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FUEL REPROCESSING : SAFETY ANALYSIS OF EXTRACTION CYCLES.

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ABSTRACT

An essential part of the safety analysis related to the extraction cycles of reprocessing plants, is the analysis of their behaviour during steady-state and transient operations, by means of simulation codes. These codes are based on the knowledge of the chemical properties of the main species involved (distribution coefficient and kinetics) and the hydrodynamics inside the contactors (mixer-settlers and pulsed columns). These codes have been consolidated by comparison of calculations with experimental results from laboratory experiments or industrial plants, covering the whole range of plant operations.

The safety analysis of an extraction cycle operation is essentially performed in two steps.

The first step is a parametric sensivity analysis of the chemical flowsheet operated : the effect of a misadjustment for each operating parameter (flowrate of feed, solvent, etc) on the functionning of the workshop is evaluated by successive steady-state calculations. These calculations help the identification of the sensitive parameters for the risk of plutonium accumulation, while indicating the permissible level of misadjustment. These calculations also serves to identify the parameters reflecting the status of the process (plutonium accumulation or not) which should be measured during plant operation.

The second step is the study of transient regimes, for the most sensitive parameters related to plutonium accumulation risk. The aim of this study is to confirm the conclusions of the first step and to check that the characteristic process parameters chosen effectively allow, the early and reliable detection of any drift towards a plutonium accumulating regime. The procedures to drive the process backwards to a specified convenient steady-state regime from a drifting-state are also verified.

The identification of the sensitive parameters, the process status parameters and the process transient analysis, allow a good control of process operation. This procedure, applied to the first purification cycle of COGEMA's UP3-A La Hague plant has demonstrated the total safety of facility operations.

I. SIMULATION MODEL OF EXTRACTION OPERATIONS

The methodology employed in developing the numerical code for modelling the extraction operations consists of four steps. First of all, the phenomenological analysis is performed. Accordingly, the main mechanisms are translated in mathematical terms. Then numerical solving of the problem is the next step. At last the code produced is then qualified by comparing the calculations with the experimental data. After these steps are successful, the code produced is used to analyse plant operations.

The numerical code ([1], [2]) used to analyse plant operation has the following main characteristics:

- **species considered** :U(VI), U(IV), Pu(IV), Pu(VI), Pu(III),HNO_3, HNO_2, Zr, Tc, $N_2H_5NO_3$, TBP, diluent
- **mechanisms simulated** : partition of species, transfer kinetics, kinetics of redox reactions
- **extractors simulated** : mixers-settlers, pulsed columns

This code was qualified by comparing calculations with experimental data from laboratory experiments performed in mixer-settlers and pulsed columns, in steady and unsteady states conditions, and also with operating results of French industrial facilities.

II. EXAMPLE OF QUALIFICATION

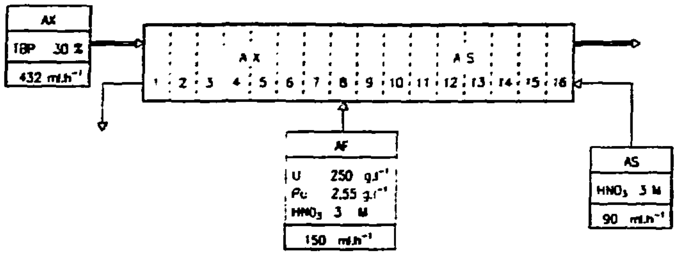
The following describes an example of a code qualification experiment, performed in a set of mixer-settlers.

The experiment consists of three steps. In the first, we carried out an experience based on the flowsheet shown in chart I, and recorded experimental profiles of uranium, nitric acid, and plutonium in both aqueous and organic phases at equilibrium. In the second step, the acidity of the scrub flux was decreased from 3M to 0.1M, creating a plutonium accumulation. The aqueous plutonium concentration of stages was monitored during the transient regime. The final step was the recording of the experimental profiles in the final equilibrium state.

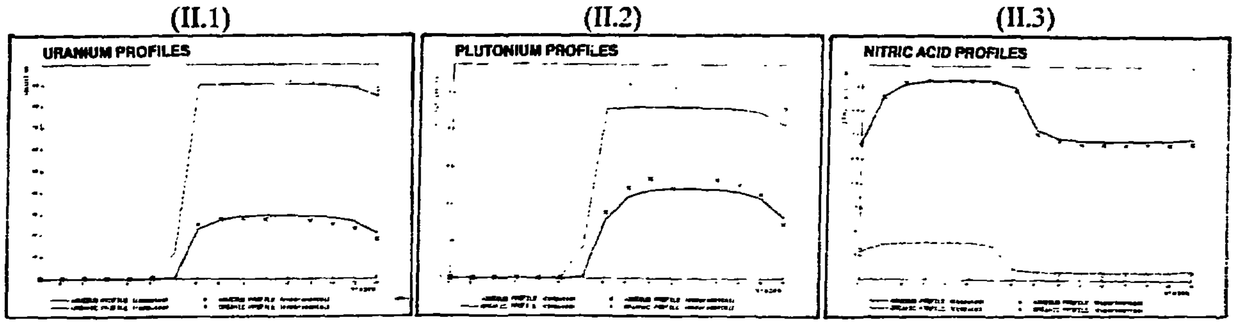
Graphs II.1, II.2 and II.3 show the comparison of computed and experimental profiles in the initial steady state. Graphs n0. III.1, III.2 and III.3 compare the computed and experimental variation of aqueous plutonium concentration in stages, and graphs IV.1, IV.2, IV.3 show the comparison of profiles in the final steady state.

