Simultaneous Optimization And Heat Integration With Process Simulator

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SIMULTANEOUS OPTIMIZATION AND HEAT INTEGRATION

WITH PROCESS SIMULATORS

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ABSTRACT

Strategies are presented for performing maximum heat integration of process streams simultaneously with flowsheet optimization in sequential process simulators. The strategies have been implemented on FLOWTRAN, using infeasible path optimization methods. Two formulations for performing the heat integration are compared in terms of reliability and computational efficiency. Results on complex process flowsheets show that this approach can produce substantial savings in raw material consumption through more efficient energy integration. Moreover, based on a simplified model, it is proved that these savings can always be achieved in recycle processes.

INTRODUCTION

Over the last five years important advances have been made in flowsheet optimization with process simulators. Effective computational strategies that are coupled with the successive quadratic programming algorithm [1,7,14] have been developed. These strategies include the feasible and infeasible path strategies for sequential modular simulators (Biegler and Hughes [2]), the simultaneous modular approach by Chen and Stadtherr [3], and the inside-out strategy by Jiraphongpan et al [9]. With these type of strategies the computational requirements for flowsheet optimization are typically equivalent to only 3 to 10 simulations of a process flowsheet. Therefore, it has now become computationally feasible to optimize chemical processes that are described by rigorous models.

Another important development that has taken place over the last decade is the development of synthesis techniques for the efficient heat integration of process streams to minimize the utility consumption (Hohmann, [8], Linnhoff et al. [10]). The application of these techniques can often produce substantial economic savings. However, an important limitation in the application of these techniques is that they assume fixed values of the flowrates and temperatures of the process streams. Therefore, heat integration techniques are usually applied once the flows and the process conditions have been established. Through this sequential procedure the interactions and the trade-offs with the overall optimization of the flowsheet are not properly accounted for.

Recently, Duran and Grossmann [4] proposed a strategy for the simultaneous optimization and heat integration of processes. The basic idea in this strategy is to include special constraints into the optimization problem to predict the minimum utility cost at any point in the flowsheet optimization path. With this scheme the flowrates and temperatures are treated as decision variables for both the optimization, as well as for the heat integration problem. This strategy has then the effect of anticipating *at the optimization stage* the economic effect of the utility cost for the heat integration. A limitation of this procedure is that the capital cost of the heat exchanger network is not accounted for. It should be noted though, that the utility cost has a much greater sensitivity in the optimization than the capital cost, as will be shown later in the numerical results.

Duran and Grossmann [4] illustrated the application of their strategy on a simplified process with a recycle which was modelled within an equation oriented framework. Their results showed that the simultaneous strategy can greatly increase the profit by reducing not only the energy consumption, but also more importantly, by increasing the overall conversion of the raw material.

It is the purpose of this paper to consider the problem of performing the simultaneous optimization and heat integration with sequential modular process simulators. Firstly, based on a simplified model for recycle processes it will be shown why the simultaneous strategy will lead to lower cost solutions that exhibit higher overall conversion for the raw material. Explicit and implicit modelling schemes will then be discussed for interfacing the heat integration problem with the flowsheet optimization. This will be illustrated with the FLOWTRAN process simulator in which several optimization strategies have been implemented by Lang and Biegler [11]. Finally, the application of this technique will be presented on two flowsheets for the production of ammonia and methanol. As will be shown, the results confirm the superiority of the simultaneous strategy over the sequential optimization and heat integration.

TRADE-OFFS IN OPTIMIZATION AND HEAT INTEGRATION

Examples in previous work by Duran and Grossmann [4] and Grossmann [6] have shown that simultaneous optimization and heat integration can lead to solutions that exhibit lower cost and higher overall conversion of the raw material. It is the purpose of this section to show that this property is in general true for recycle processes, and also to provide some insight into the nature of the economic trade-offs between the process optimization and the heat integration.

Consider the processing system shown in Fig. 1. This system consists of the following steps: (FP) Feed preparation (e.g. compression); (R1) Reaction (e.g. preheat, reaction, cooling); (S1) Recovery of liquid product and by-products (e.g. flash separation); (S2) Split for purge stream; (R2) Recycle (e.g. recompression); (PR) Recovery of final product (e.g. distillation). This processing scheme is representative of many chemical and petrochemical processes in which the feedstock contains some inerts, and the conversion per pass in the reactor is not very high.

Appendix A presents a simplified model for this processing scheme. Major assumptions include a single reaction, fixed pressure and temperature levels, near perfect separation in the liquid recovery step, and major heating and cooling requirements for the reaction step and for the recovery of the final product. Also, all the cost models are assumed to be given as linear functions of the flows in the process streams. For the sequential strategy it is assumed that nominal unit costs of utilities are given for heating and cooling requirements. For the simultaneous strategy it is assumed that unit costs for heating and cooling are lower to reflect the savings that are achieved by heat integration. Finally, since a fixed production rate is assumed, the objective function consists of the minimization of the total cost.

Based on the simplified model there are two major terms that can be identified in the cost function for the process scheme in Fig. 1 (see Appendix A). One is the net cost of the feedstock which is the cost of the raw material minus the income from the purge stream. The second term is the operating and capital cost of the process. The operating costs considered in this model are electricity costs for compression and recompression in the feed preparation and recycle steps, and heating and cooling costs for the reaction and final product recovery steps.

A representative plot of the two cost terms as a function of the overall conversion of the raw material is shown in Fig. 2. The specific functional relationships are given in Appendix A. As expected the curve for the net cost of the feedstock is convex and decreases monotonically with the overall conversion. On the other hand the curve for capital and operating expenses is convex, goes through a minimum, and tends to infinity for 100% overall conversion. Qualitatively the reason is that at low overall conversion the cost of feed preparation is high due to the large flow in the feed, while at high overall conversion the cost of the reaction and recycle is very high due to the large flow in the recycle loop. By considering nominal prices for heating and cooling, the optimal overall conversion $\bar{x}^{*}$ shown in Fig. 2 is obtained. If heat integration is performed based on the flows corresponding to the optimal overall conversion in Fig. 2, the total cost can be reduced from C^*_{SEQ}

If on the other hand heat integration is considered for determining the optimal overall conversion the effect is that the curve for capital and operating expenses will be lower as seen in Fig. 3 due to the savings in the utility costs (see Appendix A). This in turn has the effect of shifting the location of the optimal overall conversion towards a higher value, $\bar{x}'$, than the one of the sequential approach (see Fig. 3). Also, it can be clearly seen that the optimal cost C^*_{M} lies below the cost $C^{1}_{..}$ that was obtained in the sequential approach. Therefore, based on the simplified process flowsheet model that has been assumed, it is clear that due to anticipation of the efficient use of energy at the optimization stage, the simultaneous strategy will lead to designs, which, compared to the sequential approach, exhibit:

a) Higher overall conversion of the raw material.

b) Lower total cost.

Qualitatively the reason for this is that the simultaneous strategy is able to

establish the correct trade-off between the raw material, capital and energy costs. A rigorous proof of this property is shown in Appendix A by making use of the analytical expressions that were developed for the cost models.

An important assumption in the above analysis is that operating conditions such as pressures and temperatures have been assumed to be constant. However, very often some of these variables will be degrees of freedom for the optimization. This implies that since fixed pressures and temperatures are considered for the heat integration of the process streams, the final cost C^2_{SEQ} in the sequential approach will typically lie above C^1_{SEQ}. and hence will have an even greater difference with C^*_{SIM}. Therefore, as will be shown later with the examples, the differences between the simultaneous and sequential procedures will often be substantial.

Another point of interest is the fact that it is quite possible that the capital and operating cost in the simultaneous strategy will be greater than the one in the sequential approach. For example, in Fig. 3, if the sequential cost is C^1_{SEQ}, then its operating and capital cost given by point A lies below point B which corresponds to the operating and capital cost for the simultaneous strategy. If on the other hand the cost C^2_{SEQ} is obtained, its operating and capital cost given by point C lies above point B. Therefore, depending on the nature of each problem the capital and operating costs might be higher or lower in the simultaneous approach. Note that in all these cases however, the net cost of the feedstock and the total cost in the simultaneous strategy are lower.

In summary, this section has shown that in recycle processes the sequential procedure will always lead to higher cost solutions because it distorts the trade-off between the net cost of the feedstock, and the capital and operating costs. In the simultaneous strategy, since the proper economic trade-off is established, solutions with lower consumption of the raw material, lower total cost, and either higher or lower capital and operating expenses are obtained.

PROBLEM FORMULATION

In order to consider the simultaneous optimization and heat integration of a process it is assumed that this problem will be posed in the following form:

Given a process flowsheet to be optimized, the specified streams for which heat integration is intended are given by a set of n_H hot process streams $i \in H$, which are to be cooled, and a set of n_C cold process streams $j \in C$, which are to be heated. The objective is to determine an optimal process flowsheet that features minimum utility consumption (cost) for these sets of streams at a specified temperature approach ΔT_m. The flowrates and inlet and outlet temperatures of the hot streams (F_i , T_i^{in} , T_i^{out} : $i \in H$) and cold process streams (f_j , t_j^{in} , t_j^{out} : $j \in C$), must be determined optimally in the feasible space for process optimization and heat integration, given that a set of n_{HU} hot utilities $i \in HU$, and a set of n_{CU} cold utilities $j \in CU$ are available for supplying the heating and cooling requirements.

For the sake of simplicity in the presentation, it will be assumed that enthalpies are treated through equivalent heat capacities (C_i : $i \in H$) and (c_j : $j \in C$), of the hot and cold process streams, and that these streams exhibit a finite difference between inlet and outlet temperatures. Also fixed inlet temperature levels (T_H^i : $i \in HU$) and (t_c^j : $j \in CU$), are assumed for the hot and cold utilities.

The model for the optimization or synthesis of a chemical process without heat integration among process streams is assumed to be given in the form

$$\min \phi = F(x,y) + \sum_{i \in HU} c_H^i Q_H^i + \sum_{j \in CU} c_C^j Q_C^j$$

$$\text{s.t. } h(x,y) = 0$$

$$g(x,y) \le 0 \qquad (P_0)$$

$$Q_H^i = r_i^H(y) \quad , \quad i \in HU$$

$$Q_C^j = r_j^C(y) \quad , \quad j \in CU$$

$$Q_H^i \ , \ Q_C^j \in R_+^1 \ : \ i \in HU \ , \ j \in CU$$

$$x \in X \subset R^n \ , \ y \in Y \subset R^m$$

The vector of variables x represents process parameters such as pressures, temperatures, flowrates or equipment sizes; the vector of variables $y = [\ F_i\ ,\ T_i^{in}\ ,\ T_i^{out}$: all $i \in H$; $f_j\ ,\ t_j^{in}\ ,\ t_j^{out}$: all $j \in C\ ]$, represents the flowrates and temperatures of the process streams that are to undergo either cooling or else heating. The variables in x and y belong to the respective sets X and Y, which are typically given by known lower and upper bounds (e.g. physical constraints, specifications). The vectors of constraints h, g, represent material and energy balances, or design specifications. In a non-integrated process flowsheet, all of the heating and cooling is supplied with utilities that have been pre-assigned to process streams so as to ensure feasible heat exchange. The equations in P_0 involving the expressions $r_i^H(y)$: $i \in HU$ and $r_j^C(y)$: $j \in CU$, represent the specific heat balances for calculating the heating and cooling utility requirements, Q_H^i: $i \in HU$ and Q_C^j: $j \in CU$, for the non-integrated flowsheet.

The objective function ϕ is in general economic in nature involving both investment and operating costs in the term F(x,y) ; the other terms correspond to the utility costs with c_H^i : $i \in HU$ and c_C^j : $j \in CU$, representing unit costs for the respective heating and cooling utilities.

For the case of an optimal design of a given process flowsheet, problem P_0 corresponds to a nonlinear program as then only continuous variables are involved in the vectors x and y. As shown by Duran and Grossmann [4], in the simultaneous optimization and heat integration the heat balance equations for a non-integrated flowsheet can be replaced by a set of constraints that will ensure that the process streams are heat integrated in the optimized flowsheet so as to feature the minimum utility target.

The basic idea in the heat integration constraints is to postulate each inlet temperature of the process streams as a candidate for a pinch point. The minimum heating is determined by the largest heating deficits of these streams through a system of inequalities. The minimum cooling is determined by heat balance. For the case of multiple utilities, the inlet temperatures of intermediate temperatures are postulated as additional candidates for the pinch points through the inequalities. It should be noted that this model will then treat the flowrates and temperatures of the streams as decision variables for both the optimization and the heat integration. Also, by using this representation there is no need to define temperature intervals, which of course can vary during the optimization.

For the case of single heating and cooling utilities an optimal integrated process flowsheet featuring minimum heating (Cl) and minimum cooling (Q_r) requirements can then be determined by solving the following nonlinear program (see Duran and Grossmann [4]:

$$\min \quad \wedge = F(x,y) \bullet C_H Q_H \bullet c_c Q_c$$

$$\text{s.t.} \quad h(x,y) = 0$$

$$g(x,y) \pounds 0 \qquad\qquad (P,)$$

$$z^*(y) - Q_M * 0 \quad \} \quad \text{all } p \text{€} P$$

$$O(y) + Q_H - Q_c = 0$$

$$Q_H \pounds 0 \quad , \quad Q_c * 0$$

$x \in X \; . \; y \in Y$

where the total excess heat, $\Omega(y)$, and the heating deficits, $z_H^p(y)$, for the pinch candidates $p \in P$, are given respectively by the explicit expressions

$$\Omega(y)=\sum_{i \in H} F \qquad) - \sum_{j \in C} f_j c_j (t_j^{out} - t_j^{in}) \qquad (1)$$

$$z_H^p(y) = \sum f_j c_j \, [\max\{0, t_j^{out} - (T^p - \Delta T_m)\} - \max\{0, t_j^{in} - (T^p - \Delta T_m)\}]$$

$$- \sum_{i \in H} F_i C_i \, [\max\{0, t_i^{in} - T^p\} - \max\{0, t_i^{out} - T^p\}] \qquad (2)$$

where the set of pinch point candidates is given by

$P = \{p \mid T^p = T_i^{in} : \text{all } p=i \in H \; ; \; T^p = (t_j^{in} + \Delta T) : \text{all } p=j \in C\}$

Problem P_1 allows then the simultaneous optimization and heat integration of chemical processing systems. The solution to this problem will be a process featuring the optimal minimum utility consumption (Q_H , Q_C).

Note that incorporation of the minimum utility target into the process flowsheet optimization framework does not introduce any additional variables for the definition of problem P_1. It only introduces the $[\, n_H + n_C + 1 \,]$ constraints for heat integration in place of the heat balances in P_0 to determine the utility requirements for the non-integrated process. However, the price one has to pay is that one has to deal with the structural nondifferentiabiiities inherent to the max{.} functions involved in the expression (2) that defines the heating deficit functions $z^p(y)$: all $p \in P$. These nondifferentiabiiities are potentially many and arise for instance when $T_i^{in} = T^p$ or when $t_j^{out} = (T^p - \Delta T_m)$. As shown in Duran and Grossmann [4] these nondifferentiabiiities can be treated through a smooth approximation procedure with which P_1 can be solved with standard nonlinear programming algorithms.

