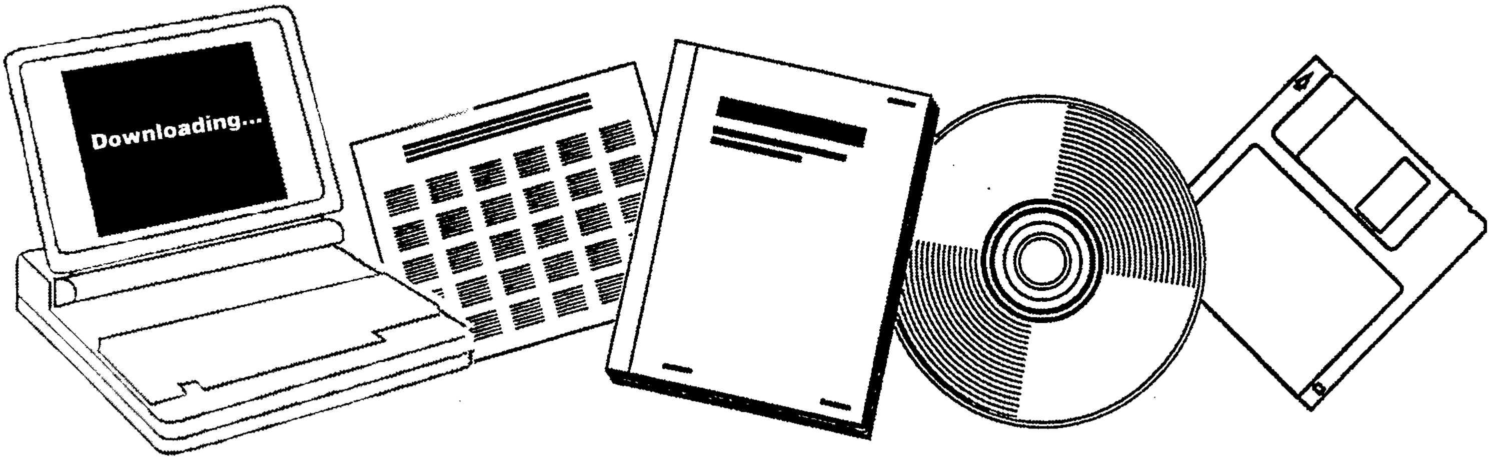
OPTIMIZATION OF COAL CONVERSION PROCESSES. MONTHLY PROGRESS REPORTS FOR THE PERIOD DECEMBER 1966--DECEMBER 1967

WEST VIRGINIA UNIV., MORGANTOWN

1967

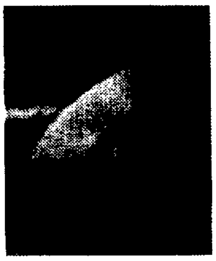


One Source. One Search. One Solution.

NTIS

Providing Permanent, Easy Access to U.S. Government Information

National Technical Information Service is the nation's largest repository and disseminator of government-initiated scientific, technical, engineering, and related business information. The NTIS collection includes almost 3,000,000 information products in a variety of formats: electronic download, online access, CD-ROM, magnetic tape, diskette, multimedia, microfiche and paper.



Search the NTIS Database from 1990 forward

NTIS has upgraded its bibliographic database system and has made all entries since 1990 searchable on **www.ntis.gov.** You now have access to information on more than 600,000 government research information products from this web site.

Link to Full Text Documents at Government Web Sites

Because many Government agencies have their most recent reports available on their own web site, we have added links directly to these reports. When available, you will see a link on the right side of the bibliographic screen.

Download Publications (1997 - Present)

NTIS can now provides the full text of reports as downloadable PDF files. This means that when an agency stops maintaining a report on the web, NTIS will offer a downloadable version. There is a nominal fee for each download for most publications.

For more information visit our website:

www.ntis.gov

U.S. DEPARTMENT OF COMMERCE
Technology Administration
National Technical Information Service
Springfield, VA 22161

FE--497-T-1

OPTIMIZATION OF COAL CONVERSION PROCESSES

Monthly Progress Reports for the
Period December 1966 - December 1967

C. Y. Wen

West Virginia University
College of Engineering
Morgantown, West Virginia 26506

Prepared for

Office of Coal Research
U. S. Department of the Interior

OCR Contract No. 14-01-0001-497

FE--497-T-1

CONTENTS

LANNED PHASING OF WORK FOR OPTIMIZATION OF COAL GASIFICATION PROCESSES

OCR CONTRACT 14-01-0001-497

1

Dec 1966

(A) Information gathering and assimilation.

To consult, collect review and classify information relating to coal gasification processes in the literature, from OCR contractors, or others.

(B) Sensitivity Study.

To study each step in the various gasification processes to determine steps and process parameters which are critical from those which are insignificant in the optimization of the processes.

(C) Development of general optimization technique involving information uncertainity.

To develop general mathematical optimization techniques applicable to the optimization of processes involving uncertain or changeable system functions, such as heat and mass transfer coefficients, kinetic and thermodynamic constants, operating conditions, marketing information, etc.

(D) Analysis and optimization of various processes.

To analyze various gasification processes and to determine the most effective optimization technique for each process and to develop a new technique, if the existing ones are inadequate.

(E) Development of computer program.

To develop comprehensive computer programs to determine optimum operating conditions of the various gasification processes and subsequent gas treatment including optimum design of reactors and purification systems, optimum coal supply, transportation, by-product utilization, etc., to arrive at the most practical and most economical gasification process.

(F) Comparison, evaluation and recommendation.

To compile and interpret the computer results in practical commercial terms and to compare the various proposed schemes recommending the most economically attracted coal-gasification processes.

(G) Final report preparation.

To prepare a complete, final technical report covering all phases of work under this contract.

PROGRAM FOR OPTIMIZATION OF COAL GASIFICATION PROCESSES

OCR CONTRACT 14-01-0001-497

Month	1966-67	1967-8	1968-9
	D J F M A M J J A S O N 1 2 3 4 5 6 7 8 9 10 11 12	D J F M A M J J A S O N 1 2 3 4 5 6 7 8 9 10 11 12	D J F M A M J J A S O N 1 2 3 4 5 6 7 8 9 10 11 12
A	G.S. 12 E. 6 D.S.C.=$10,000 O.D.C.= 1,000	G.S. 6 E. 4 D.S.C.=$6,000 O.D.C.= 800	
B	G.S. 12 E. 6 D.S.C.= 10,000 O.D.C.= 1,000	G.S.12 E. 6 D.S.C.=11,000 O.D.C.= 800	
C	G.S. 12 E. 6 D.S.C.= 10,000 O.D.C.= 1,000	G.S. 6 E. 2 D.S.C.= 4,000 O.D.C.= 800	
D	G.S. 12 E. 6 D.S.C.= 10,000 O.D.C.= 1,000	G.S.12 E. 6 D.S.C.=11,000 O.D.C.= 1,000	G.S. 12 D.S.C.=$11,000 E. 6 O.D.C.= 1,000
E		G.S.12 E. 6 D.S.C.=11,000 O.D.C.= 1,000	G.S. 18 D.S.C.= 12,000 E. 6 O.D.C.= 900
F			G.S. 12 D.S.C.= 11,000 E. 6 O.D.C.= 500
G			G.S.6 D.S.C.=$11,000 E. 6 O.D.C.= 3,000

Manpower shown in total man months:
E = Engineers
G.S.= Graduate Students

Costs:
D.S.C. = Direct Salaries and Consulting Costs
O.D.C. = Other Direct Costs

OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 2 JANUARY 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

Information gathering and assimilation have been continued. In this connection, Professors Bailie, Fan, Galli and Wen participated at the Office of Coal Research contractors meeting on January 17 and 18, 1967 at the Bituminous Coal Research, Incorporated in Pittsburgh. Some of the information from Pittsburgh Consolidation and Bituminous Coal Research have been received and they are now being reviewed.

