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INDIA

Technical Report: Automation of Fertilizer Plant*

Prepared for the Government of India by the
United Nations Industrial Development Organization,
acting as executing agency for the United Nations Development Programme

Based on the work of Dr. Irving Lefkowitz,
expert in Automation of Fertilizer Plant

Backstopping Officer: C. Gürkök, Engineering Industries Branch

United Nations Industrial Development Organization

Vienna

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* This document has not been edited.

A. Summary of activities in performance of mission

1. 17-22 July 1988

Made arrangements for the trip; reviewed literature on ammonia synthesis process and automatic control of ammonia reactor.

2. 26 July 1988

Met with UNIDO officials in New Delhi to take care of administrative matters. Introductory meetings with Dr. Hema Khurana, project leader, and other members of the AAPP staff.

3. 27-29 July 1988

Reviewed literature and current reports to become familiar with (i) the background of the ammonia project, (ii) the derivation of the mathematical model for the ammonia reactor, and (iii) experiences with the computer simulation developed for the process. Participated in discussions with Dr. H. Khurana and Dr. G.S. Varadan to identify project objectives, problem areas, and directions of effort. See Appendix I for the list of references.

4. 1-5 August 1988

Studied the Bhatia model for the ammonia reactor to check its consistency with other models described in the literature, to examine the underlying assumptions incorporated in the model, to determine ways in which the model may be validated against plant operating data, and to suggest ways that the model might be simplified with minimal impact on its effectiveness for optimizing control. Participated in on-going discussions with project principals concerning the model and its application,

the identification of potential problems and some tentative suggestions for solutions.

5. 8 August 1988

Met with Mr. A.B Misra, Operations Manager of the Bombay plant of Rashtriya Chemicals & Fertilizers Ltd. for discussions about the project. Particular objectives included: (i) ascertaining the reasonableness of various assumptions embedded in the reactor model and results of computer simulation runs based on the model, in relationship to plant operating experiences, (ii) identifying the characteristics of disturbance inputs and parameter variations in normal operating practice, (iii) identifying constraints on operating conditions which are not included in the model, and (iv) determining the feasibility of installing in the plant additional sensors deemed necessary for effective optimizing control.

Discussed with Mr. Bisra and Dr. Varadan the possibilities of conducting a special training course for plant personnel on advanced process control and optimization methods. Prepared topical outline for proposed course (see Appendix II).

6. 9-10 August 1988

Developed details of proposals for (i) simplifying the mathematical model to reduce computational effort, (ii) developing an on-line optimizing control algorithm, (iii) implementing the control scheme via a multilayer hierarchical control structure, and (iv) identifying means for on-line updating of the model used in the optimization.

7. 11-12 August 1988

Prepared preliminary report of observations, comments, and recommendations for computer control of the ammonia process. Presented report to Dr. Kharana and discussed findings with her.

8. September 1988

Prepared final report.

B. Introduction

1. Process Description

Ammonia synthesis is a basic component of the fertilizer industry, where ammonia serves both as an end product and as an intermediate in the production of other products (e.g., urea). A schematic diagram of a typical ammonia plant using natural gas feedstock is shown as Fig. 1. The plant consists of the following process units.

Primary Reformer. The hydrocarbon feedstock is reacted with steam in the presence of catalyst at high temperature to produce hydrogen, carbon monoxide and carbon dioxide.

Secondary Reformer. Sufficient air is mixed with the gases exiting the primary reformer to provide the nitrogen needed to achieve the desired three to one hydrogen-nitrogen ratio in the final synthesis gas stream. The oxygen content of the introduced air is used up in reacting with carbon monoxide and unreacted hydrocarbons.

Shift Reactor. The remaining carbon monoxide is catalytically reacted with steam to produce carbon dioxide and hydrogen.

Removal of Carbon Oxides. The product gases are passed through an absorption tower which removes most of the carbon dioxide. Any remaining traces of carbon oxides are removed through a subsequent methanation process.

Compressor. The synthesis gas is compressed to several thousand psi and then mixed with recycle gas. A fraction of the recycle gas is purged to prevent buildup of inerts (methane and argon).

Ammonia Separation. The compressed synthesis gas is refrigerated to condense out the ammonia product.

Ammonia Synthesis. The pressurized gas mixture of hydrogen and nitrogen in near-stoichiometric proportions is passed over a catalyst bed at elevated temperature. The reactor used in the Bombay plant of Rashtriya Chemicals & Fertilizers Ltd. (RCF) is of the Haldor Topsoe design; a schematic of the reactor is shown in Figures 2 and 3. Fig. 2 includes the patent description of the reactor operation; Fig. 3 shows the instrumentation on the Bombay reactor along with some representative reactor temperature readings.

The reactor design provides for radial flow through two catalyst beds with a heat exchanger mounted centrally in one of the beds. The process stream of synthesis gas is obtained by combining, inside the converter, separate feed streams: a shell stream serving to cool the converter shell, an exchange stream serving to cool the central heat exchanger, and a bypass stream for final adjustment of the temperature of the process stream. The process stream passes in succession radially through the first catalyst bed in an inwards direction, through the central heat exchanger for cooling radially, through the second catalyst bed, and finally through a heat exchanger in countercurrent flow to the main inlet flow stream.

2. Control Objectives

The overall productivity of the ammonia plant depends on efficient operation of

each of the process units as well as their effective integration with respect to production of the ammonia product. The ultimate goal is to develop a computer control system that will determine and implement optimum operating conditions throughout the plant to maximize plant objectives within allowable constraints.

The goal of the present project is computer control of the ammonia reactor to maximize yield of its ammonia product. The motivations here are (i) to contribute to overall productivity by improving the operating efficiency of a key process in ammonia production, (ii) to gain experience and expertise in the modeling, optimization, and implementation stages prerequisite to achieving the ultimate goal, on-line computer control of the ammonia plant.

There is a substantial body of literature dealing with the various aspects of the problem: modeling of the ammonia reactor from first principles, computer simulation, process control applied to the reactor, optimization methods and algorithms, and computer control methodology. Appendix I lists the publications studied and/or referenced in the course of this project.

C. Modeling of the Ammonia Reactor

The starting point for the present study was the ammonia reactor model developed by Dr. S.K. Bhatia of the Indian Institute of Technology, Bombay [4]. The model is based on first principles of reaction kinetics, mass and energy balances, etc., and particularized for the reactor presently operating in the RCF Bombay plant. It is reasonably detailed (including the effects of temperature, pressure, and composition on kinetic coefficients, thermodynamic properties, and heat-transfer coefficients), resulting in two nonlinear differential equations representing the reaction kinetics and two nonlinear differential equations representing the heat exchanger energy transfer relationships. The equations are all coupled and hence have to be solved simultaneously. Because of the strong couplings and the nonlinearities, it is difficult to assure convergence, solution accuracy, or that the solution obtained is indeed the desired optimum (among multiple solutions).

A number of important assumptions are embedded in the model derivation; these include the following.

1. The system operates in steady state, i.e., input conditions vary slowly enough that transient effects are negligible.
2. All of the process variables within the catalyst bed (e.g., temperature, flowrate, gas composition) are functions only of catalyst bed radius (i.e., they are uniform axially and with respect to height within the bed).
3. The catalyst activity is constant with time and uniform throughout the catalyst bed.

The Bhatia model seems essentially consistent with other models described in the literature (e.g., those reported by Shah [17-19], Patnaik [15], and Gaines[8], with basically similar assumptions and formulation. There are some differences among the models related to configuration and also some differences in the expressions used for heat capacities, kinetic coefficients, etc.; however, I did not consider these important with respect to the computer simulation or optimization method to be applied.

The model equations developed by Bhatia are contained in his report [4], a copy of which is appended here as Appendix III. The report also presents a method of solving the simultaneous nonlinear differential equations via an iterative computer simulation procedure.

Computer code for the simulation of the ammonia synthesis model was prepared by Bhatia in collaboration with the Bombay plant and, apparently, results of the simulation runs were considered in reasonable agreement with plant experience. Also, apparently, the iterative procedure used for solving the model equations converged satisfactorily under conditions of the tests. However, Dr. Khurana's group in New Delhi, using the same computer program, had difficulties in obtaining convergence and in replicating the results of the Bombay group. See Section D for a further discussion of this problem.

While in general agreement with the theoretical bases for the Bhatia model, I did express concern about the following issues:

1. There is no plant instrumentation for on-line measurements of many important variables associated with the ammonia reactor. The most critical lack are measurements by means of which the reactor flow streams (e.g., main, quench, and bypass flowrates) can be reliably and independently determined. Also, important deficiencies in the instrumentation are the lack of on-line sensors for input and output gas stream composition measurements. This situation poses serious consequences:

- a) Not having direct measurements of the individual flowrates, we have no assurance of steady-state conditions existing within the catalyst beds (i.e., there may occur transients in the flow distribution within the reactor that would violate the steady-state assumptions of the model).

- b) By the same token, we have no way of controlling the flowrates to desired, computer-determined optimal values, limiting the potential effectiveness of any computer control scheme.

- c) Further, the lack of direct knowledge of the flow distribution within the reactor contributes in a major way to the difficulty in solving the model equations. Because of the nonlinear couplings among the equations, the solution procedure requires guessing the flow distribution, solving the equations based on the assumed flow values,

and then iterating the procedure until agreement is reached between assumed and computed values. Obviously, if the flowrates are known *a priori* (through real-time measurements), the computational effort may be considerably reduced.

2. A corollary to Issue 1 above has to do with model validity and the evaluation of model parameters. In theory, there are enough plant measurements (catalyst bed temperatures, synthesis gas flowrate, etc.) to permit the model equations to be solved. However, this assumes that (i) the assumptions underlying the model are valid, (ii) the model itself is correct, and (iii) the parameters associated with the model are accurately determined. In reality, the assumptions are at best only approximately valid. Also, because of the complexity of the reaction kinetics and the fluid flow and heat transfer mechanisms, most of the model parameters can only be evaluated experimentally, i.e., by running tests on the reactor under operating conditions and determining parameter values which produce consistent results with respect to measured plant data. This, however, requires a degree of redundancy in the data; in particular, the amount of redundancy required increases with the number of parameters to be determined.

