annual review

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Distillation

Writing on the general subject of distillation continues to be prolific. The authors considered well over 500 publications in preparing this review. Many of the publications did not contain significant contributions or were foreign versions of material already published in English. Others turned out to have little connection with the principles or practices of distillation. Of those selected for inclusion, many contained interpretive or tutorial material thought to be of value by the present authors. A relatively few contained significant breakthroughs in our state of distillation knowledge. This combination of little advancing and much interpretation appears to result from the high state of maturity of distillation technology, coupled with a continuing need for its proper exploitation. In the future, we may expect a steady flow of distillation literature that can be justified primarily on the economic leverage of distillation practice.

This review covers publications that appeared from July 1968 through June 1969, in sequence with the previous review (*17*). Emphasis is on the practitioner audience; more fundamental material is covered in other *I&EC* reviews, particularly the one on Mass Transfer (*62*).

General Works

This year the important general works on distillation were in German. The regular literature review of Huebner (*77*) lists 312 references through mid-1968 and covers absorption and extraction in addition to distillation. The review of Hoppe *et al.* (*75*) has similar coverage and lists 286 references through 1967. A biennial monograph on chemical technology literature for 1964 and 1965 appeared in late 1967 (*112*) and should have been noted in last year's review by the present authors; the section of the monograph on distillation lists 460 references. These three German reviews place emphasis on the European literature and should be consulted by those making exhaustive searches.

Another German reference that should be of particular interest to the distillation practitioner is the fourth edition of the classic text by Kirschbaum (*84*). Off the press in mid-1969, the new edition has about 20% expanded coverage with major revisions in the areas of high-vacuum distillation and tray-type contacting device performance. This reference contains information on the performance of commercial-scale distillation devices that is not readily available elsewhere.

A final German reference is the book by Billet (*12*) that summarizes some of the author's extensive studies of distillation equipment performance and economics. This work, like those mentioned above, is not available in English translation. Those not familiar with technical German may find the distillation dictionary (*164*), mentioned last year, to be of some assistance in scanning these references.

Also appearing were some English-language books relating to distillation. The second volume of a pair by Prausnitz and coworkers (*133*) on computer calculations for vapor–liquid equilibria appeared, this one covering high pressure applications. The seventh volume of the series on "Advances in Chemical Engineering" (*39*) includes chapters on high pressure vapor–liquid equilibria and on gas–liquid dispersions. A general treatise by Pratt (*131*) on countercurrent separation operations includes a great deal of material related to distillation. Finally, the volume edited by Perry (*125*) includes a chapter on foam fractionation.

Research

There appeared to be a slight upswing in United States university research related to distillation. Of 371 thesis titles listed (*31*), 13 had a direct relationship and perhaps two dozen lent direct support. Popular areas of research continue to be column dynamics and control, and mass transfer capability of contacting devices.

Commercial-scale research in the United States is carried out largely by Fractionation Research, Inc. (FRI), and some of the results of FRI are being disclosed publicly. A motion picture showing the behavior of sieve and bubble-cap trays under high-vacuum distillation conditions was shown at the annual meeting of the A.I.Ch.E. (*179*) and may be purchased or borrowed from FRI. Also, FRI test data for Raschig ring packings were presented at the International Symposium on Distillation at Brighton, England, in September 1969. The Brighton symposium was a sequel to the important and successful 1960 symposium with the same title. Its program will be covered in next year's review.

The demise of distillation as an analytical tool was duly noted by Williams (*174*). However, distillation is not absent from the laboratory, being employed extensively for the production of small quantities of high purity material. The Williams paper reviews developments in equipment used for such purposes.

HIGHLIGHTS

- Significant papers and general works appearing in the German literature
- Impressive theoretical work on hydrodynamics in contacting devices originating in Czechoslovakia
- Large-scale fractionation research facility reported in the USSR
- Extensive activity in general, rigorous, stage calculation methods, computer-implemented
- Beginning work in modeling for simultaneous distillation and chemical reaction
- Continued trend for revival of design-oriented studies

Physical Properties

Diffusion coefficients. Models for the prediction of mass transfer efficiency in distillation require values of the gaseous and liquid molecular diffusion coefficients. For the gas phase it is usually assumed that composition has an insignificant effect on the coefficient; studies by Mrazek *et al.* (*116*) indicate that this assumption must be considered carefully. These authors showed that for the chloroform–air system at 50 °C there was a 9% variation in coefficient over a chloroform composition range of 0 to 57 mol %. This was a fairly rigorous test, and for engineering calculations the concentration-independence assumption may still be valid.

Prediction of the gaseous diffusion coefficient has been studied extensively, and several semiempirical methods have been developed. Lugg (*100*) presents new data for 147 organic and other vapors diffusing through air with related tests of nine prediction methods. He found that the Wilke–Lee equation (*173*) gave the best fit in general, but for C_5–C_9 aliphatic esters, C_5–C_8 acids, and C_5–C_8 alcohols the Hirschfelder–Bird–Spotz equation (*71*) was best. The Lugg paper includes a tabulation of the experimental diffusion coefficient values.

