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Nuclear-Hazardous Accumulations of Fuel-Containing Materials in the Destroyed Fourth Unit of the Chernobyl NPP[1]

E. D. Vysotskii, A. A. Klyuchnikov, A. S. Lagunenko*, E. M. Pazukhin, and V. N. Shcherbin

*Institute for Nuclear Power Plant Safety Problems, National Academy of Sciences of Ukraine, ul. Kirova 36-a, Chernobyl, Kiev oblast, 07270 Ukraine; * e-mail: allaguna@mail.ru*

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Abstract—The neutron incident that happened in June 1990 in the 4th Unit of the Chernobyl NPP, destroyed in the accident, at the boundary of rooms 304/3 and 305/2 is analyzed. One of possible scenarios of the incident, consistently accounting for the dynamics and self-extinguishment of the detected neutron anomaly, is suggested.

Keywords: fuel-containing materials, geometry, critical assembly, neutron anomaly, reactivity, cricicality

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In the end of June 1990, detectors of the Finish system recorded a powerful neutron anomaly. The neutron flux density (NFD) in room 304/3 adjacent to the southeastern part of subreactor room 305/2 (site of formation and flow of most of the lava streams) increased by a factor of tens within several days and exceeded 10^4 neutron cm^{-2} s^{-1} in the maximum [1]. The factors that could cause the anomaly were considered by a special commission. Its main conclusion was that the NFD spike was most probably due to a critical incident [2, 3].

In [4, 5] we put forward and substantiated the hypothesis that there are two latent accumulations of fuel-containing masses (FCMs) in the southeastern quadrant of room 305/2. These accumulations can be located in cavities melted by the lava in concrete of the floor in the southeastern part of room 305/2 in the active stage of the accident (Fig. 1). The calculations showed that the uranium content of such lava should be considerably higher than the uranium content of the other products formed by interaction of the nuclear fuel with structural materials of the destroyed unit.

These assumptions were based on the results of thermal measurements, observations in the steam-distribution corridor, results of drilling works, data of instrumental measurements, and analysis of the progress of the anomalous event in 1990.

Further studies performed in 2008 made it possible to create a model of the accumulation directly adjacent to the break (burn-through) between rooms 305/2 and 304/3 (accumulation *1* in Fig. 1a), to substantiate possible causes of its formation, and to conclude on the basis of the neutron calculation results that an FCM accumulation with the maximal fuel concentration exceeding 30% (counting on uranium) is indeed present today in the southeastern part of room 305/2 (zone with high uranium concentration) [6, 7].

There are good grounds to state that water is permanently present in this accumulation. The water surface is on the +9.100 level and higher. Discharge of warmed water is intermittently observed from the southern zone of room 305/2 starting from 1990. The time and intensity of the discharge satisfactorily correlate with the amount of atmospheric precipitations or with the appearance of condensation moisture in the unit [5].

It should be noted that the studies performed in [4–7] do not unambiguously prove the existence of a critical mass zone (CZ) within this accumulation. Therefore, it seems appropriate to consider one of possible versions of the structure of such formation that could account for the anomalous event of 1990. Of course, such model should not contradict the results of previ-

[1] Dedicated to the memory of all Chernobyl liquidators who prematurely passed away. To 25th anniversary of the Chernobyl accident.

ous observations and the existing facts. In this paper we consider such model.

GEOMETRIC AND WEIGHT PARAMETERS OF THE ACCUMULATION MODEL, INITIAL DATA, AND ASSUMPTIONS

The volume of the LFCM massif shown in Fig. 1 is about 270 m^3, and the volume of the southeastern part of the massif is about 170 m^3. As shown in [6, 7], the LFCM massif in room 305/2 contains everywhere unmelted parts of building structures and fragments of the reactor core. It was also shown that the volume of accumulation *1* (Fig. 1) is no less than 16 m^3 and the weight of the fuel in it is no less than 10 t (counting on U) [7]. In calculations of the hypothetic structure of the possible CZ, we made certain assumptions that simplify the model shown in Fig. 1, namely: (1) the geometry was set as a flat cylinder of height 0.9 and diameter 2.5 m, which actually corresponded to the volume of accumulation *1* (Fig. 2) [7]; (2) both homogeneous and heterogeneous structures were considered; (3) the free volume (porosity and fracturing)[2] were set within 25–60% of the total volume of the zone; (4) the major fraction of the lava that left the southeastern part of room 305/2 is in the large horizontal stream starting in room 304/3. Therefore, we assumed that the chemical composition of LFCMs in the zone, except uranium,[3] is identical to that of LFCMs in room 304/3; (5) the density of the porous matrix was calculated additively; (6) the fuel burn-up was assumed equal to 12.5 MW day $(kg\ U)^{-1}$, which would correspond to ^{235}U enrichment of 1.1%, taking into account the produced Pu.

