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Activity release from damaged fuel during the Paks-2 cleaning tank incident in the spent fuel storage pool

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ABSTRACT

During crud removal operations the integrity of 30 fuel assemblies was lost at high temperature at the unit No. 2 of the Paks NPP. Part of the fission products was released from the damaged fuel into the coolant of the spent fuel storage pool. The gaseous fission products escaped through the chimney from the reactor hall. The volatile and non-volatile materials remained mainly in the coolant and were collected on the filters of water purification system. The activity release from damaged fuel rods during the Paks-2 cleaning tank incident was estimated on the basis of coolant activity concentration measurements and chimney activity data. The typical release rate of noble gases, iodine and caesium was 1–3%. The release of non-volatile fission products and actinides was also detected.

1. Introduction

The activity release from the damaged fuel rods is a key question in the analysis of the consequences of reactor accidents. Different models are used for the estimation of activity release during design basis and severe accidents. However very limited measured data are available on the release from fuel rods irradiated in nuclear power plants. Some very valuable results were produced in experimental series FLASH [1] and some of the currently used models are based on these tests [2]. New data on activity release from real damaged fuel under accident conditions are useful for the validation of existing models and for the better prediction of potential consequences of reactor accidents.

In April 2003, the incident, in which severe fuel damage occured in the spent fuel storage pool, took place at the Paks nuclear power plant [3,4]. Thirty VVER-440 type fuel assemblies were treated in a cleaning tank in order to remove crud depositions from cladding surfaces. After the completion of chemical procedures the cleaning tank was operated in cooling mode with the circulation of the water of spent fuel storage pool. The flow rate of the applied pump was not enough for the removal of decay heat, and, for significant part of the incident, flow bypassing the fuel assemblies. The coolant reached saturation in the cleaning tank and a steam volume was formed in the tank. The temperature in the tank did not reach the melting point of steel, but was high enough for the intense oxidation of zirconium components. The estimated maximum temperature was between 1200 and 1300 °C. After 7 h of dry conditions the cover of the tank was opened and the surrounding water of the pool quickly cooled down the hot assemblies. The quench resulted in severe damage of the oxidised fuel assemblies. Some activity release was detected 5 h before the opening of cover and high activities were measured during the opening of tank cover. The visual observation of fuel assemblies showed strong fragmentation of all fuel assemblies. The measured data indicated that the release of isotopes with short half-life stopped after the incident. However the isotopes with long half-life were released until the removal of damaged fuel. The gaseous fission products escaped through the chimney from the reactor hall. The volatile and non-volatile materials remained mainly in the coolant and were collected on the filters of water purification system. Using the activity measurements of Paks NPP the integral activity from fuel rods can be estimated.

2. Measured data

The activity concentrations in the coolant and the release through the chimney are regularly measured at the NPP. Such data were available for the period of the incident, too. The contamination of the intact fuel assemblies in spent fuel storage pool and the steel surfaces of the structures was a very important problem after the incident and especially before the start-up of the unit [5]. However, the total activity absorbed on the surfaces was much less, than the activity in the coolant. For these reasons the surface

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activity measurements were neglected in the estimation of total release from fuel.

2.1. Coolant activity measurement

Water samples were taken for laboratory analysis from the spent fuel storage pool, from the service pit and from the primary circuit. The activity concentrations of fission products, actinides and some other isotopes activated in the reactor core were determined by γ- and α-spectrometry of coolant samples. Measurements were performed from the residual heat removal system of the spent fuel storage pool and from the water purification systems operated in the primary circuit, in the spent fuel storage pool and in the service pit. There were separate measurements from the coolant before and after the ion exchange resins of purification system and such data made possible the estimation of purification efficiency for the different isotopes.

The service pit, the spent fuel storage pool and the refuelling pool with the primary circuit were not isolated from each other during the incident (Fig. 1). The measurements taken from different locations indicated that there was an intense mixing between these pools. The fastest activity increase was detected in the service pit where the damaged fuel was located. The activity concentrations in the primary coolant reached the level of service pit with a 2 day delay (Fig. 2).