Liquid phase diffusion coefficients remain in the "difficult" field for distillation practitioners. As pointed out by Cullinan (*34*), much effort is being devoted to developing an understanding of the mechanism of the diffusion process in liquid systems while also providing reliable methods for prediction of diffusion rates in concentrated binary or multicomponent systems. The general predictive approach utilizes an equation such as that presented by Vignes (*165*):

$$D_{ij} = (D_{ij}^0)^{x_j}(D_{ji}^0)^{x_i}\left(1 + \frac{d \ln \gamma_i}{d \ln x_i}\right)$$

where the first two diffusion coefficients on the right refer to infinite dilution conditions and x refers to mole fraction of the constituents in the liquid. The activity coefficient term shows the relationship of the composition dependence to nonideality. The Vignes equation emphasizes the need for data on infinite dilution coefficients and a number of prediction methods have been developed. Experimental data for ethylene in water, taken by Huq and Wood (*78*), show the Wilke–Chang (*172*), Othmer–Thaker (*123*), and Ferrell–Himmelblau (*44*) methods to be the best of nine considered. Lusis *et al.* (*102*) have developed a new method that

TABLE I. STAGE CALCULATIONS, CONTINUOUS[a]

Subject	*References*
Binary-equivalence concept	(*68*)
Computer programs	(*66, 70, 146*)
Concentration maxima	(*9*)
Enthalpy, ideal	(*9, 110*)
Nonideal	(*65, 129, 146, 147*)
Feedplate, optimum	(*67, 148, 170*)
Heat leak, allowance for	(*65*)
Iteration schemes, algorithms	(*59, 66, 70, 135, 146, 168*)
Models, mathematical	(*9, 59, 66, 126, 143, 146*)
Multiple feeds, side draws	(*65, 146*)
New fundamental concepts	(*16, 110*)
Optimization	(*67, 146*)
Phase equilibria, ideal	(*9*)
Nonideal	(*65, 70, 146*)
Physical property models	(*70*)
Reflux ratio, minimum	(*126, 135*)
vs. plates	(*58, 98, 161, 162, 163*)
Single-stage (flash)	(*66, 70*)
State-of-the-art review	(*15, 146*)
Systems, binary	(*15, 65, 110, 129, 147*)
Complex	(*66, 143*)
Multicomponent	(*9, 16, 58, 59, 68, 70, 98, 110, 126, 146, 148, 161, 162, 163, 168, 170*)
True-boiling-point curves	(*66*)

[a] *Explanation of indexing of Table I and other reference tables: Each reference is indexed under all subjects covered, providing both single-entry and concept-coordinate access. For example, Ref. 146 discusses a computer program. The same reference also covers such subjects as iteration schemes, optimization, and state-of-the-art review.*

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TABLE II. HYDRODYNAMICS IN PACKED COLUMNS

Subject	*References*
Absorption	(22)
Boundary conditions	(40, 89, 154)
Capacity, flooding	(90, 118)
Column diameter, 4-in.	(118, 155, 156, 157)
6–8-in.	(21, 40)
10-in.	(40)
Diffusion coefficients	(155)
Distribution of liquid	(40, 89, 118, 154, 155, 156, 157)
Experimental	(21, 40, 60, 118, 155, 156, 157)
Feed distribution, liquid	(21, 155, 156)
Flow, countercurrent	(21, 51, 53, 54, 86, 87, 88, 90, 118)
Trickle	(60)
Holdup	(21, 51, 53, 54, 87, 118)
Mechanisms	(40, 60, 86, 89, 90, 118, 154)
Models, modeling	(21, 86, 87, 88, 89, 90, 154, 155, 156, 157)
Packing diameter, 0.2-in.	(21, 157)
0.4-in.	(21, 40, 118, 155, 156, 157)
0.6–1.0-in.	(21, 40, 157)
Various	(54)
Packings, mesh or wire	(54)
Raschig rings	(21, 40, 54, 118)
Saddles	(40)
Spheres	(21, 155, 156)
Various	(52)
Phases, single	(60, 89, 154, 155, 156, 157)
Two	(21, 51, 53, 54, 86, 87, 88, 90, 118)
Pressure drop	(21, 86, 87, 88, 118)
Properties of fluids	(21, 52, 157)
Residence time distribution	(22, 60)
State-of-the-art review	(86, 89)
Vacuum distillation	(52)
Wall effect	(89, 154, 155, 156)

shows considerable promise for cases where the solvent is not water; it eliminates the need for estimating an "association parameter" and represents the entire range of solute and solvent molar volumes in one equation. For aqueous systems, the Othmer–Thaker equation was found to be best.

