**Solvent Extraction Equipment
Evaluation Study
Part 2. Workshop Proceedings**

January 1977

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SOLVENT EXTRACTION EQUIPMENT
EVALUATION STUDY
PART 2. WORKSHOP PROCEEDINGS

For the Workshop on Evaluation of
Solvent Extraction Equipment given
August 5-6, 1976 at the
Battelle Seattle Research Center,
Seattle, Washington

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ABSTRACT

This is Part 2 of a three-part document that reviews the solvent extraction contactors available for use in radiochemical reprocessing plants. The three parts are: Part 1 - A Review of the Literature, Part 2 - Workshop Proceedings, Part 3 - A Summary. The main objective of the document is to provide an information base to aid in contactor selection and design of future reprocessing plants.

The Literature Review (Part 1) brings together scattered data on all major contactors in use today. It contains an annotated bibliography of the citations used in the review and a complete listing of all the citations screened prior to the review. These bibliographies should aid in determining which reports would be most useful if additional information is desired.

The Workshop Proceedings (Part 2) summarizes a workshop on the title subject held in mid-1976. The participants had considerable experience in the use of solvent extraction contactors in reprocessing plants. The purpose of the workshop was to bring together these people in order to collect, evaluate and document any information that would be helpful in the selection of solvent extraction contactors for nuclear fuels reprocessing plants.

The Summary (Part 3) is a compendium of the information presented in the Literature Review and the Workshop Proceedings. It was written to present the salient points of Parts 1 and 2 without going into as much detail. The Summary contains a listing of the references used in the Literature Review so the reader can either go directly to the citation or to Parts 1 and 2 for further information.

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PROCEEDINGS OF THE
WORKSHOP ON EVALUATION OF SOLVENT EXTRACTION EQUIPMENT

INTRODUCTION

On August 5-6, 1976, a workshop on solvent extraction equipment was held at the Battelle Seattle Research Center in Seattle, Washington. The overall purpose of the workshop was to bring together people of diverse backgrounds and experience in order to collect, evaluate, and document data on solvent extraction process equipment, control, and operation as applied to the nuclear industry. It was felt that an unstructured format geared to the participants' needs and interests would produce the maximum interchange of ideas. To identify these needs and interests, the group was asked to develop two lists: a list of their personal objectives for the workshop, and a list of the issues that they would like to see covered.

OBJECTIVES

The group developed the following list of objectives:

- Assess the current state of the technology--its main issues, problems, and deficiencies.
- Determine the potential of each of the contactor types and develop criteria for contactor selection according to flowsheet variations.
- Compare actual production plant experience (largely undocumented) with documented pilot plant experience in order to determine if production plants operate as predicted by pilot plant data.
- Identify design and operational failures, since these have not been documented as extensively as the successes.
- Provide an opportunity for improved communication among the various ERDA (Energy Research and Development Administration) contractors, each of which is working on a different aspect of the fuel recycle program.
- Formulate research and development (R&D) ideas.
- Obtain feedback on the literature search document and identify any additional articles and other sources that should be included.

ISSUES

Once the participants had established their objectives, they developed a list of the issues of importance in radiochemical processing. First, they grouped items roughly into three categories: design, performance, and auxiliary systems. Then they ranked both the categories and the issues within them in order of importance. The final tabulation of issues, presented in descending order of importance, was:

1. Performance issues
 - a. Solids handling: tolerance for solids, handling of interfacial buildup, bases for deciding which phase should be continuous
 - b. Solvent damage
 - c. Application of pilot plant work to production plant operation
 - d. Flexibility of equipment in adapting to flowsheet changes
 - e. Documentation of plant performance
2. Design issues
 - a. Remote maintenance--desirability; extent necessary
 - b. Criticality
 - c. Criteria for selection of different contactor types, including equipment availability
 - d. Instrumentation for process control and for safeguards
 - e. Design criteria
 - f. Kinetics
 - g. Documentation of the problems and advantages of the different design bases
 - h. Physical dimensions of the equipment
 - i. Flexibility of equipment for different fuel types
 - j. Ranking of design criteria in order of importance
 - k. Solvent damage as a design factor
3. Auxiliary systems issues
 - a. Feed clarification
 - b. Solvent washing
 - c. Phosphate (TBP, DBP) removal

Because of time limitations and a desire to stay within the scope of the project, it was decided to limit the discussion to these issues.

FORMAT

Following the development of the final list of issues, the group addressed the question of the most effective format for confronting these issues. It was decided that participants would remain together as a full group rather than split up into smaller groups, so that no one would miss anything that was said, and so that proponents of one point of view would have the opportunity to hear the others.

INTRODUCTORY PRESENTATIONS

Several participants gave introductory presentations about current projects dealing with some of the issues of concern listed above. These presentations are summarized below.

LWR RECYCLE PROGRAM (A. Jennings, duPont)

ERDA has recently begun putting together a program to help industry close the fuel cycle. E. I. duPont's Savannah River Laboratory (SRL) has been selected as the lead laboratory in the R&D effort for LWRs (light water reactors).

The duPont program will not be of much interest to organizations that have already built a fuel reprocessing plant or completed a conceptual design. It will be of great help, however, to those that are just starting to think about developing a conceptual design.

DuPont's approach will be to involve all of the ERDA facilities and to have the work done where the expertise resides. Thus, the program will involve an extension of work that is already going on at the various sites.

In order to bring all of this work together, the duPont study team will translate the data it collects into a conceptual design study of closing the fuel cycle. The study itself will not involve new R&D;

rather, it will develop a conceptual design based on existing technology. However, duPont has found in the past that a conceptual design study is a highly effective way of pinpointing areas that need additional R&D.

The study team includes representatives of three divisions at SRL: technical, manufacturing, and engineering. The Technical Division's task is to specify the technical requirements of the process and equipment. The Manufacturing Division, which would operate the plant if built, confirms that it would be able to operate any plant meeting these specifications. The information is then passed on to the Engineering Division, which produces the actual design.

Members of the study team hope that the present workshop will provide them with documentation of actual production plant experience for the Manufacturing Division to use in making its selection. Information on process requirements is available from the R&D currently being done; data on equipment design, however, must be obtained primarily from experience. Although a certain amount of design information can be found in the literature, such information is at least 15 years old.

To further complicate the situation, neither the Manufacturing Division nor the Engineering Division has had actual experience with pulse columns, and most of the available documentation represents pilot plant experience. What is needed is more information about how the equipment works in a production plant context.

Among the factors that will be considered in the conceptual design study is equipment reliability. Unreliable equipment can greatly increase the cost of building and operating a fuel reprocessing plant, since it results in the need for redundant equipment to keep the operation going. Since SRL's Engineering Department estimates the current cost of building a remotely maintained and operated canyon as \$1 million per linear foot, equipment design must take all cost-influencing factors into account.

The efficiency of the equipment, on the other hand, does not seem to affect cost much, at least with respect to mixer-settlers. That is, the difference between 75% and 95% processing efficiency does not seem to be important. However, pulse column experience may be different. This uncertainty underscores the lack of available data and the need for

workshops such as this one to bring out and document production plant experience.

LMFBR RECYCLE PROGRAM (B. Vondra, ORNL)

The LMFBR (liquid-metal fast breeder reactor) program deals with those areas in which LMFBR fuels differ from LWR fuels: higher burnups, larger quantities of plutonium, and different types of cladding. The problems addressed by the program occur mostly at the head end of the solvent extraction process. Since constraints on containment will probably be far more stringent than at present by the time an LMFBR plant is in operation, the program also is concerned with the complete containment of off-gases and all other effluents. Other program thrusts include the final form of the equipment and the remote aspects of the plant.