These comparisons show the good results of our calculations both in steady and unsteady state conditions.

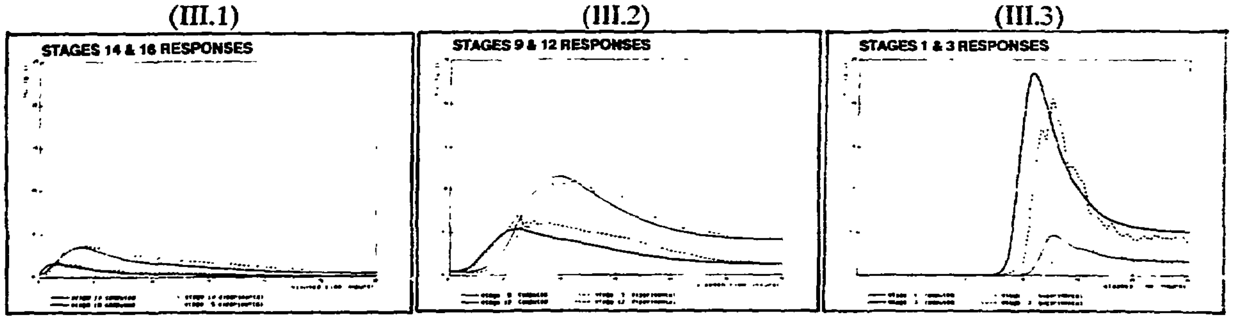
I. Experimental flowsheet



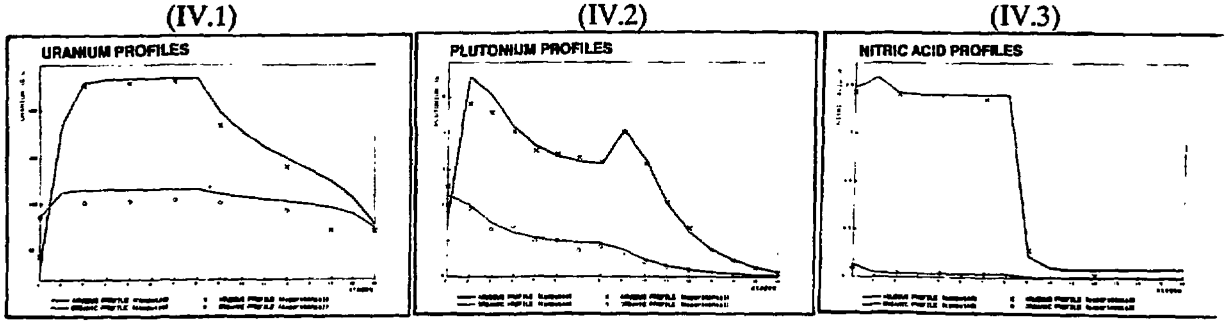
II. First step : initial steady-state profiles


(II.1) (II.2) (II.3)

III. Second step : decrease of scrubbing acidity (3M to 0,1M)


(III.1) (III.2) (III.3)

IV. Third step : final steady-state profiles


(IV.1) (IV.2) (IV.3)

III. APPLICATION TO SAFETY ANALYSIS.

The plant operation analysis using our numerical code is performed in two steps, a steady-state analysis followed by a transient analysis.

III.1. STEADY-STATE ANALYSIS

The first step is aimed to determine, for each of the operating parameters (feed flowrate, feed composition, solvent flowrate, etc) the maximum permissible misadjustment level with respect of plutonium accumulation and leakage, and to identify process status (plutonium accumulation or not) indicators . This is done by successive steady-state calculations.

This is illustrated by the calculations made for the sensitivity analysis related to solvent flowrate for the experimental flowsheet presented in chart I.

Graph V.1 shows the variation of the plutonium peak (highest aqueous plutonium concentration in the contactor) and plutonium concentration in the raffinate with the decrease in solvent flowrate in case for the flowsheet in figure I. It shows that the maximum permissible decrease is about 8%.

Graph n0.V.2 shows the variation of the plutonium peak and reflux scrub density. It can be concluded that this value is a reliable indicator of plutonium accumulation.

