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IN-DEPTH ANALYSIS OF ACCIDENTAL CRITICALITY IN A REPROCESSING PLANT

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ABSTRACT

An in-depth-analysis including probabilistic considerations has been performed for a potential criticality excursion in two large vessels in a planned reprocessing plant. Criticality safety of these components is based on limitation of uranium and plutonium concentration. The main intention of this study was to investigate the potential and probable magnitude of a criticality in greater detail and to detect possible weaknesses of criticality safety provisions. The results of the analysis show, that the calculated probability of less than $5 \cdot 10^{-4}/a^{a}$ for a criticality excursion in the most relevant rework tank may be further reduced. The peak power rework tank may be further reduced by design modifications. The peak power and total fissions of the critical excursion as assessed in a preceding analysis for licensing are conservative by factors of 10 and 4, respectively.

1. INTRODUCTION

In the Federal Republic of Germany an accidental nuclear criticality represents a design basis accident in the licensing procedure for fuel cycle facilities. Due to the requirements of the Radiation Protection Ordinance it has to be demonstrated, that the potential radiological consequences of a design basis accident shall not exceed certain dose limits. The most relevant dose limits in case of a criticality accident are a maximum permissible whole body dose of 50 mSv and a limiting thyroid dose of 150 mSv at the most exposed point outside the fuel cycle facility. These dose limits are valid for the summation of dose contributions from all relevant pathways including ingestion of contaminated food and water.

For the evaluation of a credible accident scenario and the calculation of potential radiological consequences of a criticality there are no specific regulations or standards in Germany. To establish assumptions for the accident scenario and to determine possible consequences of the accident USNRC-Regulatory Guides 3.33 - 3.35 give some support /1/. A plant-specific accident analysis is, however, necessary to determine the most important parameters for the accident analysis.

In case of the licensing procedure for the Wackersdorf reprocessing plant a plant-specific accident analysis has been performed on the premise, that this analysis should rely on conservative assumptions to ensure results to be on the safe side. In this analysis a deterministic approach has been used to evaluate upper bounds for peak power and total number of fissions in the nuclear excursion and to calculate the radiological consequences. To get more insight into the complex phenomena during a criticality accident and to determine the degree of conservatism of the deterministic accident analysis in the licensing procedure an additional in-depth analysis has been performed taking also into consideration probabilistic elements.

2. DETERMINISTIC CRITICALITY ACCIDENT ANALYSIS

The analysis of a criticality accident in the Wackersdorf reprocessing plant, which includes also a fabrication of mixed oxide LWR fuel elements, has been performed in two steps. Step 1 consisted of a systematic analysis of the equipment and the provisional operation of the plant to find out possible points and situations for a criticality accident. An important result of this analysis was, that possibilities for a criticality seem only conceivable for a chain of independent events exceeding the generally applied double contingency principle. Undetected accumulation of plutonium and incorrect transfer or spill of concentrated fissile solution have been found to be the most important initiating events. In a second step the most relevant accident scenarios probably causing the most severe consequences have been derived from accidental possibilities identified in step 1. Main criteria for this deviation have been the type and concentration of the fissile material present in the component, the shape and volume

[a] per annum.

of the component, connections to other equipment, provisional transfer volumes and rates, possible release paths to the environment and the planned retention systems in the off-gas systems. As the most relevant locations for a criticality excursion the feed storage tank with 16.8 m^3 volume and a large rework tank with 12.5 m^3 volume have been identified. Both vessels are designed for limiting concentration of fissile material. Existing experience from criticality accidents and excursion experiments has been evaluated to generate the parameters for a conservative assessment of a criticality excursion representative for accidental conditions in the plant under worst conditions. In addition calculations with a foreseeable computer program have been performed to model the time-dependent behaviour of a criticality excursion /2/. The most important results of these evaluations are presented in Table 1.