The program schedule is as follows: component development and scaled-up equipment tests are currently under way; the IPET (integrated prototype equipment test), a cold equipment test, is scheduled for startup in 1979, with a completion date of 1981; and the hot pilot plant is scheduled for startup in 1988. This schedule is expected to provide ERDA administrators with the needed data in time for their promised statement on plutonium--now scheduled for 1986.

LMFBR fuel reprocessing appears to have much in common with the LWR process. In many cases, the differences are just a matter of degree: the quantity of fission products involved, the amount of radiation, the quantity of insolubles, and the proportion of plutonium (16-22% in LMFBR spent fuel as opposed to 1% in LWR). Because of these differences, however, remote maintenance and replacement are vital in an LMFBR plant.

The LMFBR program currently does not include thorium recycling, but is considering it as a backup cycle that might be brought into use if GESMO (Generic Environmental Statement on Mixed Oxides) comes out against plutonium.

The description of the LMFBR program was followed by a brief discussion of related issues. To begin with, it was pointed out that most of the problems peculiar to LMFBR fuel reprocessing deal with storage, transportation, and other head-end operations, rather than with actual

reprocessing of the fuel. For example, fuels that have developed breaks in the cladding pose problems for fuel storage.

One question that was raised was whether flowsheets will be required to yield plutonium-free uranium or uranium-free plutonium, or whether a combined product will be acceptable. Although several options are available, a final decision cannot be made until the results of GESMO and safeguards decisions are in. If GESMO comes out in favor of the use of plutonium, a decision probably will be made to go to a breeder cycle, which in turn will result in flowsheets yielding complete plutonium segregation. However, the use of plutonium versus uranium fuel is only one of the factors that must be taken into account in determining the flowsheet product. The other is the process selected for fuel refabrication, together with the relative ease of coupling the different flowsheet products with that process.

It was also pointed out that it is important to consider the whole fuel cycle, rather than just a segment. Otherwise, fuel manufacturers may come up with a fuel that has excellent performance but is very difficult to recycle. This is in fact the case with mixed oxide fuels, which are excellent fuels but are almost impossible to dissolve completely.

EVALUATION OF SOLVENT EXTRACTION EQUIPMENT (D. Lester, PNL)

Our program on evaluation of solvent extraction equipment was begun in April 1976. The main program objective is to provide an information base to aid in contactor selection and design. Other objectives include:

- gathering and summarizing the relevant literature;
- critiquing, interpreting, and adapting the information in the literature;
- developing unpublished sources--letters, verbal comments, and experience;
- identifying needs for more information; and
- combining all of the above and presenting the information in a single, usable source.

At the time of the workshop, progress on the project included completion of the literature search, as well as organization of the workshop. The literature search involved the use of computerized data bases, hand searches, and the personal literature files of engineers with many years of experience in the field. The initial product of the search was somewhere between 1,000 and 2,000 citations. These sources were then screened by a review of abstracts and/or the citations themselves to eliminate duplications and material irrelevant to solvent extraction equipment. This screening reduced the number of citations to the 128 discussed in the literature search document (Part I of this study).

The document resulting from this stage of the project has three major parts. The first, as indicated above, includes the final literature review; it also contains an annotated bibliography and the original listing of the 1,000 to 2,000 citations. Part II consists of the workshop proceedings, and Part III comprises a summary of both the literature review and the workshop proceedings. This last volume is written so that it can be used alone if documentation is not needed.

LITERATURE SEARCH (R. Geier, ARHCO)

The purpose of the literature review was to provide a compendium of what has been done, to be used by those planning additional work in the field. Following the selection and screening process described by D. Lester (above), a summary was written for each entry, describing the equipment or system that was tested and the test results. No interpretation of the data was provided other than that of the author, and no attempt was made to evaluate authors' interpretations.

The group raised several questions as to why certain entries were or were not included. The omission of the Purex Technical Manual⁽¹⁾ was of particular concern to some participants. Although some of its equipment discussions are out of date, it contains a great deal of information that is still valid; in fact, for some purposes, such as height of transfer unit (HTU) calculations, it provides some of the best information available.

Other concerns were that 1) the document gives the same weight to purely academic treatises as to those that are applicable specifically to radioactive processing, and 2) some statements that are now considered invalid may have been included in the document due to the lack of critical review. Subsequently, suggestions were made to alleviate these concerns. The recommendations include:

- Add the Purex manual to the document to provide a balance for some of the academic material.
- Omit the incorrect conclusions.
- Include a critique as a separate document. (Some concern was expressed that the separation of the critique from the body of the literature review would tend to negate its effect; however, it was pointed out that the critique could be heavily referenced in the body of the review.)
- Include a critique section at the start of each entry.
- Include critical commentary in the form of footnotes.

All these suggestions were given consideration in the preparation of the final document.

DISCUSSION OF THE ISSUES

The remainder of the workshop dealt with a discussion of the list of issues developed by the participants.

DESCRIPTION OF CONTACTOR TYPES

Several types of contactors were considered during the workshop. The descriptions and applications of these contactors are presented in the following sections.

Pulse Columns

A pulse column cell, from the cell bottom to the cell cover block, is approximately 40 ft deep. The pulse column itself is usually shorter, and in no case exceeds 40 ft. Its exact height depends on the operation to be performed. Between the top of the cell and the canyon ceiling is a

clearance space of approximately 60 ft, making the total distance from the cell floor to the canyon ceiling over 100 ft.

Some of the participants expressed the opinion that there is no such thing as an extraction section that is too long; others felt that, for a given extraction requirement, there is an optimal length. However, most agreed that as extraction stages are added, scrub stages must also be added, which can result in a very tall column.

Design Standards

The question was raised as to whether pulse column design was still an art, or whether it could now be considered a science. Although participants agreed that it was generally preferable to be able to set up a glass column operation and make adjustments on the basis of observed performance, they felt that some standard criteria could be specified; for example, if a nozzle plate cartridge is to be used:

- Nozzle holes should be 1/8 in. to 3/16 in. in diameter.
- Nozzles should point down for organic continuous and up for aqueous continuous.
- Plate spacing should be 2 in. for extraction and 4 in. for stripping (dispersion is easier if there is more capacity in the stripping column, and the greater spacing is needed to provide adequate capacity).

The optimum amount of free area in the plates was also discussed. The amount generally used is 23%, but variations ranging from 6 to 60% free area have been tested. At 6%, a column would operate adequately; however, if flooding began, it would be impossible to return to normal operation by the usual technique of reducing throughput. Instead, a complete shutdown and restart would be required.

With respect to both nozzle hole size and amount of free area, there is a trade-off between capacity and efficiency of performance. In one of the Purex columns, plates had 33% free area; the result was good capacity but poor efficiency. When the amount of free area was lowered to 23% and hole diameters were enlarged, efficiency improved.

Two other variables to be considered in cartridge design are pulse frequency and pulse amplitude. One participant suggested that with sieve plates having 23% free area, optimal pulse frequency would be 1/sec and optimal pulse amplitude would be about one-half the distance between pulse plates. Another stated that he had experimented with a wide range of amplitudes and had found the optimum to be the distance between the plates; however, he had also found that changes in amplitude had very little effect on the efficiency of the process.

It was observed that established designs are available for pulse columns, and that, with the exception of the electrolytic column currently installed at Allied General Nuclear Services (AGNS), there have been no significant new design changes for some time.