The sensitivity study is also being continued. Two types of sensitivity studies are being conducted. One involves uncertainty of parameters when complete data are not available. Another one involves changeable parameters which vary according to time or localities.

A paper on the sensitivity analysis during optimization is being prepared. When this is completed, copies will be sent immediately to the Office of Coal Research.

Signed C. Y. Wen

C. Y. Wen, Project Leader

OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 3 FEBRUARY 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

Information gathering and assimilation have been continued. With an exception of the molten salt process of the M. W. Kellog Company, the current information on the progress of coal gasification from the OCR contractors, Institute of Gas Technology, Bituminous Coal Research, and Consolidation Coal Company, have been received regularly. These reports are now being reviewed and classified to evaluate the necessary information for optimization. The degree of progress of the first quarter on the various phases of work have been roughly estimated and are tabulated on the attached page. Although the chart indicates a slow start of the project, an acceleration is expected in a few months when new staff members report to work.

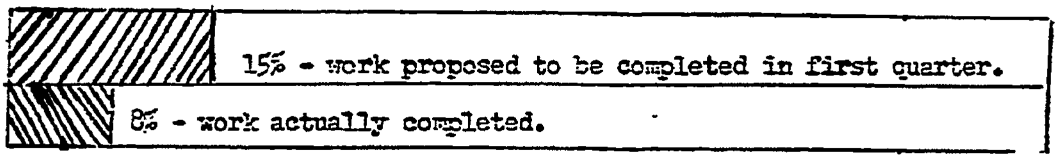
A study on optimal design on coal gasification system involving system parameter-uncertainty has been made. A report of this study is attached. With the technique developed, it is possible now to arrive at an optimal design even though some process information may be unavailable.

Signed C Y Wen
C. Y. Wen, Project Leader

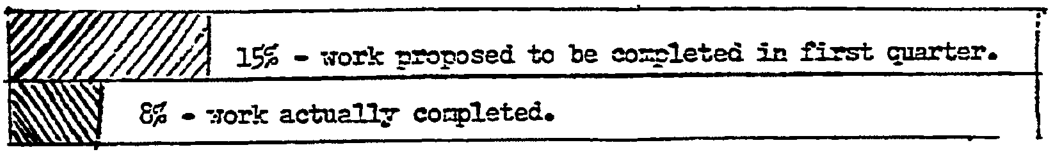
PROGRESS OF WORK FOR OPTIMIZATION OF COAL GASIFICATION PROCESSES

OCR CONTRACT 14-01-0001-497

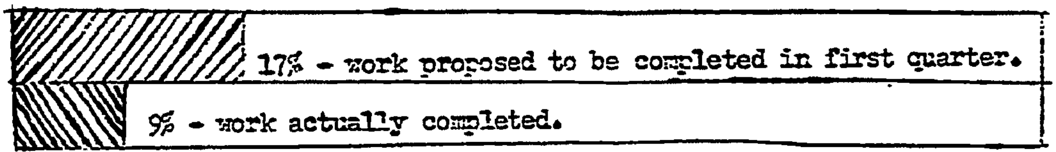
(A) Information Gathering and Assimilation.



(B) Sensitivity Study.



(C) Development of General Optimization Technique Involving Information Uncertainty.



OPTIMAL DESIGN OF COAL GASIFICATION SYSTEMS INVOLVING PARAMETER-UNCERTAINTY

Two design criteria were proposed for the optimal design of the coal gasification systems involving parameter-uncertainty. One of these design criteria may be used to obtain an appropriate decision which will keep the deviation of the objective from the optimal behavior within a certain tolerance. The other criterion assures a minimum average deviation of the objective from the optima over the range of uncertainty.

Examples were given to demonstrate the applicability of these criteria to the optimal gasification reactor design with uncertainty in kinetic constants.

Chemical Engineering Department
West Virginia University
Morgantown, West Virginia

OPTIMAL DESIGN OF SYSTEMS INVOLVING PARAMETER-UNCERTAINTY

In optimization, one seeks for an optimal policy containing various system parameters, the values of which are assumed to be accurate. But often the system parameters of chemical processes such as, kinetic constants, heat and mass transfer coefficient, etc. are unknown or known only approximately, or are varying within a certain range due to the changing operating conditions.

In order to determine these parameters accurately, a great length of time and money must be spent, often beyond the justification called for. Usually, a cost estimation is necessary before arriving at a decision whether or not to construct a certain plant. To be anyway meaningful the estimation must be obtained under the optimum conditions. In other words, to determine how profitable a chosen process is over other alternate processes, an economical comparison should be made based on each process operating at its optimum conditions. This means that it becomes often necessary to optimize the processes without accurate information of these parameters. Also in many instances, a process may have varying system parameters which are changing due to operating conditions. In these cases, a reasonable design criterion must be established. It is obvious that an optimal policy obtained based on an approximate or a specific value of the parameter would not necessarily give the best profit obtainable. Consequently, it is not appropriate to derive an optimal policy simply based on some specific value of the parameters when they involve a range of uncertainty. Therefore, some sort of design criteria must be developed so that an appropriate policy (decision) can be selected which will result in a profit (cost) close to the maxima (minima) obtainable in the range of parameters considered.

There are several decision criteria available which provide economic strategies for decision making under uncertainty (3). The most commonly used criteria for the design of processes involving parameter-uncertainty has been maximizing expected profits or minimizing expected costs (1). According to this criterion, an appropriate decision is the one which gives the largest (smallest) among all possible expected profits (costs). In other words, the optimality of a given decision is measured by the expected value of the objective function. Since we are looking for a decision which will give the profit close to that resulted from every optimal policy over the range of parameters, this criterion cannot assure us such a decision. Moreover, when a policy which will keep the deviation from the optimal behavior within a certain tolerance is to be chosen, some other kind of criterion is needed.

In searching for appropriate design criteria, we have noticed the recent work by Rohrer and Sobral (5). They proposed design criteria by introducing the so-called relative sensitivity for designers to find a single controller for many similar plants or to find a controller for a single changeable plant. These criteria may well be adapted in optimal processes design involving parameter-uncertainty.

The relative sensitivity is defined as the normalized deviation from the optimal value of the objective function (5). Let J, θ and W be objective function, decision variable and system parameter, respectively. The relative sensitivity, S, is then expressed as

$$S(W, \theta) = \frac{|J(W, \bar{\theta}) - J(W, \theta)|}{|J(W, \bar{\theta})|} \qquad (1)$$

where $\bar{\theta}$ is the optimal decision for parameter W in its admissible domain Θ, and,

$$J(W, \bar{\theta}) = \max_{\theta \in \Theta} \left\{J(W, \theta)\right\} \text{ or } \min_{\theta \in \Theta} \left\{J(W, \theta)\right\} \qquad (2)$$

Note that the optimal value of S(W, θ) is zero and that S(W, θ) is always positive. One of the design criteria proposed is to find a decision so as to minimize the maximum relative sensitivity. Namely, this decision, θ^*, is obtained from

$$S(\theta^*) = \min_{\theta \in \Theta} \left\{ \max_{W} \quad [S(W, \theta)] \right\} \tag{3}$$

In choosing θ^*, first we find the largest normalized deviation over the range of the parameter. This assures that no matter how the true value of the parameter turns out to be, as long as the true value exists in this range, the normalized deviation will not be larger than this largest value. Consequently, the decision that minimizes the maximum relative sensitivity is appropriate for the design of the system involving parameter-uncertainty, when a critical tolerance must be satisfied.

