On the Ordering of Recycle Calculations

WOOYOUNG LEE and DALE F. RUDD

University of Wisconsin, Madison, Wisconsin

Algorithms are developed for organizing large recycle calculations. In particular, we discuss the problem of determining the minimum number of recycle parameters that must be assumed to render recycle calculations acyclic. This work has significance in the evolution of computer programs for the automatic design of process systems.

An accidental approach to process calculations cannot be tolerated during the evolution of computer-aided process design programs. The computer does not possess the intuitive abilities which enable the experienced human problem solver to recover from his early errors. An orderly approach to process calculations must be available to the computer in the form of simple algorithms. The development of such algorithms has been an area of research of late.

In this paper we discuss the problem of rendering complex recycle calculations acyclic by assuming the values of certain key recycle parameters. This concept is not novel; the experienced engineer frequently approaches the material or energy balances for a recycle process thus:

The values of all recycle parameters are assigned tentative values.

The remaining calculations, which contain no unspecified recycle parameters, are then performed step by step, yielding new values for the recycle parameters.

The first two steps are repeated until the desired convergence is achieved, often with the aid of a convergence accelerator.

It is well known that a number of combinations of variables in a given problem can assume the role of recycle parameters, and that the selection of these recycle parameters determines the difficulty of the iterative calculations.

The novel aspect in this work involves the development of simple procedures for determining the minimum number of recycle parameters in complex recycle processes. We are concerned with setting up difficult problems for easier solution.

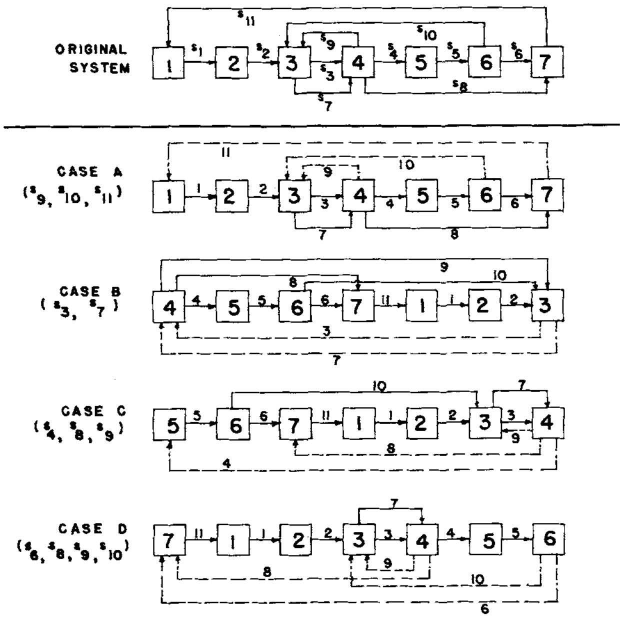


Fig. 1.

For example, the system shown in Figure 1 may be rendered acyclic in a number of ways by the specification of sets of recycle parameters. Case B may result in simpler calculations, since it requires that a minimum number of recycle parameters be assigned tentative values. While this area of study has received attention (*1*, *4*), no complete method of analysis had evolved for detecting the true recycle in a complex process.

MINIMUM NUMBER OF TEARS

Two cases are distinguished: recycle streams defined by a single variable and recycle streams defined by one or more variables. The analysis in this section is limited to systems in which the recycle streams are defined by a single variable. In a later section we modify the analysis to include multiple variable streams (for example, streams which carry information in terms of compositions, enthalpy, and mass flow rate). Throughout this paper we use the term *tearing* to refer to the assignment of a value to a recycle stream; such an assignment tears or breaks the recycle of information during computation (*2*).

Tracing Cycles

The analysis in this paper is preceded by the identification of the cycles in the system. Norman (*3*) has shown how to locate all cycles in a system; however, we need to know how the cycles are formed and how they are related to one another. Steward (*5*) has shown that the spanning tree technique gives this information but that combinatorial problems arise. As a starting point in this work we assume that some method is available to trace out all cycles.

