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Krypton Absorption in Liquid CO_2 (KALC): Effects of the Minor Components N_2, CO, and Xe

T. M. Gilliam
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T. M. Gilliam, V. L. Fowler, and D. J. Inman

ABSTRACT

Results are presented for the fourth major campaign for quantifying krypton removal from simulated High-Temperature Gas-Cooled Reactor reprocessing off-gas by the Krypton Absorption in Liquid CO_2 (KALC) process. This process utilizes the high solubility of krypton in liquid CO_2.

Mass transfer experiments for the absorption, fractionation, and stripping operations of the KALC process indicate that the addition of N_2 and CO do not alter the mass transfer characteristics exhibited by O_2 and krypton in the basic CO_2-O_2-Kr system. Decontamination factors for xenon in the absorber and stripper were several orders of magnitude less than those for krypton under similar conditions. Indications are that the fate of xenon is controlled by the heat input to the stripper reboiler. Experiments on the solubility of O_2 and CO indicate that CO is more soluble than O_2 at temperatures below -21°C.

1. INTRODUCTION

Development work is being performed on the Krypton Absorption in Liquid CO_2 (KALC) process as part of the High-Temperature Gas-Cooled Reactor (HTGR) Fuel Recycle Program.[1,2] The KALC process is being developed as a means of removing and concentrating ^{85}Kr from the off-gas generated during the reprocessing of HTGR fuel elements. An estimated composition of the generated gas stream after filtration is given in Table 1.[3] For a plant processing 1 metric ton/day, the numbers

Table 1. Estimated burner off-gas composition for head-end reprocessing of one Fort St. Vrain fuel block (6-year exposure, 150-day cooling)

Major component	kg	std m^3/min	Mole fraction
CO_2	555	282	0.897
O_2	34	24	0.075
N_2	11	9	0.028
Contaminant	ppm (total)	ppm (radioactive)	Ci
Krypton	16	1.2	572.0
Tritium	2.4×10^{-2}	2.4×10^{-2}	9.8
Iodine	3	9.2×10^{-7}	0.2
Xenon	61	0	0

given would need to be multiplied by 100 because 100 blocks would be processed per day. After being discharged from the reactor, the spent fuel is "cooled" for 150 days to allow short-lived fission products to decay, and the major radioactive component remaining in the off-gas is then ^{85}Kr. Other components may be present in the off-gas throughout the reprocessing train; eventually, however, the final cleanup problem will involve removing part-per-million concentrations of Kr from the off-gas, which will consist mainly of CO_2, but will also contain (5 to 15%) lighter gases such as O_2, CO, and N_2, and part-per-million concentrations of Xe.

In previous campaigns (II and III),[4,5] experiments were conducted to quantify the mass transfer characteristics of the basic CO_2-O_2-Kr KALC system. This report presents the study of the effects of N_2, CO, and Xe on the mass transfer of Kr and O_2. The experiments were directed toward answering two questions: (1) does the presence of N_2, CO, and/or Xe alter the mass transfer data for the CO_2-O_2-Kr system in either the absorption, fractionation, or stripping steps; and (2) are the relative solubilities of the various components, as defined by the Mullins computer model,[6] essentially correct?

The experiments described will hereafter be referred to as Campign IV (i.e., experiments performed between August 1, 1976 and December 31, 1977).

2. MASS TRANSFER STUDIES

The Experimental Engineering Section Off-Gas Decontamination Facility (EES-ODF; see Fig. 1) used in these experiments has been described previously.[7,8] Therefore, discussion of the facility and its operation will

