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CALCULATIONS ON THE PERFORMANCE OF THE KALC PROCESS

M. E. Whatley

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CALCULATIONS ON THE PERFORMANCE OF THE KALC PROCESS

M. E. Whatley

APRIL 1973

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
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CALCULATIONS ON THE PERFORMANCE OF THE KALC PROCESS

M. E. Whatley

ABSTRACT

The KALC (Kr Absorption in Liquid CO_2) process for removing Kr from the burner off-gas generated in the reprocessing of HTGR fuel elements was shown to be feasible in a calculational study using a mathematical model in the form of a computer program. The KALC process uses liquid CO_2 to scrub Kr from the burner off-gas which contains about 90% CO_2, some O_2, N_2, and CO_2, and traces of Kr and Xe. It operates at pressures from 13 to 20 atm and temperatures from -20°C to -40°C. This study did not include Xe. The absorber-fractionator system, which separates Kr from the other light gases in the CO_2 solvent, was found to require close control for satisfactory performance. A good variable for adjusting performance, and the one used in this study, is the reboiler vapor rate at the bottom of the fractionator. This study also showed the need for some condensation at the feed point, and a molar scrub rate at least 20 times the feed rate. The stripper-rectifier system, which removes Kr and other light gases from the solution produced by the absorber-fractionator, was studied separately. This system showed no critical variables, performing better at higher reboiler vapor rates and easily yielding Kr concentrations of several percent in the product. The main condenser load for this system should be at the feed point rather than in the final condenser in order to keep flows in the rectifier section small. Significant variables in the operation of KALC were examined, and their effects on overall performance were noted and discussed.

---

1. SUMMARY

The KALC (Kr Absorption in Liquid CO_2) process was conceived specifically for the removal of Kr from the burner off-gas generated in the reprocessing of HTGR fuel elements. The process also has potential for removing other contaminants such as Xe, ^3H, I_2, and possibly particulates; in addition, it now seems possible that the KALC process or modifications thereof may be applied to off-gas problems beyond those for which it was initially designed. This report, however, deals only with the original

and singular purpose. The process requires that the feed gas be predominantly CO_2 plus, perhaps, 10% light gases such as O_2, N_2, CO, and contaminated with trace amounts of Kr. The gas is compressed and scrubbed with clean liquid CO_2 in an absorber tower above and connected to a fractionation tower through a condenser. A reboiler below the fractionator provides vapor which serves to fractionate the O_2, N_2, and CO from the Kr, producing a liquid CO_2 stream containing the Kr and only small amounts of the light gases. This liquid stream is fed to another system which removes the Kr and concentrates it. The stripper-rectifier system consists of a small, final condenser located above a rectifier tower which is separated from a stripper tower by a larger, main condenser. Below the stripper is a reboiler.

A computer program was written in which the best available equilibrium data on the solubilities of Kr, O_2, N_2, and CO in liquid CO_2 are incorporated. An equilibrium stage model was developed which sequentially and repetitively drew mass and enthalpy balances around each stage in the cascade while satisfying the equilibrium condition. The absorber-fractionator system was studied separately from the stripper-rectifier system. This report is basically an organization of many series of cases. Within each series all parameters were held constant except the reboiler vapor rate, which was varied from case to case. It was necessary to select from among all of the calculated variables those dependent variables which would best characterize system performance for correlating the results. The decontamination factor (DF) and concentration factor (CF) were selected for the principal dependent variables for the absorber-fractionator system, and the loss of Kr to the exit liquid CO_2 stream was chosen for the stripper-rectifier system.

It was established that the KALC process is feasible, at least on paper. In order to meet the requirement of a DF of several hundred and a CF of at least a thousand, the absorber should have at least 22 theoretical stages, the fractionator at least 16, the stripper at least 14, and the rectifier at least 6. The scrub rate must be at least 20 times the feed rate when the feed contains as much as 10% light gases. The fractionator reboiler vapor rate will be between 2.5 and 4 times the feed

rate for best operation and will require adjustment to within ±0.2% to maintain satisfactory operation under any given set of conditions. Condensers are required at the feed stage of both the absorber-fractionator system and the stripper-rectifier system.

The absorber-fractionator system operates most effectively at a pressure of about 20 atm, but the stripper performs better at about 15 atm or less. The performance of the stripper-rectifier is simpler than that of the absorber-fractionator system. It can be expected that the Kr content of the stripped liquid CO_2 (which serves as scrub for the absorber) will routinely be less than 10^{-6} times the Kr content of the stream entering the stripper. The other light gases will be more completely removed. About half of any Kr in the scrub stream will report to the decontaminated gas leaving the process. The required boilup rate for the stripper-rectifier system is about 30% of its feed rate (and perhaps six times the feed to the absorber). The bulk of the condensation load should be assumed by the feed stage condenser, leaving only about 10% of the heat for the final condenser, which operates at a significantly lower temperature.

The results of this study form an important link between the early, highly simplified calculations upon which the decision to pursue development was based and the experimental engineering development program that is just emerging. A more accurate understanding of the effects of the important design and operating variables will assist in the design of experiments, and subsequently, experimental data may be interpreted with the assistance of the computer model of the process.

2. INTRODUCTION

The KALC process is a significant part of the HTGR fuel recycle program. The original decision to study liquid CO_2 as a medium for fractionating the HTGR fuel reprocessing off-gas to effect its decontamination was made on the basis of highly simplified calculations using Raoult's law. Several such calculations indicated process feasibility. Since O_2, N_2, CO, and Kr would all be well above their critical temperatures at process conditions, the calculations required the choice of a

method for extrapolating vapor pressures; results based on the various methods all were somewhat different. None of these calculations took into account the nonidealities of the system. The only documented calculation was made by Leuze,[1] who assumed isothermal conditions. Other calculations, made assuming Dalton's law, accounted approximately for temperature variations within the towers. All of the calculations assumed constant molal overflow. The approximate nature of these calculations and their inherent uncertainty were understood at the time. The next step, however, was not simple. Improvement upon these preliminary calculations required a treatment which took heat effects into account and used actual equilibrium data.

Thermodynamic equilibrium data for O_2, N_2, and CO in CO_2 were accumulated from the literature. Distribution data on Xe are not yet available. Distribution data on Kr were obtained within Oak Ridge National Laboratory (ORNL) by Notz and Meservey.[2,3] At this point, work on the calculational analysis of KALC was temporarily stopped. The effort required to correlate the information into a model compatible with the computer program required for the calculation was subordinated to the more pressing task of initiating a meaningful experimental program.

It was intended that the feasibility of KALC be demonstrated in the Rare Gas Removal Pilot Plant at the Oak Ridge Gaseous Diffusion Plant[4] in a series of four campaigns. Unfortunately, this intention was not realized. Only one preliminary campaign of three weeks' duration was actually executed. It was the purpose of this campaign to gain experience in moving and handling liquid CO_2. Although it was not part of the original plan, radiokrypton was introduced during the last phases of this campaign. A few scouting runs gave, at best, only a rough indication of the probability of KALC's success. Significant DFs and product concentrations were not achieved simultaneously (they were achieved separately), and no reportable system performance data on the plant operation were produced.

Upon resumption of the calculational study of KALC, the most important objective was still to verify feasibility. Equilibrium and thermodynamic data were correlated and incorporated into a computer program model. The

program was used to study the effects of the number of stages, flow rate, temperature, operating pressure, etc., on the performance of the system. Results of these studies, which are presented in this paper, indicate the attainment of the objective.

The level of confidence that one may place in a theoretical study such as the one presented in this report merits discussion. It is hoped that computer calculations will subsequently be used in concert with the experimental program, resulting in a validation of conclusions from the calculations and providing a guide for the more expensive experimental work. In the interim, however, it should be noted that the calculations are as accurate as the assumptions upon which they are based. The more important of these assumptions are:

(1) The equilibrium stage model is valid.

(2) The literature equilibrium data on O_2, N_2, and CO are accurate.

(3) The Kr distribution data from Notz and Meservey are accurate.

In each case a high order of confidence is warranted.

Assumptions relative to the heat of solution of the light gases in liquid CO_2 are known to be in error. These are used only in enthalpy balances and, because of the small concentrations of light gases in the liquid phases, will affect the flow rates to a negligible extent.

If later information changes the magnitude of some numbers used to describe the system, the reported calculated numbers are expected to change only slightly. The conclusions presented here are thought to be at least qualitatively correct.

3. DESCRIPTION OF THE KALC PROCESS

The off-gas from the reprocessing of High Temperature Gas-Cooled Reactor (HTGR) fuel elements is unique among processing plant off-gases because the graphite and other carbon components of the HTGR fuel will be burned.[5-7] Hence the chief component of the off-gas is CO_2. Each HTGR fuel block produces about 11,000 scf. The burning operations are

performed at temperatures between 700 and 1000°C with a slight excess of oxygen to avoid the formation of CO. However, complete absence of CO cannot be assumed at this time. Commercial oxygen is used rather than air to avoid handling large quantities of N_2, but some inleakage of N_2 is unavoidable. Krypton, xenon, and tritium are quantitatively released in the burning step. Although significant amounts of particulates will be present in the burner off-gas, this presentation is not concerned with their removal.

The problems of concern here are the separation and concentration of the Kr. After a thorough literature search and an evaluation of alternatives, a process using liquid CO_2 as a solvent was selected for development. This process, called KALC, operates at pressures from 13 to 20 atm and at temperatures from approximately -40°C to -20°C.[3,8] The flowsheet, in simplified form, is shown in Fig. 1.

The basic system consists of an absorber-fractionator portion and a stripper-rectifier portion. In the absorber-fractionator system, the Kr is selectively absorbed in the liquid phase, while gas components with solubilities less than Kr (e.g., O_2, N_2, and CO) are returned to the gas phase and discharged from the top of the absorber along with much of the CO_2. Gases more soluble than Kr (e.g., Xe) follow the liquid CO_2 to the stripper-rectifier portion where the Kr is removed and concentrated.

The CO_2 from the bottom of the stripper would contain components of low volatility. Xenon could be allowed to accumulate in the CO_2 to the point where it would eventually be discharged from the top of the absorber, or it could be removed by a separate distillation operation on the recycle CO_2 stream. Since at this writing no data are available on the behavior of Xe in a CO_2 system, Xe is omitted from the analysis. Because water is very much less volatile than CO_2, it is expected that tritium would be concentrated in stripping; this subject is also ignored here.

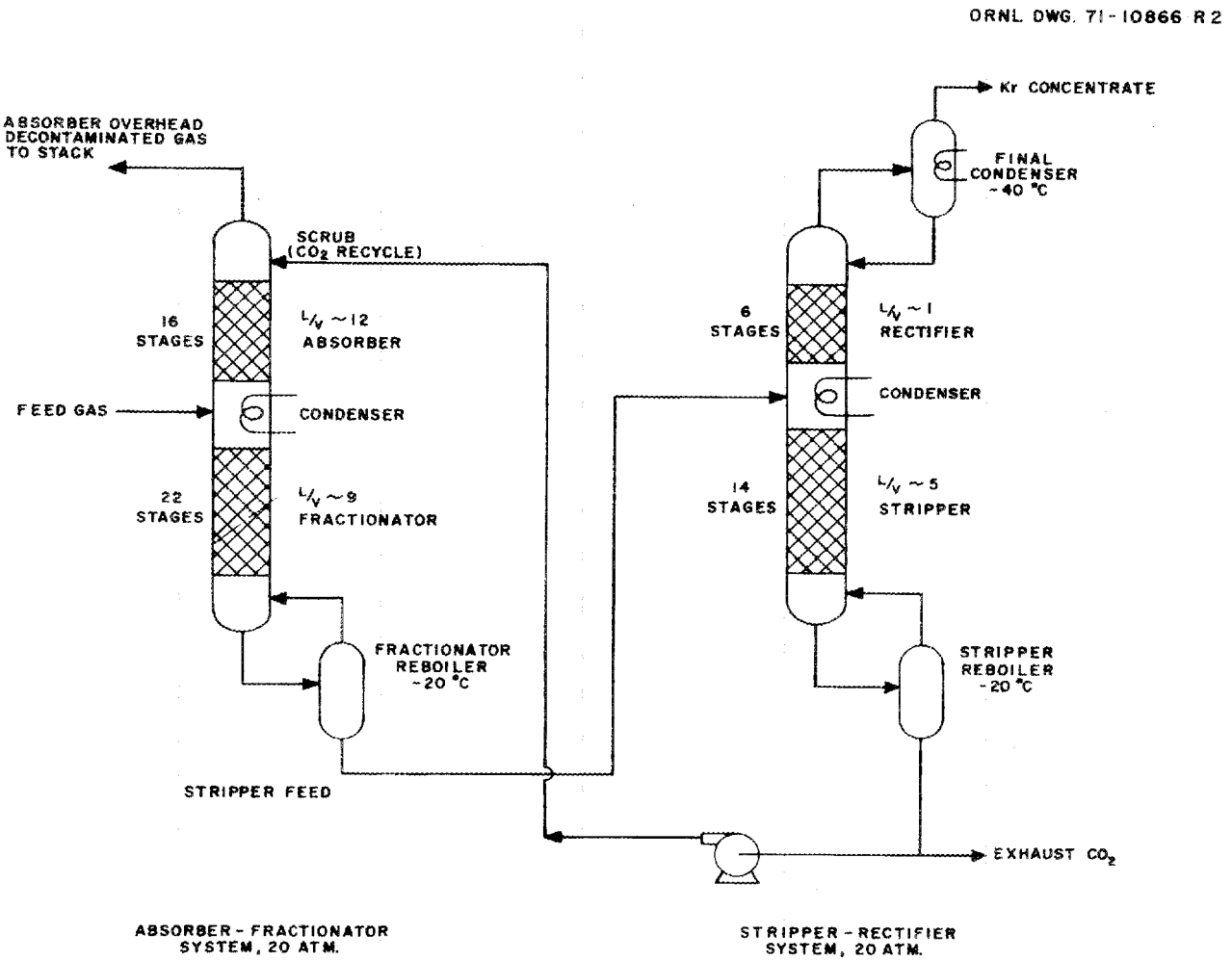

Fig. 1. Simplified Flowsheet of the KALC Process.

4. EQUILIBRIUM DATA

4.1 Solubilities of O_2, N_2, and CO in Liquid CO_2

The literature provided a reasonable amount of data on the solubility of O_2 in liquid CO_2,[9-11] somewhat less on N_2 in CO_2,[9,11,12] and only a very small amount on CO in CO_2.[13] No data on Xe in CO_2 were found. Most of these data were obtained at high concentrations, and hence at higher pressures, than are of interest to this work. When the light gases comprise less than 50% of the gas phase, at total pressures less than 50 atm, the liquid phase contains less than 10% dissolved light gases. As the concentration of light gases decreases from this level, data become sparse and their accuracy diminishes; however, theoretical considerations have helped us to extrapolate the available information into low-concentration regions.

Equilibrium data from the literature have been fitted to a model which, over the range of conditions of interest, describes the KALC system quite satisfactorily. Of necessity, this model is based on binary system data. The only ternary data obtained under conditions close to those used for the KALC system were for the O_2-N_2-CO_2 system at pressures higher than 50 atm. Even though they were out of the proper range, these data and equilibrium numbers calculated from the model were in reasonable agreement. The model is most simply expressed in the following form: given a liquid phase composition and a temperature, calculate the total pressure and the vapor phase composition.

The total pressure was found to fit the empirical relation

$$P_t = P^*_{CO_2} + \Sigma a_i x_i + \Sigma b_i x_i P^*_{CO_2} + \Sigma x_j \Sigma c_i x_i , \qquad (1)$$

where

P_t = total pressure (atm),

$P^*_{CO_2}$ = saturated vapor pressure of CO_2 at system temperature (atm),

x = mole fraction in liquid phase,

i,j = index subscripts applied to all components <u>except</u> CO_2,

a,b,c = empirically determined constants.

The compressibility of CO_2 vapor is such that a correction for nonideality is required. All other components have critical points sufficiently removed from KALC conditions that gas-phase ideality can be assumed. A reasonable form for the gas-phase composition then becomes:

$$y_{CO_2} = (p_{CO_2}/Z)/[\Sigma p_j + (p_{CO_2}/Z)] \tag{2}$$

$$y_i = p_i/[\Sigma p_j + (p_{CO_2}/Z)], \tag{3}$$

where

Z = the compressibility factor for saturated CO_2 at the temperature of the system,

p = partial pressure,

y = mole fraction in the vapor phase.

The partial pressures are defined so that

$$P_T = p_{CO_2} + \Sigma p_i. \tag{4}$$

Because the concentration of the dissolved light gases is small, Raoult's law was applied to the CO_2. It was necessary, however, to make a Poynting-type correction of the CO_2 vapor pressure. As an expedient in obtaining a better fit, the molar volume of the liquid phase was not used; instead, a fictitious molar volume about a factor of 3 times the real molar volume was employed.

The partial pressure of CO_2 is, then,

$$p_{CO_2} = P_{CO_2} (1 - \Sigma x_j), \tag{5}$$

where

$$P_{CO_2} = P^*_{CO_2} \exp [k(P_t - P^*_{CO_2})/T], \tag{6}$$

and

P_{CO_2} = effective vapor pressure of CO_2 (atm),

T = absolute temperature (°K),

k = a determined constant = 1.7 °K/atm.

The partial pressure of the ith component is given by:

$$p_i = \phi_i[1 + (P^*_{CO_2} - P_{CO_2} (1 - \Sigma x_j))/\Sigma\phi_j], \tag{7}$$

where

$$\phi_i = a_i x_i + b_i x_i P^*_{CO_2} + c_i x_i \Sigma x_j \ . \tag{8}$$

For a binary system, Eq. (7) reduces to:

$$p_i = P_t - (1 - x_i) P_{CO_2} . \tag{9}$$

These equations comprise a set from which equilibrium can be calculated. Table 1 gives the numerical values for the constants, along with the expressions used in this study to compute the compressibility factor, Z, and the saturated vapor pressure of CO_2, $P^*_{CO_2}$.

4.2 Solubility of Krypton in Liquid CO_2

Accurate understanding of the behavior of Kr in the system is essential to the success of a computer simulation of the KALC process. Since the beginning of the work on KALC we have had Kr distribution data from KFA Jülich. However, deviations of these data from certain theoretical predictions[14] made us sufficiently skeptical to undertake our own experimental determination. Measurements, made at ORNL by Notz and Meservey,[2,3,7] disagreed significantly with the Jülich data.[15] The ORNL technique involved an in-situ measurement, counting tracer ^{85}Kr, and results of replicate runs were in good agreement. Validity of the technique was established by comparing a determination of the solubility of Kr in water with previous measurements reported in the literature; the values checked closely. Further, the data on Kr distribution from Notz and Meservey are not inconsistent with applicable theoretical predictions. These data are shown in Fig. 2.

The distribution data on Kr were taken at tracer level and reported as $K = y/x$. It was necessary, for incorporation into the computer program, that these data be expressed in the same form as used for the other light gases. By employing Eqs. (3), (9), and (6), and neglecting terms that vanish at small values of x, we can write

$$\frac{y}{x} = K = \frac{P^*_{CO_2} \left(1 - \frac{k\phi}{Tx}\right) + \frac{\phi}{x}}{(P^*_{CO_2})/Z} , \tag{10}$$

Table 1. Coefficients Used to Correlate Equilibrium Data for the KALC System

Component i	a	b	c
O_2	532.33	-2.7153	-494.72
N_2	720.22	-5.9729	-553.88
CO	484.39	-1.8473	-395.10
Kr	188.0	2.80	+3000.0

Total pressure = $P_t = P^*_{CO_2} + a_i x_i + b_i x_i P^*_{CO_2} + c_i x_i \Sigma x_j$.

Saturation vapor pressure of CO_2:

$$\ln (P^*_{CO_2}) = -8.175626 + 26.27742 \left(\frac{T}{600}\right) - 23.76839 \ln \left(\frac{T}{600}\right) - 8.62709785 \left(\frac{600}{T}\right).$$

Compressibility of CO_2 = $Z = 3.21203 - 0.029634967T + 1.3757 \times 10^{-4} T^2 - 2.2960 \times 10^{-7} T^3$.

T = absolute temperature (°K).

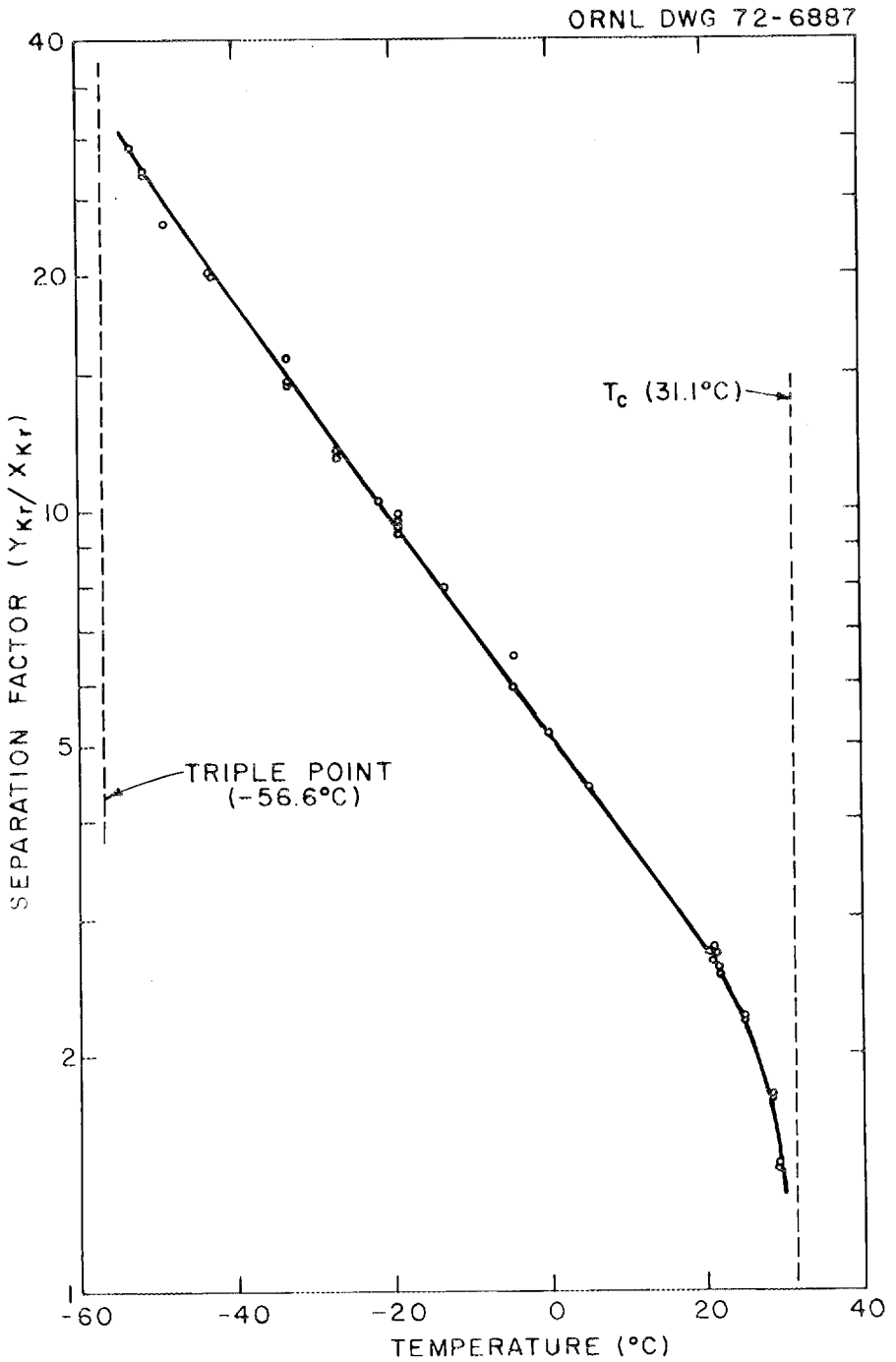

Fig. 2. Equilibrium Distribution of Krypton Between Liquid and Vapor Phases in Saturated CO_2.

where, for Kr,

$$\phi = ax + bx\, P^*_{CO_2} + cx^2. \tag{8a}$$

Clearly, the term cx^2 must be neglected; thus, by solving Eq. (10) for ϕ/x, we obtain:

$$\frac{\phi}{x} = a + b\, P^*_{CO_2} = \frac{P^*_{CO_2}\left(\frac{K}{Z} - 1\right)}{1 - P^*_{CO_2}\frac{k}{T}} \quad . \tag{11}$$

The term on the right in Eq. (11) can be evaluated from the data presented, making it possible to evaluate the constants a and b for Kr by using standard regression techniques. The constant c is relatively unimportant, although a large positive value for it apparently tends to keep the value of K constant in the presence of large amounts of other light gases. A few exploratory experimental data points are indicative of this effect. The constants for Kr shown in Table 1, when applied through the computer equilibrium program, reproduce the curve shown in Fig. 2 within experimental error over the temperature range -45 to 0°C.

