR Code PEPIN., Rapport DMT/SERMA/LENR