The neutron anomaly was recorded in room 304/3 in the period from June 19 to July 3, 1990. Presumably, it was caused by filling of the melting-through zone in room 305/2 with water. By that time, the temperature on the periphery of the accumulations decreased to 70–80°C, and in the second half of June the amount of precipitations corresponded to the 1.5-month norm. The water ingress into room 305/2 was detected already in April–May, when the neutron detectors arranged in wells that were drilled in the lower reinforced-concrete floor of room 305/2 from the west in the region of the +9.000 mark were poured over with water and failed [5]. Today this floor is termed subreactor plate.

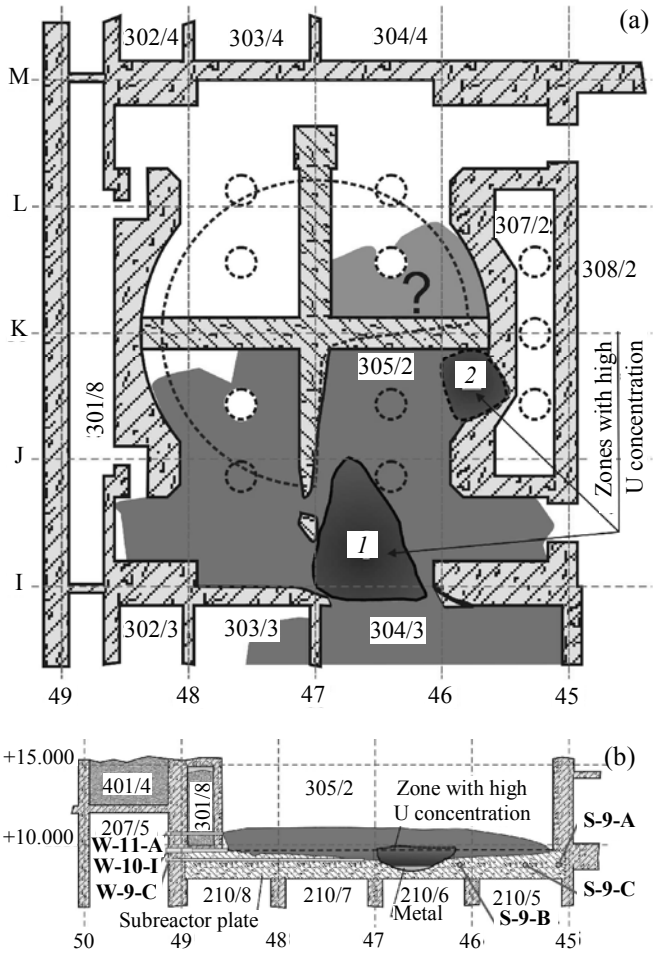

Fig. 1. Massif of lava-like fuel-containing masses (LFCMs) in room 305/2: (a) location of zones with high uranium concentration on +9.700 level and (b) section of the LFCM massif along I_{+1650} row. The well designations are printed bold.

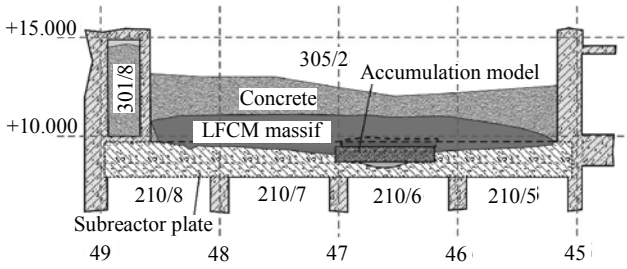

Fig. 2. LFCM massif in room 305/2 and position of the model of the possible critical mass zone.

Water–uranium systems with low-enrichment fuel are characterized by a narrow range of optimal water content when the system occurs between the first and

[2] Such a structure can be obtained, e.g., in the course of synthetic glass making by quenching of the melt in the step of vigorous gas release.

[3] Uranium was introduced additively.

