Prediction of Iodine Activity Peak During Refuelling

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Introduction

The increase of fission product activities in the primary circuit of a nuclear power plant indicates the existence of defects in some fuel rods [1]. The regular measurements of different radioactive isotopes provides detailed information on the activities of the most important elements, including iodine as well. The presence of leaking fuel elements leads to the increase of the activity of iodine isotopes in the coolant during reactor shutdown or power transients in comparison with normal stationer operation [2].

The power change leads to the cooling down of the fuel and results in the fragmentation of the UO_2 pellets, which facilitates the release of fission products from the intergarnular regions. The depressurisation of the primary system creates a driving force for the transfer of isotopes from the fuel into the coolant [3]. If steam enters the fuel through the defect in the cladding its evaporation enhances the release [4]. Furthermore the injection of boric acid after shut down will increase the primary activity, due to the solution of deposited fission products from the surface of the core components.

The deterministic simulation of the mechanisms leading to iodine spiking includes such a level of uncertainties, that makes very difficult any realistic power plant application. That is why the calculation of this phenomena usually is based on the evaluation of activity measurements and power plant data. Expert systems are used in several nuclear power plants for the evaluation of activity measurements under normal operation and for the estimation of the number of leaking fuel elements, the failure size and the amount of tramp uranium in the primary circuit water [5][6][7][8]. The estimation of iodine spiking peak during reactor transients is based on correlation with operating parameters, such as reactor power and primary pressure [9][10][11].

The approach used in the present method was originally developed by the EPRI and later applied for CANDU reactors [6]. The VVER-440 specific correlations were determined using the activity measurements of the Paks NPP and the data provided by the Russian fuel supplier. The present method is used for the evaluation of the iodine isotopes, as well as the noble gases.

The method developed for the Paks NPP consists of two main steps:

1. Steady state calculation. Using the release-to-birth ratio R/B of at least three iodine isotopes the parameters of a diffusion equation and the number of leaking fuel elements are determined under normal operation. Simplex optimum calculation method is used for the numerical fitting of model parameters.
2. Transient calculation. The iodine spiking history is calculated considering the effects of power change ratio, increase rate of boric acid concentration and pressure decrease rate. Different coefficients were determined for each contributing effect and the correction takes into account the activity level before the transient.

Steady state model

The first step of the calculation is used for the determination of release-to-birth rate ratio R/B for the unit volume of the primary circuit water on the basis of the activity measurements.

$$\left(\frac{R}{B}\right)_{measured} = \left(\frac{\lambda+\beta}{\lambda}\right)\frac{C_m V_L}{FY}\exp(\lambda T_r) \qquad (1)$$

R – release rate (atom/s)
B=FY – birth rate (atom/s)
F – fission rate (fission/s)
Y – fission yield of the given isotope (atom/fission)
λ - decay constant, (1/s)
β - purification rate constant (1/s)
V_L –volume of the primary circuit (l)
T_r – transport time between the core and the place of sampling (s)

C_m - activity concentration of the given isotope at the time of sampling (Bq/l)

The number of fissions is calculated considering the amount of ^{235}U and ^{239}Pu in the fuel. According to plant specific ORIGEN calculations the average amount of ^{235}U varies between 21.5-14 g/kg, while the amount of ^{239}Pu between 3.5-5.5 g/kg for an equilibrium core and during a 300 day cycle.

The second basic equation of the model determines the R/B ratio supposing that the primary water activity has two sources: release from the leaking fuel element and from the tramp Uranium.

$$\frac{R}{B} = \left(\frac{\varepsilon}{\varepsilon + \lambda} \right) \frac{A}{\sqrt{\lambda}} H + c \qquad (2)$$

with $A = 3x\sqrt{D}$,

ε - escape rate from the fuel-to-cladding gap, (1/s)

D – diffusion coefficient inside of the fuel pellet,

x – number of leaking fuel elements,

H – precursive diffusion factor.

