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Key words: defective (failed) fuel rod, radioactive fission products (radioactive nuclides), coolant activity, research nuclear reactor, mechanistic code, code verification.

Abstract

The OECD/NEA/IAEA IFPE database on experiments at Siloe research reactor (Grenoble, France) is discussed in brief. Release of radioactive fission products from defective fuel rods was investigated in the experiments at Siloe reactor. Experiments are chosen from the database which are close to conditions of defective fuel operation in WWERs. The chosen experiments were used for verification of the mechanistic RTOP-CA code developed to model the activity release from failed WWER fuel rods during reactor operation.

Verification of the RTOP-CA code against the OECD/NEA/IAEA IFPE database on activity release from defective fuel rods

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INTRODUCTION

Fuel failures are possible during operation of nuclear utilities. Failures lead to release of radioactive fission products from defective fuel rods into primary coolant. For objectives of radiation safety failed fuel diagnosis is performed at nuclear power plants (NPPs). The diagnosis is performed both under operation conditions and during refueling. Analysis of fuel failures under operation conditions provides the on-line evaluation of number and characteristics of defective fuel rods (burnup, defect size). Preliminary evaluations are necessary to reduce time, optimize financial costs and reduce errors in leakage tests after reactor shutdown.

The up-to-date methods of failed fuel diagnosis are based on measurements of specific coolant activity for reference radioactive nuclides and application of computer codes for data interpretation. Nowadays, mechanistic computational codes are developed for purposes of failures diagnosis. The DIADEME code is developed in France for PWRs [1,2]; the RTOP-CA code is developed for Russian WWER reactors [3,4]. The engineering code RELWWER [5] being used for WWERs till present time is based on correlations.

The mechanistic code RTOP-CA provides a self-consistent modelling of activity release into primary circuit at WWERs (radioactive nuclides of iodine, caesium, xenon, krypton). The code is based on physical models. Three main model groups describe the following physical processes in failed fuel rods:

- accumulation and behavior of radioactive fission products (RFPs) in fuel, RFP release from fuel due to diffusion and knockout;
- mass transfer of fission products inside failed fuel rod and mass exchange with coolant;
- behavior of RFPs in primary circuit during reactor operation with taking into account coolant purification at filters, contribution of tramp uranium to overall activity level, possible adsorption of nuclides at surfaces in the core.

At present time physical models of the RTOP-CA code have been separately verified using a wide database of in-pile and out-of-pile, full- and small-scale experiments. Integral verification of the code is in progress. Database for integral verification includes experiments on irradiation of artificially defected fuel rods in research reactors and data on primary coolant activity at NPPs with WWER-type reactors.

For detailed verification of mechanistic codes it is important to carry out experiments under controlled conditions. If matching of calculations with data makes it necessary to vary a great number of unknown parameters than "verification" loses its significance. In respect to verification of mechanistic codes, experiments in research reactors are of the most interest. In this case it is possible to achieve prototype conditions of irradiation and to control all dominant experimental parameters.

Open literature data on activity release from defective fuel rods under determinate conditions are scarce [6,7]. A wide experimental program on irradiation of defective fuel rods has been carried out in Siloe research reactor in France. This body of data is largely unpublished but with the agreement of EDF and FRAMATOME was included in the OECD/NEA/IAEA International Fuel Performance Experimental (IFPE) database [8-20].

A brief description of the experimental program at Siloe research reactor is given in the present paper. Experiments have been chosen which were carried out in conditions close to operational regimes of failed fuel rods in WWERs. Data of chosen experiments were compared to the RTOP-CA calculations on activity release.

Experiments in Siloe reactor.

Experiments in Siloe reactor have been performed with shortened fuel rods in two water loops called Bouffon and Jet Pompe. Most of the rods were artificially defected. List of experiments is shown in table 1.

Table 1. List of experiments in Siloe reactor.

Experiment	Water loop	Experiment	Water loop	Experiment	Water loop
CYFON-1	<i>Bouffon</i>	EDITH-2	Jet Pompe	CRUSIFON-2	<i>Bouffon</i>
CYFON-2	<i>Bouffon</i>	EDITH-5	Jet Pompe	CRUSIFON-5	<i>Bouffon</i>
EDITH-1	<i>Bouffon</i>	CRUSIFON-1bis	<i>Bouffon</i>	EDITHMOX-1	Jet Pompe

The CYFON experimental series investigated the release of fission products from failed fuel rods during variable power/load following. In the EDITH series period of power variations was increased in order to approach the steady state release. The CRUSIFON series investigated the behaviour of fuel rods which were initially intact but artificially defected during irradiation in Siloe.

The Bouffon loop consists of two vertical tubes connected at both ends to form a continuous circuit for pressurised water. The experimental fuel rod is situated in the bottom of one tube below a heater. Due to thermosyphoning the heater provides an up-current of cooling water over the experimental fuel rod. A small portion of water flow is diverted into an out-of-pile circuit with filters, gas purification system and measuring equipment. Activity of fission products is detected on-line by gamma spectrometer. In addition, water sampling is possible with measurement of activity in more detail. A scheme of the Bouffon water loop is shown in Fig.1. For main characteristics of the loop see table 2. The Bouffon operating regime is more typical of BWRs.

The Jet Pompe loop comprises two co-axial vertical tubes (Fig.2). The experimental fuel rod is situated in the inner tube and coolant flow over it is provided by a steam injector pump at the bottom of the same tube. Jet Pompe characteristics are listed in table 2. Comparison of technical specifications for the Jet Pompe and

Table 2. Characteristics of water loops in Siloe reactor.