In the present case, there are several parameters that have to be determined empirically [e.g., catalyst activities, fugacity coefficients, heat transfer coefficients, fouling factor, effectiveness factor, etc. (see equations in Appendix III)]; however, there are insufficient measurements of reactor variables to make these determinations uniquely. This leads to the contention that we have no explicit assurance of the model's validity with respect to its particularization to the Bombay reactor and with respect to its use in the derivation of an optimizing control algorithm. We note, in particular, that with more "adjustable" parameters than measured variables available for correlation (as appears to be the case here), it is possible to choose parameter values so that the model agrees with observations, yet yields incorrect results when used to extrapolate to predicted "optimum" conditions.

3. In view of the above issues and the uncertainties they invoke, it is reasonable to question the degree of detail incorporated in some aspects of the model and the lack of precision in other aspects. For example, fairly exact high-order expressions for molar specific heat and heat of reaction (as functions of temperature and pressure) are used in the model despite the degree of uncertainty with which the temperature distributions through the catalyst beds are known or determinable. On the other hand, the model assumes that catalyst activity is fixed and that temperatures, compositions, etc. vary only as a function of radius in the catalyst bed; these assumptions are at odds with plant observations (as noted, for example, in the table of observed catalyst bed temperatures included in Fig. 3), reported plant experiences with local "hot spots", known tendencies for channeling in catalyst beds, effects of catalyst degradation on catalyst activity, etc.

4. There are a number of benefits of model simplification besides the obvious one of reduced computation time (which is particularly important in this case because of the iterative solution procedure required and because of the objective of on-line implementation). These benefits include better convergence properties, higher (computational) accuracy, and possibly fewer spurious optima. In addition (and this is particularly important with respect to model adaptation), the simplified model is typically more transparent with respect to the dominant cause and effect relationships in the system and, consequently, more amenable to the identification of one or two "tuning" parameters.

D. Computer Simulation of the Reactor Model

Computer simulation, in the sense used here, refers to the coding of the model equations into the computer and their solution to yield the set of output variables for a given set of input conditions, i.e., the sets of manipulated and disturbance inputs. The outputs of interest are the temperatures and compositions of the gas streams exiting the two catalyst beds, the temperatures of the streams exiting the reactor heat exchangers, and the temperature, composition and yield of the product stream exiting the reactor. The manipulated (or control) inputs consist of the flowrates of the input gas flow streams (main, quench and by-pass flows); the set of measurable disturbance inputs include the temperature, pressure, composition, and flowrate of the synthesis gas feed stream to the reactor. We note that, in addition to these measurable inputs, there are a variety of disturbances which are not measurable (in this system) and, hence, cannot be explicitly accounted for in the simulation (e.g., variations in gas flow distribution through the catalyst beds, effects of changing ambient conditions, effects of changing heat transfer coefficients, etc.). Accordingly, these disturbances have to be factored into the simulation in terms of their average effects as estimated from observed input-output data.

Assuming the model is correct (including correctly evaluated parameters), problems may arise in the simulation due to (i) errors in coding, (ii) improper mesh size used in the numerical integration, (iii) convergence to incorrect solutions because of multiple equilibria or ill-conditioning of the equation set due to the nonlinearities in the model, or (iv) lack of convergence (or convergence to spurious equilibria) due to the nature of the iterative procedure used in the solution process.

Unfortunately, the Delhi group did experience difficulties in reproducing Bhatia's simulation results (during the period of my assignment). Since satisfactory results with the simulation had already been reported, my conjecture as to the source of the difficulty centered on factor (i) above, and possible factor (ii). Suggested remedial actions were: (i) a careful checking of the computer program, and (ii) testing the effects of reducing step sizes in the numerical integration. It was pointed out, further, that the complexity of the model contributes substantially to the impact of each of the factors listed above and that, by appropriate simplifications of the model we not only speed up the computation, but may also significantly improve the ability of the simulation to

converge to a useful solution. We noted, in addition, that by coupling the simplifications to the inputting of additional reactor data (temperatures and flow distribution) we may be able to eliminate or at least significantly reduce the need for iterating on the solution process to achieve the overall solution.

Recommendations for Computer Simulation of the Reactor Model

1. I feel that it is absolutely essential that instrumentation be installed on the plant reactor that will permit direct real-time flowrate measurement of the three flow streams in the reactor, i.e., the main, quench and by-pass flows. This will provide the minimum capability for monitoring the flow distribution in the catalyst beds and in the internal heat exchanger.
2. I highly recommend installation of automatic controls on each of the reactor flowstreams in order to (a) enhance the assumption of steady-state operation by maintaining the flowrates at their set-point values, and (b) enable the implementation of supervisory control (i.e., the continual updating of set-points to their computed optimal values without operator intervention).
3. I concur with the proposal of installing a chromatographic analyzer that will permit on-line composition measurements of the feed and product streams. This will provide valuable additional information with which (a) to validate the model, (b) to update key "tuning" parameters of the model and, (c) as a consequence, to provide an additional basis for simplifying the model used in the computation of the optimum operating conditions.
4. The important steps at this stage of the project are: The model and, in particular, the computer code used in the solution of the model equations must be validated. This includes testing for accuracy and convergence properties of the integration scheme being used. It may be necessary, because of the nonlinearities of the model, to carefully explore the effect of the step size used in the integrations, and also the step size used in the iteration sequence of the solution procedure for solving the model equations.
5. With the model validated, a series of computer simulation runs should be conducted to determine the sensitivity of the model output to various simplifying approximations of the model. Specifically, determine the effect on the computed optimum of simplifying the temperature-dependent expressions for specific heat and heat of reaction to (i) constants evaluated at the mean temperature existing in the catalyst bed and (ii) linear functions of temperature with slope and intercept values determined so as to obtain a best fit (in a least-squares sense) to the true relationship over the temperature range existing in the catalyst bed.
6. Carry out a range of sensitivity studies based on the computer simulation of the detailed model to determine (a) sensitivity of the optimum to changes in input

conditions, e.g., feed temperature, composition, pressure, and flowrate, (b) sensitivity of reactor yield to changes in the optimizing control variables (e.g., main, quench and bypass flows), (c) sensitivity to changes in catalyst activities and changes in heat transfer coefficients for the heat exchangers (over the ranges of expected variations).

7. Based on the results obtained in steps 5 and 6 above, determine how the model may be simplified without any significant sacrifice of performance. This requires consideration of how the model is to be updated to fit current operating characteristics of the reactor, i.e., key parameters of the model have to be identified and associated with specific on-line measurements. It is essential, at this stage, to assure that the updating algorithm to be employed converges rapidly to a unique solution and that the resulting model yields a solution to the optimization problem of appropriate accuracy.

8. At this stage, it is not necessary to apply the fastest or most efficient code for the optimization, only a method that will give accurate results in a reasonable time. Because the model (at this stage) is in the form of a set of nonlinear differential equations, I would employ a fast-time simulation to generate output values corresponding to the inputs determined by the optimizer. A nonlinear programming package (based say on GRG2) that makes use of gradient values determined by perturbing the process model (rather than analytically derived) is suggested. Thus, the fast-time simulation provides both output values and gradient information.

E. Optimizing Control Function

As noted earlier, the current project addresses only a subproblem of the overall plant optimization, i.e., maximizing the ammonia yield of reactor process, where the manipulated inputs consist of the quench and bypass flowrates and the exogenous (or disturbance) inputs include the total flow rate, temperature, pressure and composition of the synthesis gas feed stock.

While the physical inputs to be manipulated by the control system are the gas flowrates, there are advantages to considering the gas temperatures at the inlets to the two catalyst beds as the actual decision variables to be determined by the optimizer. Some of the reasons for this choice are:

(i) The computed temperatures can be implemented on the reactor by manipulating the gas flowrates via direct feedback control loops (see Section F, Direct Control Function).

(ii) Computer simulation of the reactor model requires the simultaneous solution of six nonlinear differential equations (two for each catalyst bed and one for each internal heat exchanger) where temperature is the dominant coupling variable. We expect that the iterative process involved in the solution is much more easily carried out when the catalyst bed inlet temperatures are the decision

variables as opposed to the gas flow distribution; this should result in reduced computational effort.

(iii) Closing the loop around inlet temperature serves to minimize the effects of various disturbances which affect the relationships between gas flow and inlet temperature (e.g., effects of feed temperature, pressure and composition, ambient temperature, heat transfer coefficients, etc.).

(iv) From the nature of the model equations, we expect that the sensitivity of performance (i.e., ammonia yield) to changes in inlet temperature (with fixed flow distribution) would be significantly greater than the sensitivity to changes in quench or bypass flowrate (with all other conditions held constant).

(v) Critical constraint conditions leading to "blowout" or "hot-spot" phenomena are temperature related.

The optimizing control function reduces essentially to a computer algorithm that determines optimum values for the inlet temperatures of the two catalyst beds, given the set of current values of the exogenous input variables (e.g., synthesizer gas flowrate, temperature, pressure and composition). There are, however, a number of different approaches to developing the algorithm (see [9], [16], [20]):

(a) Use a method based on variational calculus to derive the conditions for optimum performance. This approach is ruled out, however, because of the complexity of the model equations and the need to invoke the algorithm every time input conditions change.

(b) Use a gradient-based method to locate the optimum where the local gradient is determined by perturbing the simulation model.

(c) Use a method based on successive linearization of the model and the application of linear (or quadratic) programming with step size constraints.

(d) Approximate the mathematical model by a regression-type model wherein the reactor yield is expressed explicitly as a nonlinear (e.g., polynomial) function of the decision and disturbance variables.

We tend to favor the second approach because (i) the search process is only two-dimensional, hence the gradient determination is relatively easy, (ii) the essential nonlinearities in the model equations would tend to make the linearization procedure of approach (c) above more difficult and would probably require relatively small step sizes, therefore increasing the number of iterations required to seek the optimum, and (iii) the regression model approach may introduce spurious optima, there may be a fair amount of computational effort required to determine the coefficients of the regression model, and the regression model may not lend itself to direct correlation with plant data for model updating.

The gradient approach is particularly effective in the optimizing control application because, most of the time, the process is operating in the neighborhood of a previously computed optimum which is to be updated to correct for small changes in exogenous inputs that have occurred since the last update. Thus, at the start of each update, current values of the measured process input variables are inputted to the simulation model. The local gradient is estimated by making successive small step changes in each catalyst bed inlet temperature and computing the ratio of the resulting change in reactor yield to the change in the inlet temperature.