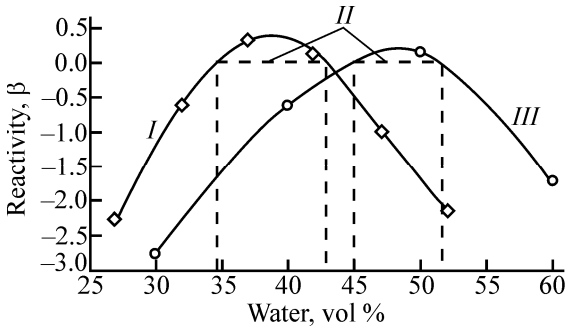

Fig. 3. Reactivity (δK in units $\beta = 0.0065$) in the optimal moistening region at 27°C: (*I*) heterogeneous structure, (*II*) optimal moistening region, and (*III*) homogeneous structure.

second critical levels. Therefore, it can be assumed with high probability that, on filling with water, the dry accumulation reached the first critical level and, under the conditions of water supply, remained in the near-critical state for at least 6 days (June 27–July 2, 1990). With further water supply, on reaching the second critical level, the system became subcritical and underwent self-extinguishment. The mechanism of retention of the system in the near-critical state was associated both by filling with water of the porous structure of the accumulation, which had high temperature, and by the negative temperature coefficients of the reactivity.

Below are the results of simulation and determination of the parameters of critical assemblies (fuel–moderator–reflector) whose geometry fits the simulation zone (Fig. 2).

SOME RADIOCHEMICAL AND NEUTRON-PHYSICAL CHARACTERISTICS OF ACCUMULATION MODEL

The results of radiochemical and γ-ray spectrometric analyses of the most representative samples taken from the accumulation zone that is simulated in this study are given in Table 1.

The neutron-physical characteristics of the accumulation were estimated on the basis of the results of simulation of critical assemblies corresponding to the volume of the cylindrical region under consideration. The effective multiplication factor (K_{eff}) was calculated using MCNP-4A program complex. In criticality calculations, we took into account the interactions of neutrons with FCMs and thermalization of neutrons in water. The calculation results are given in Fig. 3.

The reactivity calculations were performed for the multiplicating medium at room temperature (20°C). With an increase in temperature, the reactivity and water volume margins in the optimal moistening range decrease because of the effect of negative Doppler ($K_D \sim 2 \times 10^{-3}$ β/°C) and thermal expansion ($K_t \sim 3 \times 10^{-2}$ β/°C) temperature coefficients.

The geometric and mass parameters of versions of critical assemblies with the homogeneous and heterogeneous structures of multicomponent FCM composition are given in Table 2.

Table 3 shows how K_{eff} depends on the volume content of water in the assembly.

The dry assembly is deeply subcritical ($K_{eff} < 0.5$). The supply of water brings about positive reactivity, and K_{eff} increases. The assembly reaches the first criticality level and remains supercritical in the range of optimal moistening until the second critical level is reached. With the further supply of water, the assembly becomes subcritical, and the subcriticality level is restricted by the water volume that the system can take up on reaching the second critical level. At a limited volume of the assembly, higher the subcriticality at overmoistening can be reached owing of an increase in the fuel concentration. The optimal moistening range depends on the reactivity margin of the assembly and in our case is 8% of the assembly volume for the heterogeneous system and 6% of the volume for the homogeneous system.

NEUTRON ANOMALY AS CRITICAL INCIDENT

An all-wave neutron detector (ND) arranged in room 304/3 on the LFCM surface and separated from the accumulation in room 305/2 by more than a 2-m layer of black LFCMs showed an increase in count rate since June 19, 1990. Until June 27, the increase was relatively monotonic, and the background was exceeded by a factor of more than 2. On June 27, 1990, in the interval from 1 to 3 pm the count rate of the detectors increased by a factor of more than 3, after which it slightly decreased. This was followed by the next jump, in which the count rate increased by a factor of 2 in the period from 9 to 11 am. In the jump periods, the neutron flux density was measured for control with a "bare" detector (without Plexiglas cover). The results of control measurements fully correlated

Table 1. FCM samples taken at the boundaries of zones with a high uranium content from wells drilled in the subreactor plate of room 305/2. Radionuclide composition of samples