Cycle Matrix

Once the cycles have been traced, a cycle matrix can be constructed. Let *A*. *B*. *C*. ... represent the cycles and S_j the streams. The cycle matrix **C** has the elements

$$\mathbf{C} = [c_{ij}] = \begin{cases} 1 \text{ if } S_j \text{ appears in cycle } i \\ 0 \text{ otherwise} \end{cases}$$

A *cycle rank* is defined as the number of streams involved in a cycle and equals the sum of the elements in a row of the cycle matrix. A *stream frequency* is the number of cycles in which a stream appears and equals the sum of column elements. A cycle matrix augmented to include these parameters is illustrated in the following for the system in Figure 2.

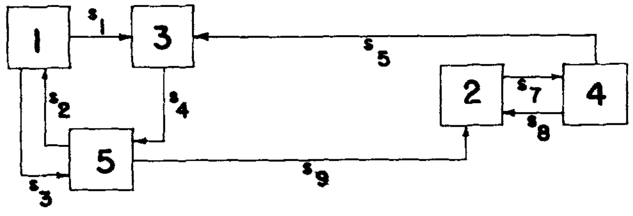


Fig. 2.

Cycles of the System

$A = S_2, S_3$
$B = S_7, S_8$
$C = S_1, S_2, S_4$
$D = S_4, S_6, S_7, S_5$

AUGMENTED CYCLE MATRIX OF THE SYSTEM

	S_1	S_2	S_3	S_4	S_5	S_6	S_7	S_8	Cycle rank
A		1	1						2
B							1	1	2
C	1	1		1					3
D				1	1	1	1		4
Stream frequency	1	2	1	2	1	1	2	1	

Any cycle initially of rank 1, a self-loop, can be eliminated only by tearing the stream involved therein. All self-loops can be eliminated from the cycle matrix, along with the attending stream columns, until all cycles have rank of at least 2. We now focus attention on problems in which the obvious recycle of self-loops has been eliminated.

Independent Column Sets

If the frequency of column j is equal to or greater than that of column k and if column j has nonzero elements in all of the rows where column k has nonzero elements, then column k is said to be *contained* in column j. In this case, selection of column j will imply that more cycles will be removed than if column k were selected, and all cycles removed by the selection of k will also be removed by the selection of j.

Therefore, if column k is contained in column j, column k can be removed from the cycle matrix without loss of generality. A set of columns is said to be *independent* when no column is contained in any other column. The removal of dependent columns reduced the size of the cycle matrix by a significant amount in the several practical problems analyzed by the authors.

Column Selection

Suppose that a cycle matrix with independent columns contains a row with only one nonzero element. The cycle corresponding to that row can be eliminated only if the column which contains that nonzero element is *selected* as a recycle variable.

Columns thus distinguished are eliminated from the cycle matrix along with the rows in which the nonzero elements of these columns appear. This procedure is continued until either all the rows have been eliminated or no rows remain with single nonzero elements.

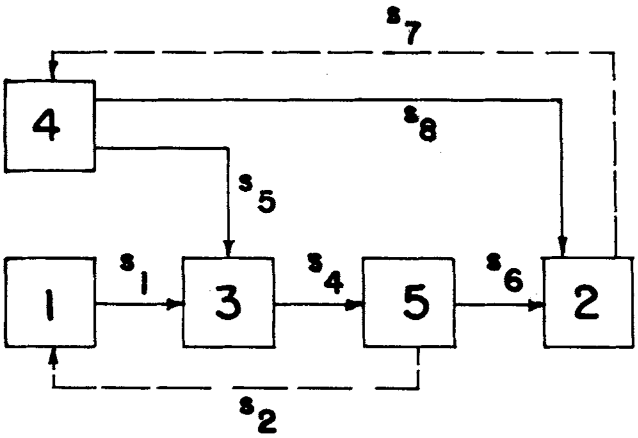


Fig. 3.

In the last matrix, notice that:

Columns S_1 and S_3 are contained in column S_2.
Columns S_5 and S_6 are contained in column S_4.
Column S_8 is contained in column S_7.

Thus, column elimination results in the following matrix:

	S_2	S_4	S_7	
A	1			(1)
B			1	(1)
C	1	1		2
D		1	1	2
	2	2	2	

Rows A and B each have only one nonzero element, and the corresponding columns are selected as the recycle streams. The precedence order of the system when the recycle streams are torn is given in Figure 3.

Should this procedure result in the elimination of all rows, the set of minimum number of recycle streams has been found. This systematic detection of cycles of unit rank is called *algorithm I*.