There are a number of well-known strategies that may be applied to determining the direction and extent of movement of the decision variables relative to the gradient direction (see [9], for example); this establishes a new test point at which the gradient estimation and movement are repeated. The process is iterated until the stopping criterion used is satisfied. The choices of strategy and stopping criterion depend on the characteristics of the performance contours, e.g., the degree of regularity of the contour lines, the presence of ridges, etc. Also important is the nature of the constraints that affect the search process and the location of the optimum.

All of the above methods are based on a computer simulation of the mathematical model of the reactor. An alternative approach uses, in effect, the physical reactor itself as the model. Specifically, the local gradient is estimated by perturbing the physical inputs to the reactor and measuring the resulting changes in ammonia yield. This approach has the obvious advantage of not requiring a mathematical model but suffers the same disadvantages as do direct hill climbing methods in general; these include: (i) direct perturbation of plant inputs is usually not favored by production personnel, (ii) real-time measurements of the objective function (e.g., yield) are often not available or are seriously corrupted by noise, (iii) dynamic lags in the process introduce errors in the gradient measurement or may require excessive time to achieve steady-state conditions, (iv) the gradient estimates may be further corrupted by the effects of various miscellaneous disturbances entering the system.

Recommendations for the Optimizing Control Function

1. Run a series of computer simulation studies to confirm (or deny) the assumption that it is better computationally to iterate on the catalyst inlet temperatures as opposed to inlet gas flowrates. Modify the suggested procedure if the result is negative. Use the results also to examine the assumption that the sensitivity of reactor yield to inlet temperature is much greater than that to gas inlet flowrate.
2. Run a series of computer simulation studies to map performance contours in the space of the decision variables for various sets of conditions on the exogenous inputs. Use the maps as a basis for choosing an appropriate gradient search strategy.

3. Run a series of input-output studies on the simulation model to map the output variables associated with each catalyst bed (i.e., exit temperature and composition) as functions of input conditions (i.e., flowrate, temperature, composition, and pressure). Determine from the characteristics of these maps (e.g., degree of curvature) the feasibility of replacing the differential equations in the model by a set of (algebraic) regression models relating outputs to inputs. If the results warrant it, develop, by least-squares fitting to the computer simulation model, a regression model relating reactor yield to the input variables. Run tests to evaluate the effectiveness (in terms of accuracy, speed, robustness) of this model as the means for deriving the optimizing control algorithm compared with the gradient-based approach described above.

4. Determine, via further discussions with plant people and literature references, what constraints are to be imposed on the optimization. These constraints may be required or useful for various reasons:

(i) to reflect physical realities, e.g., minimum and maximum flowrates determined by valve and pump capacities, and internal variables which are required to be non-negative by definition.

(ii) to reflect operating constraints on the plant which, if violated, would result in unacceptable plant operating conditions. Examples include avoiding the conditions for reactor "blowout", preventing the occurrence of "hot spots" which might damage the catalyst, etc.

(iii) to introduce "artificial" constraints whose purpose is to avoid spurious optima, to assure convergence to the desired solution, and to reduce the number of iterations required by restricting the space of allowable solutions. For example, we may determine from operating experiences and from simulation runs that reactor temperatures must lie within a limited region for acceptable operation of the plant. Another purpose served by these "artificial" constraints is to define the regions of state and operating conditions for which the model being employed is valid. This may be particularly important when using regression approximations to the model or averaged values for temperature and

On the basis of the above experimental studies and the nature of the constraints to be imposed, decide on the design of the optimizing control algorithm to be installed.

F. Hierarchical Computer Control

A three-layer (vertical) control hierarchy is recommended for implementation of the computer control system. The hierarchy is shown in Fig. 4 [12 - 14] where the following control functions are identified:

First Layer: Direct Control

Second Layer: Optimizing/Supervisory Control

Third Layer: Adaptive Control

Note that typically these layers correspond to an ordering of time scales, with the direct control algorithms operating at the highest frequency and the adaptive layer being called into play at the lowest frequency.

1. Optimizing Control

The optimizing control described in the preceding section should operate in a supervisory mode, i.e., the output of the optimization procedure determines the set-points for direct controllers that manipulate the degrees of freedom available for implementing the optimum conditions. Based on discussions with plant personnel concerning current operating practices, information in the literature and conclusions drawn from the nature of the reactor model, we have suggested that the optimizer generate the set-point for the inlet gas temperature for each of the two catalyst beds with the outputs of the two temperature controllers determining the by-pass and quench flowrates, respectively.

2. Direct Control

Since it is important that the reactor be maintained as close to steady state as possible, and because there are dynamic lags in the system and significant interactions among the three gas flowstreams, we recommend a cascade control loop for each of the temperature controllers; i.e., the outputs of the temperature controllers for the first and second catalyst beds provide the set points for flow controllers controlling the by-pass and the quench flowrates, respectively, as shown in Fig. 5.

There will be significant couplings between the two temperature control loops because (i) changes in either the quench or bypass flowrates result in corresponding changes in the main flowrate, affecting the flow distributions in both catalyst beds, and (ii) thermal feedbacks induced by the two internal heat exchangers affect both inlet gas temperatures. These interactions may be important enough that, at some later stage, we would want to take them into account through a suitably designed decoupling (or noninteracting) control algorithm. The implementation of such an algorithm is relatively easy if the direct control functions are realized through microprocessor-based control hardware. (This part of the direct control system is shown dotted in Fig. 5).

Design of the direct control algorithms depends on some knowledge of the dynamics relating inputs to outputs, the relative gains, and the cross-couplings in the process. This requires a series of transient tests on the plant reactor to identify linearized transfer functions and the effects of gross changes in operating conditions on these transfer functions (this information may already be available from plant personnel).

3. Adaptive Control

As indicated earlier, an essential element in the proposed scheme is the ability to update certain key parameters of the model by use of available process measurements (e.g., outlet temperatures of the catalyst beds). The effectiveness of this approach depends on the amount of redundant information available.

Simulation studies should be carried out to identify appropriate tuning parameters and the measured variables to be used for the updating. Sensitivity studies should be carried out to ascertain the ranges of operating space for which a given parameter update will yield acceptable results in the use of the model. The outcome of these studies may lead to ways of further simplifying the model or, conversely, may point to where a more accurate model of the effect of variations of some variable (say temperature) on the overall optimization results may be needed.

An additional independent check on the effectiveness of the model may be obtained by direct measurement of overall performance, i.e., the actual rate of ammonia production. However, this information may have to be used statistically (i.e., long-term averaging) because of dynamic lags and variations in the material holdup in the system. Another check on the model is an overall energy balance on the ammonia reactor, where the rate of ammonia production is computed by measuring the rate of thermal energy generation in the reactor.

The adaptive control function is predicated on the assumption that the mathematical model is incomplete and/or only approximates the behavior of the system and that it may be made acceptably accurate in the current operating region by proper "tuning" of certain model parameters. These tuning parameters provide the mechanism for adjusting the model so that computed values (in say a least-squares sense) agree with measured variables of the process. In effect, the adjustable parameters embody the effects of all the simplifying approximations incorporated in the model. This, of course, implies redundancy of information, i.e., real-time measurements of critical process variables which serve to "calibrate" the model.

Over time, as operating conditions change (e.g., a major change in synthesis gas flowrate or composition), or as certain plant parameters change (e.g., due to catalyst degradation or scaling of heat transfer surfaces), the model no longer tracks adequately and it is necessary to readjust the model parameters, i.e., to reinitiate action by the adaptive control algorithm. The underlying assumptions here are (i) sufficient data is available from the reactor to carry out a model update, (ii) the updating algorithm converges to correct parameter values quickly enough, and (iii) drift of the model with respect to its description of plant behavior (i.e., the required frequency of adaptive action) is relatively slow.

Recommendations for Hierarchical Computer Control

1. It is recommended that the overall control scheme be implemented in a step-by-step manner after completion of the computer simulation studies outlined in preceding sections and after achieving a feasible procedure for computing the necessary conditions for optimum ammonia yield for the reactor.
2. Prior to plant testing of the optimizing control, the means for controlling the catalyst bed inlet gas temperatures must be installed. This includes feedback controls on the two temperatures, and measurement and control of the bypass and quench flowrates, as previously mentioned. It may not be necessary to incorporate, at this early stage, the cascade and noninteracting control features – this will depend on how well the temperatures are maintained at their set point values and also how much variation is induced in the gas flowrates due to internal transients.
3. There should then be a period of limited plant testing of the optimizing control scheme wherein the computer operates only in an advisory mode, with the operator implementing the set-point changes called for. An essential ingredient of this testing phase is accurate and dependable logging of optimizer outputs and operator responses, with the operator documenting his reasons for deviating from the computer recommendations.
4. Somewhat concurrent with the plant testing of the optimizing scheme there should be some preliminary testing of the model updating scheme to be employed. Although the adaptation algorithm will most likely be implemented in an off-line batch mode for some time before it is put online, it is important that the optimization tests be based on a properly tuned model. Thus, data collected during this initial testing phase should permit an analysis of (i) the accuracy with which the model tracks actual plant variables, (ii) the effectiveness of the parameter updating scheme and the identification of potential problems due to noisy measurements, measurement lags, or ill-conditioning of the least squares algorithm (because of the nature of the data), and (iii) the overall performance of the optimizer, i.e., an assessment of the economic improvement achieved.

G. Computer Solution of Model Equations and Some Ideas on Real-Time Implementation

This section is based on the model presented by Bhatia [4], reproduced as Appendix III.

1. Notation: (Refer to Figures 6 and 7).

Process Unit 1: First catalyst bed.

Process Unit 2: Second catalyst bed.

Process Unit 3: Bottom heat exchanger.

Process Unit 4: Central heat exchanger.

M_i, M_i' Total molal flowrate entering and leaving the i th process unit, respectively, $i = 1, 2, 3, 4$.

T_i, T_i' Temperature of gas stream entering and leaving the i th process unit, respectively,
 $i = 1, 2, 3, 4$.

y_i, y_i' Composition vector of gas stream entering and leaving the i th process unit,
 $i = 1, 2, 3, 4$.

m_i, m_i' Flowrate of product stream entering and leaving, respectively, the i th process unit
(heat exchanger), $i = 3, 4$.

t_i, t_i' Temperature of product stream entering and leaving, respectively, the i th process
unit (heat exchanger), $i = 3, 4$.