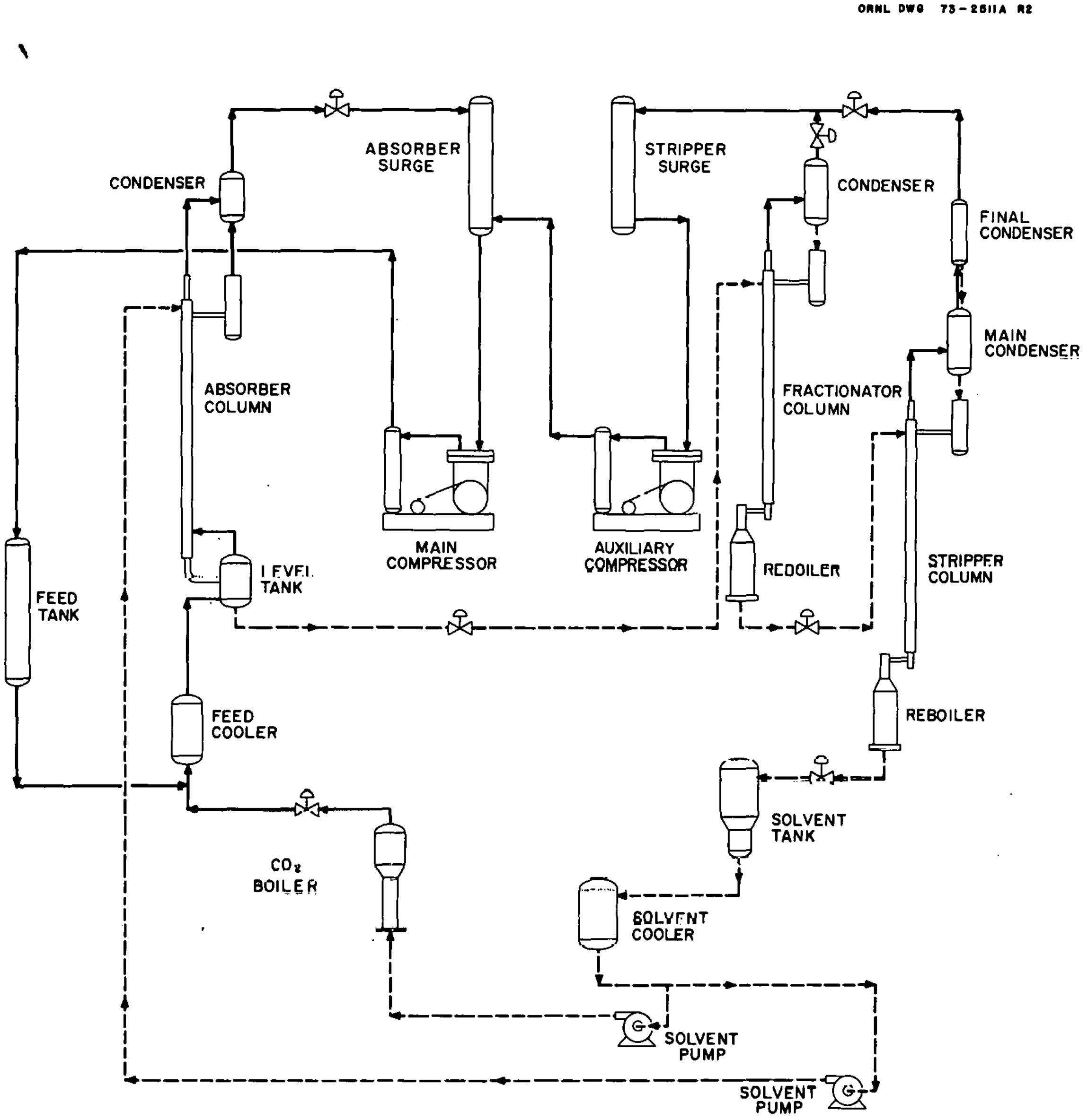

Fig. 1. Schematic diagram of the Experimental Engineering Section Off-Gas Decontamination Facility.

be limited to those areas necessary for clarification of the analysis. The addition of the third column at the beginning of this campaign[8] gave the facility greater flexibility, and both two- and three-column experiments were made in Campaign IV.

Experiments were performed to study the absorption, fractionation, and stripping operations with four system feeds: (1) CO_2-O_2-Kr, (2) CO_2-O_2-N_2-Kr, (3) CO_2-O_2-CO-N_2-Kr, and (4) CO_2-O_2-Xe. The operation and analysis of mass transfer experiments were also described previously[5,8] and will be discussed here only as an aid to the overall understanding of the analysis.

During the operations, the system feed gas is compressed to a pressure greater than the absorber operating pressure and is then routed to the bottom of the absorber. As the gas travels through the absorber, it is contacted countercurrently with a stream of liquid CO_2. The liquid scrub absorbs essentially all of the krypton, as well as significant portions of the O_2, N_2, CO, and Xe. The solute-laden liquid is routed to the top of the fractionator by an operating pressure differential (nominally 2 atm). The dissolved light gases are distilled in the fractionator and exit via the fractionator off-gas. The liquid CO_2 leaving the fractionator bottom (now containing only Kr and Xe) is sent to the top of the stripper, again by a pressure differential (nominally 2 atm). The krypton is then distilled and exits via the stripper off-gas. The liquid leaving the stripper bottom is recycled to the absorber to act as liquid scrub. To eliminate the need for large amounts of feed for these experiments, the off-gas streams from all three columns are routed to a compressor where they are combined to become the system feed. Thus the facility operates in total recycle.

In experiments using only two columns, the fractionator is valved out of the system, and the absorber liquid bottoms are routed to the top of the stripper column. The stripper can be operated as a stripper or fractionator by adjusting the boilup rate. As with three-column operations, the facility operates in total recycle.

As with all mass transfer operations, some variables are designated as independent, and others are designated as dependent. Since the analysis was performed during "steady-state" (i.e., no appreciable change in monitored variables for at least 2 hr), the choice of independent-dependent operating variables becomes academic. Therefore, the independent variables were chosen on the basis of our ability to maintain them at a desired value.

The variable of primary concern in the operation of the columns was the liquid-to-vapor ratio, the liquid rate being determined by the pump rate. The vapor rate in the absorber column was fixed by controlling the feed-gas rate. The off-gas rate was manually set to complement the feed-gas rate (nominally 80% of feed rate). The absorber pressure and temperature were then allowed to seek their own levels. The fractionator and stripper column pressures were set by controlling the amount of refrigerant to each column condenser. The fractionator and stripper temperatures were allowed to seek their own level. The reboiler heat input fixed the vapor rate.

Once these independent variables were set, sufficient time was allowed for the entire system to reach steady state. Various temperatures, pressures, liquid holdups, stream concentrations, and flow rates were monitored during the experiments and were used to indicate when steady state had been attained. As the system approached steady state operation, the various controllers were fine tuned to alleviate any controller-induced oscillations. The wide variety of conditions studied caused this to be a time-consuming operation to the extent that controller fine tuning became a major part of each experiment.

3. MASS TRANSFER DATA

Experiments using equipment of this complexity are, by nature, both difficult to control and to analyze, and generally necessitate certain engineering assumptions in the analysis of the data collected. Although many of these assumptions are based on sound engineering principles, many are based on experience gained from operating the facility. The data analysis and the assumptions associated with it are discussed in Appendix A. It should be noted that the data analysis in this report differs from that of previous campaigns in that all assumptions of component solubility (i.e., equilibrium values) were avoided, because one of the campaign objectives was to determine what effect (if any) the presence of minor components had on the equilibrium constants of O_2 and Kr.

The data were collected from experiments using both two and three columns. Whenever possible, a three-column system was used. Some experiments that could have been performed on a three-column system (e.g., absorption experiments) were performed instead on a two-column system (i.e., absorber-stripper) because installation and general shakedown of the third column (i.e., fractionator) had not yet been completed. In the case of fractionation studies, a two-column system was necessary due to the use of a new, random packing (i.e., PRO-PAK)[9] in the fractionator column. Previous experiments[8] indicated that the mass transfer characteristics of PRO-PAK packing differ from those of the Goodloe packing[10] (used in the other two columns). This would tend to limit the confidence of a comparison between fractionation studies using a PRO-PAK column with a CO_2-O_2-Kr-minor component(s) system and a Goodloe column with a CO_2-O_2-Kr system. Also, the fractionator and stripper were of different lengths (2.71 m vs 3.52 m), and the primary variable used in comparing the other systems (i.e., decontamination factor) is dependent on column height.

