

A hybrid liquid-phase precipitation (LPP) process in conjunction with membrane distillation (MD) for the treatment of the INEEL sodium-bearing liquid waste

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Received 29 November 2004; received in revised form 24 January 2005; accepted 30 January 2005

Available online 5 March 2005

Abstract

A novel hybrid system combining liquid-phase precipitation (LPP) and membrane distillation (MD) is integrated for the treatment of the INEEL sodium-bearing liquid waste. The integrated system provides a “full separation” approach that consists of three main processing stages. The first stage is focused on the separation and recovery of nitric acid from the bulk of the waste stream using vacuum membrane distillation (VMD). In the second stage, polyvalent cations (mainly TRU elements and their fission products except cesium along with aluminum and other toxic metals) are separated from the bulk of monovalent anions and cations (dominantly sodium nitrate) by a front-end LPP. In the third stage, MD is used first to concentrate sodium nitrate to near saturation followed by a rear-end LPP to precipitate and separate sodium nitrate along with the remaining minor species from the bulk of the aqueous phase. The LPP–MD hybrid system uses a small amount of an additive and energy to carry out the treatment, addresses multiple critical species, extracts an economic value from some of waste species, generates minimal waste with suitable disposal paths, and offers rapid deployment. As such, the LPP–MD could be a valuable tool for multiple needs across the DOE complex where no effective or economic alternatives are available.

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Keywords: Precipitation; Membrane distillation; INEEL sodium-bearing waste; Toxic metals; TRU elements and fission products

1. Introduction

The Idaho Nuclear Technology and Engineering facility (INTEC) located at the Idaho National Engineering and Environmental Laboratory (INEEL) site was established in the early 1950s to store and reprocess spent nuclear fuel for the recovery of uranium-235. Most reprocessing was performed on aluminum- or zirconium-clad uranium fuels [1]. Smaller quantities of stainless steel and graphite clad uranium fuels were also reprocessed. The INEEL tanks waste contains: (1) predominately nitric acid, nitrate, and sodium; (2) significant amounts of aluminum (or zirconium), and potassium; (3) appreciable amounts of sulfates, phosphates, chlorides,

and toxic metals (chromium, mercury, iron, lead, nickel, and manganese); and (4) small amounts of Transuranic (TRU) elements (plutonium, neptunium, americium, and curium) and their fission products (cesium, strontium and barium) [1].

The INEEL tanks waste is uniquely different from most tanks waste at other DOE sites (e.g., Hanford, Savannah River, and Oak Ridge) [1]. The INEEL waste is extremely acidic, while most tanks waste at other sites is very basic. The basicity of tanks waste in other DOE sites has caused many inorganic species to segregate into complex mixtures of liquids, slurries, and sludges. In contrast, inorganic species remain dissolved in the INEEL acidic tanks waste (the liquid is clear almost to the bottom of the tanks) [1].

The INEEL liquid waste has been divided into high-activity waste (HAW), and sodium-bearing liquid waste. All of the HAW resulting from the dissolution and processing of spent nuclear fuel has been calcined and stored in stainless

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Nomenclature

%AAD	percent average absolute deviation
C_F	MD bulk feed concentration
C_i	model's interaction parameters
C_m	MD feed concentration at the membrane surface
$C_{1,m}$	concentration of salt species in filtered sample
$C_{1,2}$	concentration of salt species in standard sample
J	permeate flux through MD or VMD membrane
M_i	molar concentration of individual ions
NP	number of points
p^s	vapor pressure of the aqueous solution
p^0	vapor pressure of pure water
P	precipitation fraction
R	ideal gas constant
RMSE	root mean square error
SS	objective function
T	temperature
T_F	MD feed temperature
T_{Fm}	MD bulk feed temperature at the membrane surface
T_P	MD bulk permeate temperature
T_{Pm}	MD permeate temperature at the membrane surface
v_i	molar volume of solvent i
v_w	water molar volume
V_r	solvents volume ratio (organic/water)
V_i	volume of solvent i
$x_{i,j}$	mole fraction (solubility) of species i in solvent j
$x_{i,m}$	mole fraction (solubility) of species i in mixed-solvents mixture m

Greek letters

θ_i	volume fraction of solvent i
Δ_1	salt binary-solvent interaction parameter (ternary constant)
$\Delta_{i,j}$	interaction parameter of solvent i with solvent j
Π	osmotic pressure

Subscripts

Cal	calculated
Exp	experimental
m	mixed-solvent mixture
w	water
1	salt species
2	water solvent
3	organic solvent

Superscripts

E	excess
s	solution
0	pure

steel bin sets enclosed in concrete vaults with walls up to four feet thick (interim storage). However, the sodium-bearing liquid waste still remains in storage tanks.

In the calcination process, a fluidized bed is operated at about 500 °C to convert liquid waste (evaporates water, nitric acid, and volatile species) into a dry granular solid composed mainly of metal oxides [2]. The average weight percentages of the main constituents in the calcined waste from aluminum-clad fuels are: 91% aluminum oxide; 3% sodium oxide; 1% iron oxide; 1% boron oxide; 1% sulfate; and less than 1% fission product oxides [2]. However, the average weight percentages of the main constituents in the calcined waste from zirconium-clad fuels are: 54% calcium fluoride; 24% zirconium oxide; 14% aluminum oxide; 4% calcium oxide; 3% boron oxide; and less than 1% fission product oxides [2].

The use of the calcination process is attributed to two main factors. The first factor is that the volume of the calcine waste is about seven times less than the volume of the liquid waste. The second factor is that the calcine waste is more chemically stable than the liquid waste. Thus, it is to some extent safer to store than the liquid waste. However, significant operational problems were associated with the calcination of the INEEL waste.

First, the significant presence of alkali cations (sodium and potassium) in the feed leads to form nitrate complexes that melt at temperatures below 500 °C, and agglomerate the calcine in the fluidized bed (loss of fluidization) [2]. To solve this problem, large amounts of aluminum nitrate must be added to dilute the concentrations of sodium and potassium to levels that would not affect the operation of the fluidized bed. This would reduce the overall process efficiency, and would lead to lower the depletion rate of the tanks waste (the net liquid volume to solid volume is reduced from 7 to 2). In fact, the division of the INEEL waste into HAW and sodium-bearing liquid waste is, to a large extent, attributed to this problem. As a result, it is anticipated that the remaining INEEL sodium-bearing liquid waste is unlikely to be calcined before 2012, as required by the settlement agreement (the State of Idaho, DOE and Navy) [1,2].

Second, to apply the calcination process to the remaining sodium-bearing liquid waste, it was suggested that the fluidized bed ought to be operated at higher temperatures to decompose some of the alkali cations without the addition of excessive amounts of aluminum nitrate [2]. A laboratory-scale study indicated that a feed composition with a 1.5–1.0 molar ratio of aluminum to alkali cations can be calcined at 600 °C [2]. This represents a 60% reduction in the amount of aluminum (3.1–1.0 molar ratio of aluminum to alkali cations) that must be added to the calciner feed at 500 °C (baseline operation). However, the application of the Maximum Achievable Control Technology (MACT) rule requires significant off-gas treatment and monitoring for carbon monoxide, NO_x, and products of incomplete combustion. In addition, the operation of the calcination process at higher temperatures is more energy intensive.

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