The calculations of the time-dependent behaviour of the criticality excursion in a large volume of concentrated fissile solution showed, that 10^{19} total fissions are likely to be reached until the solution starts boiling. When boiling is reached, the power level drops drastically. Subsequently the further increase of the total fissions is not remarkable beyond that number of fissions reached at the beginning of boiling. An example of this behaviour is demonstrated in Fig. 1.

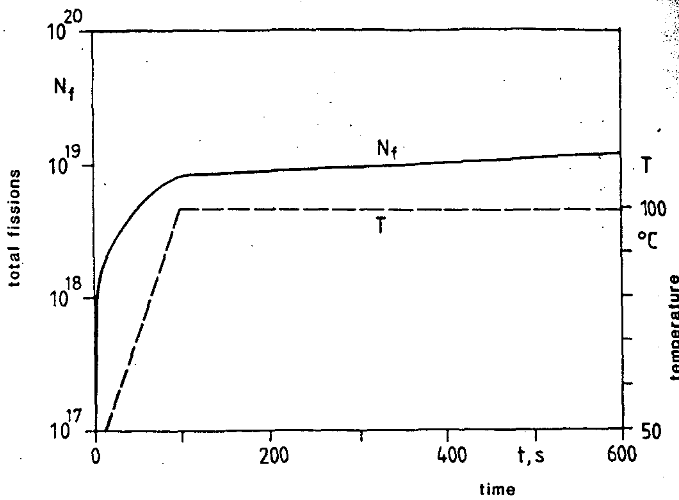

Figure 1: Total fissions and temperature for a criticality excursion in a solution

An important result of these calculations was, that even for high transfer rates of solution and high plutonium concentrations the assessed pressure for peak power was relatively low. Mechanical damage to the components therefore is unlikely. Calculations of a possible pressure propagation into the off-gas-system showed that at least two end stages of HEPA-filters will remain undamaged, because the ductwork and large components in the off-gas-system including large wash columns provide an effective suppression.

The calculations also demonstrated that a high number of total fissions should only be possible if the critical state remains stable

Table 1: Important parameters of the deterministic criticality accident analysis for the Wackersdorf reprocessing plant

	Reprocessing Main Building	MOX Fuel Fabrication
Location of accident	Feed storage vessel or rework tank	Wet Pu-conversion
Initiating event	Transfer to unsafe geometry, Too high concentration, Accumulation of Pu	Inproper batching or transfer to unsafe geometry
Volume of vessel	16.8 m^3, 12.5 cm^3	
Fissile material	Pu-nitrate-solution, 1000 l with 50 g Pu/l	Pu-nitrate-solution, 50 l with 200 g Pu/l or moderation of 100 kg PuO_2-powder
Peakpower fissions/s	$1.4 \cdot 10^{19}$	10^{19}
Total fission	10^{20}	$5 \cdot 10^{18}$
Duration of criticality	30 - 90 min	1 min; 30 min

for a longer period of time. In this case precautions have to taken to prevent a wetting of the off-gas filters by steam or condensation of water. The calculated radiation doses caused by fission products released to the environment via the stack remain lower than the limits set by the Radiation Protection Ordinance for all kinds of exposed organs.

Neutron and gamma direct radiation will be effectively shielded by the thick concrete walls of the process cell.

3. IN-DEPTH ANALYSIS

An in-depth-analysis has been performed independently from the accident analysis carried out in the framework of the licensing procedure to investigate and quantify the possibilities of a critical excursion in the feed storage vessel and the rework tank. Fig. 2 shows the principal arrangement of these two components.