and $c = F_t / 2F$

F_t number of fissions in the tramp uranium

F number of fisisons in the leaking fuel rod

Table 1.
Atomic data of iodine and noble gas isotopes

Isotope	Decay constant λ, 1/s	Fission product yield Y, %	
		^{235}U	^{239}Pu
^{131}I	9.98e-7	2.88	3.85
^{132}I	8.37e-5	4.30	5.39
^{133}I	9.26e-6	6.70	6.93
^{134}I	2.20e-4	7.71	7.27
^{135}I	2.91e-5	6.30	6.45
^{85m}Kr	4.30e-5	1.30	0.566
^{87}Kr	1.52e-4	2.52	0.987
^{88}Kr	6.78e-5	3.55	1.32
^{133}Xe	1.53e-6	6.70	6.98
^{133m}Xe	3.66e-6	0.190	0.233
^{135}Xe	2.12e-5	6.54	7.60
^{138}Xe	8.18e-4	6.42	5.12
^{135m}Xe	7.40e-4	1.16	1.69

Equation (2) consists of three unknown variables: ε, *A* and *c*. Knowing the R/B data from equation (1) the unknown variables can be determied if the data of at least three isotopes of the same element are available. It can be supposed that release of different isotopes of the same element is governed by the same mechanism and is characterised by common parameters. According to the plant measurements 5 iodine and 8 noble gas isotopes are considered and any three of them is sufficient for the evaluation (Table 1.). The calculations of iodine and noble gas isotopes are carried out separately.

The system of equations on the basis of equation (2) and applied for several isotopes is non-linear and for this reason the standard fitting procedures can not be used. In order to determine the three unknown variables a three-dimensional simplex optimum calculation was applied. The optimum function is defined as the minimum of differencies between the calculated (2) and measured (1) R/B values.

The number of failed fuel rods can be estimated according to the following expression: $x = \frac{A}{3\sqrt{D}}$. The diffusion coefficients D is applied for nominal power conditions and its value of iodine and noble gases was determined on the basis of experimental data. The failure size is estimated using ε escape rate constant. The amount of tramp uranium is calculated if the constant *c* is known from equation (2).

An example of calculated results in presented in Table 2. and Fig. 1. and 2. The measurements were taken from unit 3 of the Paks NPP in July 2000. The activity data indicated the presence of leaking fuel rod(s).

The measured data were transformed into R/B values and the optimum calculation fitted the equation (2) type curves on the data (Fig. 1. for iodine and Fig. 2.for noble gases). The determined parameters made possible the evaluation of the number of leaking fuel rods, the amount of tramp uranium and the failure size.

The calculations were carried out for both iodine and noble gas datasets and the results showed good agreement between the two calculated cases.

Table 2.
Calculated results using NPP measurement data of Unit 3, 2000.07.28

Number of leaking fuel rods		Amount of tramp Uranium, kg		Failure size,
Iodine calcu-lation	Noble gas calcu-lation	Iodine calcu-lation	Noble gas calcu-lation	mm^2
1,4	1,0	$2,4\ 10^{-5}$	$1,5\ 10^{-5}$	$2,4\ 10^{-1}$