Parameters	Bouffon boiling flow	Jet Pompe pressurized water
Channel diameter (mm)	29.5	38
Fuel rod length (mm)	350	500
Number of rods	1	1 or 4
Max length of fuel rods (mm)	400	1200
Max surface power (W/cm ²)	233	165
Circuit volume (l)	4.2	3.7
Flow rate (m ³ /hr)	0.3	1.1
Clean up rate (l/hr)	2.6	34
Rod inlet temperature (°C)	150	280
Rod outlet temperature (°C)	150	up to saturation temperature
Pressure (bar)	130	130

Bouffon loops shows that the both loops operate at equal pressure, but flow rate in Jet Pompe is three times greater and coolant temperature 280 °C is more typical of conditions in pressurised water reactors than 150 °C in Bouffon. At coolant temperature of 150 °C pellet-to-cladding gap in Bouffon fuel rods is occupied by liquid water when linear heat generation rate is below 30 kW/m. In this case radioactive nuclides inside fuel rod are transferred through liquid whereas RFP transfer through gas phase is more typical of nominal operation conditions for failed fuel rods in WWERs (see the next paragraph). Thus, conditions in the Bouffon loop do not correspond in full to mass transfer conditions in failed fuel rods which are modeled by the RTOP-CA code.

Modelling of mass transfer inside failed fuel rod by the RTOP-CA code.

The RTOP-CA model for mass transfer inside failed fuel rod takes into account the transport of radioactive nuclides in central channel of fuel stack, transport in pellet-to-cladding gap and mass exchange between gap and central channel through cracks in UO₂ pellets and through interpellet spaces. In relation to mass transfer, central hole in WWER fuel pellets is a crucial difference from PWR and BWR fuel. A separate physical model is included into the RTOP-CA code to model the mass transfer in fuel rods with solid pellets.

Central channel of fuel stack in failed WWER rods under operation conditions is always occupied with steam. In pellet-to-cladding gap steam condensation is possible on inner cladding surface with formation of water film. Film thickness is governed by local heat generation rate. When heat generation rate is low and/or fuel burnup leads to small width of pellet-to-cladding gap, the gap may be flooded with water. At present time the RTOP-CA code is verified for burnups up to 40-45 MW-days/kgU. For moderate burnups in failed fuel rods under operation conditions it can be assumed in most cases that water film does not block up the gap and RFPs are transported in it through the gas phase. A model to describe the mass transfer in water-filled gap is under development and is not included into the

current version of the RTOP-CA code. It is worth noting that even if pellet-to-cladding gap in failed WWER fuel rods under operation is flooded with water, mass transfer mechanisms in central channel of fuel stack do not change. In the case of PWRs, the qualitative change of mass transfer regime takes place for fuel rod as a whole.

The following mechanisms of mass transfer through gas phase inside failed fuel rods are considered in the RTOP-CA code. First, radioactive nuclides are transported due to molecular diffusion. Second, pulsations of coolant pressure in primary circuit result in pulsations of gas flow inside failed fuel rod. Pulsation flow rate may be significant because of low hydraulic resistance in central channel of fuel stack. This fact leads to additional mass transfer. Additional transfer is equivalent to diffusion process with effective diffusivity which is a function of pulsations amplitude and frequency.

Limitations on use of the OECD/NEA/IAEA IFPE database for verification of the RTOP-CA code.

For on-line failure diagnosis at NPPs it is common to use data on RFP activity during steady state reactor operation or during slow power changes. In such regimes more reliable modelling is possible for primary coolant activity. Transients are often accompanied with spiking effect – a sudden increase of RFP release from defective fuel rods. At present time detailed models of spiking effect are not elaborated. In most computational codes description of spikes is based on simplified assumptions and correlation dependences (e.g. [21]). The RTOP-CA code comprises a simplified phenomenological model of spiking effect, therefore activity release during power maneuvering may be simulated by the code with higher inaccuracy.

By this reason, the RTOP-CA should be preferably verified using experimental data on irradiation of defective fuel rods under steady state conditions with rare power maneuvers. Typical frequency of power maneuvering for WWERs is no more than a week. This is a consequence of reduced power consumption by industrial enterprises on weekends and holidays. A period of about a week is sufficient for most of radioactive nuclides (which activity is measured at NPPs) to establish a steady state release from failed fuel rods.

The CYFON experimental series was carried out under cyclic power changes. One day was chosen as a typical period of power variations (Fig.3). The chosen period is insufficient for establishing a steady state release from defective fuel rod for most radioactive nuclides (except for short-lived species). Upper and lower power levels were selected to provide an unstable phase state of water inside fuel rod. The purpose was to increase RFP release due to washing off by water – mainly for iodine isotopes which are adsorbed at inner cladding surface. Unstable phase composition of water in pellet-to-cladding gap leads to unstable conditions of mass transfer inside fuel rod and to unstable mass exchange with coolant. The consequence is high scatter of experimental data. The extent of data scatter can be assessed by comparing (during the same time interval) the alternative measuring techniques for detection of coolant activity – with aid of on-line detectors and by γ -scanning of water samples. Such analysis for the EDITHMOX-1 experiment (performed under the most stable conditions of irradiation power, Fig.7) shows data

scatter from 20% to 2-3 times, in some cases up to an order of magnitude. For the CYFON series data scatter is higher than that for the EDITHMOX-1 experiment.

The mentioned reasons (with taken into account atypical heat-hydraulic regime in Bouffon water loop) impose limitations on using the CYFON experimental data for verification of the RTOP-CA code. Figs.4-6 show some examples of comparison between calculations and experimental kinetics of ^{133}Xe , $^{85\text{m}}\text{Kr}$, ^{133}I activity in the CYFON-2 experiment and in the subsequent experiment EDITH-1. It can be seen that behavior of coolant activity bears an intermittent character which is a consequence of water evaporation and condensation inside fuel rod during power ramps and drops. In whole, the RTOP-CA predictions are adequate to the observed behavior of activity. Accuracy of predictions for the realized experimental conditions is noticeably lower than for steady state regime of activity release (see below – results of the RTOP-CA verification on the EDITHMOX-1 experimental data).

