Development and Performance of Centrifugal Mixer-Settlers in the Reprocessing of Nuclear Fuel

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Abstract

An 18-stage centrifugal mixer-settler, designed and developed at the Savannah River Laboratory (SRL), successfully separated plutonium and uranium from radioactive fission products at the Savannah River Site (SRS). This paper discusses the design of the mixer-settler and tests that were conducted before this equipment was installed at SRS and subsequent performance with radioactive solutions. Advantages of the centrifugal equipment over the pump-mix mixer-settler are associated principally with the fiftyfold reduction in liquid volume holdup, and include reduced exposure of the solvent to radiolytic degradation, increased nuclear safety, easier flushing, and greater operating flexibility. The unit has a capacity of over 60 gpm total flow.

In the new mixer-settler, pumping, mixing, separating, and decanting are all performed in a single device with a single rotating shaft. A particular feature of the new mixer-settler is the use of air pressure in a weir chamber to regulate remotely the emulsion position in the centrifuge. The centrifugal stages have high mass transfer efficiency; for example, an overall efficiency of approximately 95% is attained in stripping uranium from solvents. The unit has performed well since start up in October 1966. Maintenance requirements have been small, decontamination and losses are satisfactory, the solvent picks up less then one-fifth as much gamma activity as it did in the pump-mix mixer-settler, and shutdowns and startups are made rapidly without extensive flushing.

Introduction

Uranium and plutonium are recovered from irradiated actinide fuels and targets predominantly by extraction processes using kerosene-diluted tributyl phosphate as a solvent. Radiation from fission products damages the solvent and decreases the separation efficiency of the process. Because the radiation damage to the organic solvent varies with the time that the solvent is exposed to ionizing radiation and because this exposure time is determined primarily by the time required to separate the aqueous and organic phases, the use of centrifugal settling should clearly be superior to gravity settling.

The centrifugal extraction equipment available commercially was not designed for use with radioactive solutions and was not amenable to the remote maintenance procedures required for such service. Consequently, the Savannah River Laboratory (SRL) developed a centrifugal mixer-settler that met these requirements (see references). Three 6-stage assemblies were built, tested at a non-radioactive semiworks facility, and installed in SRS as part of the Purex process. The three assemblies were joined into an 18-stage bank that replaced a 24-stage bank of pump-mix mixer-settlers used for co-decontamination of uranium and plutonium from fission products. These centrifugal mixer-settlers were expected to have the following advantages relative to the pump-mix mixer-settler:

- Reduced exposure of solvent to radiation
- Reduced aqueous and solvent inventories
- Reduced space requirements
- Easier flushing for process changes
- Greater safety in handling fissionable materials
- Accommodation to a wide variety of process solutions with varying densities and viscosities

Design and Mode of Operation

Each stage of the centrifugal mixer-settler, shown in Figure 1, has a 5 HP, 1745 rpm motor with a vertical, overhung shaft to which is attached a 10-inch-diameter separating bowl and, at the bottom, a mixing paddle-pump. Both the heavy (aqueous) and light (organic) phases flow by gravity from adjacent stages and enter the pumping-mixing chamber through the tee at the bottom. The phases are mixed by the paddle and ejected at the periphery of the mixing chamber into an upper chamber where the mixture moves inward along antivortex vanes. The vanes convert some of the rotational energy to pressure so that the mixture can be injected into the bottom of the rotating bowl through a central nozzle. An inlet diversion baffle, located a short distance up in the bowl, prevents any further axial motion of the mixture and diverts it radially (small holes near the center of the baffle, however, allow axial passage of entrained air). The liquids are quickly accelerated to full rotational speed by eight radial vanes that extend the full length of the bowl. The mixed phases separate rapidly in the high centrifugal field (300-500 g), with the heavy (aqueous) phase collecting near the wall and the light (organic) phase collecting near the center. As shown in Figure 2, the organic phase flows inward and over a circular weir in the center of a baffle located at the top of the separating section, and is thrown outward through four, straight-sided, radial ducts to a collector in the stationary casing. At the bowl wall, the aqueous phase passes “under” the circular baffle, then flows inward through the four passages between the radial organic phase ducts into a pneumatically pressured weir section. After overflowing the circular weir in