H_i, H_i' Enthalpy of gas stream at pressure P and temperatures T_i, T_i' , and compositions
 y_i, y_i' , respectively, $i = 1, 2, 3, 4, F$.

X_i' Conversion of hydrogen in gas leaving the i th catalyst bed.

P Pressure of the synthesis gas entering the reactor.

C_p Specific heat of gas stream (function of temperature, pressure, and composition).

2. Catalyst Beds

Bhatia's equations (2) - (7) (see Appendix III) imply the following functional relationships:

$$X_i' = f_i (M_i, T_i, y_i, P; a_i) \quad i = 1, 2 \quad (a)$$

$$T_i' = g_i (M_i, T_i, y_i, P; a_i) \quad i = 1, 2 \quad (b)$$

$$M_i' = h_i (M_i, X_i', y_i) \quad i = 1, 2 \quad (c)$$

$$y_i = k_i (X_i', y_i) \quad i = 1, 2 \quad (d)$$

Notes: Equations (a) and (b) represent the simultaneous solution of Bhatia's differential equations (3) and (4), with the initial conditions determined from the flowrate, composition, and temperature of the gas stream entering the catalyst bed. Equations (c) and (d) are based on the material balances of Bhatia's equation (2). The parameter a_i denotes a vector of parameters associated with the i^{th} catalyst bed, including catalyst activity and fugacity coefficients, which are assumed to be constant or changing only slowly with time.

3. Heat Exchangers

Bhatia's differential equations for the heat exchanger [equations (8) through (12)] imply the following relationships:

$$T_i' = g_i(T_i, t_i, M_i, m_i, y_F, y_2', P; U_i) \quad i = 3, 4 \quad (e)$$

$$t_i' = h_i(T_i, t_i, M_i, m_i, y_F, y_2', P; U_i) \quad i = 3, 4 \quad (f)$$

Notes: Equations (e) and (f) represent the simultaneous solution of differential equations (8) and (9) with initial conditions determined by temperatures of the inlet flowstreams. We have considered here two heat exchangers, the bottom shell and tube exchanger and the central double pipe exchanger. A third exchanger unit could be added without difficulty to represent the heat loss to the surroundings, if necessary. U_i denotes the effective overall heat transfer coefficient determined from Equations (10) - (12). These are algebraic equations involving terms which are functions of temperature, pressure, compositions, and mass flowrate.

Aggregating Equations (10) - (12), we may write

$$1/U_i = 1/h_{si} + 1/h_{ti} + 1/\epsilon_i \quad i = 3, 4 \quad (g)$$

$$h_{si} = a_i Q_i(\bar{T}_i, y_F, P) * M_i^{0.6} \quad i = 3, 4 \quad (h)$$

$$h_{ti} = b_i q_i(\bar{t}_i, y_2', P) * m_i^{0.8} \quad i = 3, 4 \quad (i)$$

where U_i , h_{si} , h_{ti} denote respectively the overall, shell side, and tube side heat transfer coefficients and a_i , b_i imbed the constant parameters of the original expressions; q_i , Q_i are functions denoting the effects of temperature, pressure and composition on the thermal conductivity, specific heat and viscosity of the fluids, and \bar{T}_i , \bar{t}_i denote the average temperatures of the gas stream on each side of the exchanger, respectively; ϵ incorporates the (slowly) time-varying component of the heat transfer coefficient (e.g., the effect of fouling).

4. Interconnections

Referring to the block diagram in Figure 7 and invoking straightforward material and energy balances, we have the following equivalents and algebraic relationships:

$$M_1 = M_4' = M_4 = M_A + M_C = M_F - M_B \quad (j)$$

$$M_2 = M_1' + M_B \quad (k)$$

$$M_3 = M_3' = M_A$$

$$m_4 = m_4' = m_3 = m_3' = M_P = M_2'$$

$$T_4' = T_1$$

$$T_3 = T_F$$

$$t_4 = T_2'$$

$$t_3 = t_4'$$

$$T_P = t_3'$$

$$y_P = y_3' = y_3 = y_4' = y_4 = y_2'$$

$$Y_3 = Y_3' = Y_4 = Y_4' = y_1 = y_F$$

$$M_2 y_2 - M_B y_F - M_1' y_1' = 0 \quad (l)$$

$$M_2 H_2 - M_B H_F - M_1' H_1' = 0 \quad (m)$$

$$(M_F - M_B) H_4 - M_A H_3' - (M_F - M_B - M_A) H_F = 0 \quad (n)$$

Note that the enthalpies H_i, H_i' are determined from the specific heat of the gas stream indexed by the subscript, i.e.,

$$H_i = C_p(T_i, y_i, P)$$

$$H_i' = C_p(T_i', y_i', P)$$

5. Solution Procedure

Since the model involves coupled pairs of nonlinear differential equations plus several nonlinear algebraic equations, a solution for any given set of input and control conditions requires an initial estimate of one of the state variables followed by an iterative procedure to converge on the solution.

Bhatia describes an effective solution procedure in Appendix III (pages 6, 7). An alternative procedure that might be worth exploring is outlined below.

- (1) Measure input conditions: M_F, y_F, T_F, P .
- (2) Select control variables (setpoints): T_1, T_2 .
- (3) Assume a value for M_B .
- (4) $(M_F, M_B) \text{ ----> } M_1$ [via Eq. (j)]
- (5) $(M_1, T_1, y_F, P) \text{ ----> } X_1'$ [via Eq. (a)]
- (6) $(M_1, T_1, y_F, P) \text{ ----> } T_1'$ [via Eq. (b)]
- (7) $(M_1, X_1', y_F) \text{ ----> } M_1'$ [via Eq. (c)]

- (8) $(X_1', y_F) \rightarrow y_1'$ [via Eq. (d)]
- (9) $(M_1', M_B) \rightarrow M_2$ [via Eq. (k)]
- (10) $(M_2, M_B, M_1', y_1', y_F) \rightarrow y_2$ [via Eq. (l)]
- (11) $(M_2, M_1', T_2, T_F, T_1', y_F, y_2, y_1') \rightarrow M_B$ [via Eq. (m)]
- (12) Revise M_B in step (3) and iterate until values in steps (3) and (11) agree.
- (13) $(M_2, T_2, y_2, P) \rightarrow X_2'$ [via Eq. (a)]
- (14) $(M_2, T_2, y_2, P) \rightarrow T_2' = t_4$ [via Eq. (b)]
- (15) $(M_2, X_2', y_2) \rightarrow M_2' = m_3 = m_4 = M_P$ [via Eq. (c)]
- (16) $(X_2', y_2) \rightarrow y_2' = y_3 = y_4 = y_P$ [via Eq. (d)]
- (17) $T_4' = T_1; M_4 = M_F - M_B$
- (18) $(T_4', t_4, M_4, m_4, y_F, y_2') \rightarrow T_4$ [via Eq. (e)]
- (19) $(T_4, t_4, M_4, m_4, y_F, y_2') \rightarrow t_4' = t_3$ [via Eq. (f)]
- (20) Assume a value for M_A , $M_1 = M_A$
- (21) $(T_3, t_3, M_3, m_3, y_F, y_2') \rightarrow T_3'$ [via Eq. (e)]
- (22) $(T_4, T_3', T_F, M_F, M_B, y_F) \rightarrow M_A$ [via Eq. (n)]
- (23) Revise M_A in Step (20) and iterate until values in Steps (20) and (22) agree.
- (24) $(T_3, t_3, M_3, m_3, y_F, y_2') \rightarrow t_3' = T_P'$ [via Eq. (f)]
- (25) Compute yield = $M_P \cdot y_{P3}$
(y_{P3} = mol fraction of ammonia in the product.)
- (26) Update values of T_1 and T_2 according to the optimization method used. Return to Step (2). Iterate until yield is maximized.
- (27) Periodically return to Step (1) to update input conditions.

Three expected advantages of this procedure are: (i) the variables fixed in the solution process are T_1 and T_2 , which correspond to the control variables proposed for the optimizing control (i.e., the set point values to be implemented via cascade control on the quench and bypass flowrates), (ii) the iterative solution process follows closely the actual way that the setpoints are proposed to be implemented on the physical process (i.e., by varying M_B and M_A , respectively), and (iii) there are two iteration loops; the first loop involves only one pair of differential equations while the second loop

involves only algebraic equations. Thus, this should require much less computational effort than Bhatia's method, which involves all three sets of differential equations within a single iteration loop. It is expected that the new method will converge at least as fast as Bhatia's method, but this is something that would have to be verified.

6. Regression Model Approximation

Although the above modification of the solution procedure may result in significant reduction of computational effort, the basic difficulty with the model, that of iterating through four pairs of coupled nonlinear differential equations, still remains. This problem is removed by replacing the pair of differential equations associated with each process unit by a pair of regression equations expressing the unit's output variables as functions of its input variables. In effect, the functions f , g , h denoted in Equations (a) - (f), above, are developed as empirical expressions of their indicated arguments. The coefficients of the terms in these expressions may be determined by standard regression methods, with the "true" data provided by a series of computer simulation runs based on the exact differential equation models. Note that, for this purpose, each process unit may be treated separately. In addition, the nonlinear expressions for reaction rate, fugacities, equilibrium constant, etc. [see Equations (5) - (7) of Appendix III] may be aggregated into the final regression expressions for Equations (a) and (b). In like manner, Equations (10) - (12) of Appendix III [or Equations (g) - (i) above] may be aggregated into the final regression expressions for Equations (e) and (f).

The obvious advantage of the above approximations is reduction in computational time (or cost)— a very important consideration in the online optimizing control application. The major disadvantage is the possibility of introducing significant errors in the determination of the optimum because of the approximations (which may be exacerbated by the need for simultaneous solution of the several approximate functions). Another concern here is the introduction of false extrema because of the nature of the approximating functions. We believe that it is worth exploring this approach and determining, by a series of computer simulation runs, its feasibility for the current application.

7. Updating of the Model

As already noted, the parameters a_i , b_i , and ϵ_i in Equations (a) - (i) above are assumed to represent those attributes of the model that (i) change relatively slowly with time and (ii) are more easily and/or more accurately determined empirically from operating data. We assume further that we can do a considerable amount of aggregation of these attributes so that, in effect, the parameters are reduced to scalars or, at worst, vectors of very low dimensions (the rationale behind this has already been alluded to in Section C above).

