Chandrasekaran, S. K., A. S. Michaels, P. S. Campbell, and J. E. Shaw, "Scopolamine Permeation Through Human Skin In Vitro," *AIChE J.*, **22**, 828 (1976).
Elfbaum, S. G., and K. Laden, "The Effect of Dimethyl Sulfoxide on Percutaneous Absorption," *J. Soc. Cosmet. Chem.*, **19**, 841 (1968).
Horita, A., and L. J. Weber, "Skin Penetrating Property of Drugs Dissolved in Dimethyl Sulfoxide (DMSO) and Other Vehicles," *Life Sciences*, **3**, 1389 (1964).
Kligman, A. M., "Topical Pharmacology and Toxicology of Dimethyl Sulfoxide," *J. Am. Med. Assoc.*, **193**, 923 (1965).
Michaels, A. S., S. K. Chandrasekaran, and J. E. Shaw, "Drug Permeation Through Human Skin: Theory and In Vitro Experimental Measurement," *AIChE J.*, **21**, 985 (1975).
Scheuplein, R. J., and I. H. Blank, "Permeability of the Skin," *Physiol. Rev.*, **51**, 702 (1971).
Stoughton, R. B., and W. Fritsch, "Influence of Dimethylsulfoxide (DMSO) on Human Percutaneous Absorption," *Archs. Derm.*, **90**, 512 (1964).

Manuscript received November 23, 1976; revision received July 13, and accepted July 21, 1977.

Synthesis of Heat Exchange Networks by Mixed Integer Optimization

Process synthesis involves manipulation of the process arrangement, while studying the variables of each arrangement, to arrive at the optimal process. If each process arrangement is treated as a discrete variable, process synthesis becomes a mixed integer optimization problem. This paper examines the synthesis of heat exchanger networks using the adaptive random search procedure, which can be used to search continuous and discrete independent variables simultaneously. The means of handling the heat exchanger arrangement as a discrete variable is discussed, and the incorporation of various synthesis heuristics is presented. The results of synthesis of 2 × 2, 2 × 3, and 3 × 3 networks are presented and compared with other methods of synthesis.

R. C. KELAHAN
and
J. L. GADDY
Department of Chemical Engineering
University of Missouri-Rolla
Rolla, Missouri 65401

SCOPE

Synthesis of heat exchange networks is usually treated as a discrete optimization problem. Recent studies of these systems have successfully utilized branch and bound techniques to reduce the computational effort (Lee et al., 1970; Pho and Lapidus, 1973; Rathore and Powers, 1975). Each of these studies has been concerned with finding the optimal arrangement of the network and has not considered optimization of the individual exchangers. Improvement in the economics and different optimal arrangements might result if the quantity of heat transferred in each exchanger is optimized along with the order of the exchangers. The optimization problem then becomes mixed integer, and suitable algorithms have not been demonstrated for these problems.

The adaptive random search procedure has been successfully applied to the mixed integer reliability problem (Campbell and Gaddy, 1976). The purpose of this study is to examine heat exchanger synthesis as a mixed integer optimization problem using the adaptive random search. Examples from the literature were chosen for study so that a comparison between optimal networks could be made to evaluate any advantage in treating synthesis problems as mixed integer. The reliability and efficiency of the adaptive random search are measured and reported for three problems solved previously by Lee et al. (1970) and Pho and Lapidus (1973).

CONCLUSIONS AND SIGNIFICANCE

The adaptive random search with heuristics was found to be an effective method for solving heat exchanger synthesis problems. Simultaneous optimization of the heat transfer and exchanger sequence resulted in improved economics for two of the three examples considered.

As might be expected for more complicated problems, the search region for the mixed integer synthesis problem was found to be very poorly behaved. To aid the search, in these cases, heuristics are employed to reduce the computational effort. Termination of a stream at its desired temperature, when possible in an exchanger, was found to speed the search without impairing the accuracy.

The use of heuristics with the adaptive random search resulted in perfect reliability in solving each of the examples. Only slightly more computer time is required to solve these problems as mixed integer. The method is quite simple to apply to synthesis problems and should receive broad application to more complicated systems.

Correspondence concerning this paper should be addressed to J. L. Gaddy, R. C. Kelahan is with Amoco Chemicals Corporation, Naperville, Illinois.

Synthesis of chemical processes involves finding an equipment arrangement that will achieve the process objectives and locating at a set of independent variables that will optimize that process arrangement. Determination of the optimal process is usually approached as two separate optimization problems. First, a feasible process arrangement and its optimal conditions are found. Conventional optimization procedures are employed for this step. Second, a search for the optimal process is conducted by manipulating the process arrangement and determining new optimal conditions.

The second step often employs heuristics or discrete optimization strategies. Hendry and Hughes (1972) optimized the units in a separation system and then applied a tree search method to find the optimum order of units. Rathore et al. (1974) used dynamic programming to obtain optimal combinations of separation systems and heat exchange systems. King et al. (1972) applied heuristics in the synthesis of several processes with previously optimized unit operations.