Vapor–liquid equilibria. As usual, this review makes no attempt to list the many papers dealing with the measurement and correlation of vapor–liquid equilibrium data. Much of this work is published in the *Journal of Chemical and Engineering Data* and in the Russian *Journal of Applied Chemistry.* Also, a series of papers on VLE has reached its 40th installment in the *Collection of Czechoslovak Chemical Communications* (mostly in English).

Some general works of interest to the practitioner should be noted. The book by Prausnitz and Chueh (*133*) summarizes computational approaches to the prediction of high pressure VLE. The paper by Fleck and Prausnitz (*45*) describes a simple and useful mathematical model for estimating binary VLE. The paper by Arai *et al.* (*7*) shows how the Scatchard–Hildebrand theory of regular solutions can be useful in predicting solvent selectivity for extractive distillation. Along the same line, Tassios (*160*) presents a five-step procedure for choosing extractive distillation solvents. Finally, the review of the thermodynamics of VLE by Prausnitz (*132*) is particularly useful to those with little background in solution thermodynamics.

One specific research paper of more than passing interest is that of Zel'venskii *et al.* (*181*). These workers investigated the very dilute regions of binary systems, from several per cent down to $10^{-8}\%$. They covered 40 solutions and used radioactive tracer techniques. Although they found the simple extrapolations could lead to error, they did not find any existence of microazeotropes. They also found that the mass transfer kinetics exhibited no fundamental peculiarities in the very dilute region. This type of work is of importance to the preparation of ultrapurity materials by fractional distillation.

Stage Calculations

Continuous distillation. Table I presents a summary and index to the articles on continuous distillation. Some of the more significant contributions are discussed below.

The most comprehensive article appearing this year on computer programming of the general problem of continuous multicomponent distillation was authored by Sargent and Murtagh (*146*). This article starts with a complete state-of-the art review, proceeds with exposition of the basic mathematical models, continues with a discussion of iterative schemes and algorithms, and concludes with a discussion of computer programming. The general model will handle multiple feeds, multiple side-draws, nonideal phase equilibria, and nonideal enthalpies. There is also a discussion of optimization for the purpose of design.

Several new concepts were noted. One is the bypassing of the plate efficiency concept by Mecklenburgh and Hartland (*110*). They take issue with the concept of plate efficiency because it may vary widely through the column. Although this does not seem to be a problem to this observer, the authors proceed to develop a model which "allows for finite transfer rates and backmixing in both phases," based on a definition of "pseudo equilibrium" and "pseudo operating" lines. In the exposition of another new concept by Bogoslovskii and Shamolin (*16*), the calculation of "kinetic factors" for the process of fractionating a multicomponent mixture "via the kinetics of concentration change of one of the components" is developed.

Asano and Fujita (*9*) discuss the phenomenon of the maximizing of the concentration of a particular component on a particular plate. Their mathematical analysis provides the basis for determing whether this phenomenon will occur, and under what ranges of conditions.

Surprisingly, there is still considerable "rehashing" of old empirical and shortcut design methods. The classical method for estimating minimum reflux ratio for multicomponent systems originally developed by Underwood (*161, 162, 163*) was discussed in two articles (*126, 135*). The well-known empirical correlation between plates and reflux ratio of Gilliland (*58*) was fitted with a mathematical equation (*98*). The graphical method of Ponchon (*129*) and Savarit (*147*) for binary systems with nonideal enthalpies was extended to nonadiabatic conditions (*65*).

Batch distillation. Goldman and Robinson (*61*) report on the simulation of batch distillation with CSMP (continuous system modeling program), a program for simulating analog-type computations on a digital computer. The program assumes binary distillation, zero holdup, and the McCabe–Thiele model. The results of an example are presented in graphs and tables. This was later extended (*136*) to multicomponent systems, and to include process economics. The latter article also includes a good state-of-the-art review of the computer solution of batch distillation.

Absorption, stripping. Huckabay and Garrison (*76*) review the conventional design approaches for absorption or stripping of a single component to or from a solvent in a packed column. Models are recommended for flooding, mass transfer, and the calculation of HTU for typical packing materials. The application of the conventional method to a special type of process problem is discussed by Guerreri (*63*): the absorption of ammonia in water at high pressures in packed columns. The unusual design problems

TABLE III. HYDRODYNAMICS IN PLATE COLUMNS

Subject	*References*
Bubble size and distribution	(*137*)
Capacity, flooding	(*113*)
Column diameter, 4–6-in.	(*93, 113, 137*)
12-in.	(*25, 158*)
20-in.	(*152*)
Design of experiments	(*113*)
Entrainment	(*25*)
Experimental	(*25, 93, 113, 120, 134, 137, 152, 158*)
Froth, density	(*93, 120, 134, 152, 158*)
Height	(*93, 120, 134, 152, 158*)
Geometry variation	(*25, 93, 113, 120, 158*)
Grid plates	(*113*)
Interfacial area	(*137*)
Load variation	(*25, 93, 113, 120, 158*)
Mechanisms	(*137, 158*)
Mixing	(*134*)
Models	(*137, 158*)
Optimization	(*113*)
Photographic techniques	(*93, 137*)
Plate efficiency	(*93, 113, 134*)
Pressure drop	(*25, 120, 152, 158*)
Sieve plates	(*25, 93, 134, 152, 158*)
State-of-the-art review	(*158*)
Statistical techniques	(*69, 113*)
Tracer techniques	(*134*)

here are heat of solution, solvent vaporization, sensible heat transfer, and the application of intercoolers.

