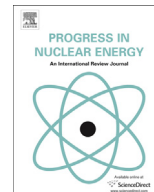




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Modeling and simulation of diluent recovery unit in PUREX solvent regeneration system

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ABSTRACT

The presented work details the modeling and simulation of distillation column for the separation of tributyl phosphate (TBP) and normal paraffin hydrocarbon (NPH) which is an integral unit in the PUREX solvent regeneration system. The vapor liquid equilibrium data of TBP + NPH system at 5.5 torr and 7.5 torr were experimentally generated using an ebulliometer and the data were subjected to thermodynamic consistency test using Herington's method. The experimental VLE data were correlated using Wilson and non random two-liquid (NRTL) models and the binary interaction parameters regressed from the data are reported. The distillation column simulation was carried out using an algorithm involving separate solver/corrector loops for the MESH variables and the NRTL model was used for predicting the vapor liquid equilibrium of TBP-NPH system. Validity of the developed code was tested by comparing the simulation outputs with the experimental results from PUREX solvent regeneration plant.

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1. Introduction

The PUREX process (Lanham and Runion, 1949) employs Tributyl phosphate (TBP) diluted with hydrocarbon as solvent is widely used for the recovery and recycle of uranium and plutonium from spent nuclear fuels. Even after six decades since its inception, due to competency of the PUREX process over various other reprocessing techniques, still it remains as the workhorse in many existing reprocessing plants. It is widely recognized that tributyl phosphate and hydrocarbon diluents are degraded due to hydrolytic and radiolytic reactions, forming activity-binding degradation products that can cause product losses, poor separation efficiencies and emulsions which could interfere with process operations (Lanham and Reilly, 1951; Goode, 1956; Mailen and Tallent, 1984). The primary degradation products (i.e. dibutyl phosphate, monobutyl phosphate, phosphoric acid and butanol) are produced from the hydrolytic and dealkylation reactions of the TBP when contacted with nitric acid. The secondary degradation products (nitroparaffins, aldehydes, ketones and carboxylic acids), on the other hand are originated from the radiolytic reactions of diluents when exposed to intensive radiation (Blake, 1968; Nowak et al.,

1972). The presence of degradation products even at concentrations of 10^{-5} to 10^{-3} M in the solvent influences the extraction performances. The most significant single parameter which affects solvent extraction process operability and product quality is the quality of the solvent. Hydraulic stability of the solvent extraction contactors and decontamination of the heavy metal from the fission products is directly traceable to the solvent quality. Hence the solvent must be purified before it is recycled back to process.

In the PUREX process, generally, the spent solvent is continuously regenerated by scrubbing with sodium carbonate/hydroxide after each pass through the process and most of the radioactivity belonging to primary degradation products is removed. Residual activity due to secondary degradation products are essentially surface tension-active agents and complexing agents of plutonium, zirconium and ruthenium, however, cannot be removed by washing these solutions as the degraded long-chain soluble organic compounds tend to remain in the solvent even after scrubbing with carbonate washing solutions and complex fission products again when the solvent is recycled. These degradation products progressively accumulate in the solvent with repeated recycling and their removal from the solvent is not well addressed as that of the primary degradation products of TBP. With time, the solvent performance characteristics become so impaired that its partial refreshment is required at intervals. In fact, the amount of organics generated in the extraction process can be reduced considerably by

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adopting a suitable solvent regeneration process.

A common method for purifying organics is distillation. The alternate method reported in literature has shown that the secondary degradation products can also be removed by passing the spent solvent through a solid adsorbent bed packed with activated alumina or base treated silica gel (Neace, 1983; Talent et al., 1984; Olson and McCray, 1986; Mailen, 1987). This bed was also shown to remove secondary degradation products of solvent. Engineering problems could be foreseen with this process since it has not been used in an operating plant. This includes the potential generation of additional radioactive solid waste, how to extend bed life and what bed regeneration techniques might be available to make the process more operable in a radiochemical plant.

Considerable effort has been devoted to the development of distillation method for PUREX-solvent purification (Sicilio et al., 1961; Clark and Nichols, 1965; Ginisty and Guillaume, 1990; Drain et al., 1996; Zilberman et al., 2002; Pandey et al., 2013), but its industrial application is limited (Drain et al., 1996). There is lack of information in the literature on the modeling and simulation of distillation column to address this issue. Process modeling is a technique used in chemical engineering process design extensively. A simulation is the representation of the real world process or system over a period of time. Further, simulation involves the generation of artificial history of the system and the observation of artificial history to draw inferences concerning of the operating characteristic of the real system. Thus, modeling and simulation can be used both as an analysis tool for predicting the effect of changes to existing system and as a design tool to predict the performance of new system under the varying set of circumstances. In addition it also minimizes the requirement of the number of experimental runs. The present work details the modeling and simulation of the distillation column for the recovery of diluent and solvent from the spent solvent.

