

State-of-the-Art Modeling of Separation Columns: A Review

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Reliable and physically consistent process models are necessary to design and optimize separation equipment. Such models are expected to deliver sufficient information on process characteristics, fill the gaps in understanding the underlying phenomena, and help avoid process malfunctions. Recent progress in computer technology has significantly increased modeling possibilities, especially concerning rigorous methods. This has resulted in high expectations for an accurate and consistent description of separation columns. However, rigorous modeling of separation equipment remains difficult. This is why the spectrum of currently applied modeling methods is large and encompasses both simplified and sophisticated concepts. The choice of an appropriate model depends on different criteria, e.g., possibility of solving the governing mathematical model, availability of process data and model parameters, proper software and computer facilities, user experience, and expectations concerning accuracy and predictivity. It is essential to clearly understand the potential and limitations of different process models. In this contribution, established and widespread modeling methods applied in the fluid separation area are reviewed and critically evaluated. In addition, the complementary modeling concept is presented.

1. Introduction

Modeling fluid separation units is a powerful means of supporting the design and optimization of chemical plants. In the last decades, rigor and complexity have grown significantly, and advanced modeling methods have been implemented in various software tools, mostly commercial ones. This has caused a certain euphoria among the users and numerous results obtained with non-disclosed commercial models have been published. This has also given rise to unreasonable hopes and an unjustified belief in commercial software. In contrast, modeling of separation units must be accomplished on a reasonable background, so that a complete agreement between the modeling targets, available data, and computer power is achieved.

The number of published works concerned with the modelling of fluid separation units is large (see, for instance, a recent review of Blahušiak et al. (2018), with a focus on short-cut methods for process selection, and an analysis of complex distillation structures by Duanmu et al. (2022)). However, in these publications, mostly specific applications are handled using certain specific modelling methods. A solid, comparative analysis of different modeling approaches highlighting their relevance, advantages, and limitations is missing. The aim of this review based on the author's long experience is to close this gap and help the reader select appropriate modeling ways. Along with the critical appraisal of individual approaches, some novel modeling concepts (hydrodynamic analogy, complementary modelling) are presented.

2. Classification of modeling methods

Outstanding achievements in describing the physical background, fluid dynamics, heat and mass transfer, and thermodynamics have opened up new modeling opportunities. This has been supported by the revolutionary progress in computer technology, allowing both advanced experimental and numerical process analysis. Nevertheless, the overall complexity of separation processes remains very high. Multiphase flows, multicomponent systems, complex thermodynamics, complex column unit design and geometry, intricate unit links, and a huge scale difference between the characteristic dimensions of the phenomena involved – often requires significant simplifications in the process description. The level and degree of specific simplifications

depend on several factors: the system's complexity to be separated, the flow pattern, the column design, the particular goals of modeling, and the availability of physicochemical data. All these factors result in high model diversity.

In Figure 1, a classification of the models applied for separation units is shown. Two large groups can be recognized – equilibrium-based and non-equilibrium-based methods. In contrast to the latter group, the first one does not include mass and heat transfer kinetics directly into the modeling framework, taking advantage that the thermodynamic equilibrium often dominates the underlying separation operation. This is particularly true for distillation operations. Many important questions regarding the process feasibility and design can be answered with just equilibrium information (Stichlmair and Fair, 1998).

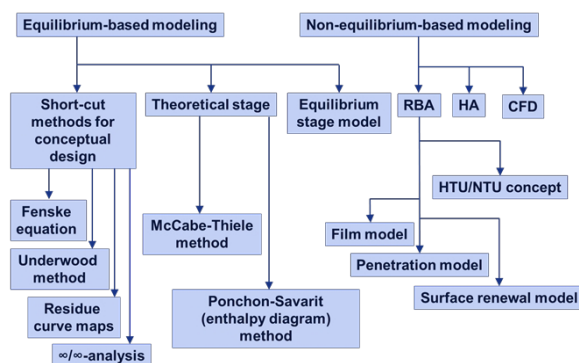


Figure 1: Classification of the modeling methods for separation processes

Non-equilibrium-based models directly include the process kinetics. They can describe column stages (see below); this approach is mostly referred to as the *rate-based approach* (RBA). Non-equilibrium modeling can also be applied outside the stage concept by considering the whole column unit or using periodic representative elements other than stages. These methods may have a very rigorous background, such as, for instance, partial differential equations of fluid mechanics and advanced numerical methods in the so-called *computational fluid dynamics* (CFD). Alternatively, simplified flow patterns can be used within the *hydrodynamic analogy* (HA) approach. Most of these modeling methods are considered more detailed in the subsequent sections.