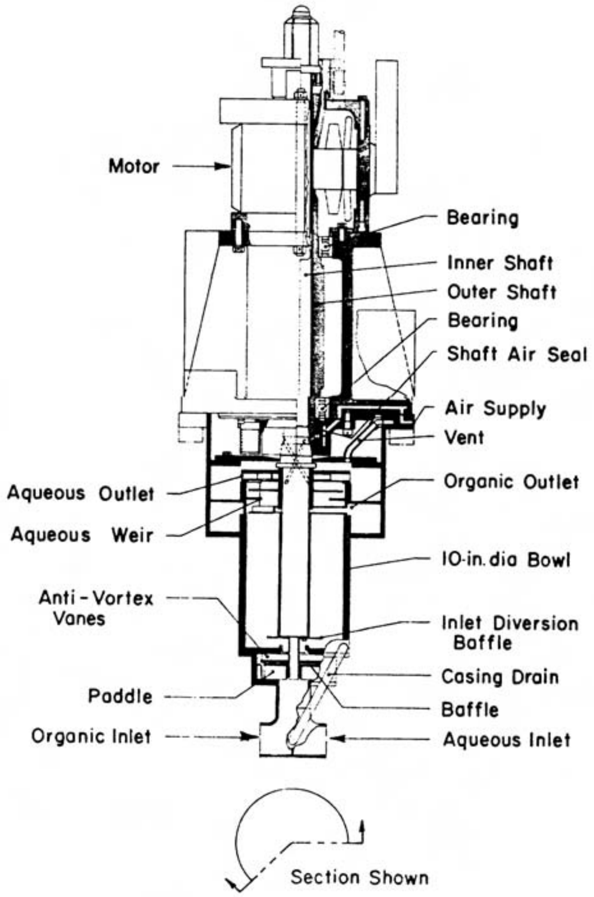

Figure 1. Centrifugal mixer-settler

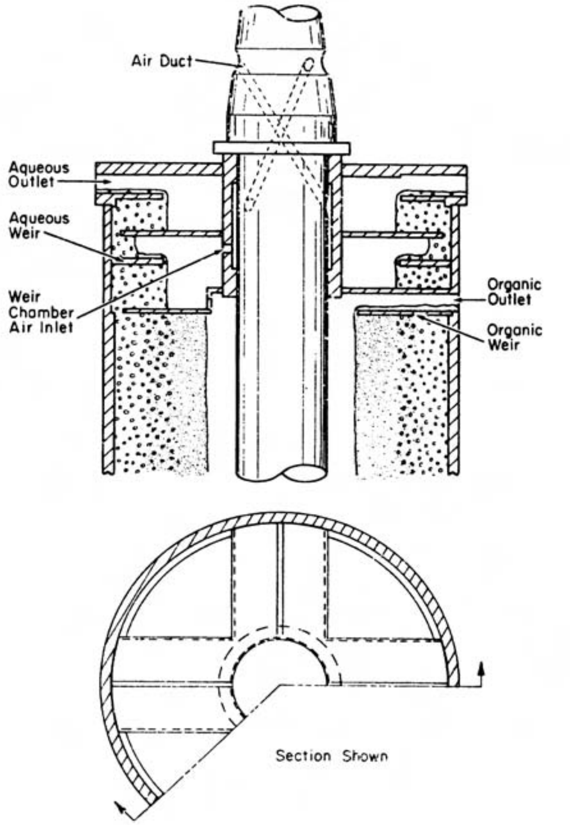

Figure 2. Outlet weir detail

this pressurized chamber, the aqueous phase passes through a circular jack leg seal and is discharged radially into its collector ring. The pressurized weir section allows remote emulsion positioning in each stage. For example, when air pressure is increased, the emulsion moves inward and the surface of liquid in the jack leg moves outward, in compensation.

The two phases flow counter-current between stages. The hold-up of each stage is approximately four gallons (three gallons in the bowl). Compressed air for the weir chamber is transferred via a “Koppers” (Koppers Company trademark) floating bushing air seal from the stationary encasement to ducts in the rotating shaft without significant pressure drop and with a continuous air leakage of less than 1.5 scfm.

Six stages are mounted to form an assembly. Each assembly, with its permanently mounted piping and wiring, and each motor of the assembly, including all bearings and mechanical air seals, are remotely replaceable. Three of the assemblies are piped together remotely to form the 18-stage extraction-scrub bank.