Owens and Maddox (*124*) authored a comprehensive paper on theoretical stage calculations for multicomponent absorption and stripping. Intended primarily as a hand method, it is based on the new concept of "terminal fractional absorption," defined as the fraction of total gas absorption that occurs on the two terminal trays (the top tray and the bottom tray). The authors found the terminal fraction absorption to be remarkably constant in the neighborhood of 80% for a large number of cases calculated by a rigorous computer program. This finding provided the basis for short-cut correlations for estimation of vapor/liquid ratio and temperature as a function of column position. This article also provides an excellent state-of-the-art review of stage calculations in connection with multicomponent absorption.

Mostafa (*115*) developed a method for multicomponent absorption calculations based on curved operating lines.

Hydrodynamics

This review year marked the appearance of seven new papers on distillation by V. Kolar, and associates, of the Czechoslovakian Academy of Sciences. This brings the total to 11 papers on distillation hydrodynamics appearing from this single source during the past three years. These papers include five on the distribution of liquid over random packing, five on two-phase countercurrent flow through beds of packing, and one starting a new series on the hydrodynamics of plate columns. These works involve a sophisticated level of theoretical modeling, experimental determination of model parameters, and experimental demonstration of model validity. These and other works in the field of hydrodynamics are covered below.

Packed columns. A summary and index to the references on hydrodynamics in packed columns is given in Table II,

The work of Kolar and associates was previously noted. The first series on packed columns covers the distribution of liquid over random packing. Part I by Kolar and Stanek (*89*) presents a state-of-the-art review, and a theoretical analysis of models. Part II by Stanek and Kolar (*154*) develops the additional model derivations required, including boundary conditions for the wall effect. Part III by Stanek and Kolar (*155*) presents experimental data in which the feed is introduced at a point, in the center, with a bed diameter sufficiently large so that the liquid never reaches the wall, thereby eliminating the wall effect. Part IV by Stanek and Kolar (*156*) employs the data from Part III to determine the model parameters. Part V by Stanek and Kolar (*157*) extends the model with new experimental data on Raschig rings of various sizes, and on fluids of varying properties.

The other Czechoslovakian series on packed columns is on two-phase countercurrent flow. Part I by Kolar and Broz (*86*) presents a state-of-the-art review, theoretical analysis, and derivation of necessary models for the flow mechanisms. Part II by Broz and Kolar (*21*) presents experimental data in an 8-in. column packed with 0.4-in. glass spheres. The feed was uniformly distributed, and measurements were made of the holdup and pressure drop. Part III by Kolar and Broz (*87*) evaluates the data from Part II for the determination of model parameters for pressure drop and holdup. Part IV by Kolar and Tichy (*90*) extends the packing models to include the flooding phenomenon. Part V by Kolar and Broz (*88*) extends the model for pressure drop over the entire range of gas flow rates, and to holdup at zero flow of gas.

Although the Kolar work to date has been most impressive theoretically, study of Table II reveals that the experimental work has been limited to small columns, small packing sizes, few packing types, and limited system properties. With these restrictions, the results at present are of limited commercial utilization. It is hoped that future publications from this source will remove these restrictions.

Plate columns. A summary and index to the literature on hydrodynamics in plate columns are presented in Table III. Pertinent comments follow.

Steiner and Kolar (*158*) have started a new series on the hydrodynamics of plate columns. In this Part I, they present a state-of-the-art review, and a discussion of various models for the prediction of froth height, froth density, and pressure drop. Also given are experimental data obtained in a 12-in. column fitted with sieve trays. An interesting finding was that froth density is not constant with respect to elevation.

Kuehler and Van Winkle (*93*) studied the height and relative density of froth on sieve trays by photographic techniques. Holdup and mixing on sieve trays were studied by Rane and Pavlov (*134*), employing a tracer technique. Photographic techniques were used by Rodinov and Radikovskii (*137*) for the determination of interfacial area and the study of the distribution of gas bubbles.

Mass Transfer and Plate Efficiency

It is well-known that distillation involves material transport between vapor and liquid phases across a boundary (the interface) under the influence of concentration driving forces. This concept provides a convenient means for discussing the literature on mass transfer and plate efficiency.

