Conf-910852--2

CONCENTRATION OF REMOTE-HANDLED, TRANSURANIC, SODIUM NITRATE-BASED SLUDGE USING AGITATED THIN-FILM EVAPORATORS

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DE91 014278

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For presentation at the
1991 AIChE Summer National Meeting, Pittsburgh, PA
August 18-21, 1991

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*Managed by Martin Marietta Energy Systems, Inc. for the U.S. Department of Energy under contract DE-AC05-84OR21400.

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CONCENTRATION OF REMOTE-HANDLED, TRANSURANIC, SODIUM NITRATE-BASED SLUDGE USING AGITATED THIN-FILM EVAPORATORS

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ABSTRACT

The Waste Handling and Packaging Plant (WHPP) is being designed at Oak Ridge National Laboratory (ORNL) to prepare transuranic waste for final disposal. Once operational, this facility will process, package, and certify remote-handled transuranic waste for ultimate shipment and disposal at the Waste Isolation Pilot Plant (WIPP) in Carlsbad, New Mexico. One of the wastes that will be handled at WHPP is the transuranic sludge currently stored at ORNL in eight 50,000-gal underground tanks. The use of an Agitated Thin-Film Evaporator (ATFE) for concentration of this waste is being investigated. Tests have shown that the ATFE can be used to produce a thick slurry, a powder, or a fused salt. A computer model developed at the Savannah River Plant (SRP) to simulate the operation of ATFEs on their waste is being modified for use on the ORNL transuranic sludge. This paper summarizes the results of the tests with the ATFEs to date, discusses the changes in the SRP model necessary to use this model with the ORNL waste, and compares the results of the model with the actual data taken from the operation of ATFEs at vendors' test facilities.

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1. BACKGROUND

Remote-handled (RH) liquid radioactive waste has been generated at ORNL by laboratory research operations as well as decommissioning and decontamination activities since the inception of laboratory operations. The waste, as generated, is primarily nitric acid contaminated with radionuclides and minor concentrations of organic liquid and hazardous metals, as defined by the Resource Conservation and Recovery Act (RCRA). This waste is collected in tanks, neutralized with sodium hydroxide, concentrated by evaporation, and stored for future processing and disposal. Upon cooling, the concentrated waste separates into sludge and supernate phases. The supernate is ~4 to 5 *M* sodium nitrate contaminated with soluble radionuclides, primarily ^{137}Cs and ^{90}Sr (see Table 1), while the sludge consists of precipitated carbonates and hydroxides, primarily calcium carbonate and magnesium hydroxide. (Current data on sludge composition are preliminary and have not been approved for publication.) Because radioactive actinides, such as transuranic (TRU) elements, and most metals are insoluble in alkaline liquid waste, these constituents also precipitate.

Table 1. Remote-Handled Mixed Waste Supernate Composition*

CHEMICAL COMPOSITION		RADIOLOGICAL COMPOSITION	
COMPONENT	**(mg/L)**	**COMPONENT**	**(Bq/L)**
Sodium	92,300	Gross α	1.4×10^5
Potassium	19,000	Gross β	6.5×10^8
Calcium	1,800	^{90}Sr	5.4×10^7
Magnesium	297	^{137}Cs	5.3×10^8
Strontium	19	^{134}Cs	5.5×10^6
Barium	2.7	^{60}Co	3.6×10^6
Aluminum	14		
Chromium	2.7		
Lead	2.9		
Nitrate	280,000		
Carbonate	3,600		
Chloride	31,00		

*The supernate has a pH of 8 to 13.

Prior to 1984, concentrated liquid waste was mixed with grout and injected into a shale formation via the hydrofracture process. The State of Tennessee Underground Injection Control Regulations required that this process be discontinued, and since disposal by hydrofracture has been discontinued, no new method for treatment and permanent disposal of the TRU, mixed liquid waste exists.

The Department of Energy (DOE) has established a policy to dispose of all TRU waste in WIPP, a deep geologic repository located in Carlsbad, New Mexico.[1] The WIPP is preparing to receive contact-handled TRU waste by implementing a 5-year test plan for the safe receipt and storage of the waste. Receipt of remote-handled TRU waste at the WIPP is also included in the DOE policy, but a firm schedule has not been established.