Further Selection of Columns

Should algorithm I not terminate, a reduced cycle matrix will result with the following properties: the cycle ranks are greater than one, and all of the columns are independent.

The problem of finding the minimum number of recycle streams now becomes the problem of finding one of the smallest sets of columns whose nonzero elements appear in every row of this matrix. In selecting potential recycle streams, it seems obvious that columns of high frequency are preferred to those of low frequency. In fact, the column with the highest frequency should be considered as the prospective recycle when one is concerned with reducing the size of a block by a single tear. Let us consider the original system in Figure 1 for which algorithm I does not terminate.

The cycle matrix of the system is

$\underline{C}$ =	S_1	S_2	S_3	S_4	S_5	S_6	S_7	S_8	S_9	S_{10}	S_{11}	Cycle rank
A			1						1			2
B							1		1			2
C			1	1	1					1		4
D				1	1		1			1		4
E	1	1	1	1	1	1					1	7
F	1	1		1	1	1	1				1	7
G	1	1	1					1			1	5
H	1	1					1	1			1	5
Column frequency	4	4	4	4	4	2	4	2	2	2	4	

By column elimination, we have the following reduced matrix.

	S_2	S_3	S_5	S_7	S_9	
A		1			1	2
B				1	1	2
C		1	1			2
D			1	1		2
E	1	1	1			3
F	1		1	1		3
G	1	1				2
H	1			1		2
	4	4	4	4	2	

An augmented column is now added to the right of the reduced cycle matrix as shown below. The elements in this column are the stream numbers of the zero elements of the rows. For example, the first element of the column is [S_2, S_5, S_7], since the first row in the cycle matrix has three zero elements in columns S_2, S_5, and S_7.

	S_2	S_3	S_5	S_7	S_9		Augmented Column
A		1			1	2	S_2 S_5 S_7
B				1	1	2	S_2 S_3 S_5
C		1	1			2	S_2 S_7 S_9
D			1	1		2	S_2 S_3 S_9
E	1	1	1			3	S_7 S_9
F	1		1	1		3	S_3 S_9
G	1	1				2	S_5 S_7 S_9
H	1			1		2	S_3 S_5 S_9
	4	4	4	4	2		

This first element indicates that cycle A will *not* be removed if the columns S_2, S_5, and S_7 are selected. It also implies that cycle A cannot be removed by the selection of any subset of this set. Notice that a number of subsets can be generated from this set. Should any of these subsets be selected, at least one more column must be selected to remove cycle A and to achieve an acyclic system. Notice that the set of three columns S_2, S_3, and S_7 removes all of the cycles. Hence, we are sure that three tears will be enough to reduce the recycle system to an acyclic system. However, is there any set of two recycle streams which will remove all of the cycles It is obvious that a set of two streams which can be generated by any element of the new column will not be able to remove at least one of the cycles.

If there is a set of two streams which cannot be generated by any element of the augmented column, such a set will remove all of the cycles of the matrix. Returning to our example, the set of columns S_3 and S_7 has been found to be the only set of two streams which could not be generated by the elements of the augmented column. Therefore, we have found that 2 is the minimum number of tears and the recycle streams are S_3 and S_7.

	S_2	S_3	S_5	S_7	S_9		Augmented Column	Sets of two streams generated by the Aug. Column
A		1			1	2	S_2 S_5 S_7	S_2S_5; S_2S_7; S_5S_7
B				1	1	2	S_2 S_3 S_5	S_2S_3; S_2S_5; S_3S_5
C		1	1			2	S_2 S_7 S_9	S_2S_7; S_2S_9; S_7S_9
D			1	1		2	S_2 S_3 S_9	S_2S_3; S_2S_9; S_3S_9
E	1	1	1			3	S_7 S_9	S_7S_9
F	1		1	1		3	S_3 S_9	S_3S_9
G	1	1				2	S_5 S_7 S_9	S_5S_7; S_5S_9; S_7S_9
H	1			1		2	S_3 S_5 S_9	S_3S_5; S_3S_9; S_5S_9
	4	4	4	4	2			

The only set of two streams which cannot be generated by the augmented column is: [S_3 , S_7]