The formulation P_1 can be extended to the case when several hot and cold utilities are available, e.g. fuel, hot gases, steam at various pressures, cooling water, refrigerants. For the case of multiple utilities, a selection among them has to be made for their feasible and economic use to satisfy the utility demands. Since in this case pinch points may also arise due to the presence of utilities whose inlet temperatures fall within the temperature range of the process streams, additional deficit constraints must be included in the model to ensure feasible heat exchange for these "intermediate" utilities whenever they are selected.

The heat loads of the different hot and cold utilities are represented by the variables $u = \{ Q^i_H : i \in HU,\ Q^j_C : j \in CU \}$, where HU and CU are the respective hot and cold utilities index sets. For given sets of utilities, the hot utility $h \in HU$ with highest inlet temperature, and the cold utility $c \in CU$ with lowest inlet temperature, can be identified such that they bound the entire feasible range of temperatures for heat integration. The remaining utilities can potentially lead to pinch points. These sets of "intermediate" hot and cold utilities will be denoted by the sets $HU' = HU \backslash \{h\}$ and $CU' = CU \backslash \{c\}$, respectively.

Incorporation of the utility target in the presence of multiple utilities yields the following nonlinear programming problem (see Duran and Grossmann [4]h

$$min <j> = f(x.y) + \wedge_{i(HU} c^*_H O\wedge + \sum_{jfCU} c^j_c Q^j_c$$

s.t. h(x,y) = 0

g(x,y) £ 0 *(P₂)*

$z^p_H(y,u) - Q^*_H$ £ 0 } *all p∈P'*

CXy.u) + *Cf* - *(f_* = 0

u = *[Q*$_H$: */6//6/* . Qj.$_C$: *jGCU)* € R^{m}

x 6 Y , y G Y

where CXy.u) and z^*_H(y,u) : all p€P' are given by.

$$\Omega(y,u) = \sum_{i\in H} F_i C_i (T^{in}_i - T^{out}_i) - \sum_{j\in C} f_j c_j (t^{out}_j - t^{in}_j) + \sum_{ifHU'} Q^i_H - \sum_{jfCU'} 0>_c \quad (4)$$

$$z^p_H(y,u) = QSIA(y,u)^p - QSOA(y,u)^p \quad (5)$$

$$QS0>4(y,u)^p = \sum_{i\in H} >F.C.tmaxiO.f^n_i - W\text{-}maxiO.T^{0}"_i - 7^*\}] \quad (6)$$

$$+ \sum_{i\in HU'} Q^i_H [max\{0, T^i_H - T^p\} - maxiO.f_M -1-7^*\}]$$

$$QSIA(y,u)^p = \sum_{j\in C} /.ct/iMxiO.^*?^{ut}_{l\,i\,t} - (^pT\text{-}Ar_m)\} max\{0 \wedge^{in}_i - (7^*\text{-}Ar_m)\}] \quad (7)$$

$$+ \sum_{j\in CU'} Q^j_C [max.\{0, T^j_C + 1 - (T^p - \Delta T_m)\} - max\{0, T^j_C - (T^p - \Delta T_m)\}]$$

and where the candidate pinch temperatures are given by

P' = { p | $T^p=T^{in}_i$: p=iGH, r«T»$_M$: p=iGHU', $T^p=(t|^n_j$ ^AT$_m$) : p^jGC, $T^P=(TJ._C$ -AT$_m$) : P=j€CU' } (8)

An optimal solution to problem P_2 will then determine an economic process flowsheet that will feature optimal heat integration in the presence of multiple utilities. Note that, as in the case of problem P^ structural nondifferentiabilities arise in problem P_2 due to the max functions in the expressions that define the heating deficits. These nondifferentiabilities can also be treated through a smooth approximation procedure.

INCORPORATION INTO PROCESS **SIMULATORS**

Problem P_1, and the more general problem P_2 for multiple utilities, correspond to explicit formulations for equation oriented simulators. For sequential modular process simulators the only difference is that the set of nonlinear process equations are given in implicit form. Therefore, any sequential modular process simulator with optimization capability can handle problems P, and P?.

In this paper the FLOWTRAN process simulator was used for performing the simultaneous optimization and heat integration. The optimization capability in FLOWTRAN was installed by writing a type 2 (convergence) block. The structure and argument list for this block, called SCOPT, is the same as the existing recycle convergence block, SCVW. Because no code was changed in FLOWTRAN, the direct loop perturbation strategy was implemented as the most straightforward way for evaluating gradients. Since FLOWTRAN generates and compiles a FORTRAN main program at run time, it can easily accommodate in-line FORTRAN and user written subroutines as part of the input data. This, in turn, allows the optimizer to evaluate partial flowsheet passes if needed for gradient evaluation. Also, user specification of the optimization problem can be made simply by adding a few lines of in-line FORTRAN to the input data for the simulation problem.

Due to the structure of the algorithms and in-line FORTRAN capabilities, the optimization implementation allows the following solution strategies:

- "black-box" optimization

- simultaneous convergence of recycle streams and design constraints using either Broyden or Newton methods.
- infeasible path optimization (IP)
- complete feasible variant optimization (CFV)
- partially converged flowsheet optimization with an embedded Broyden algorithm (EBOPT)

All of these use Successive Quadratic Programming (SQP) as the "driver" for optimization and convergence of equality constraints. Depending on the nature of the flowsheet optimization problem, different strategies could be applied for more efficient performance.

The formulation for simultaneous optimization and heat integration can be used directly by the optimization implementation on FLOWTRAN. To take full advantage of this approach, however, the inequalities that describe the heat flows need to be consistent with the physical properties used in the process simulation. Consequently, the equations use enthalpies from FLOWTRAN directly, although pinch points are still restricted to inlet process stream temperatures. The evaluation of the heat integration was implemented by writing new heat exchanger cost blocks in FLOWTRAN. At the end of the flowsheet calculation sequence, these provide calculation of the F_iC_i, f_jc_j, terms in formulations P_1 and P_2. However, since the optimization problem has more complex expressions and has a different structure than the one considered in Duran and Grossmann [4], it is instructive to analyze the simultaneous formulation for sequential modular simulators.

IMPLICIT VS. EXPLICIT FORMULATIONS

Problem P_2 is expressed in terms of process equations and inequality constraints for all of the pinch candidates (inlet temperatures of streams). With sequential modular simulators, however, a reduced set of nonlinear tear equations results in P_2. The *explicit* treatment of heat integration inequalities introduces potentially large number of nonlinear constraints in the flowsheet optimization problem (number of process streams • number of intermediate utilities • 1). Thus, the quadratic program that is solved at each iteration for this formulation may be more prone to less efficient performance. An alternate *implicit* formulation that eliminates the pinch candidate constraints can be derived by noting the following points:

- At a given SQP iteration with fixed flows and temperatures, problem $?_2$ becomes a linear program in terms of the heat loads of the multiple heating and cooling utilities (see Duran and Grossmann, [4]).

- Assuming unrestricted utility heat loads, the above linear programming model can be solved through a simple recursive algorithm as will be shown below. Hence, this problem can be solved implicitly within a "black box" in the process simulator.

However, nondifferentiabilities may arise when the pinch point transfers from inlet temperature to another. In applying this implicit formulation it is assumed that the temperatures of the inlet streams are sufficiently far away from each other so that the derivatives exist locally and can be evaluated at each iteration. For many process problems pinch points are determined by a single stream and therefore this will often not be a poor assumption. We also see that this implicit formulation extends naturally to include multiple utilities under rather mild assumptions.

IMPLICIT FORMULATION - SINGLE UTILITY CASE

Problem P_y deals only with unrestricted matches and is formulated so that nondifferentiable functions are expressed explicitly through active inequality constraints. As shown by Duran and Grossmann [4], for fixed flows and temperatures a straightforward formulation can be derived by setting

$Q_{..}$=max {<}

directly, and obtaining Q_c from the total heat balance. A simple algorithm for this case can be stated as follows:

1. At a given SQP iteration with flows and temperatures specified, calculate the heat capacity flowrates from the process simulator using:

$$f_j c_j = (h(t_j^{out}) - h(t_j^{in}))/(t_j^{out} - t_j^{in})$$

$$F_i C_i = (H(T_i^{out}) - H(T_i^{in}))/(T_i^{out} - T_i^{in})$$

where H(T) and h(T) are enthalpies of hot and cold streams, respectively, at temperature T.

2. Define the set P in equation (3) by selecting the following temperatures as pinch candidates:

- inlet temperatures of hot process streams
- inlet temperatures of cold process streams plus ΔT_m

3. Using equations (1) and (2) calculate Q_H and Q_c from

$$Q_H = \max_{p \in P} \{z_H^p\}$$

$$Q_c = \Omega(y) + Q_H \qquad (9)$$

These two quantities give the minimium utilities for the flows and temperatures at the current iteration. Equation (9) is equivalent but not identical to the problem table formulation of Linnhoff et al. [10]. Also, note that in step 1 the flowsheet need not be converged at a given SQP iteration. In fact, the implementation in FLOWTRAN treats this step no differently than standard utility calculations within its cost blocks. Also the above algorithm allows for the existence of multiple pinch points since the max operation over the pinch candidates p does not necessarily lead to a unique solution. This point becomes important in the next section when dealing with multiple utilities.

IMPLICIT FORMULATION - MULTIPLE UTILITIES

The solution of the linear programming problem for multiple utilities that arises in P_2 for fixed flows and temperatures, can be avoided by extending the above simple algorithm to the multiple utility case. For this, it is important to note the following points:

1. For unrestricted matches, it can easily be shown through T-Q curves that the total minimum utility requirement is the same for the single or multiple utility case.

2. Generally, the cost of hot (cold) utilities is monotonically nondecreasing (nonincreasing) with increasing temperature.

One can treat the multiple utility case therefore as a staged set of single utility calculations with the intention of introducing intermediate utilities at intermediate points on the T-Q curves. To do this consider the T-Q curves in Figure 4. An intermediate heating utility is available at T^ while an intermediate cooling utility is available at V_Q. Using points 1 and 2 it is advantageous to displace some of the hot (cold) utility at the highest (lowest) level with an intermediate utility. Substitution with an intermediate utility can be done graphically by considering only those parts of the T-Q curves above (below) the heating (cooling) utility temperature T^ (T^). For example, in Figure 5 addition of the intermediate hot utility at T^ amounts to shifting the hot stream curve above T^ until the hot and cold streams form another pinch point (at or above T^) or the hot utility at the highest level is entirely displaced by the intermediate utility.

It is easy to see that this procedure is equivalent to the minimum utility calculation using max(z£) if one only includes those streams above (below) the intermediate heating (cooling) utility temperature. Because of this equivalence one can expand the algorithm for the single utility case to multiple utilities as follows.

1. At a given SQP iteration with flows and temperatures specified, calculate the heat capacity flowrates from the process simulator using:

$$f_j c_j = (h(tp- MtpMtp - t_j^{in})$$

$F_iC_i = (H(Tp- HtTp\^r_i^{1*} - T_i^{in})$

2. Select the following temperatures as pinch candidates (p6P°):

- inlet temperatures of hot process streams
- inlet temperatures of cold process streams plus AT_m

3. Set k = 0. Define H_Q and C_Q as the set of hot and cold streams for heat integration.

4. Using equations (1) and (2) for the index set P° = {j 6 C_Q

and i € H_Q} calculate z_H^* and O(y) to determine

$G_h^\circ = \max_{p \in P^O} lz_H J$

$Q_c^\circ = CXy) + 0_n^\circ$

These are the overall minimum utilities at the current iteration. If no intermediate utilities are available, stop. Else, go to step 5.

5. Set k = k * 1 and let T^p be the lowest current pinch point. Let

$T^* = \max_{jtcu''} \{P_c\}$

$\hat{y} = ar\^\max_{j \in Cu''} \{T^j_Q\}$

where CU^{11} = {l | l G CiJ and T[£ T - $A7M_m$*

If none is available, set k = 0 and go to step 8. Else, define H_k and C_k as the set of inlet stream temperatures at or below (T^k • AT_m) and T^k, respectively. Also define pinch point candidates as:

P^k = fi 6 H_k, j € C_k, k}

6. Using p 6 P^k calculate:

$cf_c = \max_{p \in P^k} iz_H P)$

$Q_c^k = Q_c^{k-1} - Q_c^{\hat{j}}$

Note that if $Q_c^{\hat{j}}$ > 0, a new pinch point has been created at or below (T^k • AT_m).

7. If $Q^*_c = 0$, set $k = 0$ and go to step 8. Else return to step 5.

8. Set $k = k + 1$. Let T^p be the highest current pinch point and let:

$$7^* = \min_{if\ HU''} \{f_H\}$$

$$\hat{I} = arg \min_{if\ HU''} \{P_H\}$$

where HU" = {/ | i £ Hi/ and f_H Z f)

If none is available, stop. Else, define H^k and C^k as the set of inlet stream temperatures at or above T^k and T^k - AT , respectively. Also, define

$$P^k = \{j \in C_k,\ i \in H_k,\ k\}$$

9. Using $p \in P^k$ calculate:

$$Q^k_h = \max_{p \in P^k} \{z^p_H\}$$

$$Q^{\hat{i}}_H = Q^{k-1}_h - Q^k_h$$

Note that if $Q^{\hat{}} > 0$ a new pinch point has been created at or above T^k.

10. If $Q^k_h = 0$, stop. Else, return to step 8.

Note that this algorithm is recursive in nature and fairly straightforward to implement. In steps 6 and 9, we see the effect of intermediate utilities displacing more expensive utilities and thus reducing the utility cost. Also, care is taken to introduce hot (cold) utilities above (below) the highest (lowest) pinch as they are unnecessary elsewhere. A small example that illustrates the algorithm is given in Appendix B.

COMPARISON OF IMPLICIT VS. EXPLICIT STRATEGIES

For fixed flows and temperatures both the implicit and explicit strategies produce identical results, and these are also equivalent to the unrestricted, multiple utility formulations described in Linnhoff et al [10] and Papoulias and Grossmann [13]. The main differences between the implicit and explicit strategies lie in their treatment of nondifferentiabilities that occur as a result of the max operator. As shown in Appendix C, the minimum utility costs calculated by the implicit formulation are nondifferentiable functions if, during the course of the optimization, different stream inlets define the pinch point. This problem is handled automatically in the explicit formulation simply because a different active set of the constraints given in P_2 is chosen. On a small problem optimized with an equation-oriented procedure, Duran and Grossmann [4] demonstrated the effectiveness of the explicit formulation. Other recent work on MINLP synthesis problems by Kocis and Grossmann has confirmed this observation. It should be noted though that the computational requirements are typically 50-100% higher when compared to the case when no heat integration constraints are included.