The similarity between mass transfer and the transfer of heat and momentum has been described many times; the review by Fulford and Pei (*49*) provides a convenient summary of these transfer processes with emphasis on their common mathematics. In an unusual paper, Rony (*142*) continues a series on the "extent of separation" and discusses diffusion and rate control in an equilibrium stage; his index for covering nonequilibrium is "extent of segregation." For gas phase transfer, Vivian and Schoenberg (*166*) showed the equivalence of gas desorption and vaporization for determining coefficients, but pointed out that liquid surface lifetime can vary widely between the two processes. Lusis (*101*), on the other hand, pointed out that the variation of interface concentrations in distillation places restrictions on the use of rate data obtained in constant-composition vaporization experiments. Wasan and Wilke (*169*) concluded that the effect of the concentration level of nondiffusing species on gas phase transfer is the same in turbulent transport as in molecular transport. The importance of heat effects on transfer rates was noted by Bourne and Coggan (*18*), and computations for determining temperature effects on the value of the gas phase mass transfer coefficient were discussed by Mueller (*117*).

For liquid phase transfer, Yoshida and Arakawa (*180*) reported a pressure effect on the transfer coefficient for oxygen in water. A tentative explanation was that pressure influenced the surface renewal characteristics at the interface. Pressure is ordinarily neglected as an influence on liquid phase transfer rate. Ho and Prince (*72*) showed mathematically that the liquid phase transfer coefficient for bubbles with a size distribution may be estimated within reasonable error on the basis of the average surface-volume mean diameter.

Concentration driving force for mass transfer is strongly influenced by axial mixing tendencies of the flowing streams. Gunn (*64*) reviewed mixing of liquids and gases flowing through packed beds, but did not consider the case where the phases coexist. Piterskikh (*127*) presented liquid mixing data for a 0.3- × 1.0-m sieve tray, based on KCl tracer measurements. Furzer (*50*) and Aleksandrov (*2*) analyzed the effect of vapor distribution on the efficiency of cross-flow trays. Mellish (*111*) cautioned against the use of axial mixing data for packed columns at low throughput when designing for the practical situation of operation above the packing "load point." Other mixing studies (*81, 82*) were concerned with general mathematical models.

As usual, a great many papers dealt with absorption accompanied by chemical reaction. For the case of distillation, such studies are mainly useful in their elucidation of interfacial area and basic mass transfer concepts. In some cases, distillation is accompanied by reaction (*33*). For the most part, however, the effort toward establishing effective interfacial area is of most importance; in this regard Linek and Mayrhroferova (*99*) have provided a useful review and Jhaveri and Sharma (*80*) have presented experimental area data for a small packed column. Other area data for sieve plates were presented by Rodionov and Vinter (*140*) and Pohorecki (*128*). The other papers on absorption-reaction (*32, 35, 42, 69, 83, 119, 122, 141*) were concerned with mathematical aspects of combining diffusion with reaction in first- and second-order situations.

Specific efficiency data on packed contacting devices were reported. Special packings covered were polygonal helicoids made from wire (*144*), cellular ceramic spheres (*26, 27*), zigzag plates with vertical corrugations (*19*), and small metal or glass spirals (*52*). In each case the authors found areas of operation where their packings appeared to have advantages. The influence of liquid distribution of liquid phase mass transfer rate in a 0.5-m packed column was found to be minimal (*57*). Roughness of packing or plate surface, on the other hand, was shown to have a significant effect (*36, 176*). A method for predicting packed tower transfer rates, using graphs, was presented (*76*). In general, there appears to be much interest in packed columns for distillation applications.

Efficiencies of 150-mm diameter Turbogrid trays, operating with the ethanol–water system, were reported by Foeldes and Evangelidi (*46*) and later correlated by a simple equation involving Froude number, pressure, and liquid–gas ratio (*47*). The effect of vapor density on the efficiency of grid trays was studied (*139*); the total effective area was increased with increasing density which accounted for increased efficiency. Another paper on grid trays (*55*) showed that glass balls between the trays provided a fluidized-bed effect and markedly improved efficiency at high load.

For cross-flow trays Sharma *et al.* (*149*) investigated efficiencies of a 20.5-cm^2 bubble-cap tray, using absorption techniques; a correlation of the data was developed. Rodionov and Radikovskii (*138*) found that the height of the froth level on a sieve plate affected interfacial area but not mass transfer rate. Boyarchuk *et al.* (*20*) found froth densities of about 0.4 for bubble-cap trays with methanol–water and concluded that the mass transfer coefficient should be related only to weir setting and not to total height of froth. Diener and Gerster (*38*) measured point efficiencies in a small column designed such that gas phase would be controlling and liquid mixing would be complete. A prediction method was developed which checked both binary and ternary data quite well. Slobodyanik *et al.* (*153*) studied the orientation of openings on a small jet tray and found that vapor directed to give tangential action gave highest transfer rates.

