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# A steady-state model for the activity release of unstable fission gases from defective WWER-440 fuel rods

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It is important from the viewpoint of radiological safety and fuel management to estimate the amount of unstable fission products present in the gap and plenum region, as well as, to know the amounts of unstable gaseous and volatile fission products released from defective fuel rods to the coolant during normal power operation. In paticular, the continuous monitoring of the number of gas-leaking fuel rods and the contamination level of the primary coolant by the fission gas products is important. The steady- state release of gaseous radioactive fission products (Xe, Kr) from defective WWER-440 fuel rods has been studied. The steady-state release of active noble gases from dfective fuel rods is described in terms of a diffusion model, while the transport of fission gases in the fuel-to-cladding gap is represented by a first order rate process. A computer program; DEFUS-01 has been developed to perform the required activity release calculations. The model gives a reasonable agreement with the published measured data.

إنه من المهم من وجهة نظر الأمان الإشعاعي و إدارة الوقود القيام بتقدير كمية نواتج الانشطار الغير مستقرة المتجمعة داخل الفراغات الموجودة بوحدة الوقود. و كذلك معرفة كمية الغازات المتطايرة و الغير مستقرة المنبعثة من وحدات الوقود المعيبة إلى ماء التبريد خلال فترات التشغيل العادية. عمليا, من المهم جدا المراقبة المستمرة لوحدات الوقود التي يحدث منها تثريب للغازات و كذلك مراقبة المستوى الإشعاعي في الدائرة الأولى الذي تحدثه نواتج الانشطار الغازية. و قد تم بصفة خاصة في هذا البحث دراسة انبعاث نواتج الانشطار الغازية (Xe, Kr) من وحدات الوقود المعيبة , حيث تم توصيف انبعاث الغازات النبيلة النشطة من وحدات الوقود المعيبة لمفاعلات (WWER-440) أثناء الحالة الثابتة باستخدام نموذج الانتشار , في حين تم تمثيل انتقال نواتج الانشطار الغازية من الوقود إلى الفراغات الموجودة داخل وحدة الوقود بواسطة معادلات تفاضلية خطية من الدرجة الأولى. و قد تم عمل برنامج حاسوب DEFOS-01 للقيام بالحسابات المطلوبة للنشاطية الإشعاعية المنطلقة من قضبان الوقود المعيبة. وقد أعطى هذا النموذج توافقا معقولا مع النتائج المعملية المنشورة.

**Keywords**: Fuel performance, Defective fuel rods, Gap activity, Coolant activity, Iodine-spiking time

## 1. Introduction

In the extensive literature in the field of fission gas release, relatively little attention has been paid to the transport phase in the gap of a defective fuel rods. Physical release processes for defective fuel have generally been deduced from analysis of coolant activity measurments in nuclear reactors and in reactor loop experiments. During the irradiation (burn-up) of uranium dioxide ($UO_2$) fuel, volatile fission products (e.g., noble gases) are partially released from the fuel matrix into the gap between the pellets and cladding. In the event of cladding failure, the radioactive fission products can migrate along failure cladding gap toward the defect size, with eventual release into the primary coolant. Therefore, the nuclear fuel tightness of nuclear reactors is a necessary requirement of their success fuel operation and safety. Because the fission gas products are the main shares of the total coolant activity, the information of their quality and quantity in the coolant would be valuable. It is important to realize that the models of fission products release which are employed to the fuel in failed rods differ markedly from these prevalling in intact rods [1-2].

The release behavior of the various fission products differs greatly but nevertheless leads to specific activities in the coolant which are constant in time provided that the reactor is operated at constant power and that the mode of operation of the purification system and the number and type of fuel rod defects do not change during the period under consideration [3]. The activities of numerous radioisotopes is

routinely measured during the operation of power reactors.

