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A method for modeling a catalytic distillation process based on seepage catalytic packing internal

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HIGHLIGHTS

- A novel seepage catalytic packing internal (SCPI) was researched in this study.
- A rigorous mathematical model was developed to predict composition profiles.
- Graphical User Interface design and corresponding port design were implemented.
- Some new solution methods for solving the model were proposed in the paper.

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ABSTRACT

A method for the modeling of a seepage catalytic packing internal (SCPI) in a catalytic distillation column was developed and applied to the simulation of a catalytic distillation (CD) process. Based on the structure of the internal, rigorous mathematical models consisting of differential algebraic equations were developed to predict the temperature and composition profiles in the CD column. Based on the gPROMS platform, the gPROMS language was used to compile three modules, one for the reaction and the other two modules for hydrodynamic calculations and heat and mass transport calculations. In addition, a graphical user interface (GUI) design and the corresponding port design were implemented for each unit module to clearly represent the flow-sheet of the catalytic distillation process. According to the characteristics of the model, a new approach was proposed to avoid convergence difficulties and to improve the stability of the calculations. This approach was demonstrated to be an efficient method, which can reduce the simulation time and still accurately predict the results. Mathematical models and corresponding programs of reaction, separation, and the flow-sheet of CD process were validated. The simulated results were in good agreement with the experimental results, indicating that the simulation methods established in this paper are feasible. Furthermore, the gPROMS is a reliable and suitable technique for studying the catalytic distillation process of SCPI.

1. Introduction

Reactive distillation (RD) is one of the most important industrial applications of the multifunctional reactor concept, which combines chemical reaction and distillation in a single column. Recently, it has drawn considerable attention because of its potential advantages, especially for equilibrium-limited reactions. Several comprehensive reviews on the introduction of RD processes have been published in the last decade (Doherty and Buzad, 1992; Harmsen, 2007; Sakuth et al., 2001; Taylor and Krishna, 2000; Towler and Frey, 2001). RD processes can be divided into homogeneous and heterogeneous processes (Noeres et al., 2003; Parra et al., 1994). The latter is often referred as catalytic distillation (CD), in which the reaction is catalyzed by a solid catalyst. CD processes permit an optimum configuration of the reaction and separation zones in a RD column, avoiding the expensive recovery of a liquid catalyst. The design of the internals for CD processes is more critical than for conventional non-reactive counter-current vapor–liquid processes (Sakuth et al., 2001; Taylor and Krishna, 2000). The appropriate choice of internal characteristics, such as liquid residence time distribution (RTD), liquid hold-up, separation efficiency and pressure drop, can make CD processes feasible and efficient.

A seepage catalytic packing internal (SCPI), consisting of catalyst containers with avert-overflow baffles and corrugated

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metal sheets, is a better internal for catalytic distillation columns (Gao et al., 2012). SCPI is manufactured by the National Engineering Research Center of Distillation Technology of China and is packed in catalyst reaction zone. The structure of the SCPI used in the experimental column has been described in detail by Li et al. (2008), and the gas–liquid flow characteristics in the SCPI are shown in Fig. 1. The liquid flows downward, driven by its own gravity, uniformly through the corrugated metal sheets and catalyst container, whereas the gas flows upward, driven by a pressure gradient, only through the corrugated metal sheets. This design method can avoid contact between the gas and liquid in the catalyst bed and can reduce the pressure drop of the column. By changing the width ratio ($R_{CC/CMS}$) of the catalyst containers (CC) relative to the corrugated metal sheets (CMS), the SCPI can be suitable for various reaction and separation zones in different catalytic distillation processes.

We have conducted a lot of basic research work on the new internal SCPI. Cold model experiments in the column (600 mm in diameter × 1500 mm height) have been performed by Gao et al. (2012) to evaluate the flooding behaviors and pressure drop. The results show that the SCPI has a lower pressure drop and a higher operational capability than conventional column internals for heterogeneous catalytic distillation processes. Based on the hydrodynamic experiments, different simulation strategies have been proposed by Li et al. (2012) to predict the dry pressure drop, the irrigated pressure drop, and the height of the liquid above the catalyst bed. The results show that these methods are reliable in the design and scale-up of the SCPI in catalytic distillation columns. An ideal catalytic distillation internal not only has advantages in hydrodynamic characteristics, such as low pressure drop, high flux and high liquid holdup, but also needs to have a better matching relationship between the reaction and the separation. To better understand the internal and give full play to its functional advantage, it is necessary to establish rigorous mathematical models for describing the catalytic distillation process.