3. Equilibrium-based modeling

3.1 Short-cut methods

Short-cut methods have long been a vital modeling tool for understanding the basics of the separation process, and they are especially well-elaborated for distillation (Kenig and Blagov, 2014). They are often based on a deep theoretical analysis of process behavior. Hence, they can still serve useful purposes, for instance, when accurate phase equilibrium and enthalpy data are missing, or when (nearly) ideal solutions are considered. Besides, these methods often represent excellent tools for various theoretical studies of separation processes. Short-cut methods are not aimed at obtaining complete process information, e.g., temperature and composition profiles along the column, as more rigorous methods do. Instead, they permit a quick calculation of several specific process characteristics. These characteristics mainly represent the special limiting cases concerning separation limit (e.g., the feasibility of a selected desired separation), investment costs limit (e.g., minimal number of stages required for a specified separation), and operating costs limit (e.g., minimum reflux ratio).

For separation processes accompanied by instantaneous chemical reactions, it is always possible to transform the original concentration space using the approach suggested by Ung and Doherty (1995), so most short-cut methods available for conventional operations can be used. This technique has been further elaborated in Daniel and Jobson (2007) using the so-called boundary value method to handle reactive distillation processes.

3.2 Equilibrium stage

The treatment of a separation column as a cascade of similar segments (the so-called stages) is a common feature of its modeling, as it helps to reduce the description of a typically very large object (column) to a sequence of repeating smaller objects (stages). The stages are identified with real trays or segments of a packed column. This concept has resulted in several different theoretical models for conventional separation processes. These models are based on mass and heat balances combined with thermodynamic equilibrium information, with a wide range of physicochemical assumptions and accuracy.

The equilibrium stage concept assumes that the liquid and vapor phases can exchange species and energy fast enough to attain thermodynamic equilibrium between the leaving streams within one stage. This method is largely used for many practical tasks. Furthermore, the underlying concept of the equilibrium stage modeling has been extended to govern reactive systems (Kenig and Blagov, 2014).

The main disadvantage of the equilibrium-based methods is their insufficient link to actual column design. For instance, it is difficult to predict the column performance accurately without involving mass transfer kinetics and fluid dynamics. This information is particularly important when column internals should be optimized. Equilibrium-based methods usually employ lumped coefficients, which roughly estimate the deviation between real (non-equilibrium) and ideal process behavior, for instance, stage efficiencies. However, this method is not always sufficient, especially in the case of complex systems with several components.

4. Non-equilibrium-based modeling

Non-equilibrium-based modeling should be considered an extension of the process consideration toward more rigor and physically consistent treatment of complex phenomena in separation units. Here, mass and heat transfer rates are directly considered. However, it is tough to fully capture their sophisticated dependence on column internals, dynamic fluid regime, and physicochemical system; therefore, experimental assistance is required. Experimentally obtained correlations mostly realize this for mass and heat transfer coefficients and specific interfacial contact area (Wang et al., 2016).

Some non-equilibrium modeling methods have been developed in the last decades that do not require mass and heat transfer coefficient correlations (Kenig, 2008). Such methods (CFD, HA approach) go beyond the stage concept, considering, where possible, either an entire separation unit or its periodic elements.

4.1 HTU-NTU concept

This simple approach is applicable to packed columns for which tray efficiencies do not apply. It allows the column height to be determined as a product of two values, HTU (height of a transfer unit) and NTU (number of transfer units). The NTU value can be determined graphically or numerically by integrating an inverse of the mass transfer driving force over the column height. It does not depend on the unit itself and reflects mixture separation difficulty. The HTU value depends on the column load, internals-related geometrical parameters, and empirical mass transfer correlations. The total mass transfer resistance is often assigned to one of the contacting phases (Sherwood et al., 1975). In most cases, an assumption of a constant molar overflow is applied.

In the literature, correlations for HTU values for various internals and process conditions can be found (e.g., (Hegely et al., 2017)). However, it should be mentioned that the interfacial area and mass-transfer coefficients depend upon the mass flow rates, which may vary significantly, even if the molar flow rates remain constant. Thus, the constancy of the HTU-values should be used with care. Further details on the application of the HTU-NTU concept can be found elsewhere (Kenig and Blagov, 2014).

4.2 Film model and its extensions

This model is central within the non-equilibrium stage modeling. Initially proposed by Lewis and Whitman (1924), the film model approaches its 100-year-jubilee. This model assumes that the resistance to mass transfer is concentrated entirely in thin films adjacent to the phase interface and that mass transfer occurs within these films by steady-state molecular diffusion alone. Outside the films, in the fluid bulk phases, the level of mixing is assumed so high that there is no change in composition at all. This means that in the film region, one-dimensional diffusion transport normal to the interface takes place.

The film model has some similarities with the phenomena at many real fluid elements in two-phase systems, e.g., fast change of the main process variables (composition, temperature) close to the interface and their significantly slower change far from the interface. However, the flow pattern in the film model is strongly simplified, and no feasible momentum equation can be used here. The films represent model elements rather than any kind of real film flow. Their thicknesses cannot be measured; instead, they are estimated using mass transfer coefficient correlations – also known as Sherwood correlations (Di Natale et al., 2021).