ORNL has generated and is storing about 90% of the current inventory of DOE RH TRU waste. A WHPP is proposed as a line-item project, and it will process and solidify this RH TRU waste as well as repackage solid RH TRU waste for shipment to the WIPP. The preliminary conceptual design report (PCDR)[2] for the WHPP was completed in May 1989, and a draft conceptual design report (CDR)[3] was completed in October 1991. The design basis of the plant includes remote operation and maintenance.[4] The project is currently being held pending resolution of waste form criteria and funding issues.

To dispose of solid RH TRU waste at the WIPP, the processed waste must meet the waste acceptance criteria, which include: <1 wt % free liquid, limited gas generation, limited particulate matter <10 μm, and radionuclide concentration >100 nCi/g TRU and <500 total Ci/drum.[5] No leach requirements have been established because the waste containment philosophy is based on the prediction that primary confinement is provided by the geologic repository. However, regulatory aspects of the WIPP must be negotiated before the waste acceptance criteria can be finalized.

1.1 SLURRY SOLIDIFICATION PROCESS DESCRIPTION

The slurry processing steps included in the WHPP are as follows: (1) sludge mobilization, (2) evaporation, and (3) solidification. First, the sodium nitrate supernate and insoluble solid sludge will be removed from storage tanks and transferred to the WHPP. In order to package and certify the Melton Valley Storage Tank (MVST) sludges for shipment to the WIPP, the sludges must be concentrated and solidified. Two technologies are being considered for concentration and solidification of the sludge from the MVSTs: (1) conventional evaporation using an ATFE and (2) microwave technology. Vendor tests with ATFEs, using synthetic waste to simulate the transuranic sludges, have demonstrated that in a single pass the ATFE can be used effectively to produce a waste form that is a thick slurry or paste, a powder, or a melt that forms a fused salt upon cooling.[6] Preliminary development efforts have indicated that microwave technology can be used to melt the waste; however, the ATFE must first be used to remove the majority of water from the waste before microwave treatment. Because vendor tests indicate that the ATFE can also melt the waste, further development may show that a microwave is not required. Development efforts will determine if WHPP will contain both an evaporator and a microwave system or if the process will rely on one technology or the other.[2,7] A pilot-scale ATFE has been procured and will be evaluated at ORNL's WHPP Development Facility (WDF) for the evaporation of simulated waste to varying concentrations. The evaluation of the ATFE will be used to assess the unit for continuous long-term processing of the MVST sludges and to determine the optimum operating conditions for the ATFE. A computer model will be used to establish process parameters important in determining the optimum operating conditions.

1.2 THE SAVANNAH RIVER PLANT COMPUTER MODEL

A computer model was developed at the SRP to simulate the operation of ATFEs (of any size or configuration) for the concentration of SRP waste.[8] Because the SRP waste is similar in composition to the MVST sludges, this model is being evaluated for modeling the ATFE in a pilot-scale operation. If the SRP code cannot be used to adequately model the pilot ATFE, modification of the SRP code or development of a new model may be required to simulate the operation of the evaporator on the MVST sludges. This report discusses the use of a pilot-scale ATFE, gives an overview of the SRP computer model, describes the modifications made to the SRP model for the MVST waste, and compares this "preliminary" modified model with data taken from vendor tests using an ATFE on simulated MVST sludge.

1.3 AGITATED THIN-FILM EVAPORATORS

A schematic diagram of an ATFE is presented in Fig. 1. These evaporators use a rotor moving at a high speed to mechanically agitate and spread a turbulent thin layer of solution over the entire heated surface. The unique feature of an ATFE is not the thin film itself but rather the mechanical agitator device for producing and agitating the film. The agitation at the heat-transfer surface promotes heat transfer and maintains precipitated or crystallized solids in a manageable suspension without fouling the heat-transfer surface. This capability makes ATFEs particularly suited for volume reduction of radioactive wastes that contain suspended solids.

There are two general types of ATFEs — vertical and horizontal. In the vertical unit, feed is transported through the heated zone by gravity and falls in a helical pattern because of the action of the rotor. In the horizontal unit, the holdup of process fluid is independent of gravity and controlled by rotor design