Design Problems

One problem that can occur with pulse columns is that, after a period of time, plates can become dispersed-phase wet and permit large globules of dispersed phase to go through, thereby lowering performance. There are two ways of dealing with this problem. One is to keep the plates wet all the time, even during shutdown; the other is to flush the columns periodically. At one point, it was thought that columns should be flushed every 30 days. Because this practice generated a great deal of waste, flushing was later instituted on an as-needed basis, as indicated by instrumentation. In general, it is felt that flushing should be done at least every 6 months, and sometimes more frequently. It does not interfere substantially with batch integrity, which can be maintained within about 1 kg. About 5 days' turnaround time is required for a complete plant flush: 3 days to complete the emptying and shutdown phase, and 2 more days to purge the column to the point where cross-contamination is sufficiently minimized.

Another problem that can occur with pulse column design has to do with the diameter of the line connecting the pulse generator to the column. For a mechanical pulser, if the line is too small, cavitation can result. For an air pulser, however, this is not a problem.

Centrifugal Contactors--Robatel^(a) Type
(G. Benedict, General Atomic)

The Robatel centrifugal contactor is capable of driving eight to ten stages with a single motor. Figure 1 shows an eight-stage Robatel unit with a variable-speed, 5 horsepower DC motor and an operating range of 1500 to 3200 rpm. The unit has a total flow capacity of 3 liters per minute (ℓ/min), and a solvent flow capacity of around 1.5 ℓ/min. The outside diameter is 8 in. and the bowl diameter is 6 in. The Robatel centrifugal contactor is the only commercially available unit designed for nuclear service.

General Atomic has not been altogether happy with this type of contactor. Although it is not as mechanically complicated as other centrifugal contactors in some respects, it is more so in others. It has a low tolerance for solids, a high cost (\$51,000), and a relatively high holdup time (1.6 min for the total eight-stage organic phase holdup). Furthermore, when it is run at high speeds with low flow, it will overheat. If such a unit were run at very low flow and high rpm, it could be heated above the flash point of the solvent. This overheating problem places severe restrictions on a plant using such a contactor: the unit cannot be operated at less than one-half its rated capacity and, for best results, should not be run below its rated capacity at all. A variable-speed motor is required if such a unit is to be used with variable flow rates.

A recent report has indicated that interstage leakage can occur along the shaft of the unit when polyethylene seals are used. General Atomic is planning to run stage efficiency tests to determine whether such leaks are occurring. It is likely that, for high-level radioactive service, the polyethylene seals will be replaced with stainless steel rings.

Centrifugal Contactors--Savannah River Type
(H. Bethmann, duPont)

Figure 2 shows the duPont-designed centrifugal contactors that are currently in use at the Savannah River Plant and Laboratory. These units

(a) Manufactured by Robatel-SLP1.

FIGURE 1. Robatel Centrifugal Contactor

FIGURE 2. Savannah River Centrifugal Contactor

are sometimes called centrifugal mixer-settlers, since they have discrete mixing and settling stages. A unit of this type costs around \$350,000, including the motor. The contactors are installed in groups of six, known colloquially as "six-packs" (Figure 3).

Design details are shown in Figure 4. In each unit, the shaft, weirs, and bowl rotate. In the extraction process, the aqueous and organic phases first mix and then separate as they travel upward, each discharging into separate annular rings.

Each motor is independently removable, and the flange, air seal, and hollow shaft come out. The hollow shaft is connected to the fixed shaft at a tapered joint; the bowl, paddle, and fixed shaft to which they are attached remain in the contactor.

Interface Control

Interface control is achieved through air pressurization in a weir chamber, which makes it possible for the unit to handle a wide range of

FIGURE 3. "Six-Pack" Arrangement

TBP concentrations--a flexibility that most contactors do not have. Although the pressure is controlled six stages at a time, instrumentation can measure pressure variations at each individual stage, so that if a seal leaks, for example, it can be detected. The pressure does not have to be exact, but must remain within an allowable range.

Reliability and Repair

The contactors have been in service for primary decontamination of Purex feed since October 1966. During this time, they have accumulated around 1,000 hot-feed operating days, with daily flows ranging from 10 to 20 tons. There have been about 27 mechanical failures--primarily of the mechanical seals and bearings, with a few motor failures. However, the number of seal problems encountered is lower than might be expected, given the frequency with which the units are shut down and started up again. Furthermore, of the 18 stages in use at SRL, no one stage has had more than four failures, and the three most radioactive have never failed at all.

FIGURE 4. Design Details of SRL Centrifugal Contactor

When a failure occurs, the operation is shut down, the motor is removed, and a spare motor is put on. Before repair can begin, the motor must be decontaminated. Rough decontamination is done remotely in a decontamination facility, while final decontamination is performed by hand. Repairs are usually completed within approximately 2 hours; the motor is then kept as a spare.

Each six-pack has a wiring harness, with a power supply jumper attached to each harness. Insulation failures have occurred on both jumpers and harnesses for all three six-packs; in all cases, repair and replacement have been successful. Figure 5 shows a six-pack that is ready to be installed in the cell.

Of all the failures that have occurred, only two could be considered major. These occurred in stages 1 to 6 and in stages 13 to 18 (the coldest and hottest stages), and stemmed from cracks in the piping caused by fatigue and vibration (see Figure 6). More bracing was added during repair to correct the situation. In addition, a T weld is now being used instead of a saddle weld.

Applications in the Partitioning Process

Use of these contactors in the partitioning process has been examined in the laboratory. The only problem seems to be elimination of the nitrite; once this is accomplished, reduction of Pu IV to Pu III can be accomplished very rapidly.

Use as Only Contactor Type in Plant

There are both pros and cons to using these units as the only contactors in a plant. Cost is a negative factor, since pump mixer-settlers, for example, are far cheaper. On the other hand, quick startup is a definite plus. With respect to the failure rate, opinions are mixed: some say that extrapolation of the failure rate to a production plant situation indicates that it would not be possible to keep a plant running; others feel that the failure rate has not been unacceptable.

FIGURE 5. "Six-Pack" Prior to Installation in the Cell

FIGURE 6. Fatigue Cracks in Piping

Design Improvements

It was pointed out that this contactor was intended to be an experimental unit, and many design improvements are still possible. The bowl, for instance, although a rotating part, at present cannot be taken out. If just two plates were made removable, however, the design could be simplified considerably: the bowl could be made removable; the hollow shaft could be eliminated; and the rotating seal could be replaced with an end seal, which would be far less likely to fail.

Centrifugal Contactors--Argonne Type (G. Bernstein, Argonne)

Argonne National Laboratory (ANL) developed two modifications for the SRL centrifugal contactor. The objective was to make a contactor that had a high capacity within dimensions that were critically safe. A diagram of an SRL centrifugal contactor is shown in Figure 7. In the initial ANL design, the basic features of the SRL contactor were retained. However, the rotor diameter was reduced to 4 in. so that it could fit within a casing that had a maximum diameter of 5-1/2 in.--a diameter that is considered to be critically safe for all concentrations of plutonium solutions. The rotor is 15 in. long, including a 12 in. settling length, and can run at speeds up to 3500 rpm. (In contrast, the large SRL contactor has a rotor that is 10 in. in diameter; 17 in. long, including a 13 in. settling length; and operates at 1750 rpm.) The capacity of the ANL unit is more than 15 gallons per minute (gpm) at 3500 rpm.

Annular Design

The ANL contactor was designed so that it could be converted into a simpler form, called the annular centrifugal contactor (Figure 8). This design is adaptable to large and small contactors. To effect the conversion, the mixing chamber, paddle, and shaft are removed, and the aqueous and organic streams are fed directly into the annulus between the rotor and the casing, where they are mixed by skin friction.