Coordinates of sampling point, well	Nuclide activity, MBq g^{-1} (sample), recalculated on April 26, 1986										U, %	Burn-up, MW day (kg U)$^{-1}$	
	^{144}Ce	^{137}Cs	^{134}Cs	^{106}Ru	^{125}Sb	^{154}Eu	^{238}Pu	$^{239}Pu + ^{240}Pu$	^{242}Cm	^{244}Cm	^{241}Am		Cs data	Pu data
47, I_{+3000}, +9.100, W-9-F	1.1×10^3	1.5	6.3×10^{-1}	7.1×10	–	–	–	–	–	–	–	4.6	8.83	–
47_{-700}, J_{-1000}, +9.100, W-9-G	1.0×10^3	2.6×10	1.3×10	1.7×10	2.8×10^{-1}	1.5	–	–	–	–	–	4.1	10.7	–
	8.9×10^2	2.5×10	1.2×10	1.3×10	2.4×10^{-4}	1.4	–	–	–	–	–	3.5	10.7	–
	8.0×10^2	2.6×10	1.3×10	1.1×10	1.6×10^{-1}	1.6	–	–	–	–	–	3.2	11.2	–
	6.9×10^2	1.9×10	9.2	1.1×10	6.3×10^{-2}	1.1	–	–	–	–	–	2.3	10.6	–
47_{-700}, J_{-1000}, +9.100, W-9-G	8.8×10^2	2.6×10	1.3×10	4.0×10	2.5×10^{-1}	1.5	–	–	–	–	–	3.5	11.1	–
47_{-500}, J_{-2000}, +9.100, W-9-J	1.6×10^2	4.7	2.8	9.7×10^2	2.9	3.3×10^{-1}	–	–	–	–	–	0.6	13.2	–
	4.4	7.9×10^{-1}	4.6×10^{-1}	1.2	1.9×10^{-2}	2.4×10^{-2}	–	–	–	–	–	0.02	13.0	–
	1.1×10^3	2.9×10	1.7×10	2.4×10	–	2.2	–	–	–	–	–	4.2	12.5	–
	1.3	3.5×10^{-2}	2.2×10^{-2}	1.7×10^{-1}	2.5×10^{-2}	2.5×10^{-3}	–	–	–	–	–	0.01	13.9	–
47_{-1700},J_{-2000}, +9.100, W-9-J	1.2×10^3	2.9×10	1.7×10	3.0×10	3.9×10^{-1}	2.1	–	–	–	–	–	4.7	12.5	–
	1.3×10^3	4.7×10	2.5×10	1.7×10^2	1.8×10	2.2	–	–	–	–	–	5.3	11.4	–
	1.3×10^3	3.2×10	1.9×10	1.9×10	7.7×10^{-1}	2.2	–	–	–	–	–	4.9	13.2	–
47_{+400}, J_{-1000}, +9.100, W-9-G	2.6×10^2	1.3×10	7.6	2.5	7.9×10^{-1}	5.1×10^{-1}	7.9×10^{-2}	1.5×10^{-1}	2.7	1.8×10^{-2}	9.7×10^{-3}	6.2	12.7	13.4
47_{-700}, J_{-1000}, +9.100, W-9-G	7.8×10^2	–	–	–	–	–	2.5×10^{-1}	6.5×10^{-1}	5.7	3.8×10^{-2}	3.6×10^{-2}	–	–	11.3
47_{-900}, J_{-1000}, +9.100, W-9-G	5.4×10^2	–	–	–	–	–	2.8×10^{-1}	5.8×10^{-1}	6.3	4.8×10^{-2}	3.3×10^{-2}	–	–	12.8
47_{-1700}, J_{-2000}, +9.100, W-9-J	1.3×10^3	2.9×10	1.6×10	1.3×10	3.1	2.0	3.5×10^{-1}	7.5×10^{-1}	1.4×10^{-1}	9.8×10^{-2}	5.6×10^{-2}	8.8	11.6	12.5
46_{+2600}, L_{500}, +9.300[a], W-9-T	9.9×10^2	5.4	3.2	–	–	1.0	5.2×10^{-1}	1.0	2.2	1.2×10^{-1}	7.4×10^{-2}	5.7	13.1	13.0

[a] Base of the wall between rooms 304/3 and 305/2.