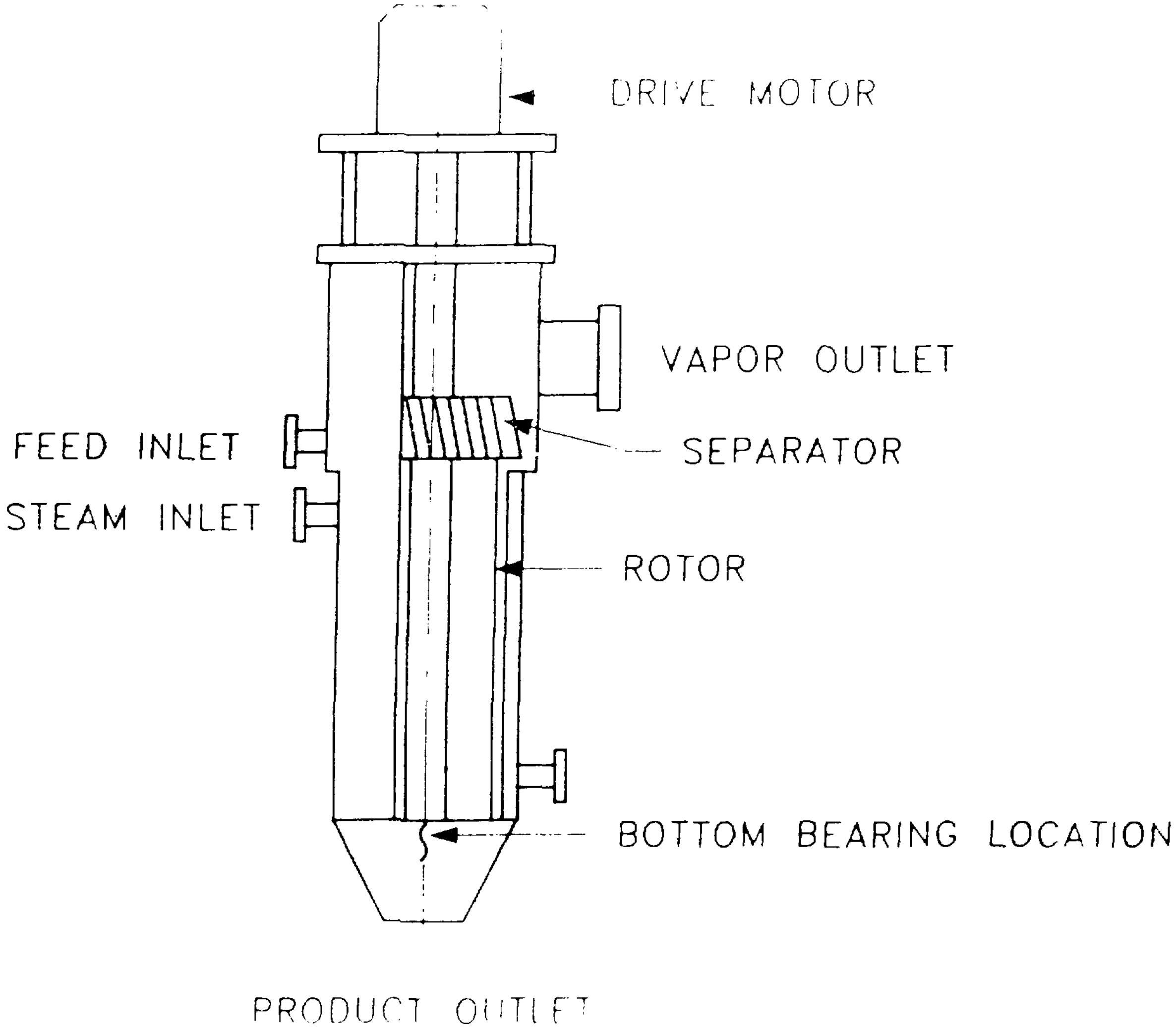

Fig. 1. Typical vertical agitated thin-film evaporator.

The SRP computer model was developed to simulate the steady-state operation of ATFEs on three distinct types of SRP wastes — Purex waste, decladding waste, and HM waste.^ Purex waste is an aqueous waste from the processing of dissolved uranium metal through the Purex solvent extraction process. It is high in $NaNO_3$, with lower concentrations of Na_2CO_3, Na_2SO_4, NaOH, and $NaAlO_2$. Decladding waste is an aqueous waste from NaOH dissolution of aluminum cladding on fuel elements. It is high in NaOH and $NaAlO_2$ with moderate concentrations of $NaNO_3$ and $NaNO_2$. HM waste is an aqueous waste from the processing of codissolved aluminum and uranium through solvent extraction, similar to Purex waste but higher in NaOH and $NaAlO_2$. Physical properties of these wastes as a function of temperature and/or concentration were included in the model. The physical properties include the specific gravity, heat capacity, thermal conductivity, viscosity, heat of vaporization, heat of crystallization, and the solubility product. The use of different wastes would require that additional physical properties be incorporated in the model.