The EDITH-2 experiment was performed in Jet Pompe water loop. A hole of 0.3 mm in diameter was drilled in the upper plenum of fuel rod. Irradiation history included longer intervals of steady state operation. The OECD/NEA/IAEA IFPE database comprises data on activity kinetics for short-lived nuclides ^{134}I , ^{87}Kr , ^{138}Xe at several stages of irradiations. For nuclides with longer lifetime the release rate (R/B) is recorded for several time points. The majority of records for the R/B ratio was done after power ramps when activity level is still influenced by the spiking effect. Only the date of the R/B measurement is indicated in the database. Under transient conditions this fact entails a considerable uncertainty in time which hampers the adequate comparison between calculation and experimental results.

In the EDITH-5 experiment the defect was a fatigue crack along the fuel column. It was characterized prior to irradiation by measuring its conductance for water and helium as a function of pressure. Crack length was found to be about 3 mm and its width was estimated to be $\sim 1 \mu\text{m}$. In addition these tests demonstrated that crack opening depends on stress conditions in cladding – estimated crack width changed with difference between pressures inside and outside fuel rod. Variation of crack opening under operation in different power regimes can lead to unpredictable changes of RFP release rate. The EDITH-5 experiment included several stages of fuel rod irradiation. Activity release was feeble, on-line γ -detectors recorded the signal with large fluctuations on background level. Only activity spikes were clearly seen during power ramps and drops. Several water samples were taken at the second stage of irradiation. Activity in these samples significantly differed from records of on-line detector. Due to possible variation of crack state in course of the experiment and considerable uncertainty of data on RFP release between activity spikes, the EDITH-5 experiment was declined to be used for verification of the RTOP-CA code.

In the CRUSIFON series fuel rods were artificially defected in course of irradiation. Defects were the through-wall cracks in cladding. Defect hydraulic resistance was measured only for the CRUSIFON-5 fuel rod. Hydraulic resistance decreased with growing pressure gradient across cladding wall. It may be the consequence of crack opening with increasing stresses. For the other CRUSIFON cases the IFPE database does not include any information about effective hydraulic size of defects. Defect size is one of the most important parameters which govern RFP release from failed fuel rods. With this parameter uncertain, the CRUSIFON experiments are not suitable for verification of the RTOP-CA code.

Verification of the RTOP-CA code on EDITHMOX-1 experimental data.

The most appropriate for verification of the RTOP-CA code is the EDITHMOX-1 experiment. The OECD/NEA/IAEA IFPE database [20] as well as ref. [7] include a detailed description of the EDITHMOX-1 experimental parameters. A particular feature of the EDITHMOX-1 program compared to the other experiments with defective fuel rods in Siloe reactor lied in long periods of steady state irradiation conditions (Fig.7). Study of activity release was performed in Jet Pompe water loop. Both these factors in combination make the EDITHMOX-1 experiment the most typical of operation conditions for failed fuel rods at NPPs with WWER-type reactors.

Release of radioactive nuclides from failed MOX-fuel rods was studied in the EDITHMOX-1 experiment. A cylindrical hole of 0.3 mm in diameter was drilled in cladding approximately in the middle of the fuel rod. RFP release rate was measured on-line for several values of heat generation rate $\sim 10, 15, 20, 26$ and 30 kW/m . Also water samples were γ -scanned after being taken from the loop circuit. In the beginning of the first stage of irradiation linear heat generation rate was 8.6 kW/m and RFPs were released from fuel rod with pellet-to-cladding gap filled with liquid water. Data on activity release during this time interval were not used for verification of the RTOP-CA code.

Comparative study reveals no significant differences in UO_2 - and MOX-fuel behavior under in-pile conditions for burnups up to $\sim 40 \text{ MW}\cdot\text{days/kgU}$ [22]. Framework of the RTOP-CA code makes it possible to perform calculations for MOX-fuel. In calculations for the EDITHMOX-1 experiment initial isotopic composition of fuel pellets was specified according to typical data on MOX-fuel for PWRs [23]. Enrichment of pellets with Pu isotopes was 7.8% in the experiment. It is known that thermal conductivity of $(\text{U,Pu})\text{O}_2$ pellets is several percent lower than that of normal UO_2 fuel [24]. In ENIGMA computational code this difference is taken into account by introducing proportionality factor $\lambda_{\text{MOX}} = 0.92\lambda_{\text{UO}_2}$ [25]. The ENIGMA approach was also applied in calculations with the RTOP-CA code for the EDITHMOX-1 experiment.

Experimental data on release rate (R/B) of various radioactive nuclides are shown in Figs.8-11. Figures correspond to different time points during the first stage of irradiation in Siloe reactor. Both the data of on-line activity measurements and data obtained by water sampling are shown. Figs.12-16 demonstrate dependence of RFP release rate upon linear heat generation rate. Data on release kinetics for different radioactive nuclides are shown in Figs.17-25. Kinetics of activity release was measured only during the first stage of irradiation. For the second and the third irradiation stages the data on release rate (R/B) for several time points are only presented (Figs.7,12-16).

The RTOP-CA calculation results are shown in Figs.8-25 together with experimental data. At Figs.12-16 two calculations and two sets of experimental data points are shown for each liner heat generation rate. These points correspond to measurements of activity at fixed power level in different time intervals (see Fig.7). In Figs.8-25 it can be seen that the RTOP-CA predictions are adequate both to measured release rate and to measured kinetics of activity release of various RFPs into coolant.

Table 3. Root-mean-square deviation of calculations from the EDITHMOX-1 experimental data.