The radiological analysis of a coolant sample, which done as a routine work takes certain time and does not provide any indication to the defected fuel rods number. This can be obtained on the basis of $^{88}Kr$ activity measurment carried out by the radiation monitoring system and three actual technological parameters: $T_c$ – the coolant temperature at inlet, $P$ – the thermal power and $W$ – the coolant flow rate [4].

In this study, the steady-state fission product release model has been developed for defective WWER-440 fuel rods. The main features of the model are the calculations of the gaseous fission products (Xe, Kr) release mechanisms from the fuel into the coolant through the gap and failed cladding, with mass balance in the coolant during steady state operation.

Modeling the release of unstable noble fission gases from the defective fuel rods to the coolant can be described by the following two steps:

1. The release of unstable fission products to the fuel-to-cladding gap.
2. migration of these products to the coolant through the cladding defects.

The release from the gap to the coolant is assumed to occur by a first-order rate process through defect sites, while the transport of fission products in the fuel matrix is described by a diffusion mechanism.

## 2. Outline of the proposed model

### *2.1. Release from the fuel to the gap*

The transport of noble fission gases from the fuel to the fuel-to-cladding gap represents a source in the gap. The standard Booth [5] treatment for the release of radioactive fission products assumes a spherical $UO_2$ grain shape and radioactive-diffusion equilibrium such that the concentration of active species a uniform steady-state profile. The diffusion equation in a grain of radius $a$ is:

$$\frac{\partial C_i}{\partial t} = D_i\left(\frac{\partial^2 C_i}{\partial r^2} + \frac{2}{r}\frac{\partial C_i}{\partial r}\right) + \beta_i - \lambda_i C_i, \qquad (1)$$

where

$C_i$ is the concentration of radioactive dissolved gas atoms (atom/cm$^3$),
$\lambda_i$ is the decay constant of the isotope '$i$', and
$\beta_I$ is the gas production rate of the unstable istope "$i$" is given as.

$$\beta_i = Y' \dot{f} H, \qquad (2)$$

where

$H$ is the precursor correction factor,
$\dot{f}$ is the fission rate,
$Y^i$ is the fission yield (atom/fission), and
$D_i$ is the single gas atom diffusion coefficient (cm$^2$/s), given by the followingCorrelation [6].

$$D_i = 7.6\times10^{-10}\exp\left(-7\times10^4/RT\right) + s^2 J_v V' + 2\times10^{-40}\dot{f}, \qquad (3)$$

where $V'$ is the irradiation-induced vacancy concentration which given as:

$$V' = \frac{\alpha_s s^2}{2Z}\left\{\left[1+\frac{4KZ}{J\alpha_s s^2}\right]^{1/2} - 1\right\}. \qquad (4)$$

$\alpha$ is the fixed sink strength,
$S$ is the atomic jumb distance,
$R$ is the universal gas constant,
$T$ is the absolute temperature,
$Z$ is the number of sites arround a point from which recombination is inevitable,
$J_v$ is the vacancy jumb rate, and
$K$ is the defect production rate per atom.

The relevant physical data for each unstable noble fission product (*Xe, Kr*) is summarized in table 1.

For the steady state ($\partial C/\partial t$ = 0), the solution of eq. (1) from $t$ = 0 with an initial uniform concentration $C^0$ of intragranular radioactive gas atoms in spherical coordinates is:

$$C = \frac{\beta_i}{\lambda_i} + \frac{Ue^{-\lambda_i t}}{r}. \qquad (5)$$

Where

$$U = \frac{2}{a}\sum_{n=1}^{\infty}\exp(-(x-\lambda_i)t)\sin\left(\frac{n\pi r}{a}\right)$$

$$\times\left[\int_0^a C^o \sin\left(\frac{n\pi r'}{a}\right)r'dr' + (-1)^n \frac{\beta_i a^2}{n\pi\lambda_i}\right.$$

$$\left. -(-1)^n \frac{n\pi\beta_i D_i}{\lambda_i x}(1-\exp(xt))\right]. \quad (6)$$