In the model adaptation procedure, we assume that we have sufficient online measurements of temperatures and flowrate so that the arbitrary parameters can be evaluated by (say) least squares fitting of the model equations to the observed data [for example, the parameter a_1 can be determined by least squares fitting of Equations (a) and (b) to temperature and flowrate data obtained from Catalyst Bed 1]. Note that since the model at this stage is a simplified approximation to the true relationships, and also that the data will undoubtedly be subject to considerable corruption by noise and process variabilities, it is necessary to fit the model over a large number of data points in order that we may get useful results (hence also the assumption that the model parameters change only slowly with time). It is necessary to assume, of course, that the updating can be carried out with sufficient frequency so that the model tracks process behavior as operating conditions or process parameters change over time.

If a regression model is adopted for the online optimization, a two-stage adaptive procedure may be considered:

Stage 1: The parameters of the computer simulation model are updated as described above.

Stage 2: The parameters of the regression model are updated based on data obtained from the simulation model. It is assumed that the required period for Stage 2 is much smaller than that for Stage 1.

H. Further Observations

1. The handling of the "hot spot" problem is an issue still to be resolved. It surely needs further study to (i) ascertain the source or contributing factors to hot spot formation, (ii) to determine compensating actions to eliminate the hot spot, (iii) to determine the frequency of occurrence of the problem, (iv) to ascertain the damage caused by the hot spot in economic terms in order to judge the extent to which the problem has to be incorporated into the final optimizing control scheme, and (v) to determine how closely a hot spot can be located in the catalyst bed, given only a finite number of temperature sensors in the bed.

If the problem is sufficiently important, an approach that might be considered is to divide each catalyst bed into a number of sectors, each corresponding to a sensor location. Then the reactor model is replicated for each sector and solved to satisfy the measured conditions (i.e., temperatures) for that sector. This may provide two benefits: (i) provide information concerning the location and severity of a hot spot and hence the corrective action to be employed, and (ii) permit incorporating into the model solution the effects of different catalyst activities for different regions of the catalyst bed.

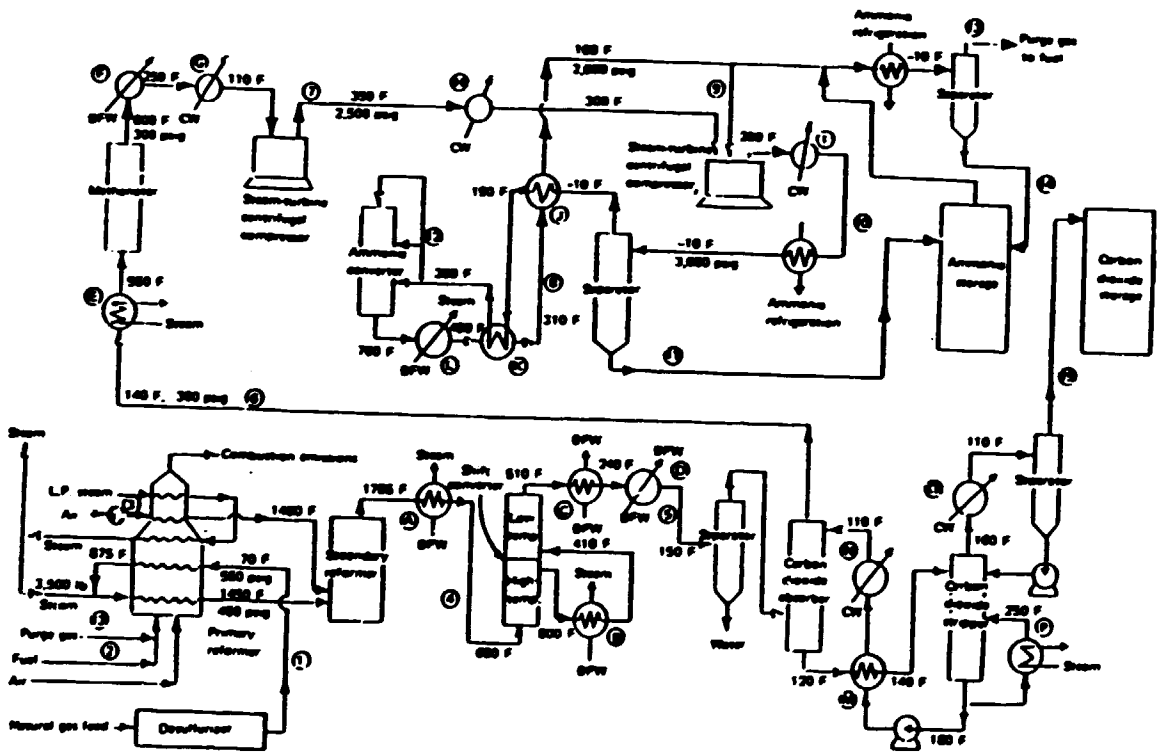
2. An alternative to the model-based optimization scheme described in this report is available if we can reliably compute the rate of ammonia production from on-line

temperature and flow measurements on the inlet and outlet streams. We can use the process itself as the model and employ a gradient search or hill climbing scheme directly on the process manipulated inputs (or the set-points of the associated direct control loops). The advantage of this approach is that we eliminate the need for the complex mathematical model; the disadvantages include dynamic lags in the measurements, noise, excessive perturbation of the inputs to generate gradient directions, etc. (see also Section E).

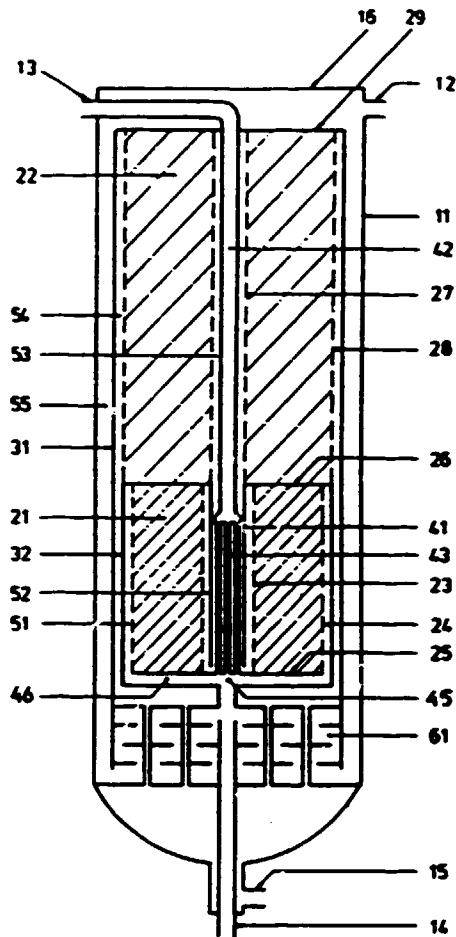
Because of the limitations of the direct hill climbing approach, I really don't believe it holds much promise for the present application. However, it seems that overall mass and energy balance relationships should provide very useful additional data for longer-term updating of the models as well as for monitoring the effectiveness of the model in the optimizing control.

3. The next step in successful on-line implementation of the reactor optimizing control is to incorporate it as a subsystem of the larger system including reformers, compressors, separation columns, etc., with each major unit provided with a local control hierarchy as developed for the reactor. We may then incorporate the subsystems into a multilevel (horizontal) hierarchy with the local optimizing controllers coordinated through a supernal unit that reflects overall objectives and overall constraints associated with the production facility.

Figure 1 Process for Producing Ammonia



Source: DOE/TIC-11118



Source: U.S. Patent 4,181,701

An exchange stream of synthesis gas of 191,450 Nm³/hr, having a temperature of about 120°C, is introduced through the inlet 13 and via the central transfer pipe 42 passes through the central heat exchanger 41. Here, the exchange stream serves to cool the process stream passed from the first catalyst bed 21 to the second catalyst bed 22. The exchange stream leaves the central heat exchanger at a temperature close to the reaction temperature and is combined with the shell stream from the bottom heat exchanger 61.

A bypass stream of synthesis gas of 40,000 Nm³/hr, having a temperature of about 120°C, is introduced through the inlet 14. At a position along the common axis of the two catalyst beds, the bypass stream combines with the shell stream and the exchange stream to form a process stream of synthesis gas of 382,930 Nm³/hr, having a temperature of 360°C. The relative amounts of the three streams which form the process stream can be adjusted during operation to obtain the desired temperature at the inlet to the first catalyst bed 21.

Via the annular space 51, the process stream is passed through the first catalyst bed 21, where its temperature is increased to 520°C because of the exothermic reaction, which causes the ammonia concentration to increase from 3.5 to 14.4 vol %. While subsequently passing through the central heat exchanger 41, the process stream is cooled to 390°C and via the annular space 53 passed through the second catalyst bed 22 under heating to 472°C, while the ammonia concentration increases to 20.8 vol %. The product stream of synthesis gas is then received in the annular space 55 and passed through the bottom heat exchanger 61 for cooling and thereafter through the outlet 15 at about 360°C.

Figure 2. Reactor Diagram with Patent Description.