3.1 Absorber Studies with N_2 and CO Present

Experiments were performed to study the absorption of Kr (in the absorber column) from the feed gas using three system feeds: (1) CO_2-O_2-Kr, (2) CO_2-O_2-N_2-Kr, and (3) CO_2-O_2-N_2-CO-Kr. In all cases, tracer levels of ^{85}Kr were used. Since the purpose of the absorber is to absorb Kr from the incoming system feed gas, all mass transfer related data are based on Kr concentrations. The absorber overall decontamination factor (DF_o) is defined as:

$$DF_o = \frac{\text{amount of Kr in the system feed-gas}}{\text{amount of Kr in absorber off-gas}} \qquad (1)$$

The liquid-to-vapor ratio, L/V, is defined as:

$$L/V = \text{liquid scrub rate/system feed-gas rate, mole/mole.} \qquad (2)$$

The absorber liquid-to-vapor ratio is an "operating ratio" and does not necessarily equal the actual column liquid-to-vapor ratio. The column absorption factor, A, is then defined as

$$A = L/VK, \qquad (3)$$

where K is the krypton equilibrium constant* determined at column pressure and average column temperature.

The mass transfer data are summarized in Table 2. A plot of overall decontamination factor vs absorption factor for the various systems studied is shown in Fig. 2. The data for the CO_2-O_2-N_2-Kr and CO_2-O_2-N_2-CO-Kr systems fall well within the data scatter for the CO_2-O_2-Kr system. This indicates that the effect of CO and N_2 on the mass transfer of Kr in the absorption step is negligible.

3.2 Fractionation Studies With N_2 and CO Present

Experiments were performed to study the fractionation of light gases (in the 3-in. stripper column) using two system feeds: CO_2-O_2-N_2-Kr and CO_2-O_2-N_2-CO-Kr. Again, tracer levels of ^{85}Kr were used. The resulting data were compared to previously reported data for the CO_2-O_2-Kr system. The main purpose of the fractionator is to fractionate the light gases from the liquid CO_2 containing Kr. Consequently, all mass transfer related data are based on oxygen (the least volatile of

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*All equilibrium constants are determined using the Glass computer model.[11]

Table 2. Summary of absorber mass transfer data

Experiment No.	Pump rate (cm^3/s)	Column pressure (MPa)	Average column temperature (K)	L/V (mole/mole)	L/VK	Overall Kr decontamination factor
C4-35[a]	34.9	2.42	257.2	8.05	1.04	3600
C4-41[a]	34.9	2.41	256.9	6.93	0.89	280
C4-48[a]	34.9	2.36	256.5	6.11	0.76	60
C4-87[a]	34.9	2.36	255.4	7.68	0.99	290
C4-304[b]	26.2	2.52	254.3	6.67	0.89	80
C4-310[b]	26.2	2.42	254.6	6.74	0.90	90
C4-1246[b]	39.3	2.32	254.3	8.05	0.99	70
C4-1264[c]	39.3	2.26	253.4	8.26	0.98	2700
C4-1272[c]	39.3	2.32	253.8	8.04	0.98	3100
C4-1280[c]	39.3	2.33	253.7	7.59	0.94	1000
C4-1290[c]	39.3	2.30	253.3	7.31	0.89	260
C4-1300[c]	39.3	2.25	252.9	7.36	0.88	130
C4-1310[c]	39.3	2.27	253.2	7.82	0.93	180
C4-1318[c]	39.3	2.31	253.4	8.47	1.03	1800
C4-1326[c]	39.3	2.28	253.7	8.81	1.06	1100

[a] Experiment with a CO_2-O_2-N_2-Kr system.

[b] Experiment with a CO_2-O_2-CO-N_2-Kr system.

[c] Experiment with a CO_2-O_2-Kr system.

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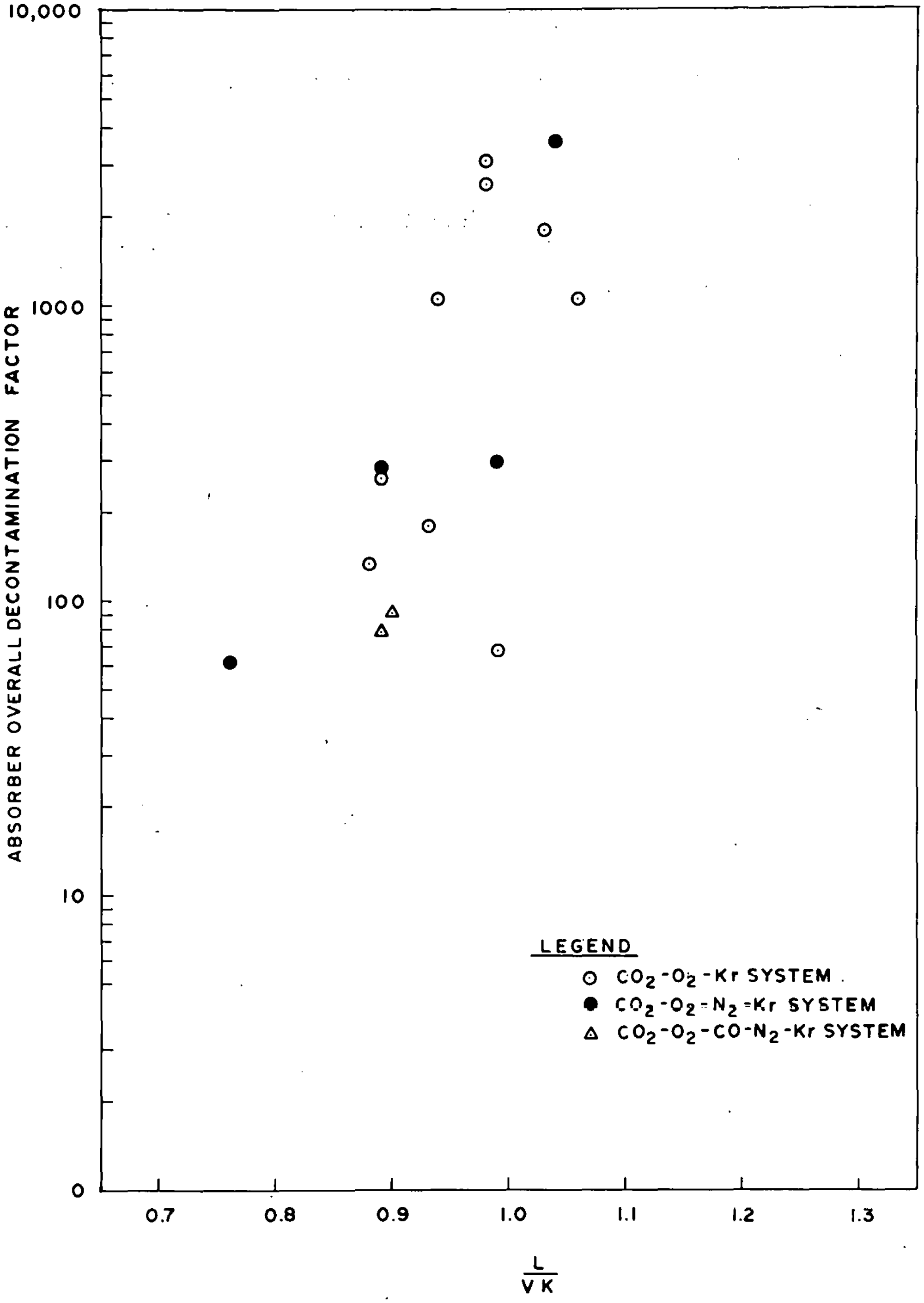