Another approach to solving the two optimization problems is to treat the process arrangement as a discrete variable and optimize all the variables simultaneously. This problem is, of course, mixed integer and requires not only a mixed integer optimization technique but also a strategy for including the equipment arrangement as a process variable.

Perhaps the most widely studied synthesis problem is that of heat exchange systems, where an optimum array of heat exchangers is to be found satisfying each stream's temperature requirements. Kesler and Parker (1969) applied linear programming with stream subdivision and subsequent integration to optimize heat exchange systems using a linear area-cost relationship. Kobayashi et al. (1971) used a similar approach allowing parallel stream splitting. Masso and Rudd (1969) considered a power relationship between exchanger area and cost and performed the optimization using heuristics and weighting factors (heuristics with learning). A branch and bound method was developed by Lee et al. (1970) for problems without stream splitting, and this was further developed and applied by Menzies and Johnson (1972). Pho and Lapidus (1973) applied a tree search algorithm to heat exchange problems and achieved very good efficiency. Rathore and Powers (1975) further developed the tree search method and considered the use of utilities before stream matching and transportation costs for the streams in between matchings. Siirola (1974) reviewed in detail many of the above methods and offered suggestions for improvement and generalization of the methods.

Each of the above methods is concerned with the ordering of the heat exchangers and does not consider optimization of the individual exchangers. The minimum allowable approach temperature in each exchanger is fixed at some predetermined value. With this restraint, finding the optimal exchanger arrangement can be considered a discrete optimization problem, the discrete variable being the process arrangement.

It is reasonable to expect that the optimum approach temperature for each exchanger should be determined by the economics of the process, since the approach temperature affects both the exchanger area and utility usage. If the approach temperatures are added as variables, the synthesis of the heat exchanger network becomes a mixed integer optimization problem. Such an approach might lead to an improvement in the economy of the system, if the optimum approach temperatures are dependent upon the ordering of the exchangers.

The purpose of this study was to examine the synthesis of a chemical process as a mixed integer optimization problem. Specifically, the adaptive random search technique, which can be applied to mixed integer problems (Campbell and Gaddy, 1976), was applied to the synthesis of heat exchange systems. The objective was to investigate problems previously solved in the literature and to compare results and efficiencies with other procedures. A comparison between the optimal networks should provide a basis for determining whether there is an advantage in treating synthesis problems as mixed integer.

ADAPTIVE RANDOM SEARCH PROCEDURE

The adaptive random search optimization technique was introduced by Gall (1966) and further developed by Luus and Jaakola (1973), Heuckroth et al. (1976), and Campbell and Gaddy (1976). The method chooses, at random, values for the independent variables in the region surrounding the last best value of the objective function.

The basic equation for determining new values for the independent variables is given in Equation (1) (Heuckroth et al. 1975):

$$x_i = x_i^* + \frac{R_i}{k_d}(2\theta - 1)^k \tag{1}$$

The search is conducted by determining new values of the independent variables from Equation (1) and evaluating the objective function. When an improvement in the objective function is found the x_i^* are replaced with new values and the search continued. As the optimum is approached the probability of sampling near the last x_i^* is increased by raising the distribution coefficient k and by reducing the sampling range (increasing k_d). Thus, the search converges toward the optimum in the same manner as a hill climbing technique.

The selection of k and k_d has an important influence on the efficiency of the method. Increasing these parameters, when near the optimum speeds the search. However, if k and k_d are increased too rapidly, convergence to a local optimum may occur.

Equation (1) can be used for continuous variables and, with slight modification, can be used for integer and discrete variables. Integers are obtained by algebraically manipulating the random number, then using the round-off characteristics of Fortran in converting from real to integer format. Evenly spaced discrete numbers can be obtained by multiplying the integers by a constant. An unevenly spaced set of discrete numbers can be obtained by assigning each discrete number to an integer obtained by the above method. With uniform random numbers, Equations (2) and (3) produce uniformly distributed integers. Nonuniform discrete variables can be obtained by a method presented by Campbell and Gaddy (1975).

Uniform integers are obtained from Equations (2) and (3):

$$R = (I_B - I_A + 1)\theta + I_A \tag{2}$$

$$I = R \text{ (Fortran statement)} \tag{3}$$

HEAT EXCHANGE SYNTHESIS PROBLEMS

The three problems used as examples were previously solved by Lee et al. (1970) and Pho and Lapidus (1973). Examples 1 to 3 are problems 4SP1, 5SP1, and 6SP1, respectively, in the above articles. In each example, there are M streams to be heated and N streams to be cooled. Each stream has an associated flow rate, heat capacity, and inlet and outlet temperatures. These data are summarized in Table 1. Example 1 has two streams to be heated and two to be cooled (2×2), example 2 is 2×3, and example 3 is 3×3.