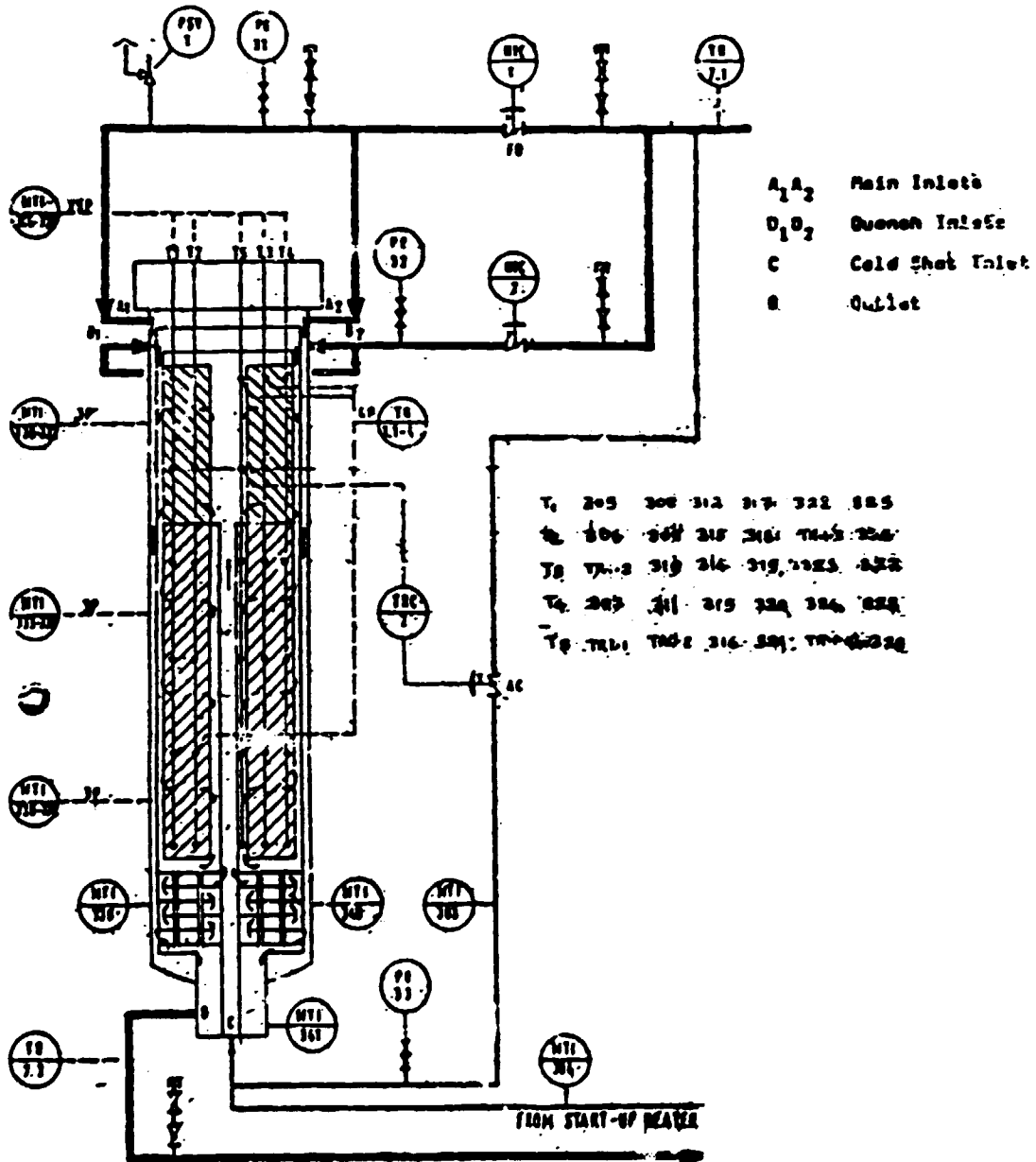
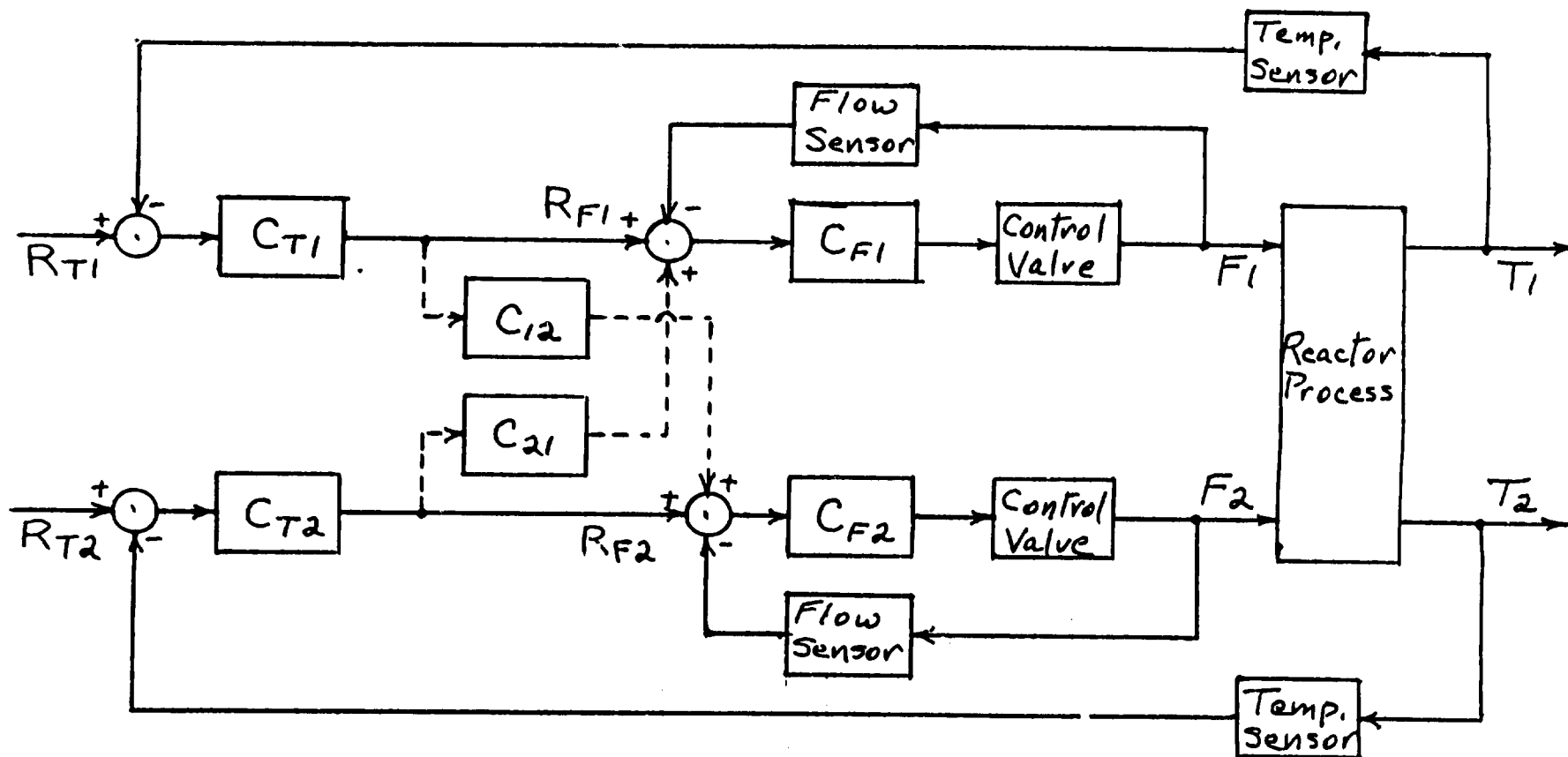


Figure 3. Reactor Diagram with Temperature Data.

Figure 4: Block Diagram of Direct Controllers for Gas Inlet Temperature.



- T_1, T_2 Inlet gas temperature, 1st and 2nd catalyst beds, respectively.
- F_1, F_2 Bypass and quench flows, respectively.
- R_{T1}, R_{T2} Gas temperature setpoints.
- R_{F1}, R_{F2} Gas flowrate setpoints
- C_{T1}, C_{T2} Temperature controllers
- C_{F1}, C_{F2} Flow controllers
- C_{12}, C_{21} Noninteracting control algorithms