Computer programs for the subset generation of a given set are readily available, and can be used to find the sets which cannot be generated by the elements of the augmented column. However, this involves some combinatorial difficulties, which are reduced in many cases by examining the frequency of the columns. A set of columns with a total frequency less than the number of the cycles of the matrix cannot remove all the cycles. For example, in the last matrix, the largest frequency is 4 and the smallest 2, and there are 8 cycles. Any set of two columns in which column S_9 is a member cannot remove all of the cycles, since the sum of frequency of this set of two columns will always be less than the number of the cycles. Therefore, while searching for sets of two columns which will remove all of the cycles, all sets of two columns which include column S_9 may be disregarded. Deleting column S_9 from the matrix, we obtain

	S_2	S_3	S_5	S_7	
A		1			(1)
B				1	(1)
C		1	1		2
D			1	1	2
E	1	1	1		3
F	1		1	1	3
G	1	1			2
H	1			1	2
	4	4	4	4	

Two rows of this matrix each now have only one nonzero element. The columns in which these nonzero elements appear correspond to the recycle streams. In general, the set (or one of the sets) containing the fewest streams which cannot be generated by the augmented column should be selected as the recycle streams. This systematic method of recycle stream selection is called *algorithm II*.

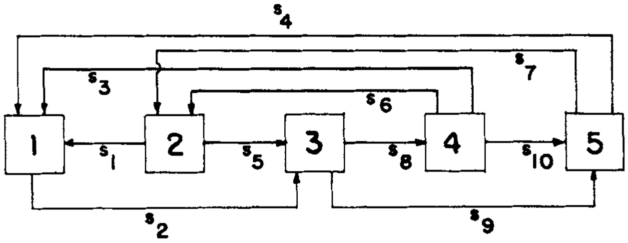


Fig. 4.

MINIMUM NUMBER OF RECYCLE PARAMETERS

In the previous section, procedures were developed for finding the minimum number of recycle streams in the case where the streams are defined by an identical number of parameters. In this section, procedures are developed for finding the minimum number of recycle parameters in the case where the streams are defined by an unequal number of parameters, such as temperature, flow rate, and composition.

Consider the example of Rubin (3) in Figure 4, in which the streams are defined by an unequal number of parameters. The number of parameters are:

Stream	No. of parameters
S_1	3
S_2	2
S_3	1
S_4	4
S_5	1
S_6	3
S_7	5
S_8	7
S_9	1
S_{10}	2

Cycle Matrix

As in the previous case, the cycle matrix of the system is constructed below. Instead of showing the frequency of columns, the variable numbers p_j are shown in the last row.

Column Elimination

Column k is said to be *strictly contained* in column j, if column k is contained in column j and if the number of variables of stream k, p_k, is not less than those of stream j, p_j. If this is the case, column k may be eliminated from the cycle matrix by the same argument which eliminated contained columns in algorithm I.

Cyclic Matrix: $\underline{C}$ =

	S_1	S_2	S_3	S_4	S_5	S_6	S_7	S_8	S_9	S_{10}	
A	1	1					1	1		1	5
B					1		1	1		1	4
C		1		1				1		1	4
D		1	1					1			3
E	1	1				1		1			4
F					1	1		1			3
G		1		1					1		3
H	1	1					1		1		4
I					1		1		1		3
p_j	3	2	1	4	1	3	5	7	1	2	

This elimination process is continued until no column of the matrix is strictly contained in any other column. For instance, S_1 of our example is strictly contained in S_2 and S_4 in S_2. Thus, the following matrix is obtained.

	S_2	S_3	S_5	S_6	S_7	S_8	S_9	S_{10}	
A	1				1	1		1	4
B			1		1	1		1	4
C	1					1		1	3
D	1	1				1			3
E	1			1		1			3
F			1	1		1			3
G	1						1		2
H	1				1		1		3
I			1		1		1		3
p_j	2	1	1	3	5	(7)	1	2	