Table 1. Process Stream Specifications

Example	Stream	Inlet temp., °C	Required outlet temp., °C	Capacity flow rate, J/s-°C
2 × 2	C_1	60.0	160.0	7 621
	C_2	115.6	260.0	6 081
	H_1	160.0	93.3	8 792
	H_2	248.9	137.8	10 548
2 × 3	C_1	37.8	204.4	11 392
	C_2	65.6	182.2	12 921
	C_3	93.3	204.4	13 027
	H_1	248.9	121.1	16 613
	H_2	204.4	65.6	13 290
3 × 3	C_1	37.8	221.1	8 438
	C_2	82.2	176.7	17 278
	C_3	93.3	204.4	13 897
	H_1	226.7	65.6	14 767
	H_2	271.1	148.9	12 552
	H_3	198.9	65.6	17 721

Table 2. Heat Exchanger Design Data

Steam pressure: (saturated)
Example 1
6 636 kN/m² (962.5 lb/in.²abs)
282.2°C (540°F)
1 527 kJ/kg (656.6 Btu/lb)

Examples 2 and 3
3 103 kN/m² (450 lb/in.²abs)
235.7°C (456.3°F)
1 785 kJ/kg (767.4 Btu/lb)

Cooling water temperature: 37.8°C (100°F)
Maximum water outlet temperature: 82.2°C (180°F)
Overall heat transfer coefficients:

Heat exchanger: 851.5 J/m²-s-°K (150 Btu/hr-ft²-°F)

Steam heater: 1 135.4 J/m²-s-°K (200 Btu/hr-ft²-°F)

Water cooler: 851.5 J/m²-s-°K (150 Btu/hr-ft²-°F)

Equipment down time: 380 hr/yr

Heat exchanger cost parameters: $a = 350$, $b = 0.6$

Annual rate of return: $\delta = 0.1$

Cooling water cost: $0.00011/kg ($0.00005/lb)

Steam cost: $0.002205/kg ($.001/lb)

* Temperatures and heats of vaporization from Keenan and Keyes (1936).

The heating or cooling requirements of each stream are to be satisfied by matching streams until no further feasible matches can be made. Steam heaters and water coolers provide any additional heating or cooling to achieve the required outlet temperature.

The goal is to obtain a minimum cost network that satisfies all the process requirements. Equation (4) is the annual cost function to be minimized:

$$Y = \delta\left[\sum_J a(A_J)^b + \sum_K a(A_{CK})^b + \sum_L a(A_{HL})^b\right] + U \quad (4)$$

The area for each heat exchanger is calculated by the log mean heat transfer equation. The fabrication cost of the heat exchangers was assumed to have a power relationship with the exchange area. The utilities cost is determined by summing the steam and cooling water requirements per year for each stream and multiplying by the cost per pound of these utilities. All design parameters necessary to solve the problems are presented in Table 2 and are identical to those used by other authors.

Several conditions were fixed in solving these problems. First, only countercurrent heat exchange is considered. A matching between any particular cold stream and hot stream is allowed to occur only once in the network. In addition, no stream splitting or merging is allowed. Transportation costs (pipes and pumping) and control costs were assumed negligible or constant. Finally, any utility usage was assumed to occur after the last match.

Following Pho and Lapidus (1973), a matrix representation of the heat exchanger network was used. A system of M cold streams and N hot streams forms an $M \times N$ array, with each position in the array representing a heat exchanger, as shown by the sample network in Figure 1. An integer J assigned to a position in the array represents the order in which the streams are to be matched. The notation at the edges of the array describes the utility requirements for the various streams.