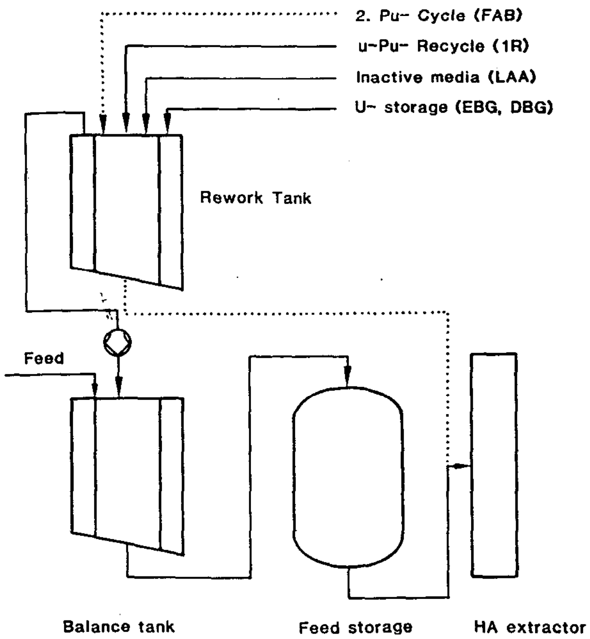

Figure 2: Principal arrangement of feed storage and rework tank

The large cylindrical feed storage tank has the task to store the balanced feed solution before transfer to the first extractor (HA-column). The feed solution resulting from the headend process is balanced and controlled for fissile material concentrations in a preceding balance tank designed as an annular slab. The maximum permissible concentrations are 360 g heavy metal (U+Pu)/l, 5 g Pu/l, the residual enrichment of uranium is max. 1.8 % U-235. The 12.5 m^3 volume rework tank is an annular tank of the same shape and volume as the balance tank. The plutonium concentration in the rework tank is restricted to maximum 5 g Pu/l. The rework tank exhibits a lot of incoming connections to other systems and two exits to the balance tank or directly to the HA-extractor. From a criticality safety standpoint the most important connections are from the 2 Pu-extraction cycle, the U-Pu-recycle and from the uranium product storage.

Actually some specific details of plant operation and of the design of components and safety devices are not yet stated. In addition reliability data for many relevant safety systems or operational procedures are scarce or lacking. Limited reliability data are available from operation of the Karlsruhe reprocessing pilot plant and from similar chemical plants. Therefore for the failure tree analysis the applied probabilities for initiating events and system or human failure can only be estimated as preliminary data. Since the intention of the in-depth-analysis was primarily to compare various sequences of events and to detect possible weak points, not to get exact probabilistic numbers, this limited data base should be sufficient for this purpose. Tables 2 and 3 present important assessed probabilities for initiating events and relevant failures.

Table 2: Assessed probabilities for initiating events

No detection of incorrect fuel element data card	10^{-1}/a
Failure of fuel element monitor before dissolution	10^{-2}/a
Dissolution of fresh fuel element	10^{-3}/a
Loss of cooling for more than 24 h, Feed storage tank	$< 10^{-3}$/a
Addition of wrong chemicals with precipitation or phase separation of fissile material, rework tank	10^{-1}/a
Too concentrated Pu-solution at the entrance of 1R-Concentration (>5 g Pu/l)	10^{-1}/a
Too high Pu-concentration in 1BOP-flow in 2.Pu-cycle (> 5 g Pu/l)	10^{-1}/a
(> 12.5 g Pu/l)	10^{-2}/a
Unexpected accumulation of Pu (10 kg)	$< 10^{-3}$/a

Table 3:
Assessed probabilities for failures

Temperature measurement	10^{-1}
Neutron monitor	10^{-1}
Evaporator control and control of limiting concentration	$5 \cdot 10^{-3}$
Measurement of Pu-concentration	10^{-2}
Double measurement of Pu-concentration, formal procedure with key valve for authorization of transfer	10^{-3}
Emptying, dilution	10^{-2}
Control of fissile material by fissily material accountancy (10 kg Pu)	10^{-3}

3.1 FEED STORAGE TANK

For the feed storage tank the following three relevant initiating events have been identified to represent a potential criticality risk:

- Dissolution of fresh fuel elements
- Loss of cooling with subsequent concentration of fissile material
- Accumulation of fissile material by physical of chemical effects.

To secure the dissolution only of spent fuel elements and to limit the concentration of fissile material the following series of checks and measurements is foreseen:

- Check of fuel element data card
- Measurement of the residual content of fissile material in the fuel element by neutron interrogation
- Double measurement of fissile material concentration, residual U-235-enrichment and plutonium isotopic composition in the fuel solution
- A second double measurement of fissile material concentration after feed clarification by a centrifuge.