Isotope	σ
^{85m} Kr	0.37
⁸⁸ Kr	0.41
⁸⁹ Kr	0.10
¹³⁵ Xe	0.42
¹³⁷ Xe	0.16
¹³⁸ Xe	0.12
¹³² I	2.11
¹³³ I	3.19
¹³⁴ I	0.46
¹³⁵ I	1.98

Root-mean-square deviation (σ) of calculated kinetic curves of activity release from experimental results was computed according to formula

$$\sigma = \left(\int_{t_1}^{t_2} (A_{exp} - A_{calc})^2 dt \right) \left(\sqrt{\int_{t_1}^{t_2} A_{exp}^2 dt} \cdot \sqrt{\int_{t_1}^{t_2} A_{calc}^2 dt} \right)^{-1}.$$

Here A_{exp} and A_{calc} are current coolant activity according to experimental records and calculations, correspondingly. Calculated values of σ are listed in table 3. Taking into consideration the level of data uncertainty in measurements it can be stated that the RTOP-CA code gives a satisfactory prediction of observed coolant activity kinetics.

CONCLUSIONS

Analysis has shown that there are limitations on using the OECD/NEA/IAEA IFPE database for verification of the RTOP-CA code. The RTOP-CA code was developed for failed fuel diagnosis during operation of WWER-type reactor for average fuel burnup up to 40-45 MW-days/kgU. With relation to integral verification of the code, most experiments in Siloe reactor have one or several of the following drawbacks:

- incomplete information for several experiments, in particular, equivalent defect size for microcracks is not specified;
- measurement of activity during spikes or when activity is still influenced by spiking effect;
- heat-hydraulic regime in Bouffon water loop leads to conditions of mass transfer inside fuel rod which could not be modeled with current version of the RTOP-CA code. In operating WWER failed fuel rods with burnup below 40-45 MW-days/kgU fission products are transported through gas phase whereas in experiments carried out in Bouffon loop pellet-to-cladding gap was filled with water.

Model of mass transfer in water-filled gap for the RTOP-CA code is under development.

The most appropriate conditions from the point of view of operation conditions for failed WWER fuel rods with moderate burnups were realized in the EDITHMOX-1 experiment. The OECD/NEA/IAEA IFPE database includes a complete information about EDITHMOX-1 experimental parameters. So it was possible to use the EDITHMOX-1 experiment for detailed verification of the RTOP-CA code. Comparison of calculations with experimental data has shown that the RTOP-CA predictions on kinetics of RFP release from defective fuel rods are in good agreement with observations in the whole range of experimental conditions.

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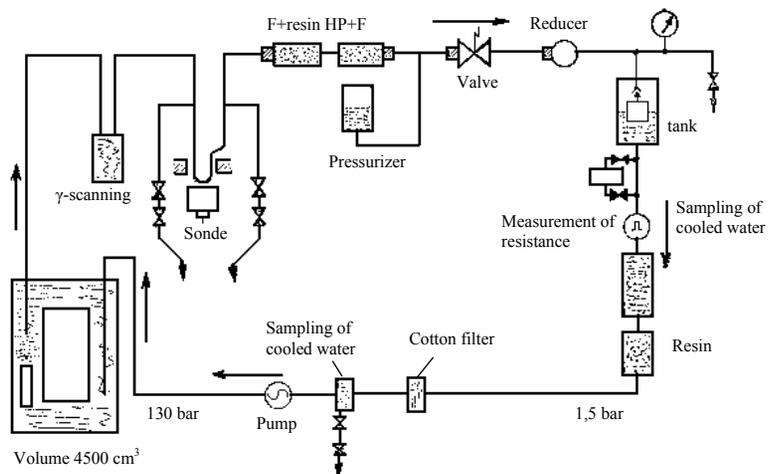


Fig.1. Scheme of Bouffon water loop in Siloe reactor.

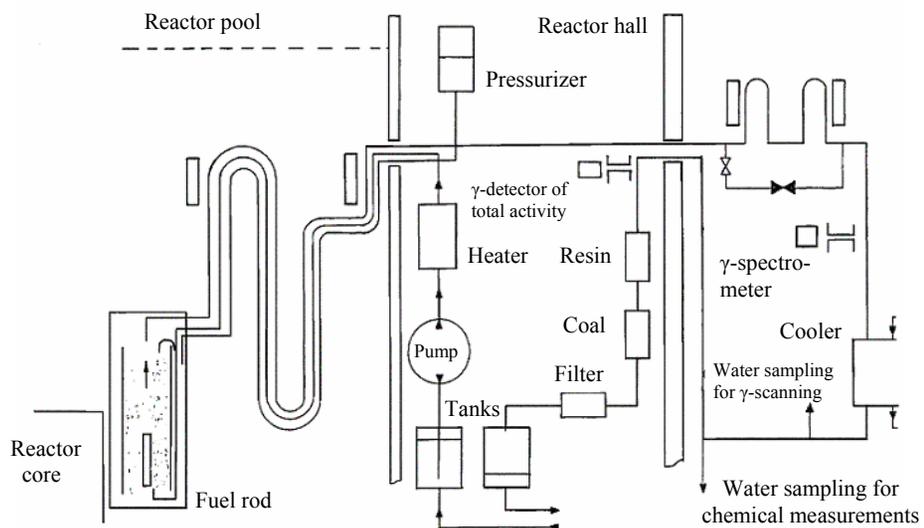


Fig.2. Scheme of Jet Pompe water loop in Siloe reactor.

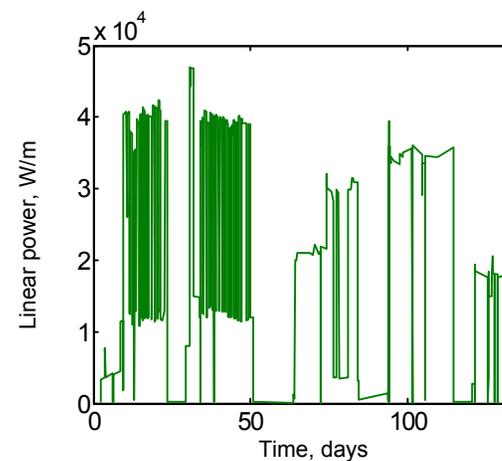


Fig.3. Irradiation history for CYFON-2 (first 50 days) and EDITH-1 experiments.