The fission gas release rate can be calculated from Fick's law as [5]:

$$R^{ig} = \frac{6D_i}{a^2}\left\{\beta_i\sum_{n=1}^{\infty}\frac{1-\exp(-xt)}{x} + C^o\sum_{n=1}^{\infty}\exp(-xt)\right\}. \quad (7)$$

For long times $((\pi D_i/2+\lambda_i)t > 5)$ the experimental terms may be neglected and the release rate reduces to a sum which sums to the steady-state Booth release:

$$\overline{R}^{ig} = \frac{3\beta_i}{\mu}\left(\coth\mu - \frac{1}{\mu}\right), \quad (8)$$

with

$$\mu = a(\lambda_i/D_i)^{1/2}, \quad (9)$$

we can calculate the accumulated concentration of surviving (i.e., non-decayed) unstable release $Q$ as follows [5]:

$$Q = e^{-\lambda_i t}\int_0^t R^{ig} e^{\lambda_i t'}dt', \quad (10)$$

which yields

$$Q = \frac{3\beta_i}{\mu\lambda_i}\left\{erf\sqrt{\lambda_i t} - \frac{1}{\mu}(1-\exp(-\lambda_i t))\right\}.$$

$$+\left(6\sqrt{\frac{\omega}{\pi}} - 3\omega\right)\left(C^o - \frac{\beta_i}{\lambda_i}\right)e^{-\lambda_i t} \quad (11)$$

Since the temperature and fission rate are varying in the fuel rod, it was divided into 0.0205 cm thich and 1 cm high. In the result of this we obtained 3630 elementary volumes of each was considered isothermal, being in the constant irradiation field of fission rate. On the basis of the partitioning, eq. (7) was used for the calculation of the fission gas release rate for each elementary volume. The total release rate of the chosen isotope "$i$" from a whole fuel rod, $R_w^i$ is given by:

$$R_w^i = \sum_{n=i}^{242}\sum_{k=1}^{15} R_{nk}^{ig}(T,\dot{f})\times 2\pi\, r_k \Delta r \Delta z. \quad (12)$$

For each elementry volume, the fission rate was obtained in terms of a linear power of the fuel rod for the assumption of cosinusoidal power distribution [7]:

$$\dot{f}(r,z) = \frac{1}{2.88}\times 10^{11}\frac{q_{10}}{\pi\left(r_2^2 - r_1^2\right)}\cos(\pi z/H_e)A(r), \quad (13)$$

where

$q_{10}$ is the the linear power in the middle of the fuel rod,
$H_e$ is the extrapolated fuel rod length,
$r_2$ is the outer radius of the fuel pellet,
$r_1$ is inner radius of the fuel pellet,
$z$ is the axial coordinate in a cylindrical system, and
$A(r)$ is the power density coefficient as a function of the pellet radius.

The distribution of the fuel rod temperature is determined by the following expressions [7].

$$T_c(z) = T_{c1} + \frac{H_e q_{10}}{\pi W c_p}(\sin(\pi z/H_e) + \sin(\pi H/2H_e)). \quad (14)$$

$$T_{kz}(z) = T_c(z) + \frac{q_{10}}{2\pi r_4 \alpha_w}\cos(\pi z/H_e). \quad (15)$$

Table 1
Isotope data for unstable noble gases [6]