Fig. 3 shows this series of controls and the specifications, which have to be met for fissile material (Table 4).

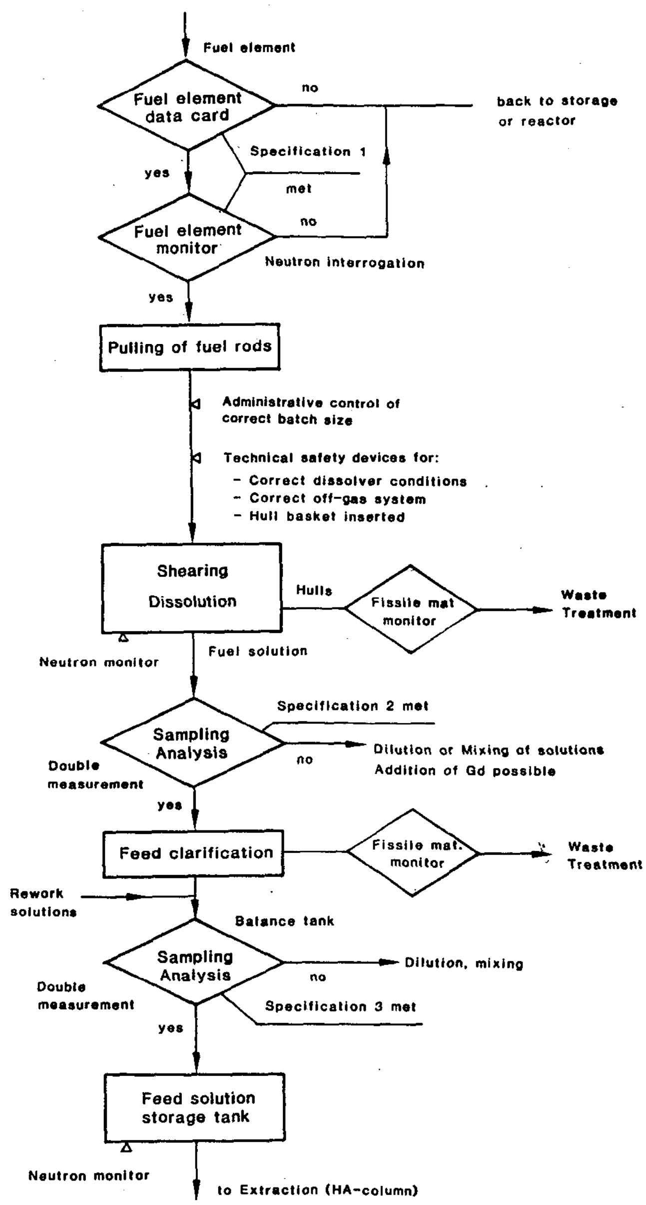

Figure 3: Control of parameters relevant for criticality safety in headend and feed clarification

The investigation of the failure tree reveals that because of those many controls the possibility of a criticality in the feed storage tank as a result of the dissolution of fresh fuel elements is very low (Fig. 4). The dissolver itself is safe by geometry and a hafnium dissolver basket even for fresh fuel.

Tab. 4: Specifications for fuel elements, fuel solution and feed solution

Specification 1 Fuel Elements	UO_2	MOX
Initial enrichment max.	4 % U-235	3.9% Pu-fiss in U(nat), max. 81.4% Pu-239 + Pu-241 in total Pu
Residual enrichment max.	1.8 % U-235	0.5% U-235, 2.6% Pu-fiss max. 77% Pu-239 + Pu-241 in total Pu

Specification 2	Fuel Solution	
U + Pu-concentration	max.	480 g/l
Pu-concentration	max.	11.6 g/l
U-235-enrichment	max.	1.8% U-235
Pu-isotopic composition	max.	77% Pu-239 + Pu-241