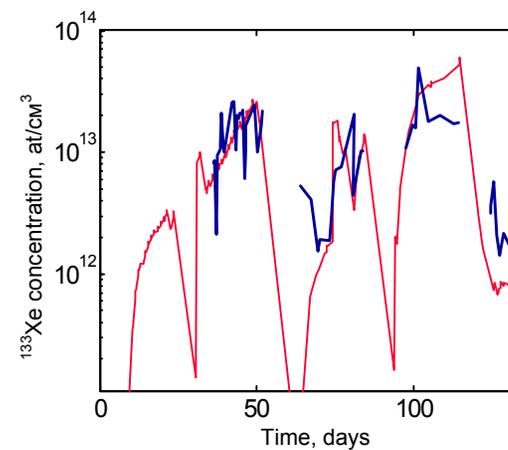


Fig.4. Release kinetics for ^{133}Xe into Bouffon water loop: — — CYFON-2 and EDITH-1 experimental data, — — RTOP-CA calculation.

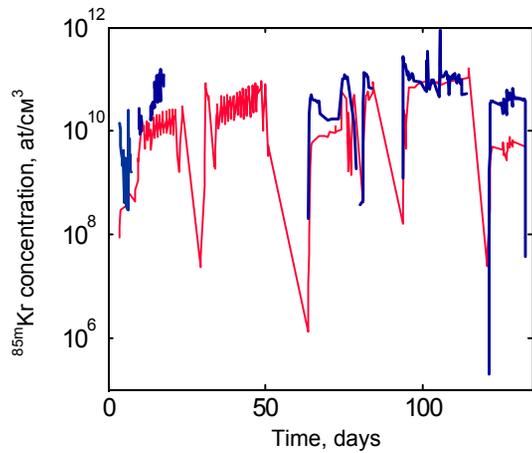


Fig. 5. Release kinetics for ^{85m}Kr into Bouffon water loop: — CYFON-2 and EDITH-1 experimental data, — RTOP-CA calculation.

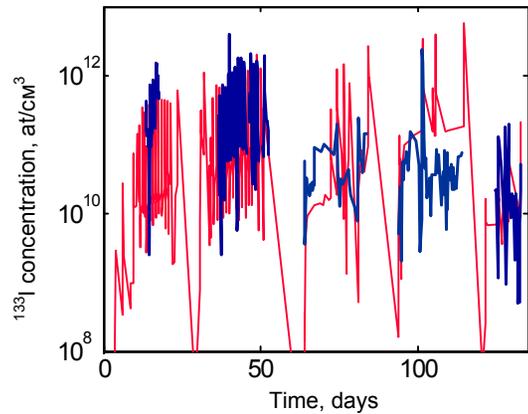


Fig. 6. Release kinetics for ^{133}I into Bouffon water loop: — CYFON-2 and EDITH-1 experimental data, — RTOP-CA calculation.

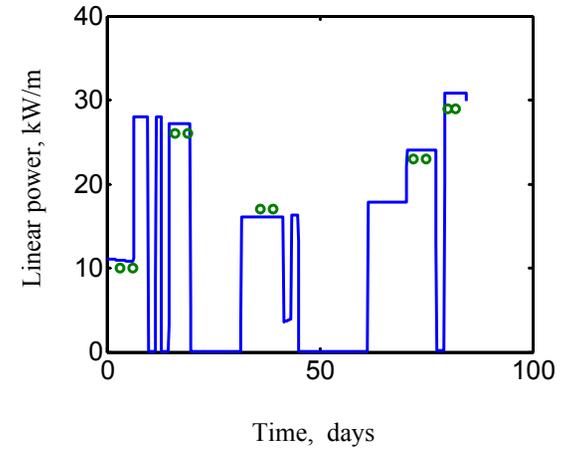


Fig. 7. Irradiation history in three irradiation cycles of the EDITHMOX-1 experiment. Markers show the time points when RFP release rate (R/B) was experimentally measured.

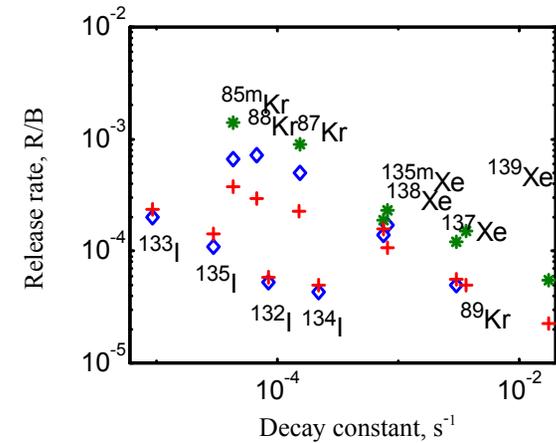


Fig. 8. Release rate for different radioactive nuclides from defective fuel rod; heat generation rate 10.36 kW/m, the third day of irradiation in Jet Pompe water loop: + — the RTOP-CA calculation, * — data of on-line activity measurements, \diamond — experimental data for activity in water samples.