The highest fission product concentrations were measured in the coolant for ^{131}I, ^{133}Xe and ^{140}Ba, which reached 10^8 Bq/l. The maximum concentration of ^{132}I, ^{134}Cs, ^{137}Cs, ^{140}La and ^{132}Te was above 10^7 Bq/l, while the concentration of ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{136}Cs, ^{141}Ce and ^{144}Ce was in the order of 10^6 Bq/l. The activity concentration peaks were detected after the opening of the cleaning tank, but high activities were measured after several days, too

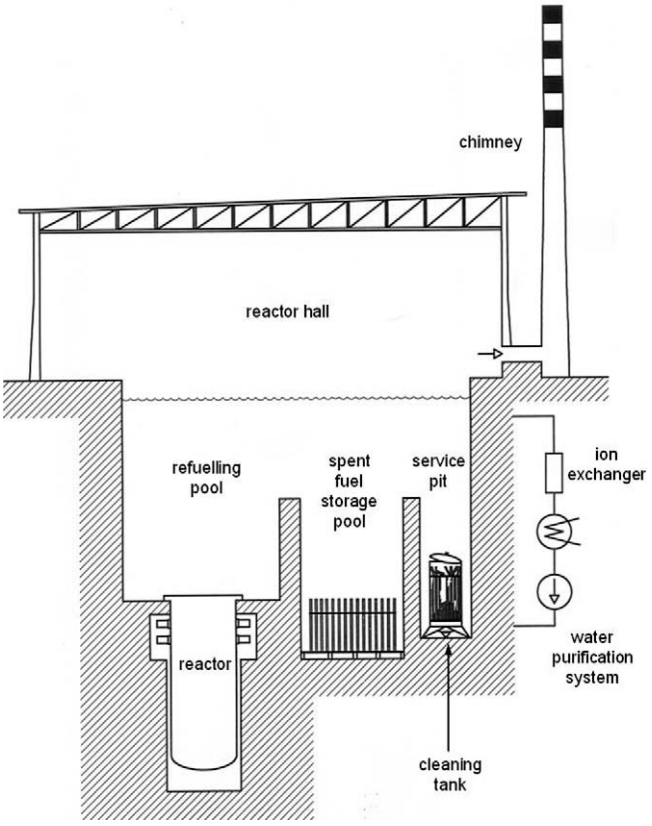

Fig. 1. Schematic view of the main technological systems during the Paks-2 incident in the spent fuel storage pool.

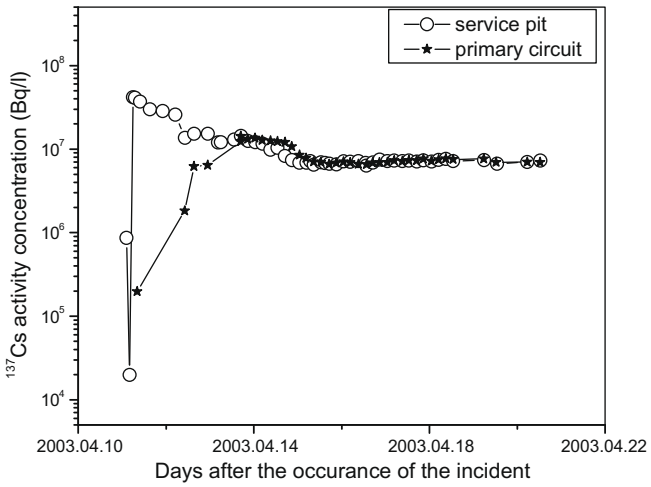

Fig. 2. Measured ^{137}Cs activity concentrations in the service pit and in the primary circuit.

(Fig. 3). The first 2-week period was characterised by the highest activity release from the fuel. After that the activity concentrations of fission products significantly decreased and the further activity from fuel was characterised by leaching process in wet storage conditions. The damaged fuel was stored in the service pit and in the cleaning tank for more than 3 years and the activity release from fuel was continuous. However the activity release was lower and for this reason only the first 2-week period was assigned to the incident and the following interval was considered as the period of wet storage of damaged fuel (Figs. 3 and 4).

Beyond fission products several actinides (^{239,240}Pu, ^{238}Pu, ^{241}Am, ^{244}Cm, ^{242}Cm) were also detected in the coolant immediately after the incident. Their concentrations were much lower; the typical value was ≈10 Bq/l (Fig. 4). Contrary to fission products the activity concentrations of actinides did not decrease after the first 2 weeks, but was characterised with the same level even in several months after the incident.