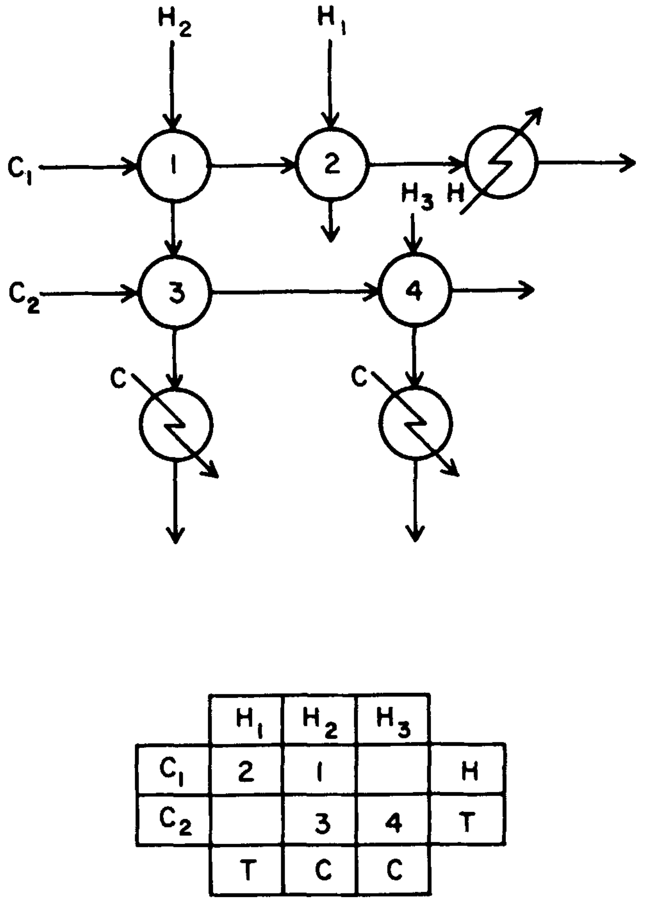


Fig. 1. Sample network with matrix form.

APPLICATION OF THE ADAPTIVE RANDOM SEARCH

The random search technique was used both in the optimization of the heat transfer in each exchanger Q_J and in choosing the sequence of matching process streams. For M cold streams and N hot streams there are MN exchangers, each identified by an integer, J (1, 2, 3, . . ., MN). Streams were identified by assigning an integer C_K (K = 1, 2, 3, . . . , M) to each of the cold streams and an

Table 3. Comparison of Heat Exchanger System Costs

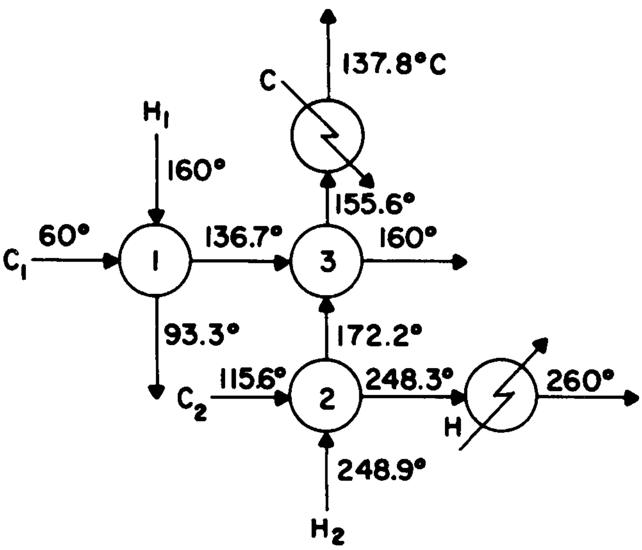
Example	Minimum cost ($/yr) ARS this study	Pho and Lapidus (1973) and Lee et al. (1970)
2 × 2	$10 634	$13 685
2 × 3	$38 316	$38 268
3 × 3	$35 048	$35 659

integer, $H_L (L = 1, 2, 3, \ldots, N)$ to each of the hot streams.

For each exchanger J, cold and hot streams were matched by selecting integers K and L using Equations (2) and (3). For example, in exchanger $1 (J = 1)$, if $K = 2$ and $L = 1$, heat transfer is indicated between C_2 and H_1. Of course, it is necessary to insure that this is a feasible match (no negative approach temperatures) and that this match has not occurred previously. Even though an integer J is assigned to each position in the array, no exchanger may be necessary, since streams are terminated when the desired exit temperature is achieved. Once the order of the network has been determined, the heat transfer in each exchanger Q_J is found using Equation (1).

RESULTS AND DISCUSSION

By allowing the heat transfer (or exchanger area) in each exchanger to vary while simultaneously varying the exchanger sequence, cost reductions were obtained in the 2 × 2 problem and the 3 × 3 problem. A comparison with results obtained by previous investigators, using fixed approach temperatures, is given in Table 3.



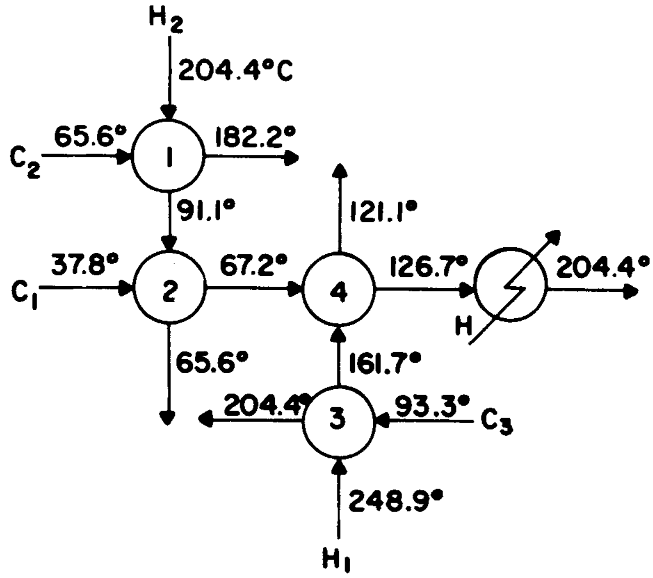
Q_1 = 586. kJ/s $\quad$ A_1 = 24.7 m^2

Q_2 = 806.6 kJ/s $\quad$ A_2 = 74.1 m^2

Q_3 = 176. kJ/s $\quad$ A_3 = 13.4 m^2

	H_1	H_2	
C_1	1	3	T
C_2		2	H
	T	C	

Fig. 2. Solution for 2 × 2 example.