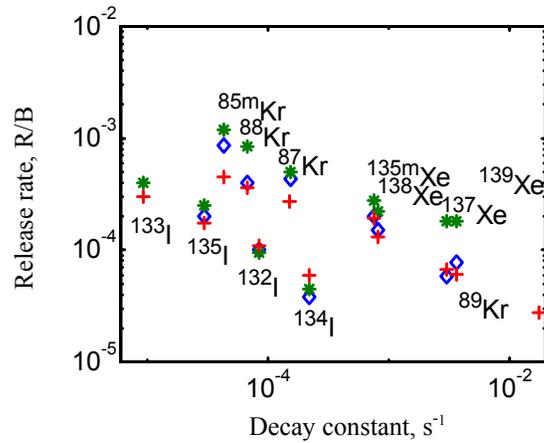


Fig.9. Release rate for different radioactive nuclides from defective fuel rod; heat generation rate 10.36 kW/m, the sixth day of irradiation in Jet Pompe water loop: + – the RTOP-CA calculation, * – data of on-line activity measurements, \diamond – experimental data for activity in water samples.

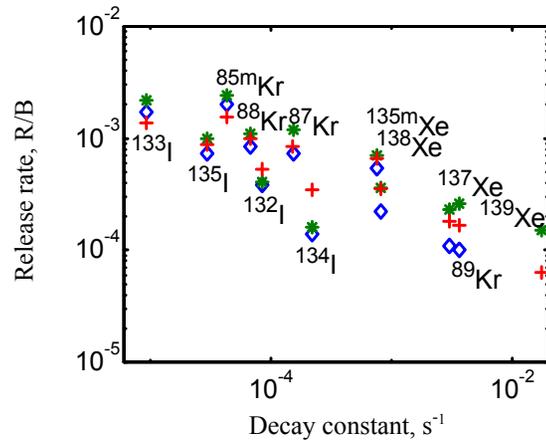


Fig.10. Release rate for different radioactive nuclides from defective fuel rod; heat generation rate 26.3 kW/m, the 16th day of irradiation in Jet Pompe water loop: + – the RTOP-CA calculation, * – data of on-line activity measurements, \diamond – experimental data for activity in water samples.

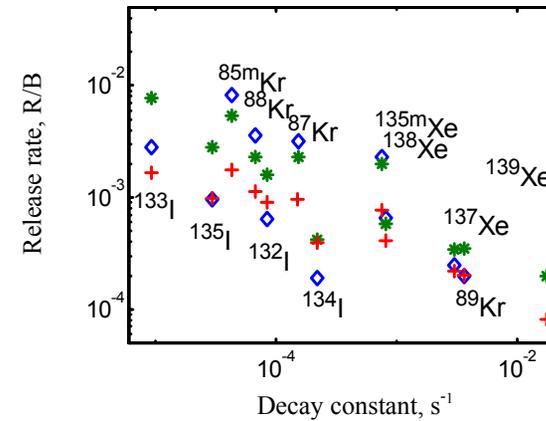


Fig.11. Release rate for different radioactive nuclides from defective fuel rod; heat generation rate 26.3 kW/m, the 19th day of irradiation in Jet Pompe water loop: + – the RTOP-CA calculation, * – data of on-line activity measurements, \diamond – experimental data for activity in water samples.

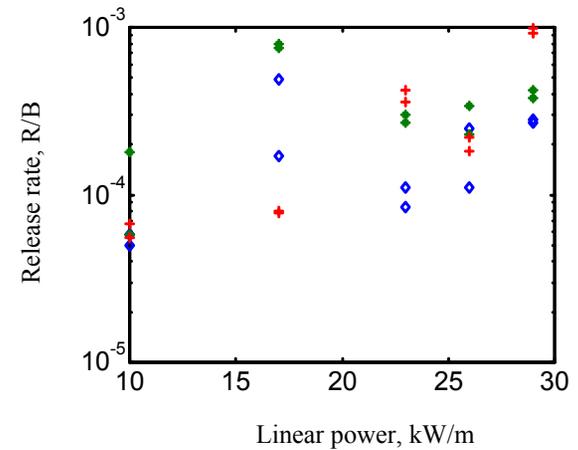


Fig.12. Release rate of ^{137}Xe into coolant as a function of heat generation rate: + – the RTOP-CA calculation, * – data of on-line activity measurements, \diamond – experimental data for activity in water samples.

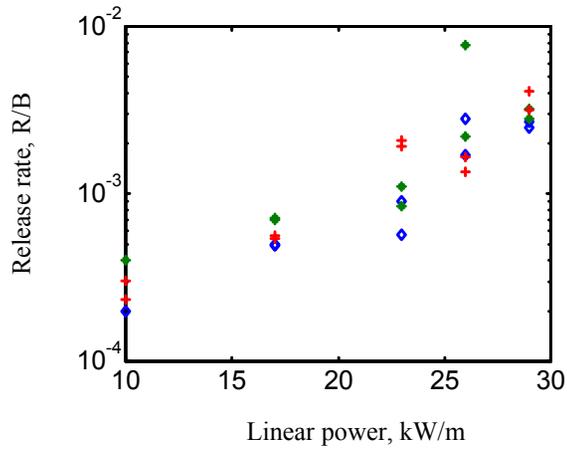


Fig.13. Release rate of ^{133}I into coolant as a function of heat generation rate: + – the RTOP-CA calculation, * – data of on-line activity measurements, \diamond – experimental data for activity in water samples.

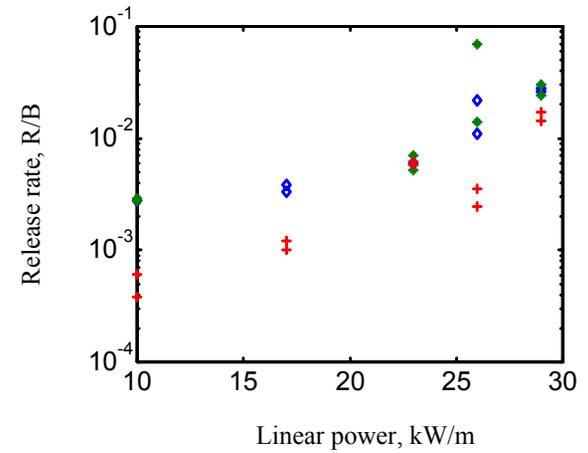


Fig.15. Release rate of ^{131}I into coolant as a function of heat generation rate: + – the RTOP-CA calculation, * – data of on-line activity measurements, \diamond – experimental data for activity in water samples.