The model calculates the following as a function of the distance from the inlet of the evaporator: (1) product temperature; (2) steam duty; (3) mass flow rate of vapor, solution, and solids; (4) fractions of salts in the product; (5) volume percent of solids in the product; (6) boil-down ratio; (7) weight percent of solids plus solute; and (8) inside and overall heat-transfer coefficients. The heat-transfer coefficients are based on the use of steam as the heating medium in the outside jacket. As sensible heating, vaporization of water, and crystallization of solids in the process solution are taken into account, the model has the capability of evaluating the production of a slurry or a powder but presently does not include the parameters necessary to include heat of fusion for melting. Variables that can be studied with the model include the following:

(1) evaporator orientation (vertical or horizontal),
(2) feed rate,
(3) feed temperature,
(4) operating temperature (jacket steam pressure),
(5) inside diameter of ATFE,
(6) wall thickness,
(7) heat-transfer area,
(8) thermal conductivity of wall,
(9) rotor speed,
(10) rotor blade clearance, and
(11) feed composition.

1.4 MODIFICATIONS OF THE MODEL FOR USE ON ORNL WASTES

The SRP computer model was written in the FORTRAN IV language. For use at ORNL, the computer model has been modified to use FORTRAN-77, which runs on IBM and IBM-compatible personal computers.

The primary chemical component of the MVST waste is sodium nitrate. The MVST waste also contains a significant quantity of insoluble sludge in which calcium carbonate is the main constituent. The primary chemical components of the SRP Purex waste include: Na_4SO_4, Na_2CO_3, $NaNO_3$, $NaNO_2$, NaOH, and $NaAlO_2$. The SRP computer model contains physical and thermodynamic properties for these constituents. In order to simulate the MVST waste, data for sodium carbonate were replaced by data for calcium carbonate. The replacement data included heat capacity, number of cations, heat of dissolution, molecular weight, density of the solid component, and the solubility product. Evaluation of the ATFE with various concentrations of calcium carbonate will be performed in future tests.

1.5 PRELIMINARY EVALUATION OF MODEL

After the program was converted to operate with FORTRAN-77, the input data provided in the SRP report were run and the results compared with those in the SRP report. The data from the output were identical to the output data listed in the SRP report, indicating that no errors had been made in converting the program from FORTRAN IV to FORTRAN-77.

After the modifications were made, the modified program was run and the output compared with the operating data obtained from the vendor tests at Cherry-Burrell AMCA International. The ATFE used in the vendor tests was a vertical unit with an inside diameter of 10 cm (4 in.) and a heat-transfer area of 0.16 m^2 (1.7 ft^2). The input parameters used in the program are listed in Table 2.

A comparison of the process parameters measured in the vendor tests using the vertical ATFE at Cherry-Burrell with those predicted by the model is presented in Table 3. The initial boiling point predicted by the model is within ~4% of that measured for the 3.5 *M* sodium nitrate concentration used in the feed for these tests. The percentage difference between the measured overall heat-transfer coefficient and that predicted by the model varied from 3.9 to 20.8%. Because the physical properties of the waste are used to determine the heat-transfer coefficients, the differences in the predicted and measured heat-transfer coefficients may result from errors in the calculation of the physical properties of the waste feed.

The model was not able to consistently predict the split between the bottom and overhead product. The percentage difference between the measured bottoms mass flow rate and that predicted by the model varied from -1.8 to -40.3%. The percentage difference between measured overhead mass flow rate, and that predicted by the model varied from 8.1 to 35.9%. The model was also not able to consistently predict the total solids concentration of the bottom product. The

Table 2. Input parameters from the vendor tests using a Cherry-Burrell vertical agitated thin-film evaporator

INPUT	PARAMETER DESCRIPTION
1	Vertical
1	Purex
0.17-0.26	Feed rate (gpm)
22-26	Feed temperature (°C)
150.0	Product temperature (°C)
1.0	Temperature increment (°C)
120.0	Steam pressure (psig)
4.0	Inside diameter (in.)
0.0	Cladding thickness (in.)
0.1	Wall thickness (in.)
1.7	Heat-transfer area (ft^2)
0.0	Cladding conductivity (BTU·h·ft·°F)
9.4	Wall conductivity (BTU·h·ft·°F)
1796.0	Rotor speed (rpm)
25.0	Rotor-wall clearance (mils)
0.0	Concentration Na_2SO_4 (g-mol/L)
0.0	Concentration $CaCO_3$ (g-mol/L)
3.5	Concentration $NaNO_3$ (g-mol/L)
0.0	Concentration NaOH (g-mol/L)
0.0	Concentration $NaA1O_2$ (g-mol/L)