The other design criterion is to find a decision so as to minimize the average deviation from optimal behavior. The average deviation (or the expected deviation) may be defined according to the probabilistic distribution of the system parameter, namely,

$$S^E(\theta) = \int_{-\infty}^{\infty} S(W, \theta) \quad f(W) \, dW \tag{4}$$

where $f(W)$ is a density function of W and $f(W)dW$ is defined as the probability that W will fall in the range between W and $W + dW$. The proper decision according to the second criterion is then obtained from

$$S(\theta^*) = \min_{\theta \in \Theta} \left\{ S^E(\theta) = \int_{-\infty}^{\infty} S(W, \theta) \, f(W) dW \right\} \tag{5}$$

Namely, the decision θ^* is the one which minimizes the expected relative sensitivity. Different from the expected profit criterion mentioned above, the optimality of a given decision according to the criterion of Eq. 5 is measured by the expected deviation from optimal behavior. In other words, we are always trying to keep the process to perform as close as possible to the optimum over the range of uncertainty. Since the optimal performance of a process is what we are seeking for, the design criterion of Eq. 5 is more meaningful in comparison to that minimizing (maximizing) expected costs (profits). When no critical tolerance is required in the design problem, this criterion is useful for the design of a process with parameters that are (a) inaccurate or (b) varying due to the changing operating conditions.

In the following, we present two examples showing how these criteria can be applied to the optimal reactor design with uncertainty in kinetic constants.

Example 1 Optimal Temperature for an Exothermic Reversible Reaction

Recently Ray and Aris (4) have investigated the effect of the errors in the kinetic constants on the maximum reaction rate for a general homogeneous exothermic reaction. Their results allow one to decide, in order to keep the deviation from the maximum reaction rate within a certain tolerance, (a) how tight the temperature control should be if kinetic constants are known exactly, (b) how accurate the estimates of kinetic constants should be if the temperature is perfectly controlled at its optimum corresponding to the approximated values of kinetic constants and (c) the combined effect of parameter and control errors. However, a question may be asked: "What will be the best choice of the reactor temperature so that the deviation from the maximum reaction rate is within a critical tolerance when the kinetic constants are not known exactly but are known to fall within a certain range?" A reasonable criterion is to choose a temperature which minimizes the maximum deviation from the maximum reaction rate, namely, the criterion as given in Eq. 3. An illustration is shown below.

Consider a first order, reversible reaction $A \rightleftharpoons B$. Let x be the mole fraction of B, and k and k' be the forward and backward reaction rate constants, respectively. If the Arrhenius form is employed, they are expressed as,

$$k = A \exp(-E/RT), \; k' = A' \exp(-E'/RT).$$

Then the reaction rate expression is

$$r(T, x) = k(1 - x) - k'x \tag{6}$$

For an exothermic reaction, $E' > E$, at a given composition, there exists an optimal temperature for the maximum reaction rate. It may be obtained from $\partial r/\partial T = 0$, and is given by

$$T_m = (E' - E) \Big/ \left\{ R \ln \left[\frac{A'E'}{AE} \, \frac{x}{(1-x)} \right] \right\} \tag{7}$$

The maximum reaction rate is then expressed as $r(T_m, x)$ by substituting Eq. 7 into Eq. 6. The system parameters of this reaction are E, E', A and A', but they are related to the thermodynamic constants. The activation energies E and E' are related by $E - E' = \Delta H$, where ΔH is the heat of reaction, and $A/A' = K^*$ can be obtained from the equilibrium constant of the reaction, K_c, as

$$K_c(T) = K^* \cdot \exp(-\Delta H/RT)$$

The thermodynamic constants, ΔH and K_c are easier to obtain than the kinetic constants and are usually known. If an experiment is performed and a rate constant k at a certain temperature is obtained then A can be related to E. Consequently, the four parameters are now reduced to one independent parameter, E.

As a numerical example, let $\Delta H = -10{,}000$ Btu/lb mole, $K^* = A/A' = 0.0126$ and from an experiment, $A = 1690 \exp(E/2000)$. Consider E as the independent parameter which is estimated to fall in the range from 4,000 to 16,000 Btu/lb mole.

For a specific value of E and for $x = 0.8$, we can calculate the optimal temperature T_m from Eq. 7 and the reaction rate from Eq. 6. Now the appropriate temperature for this reactor design with uncertainty in E may be found from Eq. 3 as

$$S(T^*) = \min_{T} \left\{ \max_{4000 \leq E \leq 16000} \left[1 - r(T, E) \Big/ r(T_m, E) \right] \right\} \tag{8}$$

As shown in Figure 1, the minimax of the relative sensitivity, $S(T^*)$, is 10% occurring at the intersection of $E = 4{,}000$ and $T^* = 775°R$. This simply means that if we control the reaction temperature at 775°R, we will obtain a deviation of at most 10% from the maximum reaction rate for the range of E considered. Any other choice of temperature will cause larger maximum deviation.

The design criterion given above can provide not only an appropriate choice of the decision variable but also the maximum deviation from the optimal value. If this maximum deviation is more than that can be tolerated, more accurate estimates of the parameters is needed. Using the above example, if the maximum deviation of 10% is too large, the range of E must be further narrowed, for instance to between 6,000 and 14,000, then T^* becomes 780°R and the maximum deviation is reduced to 5%.

The necessary equations for finding an appropriate temperature for a general homogeneous reaction may be derived. It has been shown (4) that the optimal temperature for a general homogeneous, reversible, exothermic and elementary reaction can be obtained from