4.3 Mathematical Model

The computer program, QUEST, simulates the KALC system using an equilibrium stage model. The definition of the system for the calculation includes the number of stages above and below a feed point at which a condenser can be simulated by appropriately specifying the enthalpy of the feed. The feed rate is taken to be unity, forming the basis for other rates in the system. A feed composition is required. Conditions at the top and bottom of the cascade are specified from a set of alternatives which provide for the simulation of either an absorber-fractionator combination or a stripper, but not both simultaneously. Isobaric operation is assumed at a specified pressure. Details on input to the calculation are given in Appendix A.

A profile of the variables for each stage of the system is updated repeatedly until steady state is adequately approached. Stored variables include, for each stage: temperature, vapor and liquid rates, vapor and

liquid enthalpies, and vapor and liquid compositions. The profile is updated starting at the bottom and working up. For the calculation of each stage, vapor is received from the stage below and liquid is received from the stage above. Enthalpy and mass balances are imposed, and the equilibrium conditions are satisfied, yielding again the temperature, the vapor and liquid rates, and the composition of each phase.

Since the standard form of the equilibrium equations is such that the pressure and the vapor composition are explicit in temperature and liquid-phase composition (and since the equations cannot be made explicit in pressure, gross enthalpy, and gross composition), an iterative method was employed to solve the implicit relations. Temperature was taken as the ultimate variable for iteration. The equations were rearranged to incorporate the material balance into the equilibrium equations. The fraction of the vaporized material was calculated from an enthalpy balance; then the vapor pressure of CO_2 was extracted as a function of other system variables and used to correct the assumed temperature. The computer program to effect this is found in subroutine QUEST (see Appendix C). At the top of the cascade for certain cases, and also occasionally at the bottom, it is desirable to specify a fraction of the material reporting to the vapor phase instead of honoring the enthalpy balance. This is equivalent to specifying a reflux ratio or a boilup ratio. Subroutine QUEND serves this purpose. The algorithm is similar to that used in QUEST, but simpler.

When the light gases comprise less than 0.003 mole fraction of the material, simplifying assumptions which allow more rapid convergence on stage equilibrium are possible. Subroutines LISWT and LILEND (see Appendix C) supply this need. The vapor pressure of CO_2 is written as a FUNCTION, as is its reciprocal, the temperature at which CO_2 has a specified pressure. The compressibility factor of CO_2 vapor is also written as a FUNCTION. The enthalpies are given by subroutine ENTHAL, and the temperature at which the liquid phase has a specified enthalpy is given by subroutine TENTH.

Since it was found that the cascade profile converged slowly when the number of stages exceeded about 30 and when conditions were conducive to reflux, a subroutine was incorporated to periodically extrapolate each concentration either to an anticipated steady-state value or by several hundred iterations, depending upon the manner in which it was changing. This is subroutine ZAPPO. Control of the stage calculation is in subroutine STAGE; and control of the cascade calculation is in subroutine STEP, which also includes the convergence criteria. Input and output are in separate subroutines. The MAIN program is essentially executive.

5. STRUCTURE OF THE PARAMETRIC STUDY

The absorber-fractionator system was decoupled from the stripper-rectifier system for this study. By expending sufficient computer time, it would have been possible to consider the entire system simultaneously; nevertheless, for the reasons given below, this was not done.

In KALC, the absorber is coupled to the stripper by the scrub stream composition. Normally, Kr is completely removed by the stripper and this stream is pure CO_2. Under these circumstances the absorber-fractionator system is independent of the stripper. Only if the stripper returns significant amounts of Kr to the absorber is the coupling significant. Any O_2, N_2, or CO in the stream will be thoroughly swamped by the amounts of these components already present at the point of entry of the scrub, and can be of no consequence. If Xe had been included, it would have been transported between the two systems in large amounts, thereby complicating the calculation. Here, the effect of Kr in the scrub stream was treated as a system parameter.

The absorber-fractionator system supplies a feed stream to the stripper-rectifier system. This, again, was readily characterized by treating the stripper feed as a system parameter. Analysis of the stripper system is quite straightforward and simple in comparison with that of the absorber-fractionator system. The latter system will, therefore, receive the bulk of our attention.

5.1 Absorber-Fractionator

When the feed and scrub compositions are fixed, eight remaining independent variables (see Fig. 3) fix the system. The stages above and below the feed point comprise two of the variables; a third is the feed rate, which is arbitrarily taken as unity. The remaining five, which are essentially operating variables, include: system pressure, feed enthalpy (or intertower condenser load), scrub rate, scrub temperature, and boilup rate in the fractionator reboiler.

Variations on these are possible, widening the field of ways that the program can be used. Clearly, it is necessary to have some system of exploration which will allow characterization of the principal effects by a reasonable expenditure of effort. At the beginning of the study, it was thought that an objective function might be invented quantifying the desirable and undesirable attributes of each variable in a pseudo-economic form so that an optimization calculation could be made, producing the one best case. It was quickly realized that the status of the development of KALC and its application precluded this approach.

The selection of meaningful dependent variables involved a degree of arbitrariness. The use of the Kr DF, defined as

$$DF = \frac{\text{Kr entering with feed}}{\text{Kr leaving in effluent gas}} ,$$

is obvious. When the scrub stream contains Kr, this definition could yield values less than unity.

A complementary dependent variable indicating the extent to which the other light gases are separated from the Kr was required. For this purpose, a Kr CF was defined as follows:

$$CF = \left(\frac{\text{Kr leaving bottom stage}}{\text{Other light gases leaving stage}}\right) \Big/ \left(\frac{\text{Kr in feed}}{\text{Other light gases in feed}}\right).$$

This definition is less seriously confused by the presence of Kr in the scrub. Good system performance requires that both the CF and the DF for Kr be high. Generally, a change in an independent variable results in an increase in one of these factors and a decrease in the other. Since their relative importance has not been established, most of the results of this study are reported over a range encompassing the region of interest.

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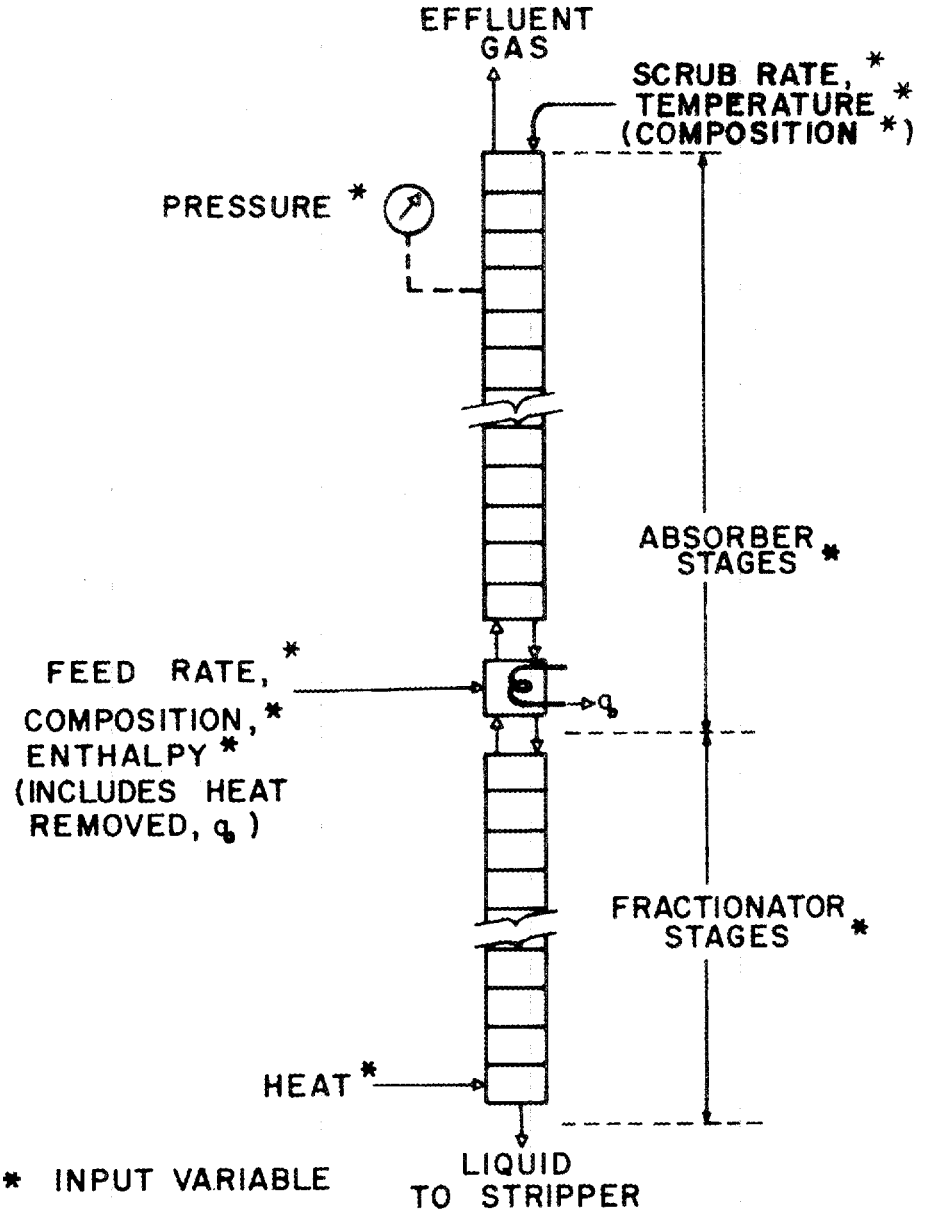

Fig. 3. Schematic Identification of Variables in Calculations on the Absorber-Fractionator System.

The feed composition was arbitrarily fixed. While this decision at first seems to neglect an important variable, its rationale is straightforward. The light gases behave similarly in the KALC system. The solubilities of O_2 and CO are quite similar, while N_2 is slightly less soluble in liquid CO_2. Since some of each gas was included, the relative behavior of the gases is observable. Krypton is present in such small amounts that the magnitude of its concentration is unimportant. The total light-gas content of the feed is important; however, since the ratio of the scrub rate to the feed rate is generally high, the effect of the light-gas content of the feed can be inferred directly from the studies in which the scrub rates are varied. Specifically, the effect of increasing the scrub rate by a factor of 2 is similar to that of decreasing the light-gas components in the feed by a factor of 2.

The number of stages in the system is a very important variable. Early attention was devoted to establishing a least number of stages which would effect a significant separation, and to gaining an appreciation for the number that might be required in applications of the KALC process. Since these studies were made while reasonable operating domains were being established for the variables (and while the program was being debugged), few of the results are presentable. Among the facts which emerged are that the system performance is markedly improved with a larger number of stages and that calculation time is greatly increased with a larger number of stages. Accepting the former, but in deference to the latter, it was decided to undertake the parameter study of the operating variables using a system with sufficient stages to give meaningful results (if not adequate to meet final KALC specifications) but not so large as to incur excessive computer charges. The standard system used 12 absorber stages and 12 fractionator stages. This system usually had a region in which the DF and the CF were each about 100. The parameter study was supplemented by calculations for systems having a larger number of stages.

It was found that the KALC system, like a large class of separation processes, had to be "tuned" to yield both satisfactory recovery and concentration. When one variable was displaced from a good operating

point, another could be altered to bring the system back to a different, satisfactory operating point. Or, if all variables but one were arbitrarily fixed, that one could be adjusted to bring the system to a relatively "best" operating point. In order for the parametric study to be meaningful, the system had to be compared at "best" operating points. This required the selection of an adjusting variable, or, as it is designated here, a "primary" variable. For this study, the boilup rate in the fractionator reboiler, V_B, was attractive. A "series" of "cases", then, consisted of a set of runs holding every parameter except V_B constant and allowing V_B to vary from case to case through the region of "best" operation. One additional aspect of the primary variable must be explained. The program is written to accept a heat input to the bottom stage, and not a vapor rate from that stage. Consistently throughout our study, this heat input was specified in terms of an equivalent number of moles of CO_2 converted from a liquid to a gas at saturation at the pressure of the system. Such a number is handy and unambiguous but actually equals the vapor rate leaving the bottom stage only when the liquid entering the bottom stage is essentially at the saturation temperature of pure CO_2. The primary variable is, therefore, the nominal V_B^*. The difference between V_B and V_B^* is treated further in Appendix B.

By restricting the scope of the study and establishing the format by which it was conducted, the parameters to be investigated were established. The most significant were the scrub rate and the enthalpy of the feed (which, as previously stated, includes the heat removal rate from the condenser between the absorber and the fractionator). The temperature of the scrub proved to be important in determining the amount of the CO_2 leaving with the effluent gas. It was studied by comparing operation at constant scrub temperature with that at a specified effluent gas rate. The effect of system pressure was established in a straightforward study, as was the effect of Kr in the scrub stream.

Table 2 lists the standard feed compositions for all series of runs. Table 3 is a summary of all series of runs made in the absorber-fractionator study; it lists all input conditions and a few of the important results.

Table 2. Standard Feed Compositions Used in Calculations

A. Absorber-Fractionator

Constituent	Mole Fraction
O_2	0.06
N_2	0.03
CO	0.005
Kr	0.00001
Xe	0.000
CO_2	0.905

B. Stripper-Rectifier

Constituent	Mole Fraction ($x10^6$) for Feed Type: I	II	III
O_2	1.0	0.1	0.1
N_2	0.4	0.04	0.1
CO	1.2	0.12	0.1
Kr	0.4	0.04	0.7
Xe	0.0	0.0	0.0
Total	3.0	0.30	1.0

Table 3. Summary of the Series of Runs Calculated for the Absorber-Fractionator System

Series	Stages in Absorber	Stages in Fractionator	System Pressure (atm)	Feed Type	Feed Enthalpy (cal/mole)	Scrub Rate	Scrub Temp. (°C)	Effluent Vapor Rate (at DF=CF)	"Best" V_B^* (at DF=CF)	DF=CF ("Best Pt.")	V_B^* When DF=100	CF When DF=100
24A001	12	12	20	Std.	-200	22	-30.0	0.907	2.786	88	2.775	48
24A002	12	12	20	Std.	+800	22	-30.0	0.920	2.460	63	2.375	12
24A003	12	12	20	Std.	-1000	22	-30.0	0.902	3.051	102	3.050	100
24A004	12	12	20	Std.	-2000	22	-30.0	0.899	3.385	120	3.389	250
24B003	12	12	20	Std.	-1000	26	-30.0	0.950	3.404	132	3.41	260
24C003	12	12	20	Std.	-1000	30	-30.0	0.988	3.749	155	3.757	340
24D003	12	12	20	Std.	-1000	34	-30.0	1.025	4.090	174	4.107	370
24E003	12	12	20	Std.	-1000	16	-30.0	0.813	2.503	59	2.378	7.6
24F003	12	12	20	Std.	-1000	18	-30.0	0.848	2.694	73	2.627	14
24A903	12	12	20	Std.	-1000	22	-29.988	0.90	3.051	102	3.050	100
24B903	12	12	20	Std.	-1000	26	-30.340	0.90	3.418	135	3.427	270
24C903	12	12	20	Std.	-1000	30	-30.574	0.90	3.780	162	3.794	398
24F903	12	12	20	Std.	-1000	18	-29.417	0.90	2.676	70	2.553	9.4
24B913	12	12	20	Std.	(-1000)	26	-30.34	0.90	(3.418)	132	—	250
24B923	12	12	20	Std.	-1000	26	-30.34	0.90	3.418	135	3.427	270
24P9AA	12	12	15	Std.	-1000	26	-36.893	0.90	2.710	143	2.719	900
24P9AB	12	12	25	Std.	-1000	26	-25.225	0.90	4.255	83	4.240	56.4
24B9F3	12	12	20	0.5 x Std.	-1000	26	-27.618	0.90	2.985	200	3.28	400
24B9G3	12	12	20	1.444 x Std.	-1000	26	-32.386	0.90	2.731	86	3.677	28
38B903	22	16	20	Std.	-1000	26	-30.792	0.90	3.499	1800	3.532	70,000

5.2 Stripper-Rectifier

The performance of the stripper-rectifier system is much less complex than that of the absorber-fractionator system, primarily because Kr is much easier to separate from CO_2 than from O_2. The format for the stripper study is similar to the absorber-fractionator study in that the heat input to the reboiler at the bottom of the stripper (V_B^*) was taken as the primary variable, and each of the other variables had its counterpart in the other system. The number of stages in the stripping section, the number of stages in the rectifying section, the system pressure, the feed composition, the enthalpy of the feed, and the effluent gas rate were considered as parameters in the study. The effluent gas rate was the only variable associated with the top of the cascade since, in the stripper-rectifier system, there is no entering scrub stream. The specification of an effluent stream as an input to an iterative calculation such as is used for this study can pose some special problems. The method by which the program used this input variable is explained in Appendix B.

The only meaningful dependent variable in the stripper system study was the fraction of the Kr entering the stripper-rectifier system which reported to the liquid stream leaving the bottom of the system (referred to as the Kr "loss"). This is a consequence of restricting the domain of feed compositions to those likely to be produced by the absorber-fractionator system (light gases, $\sim$1 ppm) and the domain of Kr losses to realistic values (losses, <1%). The O_2, N_2, and CO are all less soluble than Kr in liquid CO_2, and their losses will be much less than the Kr loss. It is clear that the effluent gas composition (and the concentration factor implied by it) is fixed by and immediately calculable from the input variables.

The feed compositions used for the study are listed in Table 2. Table 4 is a summary of all series of runs made in the stripper-rectifier study; it lists all of the input conditions and some of the results, the significance of which will be covered in the subsequent discussion.

Table 4. Summary of the Series of Runs Calculated for the Stripper-Rectifier System

Series	Stripping Stages	Rectifying Stages	System Pressure (atm)	Feed Type	Feed Enthalpy (cal/mole)	Concentration Factor ($\times 10^{-6}$)	-A	exp (A) ($\times 10^{-10}$)	-B	Mean Error in Fit (%)	Kr Loss ($V_B^* = 0.3$) (%)
S16B121	12	4	20	I	100	0.06666	24.736	0.181	11.973	5.0	0.0717
S16B122	12	4	20	I	100	0.10	24.212	0.305	11.795	3.6	0.0945
S16B123	12	4	20	I	100	0.13333	23.889	0.422	11.700	2.7	0.1149
S16B131	12	4	20	I	800	0.06666	23.571	0.580	11.014	4.0	0.0624
S16B132	12	4	20	I	800	0.10	23.363	0.714	11.085	6.1	0.0856
S16B133	12	4	20	I	800	0.13333	23.180	0.857	11.101	4.9	0.1059
S16B141	12	4	20	I	1200	0.06666	23.734	0.493	11.107	4.7	0.0614
S16B142	12	4	20	I	1200	0.10	23.390	0.694	11.090	4.9	0.0845
S16B143	12	4	20	I	1200	0.13333	23.153	0.881	11.073	5.2	0.1049
S16B231	12	4	20	II	800	0.06666	23.789	0.466	11.287	1.8	0.0729
S16B232	12	4	20	II	800	0.10	23.384	0.699	11.269	1.8	0.1066
S16B233	12	4	20	II	800	0.13333	23.097	0.931	11.256	1.9	0.1395
S16B331	12	4	20	III	800	0.06666	23.978	0.386	11.288	1.7	0.0604
S16B332	12	4	20	III	800	0.10	23.597	0.565	11.267	1.7	0.0859
S16B333	12	4	20	III	800	0.13333	23.340	0.731	11.257	1.8	0.1095
S14B122	12	2	20	I	100	0.10	18.607	82.998	10.789	7.6	6.671
S15B122	12	3	20	I	100	0.10	21.397	5.096	11.258	1.6	0.765
S17B122	12	5	20	I	100	0.10	27.239	0.0148	12.792	3.5	0.0215
S19B122	12	7	20	I	100	0.10	30.637	0.000495	14.726	3.9	0.0123
S21B122	12	9	20	I	100	0.10	30.742	0.000446	14.793	3.8	0.0122
1S16P122	12	4	18	I	100	0.100	25.740	0.0663	11.723	0.88	0.0192
2S16P122	12	4	15	I	100	0.100	28.634	0.00367	11.814	0.35	0.00122
3S16P122	12	4	25	I	100	0.100	23.664	0.528	14.252	40.5	9.32
S22BB122	16	6	20	I	100	0.10	38.636	0.0000002	18.156	5.2	0.00064
S22BB132	16	6	20	I	800	0.10	36.581	0.0000013	15.955	4.3	0.000196

6. STUDY OF THE ABSORBER-FRACTIONATOR SYSTEM

The effects produced by changing each of the important variables in the absorber-fractionator system will now be considered separately. The first would logically be the primary variable around which the study is built.

6.1 Effect of Boilup Rate on the Absorber-Fractionator System

As a primary variable for this study, the boilup rate from the bottom stage proved to be quite satisfactory. The CFs and DFs for two series of runs, believed to be nearly optimum for the KALC system, are shown in Fig. 4. Series 38B903 used 22 stages in the absorber and 16 stages in the fractionator, and series 24B903 used 12 stages in the absorber and 12 stages in the fractionator. This figure serves to illustrate the effect of increasing the number of stages as well as the effect of boilup rate. The two series are identical with regard to scrub rate, feed composition, feed enthalpy, and system pressure.

It is interesting to note a slight shift in the location of the break in the curves when the number of stages is increased. This is due to the difference in operating temperatures caused by the increased reflux possible with the additional stages.

For boilup rates below optimum, the DF is high and the CF is low. As the optimum point is reached, the CF increases sharply and the DF drops precipitously. At boilup rates greater than optimum, the DF is low and the CF is high. There is no region where the highest values of both the DF and the CF factor are realized; a relatively narrow region exists where both are acceptable. Difficulties in process control may be implied by this, but fortunately, system temperatures follow system performance sufficiently to provide an index for control. It should be mentioned in passing that, if the control problems associated with the operation of a single absorber-fractionator unit become intractable (an unexpected situation), two KALC units could be used to achieve high performance by operating in series on different sides of the critical boilup rate.

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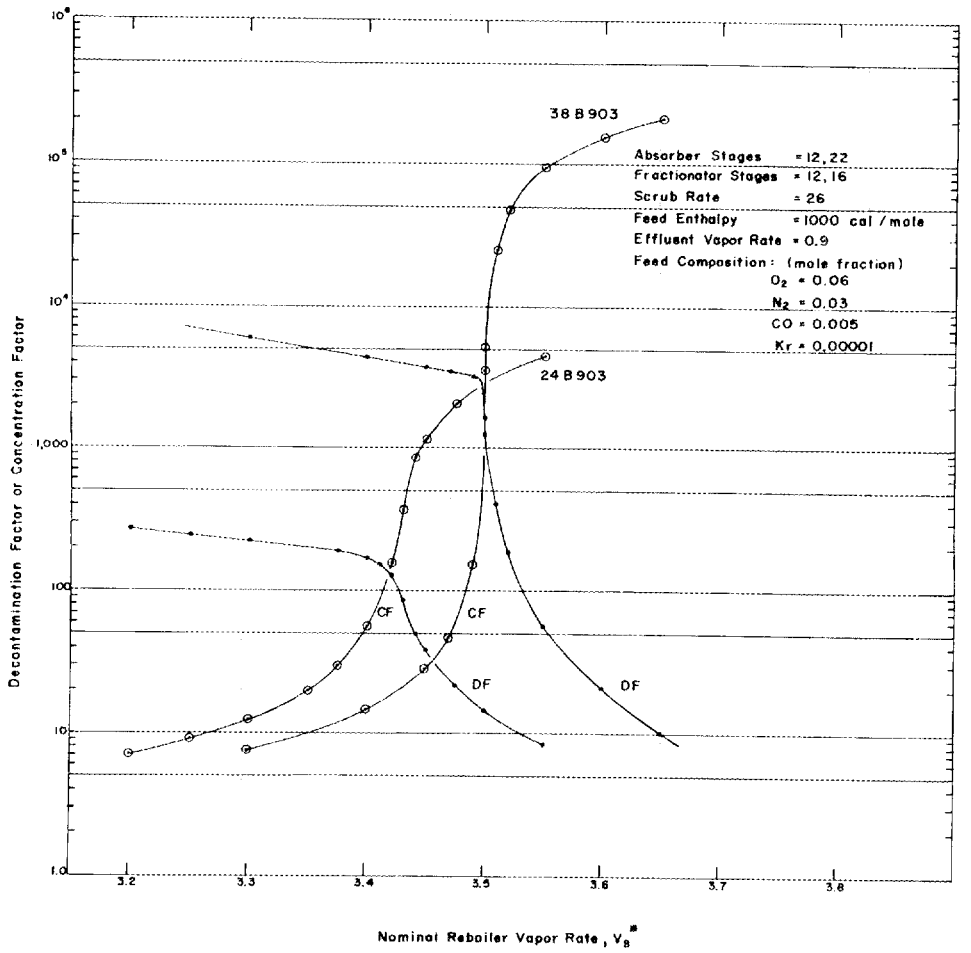

Fig. 4. Decontamination Factor and Concentration Factor Dependence on Nominal Reboiler Vapor Rate for Two Series with Different Numbers of Stages.