Alternative ways to govern column stages are the penetration model of Higbie (1935) and the surface renewal model of Danckwerts (1951). In both these models, mass transfer is described using a combination of deterministic (transient diffusion) and stochastic (residence time distribution of fluid elements) principles.

All presented models have much in common; they try to replace real phenomena at the interface with a one-dimensional simplified picture. They exclude real flow and hence exclude the momentum transport equations from the consideration. Therefore, they cannot estimate their main parameters (e.g., the film thickness or average residence time) without additional information. Thus, they are forced to use experimental correlations for the mass transfer coefficients. In this respect, they have the same weak point and should be considered of equal value for the mass transfer prediction. Nevertheless, the film model appears advantageous, since there

is a broad spectrum of correlations available in the literature, for all types of internals and systems, while for the penetration/surface renewal model, the choice is limited.

The film model can be modified to describe more complex separation units. While its extension to homogeneous reactive systems is straightforward (reaction considered either in the bulk or/and in the film), the treatment of systems with heterogeneous catalysts is definitively more sophisticated and depends both on the type of catalyst (porous or non-porous) and on its placement in the internals (pellets, catalytic bags, coating, catalytic trays). The film model can also be extended to govern dividing wall columns. In this case, two stages are combined in a larger one, with four film regions (Mueller and Kenig, 2007).

Provided that all kinetic parameters are properly evaluated, the film model and its extensions can yield reasonably accurate results. However, applying this modeling concept to equipment prototyping is hardly possible because of its strong dependence on experimental parameters.

4.3 Hydrodynamic analogy concept

An attempt to avoid using rough fluid-dynamic simplifications brought about a method called the hydrodynamic analogy (HA) approach (Kenig, 1997). This approach is an alternative way to describe the fluid dynamics and transport phenomena in processes in which the exact location of the phase boundaries is impossible. The basic idea of the approach is an adequate replacement of the actual complex fluid dynamics in a column by a combination of geometrically simpler flow patterns. Such a geometrical simplification has to be done according to experimental observations of fluid flow. Once the observed complex flow is reproduced by a sequence of the simplified flow patterns, the partial differential equations of momentum, energy, and mass transfer can be applied to govern the transport phenomena in an entire separation column.

The idea of the HA method put forward by Kenig (1997) has been successfully applied for different separation operations, among others for distillation (Shilkin and Kenig, 2005) and reactive absorption (Brinkmann et al., 2014) processes in columns equipped with corrugated sheet structured packings.

In the last few years, valuable insights into the fluid flow in the packing have been gained from the results of tomographic measurements (Bolenz et al., 2021). Analysis of tomographic images provides quantitative characteristics of the gas-liquid interfacial area, mixing points of liquid flow and fluid patterns other than liquid films. With this information, the HA models can be refined accordingly. The HA method can be used as an alternative design approach for packed columns. It is also applicable for theoretical optimization of packing geometry, as it does not depend on experimental mass transfer coefficients. This method also has a good potential for applications in which mass transfer coefficients are difficult to determine experimentally.

Recently, a new HA model has been developed to describe separation units filled with random packings (Saltén et al., 2021). The liquid flow in the real packing is captured by a combination of film, jet and droplet flow, which are assumed to be the dominating flow patterns in the bed. The overall model is more complex than for structured column internals. Nevertheless, it was successfully validated for an absorption process carried out in an industrial-scale column filled with two different random packings. This shows that the HA modeling concept can be applied even beyond the range of structured packed columns.

4.4 Computational fluid dynamics (CFD)

CFD simulation today represents a powerful approach to many physicochemical problems, and the availability of advanced commercial tools facilitates its application. In the area of single-phase flow, significant progress can be recognized. Despite that separation processes are inherently multiphase, single-phase simulations can be helpful, for instance, if pressure drop issues are studied (Atmakidis and Kenig, 2009). In contrast, multiphase flow simulations still cannot be considered mature enough to treat separation columns, especially if the separation efficiency and hence mass transfer phenomena have to be considered.

When the phase contact is very intensive, the fluid phases can be considered as interpenetrating continua, and the so-called *Euler-Euler flow modeling* can be applied (Gaurav et al., 2001). The fluid phases and their interfaces are then modeled as media continuously distributed over the computational domain, while source terms cover phase interactions in the transport equations. Using this way, some groups attempted direct Euler-Euler modeling of separation units. However, there is no physically grounded method to determine source terms encountered in the species transfer equations (the mass transfer rate per unit volume). Such source terms are necessary to describe the interfacial mass transfer. It is suggested to determine them using macroscopic models (e.g., based on the film model) and experimentally obtained correlations for mass transfer coefficients. Such an approach is certainly inconsistent.