Q_1 = 1507.5 kJ/s $\quad$ A_1 = 74.3 m^2

Q_2 = 338.4 kJ/s $\quad$ A_2 = 15.5 m^2

Q_3 = 1447.4 kJ/s $\quad$ A_3 = 30.6 m^2

Q_4 = 675.4 kJ/s $\quad$ A_4 = 18.2 m^2

	H_1	H_2	
C_1	4	2	H
C_2		1	T
C_3	3		T
	T	T	

Fig. 3. Solution for 2 × 3 example.

The arrangement for the 2 × 2 problem (Figure 2) is identical to that obtained by Lee et al. (1970) and Pho and Lapidus (1973), but the exchange areas are different. The cost reduction was achieved by allowing the heat transfer area in exchanger 2 to reach an optimum. The tendency was toward a larger exchanger: 74.1 m^2 as opposed to 29.2 m^2 in the arrangement obtained in the previous investigations. The larger amount of heat transferred resulted in less steam needed to heat C_2 and less cooling for H_2. As a rule, increasing exchanger area to decrease utilities usage was economical up to a minimum approach temperature of about 1°C. The larger exchanger in the 2 × 2 problem resulted in a savings of $3 880/yr on utilities at the expense of an additional $825 annual cost for the heat exchangers.

The optimum system obtained for the 2 × 3 problem is presented in Figure 3 and is identical to that obtained by the previous authors. The slight differences in cost obtained for the arrangement are believed due to different steam values used. The steam properties used in this study are from Keenan and Keyes (1936). Varying the heats transferred in this case did not improve the cost because in the optimum arrangement, all streams except one can be terminated at the desired temperature without

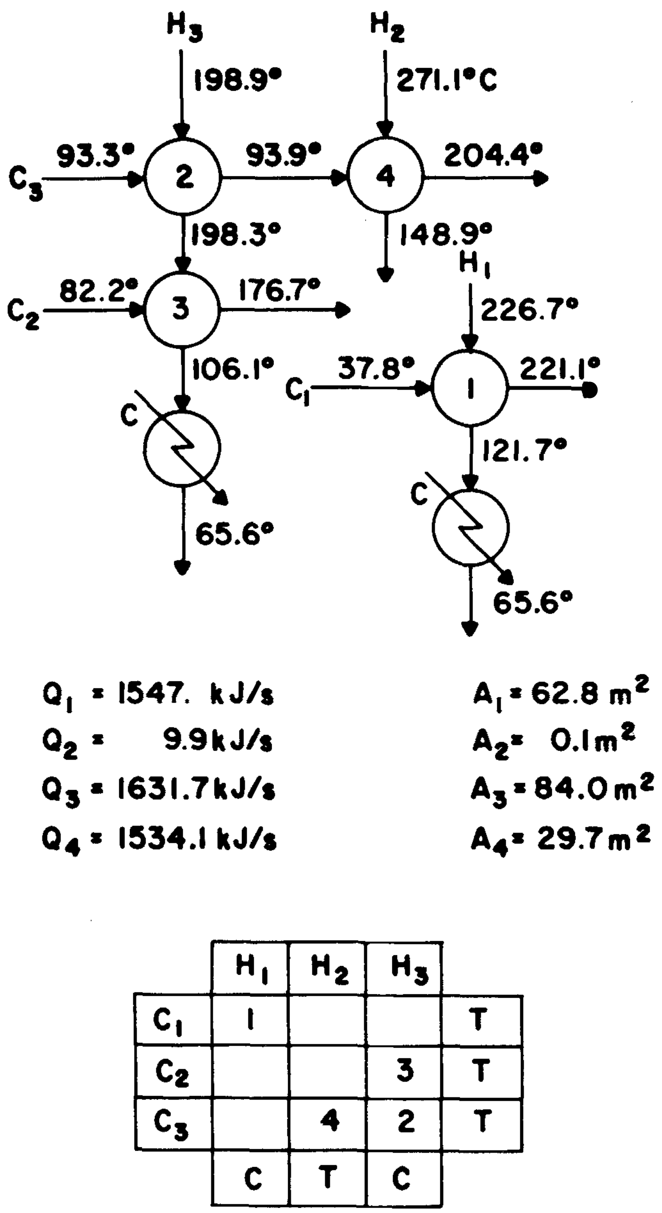


Fig. 4. Solution for 3 × 3 example.