Table 2. Mass parameters of critical assemblies

Structure	Cylinder volume, m^3	FCM volume, m^3	FCM, t	UO_2, t	U, t	^{235}U, kg
Homogeneous	17.7	7.07	33.6	22.9	20.2	231.8
Heterogeneous	17.7	8.5	35.8	21.1	18.6	213.4

Table 3. Dependence of K_{eff} of assemblies on the volume content of water

Structure	Parameter	Dry assembly	First criticality	Reactivity margin	Second criticality	Maximal subcriticality
Heterogeneous	Water volume %	0	35	39	43	52
	K_{eff}	0.427	1.0	1.0026 (0.41 β)	1.0	0.985
Homogeneous	Water volume %	0	45	48	51	60
	K_{eff}	0.438	1.0	1.0018 (0.28 β)	1.0	0.989

with the readings of the all-wave detector. The count rates for both detectors differed insignificantly. It is known that the sensitivities of the "bare" and all-wave detectors become similar only when the neutron spectrum is moderated to the thermal level. Up to 11 am on June 29, 1990, the count rate slightly increased, which was followed by a sharp increase, and by 11 pm the count rate exceeded the background level by a factor of 60. After pouring a 1% gadolinium nitrate solution into room 304/3 at 23:37 on June 29, 1990 and at 00:50 on June 30, 1990, the count rate decreased by a factor of more than 7 over a period of 2 h, and after 35 h it returned to the background level [1]. This fact indicates that Gd scavenged thermal neutrons only in the region of their escape from the accumulation. Otherwise, the background count rate of ND should decrease, because blocking of the ND with gadolinium is equivalent to the effect of a cadmium shield. Therefore, we have reconstructed the dynamics of the anomaly on the level of the ND count rate before adding Gd. The neutron spectrum in the optimal moistening region does not change, and the ratio of thermal and fast neutrons in the flux remains constant.

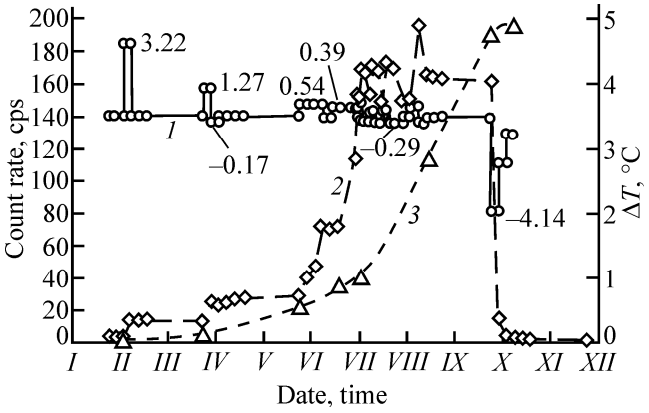

Fig. 4. Dynamics of the (*1*) introduced reactivity ($\beta \times 10^3$), (*2*) ND count rate, and (*3*) fuel temperature elevation ΔT in the course of the critical incident. Date and time: (*I*) 6/27/1990, 0:00; (*II*) 6/27/1990, 12:00; (*III*) 6/28/1990, 0:00; (*IV*) 6/28/1990, 12:00; (*V*) 6/29/1990, 0:00; (*VI*) 6/29/1990, 12:00; (*VII*) 6/30/1990, 0:00; (*VIII*) 6/30/1990, 12:00; (*IX*) 7/1/1990, 0:00; (*X*) 7/1/1990, 12:00; (*XI*) 7/2/1990, 0:00; and (*XII*) 7/2/1990, 12:00.

Figure 4 shows the reconstructed dynamics of the ND count rate in room 304/3 in the period of the neutron anomaly.

It can be assumed with high probability that the anomaly was caused by flooding of accumulation *1* (Fig. 1), and actually it reproduced the variation of the flux density of the escaping neutrons in the course of reactivity introduction with the water supply into the "dry" accumulation. The rate at which the accumulation is filled with water and, correspondingly, the rate of the reactivity introduction (positive and negative) are determined by the FCM porosity. The water supply into a hot (>100°C) porous structure leads to steam generation at the wetting front and partial repulsion of water, which sharply restricts the rate of the reactivity introduction. The power released with an increase in the number of fission events is spent for heating of the fuel and water. As a result, a competing process arises: introduction of negative reactivity owing to Doppler effect (K_D) and thermal expansion (K_t). At the same time, cold water entering into the accumulation decelerates the temperature elevation in the bulk of the accumulation and the local temperature of the fuel in the accumulation core. Hence follows that the dynamics of the development of the self-sustained chain reaction

Table 4. Parameters of the critical incindent

Parameter	Homogeneous structure	Heterogeneous structure	Notes
Neutron flux density φ, neutron cm^{-2} s^{-1}	5.6×10^7	5.6×10^7	At maximal ND count rate
Fission intensity in power keeping step, fissions per second	3.81×10^{13}	3.48×10^{13}	In step of power keeping
Total number of fissions	5.17×10^{18}	4.7×10^{18}	92 h
Energy release, MJ	172	157	
Intrinsic background, neutron s^{-1}	1.65×10^8	1.52×10^8	
Neutron multiplication	$>10^5$	$>10^5$	
Positive reactivity introduced, $10^{-3}\beta$	0.39–3.22	0.39–3.22	At process development
Negative reactivity introduced, $10^{-3}\beta$	0.17–4.14	0.17–4.14	At self-extinguishment
Optimal moistening range, vol % water	0.12	0.16	Total water volume 10 m^3
Mean temperature of accumulation, °C	70–80	70–80	During incident
Reactivity margin of accumulation, β	>1.5	>1.5	At 20°C

and the level of the supercriticality parameters attained (NFD, number of fission events, temperature) are determined by the ratio of these competing processes.