When the fluid phases in a separation column are not fully mixed, a free moving interface between them should be localized. Different strategies to handle moving interfaces are classified into moving mesh methods, fixed mesh methods, and a combination of both. In the first method, a moving mesh is used to track the interface. As the form of the interface changes, the mesh is adjusted in accordance with the change. Methods falling under this strategy are called *front tracking methods* (Ferziger and Perić, 2002). The second approach uses a fixed

(Eulerian) mesh, while the interface is tracked using different procedures, e.g., special markers or indicator functions. Such methods are called *front capturing methods*; they include marker-and-cell, surface capturing, and volume capturing approaches (Ferziger and Perić, 2002).

In the last decades, several CFD studies of two-phase systems with free moving interfaces have been carried out to investigate the wetting properties of structured packings (Amini and Nasr Esfahany, 2019). This reflects the growing interest in a detailed understanding of the flow in structured packings. Such simulations identify wetting problems for different packings and systems which largely agree with the corresponding tomographic measurements (Bolenz et al., 2021).

The development of such CFD-based methods represents an ongoing research area. However, the simulation of large-scale (reactive) separation columns appears to be too difficult, mostly because of numerical problems and conflict of different scales in the unit model. Often, the position of the phase interface is impossible to capture and the boundary conditions cannot be adequately set (Kenig, 2008). Consequently, a direct application of fluid-dynamic equations to modeling conventional separation units remains unfeasible. In contrast, CFD can be beneficial for modeling micro-separation units (Chasanis et al., 2010). In this case, the flow is more structured and the scale difference is significantly smaller than in conventional separation equipment.

5. Complementary modeling

Another important area in which CFD can successfully be exploited is determining process and model parameters in the complementary modeling concept recently suggested (Kenig, 2008). A truly complementary effect is achieved via information transfer between the different modeling approaches. For instance, CFD simulations of small periodic elements in packed columns can generate the data necessary to realize the RBA (e.g., fluid-dynamic quantities, mass transfer coefficients) in the form of numerically determined parameter correlations. Such CFD simulations are performed with very fine grids and sufficient flow resolution. The subsequent post-processing provides the required functional dependencies of the fluid-dynamic and mass transfer parameters, transferred to the RBA models. Such simulations can be called virtual experiments. They open a perspective toward largely experiment-free process optimization.

A similar link is also conceivable between the CFD and HA methods. CFD simulations provide valuable information about the turbulent flow used within the HA modeling. In this way, the number of necessary real fluid-dynamic and mass transfer experiments can be reduced, and column internals can be optimized directly “on screen”, without the need to manufacture them for the model validation. Since the HA modeling does not require knowledge of mass and heat transfer coefficients, it can also be used for their experiment-free determination and delivery to the RBA in the form of correlations.

At the moment, experiment-free design is still a vision, because the present capability of rigorous multiphase models is not sufficient. However, there exist some examples that bring optimism. Above all, this is related to the impressive progress in developing multiphase CFD-based models and computer facilities. This allows simulation-based analysis covering a broad spectrum of process variables and yields parameter correlations capable of competing with the corresponding experimental correlations.

A methodology for developing optimal internals based on a combination of CFD and RBA in a unique optimization algorithm was suggested by Egorov et al. (2005). A part of this procedure is sensitivity studies performed with a rate-based process simulator. The internals can then be modified in accordance with the results of these studies, and the modified internals can be further investigated by CFD simulations to derive relevant correlations for the RBA simulator. After several iterations, internals perfectly fitting the process criteria can be virtually created and afterward manufactured. In the long-term perspective, this method can be regarded as a way toward virtual prototyping of new, process-specific internals for separation units, provided that chemical companies and manufacturers of the internals are interested in collaborating.

Another successful demonstration of the complementary modeling concept is developing a new structured packing best suited for post-combustion CO₂ absorption. Here, even three different approaches, namely CFD, HA and RBA were employed, each delivering complementary information on the packing performance. The details of this impressive application example are given in (Kenig, 2017).

6. Conclusions

Fluid separation units can be modeled by very different methods. In this paper, a classification and a systematic overview of these methods are given. The entire spectrum of modeling accuracy is covered, starting from shortcuts and finishing with the most rigorous CFD approaches.

Rigorous modeling of fluid separation units remains a challenge due to the multiphase flow pattern that is difficult to capture. Despite recent significant progress, CFD modeling of large separation columns is hardly possible. In contrast, it can be successfully applied to the description of small-scale equipment units.

CFD can be reasonably well employed within the complementary modeling concept, for the evaluation of model parameters (virtual experiments). The complementary modeling implies a proper model choice, with due account of data, resources and criteria. Moreover, this permits the synergistic application of different approaches and suggests a promising modeling way for the coming decades.

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