The unacceptable stream from the second unit could be recycled back to the first. Since the necessity for multiple units seems improbable, the problem receives no further attention in this report.

The explanation of the sharp change in performance of the system at a critical boilup rate is not simple. Figures 5-7 show the concentration profiles for three successive cases (25, 26, and 27) of series 24B003 in the critical region. The boilup rates are 3.39, 3.40, and 3.41 moles per mole of feed, respectively. Series 24B003 is identical to series 24B903 except that the scrub temperature was held at -30°C rather than the effluent gas rate being held at 90% of the feed rate. The resulting effluent gas rates were 0.939, 0.946, and 0.953. It is established elsewhere that this change has a negligible effect on the system. It can be seen that the Kr concentration falls to very low values at the top of the absorber-fractionator and that the light gases O_2, N_2, and CO_2 fall to very low concentrations at the bottom. The light gases are refluxed and accumulate in the proximity of the feed point. Krypton, on the other hand, accumulates at a point about one-third of the way down the fractionator. The accumulation of the light gases causes the temperature to decrease, which, in turn, affects the distribution of both the light gases and Kr. In Fig. 5, the lower boilup rate permits a higher reflux of light gases, which raises their profiles in the fractionator and allows more light gases to accompany the Kr. As the boilup rate is increased, the concentration profiles for the light gases are lowered, resulting in higher CFs. However, the concentration profile for krypton is raised by about 25%, resulting in the discharge of 25% more Kr from the top of the absorber. Although the differences among Figs. 5-7 appear slight, they do, in fact, account for an increase in CF from 55.8 to 261 and a decrease in DF from 164.5 to 99.5.

The effect of small increases in the boilup rate over this critical region is also emphasized by the temperature profiles shown in Fig. 8. The temperatures in the center of the column are about -32°C, while those at the bottom of the fractionator and on top of the absorber are -19°C and -24°C, respectively. The differences between temperatures for cases 25-27 are apparent, and each generally coincides with the appropriate

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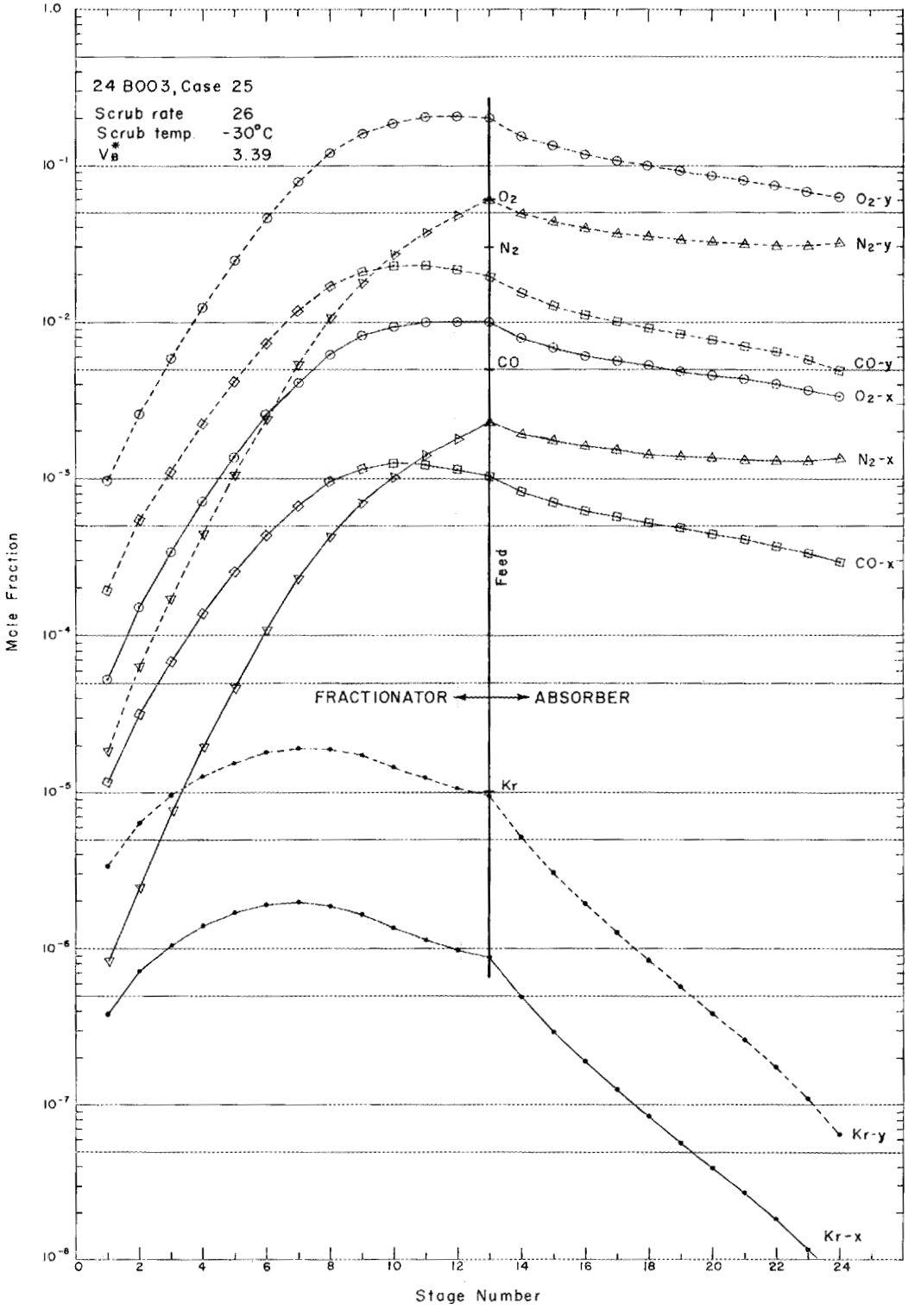

Fig. 5. Concentration Profiles from Series 24B003, Case 25.

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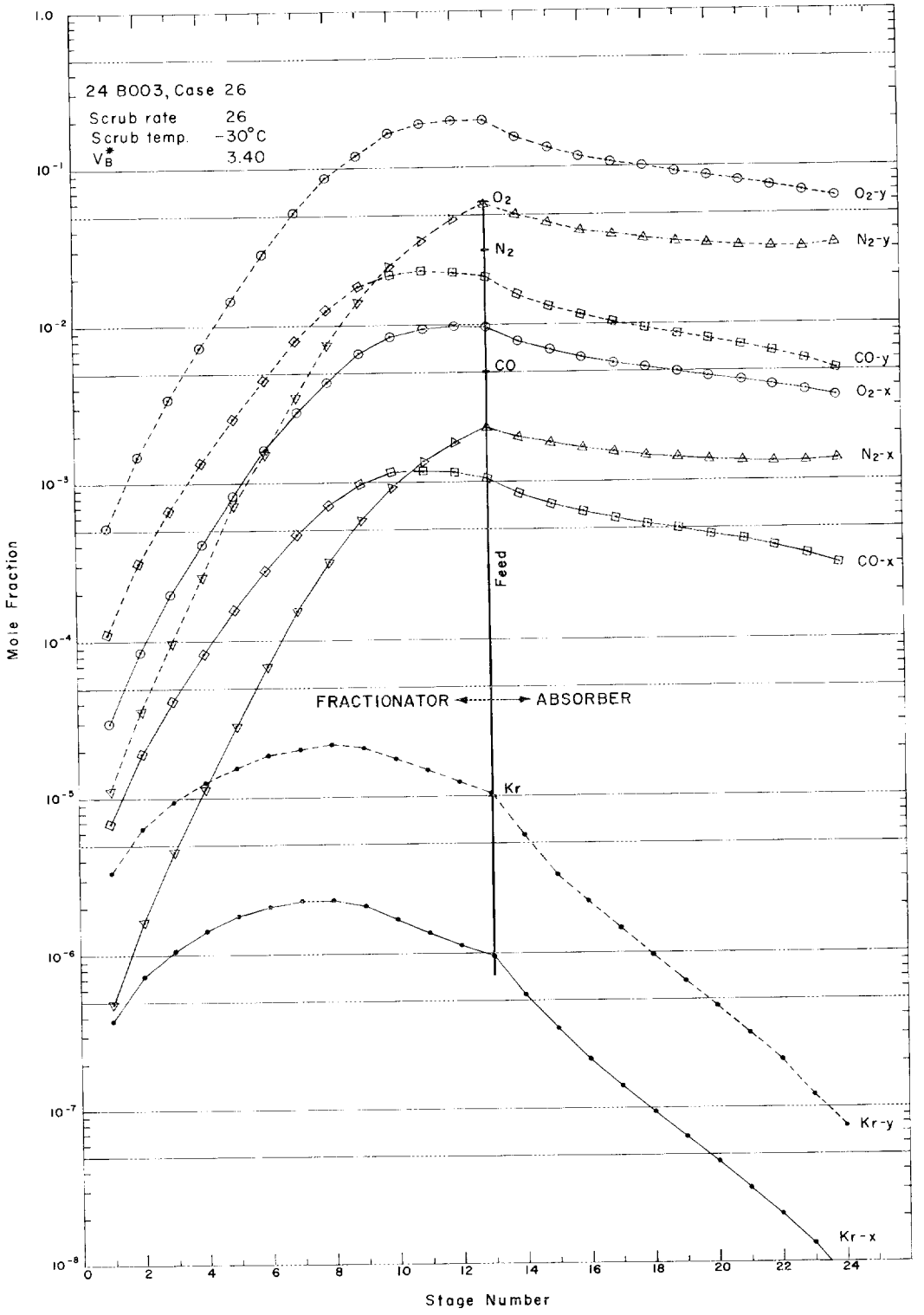

Fig. 6. Concentration Profiles from Series 24B003, Case 26.

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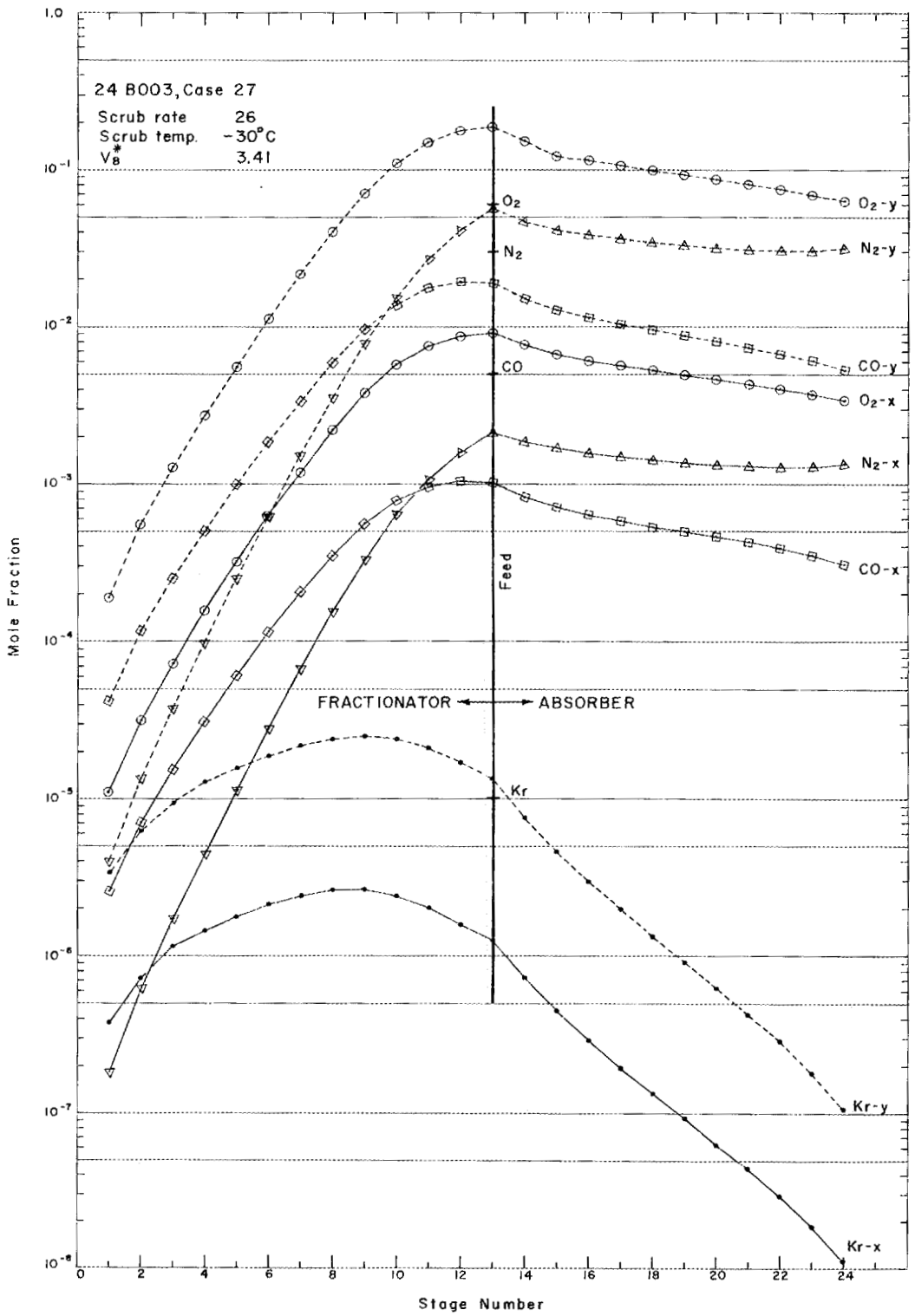

Fig. 7. Concentration Profiles from Series 24B003, Case 27.

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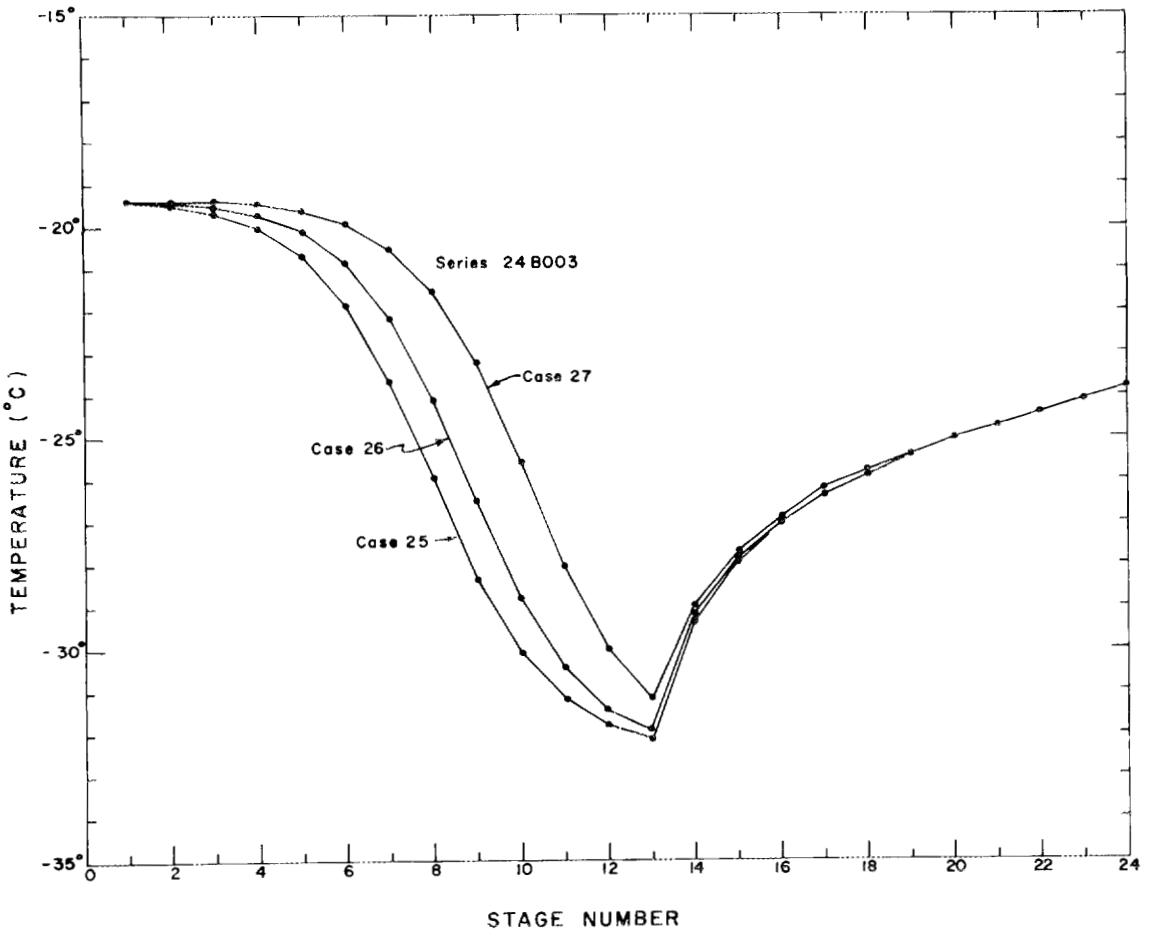

Fig. 8. Temperature Profiles from Series 24B003.

concentration profile. The temperature gradient in the absorber does not level off, implying that the addition of more stages would result in a lower temperature at the center of the cascade. This is not the case, however, since series 38B903 had about the same temperatures near the feed point and showed pronounced leveling at the top of the absorber. On the other hand, a temperature effect causes the slight shift in the location of the break in the curves from series 24B003 to 38B903.

The extraction factors for cases 25 and 27 are plotted in Figs. 9 and 10. The extraction factor is defined as a product of the distribution coefficient and the ratio of flow rates. When the extraction factor for a component on a given stage is greater than 1, that component tends to progress up the cascade; when it is less than 1, it tends to move down the cascade. Ideally, the extraction factor for the light gases would always exceed 1 and the extraction factor for Kr would always be less than 1. As shown in Figs. 9 and 10, the temperature variation in a cascade results in a variation in the extraction factors such that this ideal situation is not realized. For case 25 (with the lower boilup rate), the Kr extraction factors are less than 1 for the whole cascade except at the very bottom, but the extraction factors for O_2 and CO dip below 1 for several stages close to the feed point. When the boilup rate is increased (case 27), the extraction factor for O_2 does not drop below 1, and the extraction factor for CO_2 is less than 1 for only two stages. The extraction factor for Kr, however, is significantly increased in the lower part of the fractionator and slightly exceeds 1 for several stages.

6.2 Effect of Intercolumn Condenser in Absorber-Fractionator Operation

A condenser placed between the fractionator column and the absorber column can be considered an equilibrium stage, and introduction of the feed to this stage entails no significant loss in generality. The input to an enthalpy balance around this stage involves the liquid entering from above, the vapor coming from below, the entering feed, and the heat removed by the condenser coolant. Total input enthalpy must be accounted for in the exit vapor and liquid from the stage. Since the same effective

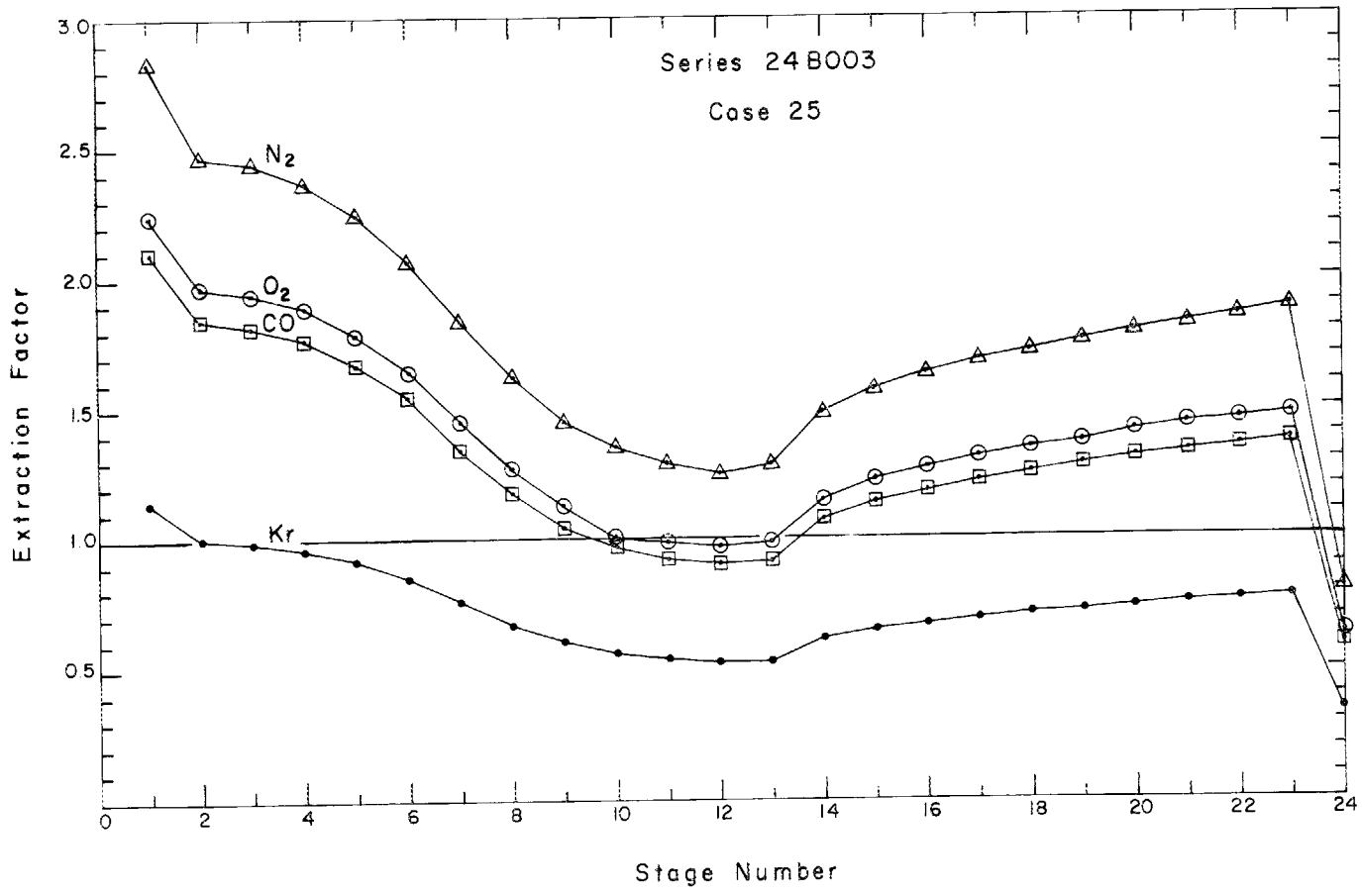

Fig. 9. Extraction Factors from Series 24B003, Case 25.