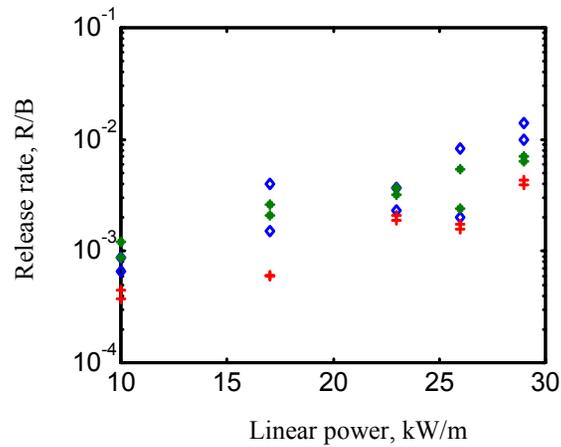


Fig.14. Release rate of $^{85\text{m}}\text{Kr}$ into coolant as a function of heat generation rate: + – the RTOP-CA calculation, * – data of on-line activity measurements, \diamond – experimental data for activity in water samples.

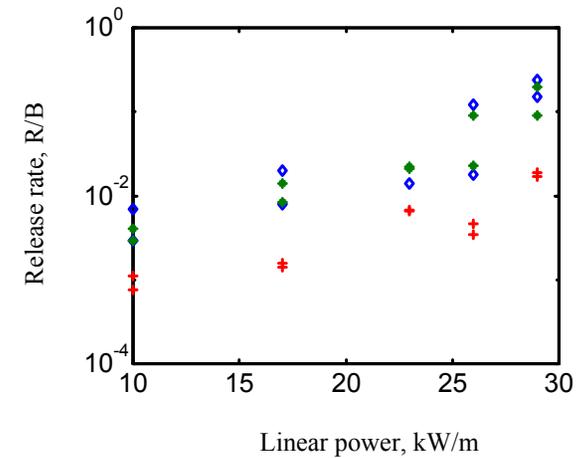


Fig.16. Release rate of ^{133}Xe into coolant as a function of heat generation rate: + – the RTOP-CA calculation, * – data of on-line activity measurements, \diamond – experimental data for activity in water samples.

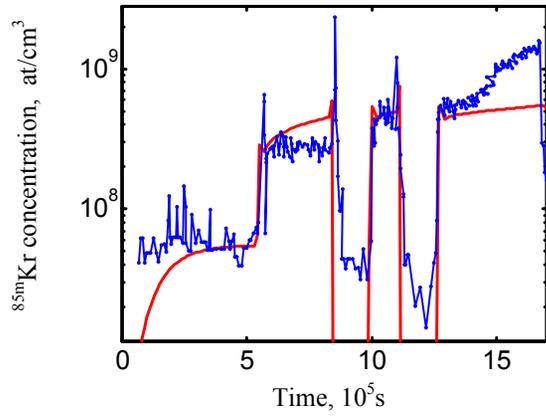


Fig.17. Kinetics of ^{85m}Kr release into coolant at the first stage of irradiation in the EDITHMOX-1 program: —•—•— experimental data; — the RTOP-CA calculations.

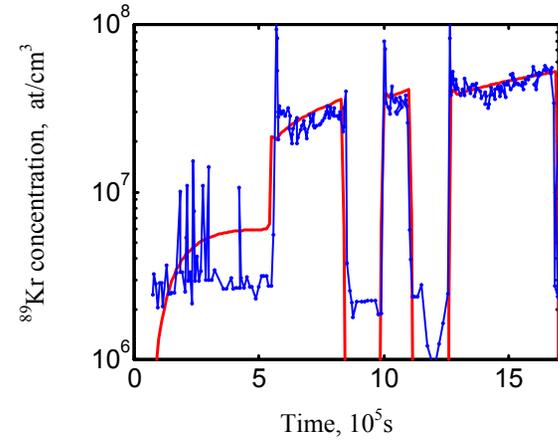


Fig.19. Kinetics of ^{89}Kr release into coolant at the first stage of irradiation in the EDITHMOX-1 program: —•—•— experimental data; — the RTOP-CA calculations.

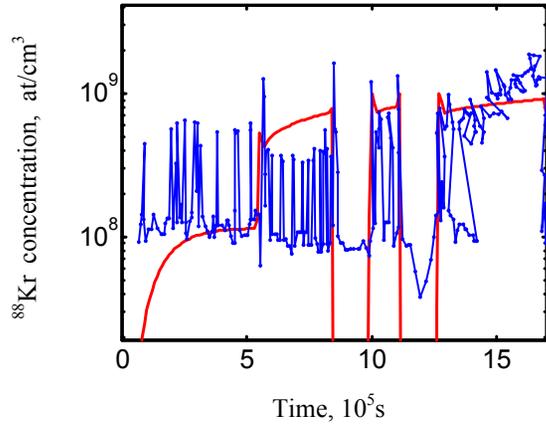


Fig.18. Kinetics of ^{88}Kr release into coolant at the first stage of irradiation in the EDITHMOX-1 program: —•—•— experimental data; — the RTOP-CA calculations.