| Isotope | Decay constant, $\lambda_i$ ($s^{-1}$) | Fission yield, $Y^i$ | Precurso rcorrection factor, H |
|---|---|---|---|
| $^{85m}Kr$ | $4.30\times10^{-5}$ | 0.0130 | 1.019 |
| $^{87}Kr$ | $1.52\times10^{-4}$ | 0.0252 | 1.016 |
| $^{88}Kr$ | $6.78\times10^{-5}$ | 0.3350 | 1.002 |
| $^{133}Xe$ | $1.53\times10^{-6}$ | 0.0670 | 1.205 |
| $^{135m}Xe$ | $7.65\times10^{-4}$ | 0.0110 | 6.373 |
| $^{135}Xe$ | $2.12\times10^{-5}$ | 0.0654 | 3.353 |
| $^{137}Xe$ | $3.02\times10^{-3}$ | 0.0606 | 1.072 |
| $^{138}Xe$ | $8.18\times10^{-4}$ | 0.0642 | 1.003 |
| $^{131}I$ | $1.00\times10^{-6}$ | 0.0238 | 1.000 |
| $^{133}I$ | $9.26\times10^{-6}$ | 0.0670 | 1.100 |
| $^{135}I$ | $2.91\times10^{-5}$ | 0.0630 | 1.000 |

$$T_{kw}(z) = T_{kz} + \frac{q_{10}}{2\pi r_4}\cos(\pi z/H_e)\frac{r_4}{\lambda_k}\ln(r_4/r_3). \quad (16)$$

$$T_{r2}(z) = T_{kw} + \frac{q_{10}}{2\pi r_4 \alpha_s}\cos(\pi z/H_e)\frac{r_3}{r_2}. \quad (17)$$

$$T_r(r,z) = T_{r2}(z) + \frac{A(r)q_{10}}{4\pi(r_2^2 - r_1^2)}\cos(\pi z/H_e) \times\left(r_2^2 - r^2 - 2r_1^2\ln(r/r_1)\right)/\lambda_p, \quad (18)$$

where

$T_{c1}$ is the average coolant temperature at the inlet,
$T_c(z)$ is the coolant temperature,
$T_{kz}(z)$ is temperature at the outer radius of the cladding ($r_4$),
$T_{kw}(z)$ is the temperature at the inner radius of the cladding ($r_3$),
$T_{r2}(z)$ is the fuel temperature at the pellet radius ($r_2$),
$H$ is the fuel rod length,
$W$ is the coolant mass flow rate,
$r_1$ is the the inner radius of the fuel pellet,
$r_2$ is the the outer radius of the fuel pellet,
$r_3$ is the inner radius of the cladding,
$r_4$ is the outer radius of the cladding,
$\alpha_w$ is the heat transfer coefficient from the side of the water,
$C_p$ is the coolant specific heat capacity, and
$\lambda_p$ is the fuel thermal conductivity, given by the following formula [6].

$$\lambda_p = \frac{1}{0.258T + 3.77} + 1.1\times10^{-6}T + 1.01\times10^{-13}T^{-3}\exp\left(7.2\times10^{-4}T\right). \quad (19)$$

The maximum linear power of the fuel rod was calculated [7]

$$q_{10} = \frac{P_{th}C_{efr}C_{efz}}{n_k H}, \quad (20)$$

where

$P_{th}$ is the reactor thermal power,
$C_{efr}$ is the radial form factor,
$C_{efz}$ is the axial form factor, and
$n_k$ is the number of fuel rods in the reactor core.

### 2.2. Total activity in the gap

If we assume that the release rate of unstable noble fission gases, $R_w^i$, is constant during irradiation. Then, the number nuclide $i$ in the gap, $N_g^i$, is obtained from the following expression [8].

$$\frac{dN_g}{dt} = R_w^i - \lambda_i N_g. \quad (21)$$

For steady-state operating conditions, equilibrium number of unstable noble fission gases is $R_w^i\lambda_i$. Then the gap activity of fuel rod per unit active length, $A_g$, is calculated by.

$$A_g = \lambda_i N_g = R_w^i. \quad (22)$$

At equilibrium condition the release rate of noble gases to the gap is equal to the rate of decaying in the gap. Therefore, the total activity in the gap from the whole fuel rod is given as.

$$A_t^g = R_w^i L, \tag{23}$$

where $L$ is the total active length of the fuel rod.

