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SIMULATION OF URANIUM/PLUTONIUM SPLITTING IN A PULSED COLUMN IN THE PUREX PROCESS

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SIMULATION OF URANIUM/PLUTONIUM SPLITTING IN A PULSED COLUMN IN THE PUREX PROCESS

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The reductive plutonium stripping operations carried out in the Purex process, such as uranium/plutonium splitting, are sometimes difficult to get under control. When these operations are performed in pulsed columns, added to the complexity of the chemical reactions involved, are the specific features of this type of contactor, whose performance is governed by a set of equally complex factors.

The perfect mastery of this process, both in the design phase and in the operating phase, requires the availability of a simulation tool incorporating the various aspects of the operation. The French Commissariat à l'Energie Atomique accordingly set up a research programme aiming to develop a mathematical model simulating the behaviour of the different species concerned in unsteady state conditions. This modelling was carried out in several steps :

- elaboration of a preliminary model, simulating uranium/plutonium splitting in mixers/settlers (already reported elsewhere <1>) ;
- elaboration of a model for simulating simple extraction operations in a pulsed column, accounting for the specific features of this contactor ;
- build-up of the final complete model.

* * *

The modelling of the uranium/plutonium splitting operation in mixers/settlers required the following in succession :

- the inventory of the chemical species to be taken into account (nine were selected : U(IV), U(VI), Pu(III), Pu(IV), HNO_3, HNO_2, $N_2H_5^+$, Tc(ox), Tc(red));
- the compilation of data concerning the partition of each of these species between the aqueous nitric phase and the solvent (30% TBP, diluent) (interdependent partition, described by a semi-empirical formula for the extraction mechanisms) ;
- the inventory of the chemical redox reactions ('functional', 'interfering' or 'useful' reactions) involving the various components, in both phases ;
- the formulation of the kinetic aspects relative to the interfacial transfer of the species and the main chemical redox reactions involved.

These details were obtained either from the compilation of published results, or, above all, from experimental determinations made in the CEA laboratories. They hence provide the basis for a model simulating a partition operation with uranous nitrate in a compartmented extractor. This model was validated successfully by comparing the results of its operation :

- with results of specific experiments performed in a suitable alpha installation (laboratory mixers/settlers) ;
- with operating results of industrial units.

The modelling of simple extraction operations (not involving chemical redox reactions) **in a pulsed column** required consideration of :

- Data relating to partition and to the transfer kinetics for the species concerned (U(VI), Pu(IV), HNO_3), previously obtained ;
- Mechanisms specific to liquid/liquid extraction columns which are reflected on the whole by axial mixing effects in both phases.

We thus developped a multi-component dispersion model which, after the determination of the axial dispersion coefficients, allows the calculation of simple extraction operations encountered in nuclear fuel reprocessing.

The validation of the model required the use of extensive experimental facilities, ranging from laboratory pilot plants to prototype industrial installations. The conducted tests consisted in the simultaneous determination of :

- concentration profiles in both phases, for different extraction operations ;
- axial dispersion coefficients by injection of tracers into each phase.

The lower performance observed at low flow rate is well simulated by the model. Figure 1 shows the variation in the mean HETS as a function of total specific flow rate, calculated for the same set of parameters. The comparison of the experimental and calculated concentration profiles (Figure 2) is also satisfactory.

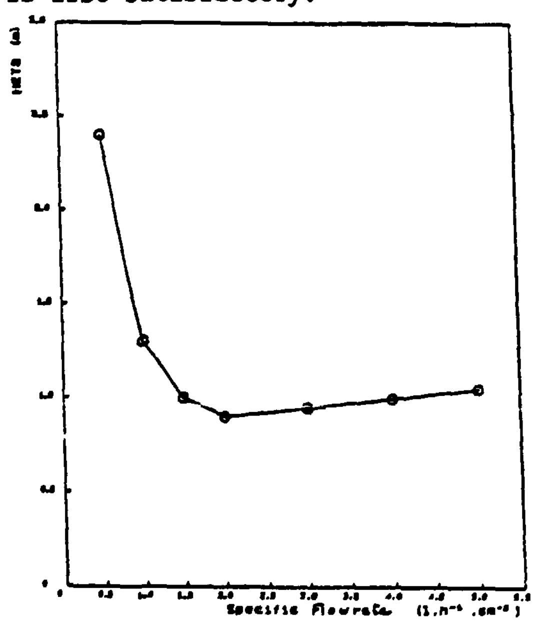

Figure 1 : Mean HETS(*) versus total specific flow rate (uranium extraction)

(*)HETS : Height Equivalent to Theoretical Stage.

