Modeling and simulation of ^{13}C isotope separation with cryogenic distillation

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Abstract

In this paper, the column for cryogenic (^{13}C) isotope separation is modeled and simulated. The complete dynamics of the column are represented by nonlinear partial differential equations, which depend on the time as well as the spatial coordinates. The proposed solution is to identify a fractional-order model from frequency response or from time response of the system. The model is validated by the Delf model. The simulation results confirmed the reference results. Results show that the ^{13}C concentration after 60h reached 2.7% and depleted ^{13}C contains 0.5%.

Keywords: Isotope separation column, Mathematical modeling, Carbon-13, Cryogenic distillation

Introduction

A great number of chemical elements are mixtures of various isotopes. For example, (^{12}C) with mass number 12 is the basic carbon element, with a concentration of 98.9% at, while (^{13}C) is the isotope with mass number 13, with the natural abundance of 1.1%at. If some chemical compounds with a higher abundance of (^{13}C) are available, detection of compounds with a higher concentration of (^{13}C) allows valuable qualitative and quantitative measurements, very important in scientific research and industrial applications [1].

The isotope separation techniques are based on the isotope effects of different isotopic compounds that arise from the differences in the nuclear properties of the isotopes. Distillation columns are well-known apparatuses/equipment to separate raw materials and products in different applications. The separation of the mixtures is done in this column by a counter-current contact between an upward gas and a downward liquid stream. Distillation columns can be categorized into two groups of packed bed and tray based on the angled amongst liquid and gas. It is worth mentioning that in pack type columns commonly two kinds of packing including the structure and random packings are applied. For large-scale separation towers, the best selection is structured packing [2, 3].

Numerical investigation of the fluid flow in the distillation towers is extensively considered recent. A general and insightful review of both experimental and theoretical work on the isotope effects with emphasis on the vapor pressure isotope effect is provided by Jancso and Van Hook in [4]. The general theory of multistage isotope separation processes was developed by Cohen [5], treating issues like hold-up, enrichment, and equilibrium time for both ideal and real cascades. In [6] Li et al. treat the possibility of using advanced structured packing instead of common random packing used in ^{13}C separation from both productivity and reduced consumption points of view while Dulf et al. [7] present a monitoring and control system of a ^{13}C enrichment plant. Mass transfer in fluid systems and the separation processes have been studied extensively, e.g. Cussler and King [8] are excellent references, while in the field of dynamics, operation, and control of distillation columns Skogestad et al. give a comprehensive and insightful exposition in [9]. Henrietta Dulf et al. presented a general model for mod Henrietta Dulfelling and simulation of carbon isotopes separation column.

The objective of this paper is to provide a modeling approach followed by the simulation of a ^{13}C isotope separation plant that makes use of the distillation of carbon monoxide.

Mathematical modeling and Simulation

Figure 1 shows the cryogenic distillation column of carbon-13 separation. The main flow rate that shown in the figure as below:

The feed flow rate (F);
Concentration (Nf) (mole fraction);
The waste flow rate (W);
Concentration (Nw) (mole fraction);
The product flow rate (P);
Concentration (NP) (mole fraction).

The mass-flow equilibrium is given by the equation [10]:

$$F=P+W \text{ [moles/sec]} \quad (1)$$

The (^{13}C) isotope balance gives:

$$F.Nf=P.Np+W.Nw \quad (2)$$

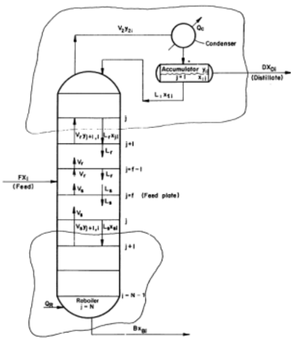

Figure 1: schematic of the separation column

For $P=0$, the column operates at "infinite reflux" (or "total reflux"). To separate the (^{13}C) isotope with the mole-fraction of (NP), at the flow rate P (mole/sec), the required feed flow rate is given by the equation:

$$F = \frac{N_P - N_w}{N_f - N_w}.P = \frac{N_P - N_w}{N_0 - N_w}.P \quad (3)$$

resulting in a "waste" flow rate:

