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ESTIMATING REPROCESSING PLANT IN-PROCESS INVENTORIES BY SIMULATION

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ABSTRACT

The Safeguards Systems Group’s generic simulation program FacSim was used to model the operation of the proposed Rokkasho Reprocessing Plant during an operating cycle consisting of a start-up phase, a period of steady-state operation, and a flush-out phase. The simulation results give a detailed account of nuclear material inventories in various process vessels as a function of time. As expected, it is found that the pulsed columns and the concentrator determine the rate at which the system responds to feed variations and transients; but the in-process inventory is dominated by the contents of the concentrator and tanks, and particularly by the contents of the tanks downstream from the concentrator. The results of the simulation were used for statistical studies of diversion detection, as described elsewhere in these Proceedings.

INTRODUCTION

Japan currently obtains about 30% of its electric energy from nuclear power plants and plans to obtain an even larger fraction of its electric energy from this source in the future. Japan Nuclear Fuel Limited (JNFL) is currently constructing the Rokkasho Reprocessing Plant (RRP) to recover the uranium and plutonium in the spent fuel from the Japanese power reactors. RRP will have a capacity of 800 metric tons of heavy metal per year and is expected to begin receiving spent fuel in 1996 and to commence reprocessing operations some time after the year 2000. RRP will be the first large-scale reprocessing plant under full-scope International Atomic Energy Agency safeguards.

Members of the safeguards groups at Los Alamos National Laboratory are working with JNFL personnel to develop appropriate measurement methods for use at RRP, to estimate the plutonium accounting accuracies that can be achieved, and to determine near-optimal statistical methods for monitoring balance closure results. As one part of this effort we have developed a simulation model of RRP that allows us to estimate the amounts of material present in the various process vessels as functions of time. This paper describes the simulation method and the results obtained in the initial phase of the study. These methods and results are being refined.

FACILITY DESCRIPTION

The RRP design is based on the PUREX process, which uses a chemical method called solvent extraction to separate the uranium and plutonium in the spent fuel both from the fission products in the spent fuel and from one another. The key process element for solvent extraction in most current large-scale reprocessing plants, including RRP, is the pulsed column. A schematic representation of one type of pulsed column is shown in Fig. 1. During operation a nitric acid or aqueous stream enters the column near the top, flows down the shaft of the column, and exits near the bottom of the column. Simultaneously, a kerosene-like solvent or organic stream, often containing tributyl phosphate, enters the column near the bottom, flows up the shaft, and exits near the top of the column. A pulsing action produced by a pump or by compressed air causes the immiscible, counter-flowing aqueous and organic streams to mix thoroughly in the shaft section of the column, which is filled with baffle plates to aid the mixing. By adjusting the molarity of the nitric acid, the oxidation states of the uranium and plutonium ions in solution, and the presence or absence of tributyl phosphate in the organic stream, it is possible to cause the uranium and plutonium to change from the aqueous...
stream or phase to the organic stream (extraction); from the organic stream to the aqueous stream (stripping); or to remain in same stream (washing or scrubbing). These pulsed-column characteristics can be used to separate the uranium and plutonium from fission products (purification) and from each other (partitioning).

The process flows for RRP are shown in Fig. 2, and are typical of those in large-scale reprocessing plants. Because plutonium accounting was our main interest, the simulation studies for RRP were limited to the part of the process between the input and output accountability tanks. Therefore, we do not discuss the head-end processes (spent-fuel pond and transport, shear, dissolver, centrifuge, etc.). Also, uranium flows after the partitioning column are not treated.

The facility design for RRP calls for the input accountability tank to be filled at periods somewhat greater than a day with a nitric-acid solution that contains dissolved uranium, plutonium, and nongaseous fission products from spent fuel. The nominal plutonium concentration in this dissolver solution is about 1% of the uranium concentration. After an accountability measurement is made, the dissolver solution flows through several buffer tanks to the first purification cycle, which consists primarily of a series of pulsed columns in which the major fission product contaminants are removed and the uranium is separated from the plutonium. The uranium stream then follows a separate process path that is not considered here. The plutonium solution leaving the first purification cycle passes through a series of buffer tanks to the second plutonium purification cycle, where traces of uranium and additional fission products are removed, and then through another series of buffer tanks and a third purification cycle. In addition to removing fission products from the plutonium, the purification cycles also increase its concentration. The plutonium solution from the third purification cycle passes through a series of buffer tanks to the concentrator, where the plutonium concentration is increased still further to a final value that is about 90 times greater than its initial value in the input accountability tank dissolver solution. Plutonium solution from the concentrator is collected in a receiving tank and then transferred to the
output accountability tank for measurement. Fission products removed from the uranium and plutonium streams in the various purification steps are sent to waste treatment processes, and are not considered here.