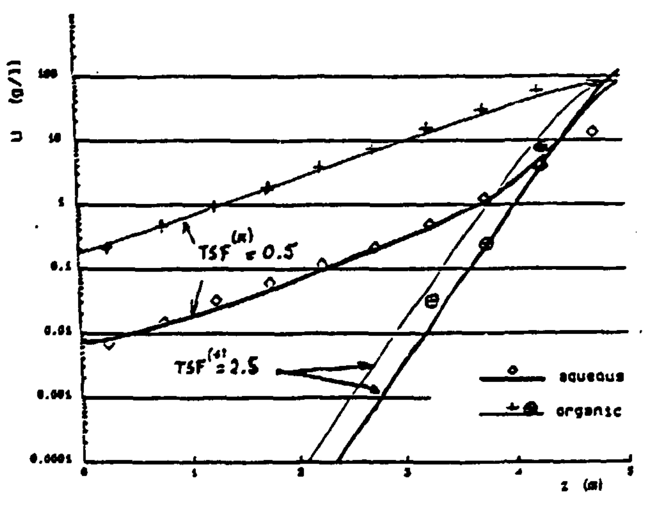

Figure 2 : Calculated and on) experimental U(VI) profiles (extracti

(*) TSF : Total Specific Flow rate ($l.h^{-1}.cm^{-2}$)

The completion of the foregoing two developments served to undertake the construction of a model able to simulate the metabolism of the different chemical species involved in a reductive stripping operation on plutonium in a pulsed column. The use of this apparatus as compared with mixers/settlers leads to signifiant differences in the operation of uranium/plutonium splitting. The competition between the various chemical reactions is intensified, both phases remaining in permanent contact. A number of relatively negligible mechanisms in mixers/-settlers assume greater importance here. This meant that the corresponding kinetic data used in the first model had to be ajusted.

The model thus obtained served to reproduce the behaviour described above, and yields very satisfactory results, as shown by the successful comparisons of the computer code with the reference experiments performed in a suitable test facility (Figure 3).

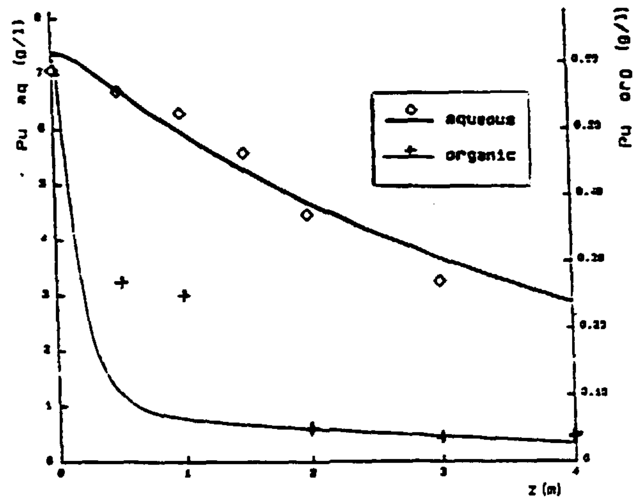

Figure 3 : Calculated and experimental Pu profiles (U/Pu splitting)

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Hence we now have an qualified tool for calculating the most complex extraction operations conducted in the Purex process. Its field of application extends to :

- The optimized definition of process flow sheets as well as equipment ;
- The simulation of incidental operations in order to predict possible developments, the means for detecting them (optimization of control systems), and the measures required to return to the normal state : for example, the model helps to carry out the safety analysis which may be required to guarantee the safe operation of the facility concerned.

Finally, it provides a solid basis of knowledge that could be exploited advantageously, both for training operators and for the elaboration of efficient tools for aid in facility control (expert system).

REFERENCE :

<1> Boullis B. and Baron P.
Modelling of uranium/plutonium splitting in Purex process
I. Chem.E.Conf.Extraction '87, Dounreay, June 1987.