Specification 3	Feed Solution	
U + Pu-concentration	max.	360 g/l
Pu-concentration	max.	5 g/l
U-235-enrichment	max.	1.8% U-235
Pu-isotopic composition	max.	77% Pu-239 + Pu-241

Similar investigations have been made for loss of cooling and accumulation of fissile material. In the case of loss of cooling the possibility to reach criticality is very remote, if real geometry of the tank and the presence of fission products is taken into account. Similar considerations are valid for undetected accumulation of fissile material. A criticality risk is conceivable only for a separation of uranium with low enrichment and plutonium. Calculated cumulative probabilities for a criticality in the feed storage tank are:

- Dissolution of fresh fuel elements $1.2 \cdot 10^{-7}$/a
- Loss of cooling $< 1.1 \cdot 10^{-5}$/a
- Accumulation of plutonium $< 1.2 \cdot 10^{-6}$/a

3.2 REWORK TANK

In comparison to the analysis for the feed storage tank the failure trees for the rework tank and the conditions for a criticality are far more complex. Whereas an accidental transfer of concentrated uranium product back to the rework tank represents no criticality problem (U-235-enrichment ≦ 1.8 %) solutions with relatively high plutonium concentrations may be tranferred from the 2. Pu-cycle or the U-Pu-concentration (1 R). If dilute rework solutions or solutions containing low enriched uranium are present in the tank, critical conditions cannot be reached even for large accidental transfers of plutonium solution into the rework tank. If the rework tank is completely or nearly empty, criticality may be possible. Fig. 5 shows typical results for transfers of plutonium solutions with 12.5 and 50 g Pu/l.

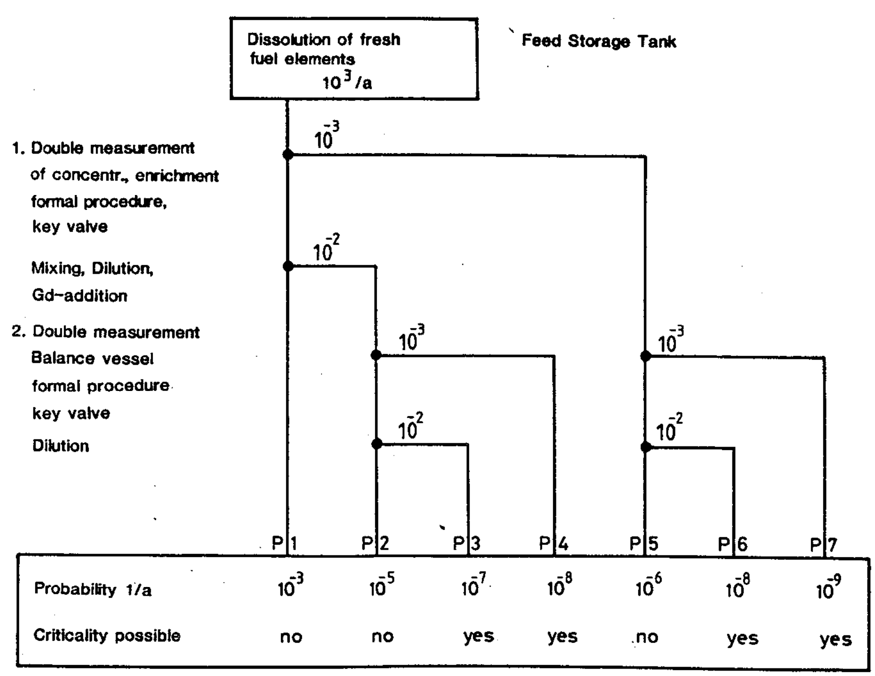

Figure 4: Probabilities for a criticality in the feed storage tank as a result of dissolution of fresh fuel elements.