Other studies related to distillation efficiency dealt with the collection of data on commercial systems, using an analog–digital system connected to a digital computer (*3*), data for a 2.0-m commercial naphtha fractionator (*105*), and surface tension effects on efficiency (*43*).

TABLE IV. CONTACTING EQUIPMENT

Subject	*References*
Capacity, flooding	(*1, 10, 13, 14, 26, 27, 30, 94, 114, 144, 159, 183*)
Column diameter, 3-in.	(*26, 27*)
8-in.	(*1, 144*)
1-ft	(*150, 184*)
2-ft	(*13, 85, 150, 159*)
3–4 ft	(*1, 10, 13, 14, 91, 114, 150*)
7-ft	(*94*)
14–17 ft	(*48*)
Comparisons of equipment	(*10, 11, 13, 14, 26, 27, 30, 48, 85, 91, 114, 144, 159*)
Economics	(*10, 11, 13, 14, 48*)
Efficiency	(*1, 10, 13, 14, 26, 27, 48, 85, 91, 144, 159*)
Entrainment	(*10, 30, 85, 94*)
Experimental	(*1, 10, 11, 13, 14, 26, 27, 30, 48, 85, 91, 94, 97, 114, 144, 159, 183, 184, 185*)
Froth height, downcomer	(*94*)
Tray	(*48, 94*)
Geometry variations	(*10, 14, 85, 94, 184, 185*)
Holdup	(*144*)
Large holes	(*94*)
Load variations	(*1, 10, 13, 14, 26, 27, 30, 48, 85, 91, 94, 144, 183, 184*)
Models	(*10, 13, 184*)
Optimization	(*10, 11, 13, 14*)
Overflow devices, without	(*1*)
Packings, ceramic, low-density	(*26, 27*)
Mesh/gauze	(*13, 97*)
Polygonal helicoids	(*144*)
Pall rings	(*13*)
Raschig rings	(*97*)
Vertical, parallel-plate	(*183*)
Plates, bubble-cap (BC)	(*91*)
Combination BC/sieve	(*30*)
Combination valve/sieve	(*10, 11, 14*)
Film, 2-zone	(*185*)
Grid	(*13, 159*)
Jet	(*91, 114*)
Linde	(*48*)
Multiple liquid zones	(*114*)
Pipe	(*85*)
Sieve	(*85, 94*)
Uniflux	(*91*)
Valve	(*1, 10, 11, 13, 14, 85, 184*)
Pressure drop	(*10, 11, 13, 26, 27, 48, 85, 94, 144, 159, 183, 184*)
Specific-volume concept	(*10, 11, 13*)
State-of-the-art review	(*13*)
Test installations	(*150*)
Test systems	(*97*)
Vacuum operation	(*10, 13, 14, 48*)
Venturi nozzles	(*10, 11, 14*)
Weeping, dumping	(*85, 94*)

Contacting Equipment

The summary and index to articles oriented toward contacting equipment are given in Table IV. Discussion of selected papers follows.

Billet (*13*) presents a comprehensive state-of-the-art review of the commercially available contacting devices. Experimental data are given from tests of these devices on the ethylbenzene/styrene system at 100-mm Hg abs in columns 2 and 3 ft in diameter. Plate efficiency and pressure drop data are correlated as a function of load for valve trays, grid trays, Pall rings, and gauze packing. An economic optimization is developed based on the concept of "specific volume" as the objective function.

Lemieux and Scotti (*94*) offer important new data on the performance of commercial-scale sieve trays. A unique feature is the testing of sieve trays with large holes, ranging from 0.5 to 1 in. in diameter. Data reported include flood capacity, pressure drop, entrainment, tray froth height, weeping/dumping, seal pot level, and downcomer froth level, all (except flooding) as a function of loading. The simulator had a cross-section of 7 ft^2.

The first public dislosure of the design and performance of Linde (Linde Division, Union Carbide Corp.) proprietary trays was presented by Frank, Geyer, and Kehde (*48*). The installation is in a distillation column separating styrene and ethylbenzene at a still-head pressure of 50-mm Hg abs. The column ranged from 14.5 to 17 ft in diameter. Mechanical details of the Linde tray are illustrated in drawings. The experimental data presented include pressure drop, plate efficiency, and froth height as a function of load. Economics are also discussed.

In an article by Sheinman *et al.* (*150*), a commercial-scale distillation equipment research facility is described at the State Scientific Research and Design Institute for Petroleum Equipment, in the USSR. This installation contains eight test units with diameters ranging from 1 ft to 4 ft. The tests can determine such items as the hydraulic resistance of a plate or its component parts, liquid entrainment, operating ranges of a plate as a function of entrainment or flooding, static level of the liquid on the plate, longitudinal liquid mixing, concentration distribution, and mass transfer on the plate. It is assumed that this is similar to the commercial-scale distillation test facility of Fractionation Research, Inc., in the U. S., the Czechoslovak Academy of Science in Czechoslovakia, and the Badische Anilin-und Soda-Fabrik AG, Germany.