The current value of the accumulation reactivity is determined by the sum of the contributions from the change in the water amount and from an increase in the temperature of the fuel and accumulation as a whole:

$$\delta K_\Sigma = \delta K(H_2O) - K_D \Delta T_1 - K_t \Delta T_2,$$

where T_1 is the fuel temperature and T_2, accumulation temperature, °C. The rate of the increase and decrease in the power depends on the sign and value of the resultant δK_Σ in each step of the incident.

Figure 4 shows on the time scale the characteristics of the steps of the development, keeping on the power, and self-extinguishment of the critical incident. The amount δK_Σ of the introduced reactivity was determined from the time derivative of the ND count rate in each step of the increase and decrease in the power. The parameters determining the temperature growth were estimated for critical assemblies simulating the accumulation (Table 1).

In each step of the development, in each power jump, the accumulation passed into the supercritical state with the positive reactivity determined by the current fuel temperature. Steam generation at the wetting front caused drop of the positive reactivity as a result of water repulsion. After each drop, the time during which the power was kept on a constant level was determined by the competing processes: introduction of the positive reactivity with the return of the wetting front and introduction of the negative reactivity (mainly due to Doppler effect) with an increase in the fuel temperature.

With the supply of water and increase in temperature, the positive reactivity margin decreases, and the optimal moistening (supercriticality) range becomes narrower. In Fig. 4, the reactivity introduced at power jumps decreases and the intervals of its keeping constant become longer. The development of the process stopped when the amount of water in the accumulation exceeded the optimal moistening volume and the propagation of the wetting front became accompanied by introduction of the negative reactivity. Now the water repulsion in the course of steam generation at the wetting front returned the system to the supercritical state, and the water supply returned it to the subcritical satet.

The step when the power remained constant and temperature gradually increased lasted for 35 h when the overmoistening and negative temperature coefficients of reactivity ceased to be compensated by water discharges accompanying steam generation.

In Table 4 we present the supercriticality parameters interpreted for critical assemblies that simulate the accumulation. The calculated NFD attenuation factor in the 2-m layer of black LFCMs separating the ND and source was 2×10^4. The maximal NFD value of 10^4 neutron cm^{-2} s^{-1} was estimated from the ND count rate, assuming deformation of the neutron spectrum to the thermal spectrum [1]. Therefore, the NFD value of 5×10^7 neutron cm^{-2} s^{-1} was taken as the mean value in the step of keeping of the maximal power in the course of the incident.

The temperature of the flooded accumulation by the start of the incident was 70–80°C and was maintained by the residual heat release (RHR) from the spent nuclear fuel. By now, the accumulation temperature decreased to 30–40°C, and it continues to decrease in accordance with a decrease in RHR. Under the conditions of overmoistening, this factor leads to an increase in subcriticality and competes with the Doppler effect. At the subcriticality due to overmoistening estimated at less than 2β, only a decrease in the water volume in the accumulation and an increase in the temperature can lead to a critical incident, but the mechanisms of its development will be different, and always with slow kinetics.

Thus, we have considered a hypothetical version of the structure of a latent LFCM accumulation at the break (burn-through) between rooms 305/2 and 304/3. The nuclear fuel concentration in this accumulation should be no less than 30% (counting on U). The revealed accumulation always contains water, and its level is at a height mark of +9.100 or above. The neutron calculations that we performed showed that this hypothesis of the structure of the latent LFCM accumulation satisfactorily accounts for the dynamics of the anomalous event in June 1990 and allows this event to be classed as critical incident. The generally accepted conservative approach to nuclear safety problems, as applied to analysis of the condition of the latent possible lava accumulation in room 305/2, gives grounds to state that today this structure is a potentially hazardous critical mass zone requiring systematic monitoring and further in-depth studies to ensure the nuclear and radiation safety of the Shelter.

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