Fig. 2. Relationship between absorber DF_o (based on Kr) and absorption factor for various system feeds.

the light gases). Table 3 presents a summary of the fractionator data and similar data from Campaign III.[5]

The column decontamination factor (DF_c) shown in the table is defined as

$$DF_c = \frac{\text{amount of oxygen in the liquid entering the column packing}}{\text{amount of oxygen in the liquid leaving the column packing}} ; \quad (4)$$

the column stripping factor (A^{-1}) is defined as

$$A^{-1} = VK/L , \quad (5)$$

where

V = column vapor rate, moles/min,

K = oxygen equilibrium constant based on column pressure and average temperature, and

L = column liquid rate, moles/min.

Figure 3 presents a plot of fractionator decontamination factors vs stripping factors for the various systems studied. The data for the CO_2-O_2-N_2-Kr and CO_2-O_2-CO-N_2-Kr systems fall within the data scatter for the CO_2-O_2-Kr system, indicating that the presence of CO and N_2 has a negligible effect on the mass transfer of O_2 in the fractionation step.

3.3 Stripping Studies with N_2 and CO Present

Experiments were performed to study the stripping of Kr (again in the 3-in. stripper column) using CO_2-O_2-N_2-Kr and CO_2-O_2-N_2-CO-Kr system feeds. Again, tracer levels of ^{85}Kr were used. A comparison

Table 3. Summary of fractionator experiments

Experiment No.	Pump rate (cm^3/s)	Reboiler heat load (kW)	Column pressure (MPa)	Average column temperature (K)	VK/L	O_2 decontamination factor
C4-124[a]	34.9	0.73	2.03	254.0	1.20	
C4-130[a]	34.9	0.64	2.03	254.0	1.05	30
C4-135[a]	34.9	0.66	2.03	254.3	1.09	210
C4-139[a]	34.9	0.66	2.03	254.0	1.07	80
C4-143[a]	34.9	0.74	2.03	253.4	1.20	260
C4-148[a]	34.9	0.71	2.03	254.2	1.16	290
C4-316[b]	26.2	0.52	2.03	254.1	1.12	60
C4-323[b]	26.2	0.54	2.03	253.9	1.15	260
C4-329[b]	26.2	0.55	2.03	254.0	1.18	130
1206[c]	26.2	0.97	2.03	253.7	1.39	90
1222[c]	39.3	0.80	2.03	253.3	1.16	30
1230[c]	39.3	0.87	2.03	253.3	1.26	50
1246[c]	39.3	0.93	2.03	253.7	1.34	200
1254[c]	39.3	0.96	2.03	253.3	1.39	1100
1270[c]	39.3	0.82	2.03	253.0	1.20	200
1286[c]	39.3	0.87	2.03	253.6	1.27	570
1302[c]	34.9	0.80	2.03	253.5	1.30	1200
1310[c]	34.9	0.76	2.03	253.4	1.23	350
1318[c]	34.9	0.71	2.03	253.4	1.18	80
1368[c]	39.3	0.82	2.03	253.1	1.20	80
1386[c]	39.3	0.90	2.03	253.0	1.32	480
1396[c]	43.7	0.99	2.03	253.4	1.30	830
1404[c]	41.5	0.96	2.03	253.9	1.32	530
1425[c]	41.5	0.89	2.03	253.2	1.23	300
1433[c]	41.5	0.86	2.03	253.1	1.19	90
1442[c]	41.5	0.81	2.03	252.7	1.12	20
1450[c]	41.5	0.97	2.03	249.8	1.49	4000
1460[c]	41.5	0.92	2.03	250.8	1.40	1050
1484[c]	39.3	0.87	2.03	252.5	1.28	120
1485[c]	39.3	0.86	2.03	252.3	1.26	80
1494[c]	39.3	0.87	2.03	253.2	1.27	170
1501[c]	39.3	0.86	2.03	252.8	1.25	120

[a] Experiments with a CO_2-O_2-N_2-Kr system.

[b] Experiments with a CO_2-O_2-N_2-CO-Kr system.

[c] Campaign III data with a CO_2-O_2-Kr system (from ref. 5).