$$\tau_m = I + 1/n \ln(1 + 1/p) \tag{9}$$

and the relative sensitivity of the maximum reaction rate is

$$S(\tau; n, p) = 1 - r(\tau; n, p)/r(\tau_m; n, p)$$

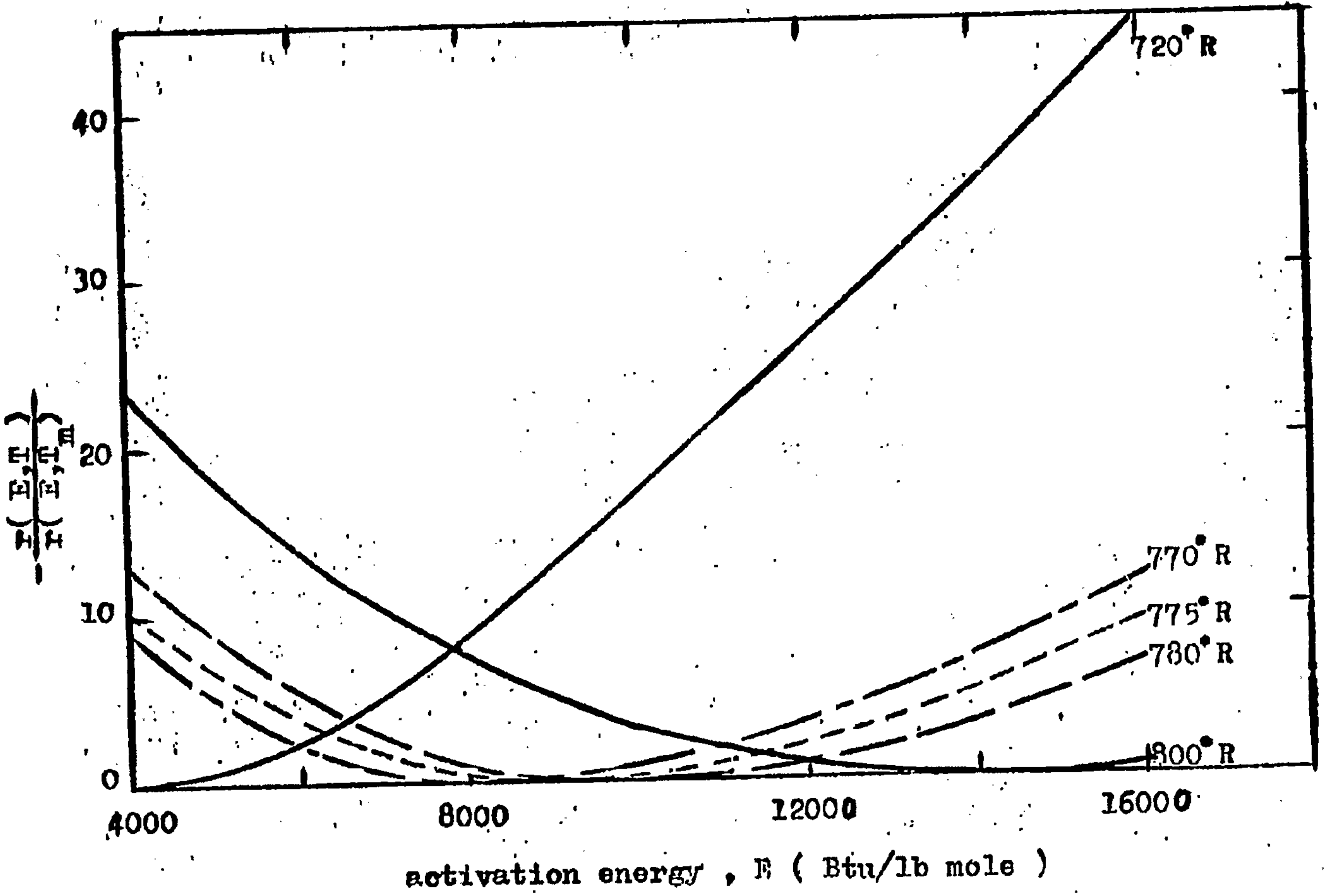


Figure 1. Relative sensitivity of maximum reaction rate.

$$= 1 + p \exp[-n(1 + p)(\tau - \tau_m)] - (1 + p) \exp[-np(\tau - \tau_m)] \qquad (10)$$

where

$\tau = -\Delta H/RT$, $X = \ln(\Pi C_j^{\alpha_j}/K^*)$, $n = (\gamma_j - \beta_j)/\alpha_j$ and $p = -E/n\Delta H$

Substituting Eq. 9 in Eq. 10, one obtains

$$S(\tau\, ;\, n, p) = 1 + \frac{(1+p)^{1+p}}{p^p} \exp[-np(\tau - X)] \cdot \left\{\exp[-n(\tau - X)] - 1\right\} \qquad (11)$$

where n and p are independent parameters of the reaction which are related to the reaction orders and the activation energy, respectively. These two parameters must be obtained experimentally, and are usually very difficult to determine accurately. Their values, however, may be estimated within a certain range, as $n_* \leq n \leq n^*$ and $p_* \leq p \leq p^*$.

Then the design criterion of Eq. 3, becomes

$$S(\tau^*) = \min_{\tau} \left\{ \max_{\substack{n_* \leq n \leq n^* \\ p_* \leq p \leq p^*}} [S(\tau\, ;\, p, n)] \right\} \qquad (12)$$

Numerical computation has shown that the maximum of $S(\tau\, ;\, p, n)$ for any τ occurs at one of the four points: (p^*, n^*), (p^*, n_*), (p_*, n^*) and (p_*, n_*). This makes the task of finding a minimax much easier since Eq. 12 may now be reduced to

$$S(\tau^*) = \min_{\tau} \left\{\max[S(\tau ; p^*, n^*), S(\tau ; p^*, n_*), S(\tau ; p_*, n^*), S(\tau ; p_*, n_*)]\right\} \qquad (13)$$

To obtain τ^*, one has only to calculate those four values of S for each τ and find τ^* which corresponds to the smallest value among the maximum S.

Example 2 Optimum Over-Design of a Reactor

To illustrate the application of the expected relative sensitivity criterion of Eq. 5 and to compare it with the expected cost criterion, we consider the following numerical example originally considered by Levenspiel (2) and later used by Kittrell and Watson to illustrate the expected cost criterion (1).

Consider a reaction $A \xrightarrow{k} R$ with rate $r = kC_A$ where $k = 0.2(\text{hr})^{-1}$. The production rate of R is 100 g-moles/hr. The feed consists of a saturated solution of A(C_{Ao} = 0.1 g-mole/l.). The cost of the reactant in the saturated solution is c_a = \$0.50/g-mole of A. The cost of the complete mixing reactor including installation, auxiliary equipment, instrumentation overhead, labor, depreciation, etc., is c_b = \$0.01/hr.l. What size of reactor, feed rate F_{AO}, and conversion X_A should be used for optimum operations? The total hourly cost is

$$c_t = Vc_b + F_{AO}\, c_a = 0.01\, V + 0.5\, F_{AO} \tag{14}$$

For the complete mixing reactor,

$$V = \frac{F_{AO}\, X_A}{kC_{AO}(1 - X_A)} \tag{15}$$

Noting $F_{AO} = 100/X_A$, the total cost can be expressed as a function of V and k,

$$c_t(V, k) = 0.01\, V + 50\, kV/(kV - 1000) \tag{16}$$

The optimum volume, $\bar{V}$, is obtained by letting $\frac{dc_t}{dV} = 0$, namely,

$$\bar{V} = 1000\,(1 + \sqrt{5k})/k \tag{17}$$

The optima are found to be:

$$X_A = 0.5,\ F_{AO} = 200 \text{ mole A/hr},\ V = 10{,}000 \text{ l.},\ c_t = 200 \text{ \$/hr.}$$

The optimum reactor size is 10,000 liters provided we have confidence in the accuracy of the rate constant, k. However, k is determined experimentally and is known to fall in a certain range with some probability density function. Kittrell and Watson (1) have solved such parameter-uncertainty problem by applying the expected cost criterion. The optimum designed volume, V^*, was obtained from

$$\min_{V} \left\{ \int_{-\infty}^{\infty} c_t(V, k)\, f(k)\, dk \right\} \tag{18}$$

where f(k) is a density function for k. If k is distributed rectangularly in the interval,(0.2 - Δk) to (0.2 + Δk), f(k) is defined as follows:

$$f(k) = \begin{cases} \dfrac{1}{2\Delta k} & 0.2 - \Delta k \le k \le 0.2 + \Delta k \\ 0 & \text{otherwise} \end{cases} \tag{19}$$

While applying the expected relative sensitivity criterion, the optimum designed volume, V^*, is obtained from

$$\min_{V} \left\{ \int_{-\infty}^{\infty} S(V, k)\, f(k)\, dk \right\} \tag{20}$$

where S(V, k) is the relative sensitivity of the total cost, namely,

$$S(V, k) = \frac{c_t(V, k)}{c_t(\bar{V}, k)} - 1 \tag{21}$$

Assuming that k is rectangularly distributed, the optimum over-design was obtained from expected relative sensitivity criterion of Eq. 20, as shown in Table 1, and is compared with that obtained by Kittrell and Watson (1) using expected cost criterion of Eq. 18. It is seen that the present criterion gives lower over-design than the expected cost criterion. The difference becomes larger as the range of uncertainty increases. The reason why the present criterion gives lower over-design is that a reactor volume is searched by minimizing the deviation from minimum cost corresponding to each k in the

Table 1. Comparison of optimum over-design obtained from expected cost criterion and expected relative sensitivity criterion.