TABLE 4. EFFICIENCY OF ARS

Example	No. runs	Average function evaluations to 0.1% of optimum: Basic	Termination heuristic	Termination heuristic with initial matching heuristic
2 × 2	50	9 600	209	279
2 × 3	50	8 200*	13	17
3 × 3	50	8 800†	1 568	1 944

* Five runs, efficiency to 11% of optimum.
† Five runs, efficiency to 15% of optimum.

external heating or cooling. Thus, the transfer of heat and the exchanger areas are fixed.

The optimum arrangement for the 3 × 3 problem differed from that obtained by the previous authors. The optimal arrangement found in this study is shown in Figure 4. This network resulted in a savings of $611 over the arrangement obtained in previous studies. In the new arrangement, the consideration of lower approach temperatures resulted in the elimination of a heat exchanger and a water cooler.

It may be concluded that treating the heat exchange synthesis problem as a mixed integer problem by varying the heat transfer areas can result in a significant improvement in economics in some cases. Savings of 30% in the annual cost were possible in one example by reducing the utilities consumption. It does not appear that finding the optimal network, followed by optimization of the exchanger areas, would always lead to the optimal solution. This procedure could be followed successfully for the simplest problem, example 1, but would fail for the more complicated problem, example 3.

EFFICIENCY OF THE ADAPTIVE RANDOM SEARCH

Efficiency in this study is based on the number of function evaluations required to reach 0.1% of the optimum. Since a random search procedure is used, the efficiencies obtained are also random. Consequently, fifty runs (except where noted) were performed for each example to obtain a reliable value for the average efficiency. The efficiencies are reported with a perfect reliability (except where noted); that is, each of the fifty runs converged to within 0.1% of the optimum.

Comparisons of efficiency with other authors is difficult not only because of the different techniques used, but also because additional variables were considered. Computation times ranged from 0.14 to 44.0 on the IBM 370/168 computer for the three examples considered, which compares favorably with computation times reported for other methods.

The Basic Search Technique

Efficiencies for the three examples using the basic algorithm are presented in Table 4. The basic method was able to find the optimum only for the 2 × 2 example. The difficulty encountered in solving the more complex problems is due to the necessity of searching over a very large number of possible networks. There is no correlation between exchanger arrangement and heat transfer in each exchanger. Therefore, in order to obtain an improved arrangement during the course of the search, an entirely different set of Q_J must be obtained. Consequently, locating the optimum becomes nearly an exhaustive search. The inefficiency is also due to the search for Q_J being performed over a wide range even when a stream could be terminated. Consequently, the method cannot be applied effectively to any but the simplest networks without a set of rules to improve the efficiency.

Termination of a Stream Heuristic

In order to improve the efficiency of the random search in application to heat exchange synthesis problems, a heuristic was invoked which would allow stream termination during the search. A stream was terminated if its desired outlet temperature could be achieved in an exchanger, thus eliminating the need to search for the optimal quantity of heat transferred in that exchanger. With this heuristic, no auxiliary exchangers are required, a condition which should result in an optimal arrangement. However, if allowing a stream to terminate results in a very small approach temperature, the cost of the large exchanger area might not be offset by the reduced utilities cost. The results with the 2 × 2 problem suggest a limiting approach temperature of 2°C below which the heuristic was not applied.

The heuristic was applied to all three example problems. In using the heuristic, first it was determined which of the two streams in a match might be terminated. Then the resulting heat transfer equation was checked to make sure there was not a negative approach temperature or an approach temperature of less than 2°C.

The efficiencies using the above heuristic are reported for the three examples in Table 4. As noted, the efficiencies for each example were significantly improved using the stream termination heuristic. The greatest improvement was noted in the 2 × 3 case, where the optimum was located as soon as the optimum sequence was selected. This sequence allowed termination of all streams except one, resulting in all the Q_J being fixed. In the 2 × 2 case, the optimum sequence resulted in all the Q_J except one becoming fixed. In the 3 × 3 case, the optimum sequence fixed all but two of the Q_J.

For the 3 × 3 problem, an alternate arrangement was obtained for about 15% of the runs at 0.1% of the optimum. This arrangement is shown in Figure 5. It is very similar to the optimum arrangement, with identical utility costs and only slightly higher exchanger fabrication costs. The total cost for the system is $35 053 as opposed to $35 048 for the optimal arrangement.

It may be concluded that the stream termination heuristic definitely speeds the search for the optimal heat exchange system. This heuristic does not impair the accuracy of the answer obtained, allowing convergence to the optimum with perfect reliability in all the trials made.