Instrumentation

Each assembly has one control system to supply air at a controlled pressure to the weirs of its six stages. The pressure and total airflow to all six units are recorded in the central control room. Flows to individual stages are indicated by rotometers located just outside the shielding wall. Failure of a seal causes an increase in the total air flow, and the defective seal is then identified as the one with the greatest air consumption, as shown on the individual rotometers.

The power supply to an assembly is divided between two circuits, each serving three stages. The current in each circuit and the difference in circuit between the two circuits are measured and indicated on the central control panel. Motor failure or improper hydraulic operation of a stage causes an unbalanced load and a measurable current difference.

The 3 streams to the 18-stage extraction-scrub bank are the radioactive aqueous feed to a center stage of the bank, an organic extractant to one end of the bank, and an aqueous scrub to the other end of the bank. Successful operation of the process depends upon controlling the flow ratio of the phases to maintain a high uranium concentration in the central stages of the bank. Flow of each stream is controlled by a conventional loop system with a flow measuring device (e.g., a rotometer), a recorder, and a stack controller that delivers an output signal to an air-operated valve or a variable speed pump. The extractant flow is set manually at a value corresponding to the desired processing rate. Flows of the two aqueous streams are controlled relative to this extractant flow by a ratio controller in the controlled loop of each aqueous stream. The ratio of scrub-to-extractant flow is set at a predetermined value and can be changed manually. The ratio of feed to extractant flow is adjusted automatically according to the specific gravity measured in the aqueous phase between Stages 12 and 13. The adjustment keeps the uranium concentration at a specified value. Specific gravity is measured by the differential pressure between two air-purged dip tubes at different depths in a small bypass pot.

An automatic shutdown system stops all feeds and the mixer-settler motors whenever any of the following abnormal conditions occur:

- Low flow of any incoming stream
- High specific gravity in Stage 12
- Low pressure air supply to the weirs
- High differential motor current

Bypass circuits are provided for startup, flushing, and maintenance.

Hydraulic and Mass Transfer Characteristics

Testing the hydraulic capacity of the centrifugal mixer-settlers with radioactive solutions was not feasible because of the inconvenience of any errors. Therefore, the hydraulic and mass transfer characteristics of the numerical 18-stage bank were evaluated at the Semiworks with nonradioactive solutions that simulated those used in the Purex extraction-scrub service. The first test reproduced the low uranium conditions of the waste end of the bank. The flows ranged from rates equivalent to processing 8 tons per day of uranium to 27 tons per day (60 gpm). A maximum processing rate of 14 tons per day was obtainable in the 24-stage bank of pump-mix mixer-settler that the 18-stage centrifugal mixer-settler replaced.

Tests more truly simulating the Purex extraction-scrub conditions were made by introducing a feed of unirradiated natural uranium at Stage 10. The resulting change in relative phase densities required higher air pressures on the weirs for satisfactory performance than did the solution without uranium.

Improper air pressure allows entrainment to pass from one stage to the next, where the entrained phase is separated and returned to the first stage. The increased power required for this internally circulating flow is detected easily and consequently serves as an indicator for improper air pressure.

The mass transfer performance of the 18-stage bank was measured during the tests with unirradiated uranium at processing rates equivalent to 8 to 16 tons per day. The cocurrent stage efficiencies for extraction approached 100%; less than 0.03% of the uranium remained in the waste stream, and less than 0.3% entrainment occurred in each end stream. Each mass transfer test was followed by a Purex 1C-bank operation of the centrifugal mixer-settlers at a rate equivalent to 10 tons per day to back-extract the uranium from the organic phase; the overall mass transfer efficiency was approximately 95%.

Additional Operating Features

Important additional features that apply to the 18-stage unit were previously established with a 4-stage prototype:

1. The hydraulic capacity of centrifugal mixer-settlers is not adversely affected by aqueous-phase acid concentrations as low as 0.01M nitric acid, in contrast to the behavior of pump-mix mixer-settlers, which could not operate below 0.1M nitric acid because of the stability of the resulting emulsion.

2. The method for detecting a motor failure and the procedure for flushing a unit containing an inoperable stage are satisfactory.