The first tests of the new design indicated that the annulus was too small, causing throughput to be restricted to 5 gpm at all speeds. Any

FIGURE 7. Experimental Long-Rotor Centrifugal Contactor (Savannah River Type)

FIGURE 8. Schematic Drawing of Annular Centrifugal Contactor

increase over this flow rate resulted in flooding and contamination of the organic phase. To solve this problem, the bottom section of the unit was removed and a new section was installed. This section provided a larger annular gap (0.55 in. versus 0.275 in.) and had feed stream inlets located closer to the bottom. It was attached to the upper section of the contactor with a flanged connection to permit conversion to the SRL design.

These changes increased capacity significantly. The unit has run at as high as 15 to 17 gpm; however, this rate was achieved with organic-to-aqueous ratios that are not realistic for the Purex process. The unit is capable of processing about 1 tonne/day of reactor fuel under Purex conditions. At a rate of 10 gpm, the residence time for both the organic and the aqueous phase would be about 3 sec/stage. Normally, equal volumes of organic and aqueous phases are held up in the rotor, but the interface position can vary, depending upon flow rates and air pressure on the weir. As long as the interface stays between the organic overflow weir and the aqueous underflow weir, however, it will not contaminate the output streams.

One effect of the design changes was steadier hydraulic operation. In the SRL unit, the mixing chamber acts as a pump and tends to cause surging in the discharged streams. The annular contactor has no mixer-pump, making flow through the rotor much more uniform.

The ANL design changes would also simplify both construction and maintenance in a unit designed for plant operation. Construction would be simplified because the rotor can be coupled directly to the drive motor. A simple rotary air seal (like the one initially used at SRL) can then be mounted on top of the motor to supply weir control air through a hollow motor shaft. This design change would eliminate the need for a complicated circumferential shaft seal. Maintenance would be simplified because the entire motor and rotor could be removed for repair, leaving the casing and piping in place. The motor could then be remotely disengaged from the rotor and replaced; the rotor cleaned up or replaced; and the entire assembly put back into the casing.

Tangential Take-Off Duct

One additional design feature, a tangential take-off duct, was incorporated into the annular design for experimental purposes. The designers wanted to see what would happen if some of the liquid was removed before it had received the full mixing treatment. It turned out that there was no difference at all in the performance. This result suggests that centrifugal contactors mix more vigorously than is necessary. This assumption is consistent with Treybal's formula, which states that good mass transfer can be achieved if $P/Q = 200$, where P is the power input in ft-lb/sec, and Q is the flow rate in ft³/sec. In a plastic model of the ANL annular design, power input measurements indicated that with 10 gpm throughput and 0.1 hp, $P/Q = 2500$, a value that is high by a factor of 10. Because a decrease in the mixing intensity would both conserve energy and reduce the tendency to form emulsions (thereby simplifying separation and permitting an increase in the throughput capacity), further examination of this question is warranted.

The tangential take-off duct can also be used to provide increased residence time in the mixing zone for fission products such as ruthenium and zirconium. The same size rotor would be used in an extended casing, which would be fed by a tangential take-off duct from the annular mixing zone.

It is possible that improved decontamination could be achieved by using contactors with differing residence times: those with shorter residence times in the extraction section of a multistage bank, and those with longer residence times in the scrub section. However, this application is still in the conceptual stage.

SOLIDS HANDLING

Redox Plant Experience (R. Smith, ARHCO)

A Redox plant experience was described to illustrate problems that can occur in connection with solids tolerance. The Hanford Redox plant was the first nuclear production solvent extraction plant built in the United States, and was capable of processing 3 to 4 tons of material per day.

FIGURE 9. Interface Control in Packed Column

A schematic design of the Redox plant's original, Raschig ring, packed-column contactors is shown in Figure 9. The disengaging section had a diameter of 12 in.--twice that of the packed section. Table 1 shows the decontamination factors that were achieved.

TABLE 1. Redox Uranium Decontamination Factor
Prior to July 1954

<u>Cycle</u>	<u>Logarithmic Decontamination Factor for Uranium</u>
1st	3.5
2nd	2.2
3rd	0.8
Overall	6.5

In 1954, extra capacity was needed, and the column diameter was enlarged to 8 in. To achieve a higher residence time for the organic phase, the disengaging section was also enlarged; however, because of space constraints, it was enlarged eccentrically. Thus, the previous concentric formation was now bathtub-shaped (see Figure 10). This change in the geometry of the disengaging section resulted in lower decontamination factors, as shown in Table 2:

TABLE 2. Redox Uranium Decontamination Factor
After New Column Installation

<u>Cycle</u>	<u>Logarithmic Decontamination Factor for Uranium</u>
1st	3.7
2nd	1.1
3rd	1.0
Overall	5.8

The poor decontamination resulted from the location of the interface. With the concentric configuration, the solids that accumulated at the interface were uniformly forced downward, so that there was very little

entrainment in the organic stream. The bathtub shape, however, created a nonuniform hydraulic condition that caused the solids to accumulate in the end section along with a pocket of stagnant scrub solution. Thus, the stagnant scrub solution precontaminated the fresh scrub solution, resulting in lower decontamination factors.

FIGURE 10. Enlarged Disengaging Section

The problem was solved by lowering the interface 3 feet into the packed section. This simple change resulted in the decontamination factors shown in Table 3. Not only were these decontamination factors better than those achieved with the original configuration, but they also made it possible for wastes to be back-cycled, which resulted in savings on essential material costs.

TABLE 3. Redox Uranium Decontamination Factor
After Lowering Interfaces

<u>Cycle</u>	<u>Logarithmic Decontamination Factor for Uranium</u>
1st	4.0
2nd	2.6
3rd	0.5
Overall	7.1

Purex Plant Experience

In the Purex plant, the 2D extraction column (final uranium cycle) was run first with a top interface and then with a bottom interface. It was found that a top interface caused a reflux of solids, resulting in lower decontamination factors. Good decontamination factors were achieved when the column was run with the organic phase continuous, allowing the solids to go out the bottom with the waste.

Thorex Plant Experience (G. Benedict, General Atomic)

At the General Atomic Thorex pilot plant, extraction pulse columns are run with a bottom interface, and solids present no problems. The solids, mostly silicon carbide and carbon materials, are finely divided and constitute 0.01 to 0.02% of the feed solution by weight. As long as the interface is at the bottom, the solids go out in the aqueous stream, with no effect at all on decontamination.

In the Thorex process for extracting thorium and uranium, the product stream from the 1BX column contains thorium and a small amount of uranium; it also contains finely divided material that cannot be removed by centrifugation--partly because of its low density and partly because of its small size. However, as long as the column is run with the organic phase continuous, the interface is kept away from the product end and these solids present no problem. In some cases, their volume has been as high as 0.4% by weight, which is enough to turn the aqueous stream black. Even in such extreme cases, however, decontamination factors did not drop; the only effect was a slight reduction in column capacity. Figure 11 shows

FIGURE 11. Interfacial Buildup

the interfacial buildup in the organic-phase-continuous IBX column, and the relative clarity of the product stream.

Formation of Interfacial Solids

No matter how clear the feed streams appear to be, solids will always appear at the interface. What appears to be perfectly clear aqueous feed can be mixed with similarly clear organic feed, and interfacial scum will still appear. No one was able to give a definite explanation for this phenomenon, although it was pointed out that there will always be siliceous materials that come from the water supply, feed impurities, and other sources. At Hanford, for example, an observed discrepancy between the plant's performance in summer and in winter was traced to the presence of humic acid in the river water at certain times--which in turn was traced to the presence of algae. The ion exchange process used for demineralizing the water had no effect at all on the humic acid. The problem was solved by the addition of trace amounts of permanganate to the water treatment system. In a reprocessing facility, where the condensate is recycled, interfacial scum accumulations may never be a problem. However, recycling poses a problem of its own: if the condensate treatment system is not adequate, impurities will be fed back into the contactor. These impurities could cause a variety of difficulties, not the least of which could be inadequate decontamination.