### 2.3. Release from the fuel-to-cladding gap to the coolant

Under the asumption that that total release rate, $R_w^i$, is constant during state-state operating conditions and that the rate of fission gas release from the failed cladding to the coolant is assumed to be a first order rate process, proportional to the number of fission gas atoms, $N_g^i$ in the fuel-to-sheath gap. Then, the mass balance for noble gas in the fuel-to-cladding gap can be written as [9].

$$\frac{dN_g^i}{dt} = R_w^i - (\nu + \lambda_i)N_g^i, \tag{24}$$

where $\nu$ = the escape-rate constant ($s^{-1}$). The release rate of fission products from the gap to the coolant, $R_c$, under steady-state operating conditions is obtained from eq. (24) as follows [10].

$$R_c = \nu N_g^i = \frac{\nu}{\lambda_i + \nu} R_w^i. \tag{25}$$

Now the balance equation for the number of fission gas atoms in the primary coolant can be written as.

$$\frac{dN_c^i}{dt} = R_c^i - \lambda_i N_c^i - \hbar N_c^i, \tag{26}$$

where $\hbar$ is the total purification rate. the term $(\lambda_i + \hbar)N_c^i$ denotes the activity of the isotope $i$ in the primary coolant $A_c^i$. During steady-state conditions, the fission gas release rate determines the activity of the isotope $i$ in the coolant.

$$A_c^i = R_c^i, \tag{27}$$

dividing eq. (27) by the volume of the primary circuit ($V$) we obtain the theoretical volumetric activity of the considered isotope ($A_i^t$) for the gas-leaking fuel rod. Subsequently dividing the measured volumetric activity of the isotope $^{88}Kr$ in the primary coolant ($A_m^i$) by the theoretical volumetric activity of the same isotope ($A_i^t$) we obtain the coefficient ($n$) describing the gas-leaking fuel rods number which can be used for evolution the volumetric activity of the other fission gas products under asumption that the fission gas has the same propertions of isotopes for the all leaking fuel rods.

$$n = \frac{A_m^i V}{R_c^i} \quad (\text{for } ^{88}Kr). \tag{28}$$

Hence the evaluation of the volumetric activity of the other isotopes ($A_i^o$) is obtaind according to the expression.

$$A_i^o = nA_i^t = n\frac{R_w^i}{V}. \tag{29}$$

### 4.2. Iodine-spiking time

The time at which the concentration of iodine in the reactor cooling ststem is at the maxximum is calculated by the follwing eq. [8].

$$T_m = \left(\frac{1}{L - k_t}\right) \ln\left\{\left[\frac{N_{co}}{JN_{go}}\left(\frac{k_t - L}{k_t}\right) + 1\right] \times \left(\frac{\lambda + L}{\lambda + k_t}\right)\right\},$$

where

$\lambda$ is the radioactive decay constant,
$L$ is the system loss rate constant,
$k_t$ is the escape rate constant,
$N_{go}$ is the total number of atoms of each isotope " $i$ " that are available for release from the fuel to cladding on shutdown, and

$N_{co}$ is the initial quantity of each isotope " $i$ " in the coolantat the time of Shutdown.

The equation indicates that the value of $T_m$ will increase with the fuel rod length ( i.e. it takes longer for the species to diffuse the extra path length) but will decrease as the iodine is removed from the system.

## 3. Results and discussions

According to the above presented algorithm, a computer program; DEFUS-01 has been written to perform the volumetric activity calculations of the noble fission gas products in the coolant of WWER - 440 on the basis of $^{88}Kr$ volumetric activity measured by the radiation monitoring system of the reactor. The value of $1.17\times10^{-4}$ Ci/l is used In the calculations to obtain the volumetric activity of the other isotopes.