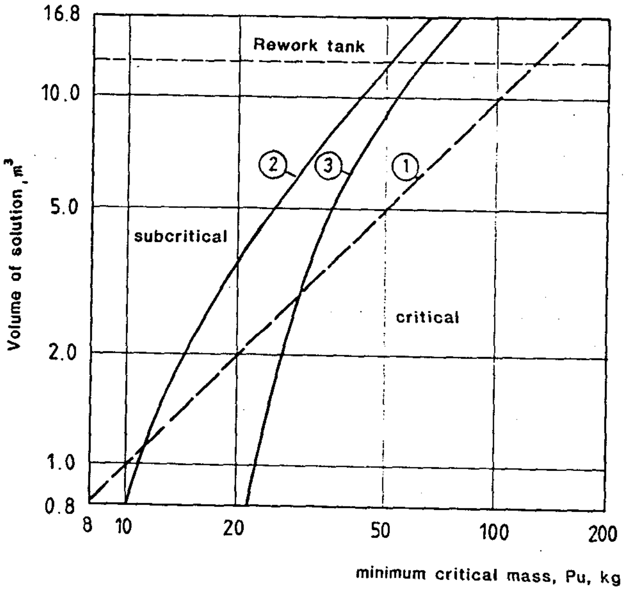

Figure 5: Condition for criticality in the rework tank

1. Minimum critical mass for minimum critical plutonium concentration (10.2 g Pu/l; 20% Pu-240)
2. Actual critical mass including 10 kg Pu from accidental transfer
3. Actual critical mass including 20 kg Pu from accidental transfer

Fault tree analysis has been performed for the connections from the 2. Pu-cycle and the 1R-concentration to the rework tank. The cumulative probabilities for a criticality in the rework tank have been calculated for accidental plutonium concentrations of ≦ 12.5 g Pu/l and ≦ 50 g Pu/l:

- Transfer from 2. Pu-cycle
  - ≦ 12.5 g Pu/l $1.1\cdot10^{-4}$/a
  - ≦ 50 g Pu/l $2.2\cdot10^{-5}$/a

- Transfer from 1R-concentration
  - ≦ 12.5 g Pu/l $2.8\cdot10^{-4}$/a
  - ≦ 50 g Pu/l $5.5\cdot10^{-5}$/a

For transfer from the 2. Pu-cycle the most relevant contribution to the cumulative probability is a failure of plutonium concentration measurement preceding the transfer. The cumulative probability for a criticality from an accidental transfer of Pu-solution from the 1R-concentration is dominated by a potential undetected malfunction of the 1R-concentrator in a way that a too high concentration of Pu is reached. Therefore the control of this evaporation is of high importance. The analysis revealed, that also in the vessels of the 1R-concentration unit following the 1R-evaporator, which are designed only for limiting Pu-concentration, a criticality may be possible. Therefore it seems mandatory to improve the control of the 1R-evaporator and possibly the design of the following components.

Various chemicals, cleaning agents and spilling solutions in case of decontamination or malfunction of the extractors might be accidentally transferred to the rework tank. Since no data are available for the frequency or possible nature of these liquids a reliability analysis for this possibility to get a criticality is very difficult. A rough estimate of this possible way to reach criticality resulted in a cumulative probability less than 10^{-5}/a. The total cumulative probability for a critical excursion in the rework tank has been calculated to less than $5\cdot10^{-4}$/a. This probability is clearly higher than for the big cylindrical feed storage tank.

4. SENSITIVITY STUDY OF CRITICAL EXCURSIONS

Additional studies have been done in the field of simulation of criticality excursions using the computer program FELIX /2/. The influence of plant characteristic effects like transfer rates and various plutonium concentrations on the resulting peak power and the number of total fissions has been investigated in model calculations. The intention of these sensitivity studies was to establish a dependence between specific parameters for the initiation of the accident and the characteristics of the resulting excursion. By that way at least a rough combination of the determined probability and the kind and magnitude of a potential criticality may be assessed.

Sensitivity studies for variations of the transfer rates and volumes of plutonium solution indicate that the capacity of transfer pumps is an improtant parameter in the assessment of the resulting peak power of the excursion. The influence of pump capacity to the total number of fissions is not so pronounced. Only for small volumes transferred to a near-critical tank the number of total fissions is influenced remarkably. Fig. 6 shows the results for a variation of pump transfer capacity on peak power and total number of fissions.