	Expected Cost Criterion (1)		Expected Relative Sensitivity Criterion	
Range of Uncertainty $2\Delta k$, $(hr)^{-1}$	Optimum designed volume, liters	Amount of over-design, %	Optimum designed volume, liters	Amount of over-design, %
0.05	10,110	1.1	10,090	0.9
0.15	11,140	11.4	10,950	9.5
0.19	12,100	21.0	11,780	17.8
0.20	12,425	24.3	12,075	20.8

range of uncertainty. This can be further discussed based on Figure 2. As shown in this figure, Curve A is the total cost over the range of k resulted from the expected cost criterion, Curve B is that resulted from the expected relative sensitivity criterion and Curve C is the locus of the minimum total cost corresponding to each k. In choosing an appropriate volume, we want to choose the resulted curve, for example, Curve B, to be as close as possible to Curve C. Therefore, the closeness between Curves B and C has to be defined in a meaningful way. The closeness may be measured by the difference between the two curves for each k. However, this does not take into account the magnitude of the cost for each k. A larger difference may be allowed if the cost is higher. Therefore, it is more meaningful to measure the closeness by the relative sensitivity, the fractional difference between Curves B and C, as given in Eq. 21. Curve B is obtained by choosing a volume such that the average of this fractional difference is a minimum. Consequently, the optimum design volume obtained from the present criterion, can assure us a smallest average deviation from the minimum total costs over the range of k considered.

Conclusions

The design criteria of Eqs. 3 and 5 have been meaningfully applied to the optimal design of the systems involving inaccuracy in the values of the parameters. The design criterion of Eq. 3 gives the minimax of the deviation from the optimal behavior, from which one can decide whether a more accurate estimate is needed by comparing the minimax with the tolerance limit. When one is seeking a decision for the system with inaccurate or changing values of the parameters in which no critical tolerance is present, the design criterion of Eq. 5 should be particularly effective.

Some examples illustrated here have demonstrated the usefulness of the proposed design criteria. The optimal temperature for a reversible and

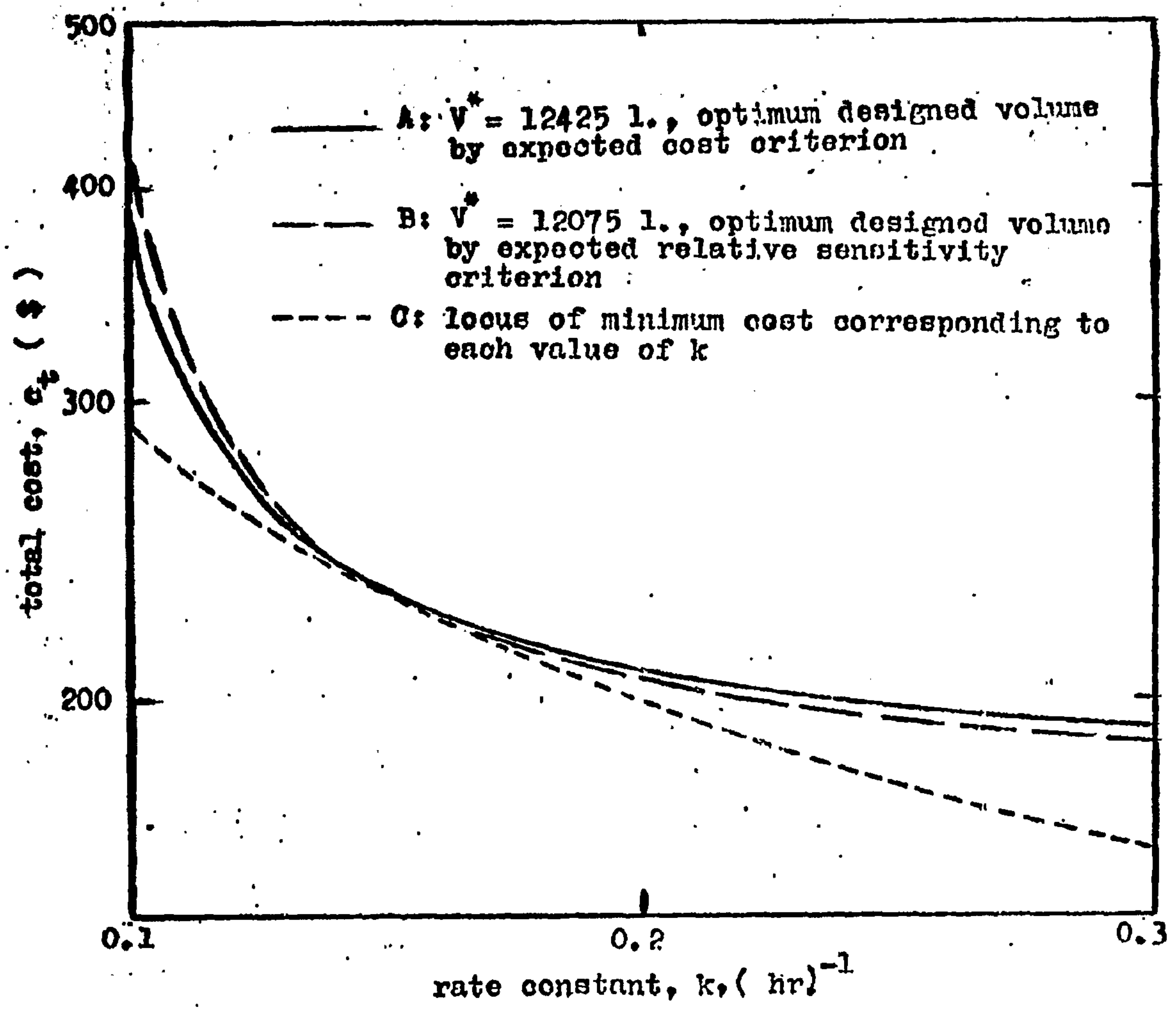


Figure 2. Deviation from minimum total costs

exothermic reaction has been determined by the proposed design criteria. The minimax of the deviation from the maximum reaction rate is 10% for ± 60% error in the activation energy. This indicates that a large error in estimates of activation energy can give the deviation from the maximum reaction rate within a few percentage, agreeing with the results obtained by Ray and Aris (4). Meanwhile, the necessary equations for finding an appropriate temperature for a general homogeneous, reversible and exothermic reaction by using the proposed design criterion have been obtained as indicated by Eq. 11 and 13.

Optimum over-design for a reactor was obtained by the expected relative sensitivity criterion. The optimum over-design so obtained is lower than that obtained by the expected cost criterion. By using this criterion, designers can make sure that the result obtained has a smallest average deviation from the optimal behavior over the range of uncertainty.

Acknowledgement

The authors wish to express their gratitude to the Office of Coal Research, Department of the Interior, Washington, D. C. for financial support.

Nomenclature

A, A' = frequency factor for forward, backward reaction

c = cost

C_A, C_j = concentration

E, E' = activation energy for forward, backward reaction

$f(W)$ = density function for W

F_{AO} = feed rate of component A

J = objective function

k, k' = reaction rate constant for forward, backward reaction

K^* = A/A'

K_c = chemical equilibrium constant

r = reaction rate

R = gas constant

S = relative sensitivity

S^E = expected value of S, Eq. 4

T, T^* = temperature

T_m = optimal temperature

V, V^* = reactor volume

w, W = parameter

x = mole fraction

X = dimensionless extent of reaction

X_A = conversion of component A

Greek Letters

α_j = stoichiometric coefficient of species j

β_j = forward reaction order with respect to species j

γ_j = backward reaction order with respect to species j

ΔH = heat of reaction

Δk = range of k

Θ = domain of decision variable

θ = decision variable

$\bar{\theta}$ = optimal decision

τ = dimensionless temperature

References

1. Kittrell, J. R. and C. C. Watson, Chem. Eng. Progr., 62, No. 4, 79 (April, 1966).

2. Levenspiel, O., "Chemical Reaction Engineering," p. 134, Wiley, New York (1962).

3. Luce, R. D. and H. Raiffa, "Games and Decisions," Chapter 13, Wiley, New York (1957).

4. Ray, W. H., R. Aris, Ind. Eng. Chem. Fundamentals 5, 478 (1966).

5. Rohrer, R. A., M. Sobral, Jr., IEEE Trans. on Automatic Control, vol. AC-10, No. 1 (1965).

OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 4 MARCH 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

Information gathering and assimilation have been continued. The monthly progress reports of the molten salt process of the M. W. Kellogg Company are now being received. Meetings and discussions, among the investigators here and personnel from the OCR contractors and the U. S. Bureau of Mines, were held. It has been tentatively decided that the optimization of the gas clean-up process and the methanation process should be conducted first since these processes are common to all the gasification processes under current studies. For this purpose, the Institute of Gas Technology in Chicago was visited and their work on the methanation project was examined. The experimental data obtained by the Institute of Gas Technology has been gathered, and examination is currently underway to determine whether or not the methanation process can be optimized at this stage.