Mistron

A technique developed at Hanford for use in the Hanford Plutonium Reclamation Pilot Plant Facility was the use of Mistron, a finely powdered talc, to counteract the lowering of capacity that tends to occur with the buildup of interfacial scum. In an organic-continuous process, the addition of Mistron to the aqueous scrub stream will raise the capacity significantly. A layer of Mistron emulsion does build up in the interface, but eventually works its way out in the aqueous raffinate. There is one disadvantage to this practice, however: the Mistron carries some organic with it, which will end up in the aqueous waste. If there is anything in that stream that cannot tolerate organic, problems could result.

Zebra Cartridges (G. Richardson, Hanford)

Zebra cartridges, which consist of alternate groupings of plastic and stainless steel sieve plates, were developed for use in the HA and 2D columns at Hanford. The cartridges, together with stainless steel nozzle plate extraction sections, permit operation with the organic phase continuous, thus flushing fission product solids to the waste end of the column. However, pilot plant tests have shown that the zebra dispersion in the scrub section traps some interfacial solids. The highest concentration of the solids occurs at the top of the cartridges, as shown in Figure 12.

Other Hanford plant columns have successfully used zebra cartridges to improve capacity and efficiency in aqueous-phase-continuous columns. For such columns, plastic sieve plates replace nozzle plates at intervals ranging from every 4 inches to every 2 feet.^(a) The effect of the plastic plates is to coalesce the fine organic dispersion into large drops that move readily upward to the next nozzle plate section, thus improving the driving force for countercurrent flow. Coalescence and redispersion also cause the formation of fresh surface area, which is believed to improve mass transfer.

Opinion about the usefulness of plastic plates was divided. One participant stated that, if capacity is to be maintained, a 5% TBP solution cannot be run organic continuous without the use of plastic plates. Another achieved good performance in terms of both operation and decontamination with no plastic plates at all in the extraction column.

There are a number of problems involved in the use of plastic plates. Some forms of plastic cannot tolerate radiation. Linear polyethylene, selected specifically because of its radiation resistance, was found to become brittle after prolonged use--even in a cold plant situation. Since pulse columns require plates that flex, this form of plastic had to be abandoned.

(a) It is important to note that zebra cartridges must be tailored to specific flowsheet conditions as determined by pilot plant testing.

FIGURE 12. Solids Holdup in the Scrub Section
with Zebra-Type Dispersion

Fluorothene has been used with some success. It has less radiation resistance than some forms of plastic, and tends to develop interfacial buildup, but it does stay intact.

Removal of Interfacial Solids

Interfacial Slurpers

Various techniques have been developed for removing interfacial scum. One, an interface "slurper," was developed in the Hanford Plutonium Reclamation Pilot Plant Facility. This device made use of a filter and coalescer to remove the solids; afterwards, the phases were separated and then directed back into the column.

Flushing

In mixer-settlers, interfacial buildup will form in spite of feed clarification, but will not affect performance.^(a) After about a year and a half of operation, however, the accumulation of solids may begin to cause plugging problems. At this point, the system is cleared out with a caustic flush. The process is as follows: 1) product removal with water; 2) flush with strong (25-50%) caustic solution; 3) water flush; and 4) acid flush. The whole process can be done in as little as 24 hours, but generally takes 2 to 3 days.

In a group of mixer-settlers, not all will require the same frequency of flushing. The 1A unit tends to need flushing the most: it may go through two or three flushes before the others need any at all. However, the entire system has to be shut down, even if only a single contactor is being flushed.

(a) It was noted that the French did experience a solids problem with their mixer-settlers, caused by zirconium DBP precipitate. They solved the problem with the addition of fluoride (see P. Faugeraf and X. Talmont, "Radiolysis and Hydrolysis of TBP and Their Effects," in Solvent Extraction Research, ed. by S. Kertes and Y. Marcus, Wiley-Interscience Publications, p. 411-423, 1969). At Hanford, however, where a laboratory study used miniature centrifugal contactors to investigate solvent stability at LMFBR radiation exposures, no zirconium DBP precipitates were observed.

Flushing of mixer-settlers is facilitated by adding a weep hole in the bottom of each of the stages, to permit complete draining.

Solids and Safeguards

It was pointed out that, even if a particular contactor type can handle solids, safeguards regulations may permit no solids in the recycle of LWR fuel because of the plutonium present in such solids. For example, a recent report⁽²⁾ on partitioning states that, following the dissolution of a sample of LWR fuel, 0.3 wt.% of the uranium in the fuel and 0.7 wt.% of the plutonium in the fuel remained in the undissolved fraction.

FEED CLARIFICATION

The general consensus was that feed clarification is desirable. Preferences were expressed for different types of equipment, but all agreed that remotability was an important requirement.

Many people still use the solid-bowl, batch centrifuge for feed clarification--the same piece of equipment that was used 25 years ago. AGNS uses a Robatel remotable batch centrifuge. General Atomic has placed an order for a Sharples continuous-flow, vertical configuration centrifuge, to be used for clarification of feed prepared from HTGR fuels. This type of feed is particularly difficult to clarify because of the large amount of silicon carbide present.

The Sharples centrifuge is similar to the Robatel except for the fact that it is continuous--that is, it discharges the sludge as it is centrifuged out. It includes a washing step, and it may be connected directly to a sludge dryer. As currently sold, the Sharples centrifuge is not remotable; however, General Atomic hopes to be able to modify it for remote removal. Its vertical configuration makes it a particularly attractive prospect for such conversion. The fact that it is belt-driven may present some complications; however, these can be handled by leaving the belt outside the cell and building a shielding plug.

The Hanford Plutonium Reclamation Pilot Plant Facility has several centrifuges, but they all have to be stopped, dismantled, and scraped out by hand.

There is a commercial continuous centrifuge available that does not have to be shut down for sludge removal. The centrifuge cracks open periodically by means of a hydraulic control, and, using hydraulic pressure, pushes the sludge out. The unit then closes and begins collection of another batch. With this type of centrifuge, continuous operation can be maintained.

Filters have been used with some success in waste solidification process development. Product filters have also worked well--for example, in separating solid plutonium oxalate from a nitrate solution. However, it was felt that such filters would not work well for feed clarification, both because of the higher radiation levels and because of the remotability requirement. Etched disc filters are another possibility; programs are currently under way to develop the technology to make them fully remotable.

SOLVENT DAMAGE

Solvent damage is frequently cited as a reason for using centrifugal contactors. It is expected that the lower residence time in such contactors will result in less damage to the solvent from radiation exposure. Shorter times could have a second benefit, as well: it is possible that the kinetics of formation of DBP and other solvent-damaging complexes may be such that they do not have time to form when residence times are kept short. It was also suggested that droplet size, which varies among contactor types, may affect the amount of radiation damage that occurs.

However, several participants felt that solvent damage is not a valid reason for selecting centrifugal contactors over other contactor types. Of the three reasons frequently cited for using a centrifugal contactor (cost-effectiveness, quick startup times, and less radiation damage to solvent), they felt that solvent damage was the weakest.

General Atomic used a tracer system to study the effects of DBP formation in the Robatel and in pulse columns.⁽³⁾ In these tests, which were aimed at reducing the deleterious effects of DBP on the system, no difference was found between the two types of contactors. One participant, however, questioned the validity of the tracer method, stating that

testing in a fully active system was necessary to determine the mechanisms involved.