SIMULATION METHOD

Simulations are often characterized as being discrete-event or continuous. In a discrete-event simulation, a series of events occurs at a sequence of simulated times, and each event may be thought of as being instantaneous (for example, arrival of a part at a machining station). In a continuous simulation, inter-related values change continuously, and it is necessary to integrate a set of differential equations that describes the system. Simulation of a reprocessing plant involves both discrete-event and continuous elements: the flow of materials through pipes that connect the process vessels begins and ends at discrete times, and between these discrete times the contents of process vessels and pipes evolve in a way that is described by a set of differential equations. The FacSim program that was used for the RRP simulation supports the description of both types of time evolution.

The assumption that there are time intervals in which the reprocessing facility can be described by a set of differential equations is actually a significant simplification compared to the true state of affairs. Even a simple tank with one input pipe and one output pipe is a three-dimensional system with both spatial and temporal variations in concentrations, fluid flows, etc. The situation is much more complicated for a pulsed column, which combines turbulent, viscous, nonsteady, multi-phase fluid flow in a complex geometry with temperature-dependent, nonequilibrium, nonvolume-preserving chemical reactions occurring at fluid interfaces. Detailed calculations of pulsed-column characteristics usually describe the column as a line of linked chambers, each treated as being internally homogeneous, and use parametrized fits to experimental data to describe phase mixing, mass transfer, and similar behavior. Even this simplified model is too computationally intensive for the simulation of RRP, where the behavior of several dozen interacting tanks, pulsed columns, and other process vessels must be calculated for simulated time periods of weeks or months. Consequently, simplifying assumptions such as the following were made to describe the process vessels in the simulation program:

1. It was assumed that the concentrations of fluid constituents entering an output pipe from a tank are equal to the average concentrations of these constituents in the tank. This approximation is probably never wrong by more than a few percent and is of course accurate once steady-state conditions have been achieved.

2. It was assumed that the concentrations of fluid constituents entering an output pipe from a pulsed column are proportional to the average concentrations of these constituents in the column. This assumption is consistent with the results of Beyerlein and Geldard, who find that the inventory of a fluid constituent in a pulsed column has a time behavior that is approximately exponential as it approaches equilibrium. The approximation is probably valid to within about 30% or better if the correct proportionality constants for the column are used.

Because many of the pulsed-column design and operating parameters were not available, it was necessary to make somewhat arbitrary (but typical) choices for the proportionality constants linking the pulsed-column output concentrations to the average concentrations. We assumed that the output concentration of a stream constituent that was being scrubbed or washed was equal to the average concentration of the constituent in the column, and the output concentration of a stream constituent that was being extracted or stripped was 10 times the average concentration of the constituent in the column.

SIMULATION RESULTS

The operation of RRP was simulated for a start-up/steady-state/flush-out cycle in which the pulsed columns were initially prepared with clean acid and solvent, twelve accountability-tank batches of dissolver solution were processed through the system, and then the process line was flushed with eight input batches of clean acid. (This is not an actual operating cycle for RRP, but illustrates the various operating modes of the plant.) At start-up the pulsed columns contain no nuclear material, and when the dissolver solution first begins to reach the columns a period of time is required for the plutonium inventories in the columns to build to equilibrium values. Figure 3 shows the plutonium inventories of several of the pulsed columns in the first purification cycle as functions of time after the
beginning of the simulation (which is 7 hours before the input accountability tank initially begins to fill with dissolver solution). The dissolver solution first enters the pulsed column Column 1, where the uranium and plutonium are extracted into the organic phase and most of the fission products remain in the aqueous stream. In Columns 2 and 3 more fission products are scrubbed from the uranium- and plutonium-bearing organic stream; and then in Column 4 the plutonium is stripped into the aqueous phase, while the uranium remains in the organic phase and exits from the simulation. Traces of uranium and additional fission products are removed from the plutonium-bearing aqueous stream in Column 5 and a mixer-settler (not shown), and the aqueous stream then enters the buffer vessels between the first and second purification cycles. The following features of the pulsed-column behaviors should be noted:

1. The first pulsed column requires somewhat more than an hour to reach an approximate steady-state plutonium inventory. Each successive column requires longer to reach equilibrium than its predecessor, because the stream entering it is "lean" for a longer period of time.