$$W = \frac{N_P - N_f}{N_f - N_w}.P = \frac{N_P - N_0}{N_0 - N_w}.P \quad (4)$$

Where N0 is the natural ^{13}C abundance. Dedicated equipment is necessary to ensure the important operation condition $W=F$ (for $P=0$). In order to define the main variables, it is useful to divide the operating variable into two categories: qualitative variables and quantitative variables. Because the feeding carbon dioxide flow rate has high purity, the main qualitative variable is the (^{13}C) isotope concentration (mole fraction). We can take out the product from the column in the different heights of the column that could affect the final product (P, Np). The quantitative variables refer to the hydrodynamic equilibrium and the thermal equilibrium. Both the quantitative and qualitative variables maybe divided into input and output variables. If (T, k) is the transfer speed and constant of the isotope exchange, the general equations of COHEN (Cohen, 1951) are, for a level (coordinate, z) in the column:

$$H\frac{\partial N}{\partial t} + L_l\frac{\partial N}{\partial z} = +T \quad (5)$$

$$h\frac{\partial n}{\partial t} - L_g\frac{\partial n}{\partial z} = -T \quad (6)$$

where (N) is the mole fraction of (^{13}C), (n) is the mole fraction of (^{12}C), (H, h) are the carbon atoms accumulated (hold-up) in a specific volume in the column, (Ll, Lg) are the acid (liquid) and oxides (gaseous) flow in counter-current expressed in atoms of carbon and (T) is given by the equation:

$$T=-k[N(1-n)-\alpha(1-N)] \quad (7)$$

Using Matlab, the author solved the nonlinear differential equations 5 and 6 for $\frac{\partial N}{\partial z}=0$ and $\frac{\partial n}{\partial z}=0$, with initial conditions N(t=0)=n(t=0)=1.1 given by the natural abundance of ^{13}C

$$H\frac{\partial N}{\partial t} = -k[(N-\alpha n) + n.N(\alpha - 1)] \quad (8)$$

$$h\frac{\partial n}{\partial t} = +k[(N-\alpha n) + n.N(\alpha - 1)] \quad (9)$$

Results and discussion

Fig. 2 and 3 show the ^{13}CO mole fraction distribution in the column with respect to both time and height. The isotope distribution was obtained by simulating the full nonlinear model for 100 discretization divisions applied to the space domain (i.e. z= 200m)and a time step (t) equal to 0.1 s. Results are validate with refrence values [10]. As can be seen from the figure 2 the ^{13}C concentration after 60h reached 2.7% and depleted ^{13}C contains 0.27%. after 60h, the concentrations were constant and shows to 120h in this figure. Figure 3 shows the concentration of 13C with the height of the column is increased. The higher the column height leads to the greater the changes in carbon-13 concentration.

Conclusions

The objective of this paper is to provide a modeling approach followed by the simulation of a ^{13}C isotope separation plant that makes use of the distillation of carbon monoxide. Results show that the model could be predict the concentration of 13C in cryogenic distillation.

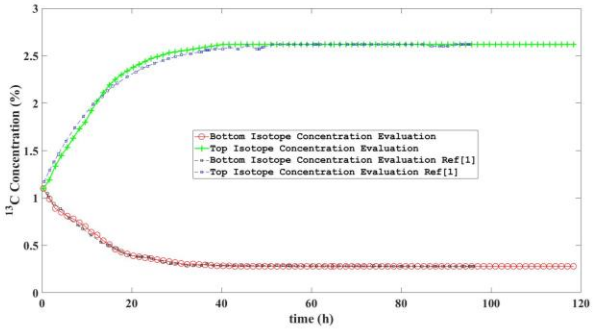

Figure 2. ^{13}C Concentration vs time

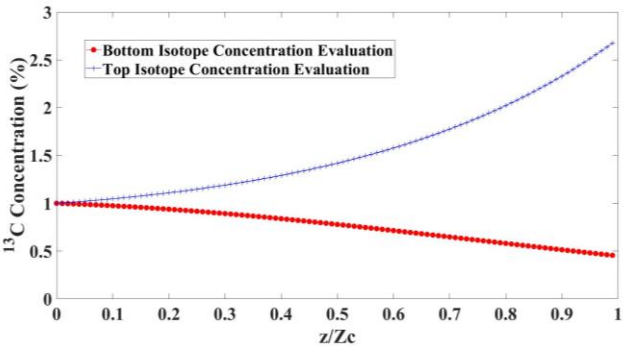

Figure 3. ^{13}C Concentration vs heigh of column

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