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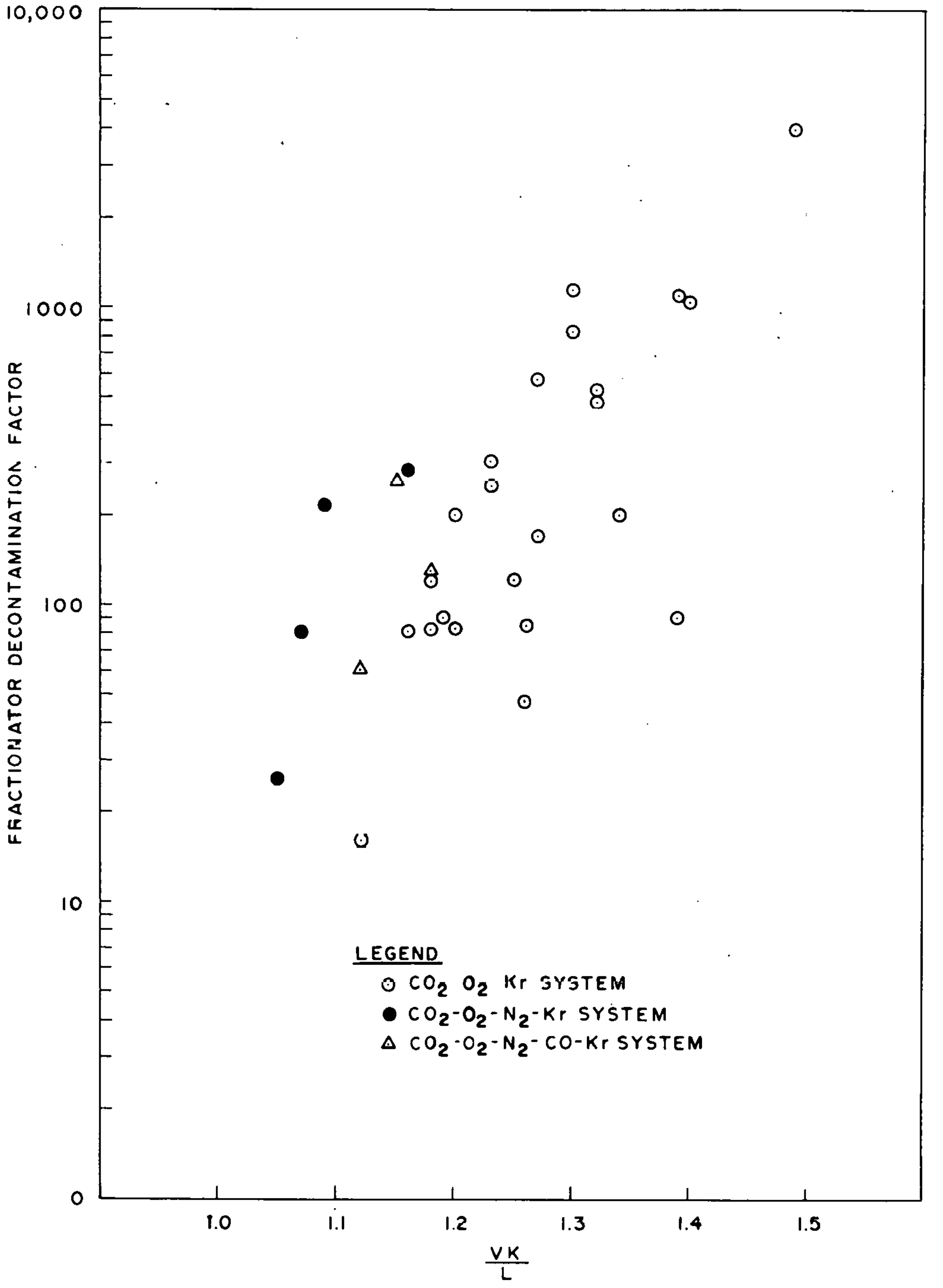

Fig. 3. Comparison of fractionator decontamination factors (based on oxygen) for various feeds.

was made between the resulting data and previously reported data for the CO_2-O_2-Kr system.

The primary purpose of the stripper is to strip Kr from the liquid CO_2. Consequently, all mass transfer related variables are based on Kr.

As with fractionation experiments, the stripping factor (A^{-1}) is defined

$$A^{-1} = \frac{VK}{L} \; ; \qquad (5)$$

but

K = Kr equilibrium constant based on column pressure and average column temperature.

The DF_o is defined

$$DF_o = \frac{\text{Kr in stripper liquid feed}}{\text{Kr in liquid leaving stripper reboiler}} \qquad (6)$$

A summary of the stripper mass transfer data and data from Campaign III[5] are shown in Table 4. Figure 4 compares the data from the CO_2-O_2-Kr system with that of the CO_2-O_2-N_2-Kr and CO_2-O_2-CO-N_2-Kr systems and shows that the data obtained with N_2 or CO present fall within the scatter of the CO_2-O_2-Kr data.

3.4 Xenon Studies

Bottled CO_2, used as the source of CO_2 in all studies, was found to contain part-per-million concentrations of Xe. Thus all data previously obtained are in the presence of Xe, which implies that the effect of Xe on the mass transfer of O_2 and Kr has already been accounted for. Consequently, the fate of Xe was studied in a three-column system

Table 4. Summary of stripper data

Experiment No.	Pump rate (cm^3/s)	Reboiler heat load (kW)	Column pressure (MPa)	Average column temperature (K)	VK/L	Overall Kr decontamination factor
C4-100[a]	34.9	1.83	1.85	251.0	1.50	90
C4-106[a]	34.9	1.49	1.84	250.7	1.26	170
C4-112[a]	34.9	1.46	1.84	250.9	1.24	300
C4-118[a]	34.9	1.16	1.84	251.0	1.01	20
C4-304[b]	26.2	1.02	1.83	250.8	1.15	120
C4-310[b]	26.2	1.10	1.83	250.5	1.25	210
752[c]	34.9	1.51	2.02	254.7	1.19	40
770[c]	34.9	1.40	2.03	254.8	1.11	20
790[c]	39.3	1.91	2.02	253.9	1.32	130
820[c]	43.7	2.12	2.03	254.0	1.29	100
832[c]	43.7	1.88	2.03	254.2	1.16	30
874[c]	39.9	1.62	1.83	250.3	1.12	20
926[c]	39.3	1.47	1.83	250.7	1.12	20
966[c]	34.9	1.30	1.83	250.1	1.12	40
978[c]	34.9	1.49	1.83	250.1	1.27	200
1056[c]	39.3	1.74	2.03	254.0	1.21	50
1130[c]	39.3	1.48	1.72	248.5	1.21	120
1148[c]	39.3	1.38	1.63	246.6	1.18	110
1158[c]	43.7	1.56	1.63	245.1	1.19	60
1168[c]	43.7	1.38	1.63	246.7	1.07	10

[a] Experiments with a CO_2-O_2-N_2-Kr system.

[b] Experiments with a CO_2-O_2-N_2-CO-Kr system.

[c] Campaign III data with a CO_2-O_2-Kr system (from ref. 5).