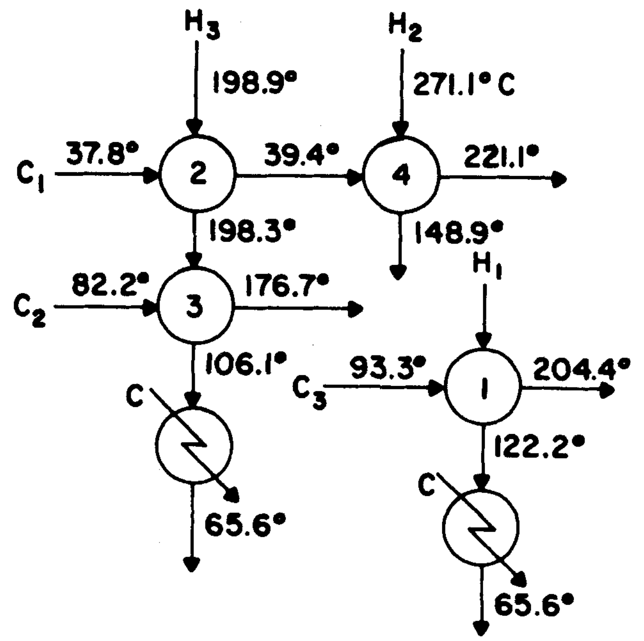
It should be noted that by using this heuristic there exists a possibility with some problems that termination on a subsequent matching would be more economical than termination at the first possible opportunity. This possibility exists for the 2 × 2 example, with H_2 being terminated instead of H_1. This system has been reported by Rathore and Powers (1975) and results in identical utility costs with a slight savings in exchanger costs, for an annual savings of $42 (about 0.4%). The insignificant saving does not seem to justify the inclusion of another variable to account for the above situation. The stream termination heuristic as used in this study should be capable of obtaining near optimal solutions on problems where the above situation can occur.

Initial Matching of Streams Heuristic

Rudd et al. (1973) suggest matching at the first exchanger ($J = 1$) in the sequence either the coldest cold stream with the coldest hot stream or the hottest cold stream with the hottest hot stream. In the three problems considered, the optimum for the 2 × 2 case follows this heuristic, those for the 2 × 3 and the 3 × 3 cases do not, although the local optimum obtained in the 3 × 3 case does follow the heuristic. An attempt was made to see if this heuristic might improve the search.

The heuristic was applied by doubling the probability of obtaining a C_K that represented a coldest or a hottest cold stream. If one of these two streams were selected, then the probability of obtaining the corresponding hot stream was doubled.

This heuristic was applied along with the termination heuristic to all three example problems. The efficiency of the method is shown in Table 4. An improvement in efficiency was not expected for the 2 × 3 case since its solution does not follow the heuristic. In the 2 × 2 and the 3 × 3 cases, the heuristic decreased the possibility of certain intermediate arrangements being selected that would have facilitated the search. Consequently, the efficiencies were worse than with the termination heuristic alone. This initial matching heuristic is of value, however, as many heat exchange network problems in industry do follow this heuristic, and an improvement in efficiency might be obtained.



Q_1 = 1544.1 kJ/s A_1 = 71.5m²

Q_2 = 12.9 kJ/s A_2 = 0.1m²

Q_3 = 1631.7 kJ/s A_3 = 84.5m²

Q_4 = 1534.1 kJ/s A_4 = 23.7m²

	H_1	H_2	H_3	
C_1		4	2	T
C_2			3	T
C_3	1			T
	C	T	C	

Fig. 5. Local optimum for 3 × 3 example.

SUMMARY

A synthesis problem is essentially a mixed integer problem that requires simultaneous optimization of process variables and unit arrangement. The adaptive random search proved to be very effective in solving the three synthesis problems considered. The variation of heats transferred resulted in cost reduction in two of the three examples. In the 2 × 2 example, the added variable resulted in large savings in utility usage with only slightly high construction costs. Solution of the 3 × 3 example resulted in a reduction in the number of exchangers required.

The adaptive random search method requires only slightly more computational time than other methods and should be considerably simpler to apply and program. Very poor efficiency was obtained with the basic method for all three examples considered. Utilization of the stream termination heuristic increased the efficiency of the method to a level comparable with other published methods. Although the initial matching of streams heuristic failed to improve the efficiency on any of the examples, its use did not severely hinder the search and perhaps would aid the search in some cases.

NOTATION

a = heat exchanger cost parameter
A_{CK} = exchange area for steam heater on stream C_K (m^2)
A_{HL} = exchange area for water cooler on stream H_L (m^2)
A_J = exchange area for exchanger J (m^2)
b = heat exchanger cost parameter
C = symbol indicating stream requires cooling water
C_K = cold stream K ($K = 1, 2, 3, \ldots, M$)
H = symbol indicating stream requires steam heating
H_L = hot stream L ($L = 1, 2, 3, \ldots, N$)
I = random integer between the values I_A and I_B
I_A = lower bound of positive integer variable x_i
I_B = upper bound of positive integer variable x_i
J = integer assigned to each exchanger in the array ($J = 1, 2, 3, \ldots, MN$)
k = distribution coefficient (positive odd integer)
k_d = range reduction coefficient (positive integer)
M = total number of streams to be heated
N = total number of streams to be cooled
N_t = number of independent variables
Q_J = heat transfer in exchanger J (Joules/s)
R = random positive real number
R_i = search region for variable x_i about x_i^*
T = symbol indicating stream requires no utilities to meet process temperature requirements
U = utilities cost per year for the network (\$/yr)
x_i = new value of independent variable ($i = 1, N_t$)
x_i^* = value of variable x_i producing the best value for the objective function
Y = cost per year for the entire system (\$/yr)
δ = annual rate of return on the investment
θ = random number between zero and one