2.2. Release through the chimney

There was significant release of gaseous fission products (mainly noble gases) through the chimney after the incident

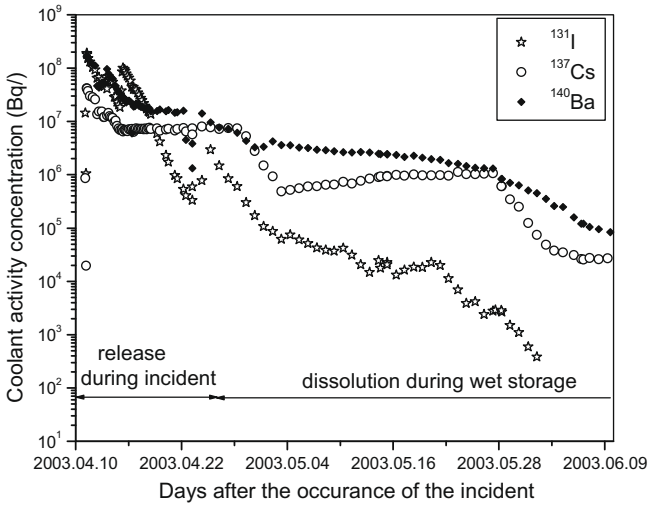

Fig. 3. Coolant activity concentrations of ^{137}Cs, ^{131}I and ^{140}Ba isotopes during the Paks-2 incident in the spent fuel storage pool.

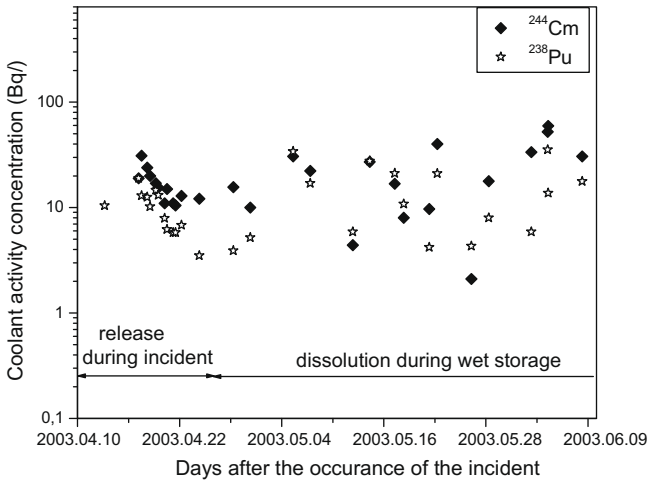

Fig. 4. Coolant activity concentrations of ^{244}Cm and ^{238}Pu isotopes during the Paks-2 incident in the spent fuel storage pool.

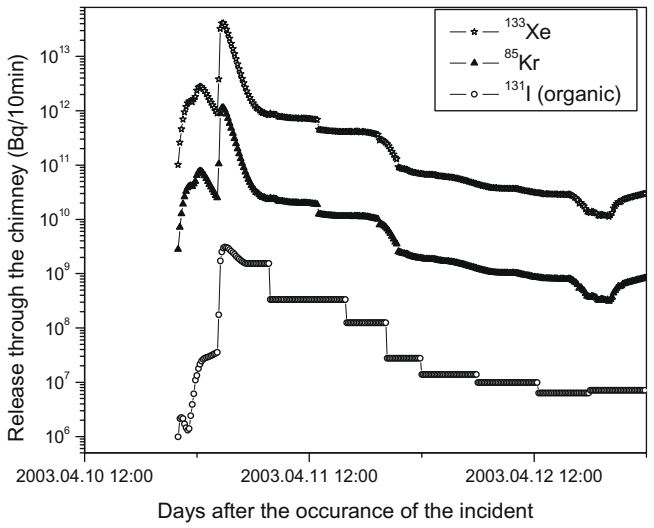

Fig. 5. Measured ^{133}Xe, ^{85}Kr and organic ^{131}I release through the chimney.

(Fig. 5). The gaseous fission products were originated from the damaged fuel. The release path of gaseous fission products started from the damaged fuel in the cleaning tank and continued in the water pool. Due to low solubility they did not remain in the coolant, but entered the atmosphere of the reactor hall. The continuous operation of ventilation system in the reactor hall removed the accumulated fission products through the chimney (Fig. 1).