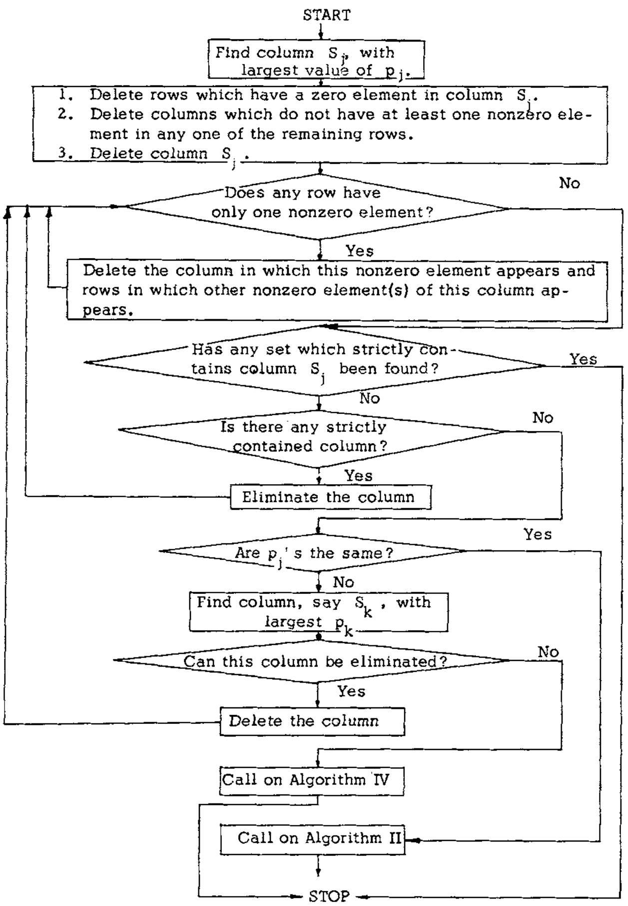
Whenever the rank of a cycle becomes 1, the column in which the single nonzero element appears corresponds to a recycle stream. This column is deleted from the matrix along with the rows in which nonzero elements of this column appear. If no cycle appears with rank 1, columns are eliminated in the following way.

If column k is contained in a set of columns and if the variable number of stream k is not less than the sum of the variable numbers of the columns in the set, we say that column k is *strictly contained in the set.* Hence, if there exists any set of columns which strictly contains column k, we may also eliminate column k from the matrix. We start eliminating columns with largest p_j.

Returning to our example, column S_8 has the largest value of p_j of the matrix $p_8 = 7$. It is readily seen that, for example, the set $[S_3, S_6, S_{10}]$ strictly contains column S_8, for this set contains column S_8 and the sum of p_j's of this set is 4, which is less than 7. Therefore, column S_8 is eliminated, and the following matrix results.

	S_2	S_3	S_5	S_6	S_7	S_9	S_{10}	
A	1				1		1	3
B			1		1		1	3
C	1						1	2
D	1	1						2
E	1			1				2
F			1	1				2
G	1					1		2
H	1				1	1		3
I			1		1	1		3
p_j	2	1	1	3	(5)	1	2	

The generation of cycles of rank 1 should be checked after each elimination. This method for finding a set of columns which strictly contains the column of the largest p_j is called *algorithm III*, and a flow diagram of algorithm III is given below.



Elimination of columns of high p_j's is continued until a cycle of rank 1 appears. The column in which the single nonzero element appears corresponds to a recycle stream. This column is deleted from the matrix along with all of the rows corresponding to other cycles which will be removed. Elimination of two more columns yields the solution of our example as follows. Elimination of column S_7 gives the following matrix.

	S_2	S_3	S_5	S_6	S_9	S_{10}	
A	1					1	2
B			1			1	2
C	1					1	2
D	1	1					2
E	1			1			2
F			1	1			2
G	1				1		2
H	1				1		2
I			1		1		2
p_j	2	1	1	(3)	1	2	

Note:

$[S_2 , S_5] >> S_7$

Strongly contained

Elimination of column S_6 gives

	S_2	S_3	S_5	S_9	S_{10}	
A	1				1	2
B			1		1	2
C	1				1	2
D	1	1				2
E	1					(1)
F			1			(1)
G	1			1		2
H	1			1		2
I			1	1		2
p_j	2	1	1	1	2	

Note:

$[S_2 , S_5] >> S_6$

If no more columns can be eliminated from the matrix and if every cycle has a rank larger than one, either one of the following situations will obtain: (i) p_j's for all j of the matrix are the same or (ii) p_j's are not the same. If case (i) occurs, algorithm II can be employed. In the case of (ii), the following procedure, *algorithm IV*, is required.

Algorithm IV

Consider the new matrix given below which has been modified for purposes of illustration so that algorithm III fails.