Table 2 presents a comparison between the calculated results of the noble fission gas volumetric activities in the primary coolant and a measured data of WWER – 440 [4]. The calculation were performed for thermal power of 1375 MW, the number of fuel rods equal to 43974, the coolant temperature at inlet equal to 269 °C and the coolant flow rate equal to 8135 kg/s. the calculated results of the model gives a reasonable agreement with measured data.

Fig. 1 shows theradial distribution of fission rate and fuel temperature. From the figure it is shown that the centerline temperature was 1425 °C and the surface temperature was 637 °C. From the figure, it is seen that the fission rate at the fuel rod surface is slightly higher than the inter part, and this difference will rise up as the fuel function of the decay constant on the basis of the known $^{88}Kr$ volumetric activity equal to $1.17\times10^{-4}$ Ci/l. these calculations were obtained for a PWR with thermal power of 1375 MW, coolant flow rate equal to 8135 kg/s and average coolant temperature equal to 542 K. The number of fuel rods is equal to 43974. The iodine-spiking as a function of time after reactor shutdown is shown in Fig. 4. The number of atoms of each isotope released after reactor shutdown is dependent on the initial inventory stored in the gap during the fuel irradiation. From the figure, it is clearly shown that the rate of escape of I-131 during shutdown is much greater than that of the shorter lived isotope I-133. Also, it is shown that the peak of the shorter lived isotope occurred earlier than the longer lived one.

Burn-up increased. The fuel and cladding temperatures as well as the fission rate are plotted in fig. 2 as a function of the fuel rod length. The radial and axial distributions are necessary for the release rate calculations of fission products from the fuel, in addition to the activity of fuel-to-cladding gap.

During periods of constant power a continuous leakage occurs which caused a slow rise to equilibrium, over a period of more than 30 days. Knowing the total activity, the total inventory of $^{88}Kr$ isotope free of the fuel can be determined. Fig. 3 presents the evaluated volumetric activity of the fission gas products in the primary coolant as a

Table 2
Comparison of the measured data [4] and the calculated results

| | Isotopes (Ci/l) | | | | |
|---|---|---|---|---|---|
| | $^{133}Xe$ | $^{135}Xe$ | $^{85m}Kr$ | $^{88}Kr$ | $^{87}Kr$ |
| Measruments [3] | $2.5\times10^{-3}$ | $6.6\times10^{-4}$ | $8.6\times10^{-5}$ | $1.10\times10^{-4}$ | $1.3\times10^{-4}$ |
| Calculations | $5.37\times10^{-3}$ | $7.64\times10^{-4}$ | $7.83\times10^{-5}$ | $1.17\times10^{-4}$ | $5.94\times10^{-5}$ |