Distillation Processes

Azeotropic and extractive distillation. Tassios (*160*) discusses the choosing of solvents for extractive distillation. A procedure of five steps is suggested for identifying likely solvents. There is discussion of the relationships among activity coefficients, selectivity, and relative volatility. Theory is offered on the nature of physical and chemical effects of various types of solvents. Alternate models are discussed for the prediction of activity coefficients. The article concludes with an example of solvent selection for the separation of cyclohexane and benzene.

A review of the design approach to azeotropic distillation is offered by Yamada, Sugie, and Abe (*178*). The degrees of freedom of azeotropic distillation in a two-column system with a one-layer reflux are analyzed, and a design method is given for determining the number of plates in each column, the optimum feed location, the loss of entrainer, and the composition of the decanter. A worked example is included.

Foam fractionation. Gerster (*56*) defines distillation as "the separation of the constituents of a ...... mixture by partial vaporization of the mixture and separate recovery of vapor and residue." He goes on to state that, in distillation "the more volatile constituents of the original mixture are obtained in increasing concentration in the vapor, the less volatile in greater concentration and the liquid residue." Lemlich (*96*) defines foam fractionation as "the technique for partially separating or concentrating dissolved material by adsorption at the surfaces of bubbles; these rise up through the solution, forming a foam that is then removed."

It should be apparent, then, that foam fractionation is not a member of the distillation family tree. However, since foam fractionation is sometimes miscalled "foam distillation," it was thought appropriate to include a few key references on this subject for elucidation. Besides the previous reference, an excellent state-of-the-art review with 147 references, on foam fractionation was authored by Lemlich (*95*).

System Dynamics and Control

The general literature on distillation dynamics and control is covered in the review by Williams (*175*). Emphasis here is on a few papers of special interest to the distillation practitioner.

Control theory for distillation systems occupied the attention of several workers. Anisimov and colleagues at the Moscow Chemical Engineering Institute described a simplified algorithm for determining optimum control policy (*4*); the basic criterion for optimization was taken to be minimum energy requirement per unit of distillate product. In another paper (*5*), the same workers described a method for determining transfer functions for columns with many plates. Optimum control policy was also developed for the extractive distillation separation of C_4 hydrocarbons in a 132 sieve plate system (*107*); this paper includes some interesting steady-state data on a Russian installation. Theory relating to the suboptimal control policy of a gas absorber was presented (*29*).

There were several experimental–theoretical studies of equipment dynamics. The behavior of a 1.0-ft packed column absorbing CO_2 in triethanolamine was reported by Ankel *et al.* (*6*); the CO_2 content of air was pulsed. Spontaneous pressure oscillations in a "flooded" packed absorber (really a packed-sparged bubble column) were related to packing layer depth, packing size, and gas and liquid loadings (*86*). The same type of column was analyzed on a frequency response basis, using a cell model for mass transfer (*109*); experimental data were included. The pulse response of a valve plate was studied (*37*), and the cell model for mass transfer was found to be applicable for this situation also. The response of a 24-plate, 8-in. benzene–acetone fractionator was studied by pulsing the reflux flow (*106*); it was found that the system could be approximated by linear first-order equations. The literature on batch fractionation was essentially nonexistent. One paper noted (*186*) dealt with the combination of reaction and batch fractionation from a theoretical viewpoint. The work led to the definition of an optimum still charge which, if exceeded, would prevent enrichment of the bottoms with the heavy component. Another paper (*92*) presented a dynamic model for a batch column.

Several papers were of direct value to the distillation practitioner without extensive automatic control background. Chan and Talbot (*28*) discussed the usefulness and limitations of two common feedback systems and one common feedforward system. Their recommendations were supported by 300 computer runs on a simple binary system. Luyben (*103*) proposed a feedforward control scheme that would manipulate feed tray location instead of reflux or boilup ratio as commonly used. The effectiveness of the scheme was demonstrated by digital simulation of a 20-plate column. McNeill and Sacks (*108*) described a bang–bang control system that was successful for a large ethylbenzene–xylene fractionator. In this scheme, overhead product flow was alternately on and off. Buckley presented two papers dealing with override controls and also containing useful information on the general problem of fractionator control. The first paper (*23*) dealt with principles of overrides for distillation, while the second (*24*) aimed at the problem of controlling a column from which a large side-stream is withdrawn.

Controlled cyclic distillation is a process which is continually upset in order to gain efficiency and capacity advantages. Commercial applications indicate deficiencies due to lags caused by the larger scale. Wade *et al.* (*167*) have investigated these scaling problems and have proposed control schemes and equipment modifications to solve them.