2. Mathematical model of the distillation column

Mathematical model of distillation column for the separation of TBP-NPH mixture is based on material balance, equilibrium vapor-liquid relationship, composition constraints and energy balance. The model equations which completely describe a theoretical stage are generally known as MESH equations and they are shown in Fig. 1. The distillation unit consists of N_S stages, where the 1st stage is a total condenser and N_S th stage is a partial reboiler. All the stages except condenser stage are assumed to be theoretical stages, i.e. the vapor and liquid streams leaving the stage are brought to thermodynamic phase equilibrium. Due to low pressure drop offered by the structured packing used in the column, the pressure along the length of the unit is assumed to be constant. Column is assumed to be adiabatic as it is well insulated and heat loss is negligible.

The model equations formulated for stages 1 to N_S are as follows:

2.1. Material and enthalpy balance equations (M&H)

For condenser ($k = 1$).

Component mass balance:

$$V_{k+1}y_{i,k+1} - L_kx_{i,k} - Dx_{i,k} = 0 \quad (1)$$

where, $i = 1, \dots, N_c$ and $k = 1, \dots, N_s$.

Enthalpy balance:

$$V_{k+1}H_{V,k+1} - L_kH_{L,k} - DH_D - Q_C = 0 \quad (2)$$

For any stage in rectification and stripping section ($k = 2, \dots, N_f -$

1 for rectification and $k = N_f + 1, \dots, N_s - 1$ for stripping section).

Component mass balance:

$$V_{k+1}y_{i,k+1} + L_{k-1}x_{i,k-1} - V_ky_{i,k} - L_kx_{i,k} = 0 \quad (3)$$

Enthalpy balance:

$$V_{k+1}H_{V,k+1} + L_{k-1}H_{L,k-1} - V_kH_{V,k} - L_kH_{L,k} = 0 \quad (4)$$

For feed stage ($k = N_f$).

Component mass balance:

$$Fz_i + V_{k+1}y_{i,k+1} + L_{k-1}x_{i,k-1} - V_ky_{i,k} - L_kx_{i,k} = 0 \quad (5)$$

Enthalpy balance:

$$FH_F + V_{k+1}H_{V,k+1} + L_{k-1}H_{L,k-1} - V_kH_{V,k} - L_kH_{L,k} = 0 \quad (6)$$

For reboiler ($k = N_s$).

Component mass balance:

$$L_{k-1}x_{i,k-1} - V_ky_{i,k} - L_kx_{i,k} = 0 \quad (7)$$

Enthalpy balance:

$$L_{k-1}H_{L,k-1} + Q_B - V_kH_{V,k} - L_kH_{L,k} = 0 \quad (8)$$

2.2. Equilibrium and summation equations (E&S)

Vapor-liquid equilibrium relationship:

$$y_{i,k} - K_{i,k}x_{i,k} = 0 \quad (9)$$

Composition constraints:

$$\sum_{i=1}^{N_c} x_{i,k} - 1 = 0 \quad \text{for } k = 1, \dots, N_s \quad (10)$$

$$\sum_{i=1}^{N_c} y_{i,k} - 1 = 0 \quad \text{for } k = 2, \dots, N_s \quad (11)$$

3. Vapor-liquid equilibrium (VLE) of TBP-NPH system

3.1. Vapor liquid equilibrium measurements

A necessary prerequisite for any realistic simulation of distillation column in the separation of TBP-NPH system is the VLE data at low pressures. Due to scarcity of reliable VLE data of the system in literature, it was experimentally generated using an ebulliometer (M/s. Abhishek scientifics Mumbai). Ebulliometer used for vapor liquid equilibrium experiments essentially consists of vaporizer, Cottrel pump, equilibrium chamber and an overhead condenser as shown in Fig. 2.

The apparatus allows recirculation of both liquid and vapor phases, and it is provided with sampling ports for both vapor condensate and residual liquid. Liquid mixture in the vaporizer is vaporized at a slow and controlled rate using a direct electrical heating system with manual controller. The vapor/liquid mixture rises through the Cottrel pump and flashes in the equilibrium chamber. The residual liquid gets collected at the liquid port, and the vapor fraction is condensed and collected at the vapor condensate port. A portion of residual liquid and vapor condensate is continuously circulated back to the vaporizer, and sampling is

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