The above studies with the explicit treatment of heat integration constraints were carried out using the MINOS algorithm for nonlinear programming. However, in the example problems considered in the next section we noted (particularly, for Case 2 of the Ammonia synthesis problem) that the implicit formulation required far fewer iterations than the explicit formulation. Therefore, with sequential modular simulators and the SQP algorithm, it is instructive to examine why the implicit and explicit formulations can perform differently.

Note that the implicit algorithm generally deals with a problem where all decision variables appear nonlinearly. Here all of the hot and cold utilities are calculated directly as dependent variables. In the explicit formulation (P_2), on the other hand, variables for the utilities (Q^J_H, Q^j_c), become part of the optimization problem and appear linearly in the objective function and constraints. For this larger problem, these variables do not contribute to the second derivatives of the Lagrange function and the Hessian matrix for this function is indefinite. However, the SQP algorithm approximates this matrix incorrectly by using a *positive definite* updating formula. While this insures that the resulting QP's are solvable at each iteration, it is possible that the rate of convergence for SQP can deteriorate. Powell [14] shows that, for SQP methods where the update formulae do not approximate the true Hessian at the solution, the rates of convergence are less than superlinear. A further example of this slow convergence is given by Yuan [16].

To improve on the convergence rate for the explicit formulation it would be worthwhile to update only the part of the Hessian matrix that deals with the nonlinear variables, w. Thus, the Hessian matrix used in the SQP algorithm would have the following structure:

$$\begin{pmatrix} B & A \\ A^T & O \end{pmatrix}$$

where $A = \nabla_{w,\bar{\Omega}} L(x,y)$ is easily calculated from gradient information and B is a positive definite approximation to $\nabla_{w,w} L(x,y)$. Because this matrix retains the structure and is a closer approximation to the actual Hessian matrix, it is likely that the performance of the SQP algorithm would greatly improve.

However, the main drawback to using the above matrix lies in the ability of the QP solver to handle an indefinite matrix at each iteration. Consequently, we leave this concept as a topic for future research. It is also interesting to note that this problem only occurs with the SQP algorithm. For example, because MINOS works in the null space of active constraints and uses projected Hessian updates, there is no deterioration of convergence properties, and consequently this problem was not encountered in earlier studies [4].

With the implicit formulation, the inequality constraints are satisfied directly by the minimum utility algorithm given earlier in the paper; in the optimization problem, the constraints are eliminated and are replaced by the actual utility cost in the objective function. Here the structure of the problem is no different from flowsheet optimization without heat integration and, as mentioned above, the only drawback to this formulation is the possible existence of nondifferentiable points in the objective function (see Appendix C).

EXAMPLE PROBLEMS

In order to demonstrate the effectiveness of the simultaneous approach for process optimization and also to compare the implicit and explicit formulations, we consider two comprehensive flowsheeting problems. Both problems involve multiple utilities and have the structure of the recycle process discussed in Appendix A. The first process, ammonia synthesis, has a high overall conversion and offers possibilities for heat integration with the reactor. The second process, methanol synthesis, has a lower conversion per pass and, consequently, requires a high recycle ratio. Here possibilities exist for heat integration with the distillation columns.

Ammonia Synthesis

The ammonia synthesis process flowsheet is presented in Figure 5. This process is a single loop design with a three-stage adiabatic flash separation. The reactor is modelled as a pseudo-homogenous plug flow reactor with rate expressions taken from the FLOWTRAN manual (Seader et al [15]). The associated cost block for the reactor also stems from the FLOWTRAN manual. A description of the optimization problem is also given in Figure 5. Here the objective function is the before tax profit over a five year life with a 15% rate of return. A number of design constraints are placed on the process to insure feasible operation and product purity. Note that instead of fixing the process feed, a constraint is placed on ammonia production and the feed streams are chosen as decision variables. The minimum temperature approach was set to 20 °F. The following two cases were considered for this problem.

Case 1. Eight decision variables specified in Table 1 were allowed to vary; the outlet pressure of the main compressor was fixed at 2159 psig.

Case 2. Nine decision variables including the main compressor outlet pressure are allowed to vary.

Both of these cases were used to illustrate the difference between the simultaneous and sequential approaches. The optimizations were run using the implicit formulation discussed above and the infeasible path optimization option implemented on FLOWTRAN.

Case 1 Solution

The optimal decision variable vector of the simultaneous and sequential solutions are given in Table 1. The results of this optimization without accounting for heat integration show a before tax profit of $24,945 x 10^6/yr. After applying heat integration to this solution, a profit of $26,915 x 10^6/yr is realized as a result of further utility savings. The simultaneous heat integration and optimization solution has a before tax profit of $27,134 x 10^6/yr or a net gain of $219,584/yr. A detailed comparision of sales and expenditures is given in Table 2. Note that the simultaneous solution has a 1.57% higher overall conversion of H_2 to NH_3 than the sequential solution and this accounts for almost a $600,000/yr reduction in raw material cost for H_2. Also, it is interesting to note that the simultaneous solution has larger capital and utility costs than the sequential solution but these are more than compensated by the more efficient use of raw materials. Finally, to compare the effect of heat integration for these two solutions, the T-Q curves for each solution are presented in Figure 6.

Case 2 Solution

Here outlet pressure for the main compressor was included as a decision variable. The simultaneous and sequential solutions for this case are given in Tables 3 and 4. The optimal decision variable vector in Table 3 shows a much lower compressor pressure for the simultaneous solution. As seen in Table 4 this has a benefical effect on both the utility and capital costs. The sequential solution is largely unchanged from Case 1. This indicates that without accounting for the possibilities of heat integration, the objective function is not very sensitive to the main compressor pressure. Consequently the advantages that a lower pressure offers at the heat integration stage are not exploited. For this case, one again observes a higher overall conversion of H_2 (94.75% vs. 93.3%). In addition, as a result of lower capital costs ($26,273/yr lower), lower utility costs ($330,708/yr lower) and lower raw material costs ($519,117/yr lower) the simultaneous solution has a greater before-tax profit of over $880,000/yr. Also, to show the effect of heat integration for these two solutions, T-Q curves for presented in Figure 7.

Finally, note that the simultaneous strategy accounts for utility costs but not capital costs associated with heat integration. As noted above, utility costs are generally more sensitive to **changes** in decision variables in the flowsheet. For the Case 2 solutions, minimum **cost heat** exchanger **networks were** generated using the MAGNETS synthesis program (Floudas et al [5]). For the simultaneous and sequential results the heat exchanger networks are given in Figures 8 and 9, respectively. The capital costs for these networks are $222,038/yr (21,001 m^2) for the simultaneous solution and $195,413/yr (17,263 m^2) for the sequential solution.

Methanol Synthesis Flowsheet

The methanol synthesis process flowsheet is presented in Figures 10(a) and 10(b). Here, a synthesis gas feed of nitrogen, hydrogen, carbon monoxide, carbon dioxide and methanol is compressed and mixed with recycle. The mixture enters a five stage quenched-bed reactor where methanol is produced according to the following overall reactions:

CO + $2H_a$ <==> CH_aOH

CO_2 + $3H_a$* <==> CH_30H + H^O

The reactor effluent is first flashed and the overhead is then absorbed with water to separate the synthesis gases from methanol. To further separate methanol from water the bottoms streams from both units are fed to a two column separation sequence. Water is then recycled back to the absorber.

The flowsheet and reactor model were taken from Meyer et al [12]. Here a system of five intermediate reactions was solved using a differential equation-based quenched bed reactor model. Using the results of this model, curves relating methanol conversion to catalyst weight were generated. These were subsequently used in the FLOWTRAN simulation model of the flowsheet. Outlet temperature of the five stage quenched bed system was found to be the maximum allowable temperature, 553 K. Because the process deals with ten chemical components, two tear streams and nine decision variables, it was decided to optimize this flowsheet using the "black box" optimization strategy implemented on FLOWTRAN. As discussed in Lang and Biegler [11], this strategy, although time-consuming, can be advantageous if the number of tear stream components is greater than the number of decision variables.

The objective function is the net present value of the total before-tax profit using a project life of 5 years and a 15% rate of return. As with the ammonia example, feed flowrate was chosen as a decision variable and a constraint of 1000 metric tons per day was imposed. Additional constraints were purity of the methanol product (99.8%) and consistency specifications on process streams. The minimum temperature approach was set to 10 °F.

It should be noted that the actual number of hot and cold streams that was determined in the sequential and simultaneous strategies are different. As shown in Fig. 10(a) there are 5 hot and 4 cold streams for the former, while as shown in Fig. 10(b) there are 6 hot and 3 cold streams for the latter.

The simultaneous and sequential solutions for the methanol process optimization are given in Tables 5 and 6. As can be seen the simultaneous strategy leads to a before-tax profit of $32,036,800/yr while the sequential strategy yields $31,005,900/yr. The optimal decision variable vector in Table 5 shows that although conversion per pass is about the same for both cases, the simultaneous case has a much higher overall conversion (71.3% vs. 65.3%y and a higher recycle ratio (7.4 vs. 5.4), because the lower cost of utilities is correctly represented in the simultaneous optimization. It is also interesting to see in Table 5 that the reflux ratio of the second column is somewhat higher in the simultaneous solution (5.06 vs. 4.89). Note in Table 6 that the profit is over $1,000,000/yr higher for the simultaneous case. This is due to an almost $6,000,000/yr savings in raw material costs. Other savings come from lower capital and utility costs for the feed compressor. For the sequential case, on the other hand, a higher purge gas credit is realized (almost $5,000,000/yr). To show the effect of heat integration for these two solutions, T-Q curves for presented in Figure 11.

Finally, the MAGNETS network synthesis program was used to generate the heat exchange networks for the simultaneous and sequantial methanol optimizations. For the simultaneous and sequential results the heat exchanger networks are given in Figures • 12 and 13, respectively. Capital costs for these networks are $342,744/yr (38,200 m^2) for the simultaneous solution and $334,845/yr (35,684 m^2) for the sequential solution. These results and the one for the ammonia problem show that the capital cost of the network is not very sensitive to the optimization formulation.

CONCLUSIONS

Based on the earlier work of Duran and Grossmann [4], a simultaneous formulation for heat integration and optimization has been developed for use with sequential modular simulators. Using the FLOWTRAN simulator, this implementation is fairly transparent to the user and simply requires the addition of special purpose cost blocks to account for the potential of the heat integration. The optimization strategy developed by Lang and Biegler [11] has been used for solving the resulting nonlinear program.

To carry out the simultaneous strategy, two formulations, the explicit and implicit, were developed and compared. As pinch points change the function describing minimum utilities can be nondifferentiable. The explicit formulation of Duran and Grossmann [4] treats these nondifferentiabilities by including additional inequality constraints and describing the minimum utilities in terms of active sets corresponding to the pinch points. However, due to the presence of linear variables in this formulation, difficulties arise in the approximation of the Hessian matrix for the quadratic programming subproblems. The implicit formulation, on the other hand, does not encounter this difficulty since it performs the minimum utility calculation directly at each SQP iteration. Inclusion of multiple utilities is also straightforward. Moreover, the implicit formulation, despite the disadvantage of possible nondifferentiable functions, appears to be more efficient for sequential modular simulators because the problem has fewer constraints.

For a generic flowsheet satisfying certain assumptions it was proved that the simultaneous formulation leads to higher profit and conversion of raw materials. These properties were further demonstrated on two complex process optimization problems, the ammonia synthesis and methanol synthesis processes. Comparing the sequential and simultaneous solutions for these processes one observes greater conversion of raw material to product, higher recycles and increases in before tax profit of up to $1,000,000/yr for the simultaneous case. Moreover, heat exchanger networks for both the simultaneous and sequential solutions require roughly the same capital cost. Therefore, the simultaneous approach presented here offers definite advantages as part of a strategy for heat integration and flowsheet optimization.

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Appendix A. On the optimal trade-off between raw material, capital and energy in recycle processes.

This Appendix will show that in recycle processes the simultaneous strategy will lead to solutions that exhibit higher overall conversion and lower cost than the sequential strategy.

For the sake of simplicity, the processing scheme shown in Fig. 1 will be considered. In order to develop a simplified model for this process the following assumptions will be made:

- Single reaction A → B with fixed conversion per pass r.
- Feedstock contains inert C with composition y^C.
- The production rate of B, P_B, is fixed.
- Fixed pressure and temperature levels throughout the flowsheet.
- Feed preparation (FP) and recycle (R1) involve only electricity demands.
- Reaction step (R1) and product recovery (PR) involve heating and cooling demands.
- Perfect split between AC/B in splitter S1.
- Fixed recovery fraction of B (β) in PR.
- Cost models are assumed to be linear functions of the flows f_i. The cost of the heat recovery network is neglected.

Based on the above, the cost models for the different items are as follows:

Net cost feedstock: $C_{NF} = C_F - I_P$, where

Feedstock: $C_F = c_F(f_0^A + f_0^C)$

Purge income: $I_P = c_P(f_6^A + f_6^C)$

Capital and operating expenses = $C_{FP} + C_{R1} + C_{R2} + C_{PR}$

where

Feed preparation: $C_{pp} = c_{Fp}(f£ * f_0^C)$

Reaction step: $C_{R1} = c_{nil}(f^* \bullet f?_1)$

Recycle step: $C_{R2} = c_{R2}(f_5^* + f£)$

Product recovery: $C_{M_{PR}} = c_B JPJfi_{PR\ D}$

The unit costs c_p, c_p, c_{pp}, c_{R1}, c_{R2}, c_{pR} are for the case of no heat integration. For the case of heat integration $c^t_{R1} < c_{R1}$, $c'_{PR} < c_{pR}$. The total cost of the flowsheet with no heat integration is then given by.

$$C = C_{NF} + C_{FP} + C_{R1} + C_{R2} + C_{PR}$$

Each of the terms in this function can be expressed as a function of x, the overall conversion of A in the feedstock to B in the product P_B. By performing the appropriate mass balances in Fig. 1 it can be shown that.

$$C_{NF}(x) = \frac{P_B}{\beta(1-y^c)}\left[\left(\frac{c_F - c_P}{x}\right) + c_p(1-y^c)\right]$$

$$C_{FP}(x) = \frac{P_B \quad c_{FP}}{\beta(1-y^c)\, x}$$

$$C_{R1}(x) = \frac{P_B c_{R1}}{fir}\left[1 + \left(\frac{y^c}{1-y^c}\right)\left(\frac{1-r}{1-x}\right)\right]$$

$$C_{R2}(x) = \frac{P_B c_{R2}}{\beta}\left(\frac{1}{r} - \frac{1}{x}\right)\left[1 + \left(\frac{y^c}{1-y^c}\right)\left(\frac{1}{1-x}\right)\right]$$

$$C_{PR} = \frac{P_B c_{PR}}{fi}$$

From the above, it can easily be shown that in the range, r £ x < 1, each of the cost terms, except the last one, is monotone in x; that is

$$\frac{dC_{NF}}{dx} < 0, \frac{dC_{fP}}{dx} < 0, \frac{dC_{R1}}{dx} > 0, \frac{dC_{R2}}{dx} > 0, \frac{dC_{PR}}{dx} = 0$$

Furthermore, since $c'_{H} < c_{R1}$,

$$C'_{R1}(x) = aC_{R1}(x). \quad \text{where } a = \frac{c'_{R1}}{c_{R1}} < 1 ,$$

$$\text{and therefore } \frac{dC'}{dx} < \frac{dC}{dx}. \qquad \text{(A1)}$$

The following property can then be established.