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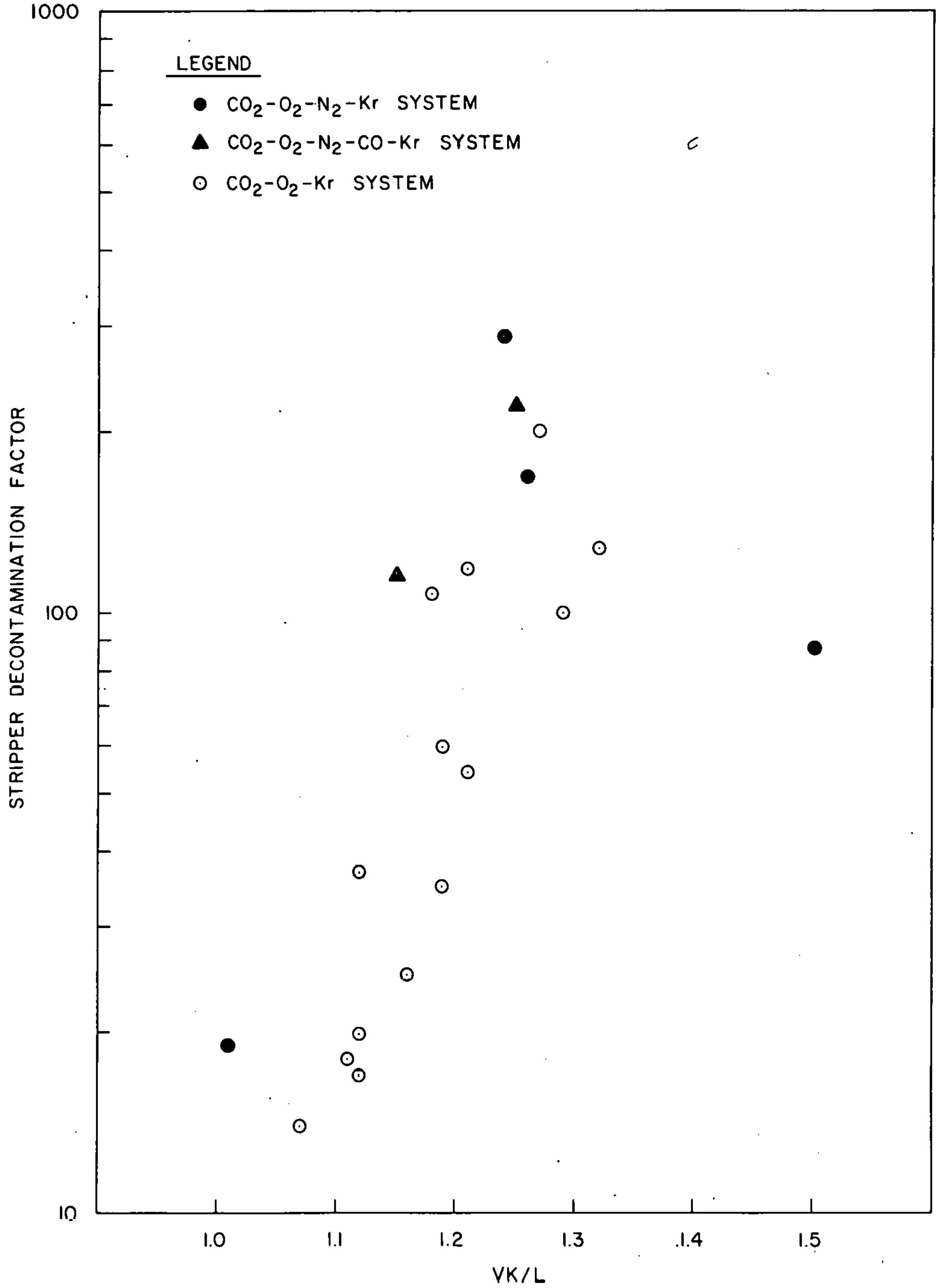

Fig. 4. Comparison between stripper decontamination factors (based on Kr) as a function of stripping factor for various feeds.

using a CO_2-O_2 system with tracer levels of ^{133}Xe. This enabled the existing beta cells to be used in determining Xe concentrations.[12]

During normal KALC operations, Xe is pinched in the fractionator, and the mass transfer of interest takes place in the absorption and stripping steps. The experiments were thus directed toward the operation of the absorber and stripper columns and a comparison of the results with the mass transfer data for Kr under similar operating conditions. However, the decontamination factor is based on Xe. For absorption studies, the analysis is identical to that described in Sect. 3.1, except that Xe concentrations and equilibrium constants were used. Table 5 presents a comparison of data taken for a CO_2-O_2-Xe and a CO_2-O_2-Kr system under similar operating conditions. Figure 5, showing the relationship between the absorber DF_o and the absorption factor, reveals an important point. Over the normal KALC operation range, the absorber decontamination factor for krypton is several orders of magnitude greater than that for Xe, because the stripper liquid-to-vapor ratio (L/V) is >4.0 under normal operating conditions. The L/V is, of course, too high to effectively strip Xe from the liquid scrub because at L/Vs >4.0 the Xe stripping factor is <1.0 (the Xe equilibrium constant, K, is a nominal 3.5). The liquid scrub to the absorber contains substantial amounts of Xe which causes Xe to pinch in the top of the absorber (i.e., the gas and liquid are essentially in equilibrium with respect to Xe). This makes the driving force for Xe absorption essentially zero at the top of the column, resulting in very little absorption of Xe from the incoming feed (thus, the low values of DF_o). In experiments C4-869 and C4-881, the stripper L/V

Table 5. Summary of absorber Xe studies

Experiment No.	Pump rate (cm^3/s)	Column pressure (MPa)	Average column temperature (K)	L/V (mole/mole)	L/VK	Overall decontamination factor
C4-838[a]	39.3	2.43	258.3	13.8	4.55	6
C4-845[a]	39.3	2.45	258.5	13.9	4.60	7
C4-852[a]	39.3	2.53	259.1	13.4	4.49	7
C4-859[a]	39.3	2.54	259.5	13.4	4.52	6
C4-869[a]	39.3	2.50	259.2	12.6	4.23	1970
C4-881[a]	39.3	2.57	260.3	11.8	4.05	30
C4-1246[b]	39.3	2.32	254.3	8.1	0.98	70
C4-1264[b]	39.3	2.26	253.4	8.3	0.98	2690
C4-1272[b]	39.3	2.32	253.8	8.1	0.98	3070
C4-1280[b]	39.3	2.33	253.7	7.6	0.94	1030
C4-1290[b]	39.3	2.30	253.3	7.3	0.89	260
C4-1300[b]	39.3	2.25	252.9	7.3	0.88	130
C4-1310[b]	39.3	2.27	253.2	7.8	0.93	180
C4-1318[b]	39.3	2.31	253.4	8.5	1.03	1790
C4-1326[b]	39.3	2.28	253.7	8.8	1.06	1080

[a] Mass transfer data based on Xe.