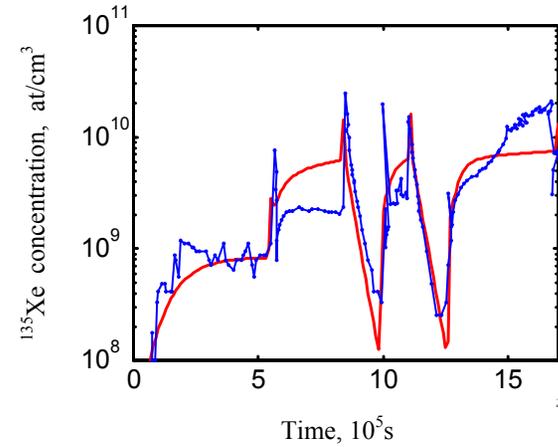


Fig.20. Kinetics of ^{135}Xe release into coolant at the first stage of irradiation in the EDITHMOX-1 program: —•—•— experimental data; — the RTOP-CA calculations.

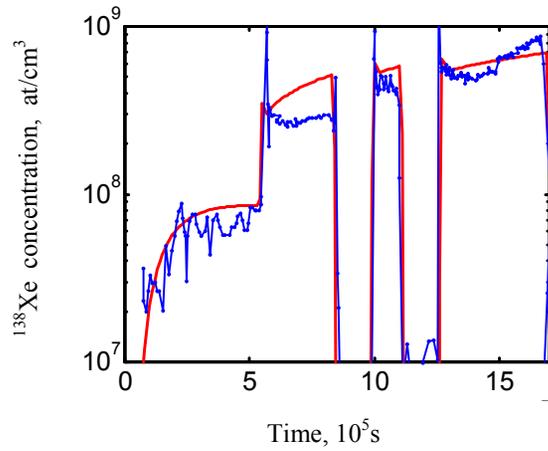


Fig.21. Kinetics of ^{138}Xe release into coolant at the first stage of irradiation in the EDITHMOX-1 program: —•—•— experimental data; — red — the RTOP-CA calculations.

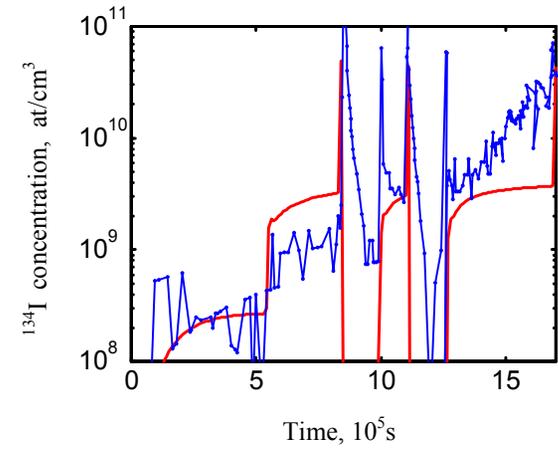


Fig.23. Kinetics of ^{134}I release into coolant at the first stage of irradiation in the EDITHMOX-1 program: —•—•— experimental data; — red — the RTOP-CA calculations.

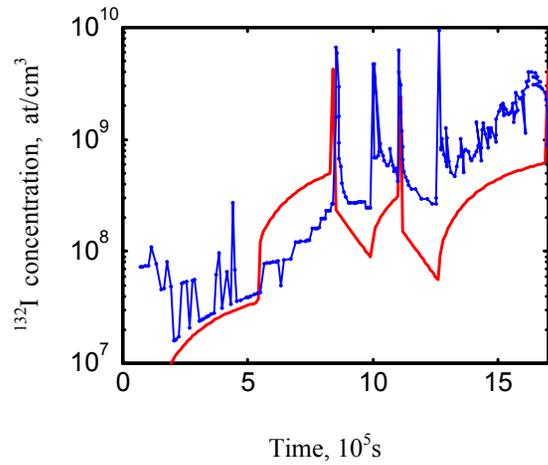


Fig.22. Kinetics of ^{132}I release into coolant at the first stage of irradiation in the EDITHMOX-1 program: —•—•— experimental data; — red — the RTOP-CA calculations.

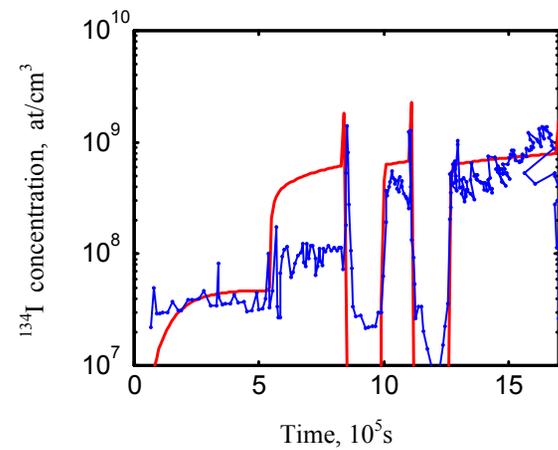


Fig.24. Kinetics of ^{134}I release into coolant at the first stage of irradiation in the EDITHMOX-1 program: —•—•— experimental data; — red — the RTOP-CA calculations.

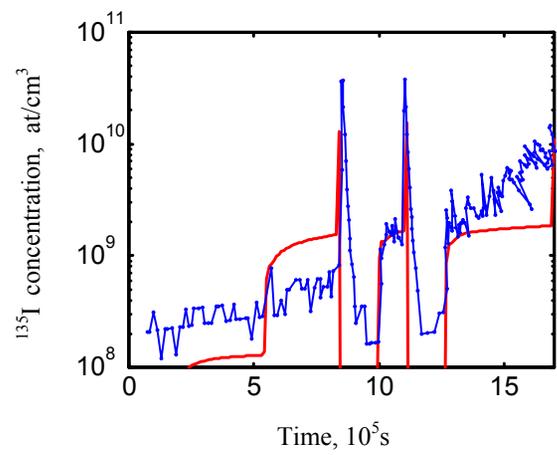


Fig.25. Kinetics of ^{135}I release into coolant at the first stage of irradiation in the EDITHMOX-1 program: —•—•— experimental data; — — the RTOP-CA calculations.