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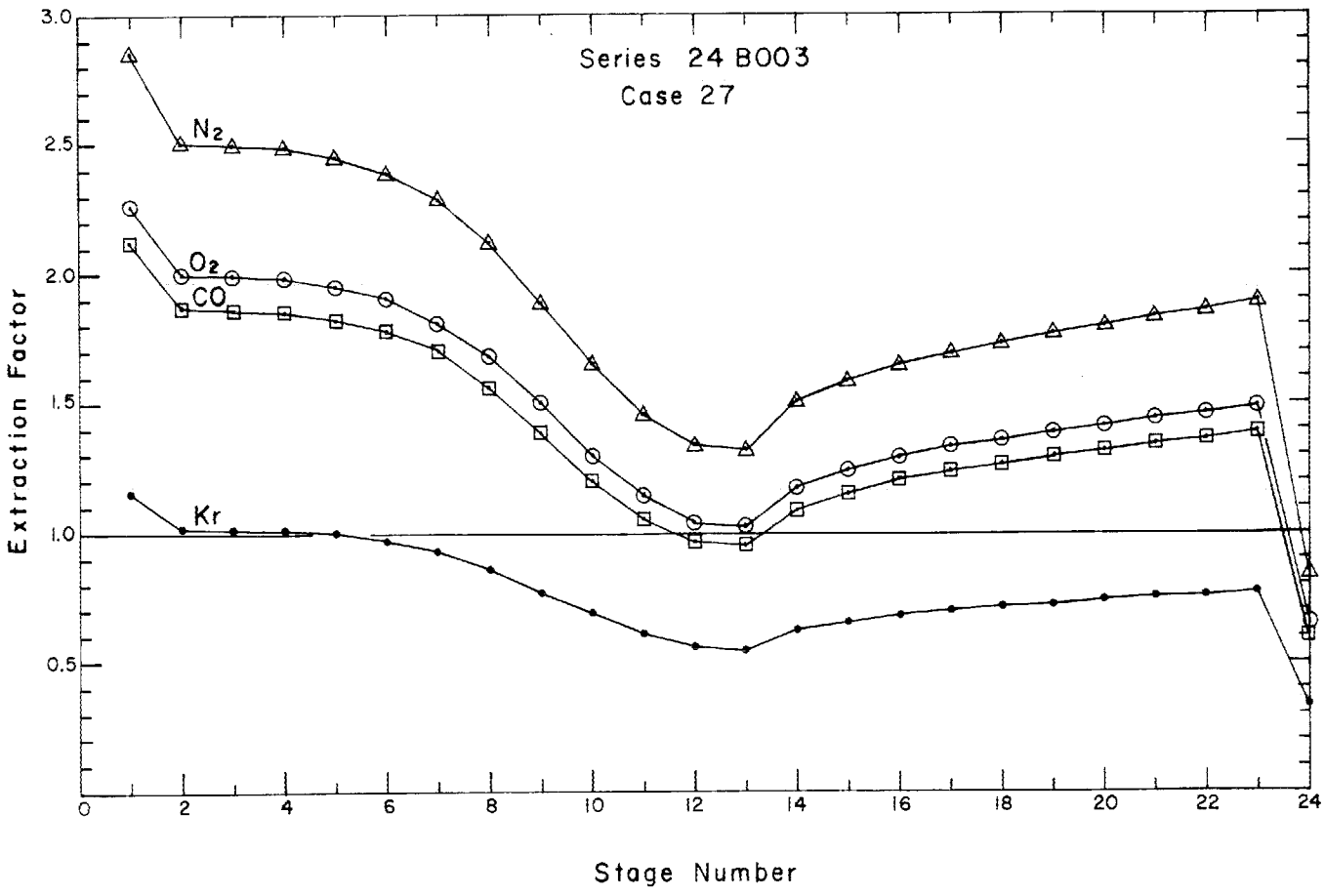

Fig. 10. Extraction Factors from Series 24B003, Case 27.

condition can be achieved by an entering feed with various enthalpies via appropriate adjustment of the heat removed by the condenser, it becomes clear that the independent specification of both of these quantities is superfluous. For this study, the heat removed by the condenser was incorporated into the enthalpy of the feed. Values of feed enthalpy above about 3600 cal/mole (depending on the system pressure and feed composition) represent superheated vapor. Values between about 600 and 3600 cal/mole represent a mixed feed (partially vaporized). Values below about 800 cal/mole can be interpreted either as a subcooled liquid or as the operation of an intercolumn condenser. Negative feed enthalpies are inherently fictitious and must represent the operation of an intercolumn condenser.

The introduction of a condenser between the fractionator and the absorber has the effect of decreasing the vapor rate in both columns; however, when a "best" operating point is obtained by increasing the boilup rate, the liquid-to-vapor (L/V) ratio in both columns is brought close to the values obtained at a "best" operating point without a condenser, and the net effect is that the L/V ratio is slightly increased in the absorber and slightly decreased in the fractionator. This results in an increased refluxing of light gases to the feed point.

Results of a parametric study varying the feed enthalpy from -2000 to +800 cal/mole has shown that there is a wide band of acceptable feed enthalpies but that an intercolumn condenser is desirable. Figure 11 illustrates this study by showing plots of DF and CF vs boilup rate for series 24A001, 24A002, 24A003, and 24A004, which are identical except for feed enthalpy. At feed enthalpies less than about -1500 cal/mole, both the DF and the CF are high at the "best" operating point, but the width of the band of acceptable boilup rates becomes intolerably narrow. In series 24A004 it is seen that the DF plummets to very low values when the boilup rate is increased only slightly beyond the "best" point. At feed enthalpies above zero the DF and CF at the "best" operating point become unattractively low.

Figure 12 shows that the feed enthalpy might have been used instead of boilup rate as the primary variable. In this study (series 24B913),

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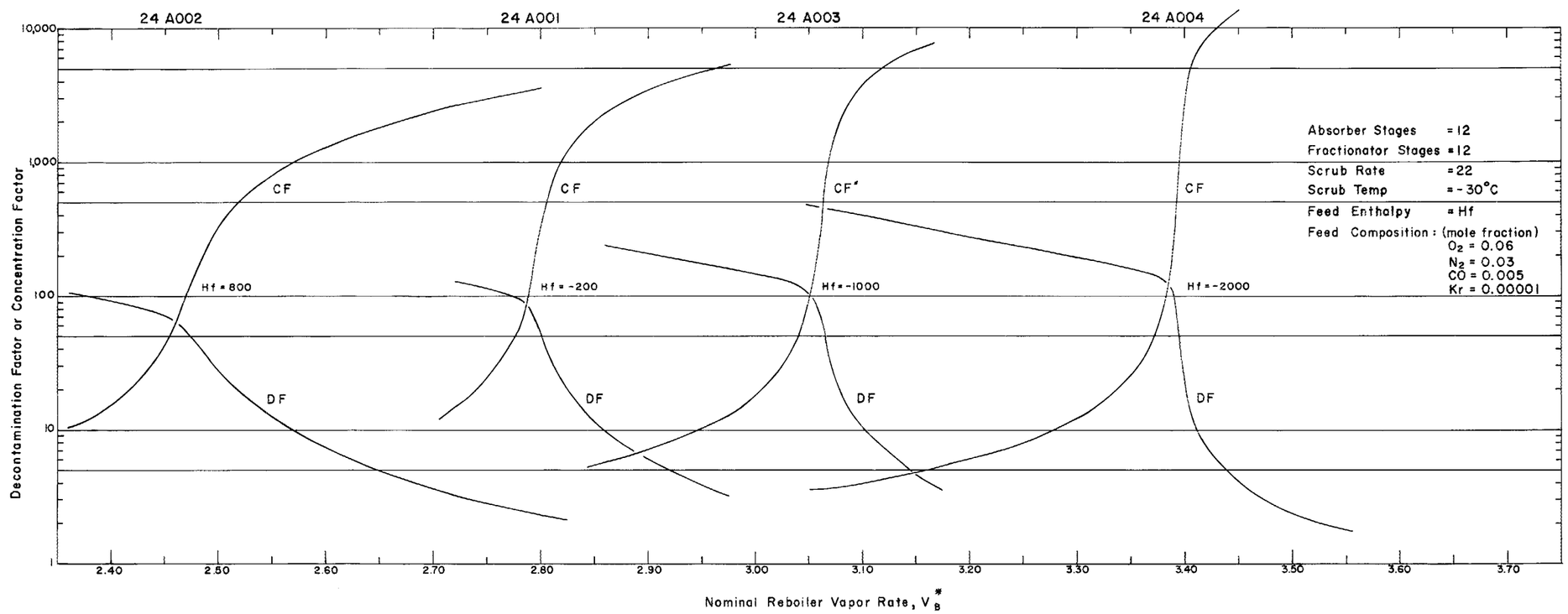

Fig. 11. Parametric Study of Intercolumn Condensation in Absorber-Fractionator Operation.

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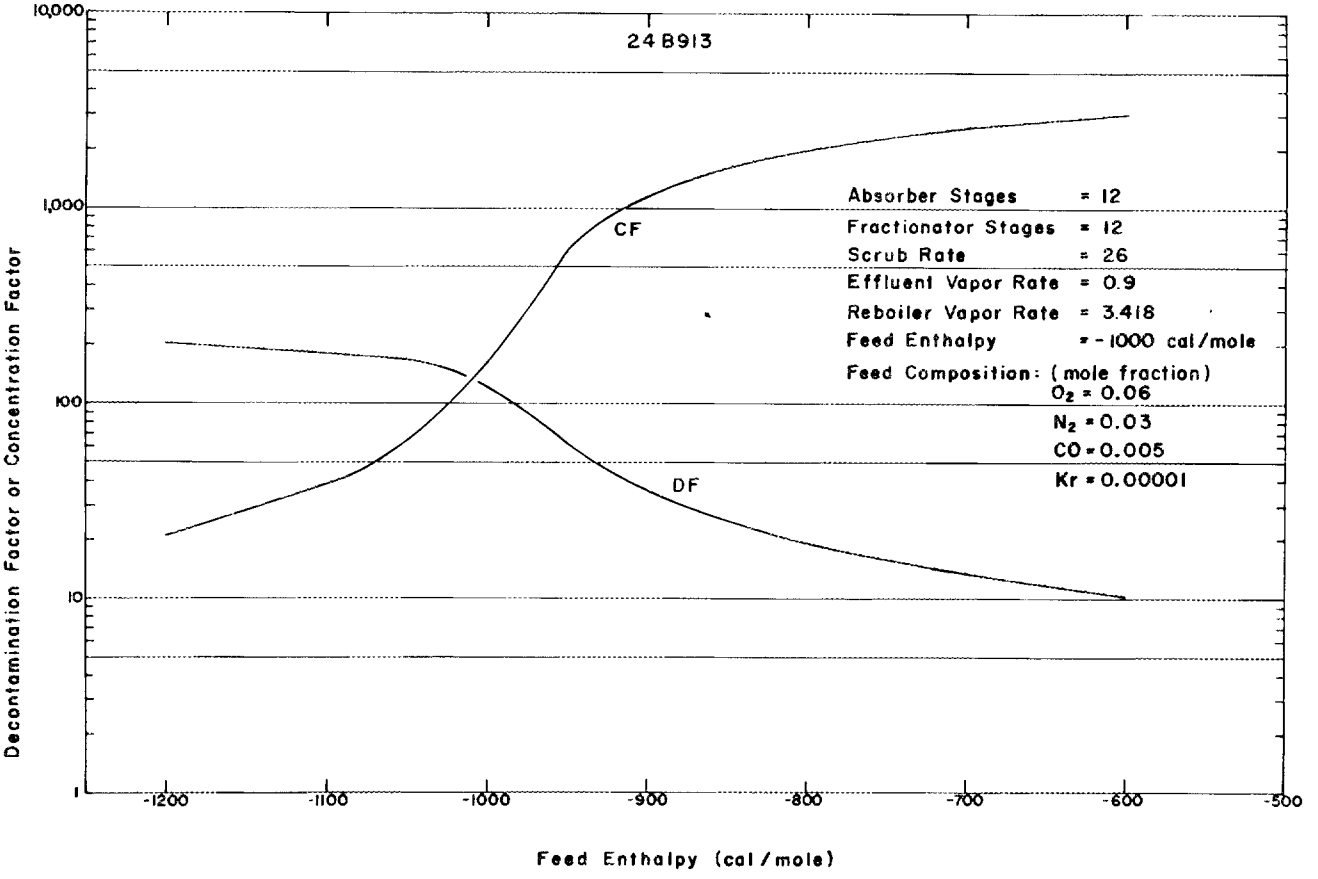

Fig. 12. Effect of Intercolumn Condensation on DF and CF (Series 24B913).

the conditions were identical to those used for series 24B903, except that the boilup rate was held constant at near the best value for that series and the feed enthalpy was varied. The form of the increase in CF, and the decrease in DF, with increasing feed enthalpy is very similar to that seen when boilup rate is used as the primary variable. Such results also imply that feed enthalpy would be a good control variable.

6.3 Effect of Scrub Rate

At a "best" operating point the boilup rate must accommodate the scrub rate in order to yield the proper flow ratios (extraction factors). Since the boilup rate increases with scrub rate, the net effect of increasing the scrub rate is to decrease all effects contributed by the feed. The most important of these are the amount of light gases in the system and the temperature effects associated with them. Because the scrub stream must be at least 16 times the feed stream, the CO_2 introduced with the feed is of little importance. The effect of the enthalpy of the feed also tends to diminish with increasing scrub rate, but not drastically.

Figure 13 shows the results of the parametric study in which the scrub rate was varied. Two sets of series are shown: the 24X003 and the 24X903 (X = A, B, C, D, E, or F). The scrub temperature was kept constant at -30.0°C for the 24X003 set, which allowed the effluent vapor rate at the "best" operating point to vary from 0.813 for 24E003 to 1.025 for 24D003. For the 24X903 set, the condition of constant effluent vapor rate was imposed, causing the scrub temperature to vary at the best operating point from -29.42°C for 24F903 to -30.57°C for 24C903. This effect is discussed in more detail later.

The variables for the series shown are all identical (subject to the comments above) with the exception of scrub rate. One concludes that the system performs more effectively at higher scrub rates. Decontamination and concentration factors at the "best" operating point decrease at the lower scrub rates, and the range of acceptable scrub rates also becomes smaller.

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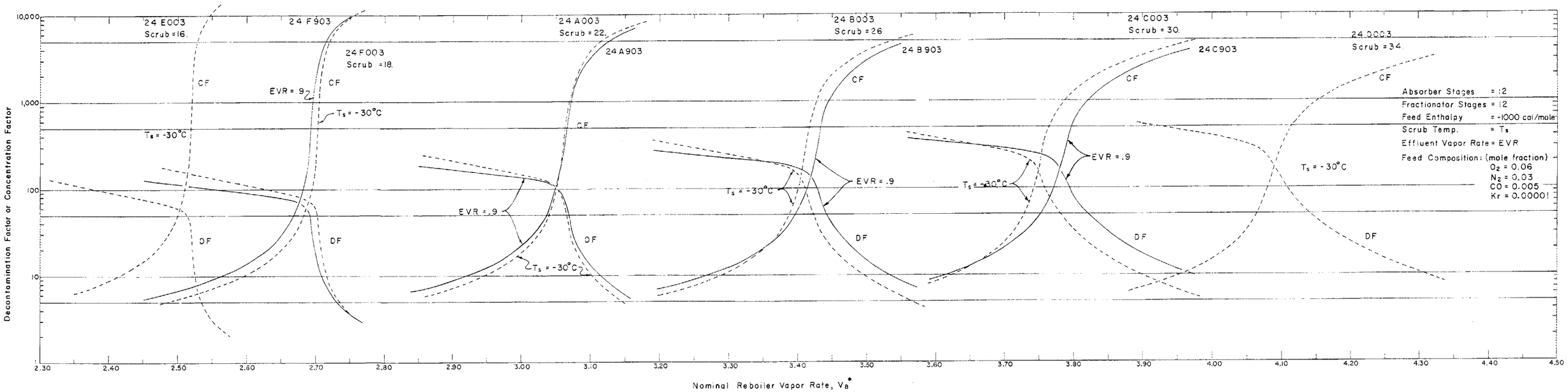

Fig. 13. Parametric Study of Scrub Rate in Absorber-Fractionator Operation.

These effects are not surprising since nonidealities are reduced by dilution at the higher scrub rates and are aggravated at lower scrub rates. The size of the system, however, is roughly proportional to the scrub rate. Economic considerations, which are beyond the scope of this work, will set an optimum scrub rate that will probably be near the middle of the range covered in Fig. 13.

6.4 Effect of Scrub Temperature

The ratio of liquid-to-vapor molar rates in the absorber ranges from about 12 to 20. The latent heat of CO_2 is about 3000 cal/mole, and the specific heat is of the order of 20 cal/mole·°C. These values imply that 12°C subcooling of the scrub stream could condense the entire vapor stream were it pure CO_2. The capacity of the scrub stream to condense and reflux vapor is exploited in the KALC system calculation to control the amount of CO_2 in the effluent gas. Characteristically, the rate at which vapor enters the top stage is 2 to 3 times the feed rate, whereas the effluent vapor leaves the top stage at about 0.9 times the feed rate.

The computer program was written to accept either a scrub temperature or a nominal effluent vapor rate. In Table 3, which summarizes all of the run conditions for the study, scrub temperatures of -30°C were specified for all runs with a series containing 0 as the fourth digit. For these the effluent vapor rate varied from run to run, yielding the tabulated value at the "best" operating point. The series having 9 as the fourth digit are those for which a nominal effluent vapor rate of 0.9 was specified. In these series, the scrub temperature was deduced and varied from run to run, yielding the tabulated value at the "best" operating point. Figure 13 shows a comparison of the series made using each mode. One must conclude that neither the scrub temperature nor the amount of effluent gas has a strong effect on the DF or CF; and, indeed, the principal importance of the scrub temperature is that it does determine the amount of CO_2 in the effluent vapor.

At a given scrub temperature in a given series of runs, the effluent vapor rate changes with the boilup rate. In a first approximation, it would be expected that the plot of effluent gas rate vs boilup rate would

have a slope of unity. Such a plot is shown in Fig. 14 for series 24B003. An increase in the boilup rate by one unit does not result in an increase in effluent gas rate by one unit because the process increases the CO_2 content of the effluent gas, which increases the equilibrium temperature on the top stage, consuming part of the available enthalpy.

Also shown in Fig. 14 is a plot of scrub temperature and effluent gas rate vs nominal boilup rate for series 24B903 (the counterpart of 24B003). Perhaps the most significant inference to be drawn is that a change of less than 2.0°C in scrub temperature is required to effect a 10% change in the effluent gas rate. It is doubtful that sufficient precision can be obtained in scrub temperature to permit the operation of a real system in the mode used for these calculations.

The deviation of the actual effluent vapor rate from the nominal effluent vapor rate in series 24B903 and the deflection in the effluent vapor rate curve in 24B003 are attributed to the fact that the temperature of the liquid leaving the bottom of the fractionator is colder than the pure CO_2 saturation temperature for boilup rates lower than the value at the "best" operating point. This is treated further in Appendix B.

6.5 Effect of Feed Concentration

Determining the effect of feed concentration on the absorber-fractionator system was not made a major part of this study because it was assumed that changing the feed concentration would be similar to changing the scrub rate. Instead, a standard feed composition was used throughout the study in order to establish the effects of other parameters. Two series of runs were made, however, in which the feed concentration was varied to test this assumption and to look for unanticipated relationships. Series 24B9F3 was identical to 24B903 except the concentrations of light gases were reduced to half the standard values. The results were expected to be similar to those obtained for a run series with a scrub rate of 52. Although such a series is not available for comparison, the expected high values of DF and CF at the "best" operating point and the smooth, insensitive relationship of DF and CF to boilup rate were attained.

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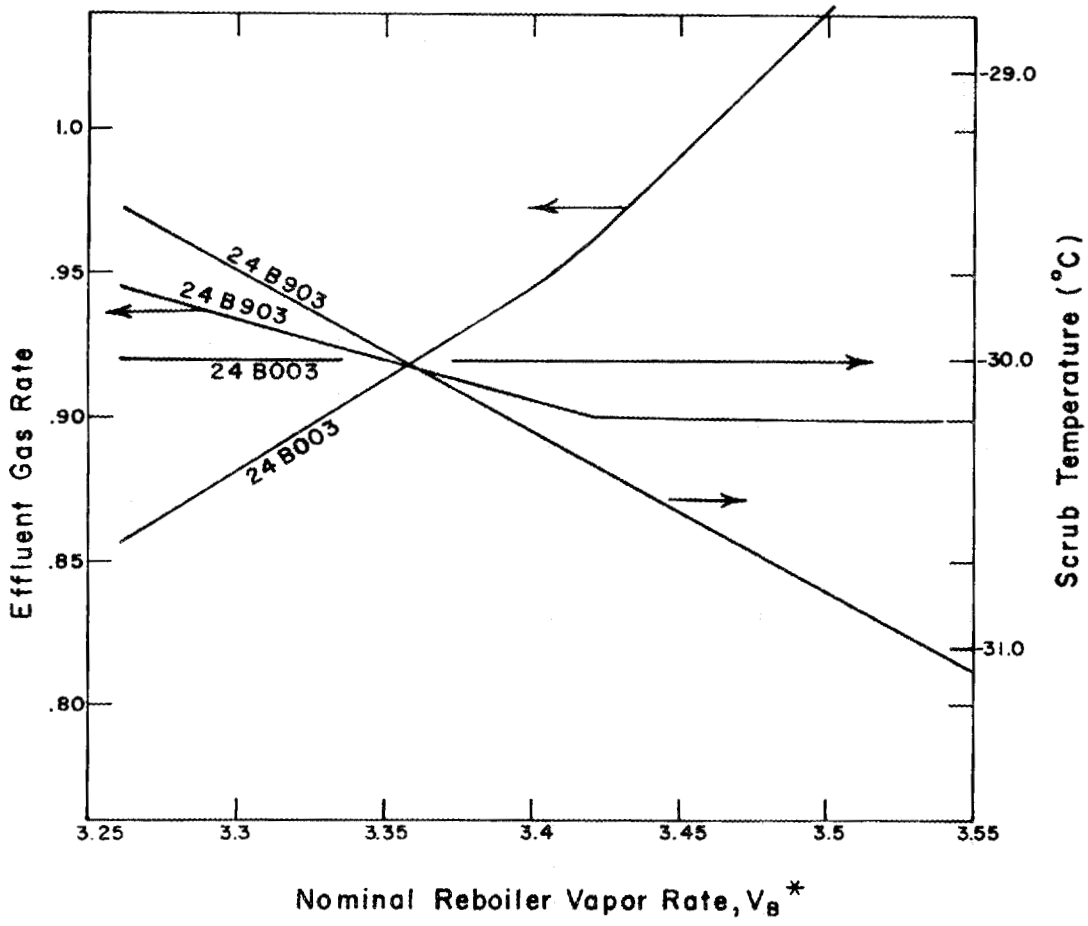

Fig. 14. Variation of Effluent Gas Rate and Scrub Temperature with Nominal Reboiler Vapor Rate.

Series 24B9G3 was identical to series 24B903 except that the concentration of each light gas was increased to 1.4444 times its standard concentration. Consequently, the results for series 24B9G3 should be similar to those for series 24F903 (if the assumption stated above is valid) since the scrub rate for 24B903 was 1.4444 times the scrub rate for 24F903. Such a similarity was established by the plots of DF and CF vs boilup rate, which, by visual comparison, are much alike. Table 5 compares series 24B9G3 with series 24F903 and 24B903, emphasizing the rate of change of DF and CF with boilup rate. Some differences between 24B9G3 and 24F903 are noted. These can be attributed to three effects:

(1) There is less CO_2 per unit of light gas entering with the feed in series 24B9G3.

(2) The effect of feed enthalpy is relatively less in series 24B9G3.

(3) The light gases are 44% more concentrated in the effluent gas of series 24B9G3, causing a lower temperature on the top stage.

Even with these effects, it is clear that series 24B9G3 more nearly resembles 24F903 than 24B903.

Although the noted effects are significant, they are not large; therefore, it is the opinion of the author that they have been sufficiently quantified for the purposes of this study.

6.6 Effect of Krypton in the Scrub Stream

It was presumed earlier that a small amount of Kr in the scrub stream to the absorber would produce a small, predictable, and easily described effect. One series of runs appears to justify such a presumption. Series 24B923 is identical to series 24B903 except that a boilup rate close to the "best" was chosen and fixed while the scrub composition was varied.

At this point, it should be established what range of concentrations of Kr in the scrub stream is considered to be reasonable. If the feed contains 10^{-5} mole fraction of Kr, then the solution entering the stripper will contain $\sim 10^{-6}$ mole fraction. The stripper will remove at least 99% of this (it will probably be designed to remove 99.99% or more). It would

Table 5. Comparison of Series 24B903, 24B9G3, and 24F903 to Establish the Effect of Feed Concentration

	Series[a]		
	24B903	24B9G3	24F903
Feed concentration	Std.	1.444 x std.	Std.
Scrub rate	26	26	18
"Best" boilup rate	3.418	3.731	2.676
Boilup/scrub rate ratio	0.1315	0.1432	0.1482
"Best" CF = DF	135	86	70
Percent increase in boilup rate required to:			
Change CF from 100 to 1000	0.906	0.563	0.563
Change CF from 100 to 10,000	>10	3.34	2.88
Change DF from 50 to 10	2.61	0.75	0.674
Change DF from 10 to 3	>5	2.69	2.21

[a]See Table 3 for run conditions.

be expected, then, that the concentration of Kr in the scrub stream would not normally exceed 10^{-8} mole fraction.

Some relevant results from series 24B923 are tabulated in Table 6. The Kr content of the scrub stream had three values: 10^{-9}, 10^{-8}, and 10^{-7} mole fraction. The largest value amounts to an input of Kr to the system by the scrub stream equal to 26% of the input by the feed. The main conclusion to be drawn is that a fixed fraction of the Kr entering with the scrub stream will report to the effluent gas. Calculations made by comparing each case with the case containing no Kr in the scrub gave 55% in this series. In simple theory, with a constant distribution coefficient and constant molal overflow, the fraction lost to the effluent gas would be equal to the extraction factor for the system. However, our system is not this simple. The extraction factor for Kr on the top stage was 0.3198; values for successive stages down the absorber were 0.7702, 0.7606, 0.7502, etc., decreasing to 0.5473 at the feed stage. The observed fraction 0.55 is not inconsistent.