Studies on the sensitivity analysis have also been continued. Sensitivity of the objective function to the variation of the systems parameters in terms of absolute sensitivity (as opposed to the relative sensitivity) is considered. The absolute sensitivity for (1) continuous process, (2) discrete process, and (3) process describable by a set of algebraic equations, are derived with numerical examples illustrating application of the method.

Signed C. Y. Wen

C. Y. Wen, Project Director

OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. ~~4~~ 5 APRIL 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

Information gathering and assimilation have been continued. A report on absolute sensitivity is being prepared which will become available in the next month's report. The absolute sensitivity analysis will yield information which can be used to differentiate the parameters and variables that are critical to the economy of the plant from those which rough estimation is sufficient for optimization.

In addition, optimization of methanation processes has been studied. The processes chosen are (1) multitube fixed bed reactor and (2) multitray fixed bed reactor. The study includes process analysis, thermodynamics and kinetics analysis, material and heat balance, economic analysis and method of optimization. The necessary equations for optimization are derived using the maximum principle.

Numerical computation will begin as soon as the necessary data are collected and accuracy checked. Gas clean-up process analysis is also started. The hot-carbonate process is being studied and the formulation of equations necessary for optimization has been started.

C. Y. Wen

C. Y. Wen, Project Director

OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 6 May 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

Information gathering and assimilation have been continued. A report on absolute sensitivity is attached. The absolute sensitivity analysis developed can be used to obtain information on how critical the parameters and variables studied are to the economy of the plant. Also attached is a report on Nakanishi's sequential optimization technique which can treat continuous, discrete and stochastic processes involved in gasification of coal.

Optimization of methanation processes is being continued. The necessary equations for optimization are being derived using the maximum principle. Gas clean-up process analysis is also being continued.

The amount of progress of the second quarter on the various phases of work have been roughly estimated and are tabulated on the attached page. The chart indicates the progress of the project to be slow, but an acceleration is expected as soon as new staff members are employed.

C. Y. Wen

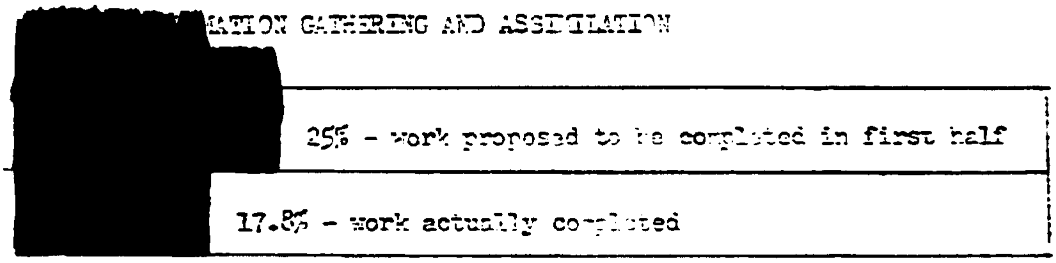
C. Y. Wen, Project Director
West Va University

Reproduced from best available copy

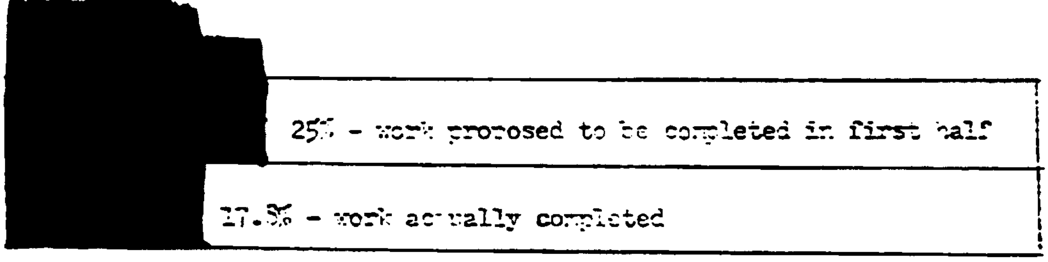
PROGRESS OF WORK FOR
OPTIMIZATION OF COAL GASIFICATION PROCESSES

OCP CONTRACT 14-01-0001-497

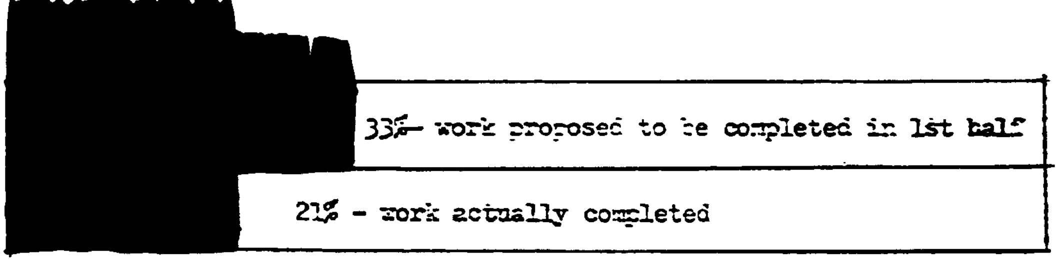
ATION GATHERING AND ASSIMILATION



(B) SENSITIVITY STUDY



(C) DEVELOPMENT OF GENERAL OPTIMIZATION TECHNIQUE INVOLVING INFORMATION UNCERTAINTY



OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 7 JUNE 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

Optimization of methanation processes is being continued. The necessary equations for a fixed bed multitray methanator have been completed. Work has been initiated on the development of a computer program to carry out the optimization. Gas clean-up process analysis is also being continued.

Our work is approaching the point where we will need the "Office of Coal Research Guide to Cost Estimating". Factors that need standardization such as plant size, labor costs, raw material costs, capitalization costs, amortization rate, etc., are needed for a meaningful comparison of processes.

C. Y. Wen

C. Y. Wen, Project Director

OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 8 JULY 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

Sensitivity analyses in mathematical models of process system have been made. The sensitivity equations necessary for the determination of the effect of the magnitude of the system parameters on the optimal performance have been derived. It was found that the change of some parameters in mathematical models could change the order of the differential equations describing the system behavior and thus the system could become degenerate. A study is being made to find a way to select an adequate mathematical model for the process to be optimized.

The optimization of methanation processes is being continued. Process analysis and economic analysis have been completed. A computer program is being written and a method suitable for optimization of the process has been selected. Some of the results of this study will be presented in next month's progress report.

Information gathering and assimilation have been continued.

Optimization of compressor-high pressure systems has been started. The objective is to find the means of reducing high pressure equipment cost which seems to be one of the major cost items.