Most of the flow streams relevant for an accidental transfer of plutonium solution are relatively small. Therefore small pump transfer capacity is provided for these transfer routes. The corresponding peak power is likely to be in the order of 10^{18} fissions and by that one magnitude less than anticipated in the conservative assessment in the licensing prodedure. In calculations for a criticality duration of 600 s the total number of fissions did not exceed a value of $2.5\cdot10^{19}$ fissions.

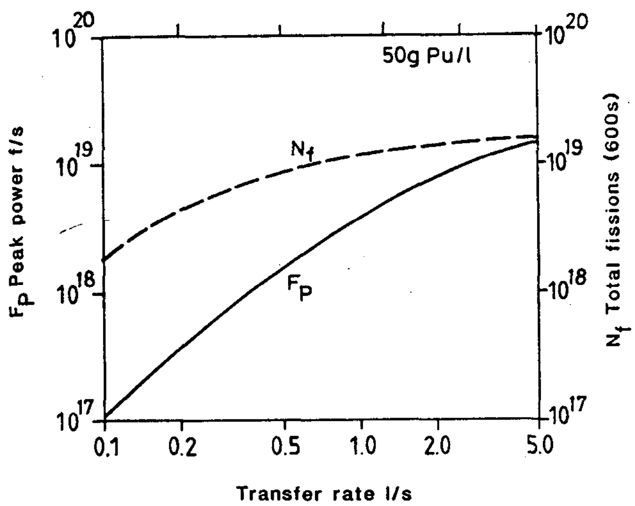

Figure 6: Variation of pump capacity for accidental transfer of plutonium solution

Responsible for this limitation is the fact that after reaching boiling temperature the power level drops at least for one order of magnitude. The variation of Pu-concentration has only a minor effect on the number of total fissions. For low transfer solution volumes and low Pu-concentration the critical system did not reach the boiling temperature within the calculated time span. In these cases, however, a release of fission products to the off-gas systems and the environment would be very small.

5. SUMMARY OF THE RESULTS

The in-depth-analysis shows that the probability for a critical excursion in the feed storage tank as a consequence of a erroneous dissolution of fresh fuel elements is very low. Most important reasons for this low probability are the series of preceding checks and measurements and the presence of low enriched uranium with only low plutonium concentrations. Higher probabilities for a criticality have been derived for the rework tank. The most relevant probability contributions result from failures in the transfer of fissile solutions from the 2. Pu-cycle and the 1R-concentration. An accidental transfer of plutonium solution from the 2. Pu-cycle may involve higher transfer rates and solution volumes than a transfer from the 1R-concentration. A cumulative probability of less than $5 \cdot 10^{-4}$/a has been assessed for a potential criticality in the rework tank. Sensitivity studies of the behaviour of critical solutions indicate that for the anticipated pump transfer capacities and solution volumes the power of the first excursion spike will be in the order of 10^{18} fissions or less. The total number of fissions will not exceed $2.5 \cdot 10^{19}$ fissions in the calculated time span of 10 minutes. For prolonged duration of the critical state only a small increase of the number of total fissions is probable after reaching boiling because of power level drop. The relevant parameters for the estimation of consequences of a criticality accident in the licensing procedure include approximately a factor of 10 of conservatism for peak power and of 4 for total fissions.

The results of the in-depth-analysis revealed some weak points where the criticality safety provisions have to be improved. To remove the relevant possibility for a criticality by transfer of plutonium solutions into the rework tank the connection from the 2. Pu-cycle has to be changed to another rework tank which is geometrically safe for high plutonium concentration. The criticality safety design of the 1R-concentration has to be improved, especially the control of the 1R-evaporator. The direct exit from the rework tank to the HA-column has to be skipped to prevent that unbalanced rework solution will enter directly the HA-column.

Generally failure tree analysis has been shown to be a very useful and systematic instrument also in the field of criticality safety analysis to detect design deficiencies.

Literature

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