The reported DFs decrease, while the reported CFs slightly increase, with increasing amounts of Kr in the scrub stream.

6.7 Effect of System Pressure

The choice of 20 atm as the standard pressure to be used for the bulk of the study was based on qualitative consideration of the requirements of a real KALC plant, balancing refrigeration requirements against structural requirements, with superficial recognition of equilibrium effects. It is gratifying that calculations support and justify this choice.

Series 24P9AA and 24P9AB are identical to series 24B903, except that they were made at system pressures of 15 and 25 atm, respectively. Plots of DF and CF against nominal reboiler vapor rate (boilup rate) are shown for the three series in Fig. 15. It is seen, first, that the boilup rates required for "best" operation are changed drastically by operating pressure. Increasing the pressure from 20 to 25 atm required an increase from 3.42 to 4.25 in the boilup rate, while decreasing the pressure to 15 atm allowed a reduction to 2.71. Further, it is seen that operation at higher pressure

Table 6. Summary of Calculations of the Effect of Kr in the Scrub Liquid

Series 24B923[a]
Kr entering with feed = 100 x 10^{-7} mole
Scrub rate = 26 (feed rate = 1)
Effluent gas rate = 0.902
Extraction factor on top stage = 0.3198

	Case			
	1 (Reference)	3	2	4
Kr entering in scrub:				
Conc., mole fraction (x 10^7)	0	0.01	0.1	1.0
Amount, moles (x 10^7)	0	0.26	2.6	26
Kr leaving in effluent gas:				
Conc., mole fraction (x 10^7)	0.878	1.04	2.47	16.8
Amount, moles (x 10^7)	0.792	0.938	2.228	15.15
Kr leaving in effluent gas in excess of reference case:				
Amount, moles (x 10^7)	0	0.146	1.436	14.36
$\frac{\text{Excess amount leaving}}{\text{Amount entering in scrub}}$		0.56	0.552	0.552
Decontamination Factor	126.2	106.9	44.85	6.6
Concentration Factor	168.7	169.2	171.0	188.7

[a]See Table 3.

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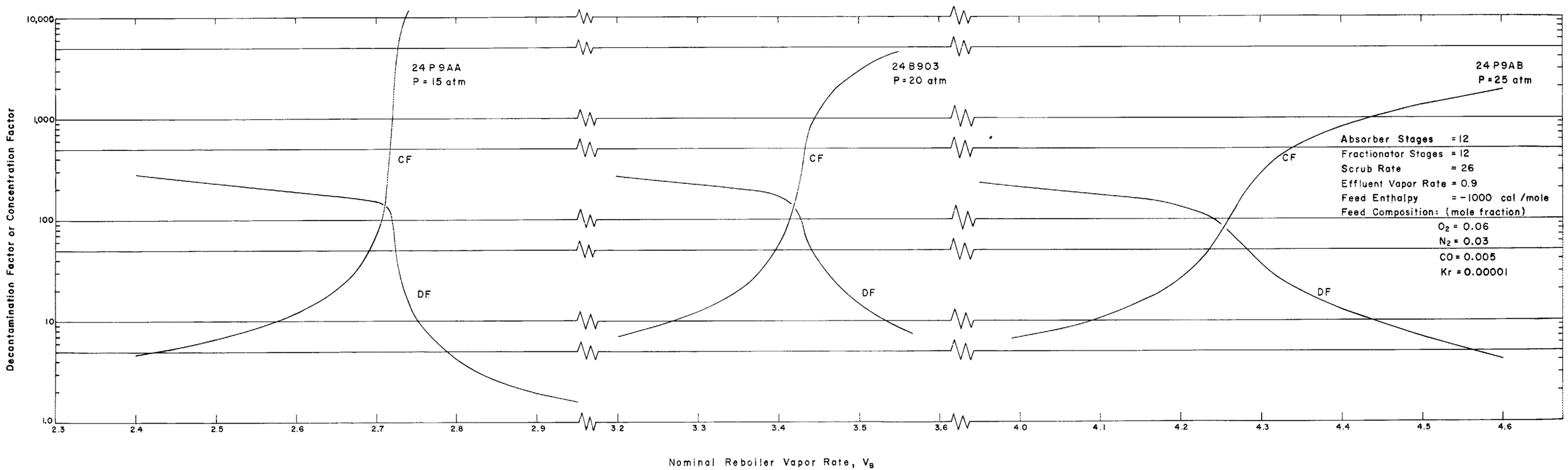

Fig. 15. Parametric Study of System Pressure in Absorber-Fractionator Operation.

results in lower CF and DF values around the "best" operating point. Operation at the lower pressure increases the "best" values of CF and DF but gives rise to a critical operating condition where a slight increase in boilup rate causes a drastic reduction in DF. Operation at the lower pressure also entails a 5 to 10°C decrease in temperature, which is probably significant.

It appears that little can be gained by using pressures either much higher or much lower than 20 atm.

6.8 Effect of Number of Stages on the Performance of the Absorber-Fractionator System

A series of absorber-fractionator runs was made to establish that the system performance improves with the addition of more stages. In series 38B903, the absorber had 22 stages and the fractionator had 16. The plot of DF and CF against nominal boilup rate for this series (see Fig. 4) compares it to series 24B903, which, except for the number of stages, was made using identical conditions. Adding 14 more stages to the 24-stage system effected about a factor of 10 improvement in both dependent variables and established a region of operation which meets the requirements of KALC as presently understood (a DF of several hundred and a CF of at least 1000.) This and other calculational runs with large numbers of stages indicate that any reasonable specifications could be met by increasing the number of stages sufficiently.

7. STUDY OF THE STRIPPER-RECTIFIER SYSTEM

Two simple conclusions quickly derivable from this study are that a stripper-rectifier system for KALC can be easily designed to perform to any desired specifications (within liberal bounds), and that the system can be expected to meet such specifications over a comfortable range of operating conditions. These conclusions are almost satisfactorily established by the examination of a single typical case. A few variables, such as the boilup rate, are seen to have a controlling effect, while some (e.g., the feed composition or effluent gas rate) have surprisingly little effect.

The stripper-rectifier system used as a point of departure for the parameter study had 12 stages in the stripping section and 4 stages in the rectifying section. As in the absorber-fractionator study, the number of stages used here is less than would be desirable in a plant. The reasons for the choice are the same as previously discussed: improved accuracy, higher concentrations, and less computer time. The feed was introduced into the lowest stage of the rectifying section, and the top stage of the rectifying section was the condenser.

The computer printout from series S16B122, case 3, is shown in Table 7. The temperature (in °C) and flow rates (L and V, referred to unit feed) are seen to be uniform in the stripping section, and the vapor rate equals V_B^*. The flow rates change abruptly at the feed point, but vary slightly in the rectifying section, as the concentration of light gases increases. The temperature in the condenser is the dew point of the effluent gas whose composition was simply fixed by the input variables. The quantities H LIQ and H VAP are the specific enthalpies of the liquid and gas phases, respectively, expressed in calories per gram·mole, referred to liquid CO_2 at -56.6°C as zero.

The concentrations (in mole fractions) of each component in the gas (y) and liquid (x) phases is given for each stage in the system in the right-hand portion of the table. The Kr concentration gradient is steep throughout the entire cascade, decreasing from several mole percent at the top to 10^{-8} mole fraction at the bottom, where the O_2, N_2, and CO are found in amounts about four orders of magnitude below that of the Kr.

7.1 Effect of Boilup Rate on the Stripper-Rectifier System

For each of the series in this study, the value of the boilup rate was varied from 0.2 or 0.25 up to 0.5 in increments of 0.05. This interval was usually ample to change the Kr loss by three orders of magnitude. In a few of the series the Kr loss exceeded 1% for the lower boilup rates; in some of the series with low feed enthalpy, the lower boilup rates were not usable because the incoming cold feed would condense all of the vapor. For a stripper-rectifier system having a fixed number of stages and

Table 7. Computer Printout from Series S16B122, Case 3.

```
SERIES  S16B122, CASE   3                                   01-18-73
ITERATIONS =  145
STAGES IN RECTIFIER =  4
STAGES IN STRIPPER = 12
FEED COMP; O2,N2,CO,KR,XE
   0.1000D-05   0.4000D-06   0.1200D-05   0.4000D-06   0.0
FEED ENTHALPY =   100.000
SCRUB/FEED =   0.0
REFLUX RATIO =  7179.620
EFFLUENT GAS = 0.000009999
COMPOSITION =
   0.1003D 00   0.4014D-01   0.1203D 00   0.3992D-01   0.0
        CO2 = 0.6993931
FRACTION KR LOST = 0.000946828
BOILER VAP RATE =   0.300
SYS PRESSURE =  20.000
```

STAGE	T	L	V	L/V	H LIQ	H VAP
16	-32.99	0.072	0.000	*******	468.59	2643.01
15	-20.04	0.077	0.072	1.074	744.61	3674.99
14	-19.36	0.077	0.077	1.004	760.00	3728.39
13	-19.32	1.300	0.077	16.783	761.06	3732.04
12	-19.32	1.300	0.300	4.334	761.14	3732.34
11	-19.31	1.300	0.300	4.333	761.17	3732.44
10	-19.31	1.300	0.300	4.333	761.18	3732.47
9	-19.31	1.300	0.300	4.333	761.18	3732.48
8	-19.31	1.300	0.300	4.333	761.18	3732.49
7	-19.31	1.300	0.300	4.333	761.18	3732.49
6	-19.31	1.300	0.300	4.333	761.18	3732.49
5	-19.31	1.300	0.300	4.333	761.18	3732.49
4	-19.31	1.300	0.300	4.333	761.18	3732.49
3	-19.31	1.300	0.300	4.333	761.18	3732.49
2	-19.31	1.300	0.300	4.333	761.18	3732.49
1	-19.31	1.000	0.300	3.333	761.18	3732.49

STAGE		O2	N2	CO	KR	XE
16	Y	0.100D 00	0.401D-01	0.120D 00	0.399D-01	0.0
	X	0.489D-02	0.151D-02	0.632D-02	0.349D-02	0.0
15	Y	0.490D-02	0.151D-02	0.633D-02	0.350D-02	0.0
	X	0.277D-03	0.680D-04	0.383D-03	0.384D-03	0.0
14	Y	0.290D-03	0.732D-04	0.398D-03	0.389D-03	0.0
	X	0.165D-04	0.331D-05	0.242D-04	0.433D-04	0.0
13	Y	0.294D-04	0.847D-05	0.397D-04	0.484D-04	0.0
	X	0.167D-05	0.382D-06	0.240D-05	0.538D-05	0.0
12	Y	0.722D-05	0.166D-05	0.104D-04	0.233D-04	0.0
	X	0.407D-06	0.743D-07	0.627D-06	0.257D-05	0.0
11	Y	0.176D-05	0.322D-06	0.272D-05	0.111D-04	0.0
	X	0.987D-07	0.143D-07	0.162D-06	0.122D-05	0.0
10	Y	0.428D-06	0.621D-07	0.704D-06	0.528D-05	0.0
	X	0.238D-07	0.275D-08	0.419D-07	0.576D-06	0.0
9	Y	0.103D-06	0.119D-07	0.182D-06	0.249D-05	0.0
	X	0.573D-08	0.527D-09	0.108D-07	0.271D-06	0.0
8	Y	0.248D-07	0.228D-08	0.467D-07	0.117D-05	0.0
	X	0.138D-08	0.101D-09	0.276D-08	0.127D-06	0.0
7	Y	0.596D-08	0.436D-09	0.120D-07	0.550D-06	0.0
	X	0.330D-09	0.192D-10	0.708D-09	0.596D-07	0.0
6	Y	0.143D-08	0.833D-10	0.307D-08	0.257D-06	0.0
	X	0.791D-10	0.366D-11	0.181D-09	0.278D-07	0.0
5	Y	0.343D-09	0.159D-10	0.785D-09	0.119D-06	0.0
	X	0.189D-10	0.699D-12	0.464D-10	0.129D-07	0.0
4	Y	0.819D-10	0.303D-11	0.200D-09	0.547D-07	0.0
	X	0.453D-11	0.133D-12	0.118D-10	0.592D-08	0.0
3	Y	0.195D-10	0.574D-12	0.508D-10	0.244D-07	0.0
	X	0.108D-11	0.253D-13	0.300D-11	0.264D-08	0.0
2	Y	0.450D-11	0.107D-12	0.125D-10	0.102D-07	0.0
	X	0.249D-12	0.470D-14	0.736D-12	0.110D-08	0.0
1	Y	0.909D-12	0.178D-13	0.267D-11	0.350D-08	0.0
	X	0.503D-13	0.781D-15	0.157D-12	0.379D-09	0.0
MATL BAL		1.0025	1.0034	1.0022	0.9988	1.0000

operating at a specified pressure, there is no other operating variable which produces an effect remotely comparable to that of the boilup rate. Fortunately, this effect yields to a relatively simple correlation.

First, it is accepted that the ratio of vapor (V) to liquid (L) flow in the stripping section is a better correlating variable than the boilup rate per se. By noting that a unit feed rate was used in the study, and that the liquid removal rate essentially equaled the feed rate (effluent gas <0.002% of feed), we have

$$\frac{V}{L} = \frac{V_B^*}{1 + V_B^*} \quad . \tag{12}$$

Figure 16 is a logarithmic plot of the krypton loss vs this flow ratio. Series S16B141 and S16B123 are typical, and all series designated by S16B_ _ _ are more or less bounded by the curves shown in the figure. The plotted points gave essentially straight lines, which led to a correlation of the form:

$$\ln\,(\text{Kr loss}) = A + B \cdot \ln\left(\frac{V_B^*}{1 + V_B^*}\right) \quad . \tag{13}$$

The constants A and B were determined by a least-squares fit. This correlation, in addition to describing the effect of boilup rate, provides us with two numbers (A and B) that are independent of boilup rate and can be used to evaluate the effects of other variables. The values of A and B for all the series comprising the study are tabulated in Table 4. Also shown is the mean error in the fit, which can be interpreted as a measure of the curvature of the data. The tabulated value of the mean error for series 3S16P122 is 40.5%, which identifies this series as unique in the study. Characterized by a high Kr loss and generally unacceptable system performance, it was included in Fig. 16 only to point out a limitation on the correlation.

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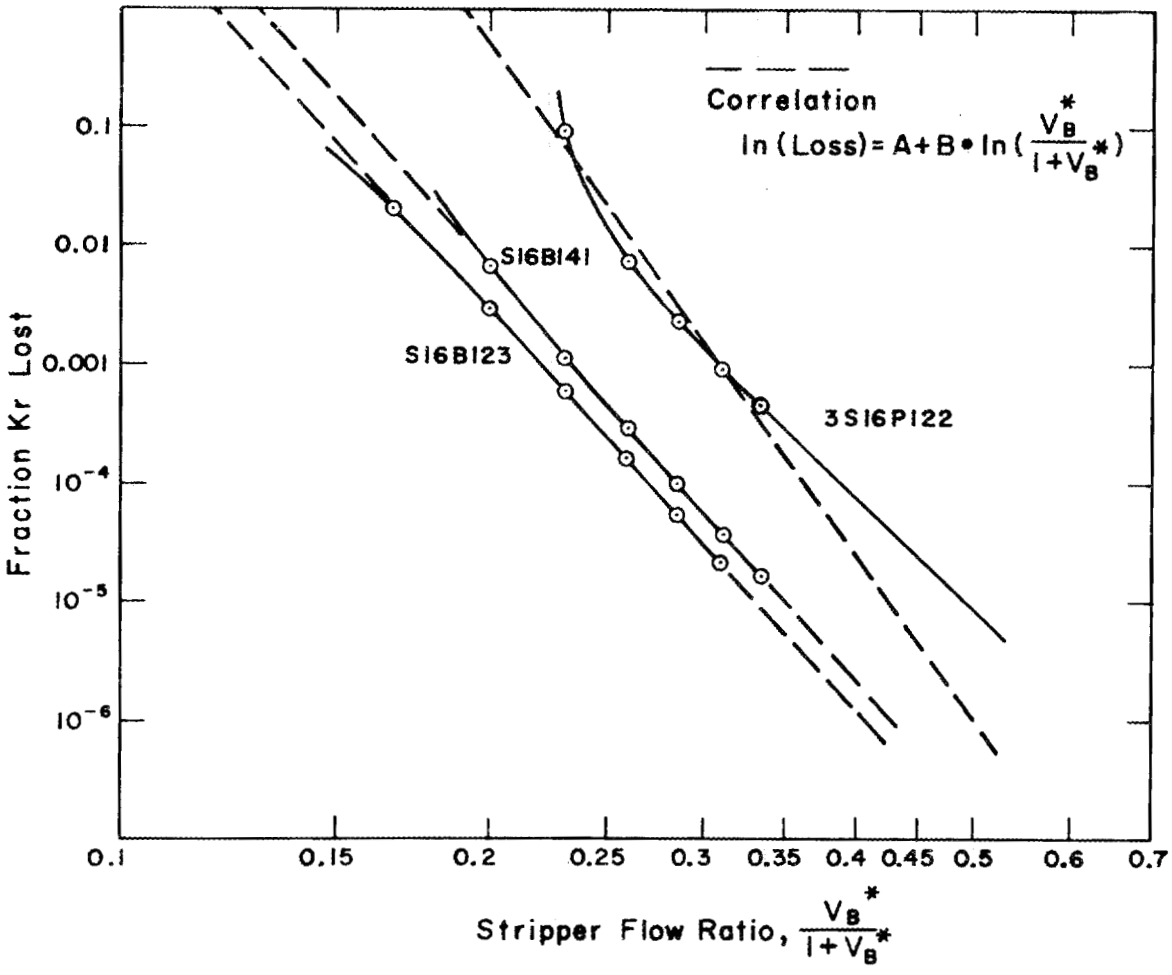

Fig. 16. Correlation of Krypton Losses from the Stripper-Rectifier System with Vapor-to-Liquid Flow Ratio in the Stripper.

It is interesting to attempt to attach some physical significance to A and B. The best approach is through the Fenske equation,[16]

$$\frac{x_B}{x_F} = \frac{E - 1}{E^{n+1} - 1} , \tag{14}$$

which describes a very simple distillation tower, of n stages, that is fed at the top with a stream containing x_F and discharges a stream containing x_B mole fraction of solute from the bottom. Both the equilibrium distribution coefficient, $K = y/x$, and the V/L flow ratio must be constant in order to give a constant extraction factor, $E = K\ V/L$. In some important respects this expression is a crude model of our system.

Further simplification and approximation are called for. By noting $E^{n+1} >> 1$, Eq. (14) can be written as:

$$\frac{x_B}{x_F} = E^{-n}(1 - \frac{1}{E}) \tag{15a}$$

or

$$\ln \frac{x_B}{x_F} = \ln (1 - \frac{1}{E}) - n \cdot \ln E . \tag{15b}$$

The slope of a plot of $\ln x_B/x_F$ vs ln V/L is obtained by taking the derivative with respect to ln E, which is the same (if K = constant) as taking the derivative with respect to ln V/L. This yields:

$$\text{slope} = -n + \frac{1}{E - 1} . \tag{16}$$

The smallest values of E for Kr in this study were about 3, introducing a varying error, not usually exceeding 0.5, in the slope. Since this modified Fenske equation has a form similar to our empirical correlation, we might expect B to be some effective number of stages. In spite of the dubious justification for such an expectation, we find that the tabulated values of B are close in magnitude to the number of stages in the stripping section.

From the form of the correlation we see that exp (A) represents a limiting Kr loss when the boilup is increased to large values. It is significant that, even under the worst circumstances, this number has a ridiculously small value.

7.2 Effect of Intercolumn Condenser in the Stripper-Rectifier System

As in the absorber-fractionator system, the effect of an intercolumn condenser was studied by varying the enthalpy of the feed to the system. Unlike the absorber-fractionator system, all of the material above the feed point will have passed the intercolumn condenser; there is no scrub stream. The liquid flow in the rectifier comes from condensed vapor and, since only a very small fraction of the vapor stream leaves the top condenser, the liquid and vapor streams are nearly equal. When the condenser at the feed point condenses most of the vapor from the stripping section, represented in the study by a low feed enthalpy, the flow rates in the rectifier are small. When no condensation occurs at the feed point, or when the feed is introduced partially vaporized, the flows in the rectifier are large.

Three feed enthalpies were used: 100, 800, and 1200 cal/mole. The highest value, when the system pressure is 20 atm, represents a condition under which about 15% of the feed is vaporized. The 800-cal/mole value is close to the enthalpy of saturated liquid CO_2, and considerable condensation occurs at 100 cal/mole. Some of the intended runs at low boilup rates were not possible with the lowest feed enthalpy because all of the vapor was condensed at the feed stage.

The results of the study of the intercolumn condenser are clearly shown in Table 4. Comparison of runs in the S16B--- group, which differ only with regard to enthalpy of feed, show that the effect of this parameter on Kr loss is negligible. Upon reflection, such results are not surprising. The rectifier section operates at nearly total reflux, regardless of the amount of condensation at the feed point (within reasonable operating bounds), and the flow rate ratio in the stripping section is set by the feed and boilup rates, independent of where condensation occurs. Only in one stage, the feed stage itself, is the flow ratio changed by the feed enthalpy. The concentrations in the effluent gas stream are fixed by the flow rate of this stream, and the liquid leaving the condenser must be in equilibrium with it. The concentration profile, then, should be essentially unaltered by condensation at the feed point.

It is significant that the flow rates in (and hence the size of) the rectifying section are determined by the intercolumn condenser. It is also significant that the feed point condenser operates at a temperature higher than that of the rectifier condenser, requiring less severe refrigeration per unit of heat removed. Apparently, then, the bulk of the condensation should be provided at the feed point, limited in application by process control or by the extreme in which the effluent gas becomes a significant fraction (perhaps >1%) of the rectifier vapor flow.

7.3 Effect of Effluent Gas Rate on the Stripper-Rectifier System

The effluent gas from the stripper-rectifier system in the KALC process is the "product" stream containing all of the Kr that has been scrubbed from the decontaminated plant off-gas stream. Ideally, this stream should be as small, and therefore as concentrated, as possible. In addition to Kr, it contains all of the light gases that entered the stripper-rectifier system, and, necessarily, it contains CO_2. Under process conditions that might be expected in HTGR fuel processing, an effluent gas from KALC containing 20% light gases, half of which were Kr, would represent a reduction in the volume of the gas containing Kr by a factor of 10^4. The Kr in a 50-scfm off-gas stream would terminate in a 140-cc/min product stream. Since the Kr is actually diluted in the absorber-fractionator system, the required concentration factor over the stripper-rectifier system must be of the order of 10^5.

For this study three stripper-rectifier CFs were used: $(2/3) \times 10^5$, 1×10^5, and $(4/3) \times 10^5$. These corresponded to effluent gas rates (based on a unit feed rate) of 1.5×10^{-5}, 1×10^{-5}, and 0.75×10^{-5}, and light-gas concentrations of 20, 30, and 40%, respectively, when standard stripper feed type I was used.

As the effluent gas flow rate was reduced and the concentration of light gases in the condenser was increased, it might have been expected that the Kr losses would be proportional to the Kr concentration in the effluent gas. Indeed, the results very nearly confirm this expectation. For example, Table 4 shows that, for each set of series where the effluent

gas concentration was varied, a doubling of light-gas concentration corresponded to an increase in Kr loss by a factor between 1.6 and 2.0; the series using the more dilute feeds approached the limit 2.0 more closely.

In considerations of accuracy, it should be noted that no multicomponent equilibrium data were available at the high Kr concentrations found in the rectifier condenser, and that the required extrapolation by the computer model here may entail more error than calculations in concentration regions for which the correlations were developed. Because of this, the study was not extended to even higher effluent gas concentrations.

7.4 Effect of Feed Concentration on the Stripper-Rectifier System

The feed to the stripper comes from the absorber-fractionator system, and its Kr and light-gas concentrations will depend on that system and its load. However, the Kr will certainly be diluted by a factor from 10 to 50 from its concentration in the original off-gas. The concentrations of the other light gases might be more or less than the concentration of Kr. The three standard feeds that were used are described in Table 2. The concentration of feed II was one-tenth that of feed I; feed III contained much less O_2, N_2, and CO than Kr.

Because all of the system except the top few stages operates at very dilute concentrations, one would not expect feed concentration to have a large effect. The data in Table 4, which summarize all the series of runs with the stripper-rectifier system, indicate a small, but probably significant, effect of feed concentration. The losses appear to be slightly higher with the more dilute feed, possibly because the solubility of Kr is slightly decreased in the rectifier condenser when the light gases are at higher concentrations.