<u>Property</u> The optimal overall conversion of the simultaneous strategy is greater than the optimal overall conversion of the sequential strategy. Furthermore, the former exhibits a lower total cost.

<u>Proof</u> For the sequential strategy the optimal conversion is obtained by minimizing the total cost with no heat integration:

$$\min_x \quad C = C_{NF}(x) + C_{pp}(x) + C_{R1}(x) + C_{R2}(x) + C_{pR} \qquad \text{(A2)}$$

Assuming that the optimum conversion occurs at an interior in r < x < 1, the necessary condition for the minimum cost is dC/dx * 0. Let $\bar{x}$ be the optimum overall conversion. Then, from (A2)

$$\left(\frac{dC}{dx}\right)_{\bar{x}} = \frac{dC_{NF}}{dx}(\bar{x}) + \frac{d}{dx}(\bar{x}) + \frac{d}{dx}(\bar{x}) + \frac{dC_{R2}}{dx}(\bar{x}) = 0 \qquad \text{(A3)}$$

Consider now the replacement of C_{RI} with C^f_{RI} and C_{PR} with C^f_{PR} (with the unit costs c^f_{R1}, c^I_{PR}) to redefine the cost function $C^I(x)$ with heat integration. Then from (A1) it follows that (A3) reduces to

$$\left(\frac{dC'}{dx}\right)_{\bar{x}} < 0$$

This then implies that there is a descent direction for C at x for x > x. Hence, if x is the optimal conversion for the cost C of the simultaneous strategy, x > x.

Finally, since x < x , and x is the optimal solution to C (x), it follows that C' (x̄) > C' (x̄'). Hence, this proves that the simultaneous strategy leads to higher overall conversion and to lower total cost.

As a corollary to this property, the following can also be established:

The optimal solution for the simultaneous strategy will exhibit:

(a) Lower net feed cost and feed preparation cost.

(b) Higher recycle costs.

This simply follows from the fact that

$$\left(\frac{dC_{NF}}{dx}\right)_{\bar{x}} < 0, \quad \left(\frac{dC_{FP}}{dx}\right)_{\bar{x}} < 0, \quad \left(\frac{dC_{R2}}{dx}\right)_{\bar{x}} > 0 \text{ and } \bar{x}' > \bar{x}.$$

Appendix B - Example for Implicit Formulation

Consider the following multiple utility example:

	FC_p (kW/K)	T^{in} (°K)	T^{out} (°K)
H1	1	450	350
H2	2	400	280
C1	2	320	480

with the following utilities

HU1 : HP steam @ 500K
HU2 : LP steam 9 430K
CU1 : Cooling @ 300K
CU2 : Refrigerant @ 270K
ΔT_m = 10 K

The algorithm for the implicit formulation requires the following relations.

$$[z_p^H]^k = \sum_{j \in C_k} f_j c_j [max \{0, t^{out}_j - (T^p - \Delta T)\} - max \{0, t^{in}_j - (T^p - \Delta T)\}]$$

$$- \sum_{i \in H_k} F_i C_i [max \{0, T_i^{in} - T^p\} - max \{0, T_i^{out} - T^p\}]$$

$$Ct(y) = \sum_{i \in H_k} F_i C_i (T_i^{in} - T_i^{out}) - \sum_{j \in C_k} f_j c_j (t_j^{out} - t_j^{in})$$

and follows the steps below:

1) From enthalpies of streams, develop the FC_p's as given above.

2) Select pinch candidates and stream sets.

C_o = {Cl} T* = {450,400,330}

H_o = {H1,H2}

3) Set k = 0 and calculate heat deficits for pinch candidates.

p	T^p	z_H^p(kW)
1	450	80
2	400	130
3	330	80

4) Calculate min. utilities.

$\Omega(y) = 20\ kW$

$Q_h^0 = max\ z_p^H = 130\ kW \qquad T_0^p = 400\ K$

$Q_c^0 = \Omega(y) + Q_h^0 = 150\ kW$

5) Set k = 1 and consider utilities below pinch.

$T^k = T_C^1 = 300K$. Consider the following streams in H_1 and C_1

	FC_p	T^{in}	T^{out}
H1	1	310	280

6) Calculate z_H^p and intermediate utility.

p	T^p	z_H^p
1	310	60

$Q_C^1 = z_H^p = 60\ kW \qquad T_1^p = 310\ K$

$Q_c^1 = Q_c^0 - Q_C^1 = 90\ kW$

7) Go to 8), all cold utilities considered.

8) Set k = 2, $T^k = T^*_H = 430$ K. Consider the following streams for H_2 and C_2.

	FC_p	T^{in}	T^{out}
H1	1	450	430
C1	2	420	480

9) Calculate z^p_H and intermediate utility.

p	T^p	z^p_H
1	450	80
2	430	100

$$Q^1_h = \max_p \{z^p_H\} = 100 \ kW$$

$$Q^1_H = Q^0_h - Q^1_h = 30 \ kW$$

10) Stop, all utilities considered.

This problem was also solved with identical results by Duran and Grossmann [4] using an LP formulation. With the above algorithm, however, exact utility costs are not required.

Appendix C - Example of Nondifferentiality in Implicit Formulation

Consider the very simple system of one hot and one cold stream. Assume that t_{in}, T_{out} and t_{out} are fixed, T_{in} is variable and $t_{in} > T_{out}$.

The following cases can be considered for T_{in}

i) $T_{in} > t_{out} + \Delta T_{min}$

$$Q_H = max\{z_H^p\} = 0 \quad , \quad T^p = t_{in} + \Delta T_{min}$$

ii) $t_{in} + \Delta T_{min} \le T_{in} \le t_{out} + \Delta T_{min}$

$$Q_H = max\{z_H^p\} = fc\ [t_{out} - (T_{in} - \Delta T_{min})] \quad , \quad T^p = T_{in}$$

iii) $T_{in} < t_{in} + \Delta T_{min}$

$$Q_H = max\{z_H^p\} = fc[t_{out} - t_{in}] \quad , \quad \text{no pinch point}$$

$Q_H(T_{in})$ can be plotted as shown in Fig. C.1. Note that this function is nondifferentiable at points where the pinch point changes. Also, since

$$Q_C = Q_H + \Omega(y)$$

$$\text{where } \Omega(y) = FC(T_{in} - T_{out}) - fc(t_{out} - t_{in}) \ ,$$

$Q_C(T_{in})$ is also nondifferentiable at the same points.

Table JL Ammonia Process z Case 1, Optimal Solution Vector

Design Variables	Bounds Lower	Bounds Upper	Starting Point	Solution Sequential	Solution Simultaneous
1. Inlet Temp. (F) of Reactor Tl	400	400	400	400	400
2. Inlet Temp. (F) of 1st Flash	65	100	65	65	65
3. Inlet Temp. (F) of 2nd Flash	35	60	35	35	35
4. Temp. (F) of Recycle	60	400	60	60	101.5
5. Purge Fraction (%)	0.1	10	1.38	0.87	0.496
6. Outlet Press, (psia) of Main Comp.	1500	4000	2173.73	2173.74	2173.74
7. Outlet Press, (psia) of Recyc. Comp.	1500	4000	2173.74	2173.74	2173.74
8. Feed Flowrate of Rich H2	2461.4	3000	2631.97	2631.13	2588.79
9. Feed Flowrate of Rich N2	643	1000	691.42	691.73	699.25
10. Ratio of H2/N2 to reactor	0.5	3.5	2.75	2.683	0.86
11. Conversion • per Pass(H2) %				40.0	53.7
12. Overall (H2) % Conversion				93.50	95.07
13. Recycle Ratio				2.31	3.32
14. Outlet Temp. (F) of Reactor				822.99	712.01
15. Inlet Temp. (F) of Preheater				159.14	145.49

Table 2. Balance Sheet for Ammonia Process - Case 1

Cost/Sales	Sequential	**Simultaneous**	Difference
I. Capital Costs ($)			
1. Main Comp.	270,940	269,180	1,760
2. Recycle Comp.	26,837	38,745	- 11,908
3. Reactor	56,430	62,345	- 5,915
4. 1st Flash Drum	34,415	52,953	- 18,538
5. 2nd Flash Drum	26,488	45,937	- 19,449
6. 3rd Flash Drum	6,248	7,357	- 1,109
Subtotal	421,358	476,519	- 55,161
II. Utility cost ($/yr)			
1. Main Comp.	2,412,984	2,389,968	23,016
2. Recycle Comp.	80,472	138,180	- 57,708
3. Cool. Utility	276,444	**476,448**	-200,004
Subtotal	2,769,900	3,004,596	-234,696
III. Raw Material ($/yr)			
1. H2-rich Feed	36,479,016	35,880,684	598,332
2. N2-rich Feed	488,796	494,088	- 5,292
Subtotal	36,967,812	36,374,772	593,040
IV. Sales ($/yr)			
1. Purge	470,288	382,823	87,465
2. Byproduct	109,126	95,580	13,546
3. Product	66,198,720	66,200,389	- 1,669
Subtotal	66,778,134	66,678,792	- 99,342
V. **Before-Tax Profit ($/yr)**	26,915,216	27,134,800	- 219,584

Table 3. Ammonia Process - Case 2, Optimal Solution Vector

Design Variables	Bounds Lower	Bounds Upper	Starting Point	Solution Sequential	Solution Simultaneous
1. Inlet Temp. (F) of Reactor Tl	400	400	400	400	400
2. Inlet Temp. (F) of 1st Flash	65	100	65	65	65
3. Inlet Temp. (F) of 2nd Flash	35	60	35	35	35
4. Temp. (F) of Recycle	60	400	60	78.7	131.575
5. Purge Fraction Øk)	0.1	10	1.38	0.92	0.541
6. Outlet Press, (psia) of Main Comp.	1500	4000	2173.73	2183.84	1583.20
7. Feed Flowrate of H2-rich Feed	2461.4	3000	2631.97	2638.79	2588.43
8. Feed Flowrate of N2-rich Feed	643	1000	691.42	688.32	687.456
9. Ratio of H2/N2 in Reactor Feed	0.5	3.5	2.75	3.5	1.616
10. Conversion per Pass(H2) %				37.30	43.37
11. Overall (H2) % Conversion				93.31	94.75
12. Recycle Ratio				2.28	3.03
13. Outlet Temp. (F) of Reactor				829.40	733.13
14. Inlet Temp. (F) of Preheater				176.82	179.61

Table 4. Balance Sheet for Ammonia Process ^ Case 2

Cost/Sales	Sequential	Simultaneous	Difference
I. Capital Costs ($)			
1. Main Corap.	271,884	220,142	51,742
2. Recycle Comp.	33,599	52,535	-18,936
3. Reactor	56,452	52,194	4,258
4. 1st Flash Drum	32,940	35,590	- 2,650
5. 2nd Flash Drum	24,726	30,595	- 5,869
6. 3rd Flash Drum	6,227	8,498	- 2,271
Subtotal	425,830	399,556	26,274
II. Utility cost i($/yr)			
1. Main Comp.	2,425,332	1,778,028	647,304
2. Recycle Comp.	112,056	216,216	-104,160
3. Cool. Utility	244,272	456,708	-212,436
Subtotal	2,781,660	2,450,952	330,708
Ill• Raw Material ($/yr)			
1. H2-rich Feed	36,573,629	36,055,127	518,502
2. N2-rich Feed	486,375	485,760	615
Subtotal	37,060,004	36,540,887	519,117
IV. Sales ($/yr)			
1. Purge	492,068	377,042	115,027
2. Byproduct	109,756	87,376	22,380
3. Product	66,227,616	66,295,515	- 67,899
Subtotal	66,829,440	66,759,933	69,507
V. Before-Tax Profit ($/yr)	26,860,000	27,743,600	-883,600

Table 5. Methanol Process, Optimal Solution Vector

Design Variables	Bounds Lower	Bounds Upper	Solution Sequential	Solution Simultaneous
1.Conversion per Pass %	1.0	100.0	16.62	16.12
2.Purge Fraction %	0.1	20.0	5.85	3.45
3.Inlet Temp. (F) of Flash F	30.0	200.0	88.58	135.15
4.Inlet Temp. (F) of 1st Column F	100.0	250.0	100.0	134.40
5.Bottom Press. (psia) of 1st Col. Psia	100.0	300.0	300.0	300.0
6.Reflux Ratio of 1st Col.	8.0	15.0	8.0	8.0
7.Bottom Press of 2nd Col. Psia	200.0	500.0	306.0	319.1
8.Reflux Ratio of 2nd Col.	2.5	15.0	4.89	5.06
9.Feed Flowrate lb-mol/Hr	10000.0	50000.0	15817	14350
OBJ. FUNC.(PROFIT PER YEAR) 10^6 $			31.0049	32.0368
OVERALL CONVERSION (Based on CO and CO2) %			65.27	71.26
RECYCLE RATIO			5.40	7.38

Table 6. Balance Sheet for Methanol Process

Costs/Sales	Sequential	Simultaneous	Difference
I. Capital Costs ($)			
1.Feed Comp.	522,346	463,368	58,978
2.Recycle Comp.	343,462	397,956	- 54,494
3.Reactor			
Reactor	65,418	65,190	228
Catalyst	540,032	533,134	6,898
4.Flash Drum	230,635	301,575	- 60,940
5.Absorber	1,478,270	2,007,930	- 528,660
6.1st Column	268,390	274,750	- 6,360
7.2nd Column	224,760	237,270	- 12,510
8 .Pump	9,910	12,036	- 2,126
II. Utility Cost ($/yr)			
1.Feed Comp.	6,335,616	5,312,160	1,023,456
2.Recycle Comp.	3,419,985	4,246,956	- 826,971
3.Pump	157,332	187,572	- 30,240
4.H&C Utility	3,250,296	2,768,472	481,824
III. Raw Material Cost ($/yr)			
1.Synthesis Gas	63,552,888	57,657,936	5,894,952
IV. Sales ($/yr)			
1.Product	87,589,068	87,958,248	- 369,180
2.Purge Gas	16,222,273	11,473,940	4,748,333
3.Waste MeOH	4,069,145	4,090,740	- 21,595
Before-Tax Profit ($/yr)	31,004,900.00	32,036,800.00	- 1,031,900

CAPTIONS OF FIGURES

Fig. 1. Processing Scheme with Recycle

Fig. 2. Optimal Overall Conversion for Sequential Strategy

Fig. 3. Optimal Overall Conversion for Simultaneous Strategy

Fig. 4. Behavior of T-Q curves with Intermediate Utilities

Fig. 5. Ammonia Process Flowsheet

Fig. 6. T-Q Curves for Ammonia Process - Case 1.

Fig. 7. T-Q Curves for Ammonia Process - Case 2.