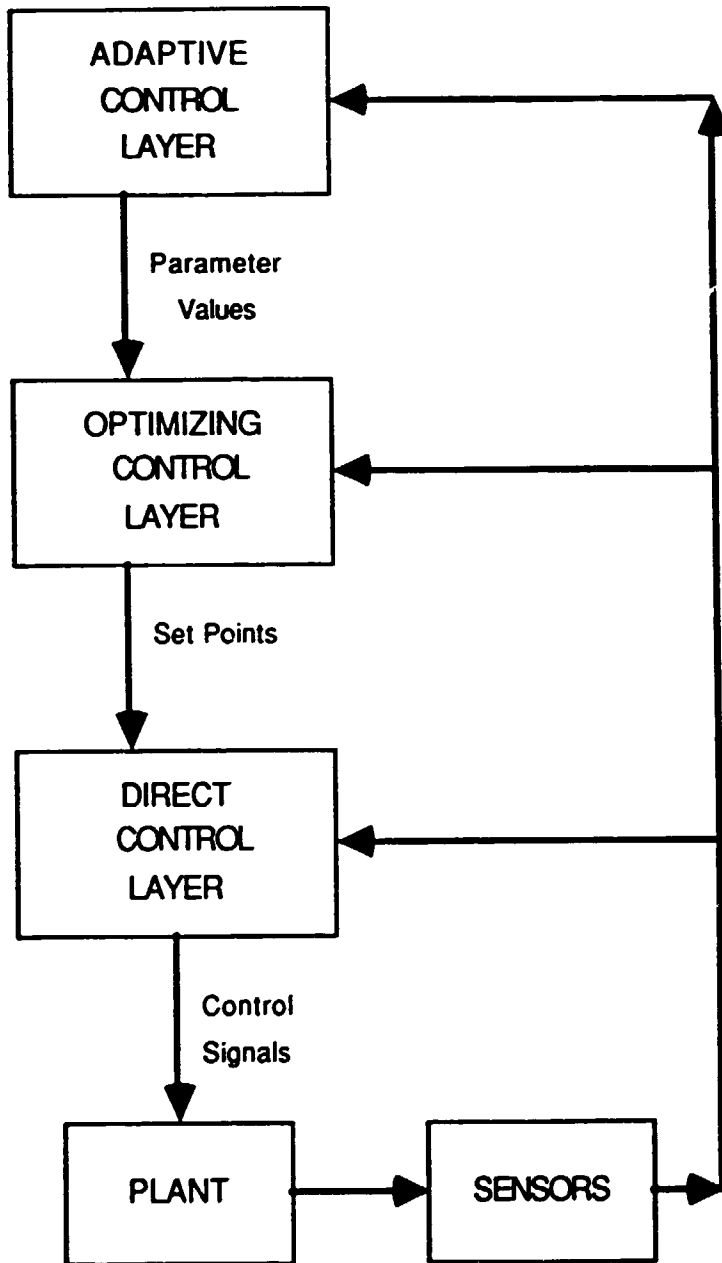


Figure 5 Multilayer Control Hierarchy

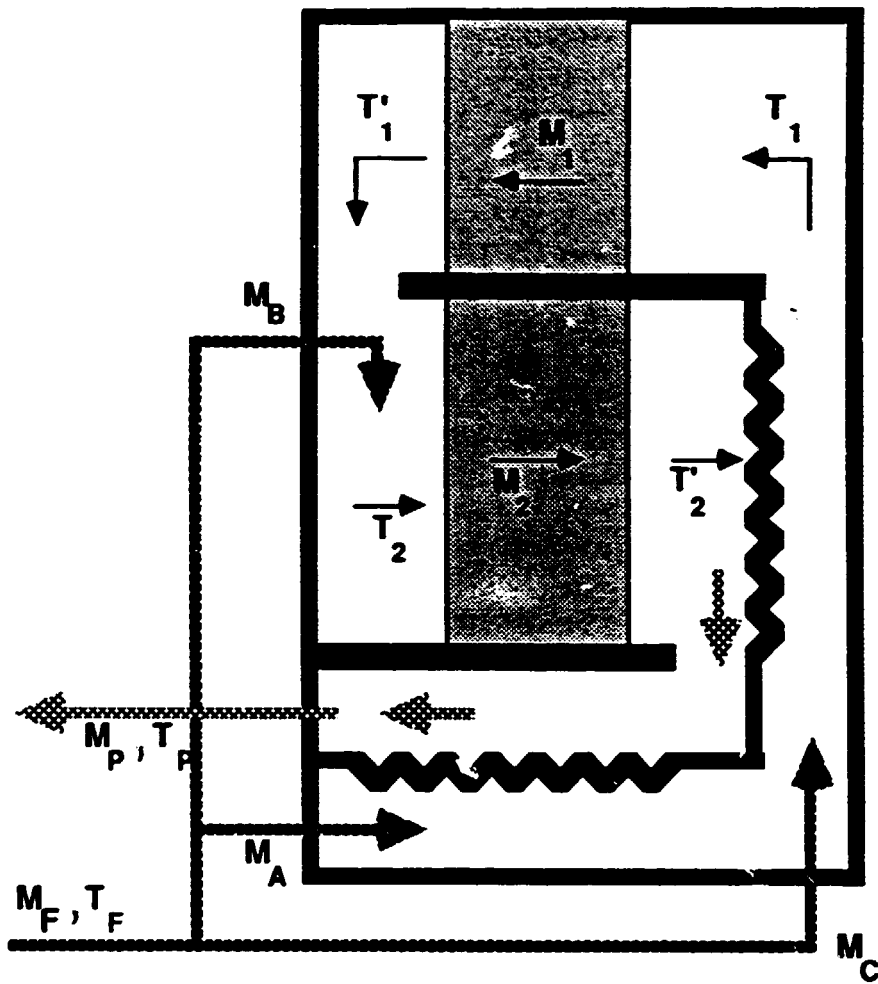


Figure.6 Simplified Schematic of Ammonia Reactor

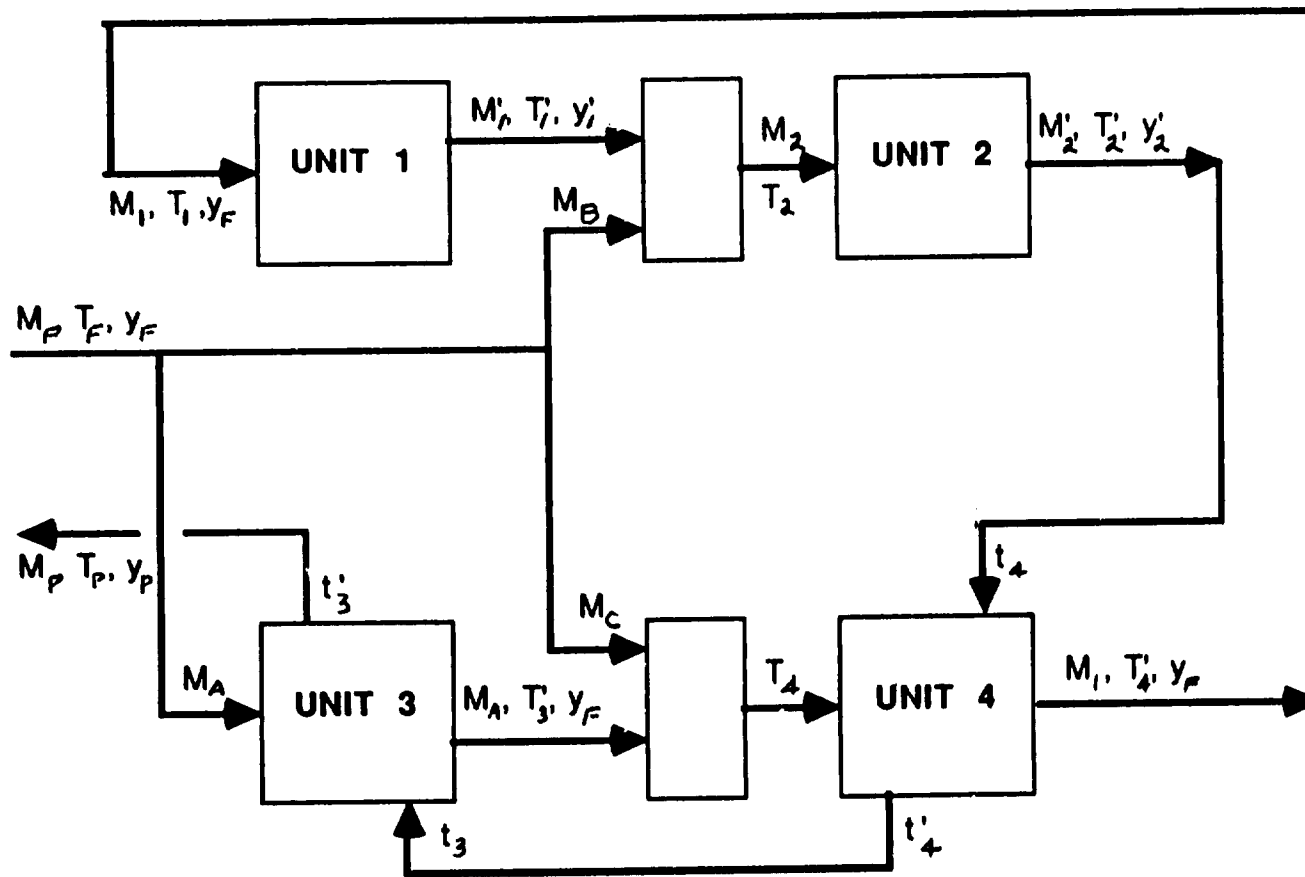


Figure 7 Flow Diagram for Reactor Relationships

APPENDIX I

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APPENDIX II

Training Modules

- A. Modelling and simulation
 - 1. Application of first principles
 - 2. Empirical methods
 - 3. Sensitivity analysis/approximation methods
 - 4. Computer simulation language/methods
 - 5. Measurement of process variables
- B. Dynamic systems and direct control
 - 1. Linear systems and linearization
 - 2. Input-output relationships/transfer functions
 - 3. Transient response characterisation
 - 4. Frequency domain characterization
 - 5. Closed-loop relationships
 - 6. Feedback control systems: algorithms, multivariable/multiloop considerations, feed forward compensation
 - 7. Design methods/performance criteria
- C. Supervisory control/higher level functions
 - 1. Optimization methods/math. programming, scheduling algorithms, etc.
 - 2. Hierarchical control structures
 - 3. Adaptive control methods-model updating
- D. Discrete event systems
 - 1. Model formulation and characterization
 - 2. Discrete event simulation languages
 - 3. Programmable logic controllers
- E. Computer implementation
 - 1. Data acquisition and storage
 - 2. Data base management
 - 3. Real-time aspects of computer control
 - 4. Discrete time algorithms/advanced control
 - 5. Microprocessor based distributed system
 - 6. Communications
- F. Selected case studies.

SIMULATION OF AN AMMONIA SYNTHESIS CONVERTER USING A MICROCOMPUTER

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ABSTRACT - A simulation study is reported for a modern radial flow ammonia synthesis converter. Model equations are formulated for the various reaction and heat exchange zones of the reactor, and solved on a microcomputer. The resulting O.D.E's are solved using a fourth order Runge-Kutta Method while a modified Newton-Raphson technique is used for convergence promotion. The model is used to identify stable regions as well as the optimum condition of operation.

INTRODUCTION AND BACKGROUND

The need for optimization of ammonia synthesis has long been apparent because of the significant increases in production rate that may be achieved with small improvements in conversion. In addition, because the reactors operate close to the stability limit it is important that adequate control systems be provided for trouble-free operation while continuously optimizing the performance. Instability in ammonia synthesis reactors arises out of their autothermal nature (comprising of internal feed-product heat exchange); and under conditions of high flow rate and low inlet temperature insufficient heat may be generated to bring the feed gas to reaction temperature, resulting in blowout [1]. Because the conditions for maximum production are only narrowly removed from the blowout condition it is imperative that adequate control be provided if optimal operation is desired. However, in order to design and implement appropriate on-line control systems it is necessary to have a mathematical model of the system so that its response to changes may be predicted. In addition, such a model will also be useful in identifying the optimal operating conditions at all levels of catalyst activity as deactivation progresses with age.

Most prior models for ammonia synthesis converters [1-6] have been devoted to axial flow systems such as the I.V.A. reactor. The present study instead focusses on a more modern radial flow reactor of Haldor Topsoe design. Such a reactor has lower pressure drop, and therefore higher effectiveness by virtue of smaller catalyst particle size. The stability region and optimal operating conditions for the reactor are identified. The model equations are solved on a microcomputer, using the Fortran IV programming language.

CONVERTER DESCRIPTION

Figure 1 provides a simplified sketch of the radial flow reactor simulated here. The feed gas entering the converter is split into three streams, with the bulk of the gas entering via stream A at the top of the reactor. The gas in stream A first flows through an outer annulus, keeping the reactor shell relatively cold, before entering the shell side of the bottom feed-product heat exchanger. The heated feed gas then passes through the central riser, after being mixed with cold bypass gas (stream C), and enters the first catalyst bed radially. Since the ammonia synthesis reaction



is highly exothermic, considerable increase in temperature of the gas mixture occurs in this bed. The gas leaving the catalyst bed at its outer radius is therefore quenched by stream B, before it enters the second catalyst bed where it flows radially inwards. Converted gas leaving the second bed then flows through an annulus surrounding the central riser, and enters the tube side of the bottom exchanger where it preheats the feed gas. The cooled product stream D leaves at the bottom of the converter.