Generally, the traditional stage concept is used to simulate reactive separation processes (RSP), either using the equilibrium-based or rate-based stage models (Taylor and Krishna, 1993; Henley and Seader, 2005). The equilibrium-based model (Noeres et al., 2003) assumes that each vapor stream leaving a tray (or packing segment) is in thermodynamic equilibrium with the corresponding liquid stream leaving the same tray or segment. In fact, this assumption is usually far from the real process conditions; therefore, the equilibrium-based theoretical description should be adjusted closer to the reality through correlation parameters such as tray efficiencies or HETS values (Henley and Seader, 2005). However, these correlation parameters often fail for a multicomponent mixture (Stewart and Prober, 1964). Contrary to the equilibrium stage model, thermodynamic equilibrium in the rate-based stage model is assumed only at the phase interface. The rate-based model is a very promising model that directly considers the actual rates of multicomponent mass and heat transfer and chemical reactions (Seader, 1989). Using different theoretical concepts (Kenig, 2000), a separate mass balance is established on each phase by means of component diffusion fluxes (Taylor and Krishna, 1993). In this sense, many models can be classified as rate-based models (Shilkin and Kenig, 2005).

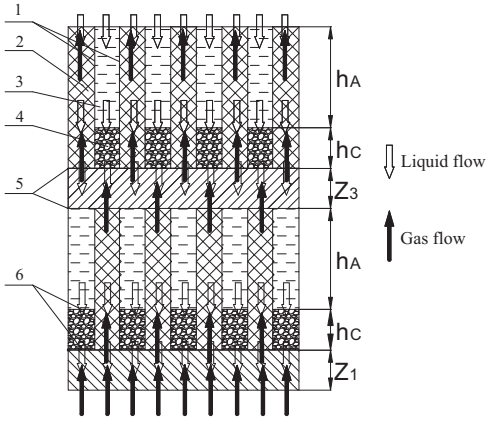

Fig. 1. The schematic diagram of an integrated element of SCPI. (1) Avert-overflow baffles; (2) corrugated metal sheets; (3) liquid layer; (4) catalyst particles; (5) the middle height of the SCPI; and (6) metal wire mesh.

The structure of the internal plays a key role in the actual reaction and separation processes. Establishing a rigorous mathematical model for the new internal is necessary (Brinkmann et al., 2010), so that simulation can be used to identify the role of the important design factors such as the size and location of the reactive and non-reactive sections during process optimization. For the internal SCPI, the processes of the reaction and separation are conducted alternately. According to the structural characteristics of the SCPI, a plug-flow reactor model is used to describe the catalyst bed reactors, and a rate-based model is used to describe the separation processes.

The purpose of this paper is to develop a rigorous model for the description of the catalytic distillation process in SCPI. First, model equations for catalytic distillation processes are built, and gPROMS language is used to compile modules of the reaction, hydrodynamics, and multicomponent heat and mass transfer. Then, to clearly represent the flow-sheet of the catalytic distillation process in SCPI and conveniently change the parameters and variables, a graphical user interface (GUI) design and the corresponding port design are proposed. Next, methods for partial differential equations are described in detail. In addition, solution methods to improve model stability and to achieve faster convergence are also proposed. Finally, comparisons between simulation results and the experimental data are drawn to verify the accuracy of the models.

2. Modeling

Establishing mathematical models on the smallest structural unit of the internal SCPI can not only facilitate the analysis of the catalytic distillation process but also significantly reduce CPU computing time. The following mathematical models established in this study are based on the smallest structural unit of the internal. The flow-sheet of the smallest internal structural unit for the catalytic distillation process is illustrated by the drawing shown in Fig. 2. The reaction and separation sections are arranged alternately (see Fig. 2). Liquid flows downward uniformly through the reaction section and the separation section. Gas flows upward from the bottom, bypassing the reaction section, into the upper separation section. The reaction apparatus is similar to a fixed bed reactor in which only a liquid phase reaction is conducted. Proper height of the avert-overflow baffles (see Fig. 1) above the catalyst bed (catalyst container) ensures that liquid seepage flows through the catalyst bed successfully without overflow. Heat and mass transfer of the gas–liquid countercurrent flow is conducted in the separation section, which is comprised of corrugated metal sheets. In the following paragraphs, we will establish reaction section and separation section models of the SCPI and then establish the flow-sheet of the internal for the catalytic distillation process.

2.1. Reaction section packed with catalyst particles

2.1.1. Model assumptions

The reaction section is similar to a fixed bed reactor, in which only a liquid phase reaction is conducted. A pseudo-homogeneous model is employed to describe the behavior of the reaction, and a “plug flow”