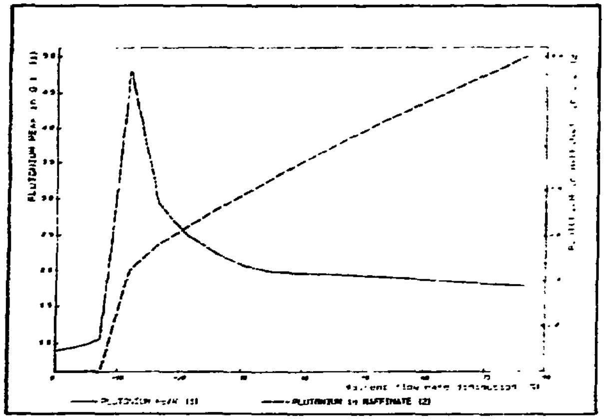
III.2. TRANSIENT ANALYSIS

The second step consists of, for the most sensitive parameters, a transient analysis of the process in case of malofunction to appreciate its dynamic response, check the reliability of the indicators selected and to elaborate procedures to drive the process backwards to a specified convenient status after the occurence of a malfunction.

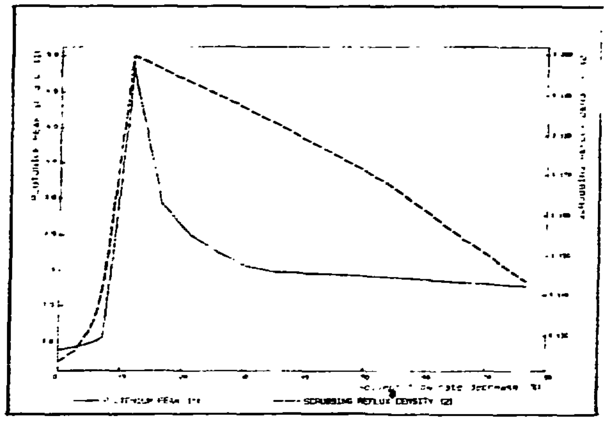
This step is illustrated by transient calculations resulting from a decrease of 20% of the solvent flow rate for the experimental flowsheet presented. Graph VI.1 shows the plutonium peak and plutonium concentration in the raffinate variation. The highest plutonium peak is reached in about 10 hours, and plutonium leakage occurs at about the same time. The quick rise of scrubbing reflux density shown in graph VI.2 shows this value to be an early indicator of plutonium accumulation.

V. Example of steady-state analysis : *flowsheet sensitivity to solvent flowrate*

V.1 Plutonium peak / plutonium leakage in raffinate

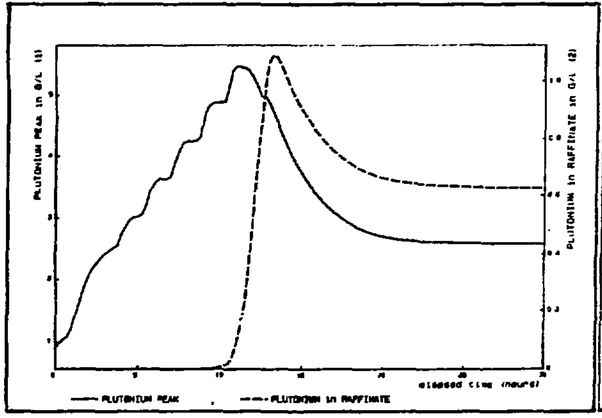


V.2 Plutonium peak / scrubbing reflux density

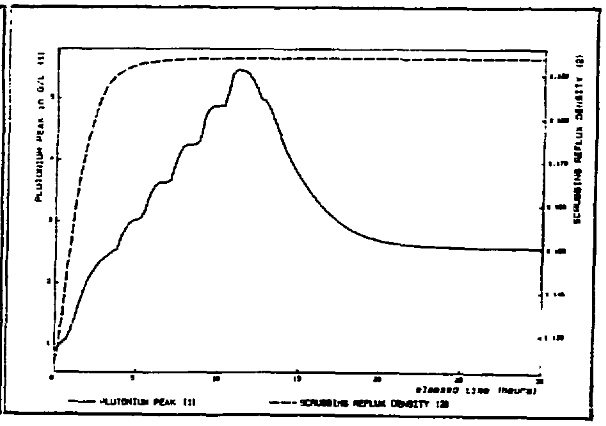


VI. Example of transient analysis : *process response to a decrease of solvent flowrate (-20%)*

VI.1 Plutonium peak / plutonium leakage in raffinate



VI.2 Plutonium peak / scrubbing reflux density



IV. CONCLUSION

A simulation model, validated by a substantial experimental data base, is an outstanding tool for the safety analysis of extraction operations.

It allows the following :

- identification of the sensitive operating parameters
- detection of early indicators, reflecting the process status
- prediction of dynamic behaviour of the process during potentially dangerous malfunctions,
- elaboration of procedures to drive the process backwards to specified convenient conditions in case of the occurence of a malfunction.

This approach, applied to the extraction facilities of the UP3-A plant (La Hague) helped to certify the validity of the operating flow sheets as well as the control methods selected, and led to conclude in the safety of their operation.

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