At Hanford, a comparison was made between centrifugal contactors and other types. The study showed no significant difference in decontamination or solvent damage in one extraction cycle, when using feed prepared from either LWR or LMFBF fuel.⁽⁴⁾

At Oak Ridge, solvent damage studies were done on the Thorex process. More solvent damage occurs in a thorium system than in a plutonium system, due to the higher radiation levels involved.

Two unanswered questions are at the crux of the solvent damage issue. One is whether a shorter residence time will significantly reduce the effects of solvent damage. If it will, as seems to be the case, and if such a reduction significantly affects performance, then there seems to be an advantage to using centrifugal contactors in the extraction section. However, these "ifs" are major ones, and no one can give a definite answer at this point. The other question is to what extent solvent damage is really a problem: that is, how much DBP can a system have in it and still continue to operate? R&D is currently under way in an attempt to resolve both of these questions.

SOLVENT WASHING

At present, solvent is washed at every pass. It was suggested that, with a shorter residence time, fewer solvent washes might be needed. This would be a highly desirable change, since washing creates a substantial amount of waste. However, as one participant pointed out, to determine whether washing is necessary with each pass may turn out to be more costly than simply to continue doing it.

One stage seems to be sufficient for solvent washing. At Hanford, for example, where solvent is circulated through a 4 ft section of packing in the top of the tank, one pass is all that is needed to clean the solvent.

Carbonate Flushes

A good basic solvent wash--for example, a carbonate wash--will dissolve DBP, break up the metal complexes, and take out much of the unfixed iodine. (Removal of the fixed portion of the iodine, however, is almost impossible.) The carbonate wash works by neutralizing the acid in the solvent: the less nitric acid there is in the solvent, the more times the wash solution can be reused. In the process of neutralizing the acid, the carbonate turns to bicarbonate, which is not basic enough to remove DBP or iodine. Consequently, it is important to make sure that the pH of the wash does not drop too low. The washing solution may either be used until all the carbonate is gone, and then be replaced on a batch basis, or it can be used in a continuous manner, in which case a constant pH is maintained by bleeding off old solution and bleeding in new solution.

Macroreticular Resin

Macroreticular anion resins are excellent cleaning agents, although they may not be economically suited for routine use. They produce a substantial amount of waste when regenerated; however, if the solvent is carbonate-washed before it is passed through this additional ion-exchange step, the ion-exchange capacity of the resin will be greatly increased and the solvent will come out as good as (or better than) a new batch. General Atomic plans to use macroreticular resin as a polishing step, to make sure that there is no U-232 left in the thorium product. Like the carbonate wash, the resin wash works by neutralizing the acid in the solvent. Consequently, the more nitric acid that can be removed from the solvent prior to the resin wash, the less resin will be used in the process.

Caustic Washes

A caustic wash can precipitate uranium, whereas a carbonate wash does not. Sometimes a combination of carbonate and caustic wash is used. In such cases, it is important that the carbonate be used first to avoid precipitation of the uranium. At Hanford, a mixed wash was tried as a means of prolonging the usability of the solvent and keeping the pH up; however, the method was not satisfactory, because of uranium precipitation.

Type of Contactor Used for Solvent Washing

Solvent washing is an easy process and can be performed in a variety of types of equipment. One participant suggested a modified turbomixer as the best type of contactor for carbonate washing. At General Atomic, however, carbonate washing is done right in the pulse column, although published studies have recommended against this operation because of erratic flooding behavior. After the washing process, the solvent is filtered.

Another participant has successfully used pulse columns for carbonate washing in nonradioactive systems, but has run into difficulties in radioactive plant operation. It was suggested that the key to successful carbonate washing in pulse columns might be the presence of solids that change the interfacial properties of the emulsion; and that thorium hydroxide, which is always present to some degree in the General Atomic Thorex work, might be the particular solid involved. Another speculation is that pulse column solvent washing can be used as long as the carbonate solution is not recirculated and does not approach bicarbonate pH.

RED OIL

Some participants have found red oil hard to make in the laboratory; others have found it easy to make. However, it appears that there is more than one kind of red oil. The type that General Atomic is unable to make with the use of an NPH diluent is a highly explosive, concentrated nitrate that detonates at 135°C. The type that SRL has made successfully is a TBP-uranium complex concentrate in a third phase. The product is yellow when pure, but turns red when it is put in solution with TBP and heated. SRL has full documentation available on this form of red oil.

VALIDITY OF DEVELOPMENT WORK FOR PLANT OPERATION

Design of Mixer-Settlers

With rotating equipment, where the stages are well mixed, scale-up is relatively easy. DuPont has been successful in going from miniature-scale

testing to full plant operation with both pump mixer-settlers and centrifugal mixer-settlers.

Design of Pulse Columns

Presumably, knowledge of pulse columns is such that a pulse column can be designed and its characteristics known with a great deal of accuracy. Nevertheless, pulse columns are not as easily scaled up (or down) as are mixer-settlers.

At Hanford, for example, serious problems occurred when a uranium extraction pulse column system with a diameter of 16 in. was scaled up to 20 in. to permit it to handle a few more tons per day. The problems, which stemmed from the channeling of the two phases, were solved by the installation of swirlplate mixers at 3 to 4 ft intervals. The instance serves as an example of the difficulties encountered with even relatively small changes of scale in pulse column systems.

Similarly, it would be difficult to go directly from hot cell work using minimixer-settlers to full-scale pulse column operation. Although many of the pulse column characteristics could be extrapolated, the HTUs would need to be verified through a cold test of the cartridge.

The rate of extraction serves as a good guide as to whether pulse column operation will be feasible. If adequate (near 100%) extraction is achieved in 30 sec or less with batch extraction, the operation probably can be done in a pulse column. On the other hand, if 2 or 3 min are required to reach equilibrium, pulse column operation probably will not be feasible, because of excessive transfer unit heights.

Solids Tolerance

Many of the problems that occur in the translation of laboratory work to full-scale production plant operation have to do with solids tolerance characteristics, which tend to change with variations in equipment size. For a more detailed discussion of these problems, see Solids Handling.

Temperature

It was generally agreed that laboratory work on temperature applies very well to full-scale operation. The production plant results are either the same as or better than the pilot plant predictions.

Use of Organic Continuous Phase in Pulse Columns

The laboratory predicted the beneficial effects of a switch to organic-phase-continuous extraction columns. The production plant results agreed almost exactly with what was predicted concerning solids handling, capacity, operating characteristics, fission product decontamination, etc.

Need for Cold Testing of Equipment

Difficulties arise in taking a design directly from the laboratory to a full-scale production plant without cold-cell testing. If there is no intermediate step to develop the necessary devices for achieving flow control, adequate agitation, etc., it is very likely that the equipment will have to be reinvented after the building of the production plant.

In the 1950s this was not so much of a problem, because production plants could also be run as test facilities. Current licensing regulations, however, do not permit this flexibility. Today, the plant that is built must be the same as the plant that was designed--or else relicensing is required. Had similar regulations been in effect when the first Purex facility was built at Hanford, that plant would have gone through several relicensings before reaching its current form.

Because the cost of building a plant today is about \$1 billion, as opposed to \$85 million to \$90 million for the Purex plant, adequate demonstration of equipment performance between the laboratory work and the production plant is vital. The interest on a billion-dollar plant could support a substantial amount of testing.

When the funding comes from the government, the necessary testing steps tend to be included: the LMFBR program, for example, includes scale-up of component development, integrated systems, and equipment. However, in the LWR programs, which have been funded by industry, plants have been built without adequate cold testing.

Feed Clarification

It was generally agreed that feed clarification equipment should be tested in a cold-plant situation before being installed in a full-scale production plant.