[b] Mass transfer data based on Kr.

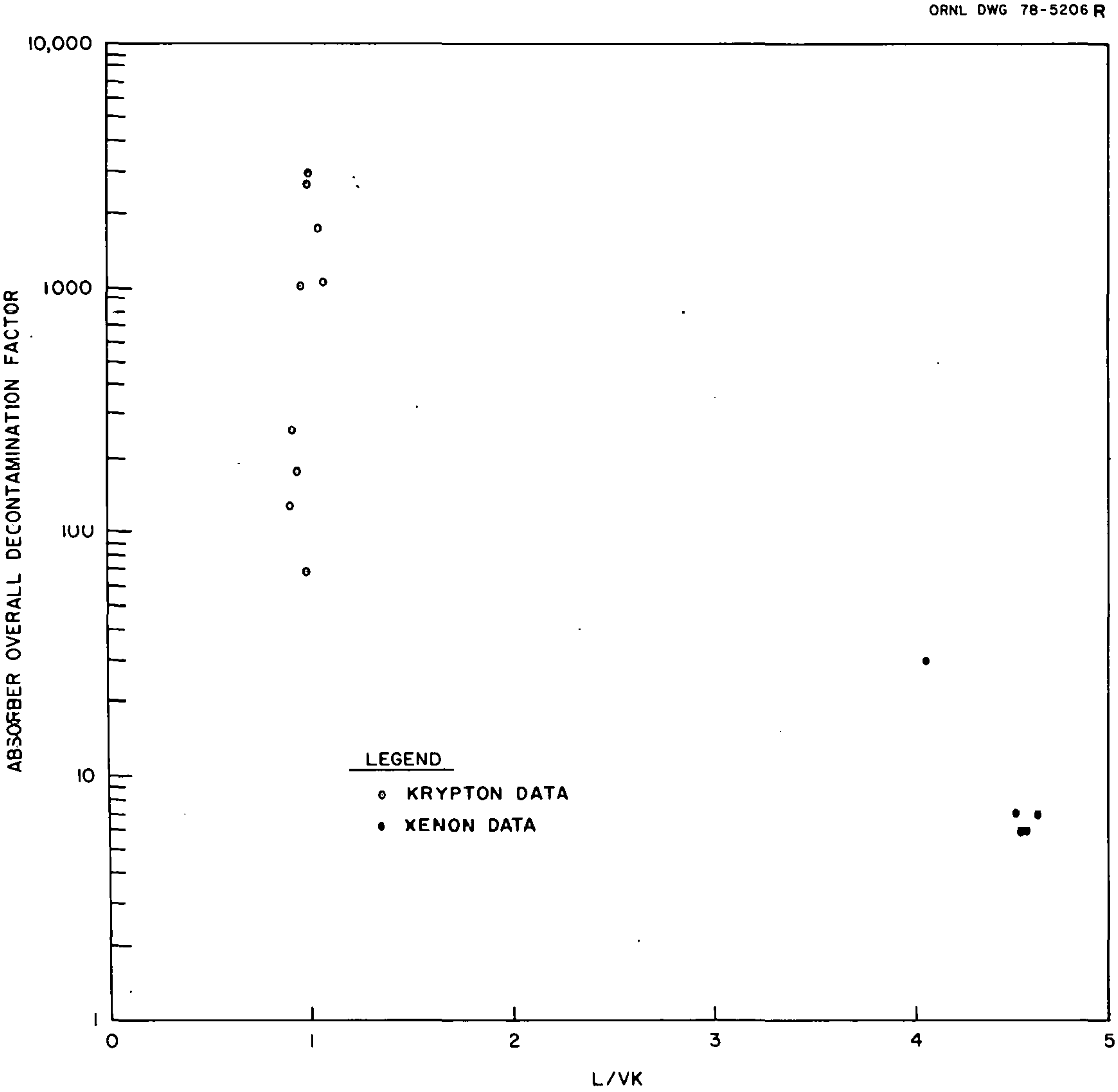

Fig. 5. Comparison between absorber overall DF for Kr and Xe as a function of absorption factor (based on Kr).

was set low enough to strip Xe from the liquid scrub which produced the high absorber DF_o values. For the absorber to absorb all (>99.9%) of the Xe in the process feed, the stripper boilup will have to be increased to produce a Xe stripping factor >1.0 (i.e., L/V <4.0). The Xe in the KALC feed will be nonradioactive and, unless it is desirable to recover Xe for its commercial value, it will not be necessary to operate the stripper at such low L/Vs (i.e., high-boilup rate).

Table 6 presents a comparison between stripping data for a CO_2-O_2-Xe and a CO_2-O_2-Kr system.[5] Again, the mass transfer variables are determined as defined in Sect. 3.3. The only exceptions are that the column decontamination factor (DF_c) is defined as

$$DF_c = \frac{\text{mass of Xe in the liquid entering the stripper packing}}{\text{mass of Xe in the liquid leaving the stripper packing}}, \quad (7)$$

and the equilibrium constant (K) used in the stripping factor refers to Xe.

Figure 6, showing a comparison of decontamination factor vs stripping factor for the two systems, illustrates behavior similar to that seen in the absorber. Under normal operating conditions (L/V >4.0), the decontamination factor for Kr is several orders of magnitude greater than that for Xe. Again, as the stripper L/V becomes less than the Xe equilibrium constant (Run Nos. C4-869 and C4-881), Xe begins to be stripped from the solvent, which indicates that the stripper can be used to strip both Kr and Xe from the liquid or to fractionate Kr from Xe.