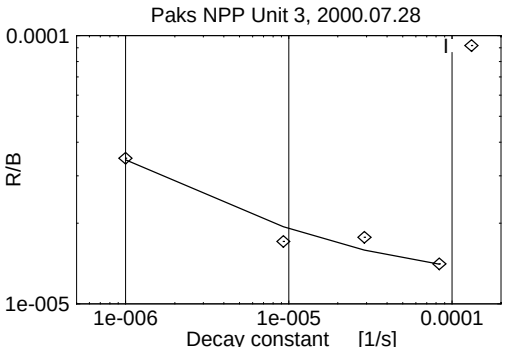

Fig. 1.
Measured (points) and fitted (lines) R/B ratio for iodine isotopes

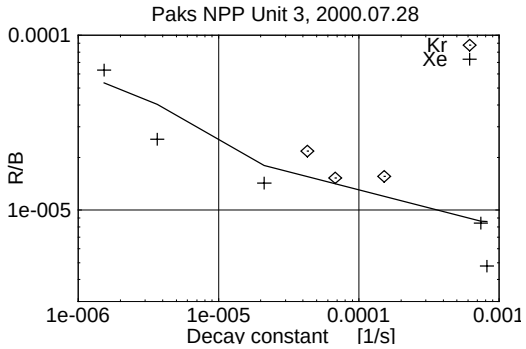

Fig. 2.
Measured (points) and fitted (lines) R/B ratio for noble gas isotopes

Spiking model

The activity concentration of the ^{131}I isotope is calculated according to (3), which was received from equations (1) and (2). Similar approach was used by EPRI [9].

$$C_m^{calculated} = \frac{FY}{V_L(\lambda+\beta)\exp(\lambda T_r)}\left(\frac{3S\varepsilon x\sqrt{SD}H\sqrt{\lambda}}{\lambda+S\varepsilon}+c\lambda\right) \quad (3)$$

It is supposed that the escape of the fission products from the fuel and their diffusion in the pellet is accelerated during the spiking phenomena due to the change of operating conditions. The effect of power (Q), pressure (P) and boric acid (c_{bor}) concentrations were taken into account in the following expression for S_s spiking factor:

$$S_s = \left(1 + a_1\frac{\Delta Q}{Q^{\max}} + a_2\left(\frac{\Delta P}{P^{\max}}\right)^n + a_3\frac{\Delta c_{bor}}{c_{bor}^{\max}}\right)e^{-T/a_4} \quad (4)$$

The coefficients a_1, a_2, a_3, a_4 and exponent n were determined according to four datasets, which were recorded during the shutdown of reactors with leaking fuel elements. The power decrease (reactor scram), boric acid concentration increase and depressurisation takes place one after another, so their effect on activity increase could be observed separately. The strongest contribution to ^{131}I activity increase was seen after the pressure decreasing actions. The exponential multiplier counts for the delay of activity release after the considered actions of shutdown procedure.

The spiking model uses the variables ε, x and c, which must be determined for steady conditions before the spiking event. Equation (3) calculates equilibrium concentration in a quasi-stationer manner, and for this reason its applicability is limited to slow transients. The typical time step of the calculation is 60 min.

Power plant spiking calculations

For the development and validation of the spiking model the measurements provided by the Paks NPP were used. The data collection covered the first days of refuelling period after reactor shutdown. Activity measurements were taken with high frequency in order to have sufficient number of experimental points for the modelling. The following data were collected and analysed:

- primary circuit pressure,
- boric acid concentration,
- core power,
- core inlet temperatures,
- activity concentrations.

Four datasets were available for evaluation purposes from unit 2 and 3 at different cycles. The characteristic ^{131}I activity concentrations for the four cases are listed in Table 3. The detailed comparison of the datasets made possible to compare the effects of different phenomena on the iodine spiking. Fig. 3-5 illustrate the change of boric acid concentration, primary pressure and core power during the first three days of the refuelling period.The data were systematised so that the zero time corresponded to the power decrease (scram). In all cases the power decrease took place in very similar manner. The increase of boric acid concentration started also approximately at the same time after reactor scram. The pressure decrease to 20 bar level was delayed for several hours in *case 2* compared to the other three cases.

The iodine activity concentrations correlated with the technological processes: activity increase was observed after reactor scram, boric acid concentration increase and pressure reduction. In *case 4* a second peak was also noted, which correlated with the second peak of pressure reduction from 20 bars to atmospheric value. The maximum value of ^{131}I activity concentration peak during spiking was the same order of magnitude for all cases. Two interesting phenomena could be mentioned:

- the activity peak in *case 2* was delayed in comparison with the other cases. It happened obviously due to the delayed pressure decrease.
- the initial ^{131}I activity concentrations were the same order of magnitude in the for cases, but the activity increase after boric acid injection was quite different. The cause of this effect probably is related to the different levels of primary circuit contamination.