3. The air-weir pressures required to control the location of the emulsion can be predicted for any system.

4. The centrifugal mixer-settler performance is not impaired by 5000 ppm of solids in feed solutions. Solids are centrifuged out of solution and accumulate as a thin layer in each stage until the rate of deposition is equal to the rate of re-suspension into the aqueous phase at the underpass baffle. Thereafter, the solids follow the aqueous phase through the bank. In contrast, many of these solids collect at the interface of pump-mix mixer-settlers until the phase separation is greatly slowed.

Plant Performance with Radioactive Solutions

The 18-stage bank of centrifugal mixer-settlers have been highly successful, operating since startup on radioactive feed in 1996. Mechanical reliability has been satisfactory with minimal remote maintenance required on defective air seals or motor bearing failures. Both of these types of failures were remedied by remotely replacing the motor-seal-bearing assembly with a spare assembly. The defective units were decontaminated, repaired, and stored for reuse. One 6-stage module was decontaminated and rewired to replace deteriorated electrical insulation, and another was remotely replaced with a spare because of a crack in the permanently mounted piping. The replaced module was decontaminated, repaired, and stored for reuse. The automatic control system has also performed well. The only change required was damping of the fluctuating signal from the specific gravity probe.

The Purex process has frequently been shut down completely over weekends, or in a few cases, for prolonged scheduled plant outages. The pump-mix mixer-settlers were prepared for shutdown by feeding a "cold" uranium solution for about four hours to purge fission products from the bank and to minimize solvent degradation during the idle period. The total shutdown time was five to six hours. In contrast, the centrifugal mixer-settlers are purged by 15 to 30 minutes of solvent and scrub flow, and shutdown is accomplished in less than an hour. Completely flushing the centrifugal bank, required to obtain product purity when the equipment is used occasionally for processes other than Purex, takes 1 to 2 hours, a marked improvement over 8 to 16 hours involved in completely flushing the pump-mix mixer-settlers. When the previous bank was started after a shutdown, a steady state operation was attained in about 16 hours. As well as it can be determined, the centrifugal bank reaches equilibrium in about 20 minutes.

Decontamination from fission products by the new 18-stage bank has been satisfactory, although somewhat less than that by the old 24-stage bank because of a deficiency of scrub stages. The short residence time and consequent lower exposure of solvent to radiation in the centrifugal mixer-settler has markedly reduced the retention of fission products in the solvent. Gross gamma activity of the solvent, both before and after washing, fell to less than one-fifth of former values for the same feed activities. This reduction permits processing more active feeds. Whereas feeds of 100 curies per liter to the pump-mix mixer-settlers caused excessive fission product retention by the solvent and drastically reduced decontamination through the first cycle, the centrifugal bank has processed feed with up to 250 curies per liter with no adverse affects. No upper limit on feed activity has been established.

The years of experience with the centrifugal mixer-settlers in radioactive service has demonstrated the expected ease of operation and reduction of solvent damage, with good mechanical reliability.

References

A number of scientists and engineers contributed to the development of the centrifugal mixer-settlers at SRL. The references of their work are listed below:

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Biography

Albert A. Kishbaugh received a Bachelor of Science degree in Chemical Engineering from Penn State University in 1952. He was employed by the Du Pont Company in 1952 and was assigned as a Research Engineer at the Knolls Atomic Power Laboratory in Schenectady, New York, while the initial construction of the Savannah River Site was begun. He transferred to the Savannah River Laboratory (SRL) at SRS from 1954 until 1971, at which time he was relocated to the Du Pont Process Section of the Atomic Energy Division in Wilmington, Delaware. While at SRL, he held the positions of research engineer, senior research engineer, and senior research supervisor. While in Wilmington, he was process superintendent in the Process Section from 1972 to 1979 and engineering manager in the Departmental Engineer's Office from 1980 to 1989, at which time he retired from Du Pont. As engineering manager, he was responsible for coordinating engineering activities between the Du Pont Company Management, SRL, SRS, and the Du Pont Engineering Department. He was employed by Bechtel Savannah River, Inc. (BSRI) from 1989 until 1992, where he held the position of design engineering resident manager at United Engineers and Constructors, Philadelphia, Pennsylvania (1989-1991) and research manager technical representative at Ebasco, Augusta, Georgia (1991-1992).