7.5 Effect of System Pressure on the Stripper-Rectifier System

The stripper-rectifier system was found to be very sensitive to operating pressure. Series S16B122 was replicated with pressures varying from 14 to 25 atm (making the S16P122 series). The amount of Kr lost to the stripper bottoms (recycled with the absorber scrub stream) changed by

four orders of magnitude over this pressure range. The results are shown in Fig. 17, where the fraction of Kr lost is plotted against the system pressure for $V_B^* = 0.3$ and $V_B^* = 0.5$. The two curves, which are approximately parallel, clearly show that the system performs dramatically better at the lower pressures. This is contrasted by the conclusion that pressure is a variable of secondary importance in the absorber-fractionator system. The only penalty for operating at a lower pressure is that the system temperature will be lower. This temperature must not approach the freezing point, -56°C, too closely; however, at 13 atm this temperature is -44°C, which allows a comfortable margin. The additional refrigeration load required by the 12°C lower temperature is probably tolerable. The scrub stream for the absorber would have to be pumped up through the pressure difference between the stripper-rectifier and the absorber-fractionator, involving an additional cost which probably would be insignificant.

It must be concluded that all factors point toward the desirability of operating the stripper-rectifier system at pressures several atmospheres lower than the absorber-fractionator system. There is only one reservation: the ease with which the stripper-rectifier system meets the required performance at a pressure of 20 atm allows little incentive to pay any price for improvement. This would apply particularly to the implicit cost of complicating the operation of the plant.

7.6 Effect of Number of Stages on the Performance of the Stripper-Rectifier System

Varying the number of stages in the system produces the expected, and largely predictable, effect. The use of additional stages in the stripping section reduces the Kr losses by an amount approximately equal to the Kr extraction factor per stage. The effect of varying the number of stages in the rectifying section is more interesting. The flow ratio in this section is very near unity (almost total reflux). This section must span the gap between the effluent gas concentration and the feed concentration. It can do so exactly for only one component. A study was made in which

ORNL DWG. 73-726 RI

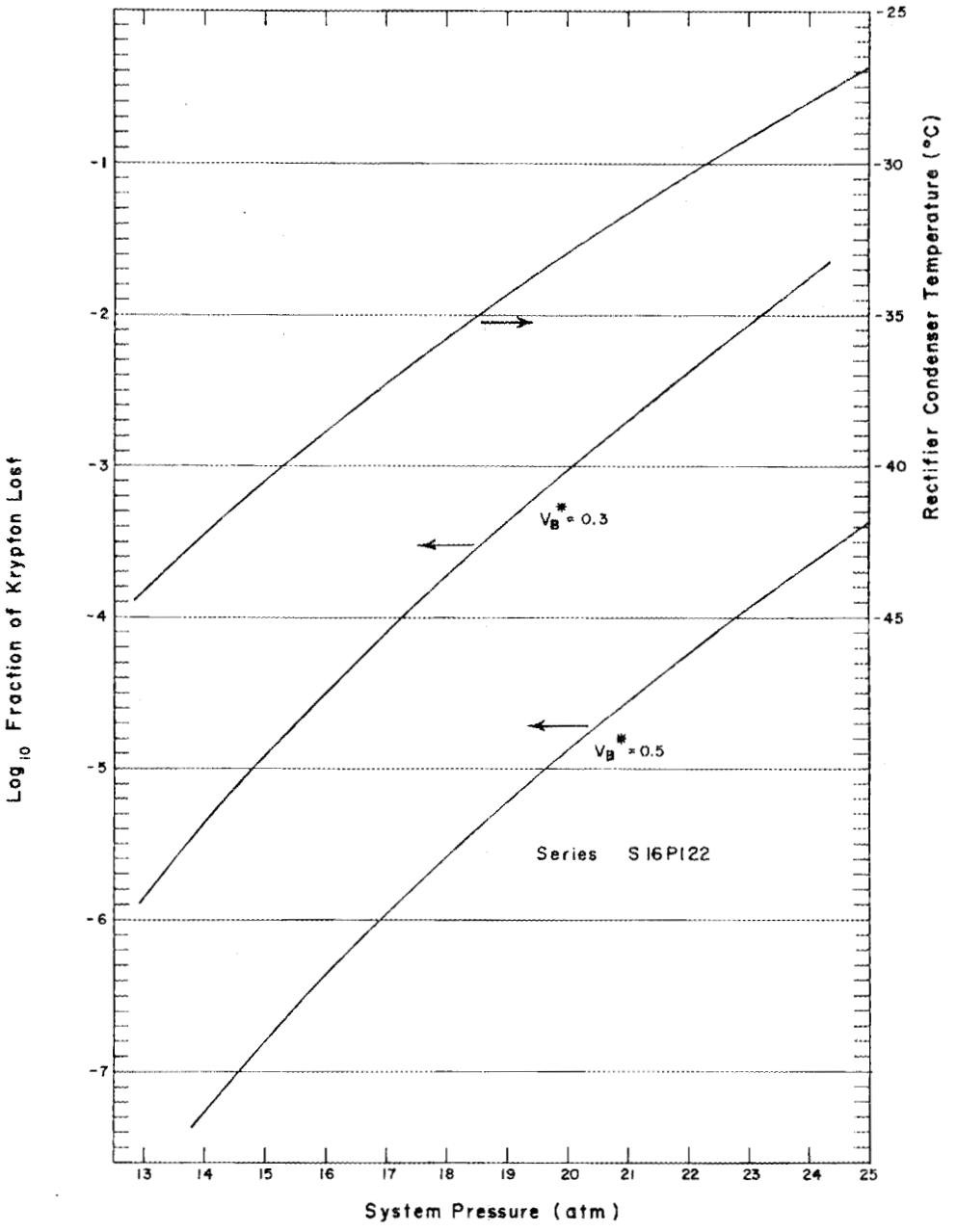

Fig. 17. Effect of System Pressure on Krypton Losses from the Stripper-Rectifier System and on Rectifier Condenser Temperature for Series S16P122.

the number of stages of rectification was varied from 2 (the smallest number admissible by the program) to 9, using series S16B122 as a reference. The results are plotted in Fig. 18. Addition of the third stage accounted for nearly an order-of-magnitude reduction in the Kr loss, and the fourth had a similar effect. The sixth and seventh stages, however, contributed little. Examination of the concentration profiles from series S21B132 (9 rectifier stages) shows that concentrations in the rectification section have "pinched" near the feed stage, and that additional stages would have contributed nothing. This effect is not likely to be altered by reasonable variations in feed composition or reasonable changes in the effluent gas concentration. The performance of the rectifier will be completely unaffected by the performance of the stripper as long as the Kr losses do not exceed a few percent. One concludes that there should be some rectifier stages, but that more than about six or seven are unnecessary.

Two series (S22BB122 and S22BB132) were run to demonstrate that systems with an adequate number of stages (but still a quite reasonable number) could reduce the Kr and light-gas content of the stripped CO_2 to a negligible value. Series S22BB122 and S22BB132 were identical except for enthalpy of the feed; the former used 100 cal/mole (sufficient condensation to reduce the vapor and liquid rates in the rectifier to 7.7% of the feed rate at $V_B^* = 0.3$), and the latter used 800 cal/mole (which resulted in about a 1.3% increase in the vapor rate from the stripping to the rectifying section). Sixteen stripping stages and six rectifying stages were used (see Table 4). At $V_B^* = 0.3$, the Kr losses were 6.4×10^{-4} and 2.0×10^{-4}%, respectively. The O_2 and CO concentrations were of the order of 10^{-16} mole fraction, and the N_2 concentration of the order of 10^{-18} mole fraction, in the stripped CO_2 stream. This more than meets every desired performance specification.

The value of the correlating parameters, A and B, for these runs is of some interest. The value of -B was expected to be about 16, or possibly a little higher, in view of the large number of stages in the rectifying section. In fact, S22BB122 and S22BB132 yielded different values: 18.1 and 16, respectively. The corresponding values of exp (A) were 2×10^{-17} and 13×10^{-17}. Although these are sufficiently close to the expected

ORNL DWG. 73-729RI

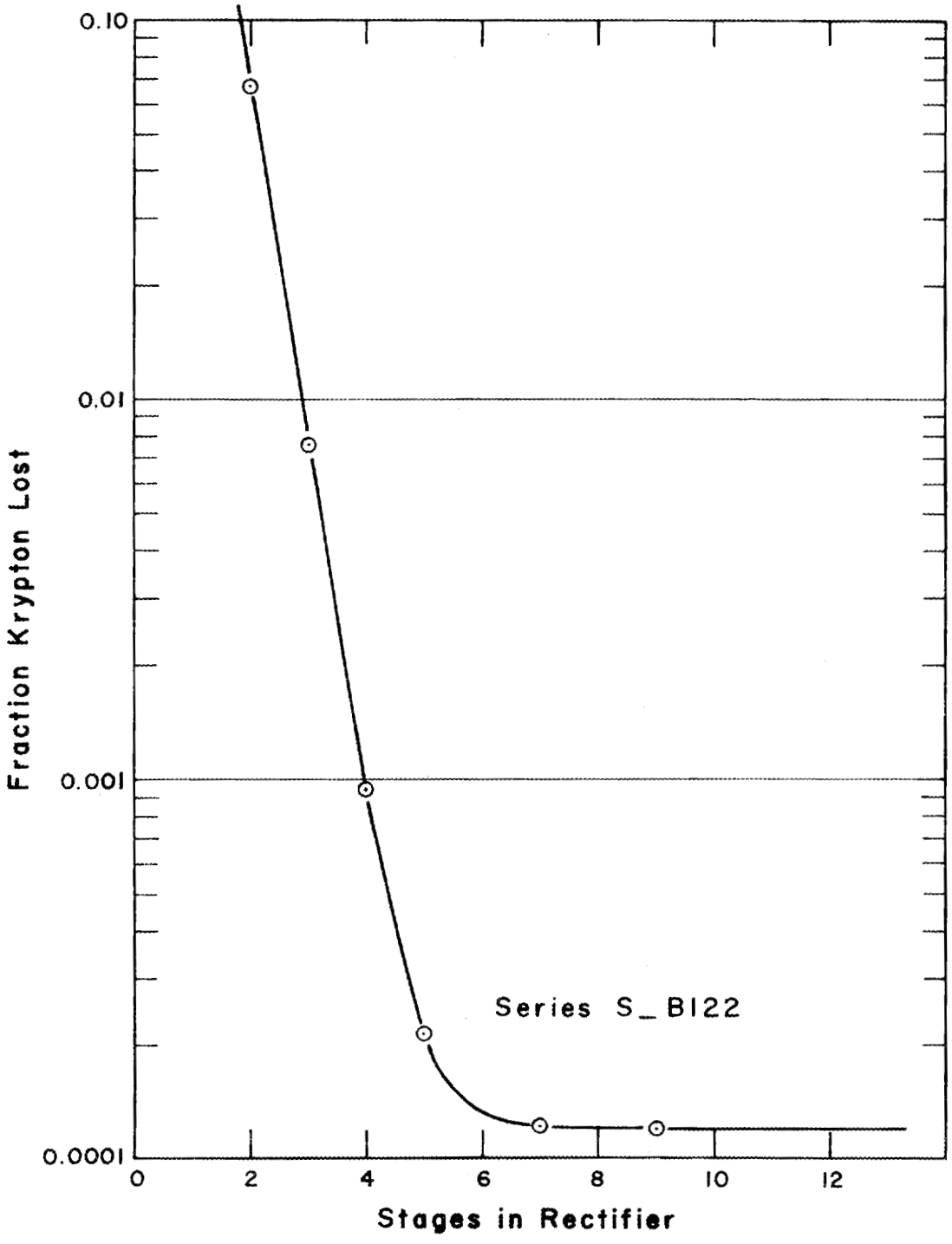

Fig. 18. Effect of the Number of Stages in the Rectifier on Krypton Losses from the Stripper-Rectifier System.

value to satisfy theory, the magnitude of the difference between them is puzzling.

8. CONCLUSIONS

This study has examined independently the absorber-fractionator system and the stripper-rectifier system of the KALC process, exploring design parameters and operating variables that:

(1) Are amenable to study by a computer program built around a steady-state model of the KALC system using less than 50 equilibrium stages.

(2) Fall within the range of operating possibilities currently thought to be plausible for KALC.

(3) Do not require inordinate amounts of computer time.

(4) Are thought by the author to be important.

No claim is made that this study is complete. Further, although the most accurate data available were used to formulate the program, its accuracy in representing the systems must be considered questionable until experimental validation (or basis for revision) is forthcoming. The results of this study do, however, allow some conclusions. The KALC system passes the test of mathematical simulation. The calculations show that, at least on paper, the process will work.

The absorber will require at least 22 theoretical stages and the fractionator at least 16. Control of the effluent gas flow rate from the absorber by control of the scrub temperature is theoretically feasible, but is too sensitive for practical application; a condenser at the top of the absorber seems to be required. The scrub rate will have to be at least 20 times the feed rate when the feed gas contains as much as 10% light gases, and will have to be higher for higher light-gas concentrations. A condenser is required at, or close to, the absorber feed stage. Apparently, there is an optimal condensation rate involving factors which were not quantified in this study. The boilup rate at the bottom of the fractionator was used as the primary variable in this study and proved to be

very important. When all the other variables are specified, a "best" operating point can be found by adjusting this rate. At higher boilup rates the DF decreases, and at lower rates the CF decreases. The allowable operating band for this variable is only about 0.2%, but the temperature profile in the fractionator provides an excellent indication of the location of the "best" operating point and can probably be used for control. The required boilup rate ranges from 2.5 to 4 times the feed rate, depending on other variables. The feed concentration is an important variable which was given only cursory attention since changes in the scrub rate and in the feed concentration produced about the same effect in the steady-state model. Feed concentration fluctuations will be large and frequent in the application of KALC to HTGR burner off-gas processing. In application, a scrub rate high enough to handle the feed most of the time will probably be fixed, and the other variables (i.e., the condenser on the absorber effluent, the condenser at the feed stage, and the boilup rate) will be adjusted to give the most effective operation under the prevailing feed load.

Conclusions from the study of the stripper-rectifier system are more straightforward. Results indicate that the stripper section will require about 14 stages to reduce the Kr concentration by a factor of 10^{-6} when the stripper boilup rate is 0.3 times the stripper feed rate. The higher the boilup rate, the cleaner the recycle stream. About six stages of rectification should be used above the feed stage, but more than about eight stages would provide no advantage. The main condenser should be located at the feed stage, reducing the flow in the rectifier section to not more than 0.1 times the flow in the stripper. Reduction of the effluent gas flow rate from the final condenser to 10^{-5} times the feed and accommodation of light-gas concentrations as high as 50% at this point seem feasible. In an actual system, this would be effected by controlling the temperature of the effluent gas. One important conclusion from this study is that operation of the stripper-rectifier system at 15 atm gives much better performance than operation at 20 atm; performance at 25 atm is correspondingly less satisfactory. This effect of pressure was not

observed in the absorber-fractionator study, where a rather obscure optimum at about 20 atm was deduced from qualitative arguments.

The decoupling of the absorber-fractionator system from the stripper-rectifier system proved to be justified. It was established that about half of the Kr, left in the scrub stream by imperfect stripper operation, reports to the decontaminated gas leaving the top of the absorber. There were no complicating subtleties. The feed to the stripper-rectifier system, produced by the absorber-fractionator system, had little effect on the performance of the stripper-rectifier system, which readily accommodated to a range of feed concentrations.

This study provides a basis for planning in the experimental phase of the program which, with the completion of the facilities presently under construction, will soon be under way. Subsequently, the computer program will be used to complement the experimental work, attempting to simulate particular experimental runs to assist in the interpretation of the data as well as to validate the computer program. If a sufficient degree of confidence in the computer program can be established, future studies will explore the operating regions of interest more thoroughly and modifications will be attempted to permit studies of system transients and economic optimization.

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10. APPENDIXES

10.1 Appendix A: Computer Program Input

The computer program, QUEST, for performing calculations on the KALC system can handle two connected towers with feed and condensation between them. The input to the program has been tailored to provide flexibility to accommodate several different modes of KALC operation. The IBM 360/91 computer, on which the program is run, is addressed through the PDP-10 system, making it convenient to format the input with only one number per record. All real numbers are read into a G10.3 field. All integers are read into an I2 field.

The first number in each case must be an integer, which, if positive, is interpreted as a case number. For the initial case in a series, the first number must be zero. The next record is the name of the series, which may be eight characters long. The following record is an integer, the case number of the first case. The subsequent two records are integers that designate the numbers of stages in the upper tower (the absorber) and the lower tower (the fractionator). (The reboiler at the bottom of the lower tower is counted as a stage.)

All remaining input numbers for a case are real. The record that follows is the system pressure in atmospheres; the subsequent five records are the composition of the feed with regard to O_2, N_2, CO, Kr, and Xe (expressed in mole fractions), respectively. The program is not capable of describing the Xe performance; thus the Xe concentration should be zero. The next record is the enthalpy of the feed (in calories per mole), which, as noted elsewhere, includes the heat removed from the intertower condenser. The subsequent record is the scrub rate, the next the temperature of the scrub (in °C), and, finally, the nominal boilup rate at the bottom of the lower column. The nominal boilup rate is given in terms of the moles of saturated CO_2 vapor, which the program converts into heat units. The calculation literally yields a vapor rate from the first stage equal to this number only when the liquid entering the first stage from the stage above is very close to the saturation temperature of pure CO_2.

The case number for each case after the initial case appears in the first record. The program retains the number of stages, the system

pressure, and the feed composition given in the initial case. It also remembers the calculated profile of compositions and temperatures from the previous case. The records following the case number are: the enthalpy of the feed, the scrub rate, the scrub temperature, and the nominal boilup rate. If it is desired to change the number of stages, feed composition, or system pressure, a zero should be entered instead of the case number, and the format for initiating a series should be repeated.

The description given above may be considered a standard case. Its variations will now be enumerated. If it is desired to specify a boilup ratio rather than a boilup rate (a heat input) to the bottom stage, a negative number is used. The magnitude of this number is interpreted as the ratio of the vapor leaving the bottom stage to the liquid entering that stage.

If a scrub composition is to be specified, then the scrub rate should be entered as zero. The scrub temperature and boilup rate follow in standard sequence. The scrub temperature should be a negative number. Following the boilup rate, five records designating the composition of the scrub are entered in the same format used for the feed. Finally, the actual scrub rate is entered. The scrub composition, once established, will be remembered and used until changed or until the series is reinitialized. The default option gives pure liquid CO_2 scrub.

If it is desired to specify the vapor rate leaving the top of the top tower rather than the temperature of the scrub, this can be done by entering a positive number between 0 and 1 for the scrub temperature. This is interpreted as the difference between the feed rate and the exit vapor rate (or the net CO_2 removal from the feed). For instance, to specify an exit vapor rate of 0.9, the number 0.1 should be entered in the record normally used for the scrub temperature. An enthalpy balance is made to arrive at the scrub temperature that yields the specified vapor rate, and this temperature is printed. It is not possible to specify a boilup ratio for the bottom stage and a vapor rate for the top. It is possible to specify a scrub composition and a vapor rate at the top.

The upper tower can be run with a condenser rather than a scrub stream. This is effected by entering a scrub rate less than 1. If the number lies between 1 and 0, it is interpreted as a condensation ratio, R, which is the ratio of the vapor leaving the top stage to the vapor entering the top stage. If the number is less than 0, it is interpreted as a condensation rate, based on the heat required to condense the given number of moles of saturated CO_2 vapor. When calculations are being made in the condenser mode, the condenser is considered a stage. The scrub temperature must be given but is ignored.

If it is desired to specify an effluent vapor rate for a stripper system (when the top stage is a condenser and there is no scrub), this can be done by setting the scrub rate equal to 0 and the "scrub temperature" to the desired value for the effluent vapor rate. The method by which this is effected is treated in Appendix B. The output is automatically modified to report the effluent gas rate, reflux ratio, and fraction of Kr lost to bottoms when a stripper mode calculation is made.

When the case number is negative, the run is terminated.

A typical input is shown in Table 8.

10.2 Appendix B: Nominal and Actual Input Variable Values

10.2.1 Nominal Reboiler Vapor Rate

The primary variable used throughout this study was the heat input to the reboiler. Using the heat equal to the latent heat of a quantity of CO_2 was thought to be more desirable than using calories per unit molar feed rate, a number whose magnitude requires some mental manipulation to interpret. The obvious basis was pure CO_2 at saturation at the pressure of the system. Thus the heat to the reboiler was specified in terms of the number of moles of CO_2 per mole of feed; when the light-gas content in the lower part of the fractionator was small, it numerically equaled the vapor rate from the bottom stage. This suggested the name "nominal reboiler vapor rate" for this number. When the liquid entering the bottom stage was colder than the saturation temperature, part of the heat

Table 8. Typical Input for the Computer Program.

```
 0            LOOP
 448903       SERIES
 1            CASE
26            ABSORBER
18            FRACTIONATOR
20.           PRESSURE
.06           O2
.03           N2
.005          CO
.00001        KR
.00           XE
-1000.        FEED ENTHALPY
26.           SCRUB
.1            EFFLUENT GAS
3.4           BOILUP
 2            CASE
-1000.
26.
.1
3.45
 3            CASE
-1000.
26.
.1
3.5
 4            CASE
-1000.
26.
.1
3.55
 5            CASE
-1000.
26.
.1
3.6
 6            CASE
-1000.
26.
.1
3.65
 7            CASE
-1000.
26.
.1
3.7
-1
*
```

was used to warm the liquid and the vapor rate was less than nominal. Since the variable used is clear and unambiguous, the relationship of the actual vapor rate to the nominal rate is of superficial significance to the study, but is interesting. No simple analysis was successful in expressing this relationship, although the ratio of actual to nominal boilup rate correlated approximately with the CF for the run. A plot of this correlation using a large sample of runs is shown in Fig. 19. The group of points is bounded below by series 24P9AA, at 15-atm pressure, and above by series 24P9AB, at 25 atm. At values of CF equal to 10, 100, and 1000, all of the values for the ratio lie above 0.95, 0.995, and 0.9995, respectively.

10.2.2 Nominal Effluent Gas Rate for Absorber-Fractionator System

When an effluent gas rate (net CO_2 removal) was specified, an appropriate temperature for the scrub stream was calculated by the following method:

(1) The effluent gas was assumed to contain all of the light gases entering with the feed; its composition, dew point, and enthalpy at the dew point were calculated.

(2) The liquid rate leaving the bottom stage was computed by an overall material balance; it was assumed to be pure CO_2 at saturation temperature at the pressure of the system. Its enthalpy was calculated.

(3) The enthalpy of the entering scrub was computed from an overall enthalpy balance. Its temperature was calculated from the specific enthalpy.

Assumptions required for steps (1) and (2) are invalid when significant amounts of light gases leave with the exit liquid from the bottom of the fractionator. At the time of input to the calculation, the scrub temperature is established; hence the deviation appears in the calculated effluent gas rate. Only a small decrease in temperature of the liquid stream leaving the bottom of the reboiler causes a significant increase in effluent gas rate above the nominal value.