Flow Sheet Feasibility

An important factor in translating laboratory results into full-scale plant design is the flowsheet itself. If the flowsheet specifies results that are difficult to obtain, the equipment cannot be expected to work without extensive cold-plant testing and development. In this regard, a participant with mixer-settler experience pointed out that he would never try to achieve an extraction factor greater than 4. Another agreed, stating that he would rather put in four stages than one, so that he would not have to worry about effects of entrainment or back-mixing.

Participants with pulse column experience pointed out that both terminology and feasibility differ according to contactor types. First, mixer-settlers are treated as having stages while pulse columns are treated as having transfer units. Second, a set of flowsheet conditions that is difficult to meet with one type of contactor may not be so hard to meet with a different type.

INSTRUMENTATION

In any plant, the proper instrumentation is important to smooth operation. At the Purex plant alone, there are some 5000 instruments, and about 1200 to 1400 instrument systems. Many of the plants are still using instrumentation that is 15 or more years old, and when these instruments need repair, parts frequently are not available. SRL has run into this problem with the Taylor transceiver devices and is starting to replace them with Fisher-Porters. Other problems arise from the need for specific types of instruments that have not yet been designed. Some of the types of instruments available, as well as those that are still needed, are described below.

Pneumatic vs. Electronic Instrumentation

Idaho Falls currently uses pneumatic equipment for controlling flow, levels, etc. The plant is now planning to convert to electronic equipment, and hopes at some point in the future to computerize. Pneumatic-to-electronic converters are now available in a highly compact form. They fit right on top of the valves, eliminating both the large converter and the extensive copper tubing that used to be required.

SRL has been using a computer for contactor flow control, although tank depletion is still the primary method.

Ratio Control

The Hanford Purex plant was built for ratio control, but the system was never used because there were too many problems with it. It was suggested that some of the problems stemmed from the use of an all-pneumatic system, and could be eliminated by converting to an electronic system.

At SRL, however, the centrifugal contactors use an all-pneumatic system and have no problem with ratio control. The solvent flow rate is fixed and the scrub is varied according to changes in the specific gravity of the solvent effluent stream: that is, as the specific gravity goes down, the feed is increased.

Flow Meters

At Idaho Falls, the low flow rates and the relatively large amount of undissolved solids have led to difficulties in finding good flow-measuring devices. The plant has no flow meter per se, as none that works reliably has been found. In lieu of a flow meter, the feed rate to the column is metered by tank depletion. A device in the feed tank automatically takes the derivative of the liquid level and converts it into a flow rate.

Feed Tank Types

At some plants it is difficult to measure feed tank levels because, at the same time as feed is going out, new feed is being put in. Other plants use dual tanks, or single tanks with noncontinuous flow.

SRL, for example, uses dual tanks for cold feed. For hot feed, it uses a single tank, but one that is filled only when depleted, rather than

continuously. The intercycle generally has separate feed tanks for inflow and outflow, but the intracycle is continuous, with no holdup.

At Hanford, individual tanks are used for scrub streams. Like the SRL hot feed tanks, they are filled only when depleted. Monitoring of the feed to the extraction columns is done by rotameter. Both the intercycle tanks and the solvent tanks have continuous inflow and outflow.

Neutron Monitors

At Hanford, neutron monitors have been installed on the 1BX column and on the feed makeup tank to determine plutonium concentration. They work well and are very sensitive; however, their primary use is to indicate change rather than to specify a particular plutonium concentration. SRL uses neutron monitors in a similar manner: to record changes that occur. When the device shows a change, a check is made to see which flow rate is wrong or which composition is incorrect.

Interface Control (G. Offut, Allied Chemical)

Interface control in pulse columns at Idaho Falls is achieved by means of a pressure weir (Figure 13). The liquid is forced to flow from the bottom of the column up and over the weirs, where air pressure is used to control the interface. A similar control is used in other plants for mixer-settlers.

Density-Measuring Devices

Various participants expressed the need for a good density-measuring device. Several use a Dynatrol; however, it was suggested that the instrument would be better if it had more sensitivity to slight changes in density. For instance, with its present capability, the presence of just a few drops of aqueous phase can produce a very misleading measurement of organic phase density.

Ultrasonic density-measuring devices were also discussed, but there are two major problems with this type of instrument. One is finding a device that will withstand radioactivity. The other is that the presence of solids in the liquid tends to throw off the measurements, since the instrument cannot differentiate between these and the wall of the container.

FIGURE 13. Pressure Weir

Safeguards

Safeguards regulations can affect instrumentation in two ways: 1) they can eliminate certain types of instrumentation that are not sufficiently tamper-resistant; and 2) they can create the need for additional instrumentation. For example, bubbler tubes might be ruled out as a means of measuring density, or they might have to be equipped with alarms that are activated by a change in density.

Current Status of Instrumentation

Most of the new instrumentation available today has not been tested in radiochemical service. There is a great need for applications development for all new instruments, particularly for testing their performance in a radioactive zone.

Perhaps the greatest need for new instrumentation is in the area of in-line chemical analysis. Instead of having to send samples to the

laboratory periodically and wait for results, continuous in-line monitoring of uranium, plutonium, and zirconium concentrations could be performed. Unfortunately, funding for any type of instrumentation development is difficult to obtain right now, but it is hoped that this situation will change.

COMPARISONS AMONG CONTACTOR TYPES

For each of the subject areas that were discussed, comparisons were made among the various types of contactors. The results of these comparisons are presented below.

Rangeability

For pulse columns, there was some disagreement as to rangeability: one participant stated that a pulse column could process anywhere from 5 to 30 tons/day, and perhaps even more. Another said 5 to 11 tons, and another said that rangeability can be calculated as a factor of 4.

Centrifugal contactors are able to handle 20 tons or more, rangeability being limited only by the flow meter. Low flows can be a problem with the Robatel, which tends to overheat in such conditions. This problem does not exist with the other types of centrifugal contactors, however. In the SRL design, power input is linear with throughput. Power is dissipated by discharging the liquid at the periphery; if there is no liquid to discharge, there is also no power being used. The ANL contactor not only has no heat dissipation problem, but it has no flow minimum. If the feed is stopped, the level in the annulus simply drops and the liquid stays in the rotor. Since no mixing is occurring at this point, there can be no overheating problems. Even with fairly low flows, good efficiency is obtained.

Setting aside these variations among different centrifugal contactor designs, it can be said that, between pulse column contactors and centrifugal contactors, there is no significant difference in rangeability.

Residence Time

Pulse column organic holdup on a theoretical-stage basis is under 2 min/stage on the average, and can go as low as 1 min/stage. The range for pump mixer-settlers is about the same--1 to 2 min/stage.

In the centrifugal mixer-settlers, organic phase holdup is 30 sec/unit. With the SRL design, as the flow rate is decreased, the residence time rises both in the mixing chamber and in the settler. With the ANL annular design, a lower flow rate results in a decrease in mixer zone holdup; thus, there is less increase in residence time than in the SRL design. However, users of the SRL units pointed out that the mixing residence time is so short (they can process 60 gpm) that this difference is not significant.

The Robatel contactor has a total holdup time of about 1.6 min for all eight stages; this is better than pulse column operation only by a factor of 5. When the flow rate of the organic phase is pushed to the maximum, the holdup has actually been below 1 min, but never as low as 30 sec. Furthermore, to get this sort of figure, the unit must be run well above the design flow rate. An advantage of the Robatel, however, is that it is commercially available right now, whereas neither the SRL nor the ANL centrifugal mixer-settlers are available to commercial users at present.

Solids Tolerance

Pulse columns are capable of handling solids in the feed, provided that plugging of the lines to and from the columns can be avoided. This is accomplished by keeping the interface away from the product end. Use of a zebra cartridge has also been found to help (see Solids Handling: Zebra Cartridges).