C. Y. Wen

C. Y. Wen, Project Director

OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 9 AUGUST 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

Optimization of methanation processes is being conducted. The computer programming for a fixed bed multitray methanator has been completed. Partial results of the computation have been obtained. However due to the fact that the University's Computer Center has been busy with the work that accompanies the close of the Summer Session and the beginning of the fall Semester and also due to the computer breakdown, we were unable to use the computer for approximately three weeks. Instead of reporting the result here, it will be compiled as a quarterly report in the September report. The sensitivity study is also being continued. A decision making criterion is proposed for systems which are sensitive to changes in parameters. This criterion is to minimize a new objective function which is composed of the original objection function and the sensitivity functions. The decision obtained from this criterion is compromisely optimum but less sensitive. This criterion is useful in making a suitable decision for a system whose performance is very sensitive to the changes in system parameters. A modified algorithm of the maximum principle is developed to obtain an optimum trajectory of sequence according to the proposed criterion. The amount of progress of the third quarter of various phases of work have been estimated and are tabulated on the attached page.

C. Y. Wen

C. Y. Wen, Project Director

PROGRESS OF WORK FOR
OPTIMIZATION OF COAL GASIFICATION PROCESSES

OCR CONTRACT 14-01-0001-497

[redacted] ASSIMILATION

46% - work proposed to be completed by third quarter

35% - work actually completed

[redacted]

30% - work proposed to be completed by third quarter

25% - work actually completed

[redacted] PTIMIZATION TECHNIQUE INVOLVING

52% - work proposed to be completed by third quarter

40% - work actually completed

[redacted] ES AND OPTIMIZATION OF VARIOUS PROCESSES

18% - work proposed to be completed by third quarter

16% - work actually completed

OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 10 SEPTEMBER 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

Sensitivity study is being made on actual methanation and gasification process. The optimization of sub-systems, namely the purification process, the shift reaction system, the hydrogen production system, and the methanation system is being made. An interim report on the progress of optimization of methanation processes is attached. It is apparent from the study, heat removal is the major problem and the cost of fin tube becomes a large portion of the total cost. It is believed that the cold shot cooling system which does not use internal heat exchanger will be better and optimization of this system is now being conducted.

C. Y. Wen

C. Y. Wen, Project Director

INTERIM REPORT ON THE PROGRESS OF
OPTIMIZATION OF METHANATION PROCESSES

Although gasification of coal to pipeline gas can be achieved by a considerable variety of routes (at least four of such routes are currently under extensive study under the sponsorship of the Office of Coal Research) it appears, any system involving conversion of coal will require additional units for conversion of excess carbon monoxide and hydrogen to methane in order to produce a gas with the heating value equivalent to natural gas.

The magnitude of methanation varies greatly depending upon as yet unsettled choice of the process in the primary gasification phases. One of the major aspects in the selection of the best way to accomplish the primary gasification process is to find the amount of methanation which must be carried out for the most economical production of pipeline gas.

In this regard, various catalytic methanation processes must be examined and compared under their optimum designed conditions. Accordingly, three likely feeds as shown on Table I are selected for the optimization study. Since it is impossible to predict the exact composition of the gaseous effluent from the primary coal gasification processes, the compositions of the three feeds listed on Table I are approximate gas mixtures likely to come from the primary gasification phase, after the subsequent adjusting of the composition by the water-gas shift reaction and purification.

Three types of simple fixed bed downflow catalytic reactors are considered. They are (1) the internal heat removal system, (2) the cold shot cooling system, and (3) the recycle system. The distinguishing features among the three systems are the manners by which heat is removed and temperature is controlled in the reactor. The goal is to evaluate their relative technical merits for prospective application. In the following the results of the economic optimization using the internal heat removal system are briefly summarized. Study of other two systems is now under way and a comprehensive report on this subject is being prepared.

The reactor for the internal heat removal system consists of multi-tray fixed bed packed with 1/4-inch Harshow catalysts. Some experimental data on reaction kinetics and activity for this catalyst are reported by I.G.T. For convenience in regeneration of catalysts, multi-tray with embedded hairpin fin tubes having equal heat transfer area in each tray is used. Details of the design of the system including kinetic analysis and economic analysis will be given in a comprehensive report and therefore will not be discussed here. In this study, only those variables that will affect the optimal design of the process are considered. The calculation is based on the plant capacity of 250 x 10^9 Btu/Day of gas having heating value of approximately 950 Btu/SCF.

TABLE I COMPOSITION OF FEEDS (MOLE PERCENT)

	Low CO Feed%	Intermediate CO Feed%	High CO Feed%
CH_4	75.4	62.0	92.1
CO	4.5	8.0	0.1
H_2	17.5	27.7	4.8
CO_2	0.2	0.2	0.3
H_2O	0.1	0.1	0.2
N_2	2.1	1.8	2.5
C_2H_6	0.2	0.2	0.2
TOTAL(lb.mole/hr)	34,107	39,792	47,750

TABLE II REACTOR TEMPERATURE, PRESSURE AND PRODUCT GAS COMPOSITION

	Low CO Cases	Intermediate CO Cases	High CO Cases
Production Rate Btu/Day	250×10^9	250×10^9	250×10^9
Operating Temp. °F	850	850	850
Operating Pressure atm	69.7	69.7	69.7
Product Gas Composition (%)			
CH_4	92.104	92.100	92.099
CO	0.156	0.098	0.124
H_2	5.085	5.167	4.748
CO_2	0.231	0.264	0.505
H_2O^*	0.0	0.0	0.0
N_2	2.425	2.371	2.524

*Dry Basis

TABLE IIIa OPTIMUM REACTOR REQUIREMENTS FOR THREE DIFFERENT FEEDS USING WATER COOLING

Reactor Diameter ft	6	6	7
Reactor Height ft	18.70	33.2	42.5
Reactor Volume ft^3	526.	937.	1,635.
Catalyst Weight lb	12,390	18,620	23,840
Total Heat Removed MM Btu/hr	140	296	602
Number of Trays	5	10	13
Pressure Drop psi	60	107	75
Total Heat Transfer Area of Fin Tubes (Based on Bare Tube) ft^2	9,800	20,600	43,700
Heating Value	950	950	950
Production Rate lb.moles/hr	28,940	28,940	28,940

TABLE IIIb COSTS OF EQUIPMENT AND UTILITIES WITH THREE DIFFERENT FEEDS USING WATER AS COOLANT

	Low CO Feed	Intermediate CO Feed	High CO Feed
Total Fin Tube Cost $	47,700	104,100	243,000
Catalyst Cost $	31,000	47,000	60,000
Reactor Cost, $	37,000	56,000	91,000
Tray Support Cost $	1,800	3,700	6,100
Control Valve Cost $	7,000	14,000	18,000
Pumping Cost $	2,300	6,200	16,200
Water Cost $/hr	11.68	24.74	50.29
Electricity Cost $/hr	0.074	0.64	4.99
Steam Recovery $/hr	122	258	524

TABLE IVa OPTIMUM REACTOR REQUIREMENTS FOR THREE DIFFERENT FEEDS USING DOWTHERM A COOLING

Reactor Diameter ft	6	6	7
Reactor Height ft	21	40	60
Reactor Volume ft^3	594	1,130	2,300
Catalyst Weight lb	12,390	18,620	23,840
Total Heat Removed MM Btu/hr	140	296	602
Number of Trays	6	12	17
Pressure Drop psi	68	129	305
Total Heat Transfer Area of Fin Tubes (Base on Bare Tube) ft^2	12,500	28,200	70,700
Total Fin Tube Volume ft^3	310	700	1,760
Production Rate lb.moles/hr	28,940	28,940	28,940
Heating Value Btu/SCF	950	950	950

TABLE IVb COSTS OF EQUIPMENT AND UTILITIES FOR METHANATION PROCESS WITH THREE DIFFERENT FEEDS USING DOWTHERM A AS COOLANT

	Low CO Case	Intermediate CO Case	High CO Case
Total Fin Tube Cost $	54,000	118,600	278,400
Catalyst Cost $	31,000	47,000	60,000
Reactor Cost $	42,000	67,000	129,000
Tray Support Cost $	2,190	4,380	8,000
Control Valve Cost $	8,400	16,800	23,300
Condenser Unit Cost $	5,080	7,740	11,510
Dowtherm Cost $	42,500	55,000	79,500
Pumping Cost $	2,260	6,210	16,220
Electricity Cost $/hr	0.074	0.64	4.99
Water Consumed Cost $/hr	3.648	7.727	15.705
Steam Recover $/hr			

OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 11 OCTOBER 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

Optimization of methanation process is being continued. Process analysis and economic evaluation of the gas purification process, the shift reaction process, and the steam - carbon reaction process are being conducted. The information necessary for the analysis of these processes have been evaluated. The sensitivity study is now near completion and application to coal gasification processes is being made.