The isotope specific data of noble gases were determined with a continuous monitoring system and these data were verified with daily ball sampling for laboratory analysis. The activity release values were recorded each ten minutes for several isotopes. The chemical form (elemental, organic or aerosol) of the released iodine was also determined for each period. The highest release was detected for ^{133}Xe isotope with 10^{13} Bq release per 10 min. The ^{85}Kr and ^{131}I activity releases reached 10^{12} Bq and 10^{9} Bq, respectively during the same sampling period. 99% of the total release took place in the first 24 h after the incident.

3. Calculation of isotope inventory

The isotope inventory of the 30 assemblies was calculated using the ORIGENARP module of the SCALE4.4a program system [6]. The calculations produced the concentrations and activities of isotopes for each assembly. The typical material composition was considered in the calculations with the following isotopes and elements: ^{234}U, ^{235}U, ^{236}U, ^{238}U, Hf, Nb, Sn and Zr. The calculations were done with 44 energy groups using gamma and neutron spectrum directories. The decay during the 13 days storage time between the reactor shutdown and the incident was calculated.

There were 11 normal and 19 follower type fuel assemblies in the cleaning tank. The followers are the lower sections of the borated steel control assemblies in VVER-440 reactors. The followers have the same structure as the normal assemblies, but they can be moved below the core during normal operation or shutdown periods.

The average burnup of the assemblies was 16 MWd/kgU. The burnup of individual assemblies varied between 11 and 27 MWd/kgU. Some assemblies were used in one cycle only, while some others spent two or three cycles in the reactor core.

The power histories for each fuel assembly were considered on the basis of operational data provided by the NPP. The fuel assemblies with similar power histories and of the same type were grouped into six groups and only one representative assembly was calculated for each group.

4. Evaluation of measured activity data

4.1. Coolant activity concentrations

The measured activity concentrations in the coolant provided the primary information for the calculation of activity release from damaged fuel. In the evaluation process the measured activity concentrations were considered as typical for the total volume of water pools connected to the cleaning tank with damaged fuel. The elapsed time after the incident was divided into short periods, which were characterised by stable technological conditions. It was supposed that the release rate was constant during these short periods. The number of periods was different for the different isotopes, normally one calculated period covered several days.

The dissolution rates (or release rates from the fuel) in Bq/h units were calculated for each period for the total mass of uranium-dioxide pellets in the damaged fuel (3.7 t UO_2). The operation of water purification system was considered in the calculation of dissolution rates using the flow rate and the efficiency values. The efficiency data were calculated using measured activity concentrations before and after the ion exchangers. The purification efficiency was calculated for each isotope and for each period.

Some examples of calculated dissolution rates are shown in Fig. 6. The scatter of received values is rather large. It indicated that the dissolution was not a stable process with constant dissolution rates, but it was changing due to different conditions. However the general trend shows the decrease of average dissolution rate in time.

The activity release from the damaged fuel into the coolant was determined as the result of integration over the period from 10th to 25th April 2003.

4.2. Gaseous release from the reactor hall

Since in the chimney the integrated values of activity release were measured for ten minute periods it was not necessary to determine release rates, but the total release could be calculated through the simple integration of the measured data over the elapsed time. Such integration was carried out for all measured isotopes for the first 3 days of the incident. The results showed that except the noble gases, the release through the chimney was for

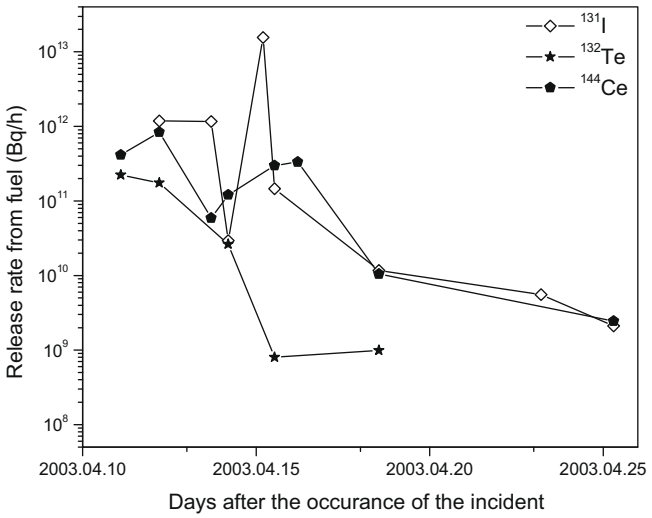

Fig. 6. Dissolution rate of ^{131}I, ^{132}Te and ^{144}Ce isotopes.

several orders lower than the release from the fuel into the coolant. Obviously the non-gaseous fission products were collected on the ion exchangers, while the gases were released through the open water surface into the reactor hall. For this reason the total release for noble gases was derived from the chimney data and for the other elements the coolant activity data were used.