2. Other things being equal, wash and scrub columns have larger inventories than do extraction and stripping columns. This is because nuclear material entering an extraction or stripping column immediately begins moving to the phase that is exiting at the same end of the column, so most of the material remains at one end of the column. In a wash or scrub column, on the other hand, the nuclear material remains in the same phase throughout its residence in the column, and is approximately uniformly distributed along the length of the column.

3. The pulsed-column inventories reach their steady-state values in a series of steps. The steps are caused by the fact that the feed tank for Column 1 is prepared with an initial heel of clean acid, so that the first batch of dissolver solution reaching the feed tank is somewhat diluted. The degree of dilution then decreases with succeeding batches.

The build-up to steady-state plutonium inventories in the second and third purification cycles takes place still more slowly, because each cycle receives a feed stream from the preceding cycle that is initially far below nominal steady-state concentration. Several days elapse between the time dissolver solution initially reaches the first purification cycle and the time the third purification cycle inventories reach approximate steady-state values. The time dependence of the total inventories of the three purification cycles during start-up, steady-state operation, and flush-out can be seen in Fig. 4. As noted before, the times are only indicative because of uncertainties about the true values of the pulsed-column parameters.

Although the pulsed columns in the three purification cycles are key determiners of the time scale for

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**Fig. 3.** Plutonium inventories (arbitrary units) in selected solvent-extraction vessels in the first purification cycle as functions of time from the beginning of the simulation.

**Fig. 4.** Total plutonium inventories (arbitrary units) of the solvent-extraction vessels in each of the three purification cycles as functions of time from the beginning of the simulation.
changes in plutonium inventory, the purification cycles contain only a small part of the total inventory in steady-state operation. This can be seen in Fig. 5, in which the upper curve shows the plutonium inventory in all process vessels between the input and output accountability tanks except those in the three purification cycles, and the three lower curves show the plutonium inventories in these cycles. Figure 5 also shows that the in-process inventory periodically undergoes rather large increases, and at longer intervals undergoes even larger decreases. The inventory increases correspond to transfers from the input accountability tank to the buffer vessel immediately following it. The source of the inventory decreases is shown in Fig. 6, in which the upper curve shows the plutonium inventory in all process vessels between the input and output accountability tanks, and the lower curve shows the plutonium inventory in the receiving vessel between the concentrator and the output accountability tank. It is seen that when the receiving vessel is full it contains almost half the plutonium inventory of the entire process. It is clearly desirable to close material balances when this receiving vessel is nearly empty and also, if possible, just before a transfer from the input accountability tank to the buffer vessel that follows it. There is not enough space here to show the plutonium inventories and transfers for all of the other process vessels, but none of these other inventories/transfers is as important as the two just mentioned.

![In-process Inventory](image)

**Fig. 5.** The top curve shows the total plutonium inventory (arbitrary units) in all process vessels between the input and output accountability tanks except the vessels in the three purification cycles, and the three bottom curves show the plutonium inventories in the process vessels that are in the three purification cycles.

![In-process Inventory](image)

**Fig. 6.** The top curve shows the plutonium inventory (arbitrary units) in all process vessels between the input and output accountability tanks, and the bottom curve shows the plutonium inventory in the receiving vessel that is between the concentrator and the output accountability tank.

**COMMENTS AND CONCLUSIONS**

The simulation makes it possible to determine the time-dependent nuclear material inventory of every process vessel in the reprocessing plant, even during transient periods such as start-up and flush-out. In fact, even during steady-state operation the determination of in-process inventories may be fairly complicated because various sections of the RRP process line operate with periodicities that are incommensurable within the time span of a facility operating cycle. Thus the inventory is not periodic even at steady state. An indication of this can be observed in Fig. 6, where it is seen that the inventory minima near 200 hours and 320 hours have different widths. Without a tool such as simulation it would be very difficult to determine the expected in-process inventory as a function of time. By means of simulation it is possible to evaluate the effectiveness of each of a variety of materials accounting strategies and anomaly-detection methods for the facility, and this has been done for RRP and is reported on elsewhere in these Proceedings.

The simulation study reported on here neglected the volumes of the pipes connecting the process vessels to one another and did not take into account certain recycle procedures that are used during facility start-up. These omissions are being corrected in the second phase
of the study that is currently in progress, and other improvements are also being made to enhance the accuracy of the simulation. It is expected that these changes will introduce small quantitative changes, but not qualitative changes, in the simulation results.

REFERENCES