	S_2	S_3	S_5	S_6	S_9	S_{10}	
A	1					1	2
B			1			1	2
C	1					1	2
D	1	1					2
E	1			1			2
F			1	1			2
G	1				1		2
H	1				1		2
I			1		1		2
p_j	2	1	(2)	3	1	2	

The largest value of p_j's is 3 for column S_6. Column S_6, however, cannot be eliminated, since there does not exist any set of columns which strictly contains column S_6.

This is seen from the fact that there is only one set of columns which contains (not strictly contains) column S_6, namely, the set of columns S_2 and S_5, and that the sum of p_j's of this set is 4, which is larger than 3. In algorithm IV, column S_6 is divided into two pseudo columns as follows.

Either

	S_6'	S_6''
A		
B		
C		
D		
E	1	
F		1
G		
H		
I		
p_j	1	2

Or

	S_6'	S_6''
A		
B		
C		
D		
E	1	
F		1
G		
H		
I		
p_j	2	1

A column can be divided into a number of pseudo columns such that at least one of the pseudo columns is strictly contained in some columns of the matrix. For instance, in the second case of the above division of column S_6, pseudo column S_6' is strictly contained in column S_2 and hence pseudo column S_6' may be eliminated from the matrix to get the following matrix.

	S_2	S_3	S_5	S_6''	S_9	S_{10}	
A	1					1	2
B			1			1	2
C	1					1	2
D	1	1					2
E	1						(1)
F			1	1			2
G	1				1		2
H	1				1		2
I			1		1		2
p_j	2	1	2	1	1	2	

Since row E has only one nonzero element and it appears in column S_2, we select S_2 as a recycle stream and delete the corresponding column from the matrix along with rows A, C, D, E, G, and H. The resulting matrix will be

	S_5	S_6''	S_9	S_{10}	
B	1			1	2
F	1	1			2
I	1		1		2
p_j	2	1	1	2	

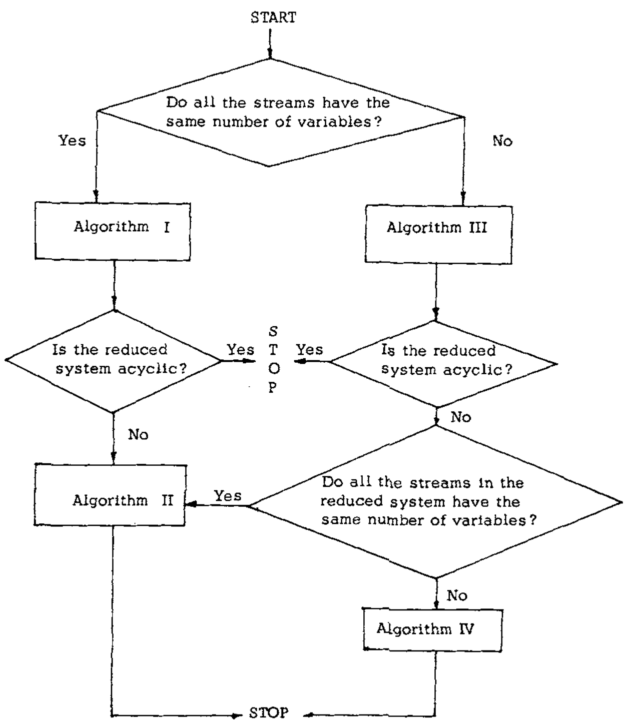
Column S_{10} is strictly contained in column S_5 and hence may be eliminated from the matrix. This makes row B have one nonzero element in column S_5. The selection of column S_5 removes all of the remaining cycles. Thus, the recycle streams which yield the optimal stage order with the minimum number of recycle parameters have been selected. Recycle streams for this case are S_2 and S_5 and the minimum number of recycle parameters is 4.

The worst case to which algorithm IV leads is a case where algorithm II can be used. That is, the values of all p_j's become the same and every cycle has rank greater than one.

CONCLUSION

We have presented some systematic procedures for finding the minimum number of recycle streams and for finding the minimum number of recycle parameters of a strongly connected system. Experience shows that these procedures enable the analysis, even by hand, of systems of fair complexity. For most of the cases considered the solutions have been straightforward without the need for algorithms II and IV. We summarize the application of these algorithms in the following diagram.

ESTIMATION OF THE MINIMUM NUMBER OF RECYCLE STREAMS



May we remark that this area is in its infancy and much more work is required before a complete strategy for attacking large problems is at hand. The computer will not have its full impact on process design until such a strategy evolves.