Table 3
^{131}I spiking datasets from the Paks NPP

Case	Unit	Cycle	^{131}I activity concent-ration before shutdown [Bq/l]	^{131}I activity concent-ration peak during spiking [Bq/l]	Ratio of ^{131}I activity concent-ration increase
1	2	14th	3753	513370	136,8
2	3	12th	4400	330250	75,1
3	3	13th	2942	269840	91,7
4	3	14th	11120	890900	80,1

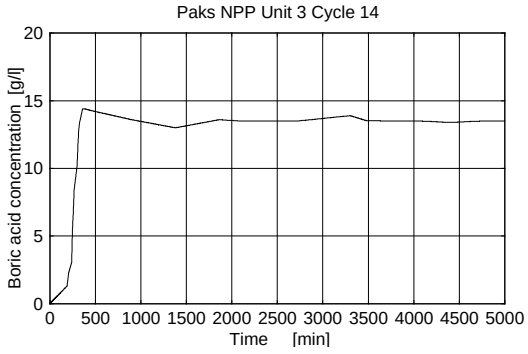

Fig. 3.
Boric acid concentration in *case 4*

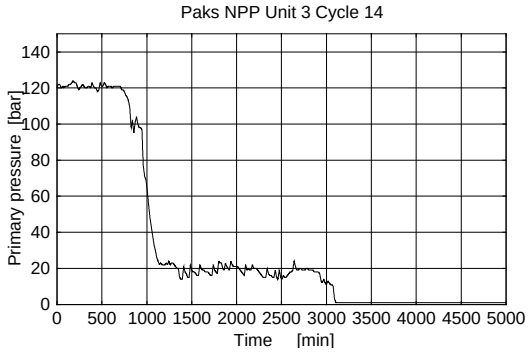

Fig. 4.
Primary pressure in *case 4*

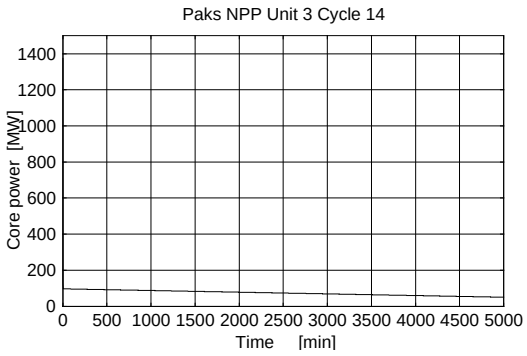

Fig. 5.
Core power history in *case 4*

The spiking model was used for the simulation of shutdown transients and good agreement was reached concerning the timing and the peak value of ^{131}I activity concentration. The measured and calulated iodine activities are presented in Fig. 6-9. The effect of boric acid proved to be the most difficult task to simulate: in *case 2* the model gave high overprediction and in *case 1* underprediction for the ^{131}I activity concentration. It seemed that there was no direct correlation between the amount of tramp uranium and the activity increase due to boric acid injection.

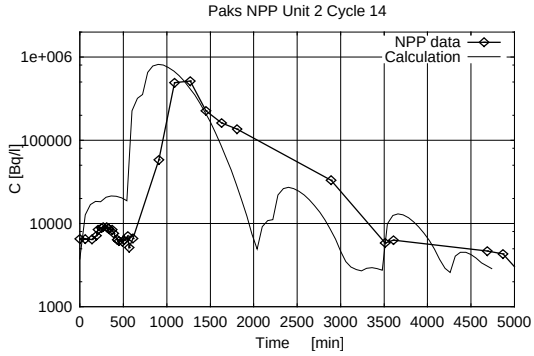

Fig. 6.
Measured and calculated ^{131}I activity concentration during shutdown period, *case 1*