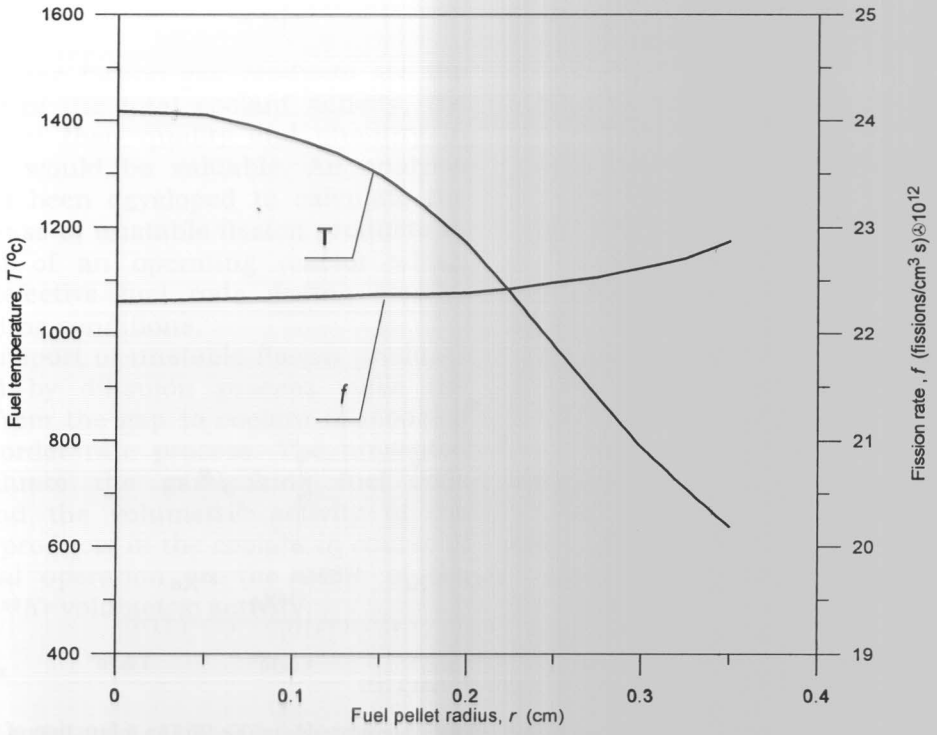


Fig. 1. Calculated fission rate and fuel temperature as a function of the fuel pellet radius.

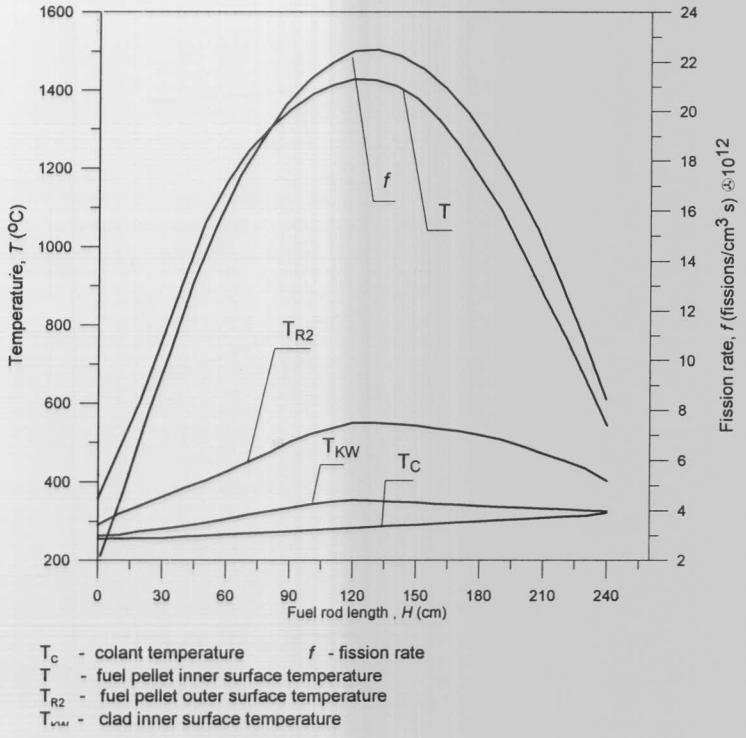


Fig. 2. Calculated foolant, cladding and fuel temperatures and fision rate as a function of the fuel length.