Table 3. Comparison of process parameters from the vendor tests at Cherry-Burrell with the modified SRP model

RUN	PARAMETER	MEASURED	MODEL	% DIFFERENCE
1	Initial boiling point (°C)	102	106	3.9
	Mass flow rate feed (lb/h)	157.5	164.1	4.2
	Mass flow rate bottoms (lb/h)	75	69.8	-6.9
	Mass flow rate overhead (lb/h)	82.5	94.3	14.3
	Total solids in product (%)	63	56	-11.1
	Overall heat-transfer coefficient (BTU·h·ft^2·°F)	503	483	-3.9
2	Initial boiling point (°C)	102	106	3.9
	Mass flow rate feed (lb/h)	136.9	142.8	4.3
	Mass flow rate bottoms (lb/h)	50.6	49.7	-1.8
	Mass flow rate overhead (lb/h)	86.25	93.2	8.1
	Total solids in product (%)	83	67	-19.3
	Overall heat-transfer coefficient (BTU·h·ft^2·°F)	497	455	-3.5
3	Initial boiling point (°C)	102	106	3.9
	Mass flow rate feed (lb/h)	120	125.1	4.3
	Mass flow rate bottoms (lb/h)	46.5	35.6	-23.4
	Mass flow rate overhead (lb/h)	73.5	89.5	21.8
	Total solids in product (%)	88	82.5	-6.3
	Overall heat-transfer coefficient (BTU·h·ft^2·°F)	489	415	-15.1
4	Initial boiling point (°C)	102	106	3.9
	Mass flow rate feed (lb/h)	101.3	105.5	4.1
	Mass flow rate bottoms (lb/h)	42.2	25.2	-40.3
	Mass flow rate overhead (lb/h)	59.1	80.3	35.9
	Total solids in product (%)	97	98	1.0
	Overall heat-transfer coefficient (BTU·h·ft^2·°F)	385	465	20.8

percentage difference between the measured total solids concentration and that predicted by the model varied from 1 to -19.3%. The differences among the measured and predicted values of the bottom flow rates, the overhead flow rates, and the bottom total solids concentrations may be due to the overall heat-transfer coefficient and physical properties calculated by the model.

It should be noted that the evaporator used in the vendor tests at Cherry-Burrell AMCA International contained only 0.16 m^2 (1.7 ft^2) of heat-transfer area. This is small for scale-up purposes and may have contributed to larger errors in the model calculations. Tests at the WDF will use a larger evaporator. Future tests will also be conducted to compare the physical properties predicted by the model with the actual physical properties of the simulated MVST sludges. The model will be modified to ensure that the predicted physical properties and heat-transfer coefficients match those in the actual operating system.

1.6 FUTURE TESTING AND MODIFICATIONS FOR THE MODEL

Several modifications to the model must be made if it is to be used to simulate operation of ATFEs on MVST sludges. Because of differences in the chemical composition of the SRP wastes and the MVST sludges, the program must be modified to accept different feed components and calculate the correct physical properties and heat-transfer coefficients as a function of concentration and temperature within the evaporator. Extensive tests with the simulated wastes will be necessary to accomplish this task. Because future plans include using hot oil to melt the sludges, the program must be modified to calculate heat-transfer coefficients for hot oil, as well as for steam. The heat of fusion must also be incorporated in the model to account for the heat required to melt the waste

2. SUMMARY

Vendor tests, using synthetic waste to simulate TRU sludge stored in underground tanks at ORNL, have demonstrated that in a single pass an ATFE can be used to produce a waste form that is a thick slurry or paste, a powder, or a melt that forms a fused salt upon cooling.

A computer model developed at the SRP to simulate the steady-state operation of ATFEs on SRP waste has been modified for use on ORNL waste. This modified model was compared with experimental data taken during vendor testing of an ATFE. The computer model was not able to consistently produce an accurate prediction of the split between the mass flows of the bottom and overhead products, the total solids concentration in the bottoms, or the overall heat-transfer coefficients. Further testing and modifications to the model during testing of the ATFE at the WHPP Development Facility will be necessary if the model is to be used. Also, additional sections for the computer model must be written to account for: (1) the acceptance of different feed components, (2) the heat of fusion in the melting of the waste, and (3) the use of either steam or hot oil as a heating medium.

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