Fig. 8. Heat Exchanger Network for Ammonia Process - Sequential Solution

Fig. 9. Heat Exchanger Network for Ammonia Process - Simultaneous Solution

Fig. 1O.(a) Methanol Process Flowsheet - Sequential

Fig. 10.{b) Methanol Process Flowsheet - Simultaneous

Fig. 11. T-Q Curves for Methanol Process

Fig. 12. Heat Exchanger Network for Methanol Process - Sequential Solution

Fig. 13. Heat Exchanger Network for Methanol Process - Simultaneous Solution

Fig. C.1. Nondifferentiable Objective Function for Implicit Formulation

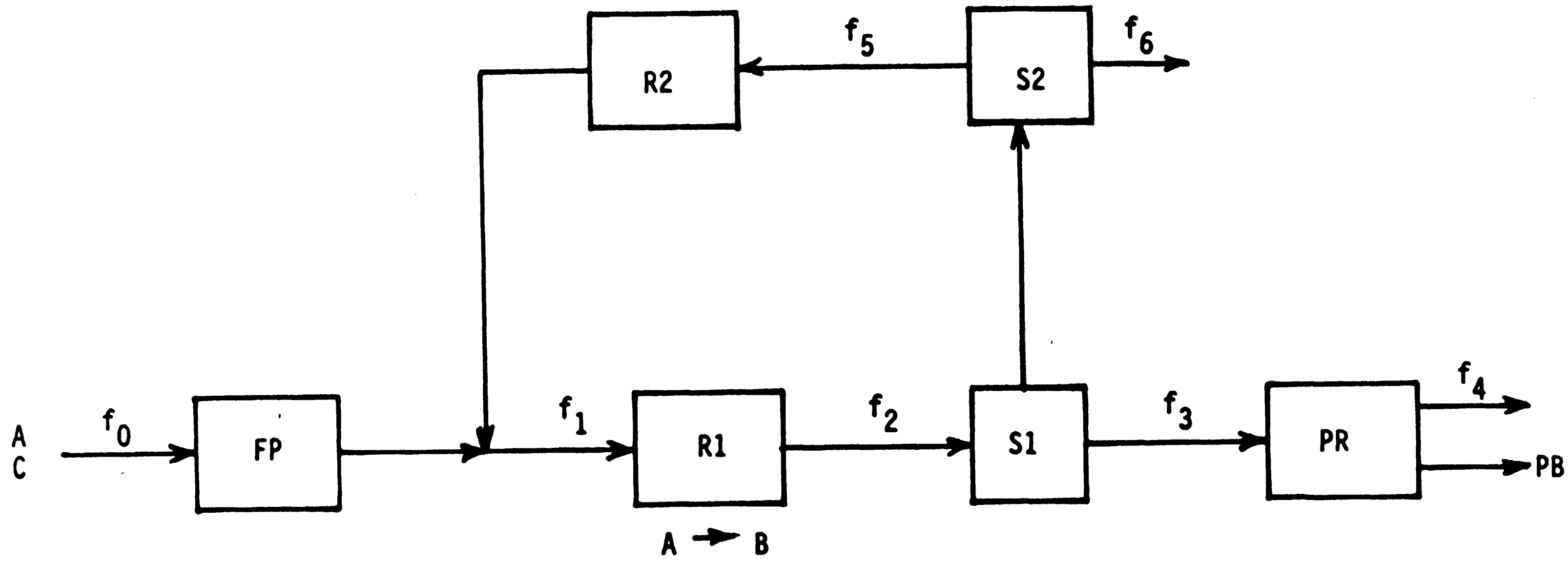


Fig. 1

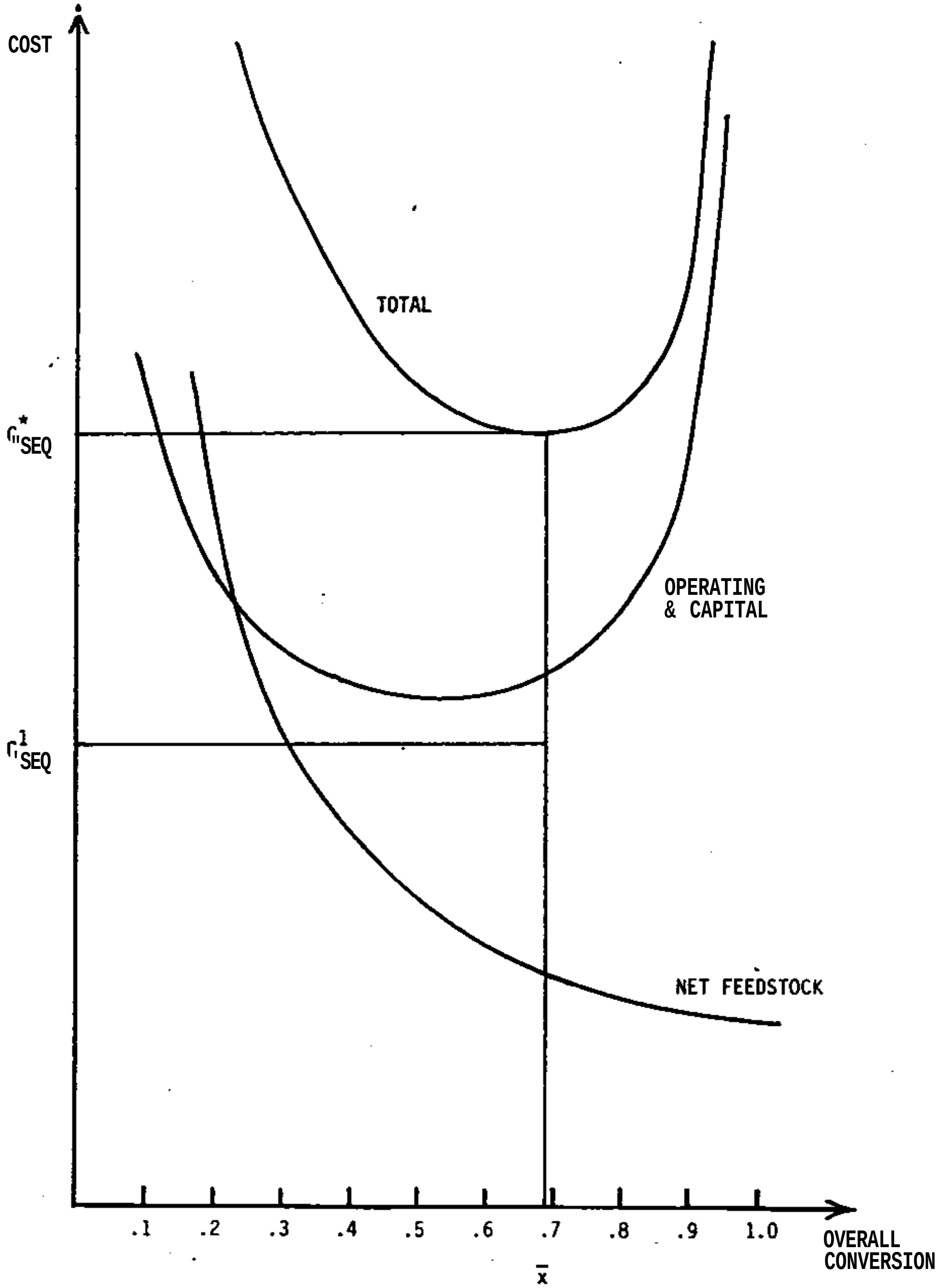
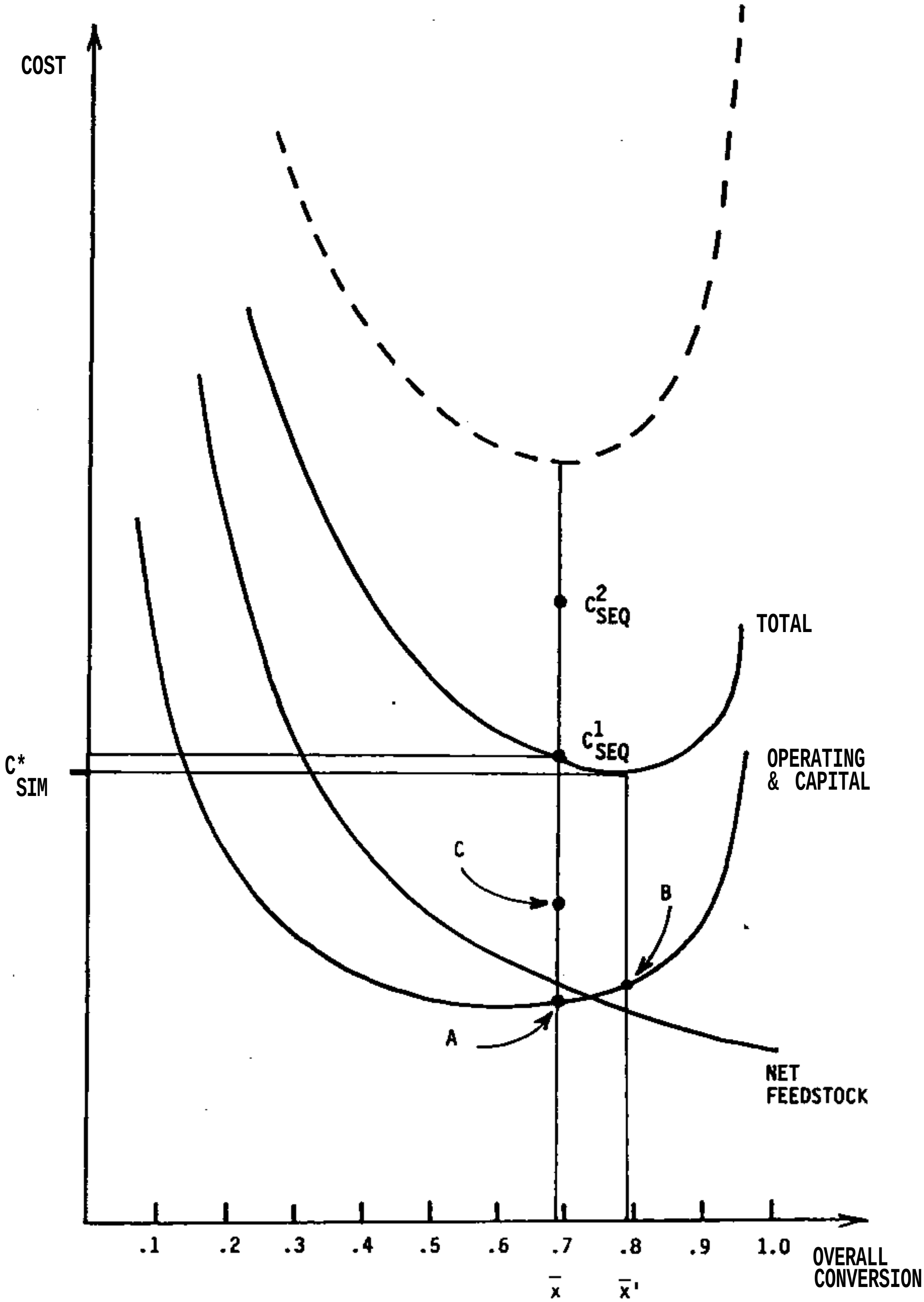

COST
TOTAL
C^*_{SEQ}
OPERATING & CAPITAL
C^1_{SEQ}
NET FEEDSTOCK
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.0
OVERALL CONVERSION
$\bar{x}$

Fig. 2



COST
C^2_{SEQ}
TOTAL
C^1_{SEQ}
C^*_{SIM}
OPERATING & CAPITAL
C
B
A
NET FEEDSTOCK
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.0
OVERALL CONVERSION
$\bar{x}$
$\bar{x}'$