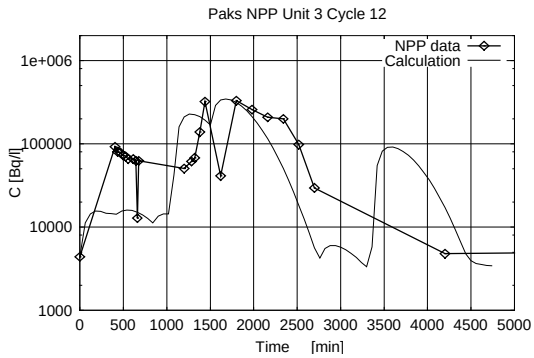

Fig. 7.
Measured and calculated ^{131}I activity concentration during shutdown period, *case 2*

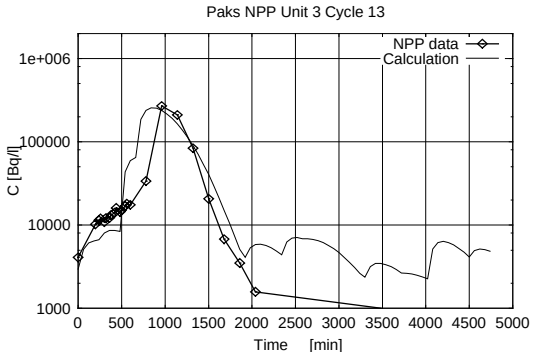

Fig. 8.
Measured and calculated ^{131}I activity concentration during shutdown period, *case 3*

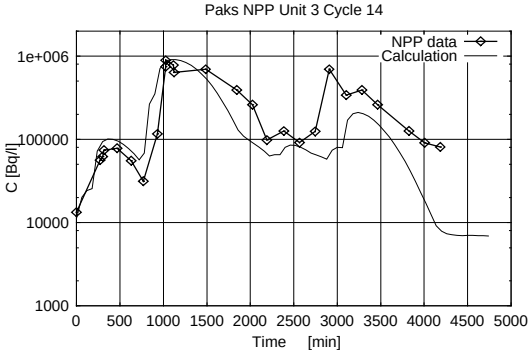

Fig. 9.
Measured and calculated ^{131}I activity concentration during shutdown period, *case 4*

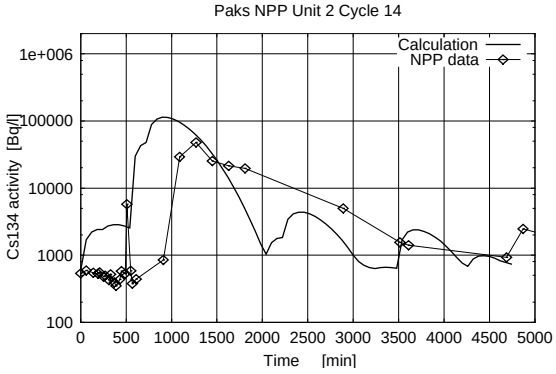

Fig. 10.
Measured and calculated ^{134}Cs activity concentration during shutdown period, *case 1*

The shutdown datasets included not only the iodine activity data but other isotopes as well. The ^{134}Cs and ^{137}Cs isotopes are important due to their biological effects. The increase of the cesium isotope activity concentrations was observed parallel to iodine spiking. However the amplitude and increase rates of Cs were different from the iodine peaks in most of the cases. In *case 1* the iodine and cesium activities showed similar behaviour and so the parameters of iodine seemed to be applicable for cesium spiking calculation as well (Fig. 10.). In the other cases larger differences were observed and the iodine specific parameters proved to be not applicable.

Conclusions

A numerical model has been developed for iodine spiking simulation and has been validated against several shutdown transients, measured at Paks NPP. The analysis of the measured data showed that the strongest contribution to activity increase was related to the pressure decrease. The algorithm proved to be capable to predict the time and the amplitude of the ^{131}I spiking peak on the basis of activity data before the shutdown and the history of the main parameters during the transient and could be used for the prediction of iodine spiking peaks during refuelling periods.

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