Distillation Design

Much of the information reviewed up to this point is pertinent to the execution of distillation design. However, design involves much more than an analysis of physical properties, stage calculations, hydrodynamics, contacting equipment and distillation processes. Design involves the combination of all the above submodels in such a way as to produce the optimum commercial design. Thus, design is concerned not only with technology, but also with optimization, economics, and design strategy.

Unfortunately, during the recent revolution in chemical engineering education, the concepts of unit operations, distillation, and design have fallen into disfavor. It is almost as if it were thought

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that the end result desired is engineering science, not scientific engineering, which is the application of engineering science to reach commercial achievements. Fortunately, last year marked a revival of interest in design-oriented studies of distillation. This year proves to continue this welcome trend.

Equilibrium stage calculations. An important special case of distillation is the calculation of the separation achieved in a single equilibrium stage for either multicomponent or complex mixtures, and either at a specified temperature or a specified system enthalpy.

Shelton (*151*) presents a model and computer program for design of the equilibrium stage for multicomponent systems, either with or without heat balance. The program includes submodels for the estimation of phase equilibria and enthalpy data for hydrocarbon systems. Output includes the equilibrium vapor and liquid compositions, and, as limiting cases, the bubble point and the dew point. Included are the complete models required, as well as the necessary programming algorithms.

Hoffman (*73*) addresses himself to the problem of equilibrium vaporization calculations for complex mixtures. His approach avoids the usual approximation of subdividing the complex mixture into a vector of discreet multicomponents. Instead, he derives a new analytical solution based on the assumption of Gaussian distribution in the differential curve. The new model provides the ability for conversion of True-Boiling-Point (TBP) curves to Equilibrium-Flash-Vaporization (EFV) curves, with the bubble point and dew point as special cases.

Because ASTM distillation curves are generally more readily available than TBP curves, a companion article by Hoffman (*74*) derives an analytical solution to the problem of converting ASTM to TBP curves.

Optimization. Porter and Ashton (*130*) discuss the problem of optimizing the design of packed columns for vacuum distillation of heat-sensitive materials. Their objective function for the optimization is minimum pressure drop per theoretical plate. By combining mass transfer theory with hydrodynamic pressure drop, the authors develop analytical expressions which can be optimized.

The optimization of the design geometry of valve trays is discussed by Nitschke (*121*).

Safety factors. The rational determination of safety factors to be employed in chemical engineering design has been sorely neglected. Welcome indeed, then, is the paper by Saletan (*145*) on the estimation of safety factors for distillation design. The method developed is based on an application of statistics and optimization of the economics involving the cost of overdesign, on the one hand, and the cost of failure to meet expected performance on the other.

Standards. Engineering standards are justified as reliable and economic solutions to repetitive or routine engineering problems. They are aimed at the lowest cost method which will provide required performance over a reasonable service life. In view of the many repetitive and routine components of a distillation process design, it is surprising that there has not been more effort in the area of standardization.

An excellent example of standardization of distillation equipment is given by Eduljee (*41*). The author documents what has been done in India for the standardization of distillation sieve plates. The situation was this: there was much more advantage to getting the job done economically than in creating new, sophisticated, custom-engineered designs. An interesting aspect of the India project is the inclusion of standards for the prediction of performance, as well as for equipment design. Although this may seem undesirable from a standpoint of our developing technology, it has led to "the selection diagram," which provides the basis for selecting the optimum available standard geometry. The article also presents in detail the models included in the standard performance prediction.

Internal appurtenances. It is not a rare circumstance for a misdesigned feed entry arrangement, packing support, or reboiler vapor return line to bottleneck the capacity of an entire distillation system. Hence, it is important that all of these internal appurtenances be understood thoroughly and designed properly. This year a number of useful articles appeared on this subject.

Jamison (*79*) provides the designer with a summary of various column and tray design techniques used successfully throughout the industry in recent years. These recommendations include reboiler vapor return arrangements, introduction of intermediate feeds, transition sections between differing tray designs, seal pan designs, arrangements for total drawoff and partial drawoff, design for unusually low liquid rates, design of 3- and 4-pass trays, and various mechanical design considerations.

Design criteria for chimney trays are offered by Wheeler (*171*). Chimney trays are used in place of enlarged downcomers for removing the liquid downflow from a column while allowing rising vapors to pass up the column, especially in packed columns.

Manovyan, Skoblo, *et al.* (*104*), discuss the pitfalls of improper trap-out tray design for fractionators with intermediate reflux generators.

Analysis of performance. A method of plotting diagrams of the lines of constant composition are proposed for interpreting the course of fractionation processes (*182*).

Air distillation. Armstrong and Schofield (*8*) review their computer program of distillation calculations for the special case of air separation in an oxygen plant. This is an example of how general design methods may be converted to special-purpose programs. This approach may be justified for an engineering organization whose concern is one exclusive type of distillation process.

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