Fig. 3

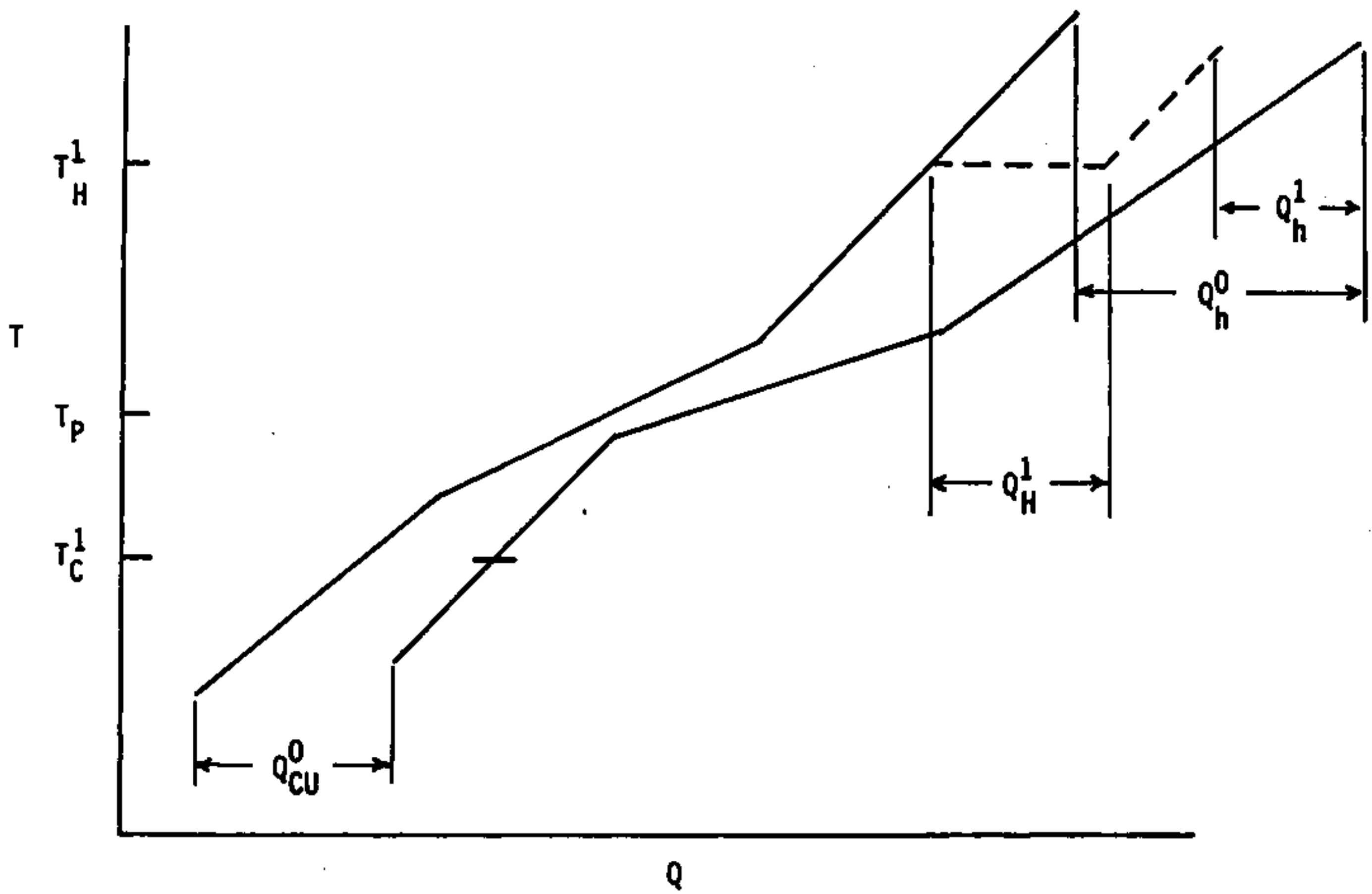
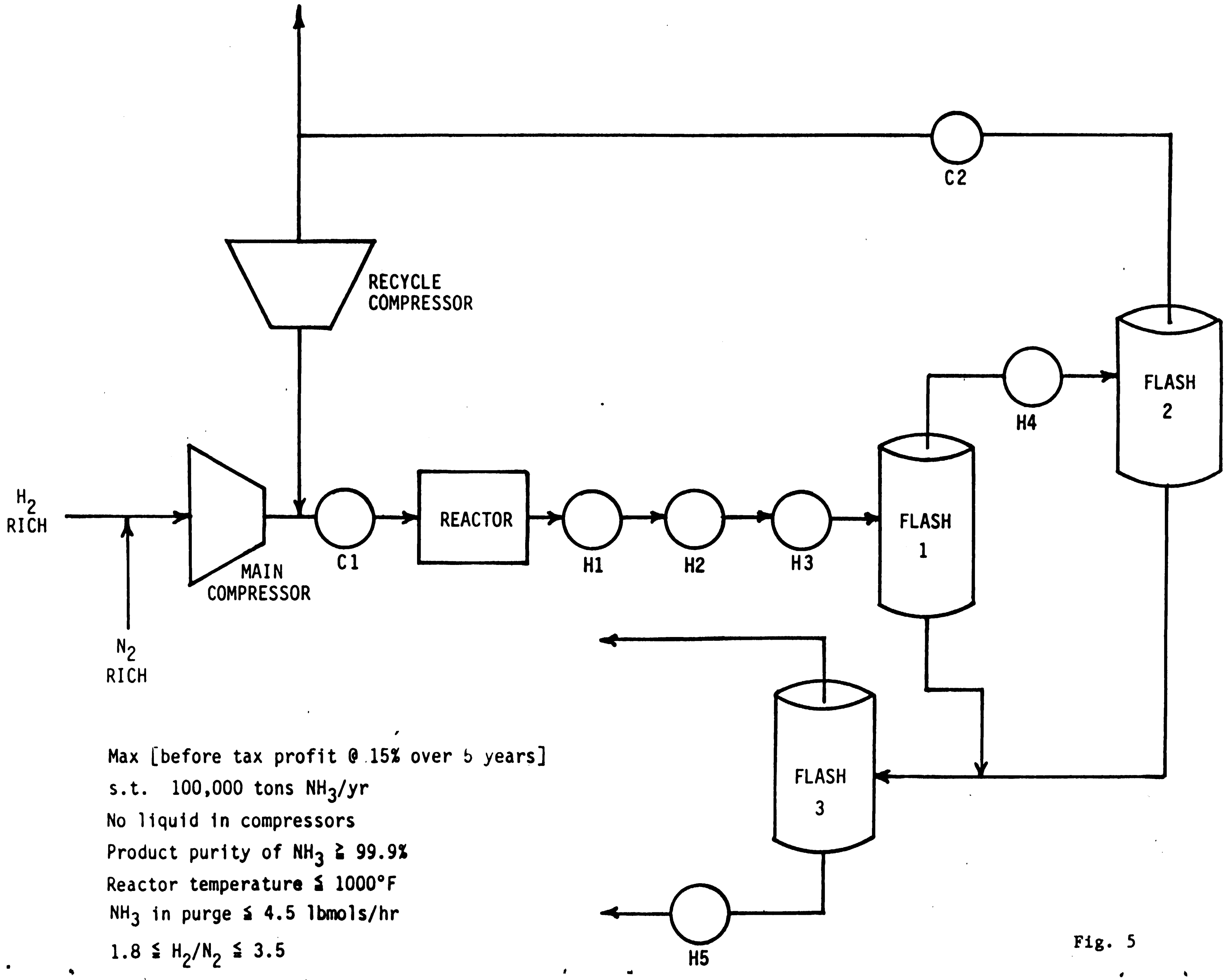


Fig 8



Max [before tax profit @ 15% over 5 years]

s.t. 100,000 tons NH_3/yr

No liquid in compressors

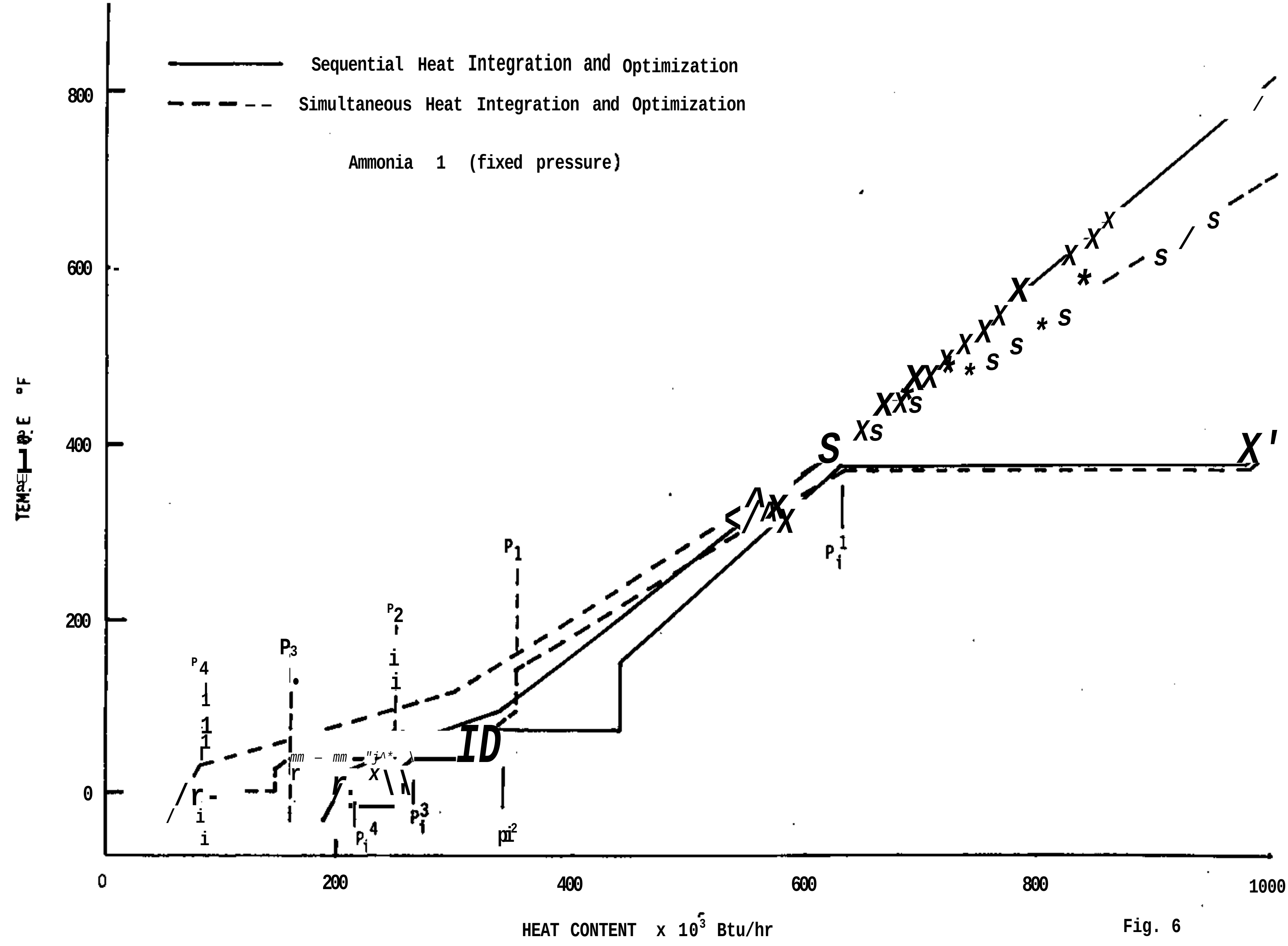
Product purity of $NH_3 \geq 99.9\%$

Reactor temperature $\leq 1000°F$

NH_3 in purge ≤ 4.5 lbmols/hr

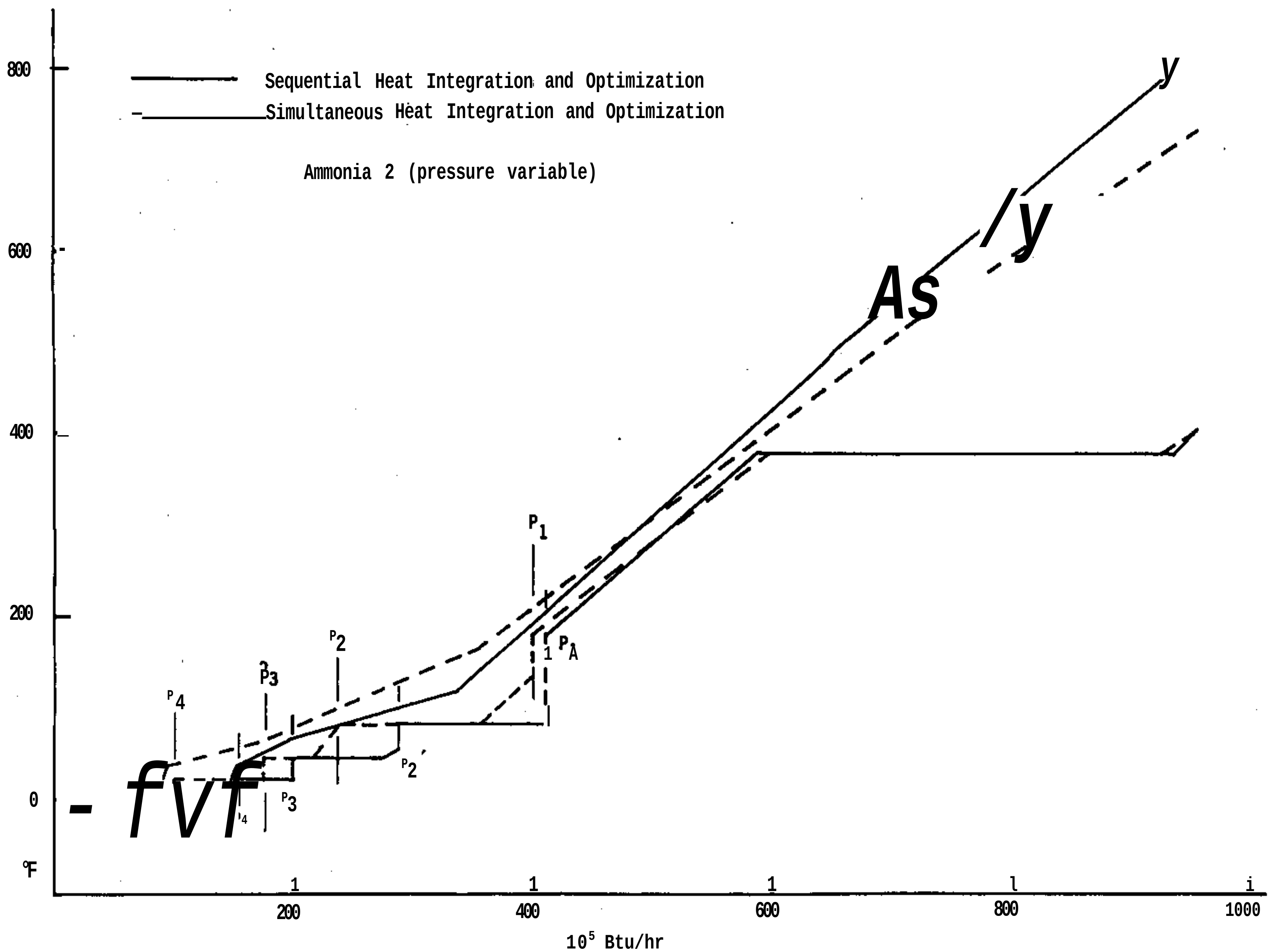
$1.8 \leq H_2/N_2 \leq 3.5$

Fig. 5



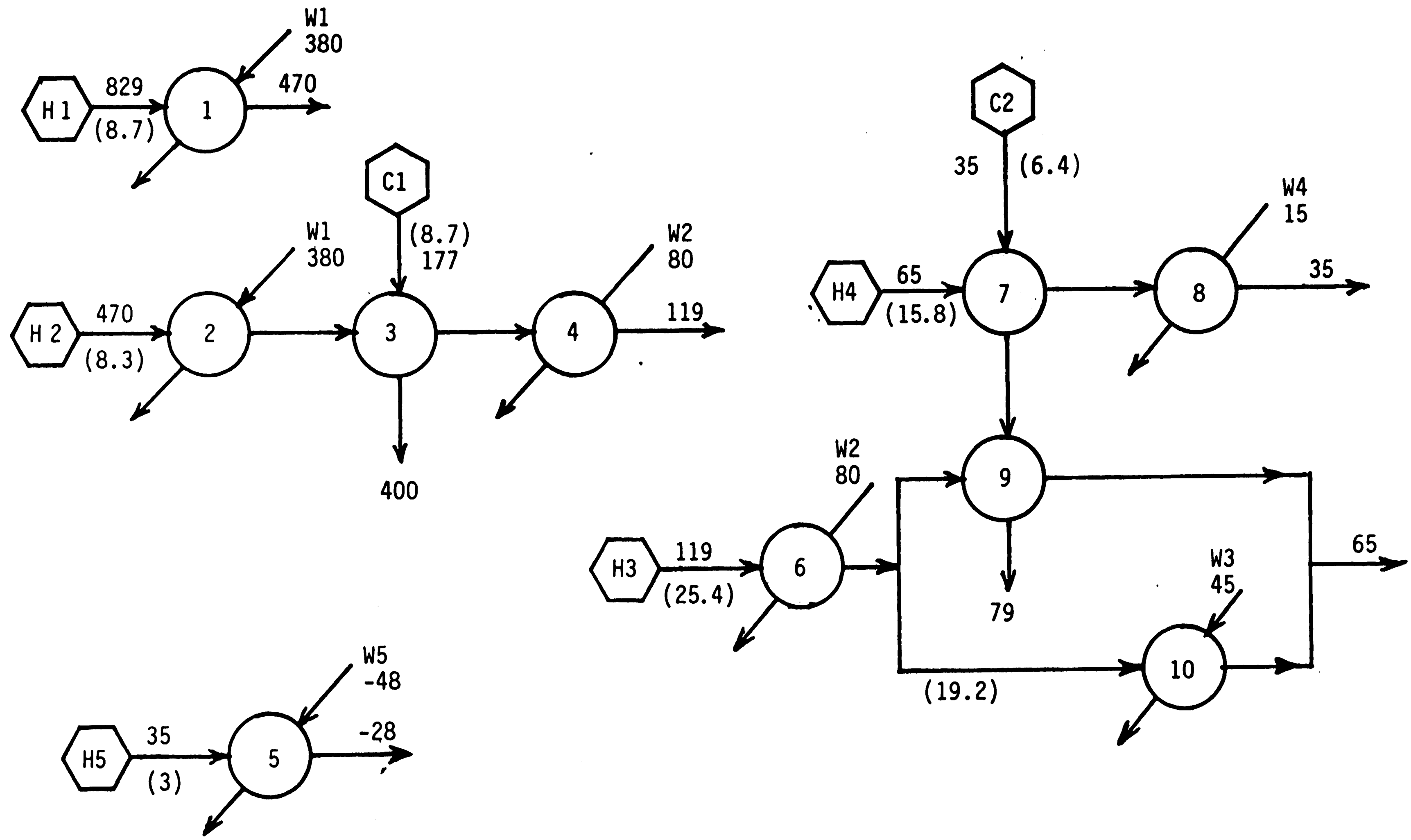
Sequential Heat Integration and Optimization
Simultaneous Heat Integration and Optimization
Ammonia 1 (fixed pressure)
800
600
400
200
0
0
200
400
600
800
1000
HEAT CONTENT x 10³ Btu/hr