C. Y. Wen

C. Y. Wen, Project Director

OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 12, NOVEMBER 30, 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

The theoretical study on the sensitivity analysis and development of a technique useful for optimization in coal gasification processes are now completed. A report entitled "Sensitivity Analysis in Optimum Process Design for Coal Gasification Processes", Paper No. V is enclosed. The technique developed is now being used to analyze the sensitivity problems in methanation processes, purification processes, shift reaction processes, and hydrogen production processes.

Optimization of methanation processes is now being completed. A report is being prepared which will become available shortly. The results indicate that the cold quench process to be much superior for intermediate CO and high CO content feeds and only slightly superior for low CO feed. The recycle process gives the most inferior results, although for low CO feeds, the difference among the three processes, heat extraction, cold quenching, and recycle, is small.

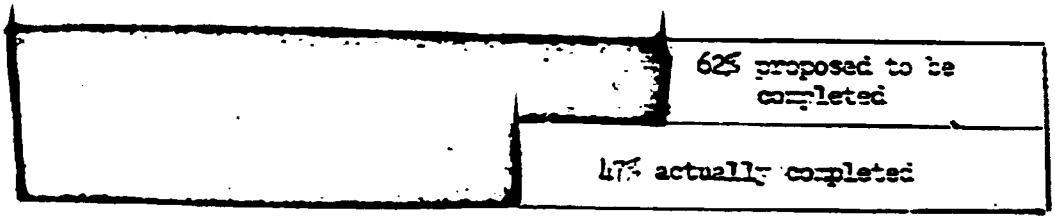
Models useful for gas purification processes are being established for optimization. Information gathering and assimilation are being continued. A chart indicating the progress of work for optimization on coal gasification progresses is enclosed.

C. Y. Wen

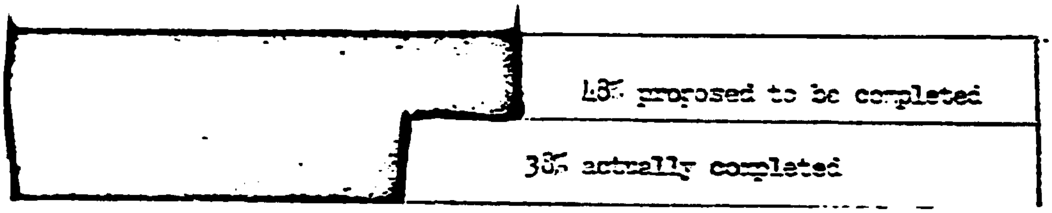
C. Y. Wen, Project Director

PROGRESS OF WORK FOR
OPTIMIZATION OF COAL GASIFICATION PROCESSES
NOVEMBER 30, 1967
OCR CONTRACT 14-01-0001-497

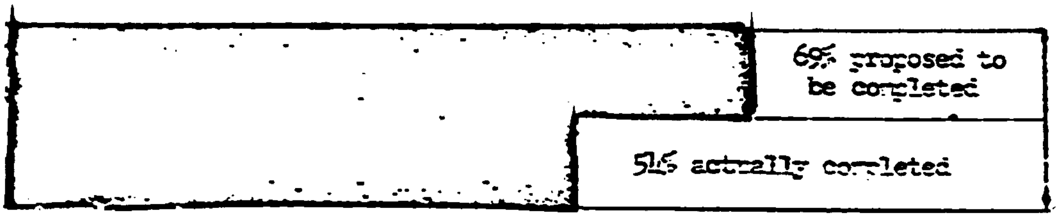
(A) INFORMATION GATHERING AND ASSIMILATION



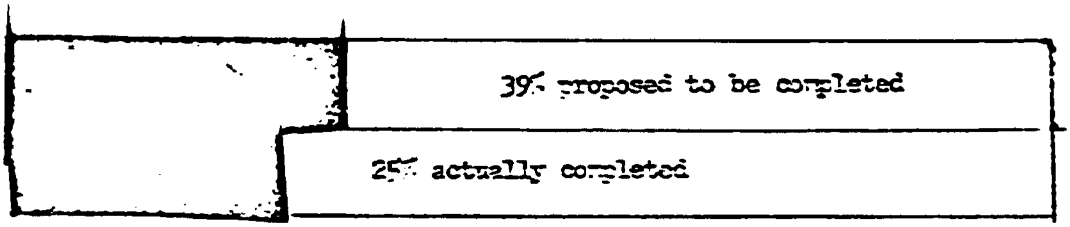
(B) SENSITIVITY STUDY



(C) DEVELOPMENT OF GENERAL OPTIMIZATION TECHNIQUE INVOLVING INFORMATION UNCERTAINTY



(D) ANALYSIS AND OPTIMIZATION OF VARIOUS PROCESSES



OPTIMIZATION OF COAL GASIFICATION PROCESSES
PROGRESS REPORT NO. 13 DECEMBER 31, 1967

to

Office of Coal Research
Contract No. 14-01-0001-497

A report of the optimization of methanation processes is being completed. The final draft is now being typed and copies will be made for distribution. The gas purification processes including the hot carbonate processes, MEADEA processes are being analyzed in terms of mathematical models useful for optimization. Detailed thermodynamic and kinetic data as well as physical constants necessary for computer programming have been assimilated. Also a kinetic study on water gas shift reaction processes and carbon steam reaction processes in the fluidized bed are being conducted. Information gathering and assimilation are being continued.

C. Y. Wen, Project Director

SATISFACTION GUARANTEED

NTIS strives to provide quality products, reliable service, and fast delivery. Please contact us for a replacement within 30 days if the item you receive is defective or if we have made an error in filling your order.

▶E-mail: info@ntis.gov
▶Phone: 1-888-584-8332 or (703)605-6050

Reproduced by NTIS

National Technical Information Service
Springfield, VA 22161

This report was printed specifically for your order from nearly 3 million titles available in our collection.

For economy and efficiency, NTIS does not maintain stock of its vast collection of technical reports. Rather, most documents are custom reproduced for each order. Documents that are not in electronic format are reproduced from master archival copies and are the best possible reproductions available.

Occasionally, older master materials may reproduce portions of documents that are not fully legible. If you have questions concerning this document or any order you have placed with NTIS, please call our Customer Service Department at (703) 605-6050.

About NTIS

NTIS collects scientific, technical, engineering, and related business information – then organizes, maintains, and disseminates that information in a variety of formats – including electronic download, online access, CD-ROM, magnetic tape, diskette, multimedia, microfiche and paper.

The NTIS collection of nearly 3 million titles includes reports describing research conducted or sponsored by federal agencies and their contractors; statistical and business information; U.S. military publications; multimedia training products; computer software and electronic databases developed by federal agencies; and technical reports prepared by research organizations worldwide.

For more information about NTIS, visit our Web site at http://www.ntis.gov.

NTIS

Ensuring Permanent, Easy Access to U.S. Government Information Assets

U.S. DEPARTMENT OF COMMERCE
Technology Administration
National Technical Information Service
Springfield, VA 22161 (703) 605-6000