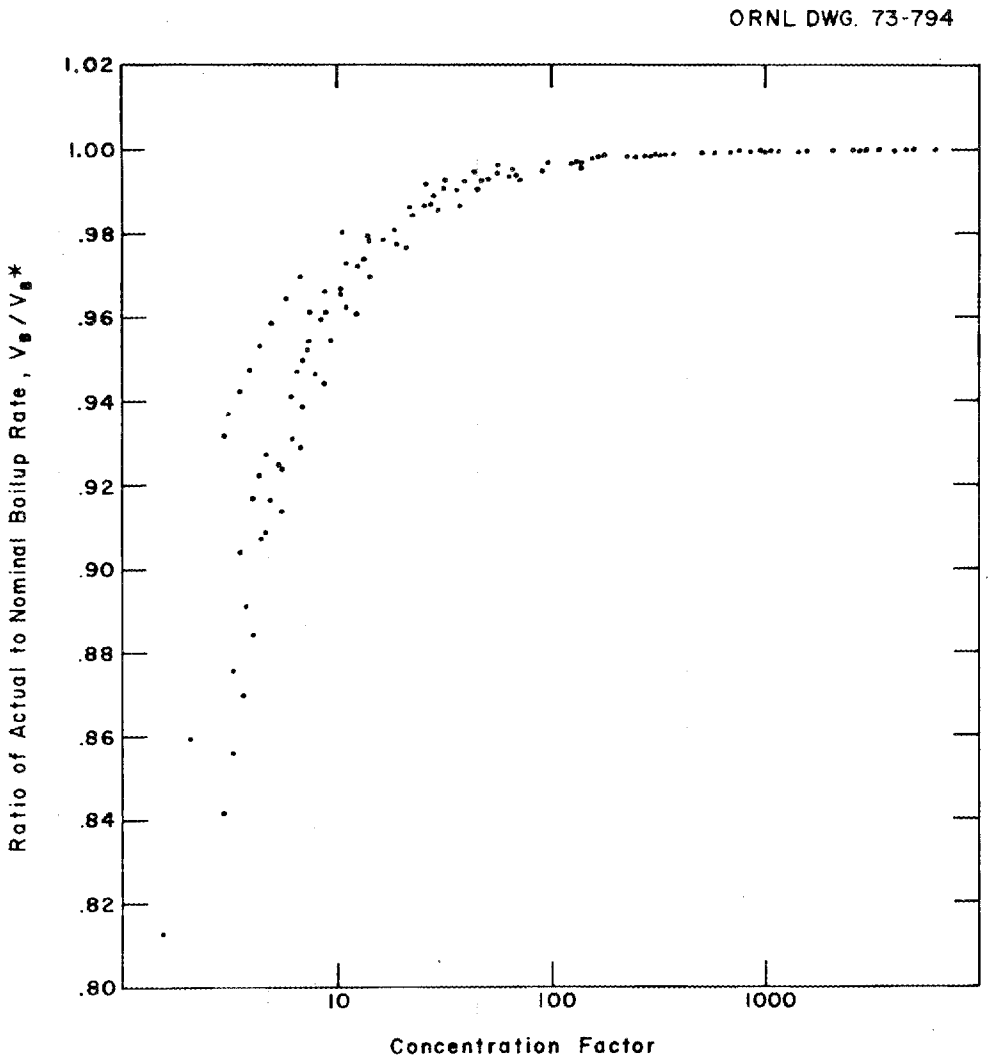

Fig. 19. Correlation of the Ratio of Actual to Nominal Reboiler Vapor Rate with the Calculated Concentration Factor.

Since the major dependent variables of the study, DF and CF, were very insensitive to effluent gas rate, and since values of the actual effluent gas rate were never greatly different from the nominal (see Fig. 14) and quite close for CF values above about 50, the simple method described above of using the input nominal effluent gas rate was thought to be appropriate.

10.2.3 Nominal Effluent Gas Rate for the Stripper-Rectifier System

Some manipulation is involved in satisfying a specified effluent gas rate from a condenser when that condenser is fed vapor by a cascade and 99.999% of the vapor is condensed. The computer program is written to allow the use of a specified number, R, on the top stage (condenser) of the stripper. This number is interpreted to be the fraction of the total material fed to the stage remaining in the vapor phase at equilibrium (normally, R is determined by an enthalpy balance). The following procedure is used to obtain R from input variables that include a specified effluent gas rate:

(1) All light gases are assumed to report to the effluent gas, and the composition of the gas is calculated from an overall material balance.

(2) The liquid phase in equilibrium with the effluent gas is calculated, along with its temperature and specific enthalpy.

(3) An enthalpy balance is made over the whole system, excluding the top stage and assuming zero light-gas concentration. This yields an approximate value for the vapor rate entering the top stage.

(4) The composition of this entering vapor is calculated from a material balance, using its approximated rate. Since all reflux ratios of interest are very high, this composition calculation is quite accurate.

(5) The dew point and the specific enthalpy of the entering vapor are calculated.

(6) An enthalpy balance (similar to step 3) is made over the whole system, excluding the top stage, using the calculated specific enthalpies to yield an accurate value for the vapor rate entering the top stage.

(7) R is set equal to the ratio of the specified effluent gas rate and the vapor rate entering the top stage.

This procedure proved to be surprisingly accurate, generating effluent gas rates that deviated as much as a few tenths of a percent from the specified rate only when the Kr losses exceeded 10%.