ACKNOWLEDGMENT

This study was supported in part by the National Science Foundation.

LITERATURE CITED

1. Cavett, R. H., paper presented at A.I.Ch.E. Pittsburgh meeting (May 19, 1964).
2. Lee, W., J. H. Christensen, and D. F. Rudd, *A.I.Ch.E. J.*, **12**, No. 6, 1104-1110 (1966).
3. Norman, R. L., *ibid.*, **11**, 450 (1965).
4. Rubin, D. I., *Chem. Eng. Progr. Symposium Ser.*, No. 37, **58**, 54 (1962).
5. Steward, D. V., *J. S.I.A.M.*, **2B**, No. 2, 345 (1965).

Manuscript received February 11, 1966; revision received May 26, 1966; paper accepted May 31, 1966.

Heat Transfer and Frost Formation Inside a Liquid Nitrogen-Cooled Tube

R. C. REID, P. L. T. BRIAN, and M. E. WEBER

Massachusetts Institute of Technology, Cambridge, Massachusetts

An experimental study was undertaken of simultaneous heat transfer and frost deposition inside a liquid nitrogen-cooled tube for a range of humidities and Reynolds numbers. The data indicate that diffusion of water vapor within the frost layer caused the frost density and thermal conductivity to increase with time. The increase was so great that the heat transfer rate became constant even while frost continued to accumulate. In a separate study the thermal conductivities of frosts formed on a cryogenic surface were measured and found to be lower than values near the freezing point of water.

Processes in which heat is transferred to a refrigerated surface with the simultaneous deposition of a frost layer are important in refrigeration, freeze-out purification of gases, cryopumping, and the storage of cryogenic liquids. Frost will form on a refrigerated surface that has a temperature below the dew point of the gas and the freezing point of the vapor. For small wall-gas temperature differences, water vapor reaches the wall by molecular and turbulent diffusional processes, and the standard correlations can be used to predict the heat transfer and frost accumulation rates (*1* to *13*). For large temperature differences, however, fog may form in the gas, and the impingement and trapping of fog particles on the surface may become the dominant mechanism of frost deposition. At the present time a rigorous analysis of this mechanism is not possible, although the conditions necessary for fog formation have been studied by Johnstone (*14*).

In natural convection to spherical and cylindrical liquid oxygen tanks (*15, 16*), the frost accumulation rate was only 10 to 30% of the predicted mass transfer rate because of the formation of a fog in the boundary layer flowing past the tank surface. In forced convection to cryogenically cooled cylinders, however, the frost accumulation rates were nearly equal to the predicted rates (*17, 18*). In both natural and forced convection frosting on cryogenic surfaces, the heat transfer rate decreased to a constant value when the air dew point was below 32°F. (liquid water could not condense on the frost surface).

M. E. Weber is with McGill University, Montreal, Canada.

The prediction of heat transfer rate with frosting on cryogenic surfaces is uncertain, not only because of the complex deposition mechanism, but also because of the scarcity of frost density and thermal conductivity data. The present experimental study was undertaken to determine the effect of a frost deposit on heat transfer inside a liquid nitrogen-cooled tube. Attention was focused upon the frost deposition rate and the effect of the frost layer on the heat transfer rate. The humidities studied were in the range where fog formation was important but where liquid water condensation could not occur. In addition, low-temperature frost thermal conductivity data were obtained for a range of frost densities typical of forced convection.

APPARATUS AND PROCEDURE

A cross section of the tubular heat exchanger is shown in Figure 1. A nitrogen-water vapor mixture flowed downward through the central tube (tube *D*), 0.62-in. I.D. and 44.5 diameters long. It was cooled externally by boiling liquid nitrogen. Frost formation occurred on the inside wall. Preceding the exchanger tube was a calming section of the same diameter and 80 diameters long.

The test heat exchanger in Figure 1 was constructed from four concentric copper tubes, the innermost tube being the exchanger tube (tube *D*). The space between tubes *A* and *B* was filled with liquid nitrogen to absorb the heat leak from the surroundings. The space between tubes *B* and *C* contained air. Horizontal plates separated tube *C* into six chambers, which contained the boiling nitrogen required to cool the walls of the exchanger tube. Gravity caused the flow of liquid nitrogen within the chambers, fresh liquid being fed