LITERATURE CITED

Campbell, J. R., and J. L. Gaddy, "Methodology for Simultaneous Optimization with Reliability: Nuclear PWR Example," *AIChE J.*, **22**, No. 6, 1050 (1976).
Gall, D. A., "A Practical Multifactor Optimization Criterion," in *Recent Advances in Optimization Techniques*, A. Lavi and T. P. Vogl, ed., Wiley, New York (1966).
Hendry, J. E., and R. R. Hughes, "Generating Separation Process Flowsheets," *Chem. Eng. Progr.*, **68**, 71 (1972).
Heuckroth, M. W., L. D. Gaines, and J. L. Gaddy, "An Examination of the Adaptive Random Search Technique," *AIChE J.*, **22**, No. 4, 744 (1976).
Keenan, J. H., and F. G. Keyes, *Thermodynamic Properties of Steam*, Wiley, New York (1936).
Kesler, M. G., and R. O. Parker, "Optimal Networks of Heat Exchange," *Chem. Eng. Progr. Symposium Ser. No. 92*, **65**, 111 (1969).
King, C. J., D. W. Gantz, and F. J. Barnes, "Systematic Evolutionary Process Synthesis," *Ind. Eng. Chem. Process Design Develop.*, **11**, 271 (1972).
Kobayashi, S., T. Umeda, and A. Ichikawa, "Synthesis of Optimal Heat Exchange Systems," *Chem. Eng. Sci.*, **26**, 1367 (1971).
Lee, K. F., A. H. Masso, and D. F. Rudd, "Branch and Bound Synthesis of Integrated Process Designs," *Ind. Eng. Chem. Fundamentals*, **9**, 48 (1970).
Luus, R., and T. H. I. Jaakola, "Optimization by Direct Search and Systematic Reduction of the Size of Search Region," *AIChE J.*, **19**, 760 (1973).
Masso, A. H., and D. F. Rudd, "The Synthesis of System Designs," *ibid.*, **15**, 10 (1969).
Menzies, M. A., and A. L. Johnson, "Synthesis of Optimal Energy Recovery Networks Using Discrete Methods," *Can. J. Chem. Eng.*, **50**, 290 (1972).
Pho, T. K., and L. Lapidus, "Synthesis of Optimal Heat Exchanger Networks by Tree Searching Algorithms," *AIChE J.*, **19**, 1182 (1973).
Rathore, R. N. S., and G. J. Powers, "A Forward Branching Scheme for the Synthesis of Energy Recovery Systems," *Ind. Eng. Chem. Process Design Develop.*, 14, 175 (1975).
Rathore, R. N. S., K. A. Van Wormer, and G. J. Powers, "Synthesis Strategies for Multicomponent Separation Systems with Energy Integration," *AIChE J.*, **20**, 491 (1974).
Rudd, D. F., G. J. Powers, and J. J. Siirola, *Process Synthesis*, Prentice-Hall, Englewood Cliffs, N.J. (1973).
Siirola, J. J., "Status of Heat Exchanger Network Synthesis," paper presented at 76th National AIChE Meeting, Tulsa, Okla. (Mar., 1974).

Manuscript received January 14, 1977; revision received July 19, and accepted August 11, 1977.

Effects of Temperature on the Crystallization of Potassium Nitrate by Direct Measurement of Supersaturation

JAMES E. HELT
and
MAURICE A. LARSON

Department of Chemical and Nuclear Engineering and Engineering Research Institute Iowa State University Ames, Iowa

A laboratory continuous mixed-suspension, mixed-product- removal crystallizer was used to study the effects of temperature on the kinetics of crystallization of potassium nitrate. A differential refractometer was used to continuously monitor the supersaturation permitting the measurement of supersaturation levels of order 10^{-3}Kg solute/Kg water. The nucleation rate exhibited an inverse relationship with temperature.

SCOPE

Attainment of the desired size distribution is one of the principal objectives in the design and operation of industrial crystallizers. The main factor which determines size distribution is the crystallization kinetics. If improvements are to be made in crystallizer design and operation, knowledge about the crystallization kinetics is needed.

Correspondence concerning this paper should be addressed to James E. Helt, North Carolina State University, Raleigh, North Carolina.