In spite of the general experience that pulse column plates do not become plugged under any circumstances, one participant described a case in which the top 2 or 3 ft of an aqueous continuous column had become

plugged.^(a) Other participants described certain situations where they felt that prior feed clarification was necessary to avoid problems. At Hanford, for example, where pulse columns are used to process first-cycle wastes that contain a high proportion of solids, wastes are always centrifuged before being fed into the columns.

A standard mixer-settler has satisfactory solids tolerance as long as the feed is centrifuged. Operation without such feed clarification has never been tried, since it is generally felt that solids would interfere with the contactor's performance. In spite of the feed clarification step, interfacial scum does form, but does not appear to affect performance.

The SRL and ANL centrifugal contactors have no trouble with solids. Solids accumulate until an equilibrium point is reached and the remainder go out with the aqueous solution. However, feed clarification is still required.

The small Robatels do not handle solids well. In the newer units, which are larger, the solids problem is less serious due to the addition of a spray flusher in the central shaft above the aqueous inlet. This flusher can be used to remove any solids that are plugging the aqueous inlet. With all Robatel units, however, a feed clarification step is necessary. The recommendation is that the centrifuge used for this purpose develop three to four times as many Gs as the centrifugal contactor. However, at the General Atomic pilot plant, the only feed clarification step being used at present is a filter in front of the contactor. With this mode of operation, the contactor is expected eventually to become blocked, but disassembly and inspection to determine the location of the plug are part of the experimental plan.

(a) It was suggested that the problem may have been caused by the formation of a DBP complex at that point. Some DBP compounds--thorium-DBP, for example--are very difficult to dissolve. Heat was suggested as one possible means of solving the problem. At General Atomic, low concentrations (.005M) of fluoride ion have been used successfully for separating thorium and DBP in the top of the thorium partition column.

Remotability

Mixer-settlers are fully remotable, but they have very few mechanical problems, and it is very rare that their motors need replacement. Wiring insulation breakdowns have occurred, but the harnesses are remotable and easily repaired.

In the SRL centrifugal contactors, the motor, flange, air seal, and hollow shaft are all removable. The ANL annular units, which have no bottom mixing chamber or paddle, are fully remotable. In the commercial-size Robatels, the internals (rotor and bowl) can be lifted out and replaced remotely; the casing remains behind.

Maintenance

Of the three contactor types, maintenance costs are probably highest for the existing SRL centrifugal mixer-settlers. However, as was pointed out earlier (see Centrifugal Contactors--Savannah River Type), the contactors should not be placed in a production plant in their present form. A much simpler unit could be designed without too much difficulty; such a unit would be far less likely to have high maintenance costs.

Operation with a Third Phase

Pulse columns have no trouble handling a third phase. This is one area in which this form of contactor may have an advantage over the centrifugal type.

The ANL centrifugal contactor probably will not work with a third phase. The Robatel can do so, but not without problems. If the flow rate is too high, the heavy organic phase can be forced out the aqueous outlet; a surging effect can also occur. If the flow is right for the size of the unit, the contactor seems able to handle the three phases, and will send both organic phases out the right pipe.

Diluent and TBP Losses

With pulse column operation, the TBP concentration tends to decline, and TBP must be added periodically to keep the TBP-diluent ratio correct. With SRL centrifugal contactors, it is the diluent that tends to evaporate and needs to be added from time to time.

Cost

Studies have indicated that the cost of the equipment going into the plant is a very small fraction of the total cost of the plant. However, the selection of the contactor type can influence the cost of the cell--for instance, a cell for equipment requiring high head room will cost more than one for equipment that is more compact.

Performance

The Robatel's stage efficiency cannot be accurately evaluated with the current flowsheet. Its users plan to run it first on thorium only and then on uranium only to make this evaluation.

In general, however, there appear to be no major performance differences between the various contactor types--they all do the job equally well. If short residence time is not required for decreasing solvent damage (see Solvent Damage), then the only advantages for the centrifugal contactor are ease of flushout, startup speed, etc.

Summary

The consensus was that all of the contactor types work, and that LWR and LMFBR reprocessing plants can operate successfully with all pulse columns, all centrifugal contactors, or all mixer-settlers. Thorium-uranium reprocessing plants, however, should use pulse columns, because of third-phase considerations.

Aside from personal preference, the deciding factor may be the layout of the particular canyon in which the contactor is to go. If there is already a 25 ft high concentrator, a pulse column may be the best choice. If space is very limited, it may make more sense to pick a centrifugal contactor. Conceptual design studies would be helpful for making such decisions.

CRITERIA RANKING FOR CONTACTOR TYPES (J. Jarrett, PNL)

The group was presented with a tentative list of criteria for contactor system selection, grouped into three categories in descending order

of importance. After some discussion, the following categorization was generally accepted:

MUST BE CONSIDERED

- Criticality control
- Safeguards control (if mandated by NRC)
- Capacity
- Performance
- Cold testing capability

VERY IMPORTANT

- Impact of contactor choice on total plant cost
- Reliability (maintenance and operating costs)
- Maintenance philosophy/mandate regarding radiation dose

IMPORTANT

- Cost of contactor plus auxiliary equipment
- Development and engineering effort needed (solvent extraction equipment is generally in good shape; auxiliary equipment needs work)
- Tolerance for solids
- Rangeability, residence time
- Availability of equipment
- Solvent damage, solvent washing (may be a "must"; R&D will tell)

It was pointed out that for a given plant in a given set of circumstances, certain criteria could change category--that is, a criterion that in general is only "important" might, for a particular plant, be a "must." However, it was felt that, for general purposes, this list presents an appropriate ranking; and that, in any case, it provides a good starting point for thinking about design considerations in new plants.

PREDICTIONS AND RECOMMENDATIONS

Participants were asked to express their opinion on (1) which contactor type is most likely to be selected for reprocessing plants of the

future, and 2) which contactor type they would recommend for such plants.

The plants in question would be:

- used for LWR fuel reprocessing,
- based on the Purex flowsheet,
- designed beginning in 1979, and
- built in the U.S. sometime in the 1990s.

The results of the poll are presented in Table 4.

TABLE 4. Contactor Types for LWR Plants of the Future

<u>Contactor Type</u>	<u>Most Likely to be Used</u>	<u>Recom- mendation</u>
Pulse column	3	6
Mixer-settler	0	0
Centrifugal contactor	1	2
Combination:		
Centrifugal contactor, pulse column	11	3
Centrifugal contactor, mixer-settler	0	2
All three types	1	0
No comment	0	3

NEED FOR MORE RESEARCH AND DEVELOPMENT

There is a need not only for development work (that is, applying work done in a given set of circumstances to a different set of circumstances), but also for research: for gaining an understanding of what is occurring and explaining it in familiar terms. Although most engineering research stopped several years ago, ignorance still exists about many aspects of the solvent extraction process. The solution to a particular problem may be known, but there may be no understanding of why the solution works, or what caused the problem in the first place. If the answers to such questions were known, it might be possible to develop a better design that would eliminate the problem. Such answers would also tend to make tasks such as pulse column design more of a science and less of an art.

The group outlined the following areas in which R&D is particularly needed:

- In-line analytical equipment for chemical analysis
- Further development of centrifugal contactors
- Feed clarification equipment
- Pulse column capacity (including criticality control)
- Solvent damage
- Applications development of commercial instruments
- TBP removal from aqueous products and wastes

It was noted that between the LWR and LMFBF programs, research is already under way on all but pulse column capacity, and a proposal to do work in this area is currently under consideration.

Aside from the question of new research, a good look should be taken at what is known now that was not known several years ago when R&D came to a halt. Much of this knowledge could be applied today to improve equipment design.

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