4.3. Calculation of released fractions

The inventory of radioactive fission products continuously decreased after shutdown. Since the incident happened 2 weeks after reactor shutdown there were no isotopes with very short half-life in the fuel. However, some fission products with several days half-lives were released from the fuel. Their inventory significantly changed during the calculated 2-week period. In order to get such released activities that could be compared with the initial inventory, corrections were applied according to the decay constants of the given isotopes. These corrected values were significantly higher for short lived isotopes than the measured data (Fig. 7). Using the corrected values of total release and the inventories for the time to the incident the released fractions could be determined.

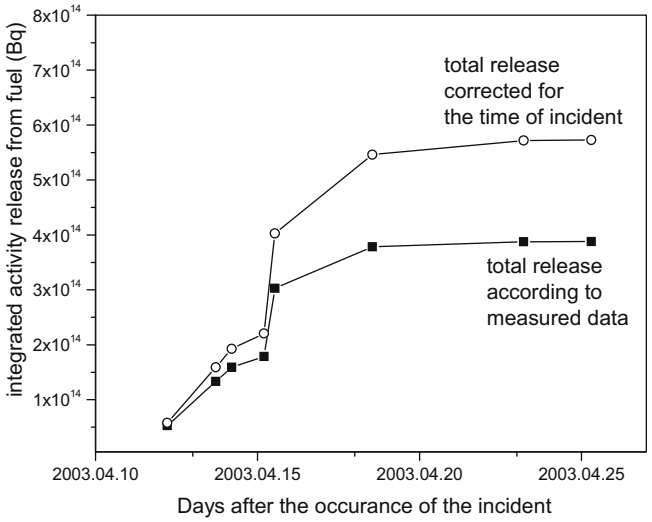

Fig. 7. Total activity release of ^{131}I isotope from fuel.

5. Results and discussion

5.1. Release rates

The highest release rates were received for the noble gases (xenon and krypton isotopes) with values between 1% and 3% (Table 1). About 1% of the volatile iodine and caesium isotopes was released. The less volatile tellurium, lanthanum and barium isotopes were released in the range of 0.08–0.26%. The lowest release with 0.02–0.06% was observed for non-volatile elements: zirconium, niobium, ruthenium and cerium. The Am, Cm and Pu release was about 0.00003–0.0001%.

It must be noted that the release rates of different isotopes of the same element were not the same. For example the release rate of ^{136}Cs was three times higher than that of ^{137}Cs. Such differences can be explained on one hand by the accuracy of measurements and data evaluation. On the other hand the release rates are not necessarily the same for the isotopes of the same element, e.g. if there were differences in the radial distribution of the given isotopes in the pellet.

5.2. Form of ^{131}I release

The measurements of the release through the chimney provided information on the fraction of elemental iodine, organic iodine and iodine in form of aerosols. The calculations showed that 77% of the total ^{131}I release through the chimney was in form of elemental iodine, 21% was organic and 2% in form of aerosols. These values were slightly changing during the measurements, but the average values were roughly the same for every day.

5.3. Comparison with design basis cases

The Paks-2 cleaning tank incident in the spent fuel storage pool can not be classified into the categories of light water reactor design basis accidents for several reasons. First of all the incident took place not in a reactor vessel, but in a special cleaning tank in the bottom of a deep water pool. The incident happened 2 weeks after

Table 1
Release rates of main isotopes during the Paks-2 incident in the spent fuel storage pool.