Fig. 6



Sequential Heat Integration and Optimization
Simultaneous Heat Integration and Optimization
Ammonia 2 (pressure variable)
800
600
400
200
0
°F
200
400
600
800
1000
10^5 Btu/hr
P1
P2
P3
P4

Fig. 7



H 1
829
(8.7)
1
W1
380
470
H 2
470
(8.3)
2
W1
380
C1
(8.7)
177
3
400
4
W2
80
119
H5
35
(3)
5
W5
-48
-28
C2
35
(6.4)
H4
65
(15.8)
7
8
W4
15
35
H3
119
(25.4)
6
W2
80
9
79
10
W3
45
(19.2)
65

Fig. 8

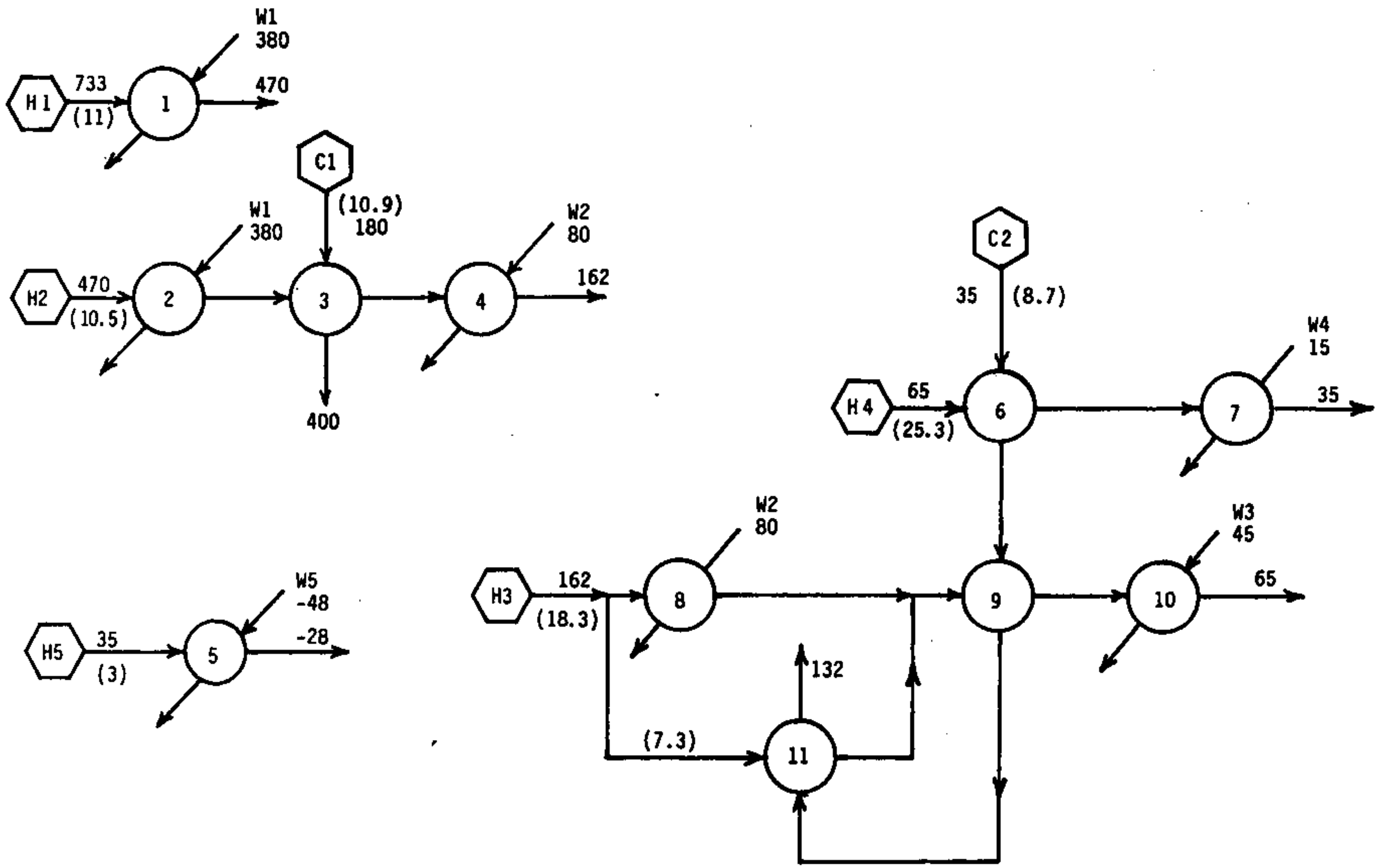

W1
380
H1
733
(11)
1
470
C1
(10.9)
180
W1
380
H2
470
(10.5)
2
3
400
W2
80
4
162
C2
35
(8.7)
H4
65
(25.3)
6
W4
15
7
35
W5
-48
H5
35
(3)
5
-28
W2
80
H3
162
(18.3)
8
9
W3
45
10
65
132
(7.3)
11

Fig. 9

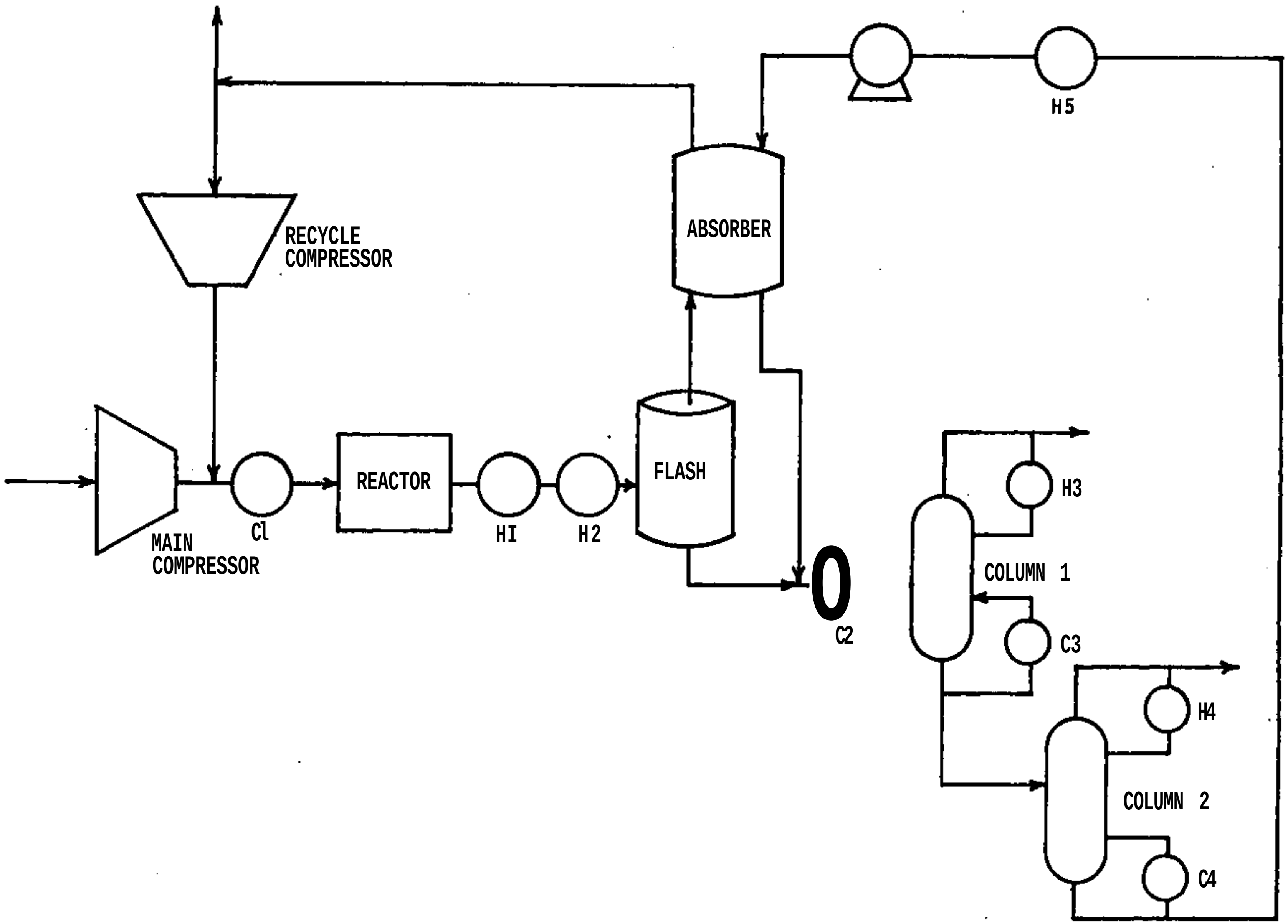

H5
ABSORBER
RECYCLE
COMPRESSOR
FLASH
REACTOR
MAIN
COMPRESSOR
Cl
HI
H2
C2
H3
COLUMN 1
C3
H4
COLUMN 2
C4

Fig. 10(a)

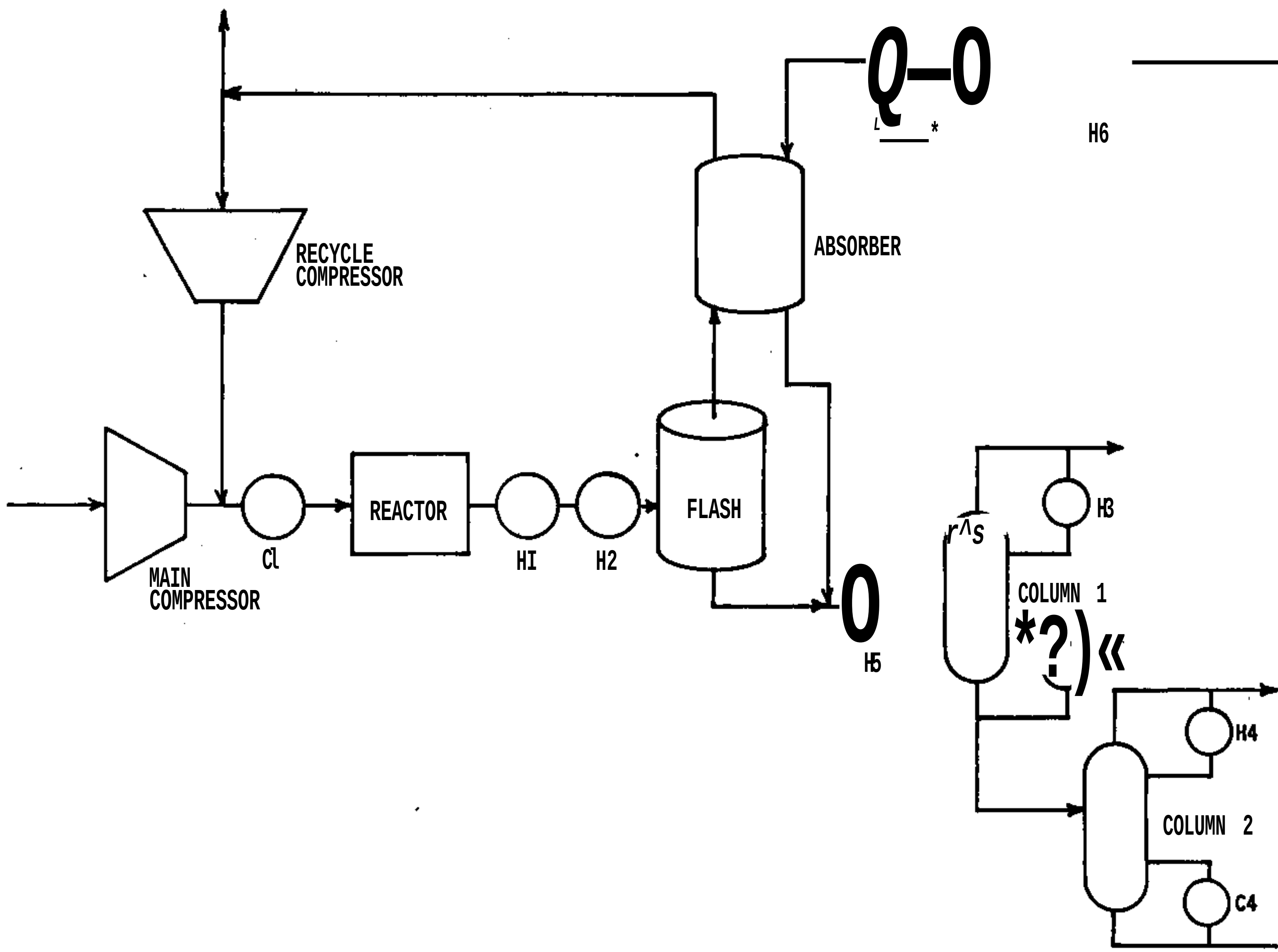

RECYCLE
COMPRESSOR
ABSORBER
H6
MAIN
COMPRESSOR
C1
REACTOR
H1
H2
FLASH
H5
H3
COLUMN 1
H4
COLUMN 2
C4

Fig. 10(b)