10.3 Appendix C: Computer Program

The listing of the computer program used in this study is given on the pages that follow.

```
C  THIS DOES A COLUMN CALC USING QUEST
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      DIMENSION BL(5)
      COMMON/FED/FEED,BFEED,XF(5)
      COMMON/LOOM/TA(52),WA(52),VA(52),XA(5,52),YA(5,52),
     1 HLA(52),HVA(52),IA(52)
      COMMON/ZOOM/XAS(3,5,52),YAS(3,5,52)
      COMMON/NUMB/NUB,NAME(2),NTOP,NF,LOOP
C  LOOP LT 0 = STOP, EQ 0 = FRESH START, GT 0 = KEEP PROFILE
      DO9JOB=1,200
      READ 150,LOOP
  150 FORMAT(I2)
      PRINT 151,LOOP
  151 FORMAT(7H1LOOP =,I3)
      IF(LOOP)30,1,1
    1 CALL SRTUP
      IF(LOOP)35,32,32
   35 PRINT 135
  135 FORMAT(' ...TOO BAD, FATAL INPUT ERROR...')
      GOTO 9
   32 NT = NTOP + 1
      IF(NTOP-50) 28,28,31
   31 PRINT 130
  130 FORMAT(' TOO MANY STAGES'/20X,' YOU BLEW IT')
      GOTO 30
   28 LOOP = 0
      LEX = 4
      DO11JIM=1,LEX
      CALL STEP
      IF(LOOP)7,7,11
   11 CONTINUE
      DO4I=1,5
      BL(I) = 1.
      IF(XF(I))4,4,2
    2 BL(I) = (XA(I,2)*WA(2)+YA(I,NT)*VA(NT))/
     1 (XF(I)*FEED+WA(NT+1)*XA(I,NT+1))
   13 DO3J=2,NT
      XA(I,J) = XA(I,J)/BL(I)
    3 YA(I,J) = YA(I,J)/BL(I)
    4 CONTINUE
      PRINT 106, (BL(I),I=1,5)
   12 DO8JUX =1,12
      JAX = 2 + JUX/4
      JIX = JAX + JUX/3
      DO20KA=1,JIX
      CALL STEP
      IF(LOOP)7,7,20
   20 CONTINUE
      DO21KA=1,3
      DO25KB=1,JAX
      CALL STEP
      IF(LOOP)7,7,25
   25 CONTINUE
      DO22N=1,NT
      DO22J=1,5
      XAS(KA,J,N) = XA(J,N)
      YAS(KA,J,N) = YA(J,N)
   22 CONTINUE
      DO24I=1,5
      BL(I) = 1.
      IF(XF(I))24,24,26
   26 BL(I) = (XA(I,2)*WA(2)+YA(I,NT)*VA(NT))/
     1 (XF(I)*FEED+WA(NT+1)*XA(I,NT+1))
   24 CONTINUE
      PRINT 106, (BL(I),I=1,5)
   21 CONTINUE
      CALL ZAPPO
  106 FORMAT(5H MATL/6H  BAL  ,5F10.4)
    8 CONTINUE
      PRINT 113
      JAX = 2*JAX
      DO27JIM=1,JAX
      CALL STEP
      IF(LOOP)7,7,27
   27 CONTINUE
      GOTO29
  113 FORMAT(' ... GROAN ...')
    7 PRINT 10
   29 CALL PUT
    9 CONTINUE
   30 CONTINUE
      STOP
   10 FORMAT (' GOT IT !!!'/)
      END
```

```
      SUBROUTINE SETUP
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      COMMON/WREF/GXY(5),HCOM,RTOT
      COMMON/NOW/P,T,X(5),Y(5)
      COMMON/ENTH/HLIQ,HVAP
      COMMON/LOOM/TA(52),WA(52),VA(52),XA(5,52),YA(5,52),
     1 HLA(52),HVA(52),TA(52)
      COMMON/FEED/FEED,HFEED,XF(5)
      COMMON/NUMB/NUP,NAME(2),NTOP,NF,LOOP
      NUP = LOOP
      IF(LOOP)30,10,20
   10 READ 151, NAME
      READ 150, NUP
  151 FORMAT(2A4)
      PRINT 111, NAME, NUP
  111 FORMAT(' SERIES ',2A4,' CASE ',I3)
      READ 150,NABS
      PRINT 101,NABS
  101 FORMAT(' STAGES IN ABSORBER',I3)
  100 FORMAT(D10.3)
  102 FORMAT(' IN FRACTIONATOR',I3)
      READ 150, NFC
  150 FORMAT(I2)
      PRINT 102, NFC
      NF = NFC+1
      NTOP = NFC + NABS
      NL = NTOP +2
      FEED  = 1.
      FF = -1.
      READ 100,RTOT
      PRINT 108,RTOT
  108 FORMAT(' SYS PRESSURE =',F8.3)
      DO2I=1,NL
    2 TA(I) = 0
      PRINT 103
  103 FORMAT(' FEED COMP: O2,N2,CO,KP,XF')
      SXFS=0.
      DO1J=1,5
      XA(J,1)  = 0.
      YA(J,1)  = 0.
      XA(J,NL)  = 0.
      YA(J,NL)  = 0.
      READ 100,XF(J)
      SXFS=SXFS+XF(J)
    1 PRINT 110,XF(J)
      IF(SXFS-1.)21,21,31
  110 FORMAT(1H ,F12.9)
   20 PRINT 111, NAME,NUP
   21 READ 100,HFEED
      PRINT 104,HFEED
  104 FORMAT(' FEED ENTHALPY =',F10.3)
      NL = NTOP +2
      READ 100,WA(NL)
      WA(NL-1) = WA(NL)
      PRINT 105,WA(NL)
  105 FORMAT (' SCRUB/FEED =',F8.3)
      READ 100, TC
C TC .LT. 0 YIELDS TEMP, TC.GT.0 IS EXIT GAS INFO
      IF(TC)13,12,12
   12 IF(WA(NL))50,51,50
   51 PRINT 114, TC
  114 FORMAT(' EFFLUENT GAS RATE =',F11.9)
      T = TVP(RTOT)
      GOTO14
   50 PRINT 109, TC
  109 FORMAT (' NET CO2 REMOVAL = ',F10.5)
      T=248.15
      TA(NL)  = T
   52 IF(TC-1.+SXFS)14,31,31
   31 LOOP = -1
      GO TO 14
   13 PRINT 106, TC
  106 FORMAT(' SCRUB DEG C =',F10.3)
      T = TC + 273.15
      TA(NL)  = T
   14 DO3J=1,5
      X(J)  = 0.
    3 Y(J)  = 0.
      CALL ENTHAL
      HLA(NL)  = HLIQ*WA(NL)
      READ 100,VA(2)
      PRINT 107,VA(2)
  107 FORMAT (' BOILER VAP RATE =',F8.3)
      T = TVP(RTOT)
      TA(2)  = T
      CALL ENTHAL
      HLBOT = HLIQ
      HVBOT=HVAP
      HVA(1)  = (HVAP-HLIQ)*VA(2)
      VA(1)  = 0.
      WA(1)  = 0.
C  IF VA(2) .LT.0, INTERPRETED AS BOILUP RATIO
      IF(VA(2))27,26,26
   27 TA(2)  = 1
      WA(1)  = -VA(2)
      HVA(1)  = HVAP-HLIQ
      VA(2)  = 1.
      IF(TC)26,29,29
   29 TC = -30.
      TA(NL)  = TC + 273.15
      T = TA(NL)
      CALL ENTHAL
      HLA(NL)  = HLIQ*WA(NL)
      PRINT 106, TC
   26 NLWA = 1000.*WA(NL)
      IF(NLWA)22,23,25
C  IF SCRUB .LT.0, TAKEN AS RATE FOR CONDENSATION
C  SCRUB=0, TC NEG, ANALYSIS REQUIRED - AND NEW SCRUB RATE
C  SCRUB=0, TC POS, NO SCRUB AND TC = EFFLUENT GAS RATE
C  IF 1 .GT. SCRUB .GT. 0, TAKEN AS FIXED P FOR CONDENSATION
   22 HLA(NL) = WA(NL)*(HVAP-HLIQ)
      WA(NL-1)  =  -WA(NL)
      WA(NL)  = 0.
      GOTO24
```

```
   23 IF(TC*(1.-TC))45,64,64
C SETUP FOR STRIPPER
   64 RF = VA(2) + (HFEED-HLBOT)/(HVBOT-HLBOT)
      IF(RF)62,62,68
   62 LOOP=-1
   68 HNET = RF*(HVBOT -HLBOT)
      IF(TC)45,67,65
   65 SXS = 0.
      DO60 J=1,5
      YA(J,NL) = XF(J)/TC
      GXY(J) = YA(J,NL)*.9905
   60 SXS = SXS +GXY(J)
      IF(1.-SXS)67,67,69
   69 P = .99
      T = TA(2) -SXS*(TA(2)-220.)
      CALL QUEND
   67 HLTOP = HLTQ
      DO61 J=1,5
   61 GXY(J) = .9905*(X(J)*RF+YA(J,NL)*TC)/(RF+TC)
      P = .99
      T = (T+TA(2))/2.
      CALL QUEND
      WA(NL-1) = (HNET-TC*HLTOP)/(HVAP-HLTOP)
      VA(NL) = TC/WA(NL-1)
      WA(NL) = 0.
      GOTO66
   45 SXS=0.
      DO6J=1,5
      READ 100, X(J)
      SXS=SXS+X(J)
    6 XA(J,NL) = X(J)
      IF(SXS-.1)44,31,31
   44 READ 100, WA(NL)
      WA(NL-1) = WA(NL)
      T = TC + 273.15
      CALL ENTHAL
      HLA(NL) = WA(NL)*HLTQ
      PRINT 112, (X(I),I=1,5)
  112 FORMAT(' SCRUB ANALYSIS =',/5D11.4)
   25 IF(WA(NL)-1.)43,43,28
   28 IF(TC)24,40,40
   40 P = .99
      T = TA(2)
      DO41J = 1,5
   41 GXY(J) = XF(J)/(1.-TC)
      CALL QUEND
      DO42J = 1,5
   42 X(J) = XA(J,NL)
      HLA(NL) = HVAP*(1.-TC)+(WA(NL)+TC)*HLBOT-HFEED - HVA(1)
      HTOT = HLA(NL)/WA(NL)
      CALL ENTHAL
      CALL TENTH
      TA(NL) = T
      TC = T -273.15
      PRINT 106, TC
      GO TO 24
   43 VA(NL) = WA(NL)
      WA(NL) = 0.
      WA(NL-1)=.01+DABS((HFEED+HVA(1)-HLTQ)*(1.-VA(NL))/(HVBOT-HLBOT))
   66 IA(NL-1) = 2
      RF = (1.-VA(NL))/VA(NL)
      PRINT 113,VA(NL),RF
  113 FORMAT(8H R-TOP=,F10.3,15H REFLUX RATIO =,F10.3)
   24 IF(LOOP)30,11,30
   11 HVA(2) = VA(2)*HVBOT
      IF(RF)7,7,63
   63 WA(2) = 1. + VA(2)
      HLA(2) = HLBOT*WA(2)
      GOTO 8
    7 HLA(2) = WA(NL-1)*HLBOT
      WA(2) = WA(NL-1) + .5
    8 NT = NFC + 1
      DO4I=2,NT
      HVA(I) = HVA(2)
      HLA(I)=HLA(2)
      WA(I)=WA(2)
      VA(I)=VA(2)
      TA(I) = TA(2)-.01
      DO4J=1,5
      XA(J,I)=0.
      YA(J,I)=0.
    4 CONTINUE
      NL = NL-1
      NT = NT+1
      WA(NT) = WA(NL)
      VA(NT) = VA(2) + .5*(HFEED-HLBOT)/(HVBOT-HLBOT)
      HLA(NT) = HLBOT*WA(NL)
      HVA(NT) = VA(NT)*HVAP
      DO5I=NT,NL
      WA(I)=WA(NT)
      VA(I)=VA(NT)
      HLA(I)=HLA(NT)
      HVA(I)=HVA(NT)
      TA(I) = TA(2) -5.
      RF = I-NT
      RF = DEXP(RF)
      DO5J=1,5
      XA(J,I) = X(J)/RF
      YA(J,I) = XF(J)+ Y(J)/RF
    5 CONTINUE
      IA(NT) = -1
   30 RETURN
      END
```

```
      SUBROUTINE PUT
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      DIMENSION BL(5),NDATE(2)
      COMMON/FEED/FEED,HFEED,XF(5)
      COMMON/WEFC/GXY(5),HTOT,PTOT
      COMMON/LOOM/TA(52),WA(52),VA(52),XA(5,52),YA(5,52),
     1 HLA(52),HVA(52),IA(52)
      COMMON/NUMB/NUP,NAME(2),NTOP,NF,LOOP
      LIP = -LOOP
      CALL IDAY(NDATE)
      PRINT 97,NAME,NUP,NDATE
   97 FORMAT(1H1//' SERIES ',2A4,', CASE ',I3,20X,2A4)
      PRINT 99,LIP
   99 FORMAT(' ITERATIONS =',I5)
      NFC = NF -1
      NABS = NTOP -NFC
      NL = NTOP +2
      IF(WA(NL))16,17,16
   17 PRINT 213,NABS
  213 FORMAT(' STAGES IN PRCTIFIER =',I3)
      PRINT 214,NFC
  214 FORMAT(' STAGES IN STRIPPER =',I3)
      GOTO18
   16 PRINT 201,NABS
  201 FORMAT(' STAGES IN ABSORBER',I3)
  202 FORMAT(' IN FRACTIONATOR',I3)
      PRINT 202, NFC
   18 PRINT 203
  203 FORMAT(' FEED COMP: O2,N2,CO,KR,XE')
      PRINT 210,(XF(J),J=1,5)
  210 FORMAT(1H ,5D13.4)
      PRINT 204,HFEED
  204 FORMAT(' FEED ENTHALPY =',F10.3)
      PRINT 205,WA(NL)
  205 FORMAT (' SCRUB/FEED =',F8.3)
      IF(WA(NL))14,13,14
   13 RF = WA(NL-1)/VA(NL-1)
      PRINT 209,RF
  209 FORMAT(' REFLUX RATIO =',F10.3)
      PRINT 212,VA(NL-1)
  212 FORMAT(' EFFLUENT GAS =',F12.9)
      PRINT 215
  215 FORMAT(' COMPOSITION =')
      PRINT 210,(YA(I,NL-1),I=1,5)
      SX=0.
      DO19J=1,5
   19 SX=SX+YA(J,NL-1)
      SX=1.-SX
      PRINT 216,SX
  216 FORMAT(10X,5HCO2 =,F10.7)
      SX=WA(2)*XA(4,2)/XF(4)
      PRINT 217,SX
  217 FORMAT(' FRACTION KR LOST =',F12.9)
      GOTO15
   14 HMC = HLA(NL)/WA(NL)
      TC = TA(NL) -273.15
      PRINT 211, TC
  211 FORMAT(' SCRUB TEMP =',F9.3)
      PRINT 206,HMC
  206 FORMAT(' SCRUB ENTHALPY =',F10.3)
   15 PRINT 207,VA(2)
  207 FORMAT (' BOILER VAP RATE =',F8.3)
      PRINT 208,PTOT
  208 FORMAT(' SYS PRESSURE =',F8.3)
      PRINT 100
  100 FORMAT(/' STAGE   T       L       V     L/V    H LIQ    H VAP'/)
C  L AND V IN MOLES PER **, H IN CAL PER MOLE
      DO1K=1,NTOP
      J = NTOP +2-K
      NST = J-1
      SHLA = 0.
      SHVA = 0.
      IF(WA(J))4,4,3
    3 SHLA = HLA(J)/WA(J)
    4 IF(VA(J))6,6,5
    5 SHVA = HVA(J)/VA(J)
      WOV = WA(J)/VA(J)
    6 T = TA(J)-273.15
      PRINT 101,NST, T,WA(J),VA(J),WOV,SHLA,SHVA
  101 FORMAT(1H ,I3,F8.2,3F7.3,2F9.2)
    1 CONTINUE
      HBAL = (HVA(NTOP+1)+HLA(2))/(HFEED +HVA(1) +HLA(NTOP+2))
      PRINT 112, HBAL
  112 FORMAT (/' HEAT BALANCE =',F7.4)
      PRINT 111
  111 FORMAT(1H1)
      PRINT 102
  102 FORMAT(//' STAGE        O2',8X,2HN2,8X,2HCO,8X,2HKR,8X,2HXE/)
      DO2K=1,NTOP
      J = NTOP +2-K
      NST = J-1
      PRINT 103,NST,(YA(I,J),I=1,5)
      PRINT 104,(XA(I,J),I=1,5)
  103 FORMAT(1H ,I3,3H  Y,5F10.3 )
  104 FORMAT(1H ,3X,3H  X,5F10.3/)
    2 CONTINUE
      DO7J=1,5
      BL(J) = 1.
      IF(XF(J))7,7,8
    8 BL(J) = (WA(2)*XA(J,2)+VA(NTOP+1)*YA(J,NTOP+1))/
     1 (XF(J) + WA(NL)*XA(J,NL))
    7 CONTINUE
      PRINT 106, (BL(I),I=1,5)
  106 FORMAT(5H MATL/6H  BAL  ,5F10.4)
      DF = XF(4)/(VA(NTOP+1)*YA(4,NTOP+1))
      BLG = 0.
      BFG = 0.
      DO9J=1,3
      BFG = BFG + XF(J)
    9 BLG = BLG + XA(J,2)
      BFG = BFG/XF(4)
      BLG = BLG/XA(4,2)
      BFG = BFG/BLG
      PRINT 111
```

```
      PRINT 107, DF
      PRINT 108, BIG,BEG
  107 FORMAT (' DF. = ',F9.2)
  108 FORMAT (' LIGHT GASES / KF =',F8.3,' , CONC. FACTOR =',F9.2//)
      PRINT 110
  110 FORMAT(//5X,'EXTRACTION FACTORS')
      PRINT 102
      DO10K=1,NTOP
      J = NTOP +2-K
      NST = J-1
      DO12I=1,5
      IF(XA(I,J)) 12,12,11
   11 BL(I) = XA(I,J)*VA(J) /XA(I,J) /WA(J)
   12 CONTINUE
   10 PRINT 109,NST,(BL(I),I=1,5)
  109 FORMAT(1H ,I3,3X,5F10.4)
      PRINT 105
  105 FORMAT(/)
      RETURN
      END
```

```
      SUBROUTINE ZAPPO
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      COMMON/FED/FEED,HFEED,XF(5)
      COMMON/LOOM/TI(52),WA(52),VA(52),XA(5,52),YA(5,52),
     1 HLA(52),HVA(52),IP(52)
      COMMON/NUMB/NUP,NAMF(2),NTOP,NF,LOOP
      COMMON/ZOOM/XAS(3,5,52),YAS(3,5,52)
      NL = NTOP + 1
      DO10NS = 2,NL
      DO8NP=1,2
      DO6J=1,5
      IF(XF(J))6,6,1
    1 IF(NP-1)8,2,3
    2 X1 = XAS(1,J,NS)
      X2 = XAS(2,J,NS)
      X3 = XAS(3,J,NS)
      GOTO4
    3 X1 = YAS(1,J,NS)
      X2 = YAS(2,J,NS)
      X3 = YAS(3,J,NS)
    4 DX2 = X3 - X2
      DX1 = X2 - X1
      IF(DX2*DX1)6,6,5
    5 IF((DX2/DX1) - .98)20,20,40
   20 XT = (X2*X2-X1*X3)/(X2+X2-X3-X1)
      GOTO45
   40 DEL = 25.*(X3-X1)
      XT = X2 + DEL
   45 IF(XT)41,41,42
   41 XT = 0.0
   42 IF((XT/X3)-25.)44,44,43
   43 XT = 25.*X3
   44 IF(NP-1)8,21,22
   21 XA(J,NS) = XT
      GOTO6
   22 YA(J,NS) = XT
    6 CONTINUE
    8 CONTINUE
   10 CONTINUE
      RETURN
      END
```

```
      SUBROUTINE STEP
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      DIMENSION CHKY(5,3)
      DATA CHKY/15*0.0/
      COMMON/RAIN/LICK
      COMMON/FED/FEED,HFEED,XF(5)
      COMMON/NUMB/NUP,NAME(2),NTOP,NF,IOOP
      COMMON/LOOM/TA(52),WA(52),VA(52),XA(5,52),YA(5,52),
     1 HLA(52),HVA(52),IA(52)
      LAX = 1 + NTOP/10
      DO13NGO=1,LAX
      LICK = 0
      DO7IT=1,5
      DO6IS=1,NTOP,2
      N = 1+IS
      CALL STAGE(N)
    6 CONTINUE
      DO5IS=2,NTOP,2
      N = 1+IS
      CALL STAGE(N)
    5 CONTINUE
      IOOP = IOOP + 1
    7 CONTINUE
C CHECK CONVERGENCE TROUBLE
      IF((1+(75*NTOP/IOOP)) - LICK) 16,16,17
   16 PRINT 130
  130 FORMAT(' CONVERGENCE TROUBLE'/)
      RETURN
C CHECK CONVERGENCE
   17 BLGS = 0.
      SBOT = 0.
      DO4I=1,5
      IF(XF(I)) 4,4,2
    2 BLG =(XF(I,2)*WA(2)+YA(I,NTOP+1)*VA(NTOP+1))/
     1 (XF(I)*FEED + WA(NTOP+2)*XA(I,NTOP+2))
      BLGS = BLGS + DABS(1.-BLG)
      SBOT = SBOT + XA(I,2)
    4 CONTINUE
      SBOT = SBOT*1.D-6 + 1.D-23
      IF(BLGS -.01) 18,18,13
   18 SCK = 0.02/IOOP
      DO8J=1,5
      Y1 = YA(J,NTOP-1)
      Y2 = YA(J,NF-1)
      Y3 = YA(J,2)
      IF(Y1-SBOT) 10,10,9
    9 SCK = SCK + DABS((Y1- CHKY(J,1))/Y1)
   10 CHKY(J,1) = Y1
      IF(Y2-SBOT) 12,12,11
   11 SCK = SCK + DABS((Y2- CHKY(J,2))/Y2)
   12 CHKY(J,2) = Y2
      IF(Y3-SBOT) 20,20,19
   19 SCK = SCK + DABS((Y3- CHKY(J,3))/Y3)
   20 CHKY(J,3) = Y3
    8 CONTINUE
      IF(SCK -.004) 14,13,13
   13 CONTINUE
      PRINT 100,IOOP
  100 FORMAT (13H ITERATIONS = ,I4)
      RETURN
   14 IOOP = -IOOP
      RETURN
      END

      SUBROUTINE STAGE(N)
C THIS MAKES BALANCE ON A STAGE AND CALLS QUEST
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      COMMON/LOOM/TA(52),WA(52),VA(52),XA(5,52),YA(5,52),
     1 HLA(52),HVA(52),IA(52)
      COMMON/PNTH/HLIQ,HVAP
      COMMON/FED/FEED,HFEED,XF(5)
      COMMON/NOW/P,T,X(5),Y(5)
      COMMON/WEES/GXY(5),HTOT,PTOT
      G = VA(N-1) + WA(N+1)
      HTOT = (HLA(N+1) + HVA(N-1))/G
      P = VA(N)/(VA(N)+WA(N))
      T = TA(N)
      DO1J=1,5
    1 GXY(J) = (VA(N-1)*YA(J,N-1) + WA(N+1)*XA(J,N+1))/G
      IF(IA(N)) 5,2,4
    2 CALL QUEST
    7 TA(N) = T
      VA(N) = P*G
      WA(N) = G-VA(N)
      HLA(N) = HLIQ*WA(N)
      HVA(N) = HVAP*VA(N)
      DO3J=1,5
      XA(J,N) = X(J)
    3 YA(J,N) = Y(J)
      RETURN
C  IA = 1 DENOTES REBOILER
C  IA = 2 DENOTES CONDENSER
    4 IF(IA(N)-2) 10,11,2
   10 P = WA(N-1)
      GOTO12
   11 P = VA(N+1)
   12 CALL QUEND
      GOTO7
    5 GF = G + FEED
      HTOT = (G*HTOT + HFEED)/GF
      DO6J=1,5
    6 GXY(J) = (GXY(J)*G + XF(J)*FEED)/GF
      G = GF
      GOTO2
      END
```

```
      SUBROUTINE QUEST
C GIVES THE EQUILIBRIUM CONCENTRATIONS AND PHASE
C  SPLIT FROM AVERAGE COMP AND TOTAL ENTHALPY
C OXYGEN = 1, NITROGEN = 2, CO = 3, KR = 4 , XE = 5
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      COMMON/PATH/LICK
      COMMON/WERT/GXY(5), HTOT, PTOT
      COMMON/NOW/P,T,X(5),Y(5)
      COMMON/ENTH/HLIQ,HVAP
      DIMENSION CPTX(5,3)
      DATA CPTX(1,1),CPTX(1,2),CPTX(1,3)/532.33,-2.7153,-494.72/
      DATA CPTX(2,1),CPTX(2,2),CPTX(2,3)/720.22,-5.8729,-553.88/
      DATA CPTX(3,1),CPTX(3,2),CPTX(3,3)/484.39,-1.8473,-395.1/
      DATA CPTX(4,1),CPTX(4,2),CPTX(4,3)/188.2,2.8,3000./
      DATA CPTX(5,1),CPTX(5,2),CPTX(5,3)/10.,0.,0./
      TC = T
      TX1 = 0.
      TY1 = 0.
      SX = 0.
      DO17J=1,5
      X(J) = GXY(J)
      Y(J) = GXY(J)
   17 SX = SX + GXY(J)
      SG = SX
      IF(SX-.003)21,21,22
   21 CALL LISWT
      RETURN
   22 SN = 0.
      SD = 1.
      DO13J=1,5
      SN = SN +(CPTX(J,1) +CPTX(J,3)*SX)*GXY(J)
   13 SD = SD +CPTX(J,2)*GXY(J)
      P = (PTOT - SN)/SD
      TMIN = TVP(P)
      IF(217.-TMIN)32,32,33
   33 TMIN = 217.
   32 T = TMIN
      CALL ENTHAL
      HL = HLIQ
      CALL TENTH
      IF(HTOT-HL)30,30,31
   30 P = 0.
      RETURN
   31 TMAX = T
      P = PTOT*(1.-SX)
      T = TVP(P)
      IF(TMAX-T)35,35,34
   34 TMAX = T
   35 T = TC
C     PRINT 103,TMIN,TMAX
  103 FORMAT (12H TEMP LIMITS,2F8.2)
      IF((T-TMIN)*(TMAX-T))28,28,29
   28 T = .4*TMIN + .6*TMAX
   29 IF(P)19,19,20
   19 P = .01
   20 SX = SX*(1.-P)*(1.-P)
      DO3NTRY =1,15
      ET = 1.0
      Z = ZCO2(T)
      P = VPCO2(T)
      PEXP = DEXP(1.7*(PTOT-P)/T)
      PHP = P
      DO11NGO=1,15
      IF(SX-SG)27,26,26
   26 SX = SG
   27 STG = SX
      CT = (1.-STG)*PEXP
      SX = 0.
      SY = 0.
      D = PTOT + P*CT*(1.-Z)/Z
      IF(STG*(PTOT-P) -1.E-8)12,14,14
   12 PSO = 0.
      GO TO 15
   14 PSO = P*(1.-CT) /(PTOT-P)
   15 DO1J=1,5
      W = (PSO+1.)*(CPTX(J,1) +CPTX(J,2)*P +CPTX(J,3)*STG)
      X(J) = GXY(J)/(P*(W/D - 1.)+ 1.)
      Y(J) = (GXY(J) - (1.-P)*X(J))/P
      SX = SX + X(J)
      SY = SY + Y(J)
    1 CONTINUE
      CALL ENTHAL
      IF(ET)18,16,16
   16 RH = DABS(R)
      R = (HTOT - HLIQ)/(HVAP - HLIQ)
C     PRINT 87,R
   87 FORMAT (F11.6)
      IF(R)5,5,6
    5 R = RH/2.
    6 IF(1.-R)8,7,7
    7 IF(0.80-SY)10,10,9
   10 R = RH*(SY+.35)
      GOTO11
    8 R = 1.0
    9 FAC = 2.*(1.-SY)/(2.-SY)
C     PRINT 102,FAC
  102 FORMAT (6H FAC =,F5.3)
      R = FAC*R + (1.-FAC)*RH
      ET = DABS((STG-SX)/SX) +DABS((RH-R)/R) -1.D-6
   11 CONTINUE
      LICK = LICK + 1
C     PRINT 101
  101 FORMAT (10H NO X FIT./)
      IF(HTOT-HLIQ)25,25,18
   25 TMAX = T
C     PRINT 103,TMIN,TMAX
      TX1 = 0.
      TY1 = 0.
      T = (TMAX + TMIN)/2.
      GOTO3
   18 SN = 0.
      SD = 1.
      DO2J=1,5
      SN = SN + (CPTX(J,1) + CPTX(J,3)*SX)*X(J)
```

```
    2 SD = SD + CPTX(J,2)*X(J)
C     PRINT 100,SN,SD
  100 FORMAT (4H SN=,F6.3,4H SD=,F6.3)
      P = (PTOT - SN)/SD
      TX2 = TX1
      TH = T
      TX1 = (T - TMIN)/(TMAX -T)
      TY2 = TY1
      T = TVP(P)
      TM = TMAX - T
      IF(TM)40,40,41
   40 TM = 0.1*(TMAX-TH)
      T = TMAX - TM
   41 TXX = T - TMIN
      IF(TXX)42,42,43
   42 TXX = 0.1*(TH-TMIN)
   43 TY1 = TXX/TM
      PHI = (TX2-TX1)*(1.-SY*SY)
      IF(PHI)46,44,46
   46 PHI = (TY2-TY1)/PHI
      IF(PHI*(50.+PHI))45,45,44
   44 PHI = -20.*SY
   45 TXX = (TY1 - PHI*TX1)/(1.-PHI)
      T = (TMAX*TXX + TMIN)/(TXX + 1.)
   24 R = DABS(TH-T) + 100.00*DABS(RHS-R)
C     PRINT 104,TX1,TY1
C     PRINT 88,R,T
  104 FORMAT(5H TX1 ,F7.3,5H TY1 ,F7.3)
   88 FORMAT (' R=',F8.5,4H  T=,F9.3/)
      IF(R -.0001)4,4,3
    3 CONTINUE
    4 RETURN
      END
```

```
      SUBROUTINE LISWT
C USED WHEN SX < .003
      IMPLICIT REAL*8(B-H),REAL*8(O-Z)
      COMMON/WEFT/GXY(5), HTOT, PTOT
      COMMON/NOW/P,T,X(5),Y(5)
      COMMON/PNCH/HLIQ,HVAP
      DIMENSION CPTX(5,3)
      DATA CPTX(1,1),CPTX(1,2),CPTX(1,3)/532.33,-2.7153,-494.72/
      DATA CPTX(2,1),CPTX(2,2),CPTX(2,3)/720.22,-5.9729,-553.88/
      DATA CPTX(3,1),CPTX(3,2),CPTX(3,3)/484.39,-1.8473,-395.1/
      DATA CPTX(4,1),CPTX(4,2),CPTX(4,3)/188.2,2.8,3000./
      DATA CPTX(5,1),CPTX(5,2),CPTX(5,3)/10.,0.,0./
      PSO = 0.
      T = TVP(PTOT)
      DO17J=1,5
      X(J) = GXY(J)
   17 Y(J) = GXY(J)
      CALL ENTHAL
      P = (HTOT - HLIQ)/(HVAP - HLIQ)
      D = PTOT/ZCO2(T)
      PUN = 1.E-12
      SX = 0.
      SN = 0.
      SD = 1.
   15 DO1J=1,5
      W = CPTX(J,1) +CPTX(J,2)*PTOT
      X(J) = GXY(J)/(P*(W/D -1.)+ 1.)
      Y(J) = (GXY(J) - (1.-P)*X(J))/P
      PUN = PUN + W*X(J)
      SX = SX + X(J)
      SN = SN + X(J)*CPTX(J,1)
      SD = SD + X(J)*CPTX(J,2)
    1 CONTINUE
      IF(SX)4,30,4
    4 DO11JOB=1,14
      P = (PTOT -SN)/SD
      T = TVP(P)
      CALL ENTHAL
      P = (HTOT - HLIQ)/(HVAP - HLIQ)
      DELP = PUN/(1.+PSO)
      PSO = P*(SX/DELP -(1.-SX)*(1.7/T*(1.+(.85*DELP/T))))
      D = (PTOT-(1.-ZCO2(T))*PUN)/ZCO2(T)
      SIG = SX
      PUN = 1.E-12
      SX = 0.
      SN = 0.
      SD = 1.
      DO3J=1,5
      W = (PSO+1.)*(CPTX(J,1) +CPTX(J,2)*P +CPTX(J,3)*SIG)
      X(J) = GXY(J)/(P*(W/D -1.)+ 1.)
      Y(J) = (GXY(J) - (1.-P)*X(J))/P
      PUN = PUN + W*X(J)
      SX = SX + X(J)
      SN = SN + X(J)*(CPTX(J,1) + CPTX(J,3)*SIG)
      SD = SD + X(J)*CPTX(J,2)
    3 CONTINUE
```

```
      IF(DABS((SIG-SX)/SX)-2.D-7)30,30,11
   11 CONTINUE
   30 RETURN
      END

      SUBROUTINE TENTH
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      COMMON/WEET/GXY(5), HTOT, PTOT
      COMMON/NOW/P,T,X(5),Y(5)
      COMMON/ENTH/HLIQ,HVAP
C GIVES TEMP AT WHICH HLIQ = HTOT
      T2 = T
      T = .98*T
      DO1J=1,10
      H2 = HLIQ
      CALL ENTHAL
      DH = H2 - HLIQ
      IF(DABS(DH) - .001)4,4,6
    4 IF(DH)5,2,5
    5 DH = (.001/DH)*DABS(DH)
    6 DT = (HLIQ -HTOT)*(T2-T)/DH
      T2 = T
      T = T - DT
      IF (DABS(DT/T) - 1.D-7)2,2,1
    1 CONTINUE
   10 FORMAT ('  TENTH = ', I3)
    2 CONTINUE
      RETURN
      END

      FUNCTION VPCO2(TK)
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      T = TK/600.
      P = -8.17562621D00 +.26277422 8D02*T
     1 -.23768393D02*DLOG(T) -.86270978 5D01/T
      VPCO2 = DEXP(P)
      RETURN
      END
```

```
      SUBROUTINE ENTHAL
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      DIMENSION CL(4,3),CV(4,3)
      COMMON/NOW/P,T,X(5),Y(5)
      COMMON/ENTH/HLIQ,HVAP
      DATA CL(1,1),CL(1,2),CL(1,3)/20.75187,-.05896506,.0013514 24/
      DATA CL(2,1),CL(2,2),CL(2,3)/6.9126566,-1.2248637E-4,1.3079E-5/
      DATA CL(3,1),CL(3,2),CL(3,3)/6.97951,-2.141268E-4,-3.75439E-6/
      DATA CL(4,1),CL(4,2),CL(4,3)/6.95,-1.8E-4,0.0/
      DATA CV(1,1),CV(1,2),CV(1,3)/2.40058552,.021725119,-.0009370143/
      DATA CV(2,1),CV(2,2),CV(2,3)/6.9126566,-1.2248637E-4,1.3079E-5/
      DATA CV(3,1),CV(3,2),CV(3,3)/6.97951,-2.141268E-4,-3.75439E-6/
      DATA CV(4,1),CV(4,2),CV(4,3)/6.95,-1.8E-4,0.0/
      TT = T - 216.58
      HLIQ = 0.
      HVAP = 0.
      SUMX = 0.
      SUMY = 0.
      DO5I=1,5
      SUMX = SUMX + X(I)
    5 SUMY = SUMY + Y(I)
      XL = 1. - SUMX
      YV = 1. - SUMY
      DO1J=1,4
      SL = TT*(CL(J,1) + TT*(CL(J,2) + TT*CL(J,3)))
      SV = TT*(CV(J,1) + TT*(CV(J,2) + TT*CV(J,3)))
      IF (J-1)2,3,2
    3 SV = SV + 3661.357
    2 HLIQ = HLIQ + SL*XL
      HVAP = HVAP + SV*YV
      XL = X(J)
    1 YV = Y(J)
      HLIQ = HLIQ + 5.0*(X(4) + X(5))
      HVAP = HVAP + 5.0*(Y(4) + Y(5))
      IF(HVAP - HLIQ - 100.)6,7,7
    6 HVAP = HLIQ + 100.
    7 CONTINUE
      RETURN
      END

      FUNCTION ZCO2(T)
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      ZCO2 = 3.2120264 + T*(-2.9634967E-2 + T*(1.375700432E-4
     1-T*2.295992279E-7))
      RETURN
      END
```

```
      SUBROUTINE QUEND
C GIVES THE EQUILIBRIUM CONCENTRATIONS FROM PHASE SPLIT AND
C AVERAGE COMP, OXYGEN = 1, NITROGEN = 2, CO = 3, KR = 4 , XE = 5
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      COMMON/PAIN/LICK
      COMMON/WERT/GXY(5), HTOT, PTOT
      COMMON/NOW/P,T,X(5),Y(5)
      COMMON/ENTH/HLIQ,HVAP
      DIMENSION CPTX(5,3)
      DATA CPTX(1,1),CPTX(1,2),CPTX(1,3)/532.33,-2.7153,-494.72/
      DATA CPTX(2,1),CPTX(2,2),CPTX(2,3)/720.22,-5.9729,-553.88/
      DATA CPTX(3,1),CPTX(3,2),CPTX(3,3)/484.39,-1.8473,-395.1/
      DATA CPTX(4,1),CPTX(4,2),CPTX(4,3)/188.2,2.8,3000./
      DATA CPTX(5,1),CPTX(5,2),CPTX(5,3)/10.,0.,0./
      TX1 = 0.
      TY1 = 0.
      SX = 0.
      DO17J=1,5
      X(J) = GXY(J)
      Y(J) = GXY(J)
   17 SX = SX + GXY(J)
      SG = SX
      IF(SX-.003)21,21,22
   21 CALL LTLEND
      RETURN
   22 SN = 0.
      SD = 1.
      DO13J=1,5
      SN = SN +(CPTX(J,1) +CPTX(J,3)*SX)*GXY(J)
   13 SD = SD +CPTX(J,2)*GXY(J)
      P = (PTOT - SN)/SD
      TMIN = TVP(P)
      IF(217.-TMIN)32,32,33
   33 TMIN = 217.
   32 TMAX = TVP(PTOT*(1.-SX))
C     PRINT 103,TMIN,TMAX
  103 FORMAT (12H TEMP LIMITS,2F8.2)
      IF((T-TMIN)*(TMAX-T))28,28,20
   28 T = .4*TMIN + .6*TMAX
   20 SX = SX*(1.-P)*(1.-P)
      DO3NTRY =1,15
      RT = 1.0
      Z = ZCO2(T)
      P = VPCO2(T)
      REXP = DEXP(1.7*(PTOT-P)/T)
      DO11NGO=1,15
      IF(SX-SG)27,26,26
   26 SX = SG
   27 SIG = SX
      CT = (1.-SIG)*REXP
      SX = 0.
      SY = 0.
      D = PTOT + P*CT*(1.-Z)/Z
      IF(SIG*(PTOT-P) -1.D-8)12,14,14
   12 PSO = 0.
      GO TO 15
   14 PSO = P*(1.-CT)/(PTOT-P)
   15 DO1J=1,5
      W = (PSO+1.)*(CPTX(J,1) +CPTX(J,2)*P +CPTX(J,3)*SIG)
      X(J) = GXY(J)/(P*(W/D -1.)+ 1.)
      Y(J) = (GXY(J) - (1.-P)*X(J))/P
      SX = SX + X(J)
      SY = SY + Y(J)
    1 CONTINUE
      IF(DABS((SIG-SX)/SX) -1.D-7)18,11,11
   11 CONTINUE
      LICK = LICK + 1
C     PRINT 101
  101 FORMAT (10H NO X FIT./)
C     PRINT 103,TMIN,TMAX
      TX1 = 0.
      TY1 = 0.
      T = (TMAX + TMIN)/2.
      GOTO3
   18 SN = 0.
      SD = 1.
      DO2J=1,5
      SN = SN + (CPTX(J,1) + CPTX(J,3)*SX)*X(J)
    2 SD = SD + CPTX(J,2)*X(J)
C     PRINT 100,SN,SD
  100 FORMAT (4H SN=,F6.3,4H SD=,F5.3)
      P = (PTOT - SN)/SD
      TX2 = TX1
      TH = T
      TX1 = (T - TMIN)/(TMAX -T)
      TY2 = TY1
      T = TVP(P)
      TM = TMAX - T
      IF(TM)40,40,41
   40 TM = 0.1*(TMAX-TH)
      T = TMAX - TM
   41 TXX = T - TMIN
      IF(TXX)42,42,43
   42 TXX = 0.1*(TH-TMIN)
   43 TY1 = TXX/TM
      PHI = (TX2-TX1)*(1.-SY*SY)
      IF(PHI)46,44,46
   46 PHI = (TY2-TY1)/PHI
      IF(PHI*(50.+PHI))45,45,44
   44 PHI = -20.*SY
   45 TXX = (TY1 - PHI*TX1)/(1.-PHI)
      T = (TMAX*TXX + TMIN)/(TXX + 1.)
   24 E = DABS(TH-T)
C     PRINT 104,TX1,TY1
C     PRINT 88,P,T
  104 FORMAT(5H TX1 ,F7.3,5H TY1 ,F7.3)
   88 FORMAT (' P=',F8.5,4H  T=,F9.3/)
      IF(E -.00001)4,4,3
    3 CONTINUE
    4 CALL ENTHAL
   30 RETURN
      END
```

```
      SUBROUTINE LTLEND
C USED WHEN SX .LT. .003
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      COMMON/WRET/GXY(5), HTOT, PTOT
      COMMON/NOW/P,T,X(5),Y(5)
      COMMON/ENTH/HLIQ,HVAP
      DIMENSION CPTX(5,3)
      DATA CPTX(1,1),CPTX(1,2),CPTX(1,3)/532.33,-2.7153,-494.72/
      DATA CPTX(2,1),CPTX(2,2),CPTX(2,3)/720.22,-5.9729,-553.88/
      DATA CPTX(3,1),CPTX(3,2),CPTX(3,3)/484.39,-3.8473,-395.1/
      DATA CPTX(4,1),CPTX(4,2),CPTX(4,3)/188.2,2.8,3000./
      DATA CPTX(5,1),CPTX(5,2),CPTX(5,3)/10.,0.,0./
      PSO = 0.
      T = TVP(PTOT)
      D = PTOT/ZCO2(T)
      PUN = 1.E-12
      SX = 0.
      SN = 0.
      SD = 1.
   15 DO1J=1,5
      W = CPTX(J,1) +CPTX(J,2)*PTOT
      X(J) = GXY(J)/(P*(W/D -1.)+ 1.)
      Y(J) = (GXY(J) - (1.-P)*X(J)) /P
      PUN = PUN + W*X(J)
      SX = SX + X(J)
      SN = SN + X(J)*CPTX(J,1)
      SD = SD + X(J)*CPTX(J,2)
    1 CONTINUE
      IF(SX)4,30,4
    4 DO11JCF=1,14
      P = (PTOT -SN)/SD
      T = TVP(P)
      DELP = PUN/(1.+PSO)
      PSO = P*(SX/DELP -(1.-SX)*(1.7/T*(1.+(.85*DELP/T))))
      D = (PTOT-(1.-ZCO2(T))*PUN)/ZCO2(T)
      SIG = SX
      PUN = 1.E-12
      SX = 0.
      SN = 0.
      SD = 1.
      DO3J=1,5
      W = (PSO+1.)*(CPTX(J,1) +CPTX(J,2)*P +CPTX(J,3)*SIG)
      X(J) = GXY(J)/(P*(W/D -1.)+ 1.)
      Y(J) = (GXY(J) - (1.-P)*X(J)) /P
      PUN = PUN + W*X(J)
      SX = SX + X(J)
      SN = SN + X(J)*(CPTX(J,1) + CPTX(J,3)*SIG)
      SD = SD + X(J)*CPTX(J,2)
    3 CONTINUE
      IF(DABS((SIG-SX)/SX)-2.D-7)29,29,11
   11 CONTINUE
   29 CALL ENTHAL
   30 RETURN
      END
```

```
      FUNCTION TVP(P)
      IMPLICIT REAL*8(A-H),REAL*8(O-Z)
      IF(P)5,5,6
    5 TVP = 216.01
      GOTO30
    6 TVP = (-1990.927)/(DLOG(P) - 10.8271)
      T = TVP/600.
      PX = P
      DO1J=1,10
      PD = -8.17562621D00 +.262774228D02*T
     1 -.23768393D02*DLOG(T) -.862709785D01/T
      VP = DEXP(PD)
      D1 =.262774228D02 -.23768393D02/T +.862709785D01/T/T
      DVP = VP*D1
      DP = (PX-VP)/DVP
      T = T + DP
      IF (DABS(DP) - 1.D-8)2,2,1
    1 CONTINUE
    2 TVP = 600.*T
   30 RETURN
    3 FORMAT ('  TVP ITERATIONS = ',I2 )
    4 FORMAT ('  D = ',F14.6,'  TVP = ',F10.3 )
      END
```

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