Isotope	Inventory (Bq)	Total release[a] (Bq)	Release rate (%)
^{85}Kr	$6.87 \cdot 10^{14}$	$2.06 \cdot 10^{13}$	3.00
^{131m}Xe	$1.10 \cdot 10^{15}$	$1.58 \cdot 10^{13}$	1.44
^{133}Xe	$5.56 \cdot 10^{16}$	$9.12 \cdot 10^{14}$	1.64
^{133m}Xe	$2.14 \cdot 10^{14}$	$3.71 \cdot 10^{12}$	1.73
^{95}Zr	$2.03 \cdot 10^{17}$	$4.98 \cdot 10^{13}$	0.02
^{95}Nb	$2.27 \cdot 10^{17}$	$3.50 \cdot 10^{13}$	0.02
^{131}I	$4.15 \cdot 10^{16}$	$5.89 \cdot 10^{14}$	1.42
^{132}I	$1.16 \cdot 10^{16}$	$8.40 \cdot 10^{13}$	0.72
^{106}Ru	$3.13 \cdot 10^{16}$	$8.69 \cdot 10^{12}$	0.03
^{103}Ru	$1.37 \cdot 10^{17}$	$6.32 \cdot 10^{11}$	0.0005
^{134}Cs	$5.69 \cdot 10^{15}$	$4.21 \cdot 10^{13}$	0.74
^{136}Cs	$1.55 \cdot 10^{15}$	$2.44 \cdot 10^{13}$	1.57
^{137}Cs	$7.22 \cdot 10^{15}$	$3.84 \cdot 10^{13}$	0.53
^{140}Ba	$1.16 \cdot 10^{17}$	$1.82 \cdot 10^{14}$	0.16
^{140}La	$1.33 \cdot 10^{17}$	$1.02 \cdot 10^{14}$	0.08
^{144}Ce	$1.20 \cdot 10^{17}$	$7.19 \cdot 10^{13}$	0.06
^{132}Te	$1.13 \cdot 10^{16}$	$2.94 \cdot 10^{13}$	0.26
^{141}Ce	$1.65 \cdot 10^{17}$	$6.39 \cdot 10^{13}$	0.04
^{239}Np	$5.04 \cdot 10^{16}$	$3.54 \cdot 10^{12}$	0.01
^{242}Cm	$9.25 \cdot 10^{14}$	$9.26 \cdot 10^{8}$	0.0001
^{244}Cm	$2.31 \cdot 10^{13}$	$3.57 \cdot 10^{7}$	0.0002
^{241}Am	$5.05 \cdot 10^{12}$	$1.00 \cdot 10^{7}$	0.0002
^{239,240}Pu	$6.73 \cdot 10^{13}$	$2.67 \cdot 10^{7}$	0.00004
^{238}Pu	$7.36 \cdot 10^{13}$	$2.34 \cdot 10^{7}$	0.00003

[a] Corrected for the time of incident.

reactor shut down, when the decay heat was very low compared to normal reactor operation. The total activity at the time of the incident was for two orders of magnitude lower, than at the time of shutdown and isotopes with short half-life were not present in the fuel. The heat-up phase lasted for several hours instead of a few minutes that is typical in the case of a loss-of-coolant accident.

However the Paks-2 cleaning tank incident in the spent fuel storage pool can be considered as an upper bound case for design basis accidents from the point of view of fuel damage. The temperature reached 1200–1300 °C, that is higher than the peak temperature estimated for a loss-of-coolant accident. The water quenching of heavily oxidised fuel assemblies led to severe fragmentation of the cladding and it made possible the release of radioactive isotopes from the gap and from the surface of fuel pellets.

It is expected that the fuel rods in the case of a loss-of-coolant (LOCA) type design basis accident will not suffer more severe damage than the fuel of the Paks-2 incident and the temperatures will not be higher than during the Paks-2 incident [7]. For these reasons it is very probable that the release rate of main isotopes from fuel would be lower during a LOCA accident than it was in the case of Paks-2 incident. However the absolute activity release from the fuel would be much larger in case of a design basis accident, because the inventory of radioactive isotopes is much larger at the moment of reactor shutdown.

6. Conclusions

The evaluation of measured activity data of the Paks-2 incident provided important information on the release of different radioactive isotopes from damaged fuel. The present work covered 2-week period after the incident. During this period 1–3% of the inventory of iodine, caesium and noble gases were released from the fuel. The release of less volatile and non-volatile fission products and actinides was also observed and the release rates were determined, the relative release rates varied between 0.00003% and 0.3%. The release into the environment was very low, due to the incident occurring in the bottom of a deep water pool. The conditions of the Paks-2 incident were more severe from the point of view of fuel damage than the conditions of LOCA type design basis accidents [1,2]. For this reason the release rates of radioactive isotopes determined in the present study for the Paks-2 incident can be used as an upper bound in the safety analysis of LOCA accidents for nuclear power plants.

Acknowledgement

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