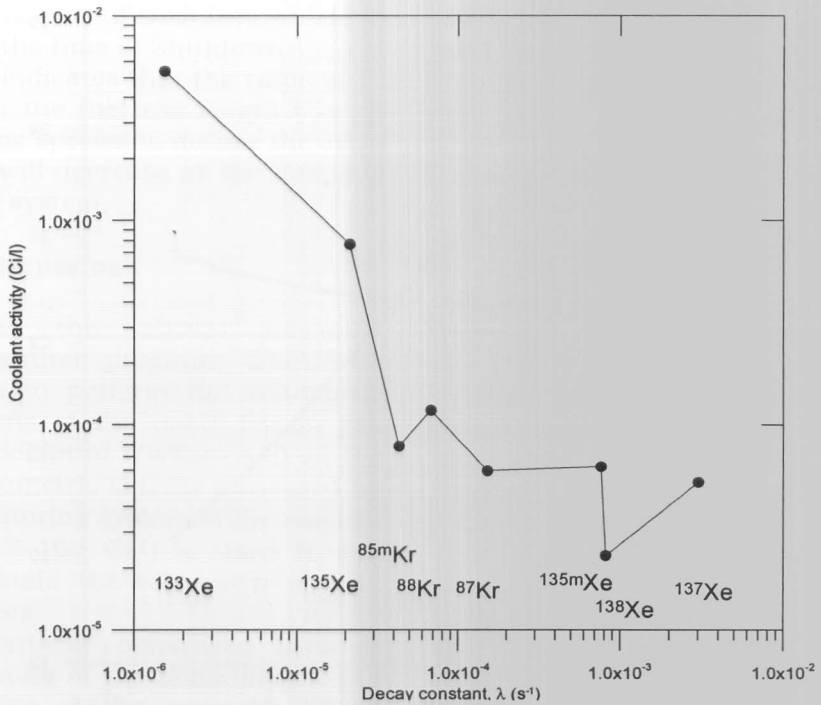


Fig. 3. Calculated volumetric activity of the gaseous radioactivity fission products (Xe, Kr) as a function of decay constant ($(\lambda)$ on the basis of $^{88}Kr$ volumetric activity equal to 1.17 $\times 10^{-4}$ Ci/l.

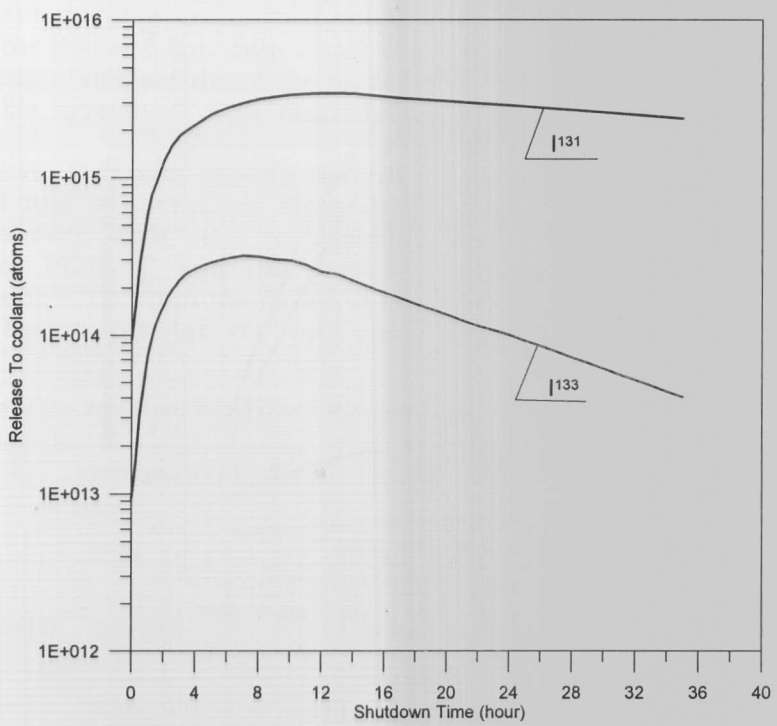


Fig. 4. Loading-spiking as a function of time after reactor shuddown.

## 4. Conclusions

Because the fission gas products are the main share of the total coolant activity, the information of their quality and quantity in the coolant would be valuable. An analysis method has been developed to calculate the activity release of unstable fission products in the coolant of an operating reactor which contains defective fuel rods during steady-state operating conditions.

The transport of unstable fission products is modeled by diffsuion process while the migration from the gap to coolant is modeled by a first order rate process. The presented model evaluate the gas-leaking fuel rods number and the volumetric activity of the fission gas products in the coolant in course of the nominal operation on the basic of the measured $^{88}Kr$ volumetric activity.

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