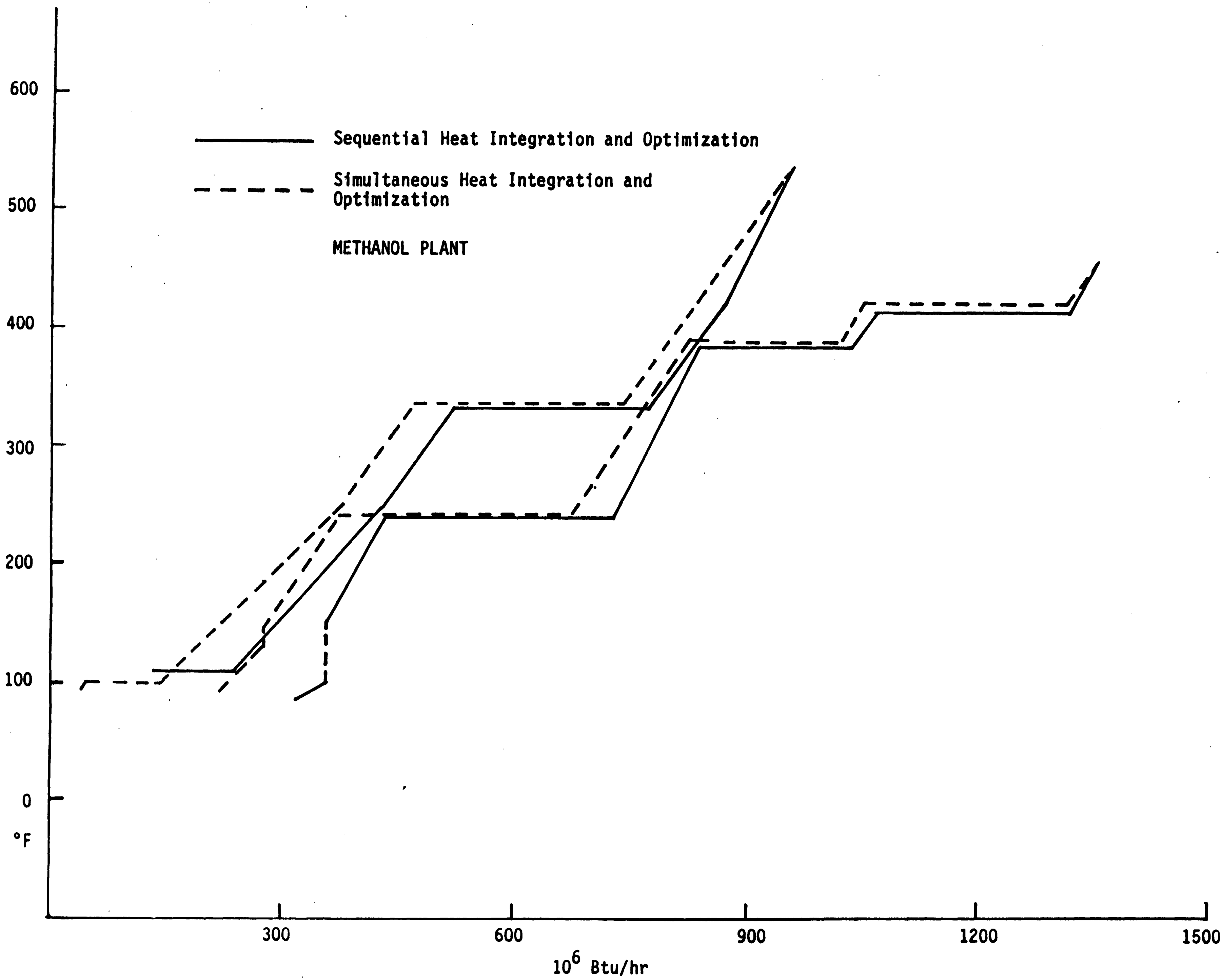

Sequential Heat Integration and Optimization
Simultaneous Heat Integration and Optimization
METHANOL PLANT
600
500
400
300
200
100
0
°F
300
600
900
1200
1500
10^6 Btu/hr

Fig. 11

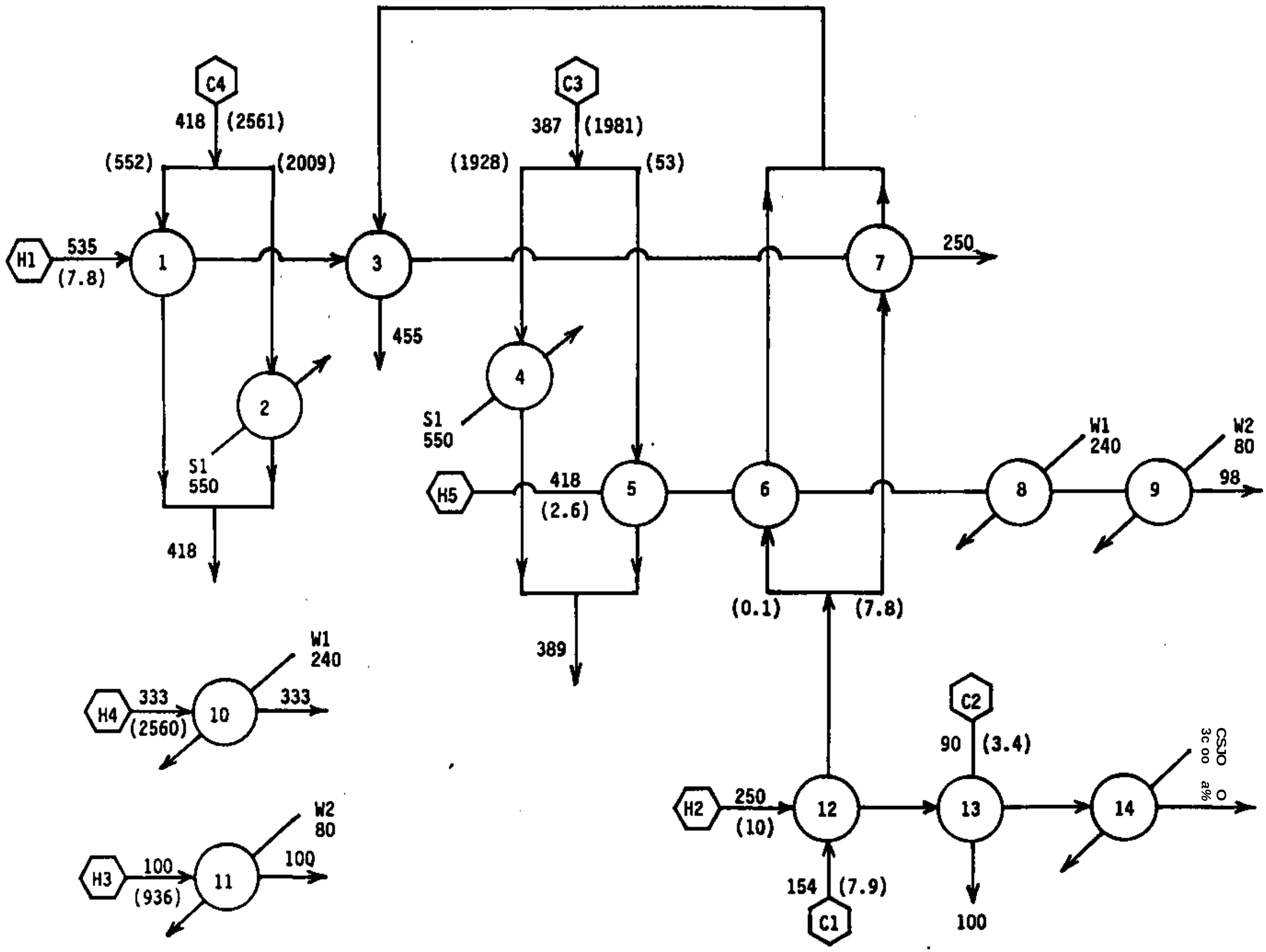
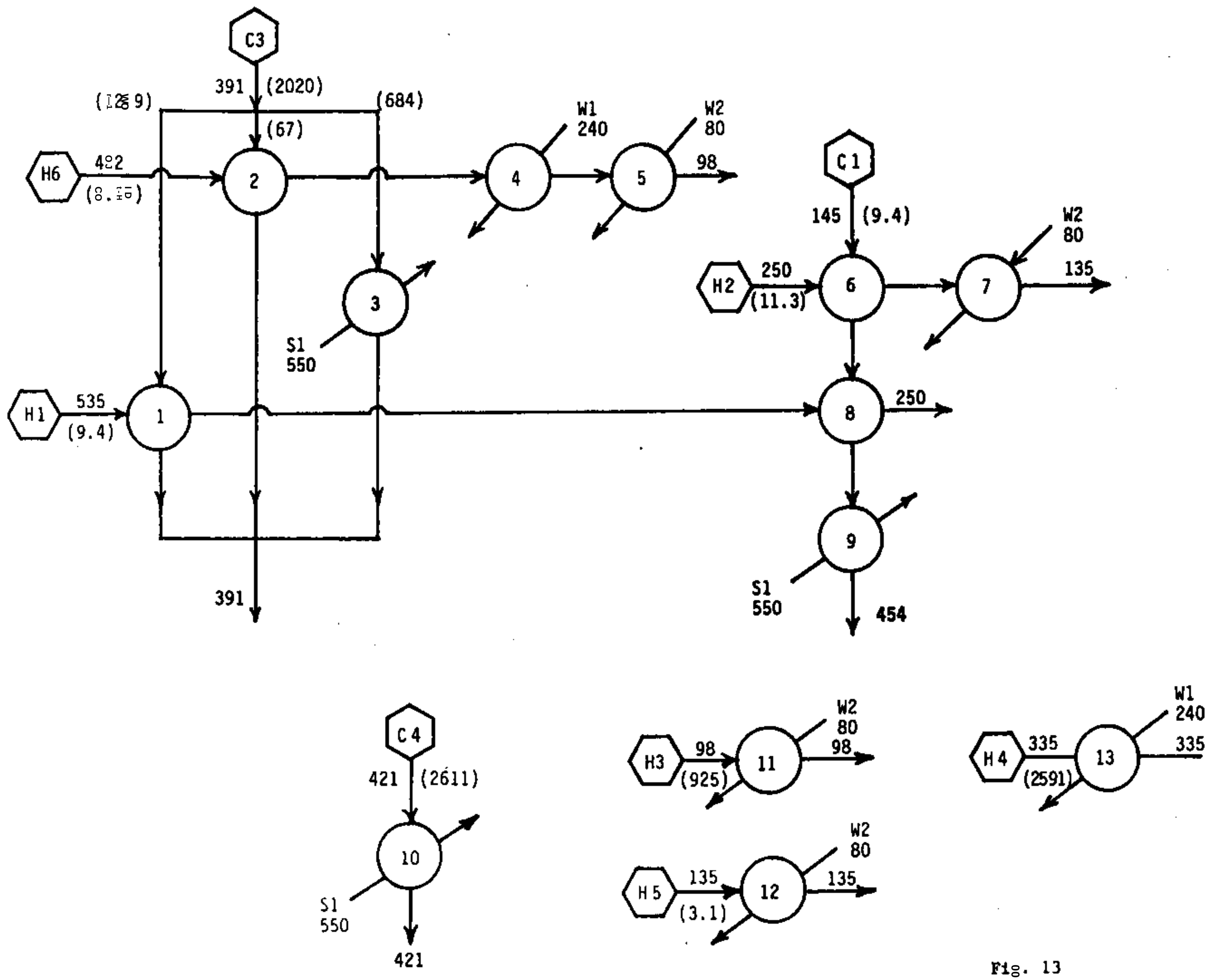

H1
535
(7.8)
1
C4
418
(2561)
(552)
(2009)
2
S1
550
418
3
455
C3
387
(1981)
(1928)
(53)
4
S1
550
H5
418
(2.6)
5
389
6
(0.1)
(7.8)
7
250
8
W1
240
9
W2
80
98
H4
333
(2560)
10
W1
240
333
H3
100
(936)
11
W2
80
100
H2
250
(10)
12
C1
154
(7.9)
13
C2
90
(3.4)
100
14

Fig. 12



C3
391
(2020)
(67)
(684)
H6
H1
535
(9.4)
1
2
3
4
5
S1
550
W1
240
W2
80
98
391
C1
145
(9.4)
H2
250
(11.3)
6
7
135
8
250
9
454
C4
421
10
421
H3
98
(925)
11
H5
135
(3.1)
12
H4
335
(2591)
13

Fig. 13

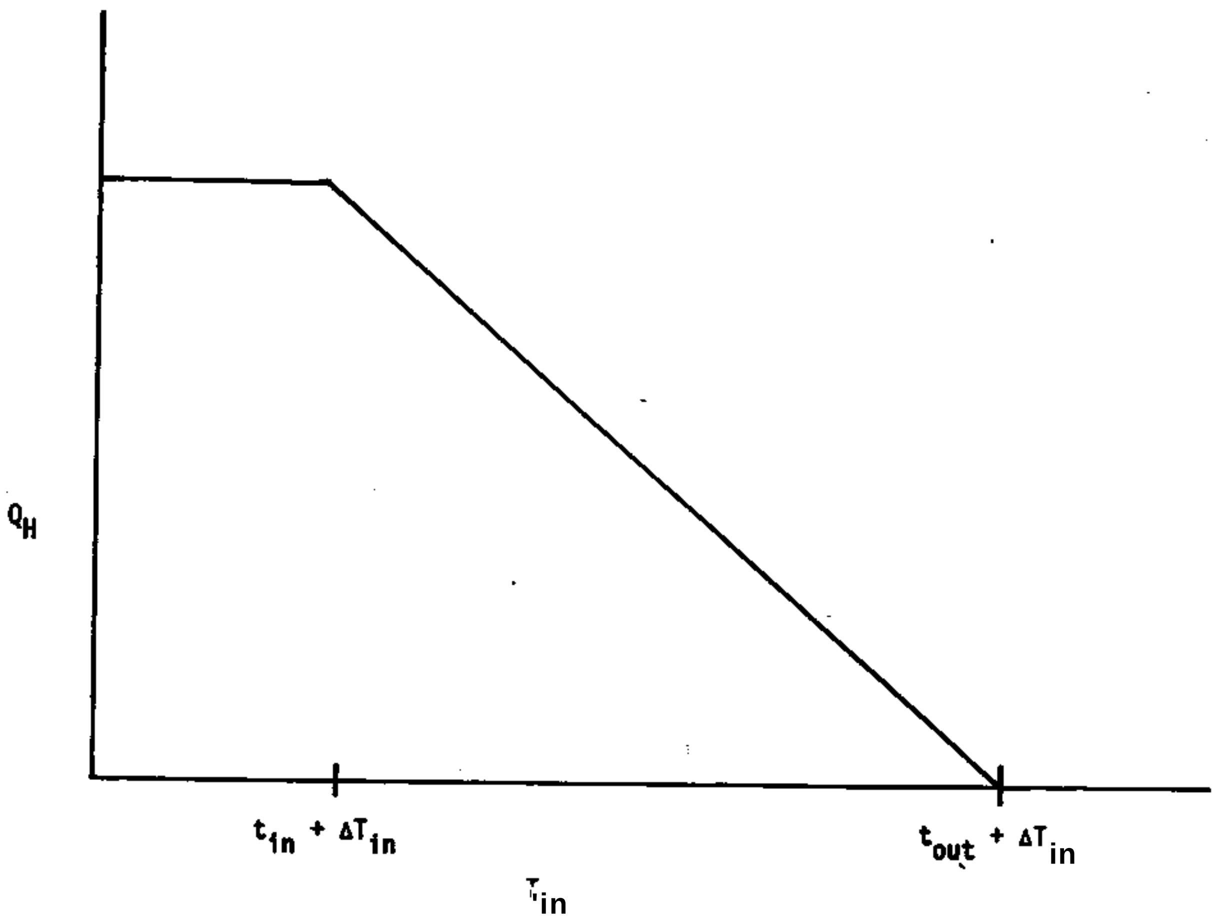


Fig. C.I