MODEL DEVELOPMENT

Modeling of the converter essentially involves development of the differential material and energy balance equations for the catalyst beds, as well as for the various mixing and heat transfer zones in the reactor. These equations are outlined below for the key sections in the reactor.

Catalyst Beds

The material balance for the five species is given as

$$\begin{aligned} F_1 &= F_{10} (1 - X) \\ F_2 &= F_{20} - \frac{1}{3} F_{10} X \\ F_3 &= F_{30} + \frac{2}{3} F_{10} X \\ F_4 &= F_{40} \\ F_5 &= F_{50} \end{aligned} \quad (2)$$

where the order of the components is hydrogen, nitrogen, ammonia, argon, and methane, and X is the conversion of

hydrogen. The rate of change of X with radius in the bed is given by the differential material balance for hydrogen:

$$\frac{dX}{dr} = \frac{2\pi rL R(X,T)}{F_1(X)} \quad (3)$$

where r is the radius, L the bed height, T the local temperature, and R(X,T) the local reaction rate of hydrogen. The above equation considers only radial gradients, and assumes no material or energy transport in the axial direction. The local energy balance yields

$$\frac{dT}{dr} = \frac{2\pi rL(-2\Delta H(T,P)/3)R(X,T)}{\sum_i F_1(X) C_{pi}(T,P)} \quad (4)$$

where $\Delta H(T,P)$ is the heat of reaction, per mole of NH_3 produced, and $C_{pi}(T,P)$ the molar specific heat of pure component i. Mixing effects on the mixture heat capacity have therefore been ignored. An expression for the heat of reaction corrected for mixing has been provided by Gaines [3], while the heat capacities are tabulated in Shah [2]. These expressions were used directly in the current work. The local reaction rate is expressed by the Temkin Pyzhev equation as modified by Dyson and Simon [7]

$$R = 1.5a_c K_2 \left[k_a^2 f_2 \left(\frac{f_1^3}{f_2^2} \right)^a - \left(\frac{f_2^2}{f_3^3} \right)^{1-a} \right] \eta \quad (5)$$

where a_c is the catalyst activity, η the catalyst effectiveness, k_a the equilibrium constant, and f_i the fugacity of component i, given by

$$f_i = \gamma_i \gamma_i P \quad (6)$$

The required fugacity coefficients γ_i for the first three components are tabulated in Rase [8], as is the expression for the equilibrium constant k_a . The rate constant K_2 for the Haldor-Topsoe catalyst has been determined by Guacci et al. [9] and is given by

$$K_2 = 10^{15.2059} e^{-42,893/1.987 T} \quad \frac{\text{kg-mole}}{\text{hr} - \text{m}^3} \quad (7)$$

where T is in $^{\circ}\text{K}$. For the same catalyst these authors also obtain $a = 0.692$. In addition, for the small particles used in the radial flow reactor (< 6 mm), the effectiveness factor is unity [10]. Finally, theoretical calculations showed a negligibly small pressure drop in the radial flow design.

Bottom Heat Exchanger

Since the specific heats are functions of temperature, the usual LMTD concept is not applicable and it is necessary to integrate the differential equations

$$\frac{dT_t}{dz} = - \frac{\dot{U} A_z (T_t - T_s)}{\sum_i F_{t1} C_{pi} (T_t, P)} \quad (8)$$

$$\frac{dT_s}{dz} = - \frac{U A_z (T_t - T_s)}{\sum_i F_{s1} C_{pi} (T_s, P)} \quad (9)$$

where U is the overall heat transfer coefficient, based on tube outer diameter, and A_z is the area per unit length. The pressure drop in the exchanger is neglected in each side.

The overall heat transfer coefficient is estimated from the individual resistances using

$$\frac{1}{U} = \frac{D_o}{h_i D_i} + \frac{1}{h_o} + \frac{D_o \ln(D_o/D_i)}{2 k_w} + \epsilon^{-1} \quad (10)$$

where k_w is the tube wall thermal conductivity, and ϵ a fouling factor taken as $0.1 \text{ cal/cm}^2\text{-}^{\circ}\text{C-sec}$. The tube side heat transfer coefficient is calculated from the usual relation

$$\frac{h_i D_i}{k_i} = 0.023 \text{ Re}^{0.8} \text{ Pr}^{1/3} \left(\frac{\mu}{\mu_w} \right)^{0.14} \quad (11)$$

while the shell side coefficient from the Donohue equation

$$\frac{h_o D_o}{k_o} = 0.2 \text{ Re}^{0.6} \text{ Pr}^{0.33} \left(\frac{\mu}{\mu_w} \right)^{0.14} \quad (12)$$

Details of the evaluation of the shell-side Reynolds number, considering the baffle geometry, are to be found in Kern [11],

and McCable and Smith [12]. Mixture viscosities and thermal conductivities at the local temperature and converter pressure are estimated using the methods presented in Reid et al. [13]. Briefly, for viscosity, pure component low pressure values are first calculated by the Chapman Enskog theory, and combined to give the mixture viscosity using the Brokaw method. The mixture viscosity is then corrected for pressure by the Dean and Stiel correlation. For conductivity, low pressure pure component values are first estimated by the Eucken formula, then combined to yield the mixture thermal conductivity by the Mason-Saxena method, before finally applying the Stiel-Thodos correction for pressure.

Other Zones

Energy balance equations for the mixing region after the shell-side of the heat exchanger, and the quench zone after the first catalyst bed, are straightforward and may be found in the prior work cited above. Some heat loss occurs from the outer cold gas annulus to the atmosphere, and this was found to be controlled by the outside heat transfer resistance. This heat transfer coefficient is calculated using

$$\frac{h_o D_o}{k} = \psi_o \left(\frac{D_o G}{\mu} \right) \quad (13)$$

in which the function $\psi_o (D_o G/\mu)$ is obtained from the curve provided by McCadams [14]. Heat transfer between the outer annulus gas and that leaving the first bed is negligible because of the insulating material separating them. Finally, some heat exchange can occur between the gas leaving the second catalyst bed and the gas flowing in the central riser tube. This section is modeled as a double pipe heat exchanger, but with continuous addition of mass in the annular section. The overall heat transfer coefficient is obtained from Equation (10) with individual coefficients calculated from Equation (11). For the annulus side h_o is evaluated at the average flow rate, and using the equivalent hydraulic diameter [12].

SOLUTION METHOD

Starting with a knowledge of the inlet gas composition, temperature and pressure, as well as the distribution of feed between main inlet, quench and cold bypass, it is now necessary to solve the model equations described above to

predict the outlet gas temperature and ammonia production rate. To this end an iterative procedure was devised as follows:

- (1) Assume inlet temperature to first catalyst bed.
- (2) Solve the equations for the first catalyst bed, quench zone, and second catalyst bed, successively in that order, using the outlet conditions in each region as those for the inlet to the next.
- (3) Using the outlet conditions from the second catalyst bed for the annulus side, and the inlet conditions to the first catalyst bed for the tube side, integrate equations for the riser heat exchanger as an initial value problem and obtain temperatures at the bottom of this region.
- (4) Using the riser bottom temperature calculated above back out the shell side top temperature of the feed-product heat exchanger by material and energy balance for the mixing with cold bypass gas.
- (5) With tube side top temperature calculated in step (3) and the shell side top temperature obtained in step (4) solve the O.D.E's for the feed-product exchanger to get conditions at the bottom of this unit.
- (6) After accounting for heat losses to the surroundings calculate temperature of the stream entering the bottom heat exchanger shell side.
- (7) Compare the shell side inlet temperatures calculated in steps (5) and (6). If convergence has not occurred guess a new temperature at inlet to the first catalyst bed, go to step (2), and iterate.

To accelerate convergence of the above algorithm a modified Newton-Raphson procedure was used. All O.D.E's encountered for any region were solved using a fourth order Runge-Kutta method with 10 steps.

RESULTS AND DISCUSSION

The above model was programmed in Fortran IV, and the computations performed on an 8-bit Zenith Aurelec micro-computer. It was found that convergence was usually obtained in 3 iterations, with each iteration requiring about 5 minutes. Over a certain range of operating conditions as many as three steady states were identified. Of these the lowest conversion steady state involved negligible ammonia production and is not

a feasible condition. The two higher conversion steady states are illustrated in Fig. 2 in terms of an inlet-outlet temperature relation. As is commonly known in reaction engineering, and in prior studies of axial flow NH_3 converters, the left branch of this curve corresponds to unstable states, so that only the high conversion right branch yields stable and feasible operating conditions. Below 149°C inlet temperature the only solution is the negligible conversion case discussed above, but not shown in Fig. 2. This value of 149°C is therefore the minimum inlet temperature for stable feasible operation, and is called the blowout temperature.

Figure 3 shows the effect of inlet temperature on NH_3 production rate on the right branch of Fig. 2, and demonstrates the existence of an optimum. At the catalyst activity ($a_c = 0.45$) chosen for the calculations maximum ammonia production is obtained at an inlet temperature of about 151.8°C . This point, as shown in Fig. 2, is only slightly removed from the blowout condition, demonstrating the need for close control of the reactor if trouble-free operation is desired while optimizing performance.

NOMENCLATURE

A_z	area per unit length
a_c	catalyst activity
C_{pi}	molar specific heat of species i
D_o	outside diameter
D_i	inside diameter
f_i	fugacity of component i
F_i	molar flow rate of species i
F_{i0}	molar flow rate of species i at inlet to bed
F_{si}	shell side flow rate of species i
F_{ti}	tube side flow rate of species i
G	mass flux

h_i	inside heat transfer coefficient
h_o	outside heat transfer coefficient
ΔH	heat of reaction
k_a	reaction equilibrium constant
K_2	reaction rate constant
k_i	thermal conductivity of inside fluid
k_o	thermal conductivity of outside fluid
k_w	thermal conductivity of wall
L	length
P	pressure
Pr	Prandtl number
r	radial position
$R(X,T)$	reaction rate
Re	Reynolds number
T	temperature
T_t	tube-side temperature
T_s	shell-side temperature
U	overall heat transfer coefficient
X	conversion of H_2
y_i	mole fraction of i
z	axial position

Greek Symbols

ϵ	fouling factor
η	effectiveness factor
γ_i	fugacity coefficient
μ	viscosity
μ_w	viscosity adjacent to wall

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