Table 6. Summary of stripper xenon studies

Experiment No.	Pump rate (cm^3/s)	Reboiler heat load (kW)	Column pressure (MPa)	Average column temperature (K)	VK/L	Column decontamination factor
C4-838[a]	39.3	1.94	1.83	250.7	0.49	1
C4-845[a]	39.3	3.00	1.83	250.7	0.70	1
C4-852[a]	39.3	3.57	1.83	250.5	0.80	1
C4-859[a]	39.3	4.02	1.83	250.6	0.88	1
C4-869[a]	39.3	6.96	1.83	250.3	1.29	200
C4-881[a]	39.3	4.63	1.83	250.6	0.98	2
752[b]	34.9	1.51	2.02	254.5	1.19	100
770[b]	34.9	1.40	2.03	254.6	1.11	60
790[b]	39.3	1.92	2.02	253.9	1.32	470
820[b]	43.7	2.12	2.03	254.0	1.29	510
832[b]	43.7	1.88	2.03	254.2	1.16	120
874[b]	43.7	1.62	1.83	250.3	1.12	70
926[b]	39.3	1.47	1.83	250.7	1.12	70
966[b]	34.9	1.30	1.83	250.1	1.12	170
1056[b]	39.3	1.74	2.03	254.0	1.21	270
1130[b]	39.3	1.48	1.72	248.5	1.21	490
1148[b]	39.3	1.38	1.63	246.6	1.18	560
1158[b]	43.7	1.56	1.63	245.1	1.19	340
1168[b]	43.7	1.38	1.63	246.7	1.07	60

[a] Mass transfer data based on Xe.

[b] Mass transfer data based on Kr. (Taken from Campaign III; see ref. 5).

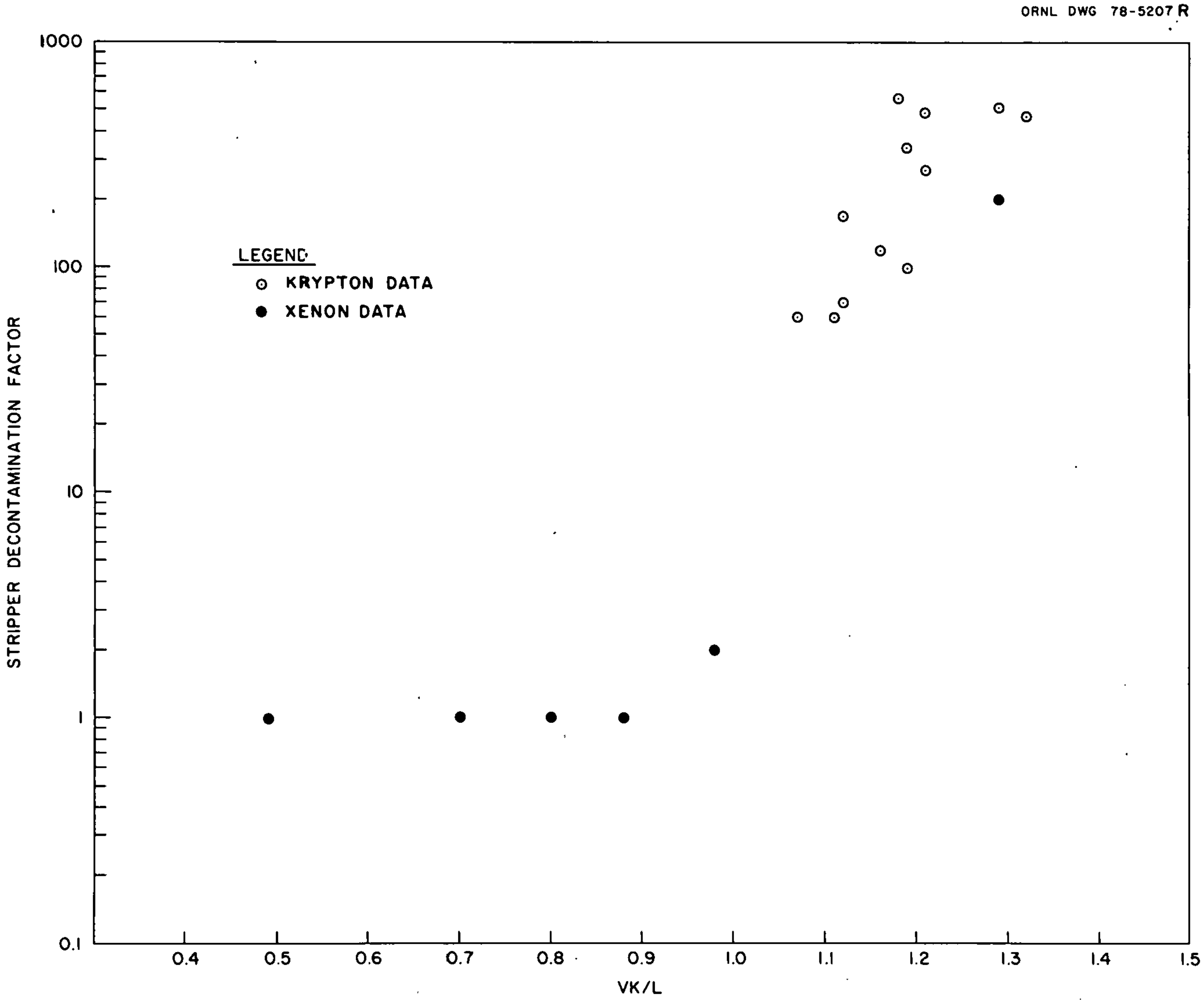

Fig. 6. Comparison between stripper DF for Kr and Xe as a function of stripping factor (based on Kr).

4. SOLUBILITY STUDIES

The Mullins computer model[6] has used published literature data to predict Henry's law constants of the various KALC components over a wide temperature range (see Fig. 7). As indicated by the figure, at temperatures warmer than -42°C, $K(N_2) > K(CO) > K(O_2) > K(Kr) > K(Xe)$. Both two- and three-column experiments were performed to verify the relative volatility of each component. The model predicts solubility behavior over a temperature range of -55 to 10°C. The experimental facility does not possess sufficient flexibility to vary experimental conditions over that range. Consequently, experiments were carried out over a very limited temperature range near -20°C. It has been shown in previous sections that the presence of CO or N_2 caused a negligible effect on Kr or O_2 mass transfer. The data treatment presented here utilizes the assumption of no interaction. For a more detailed discussion of the data analysis, see Appendix B.

Three-column experiments were performed to quantify the relative solubilities of CO and O_2. It is important to know the relative solubilities, because operations must be controlled to eliminate the accumulation of explosive mixtures of O_2 and CO (see Appendix C). The three-column data use the fact that if $K(CO) > K(O_2)$: (1) O_2 will be preferentially absorbed (over CO) in the absorber; (2) CO will be preferentially "stripped" (over O_2) in the fractionator; and (3) greater quantities of O_2 (over CO) will be sent to the stripper. Thus, if $K(CO) > K(O_2)$, the concentration factor (CF) for CO will be less than that for O_2. The CF is a measure of the solubility of a component in

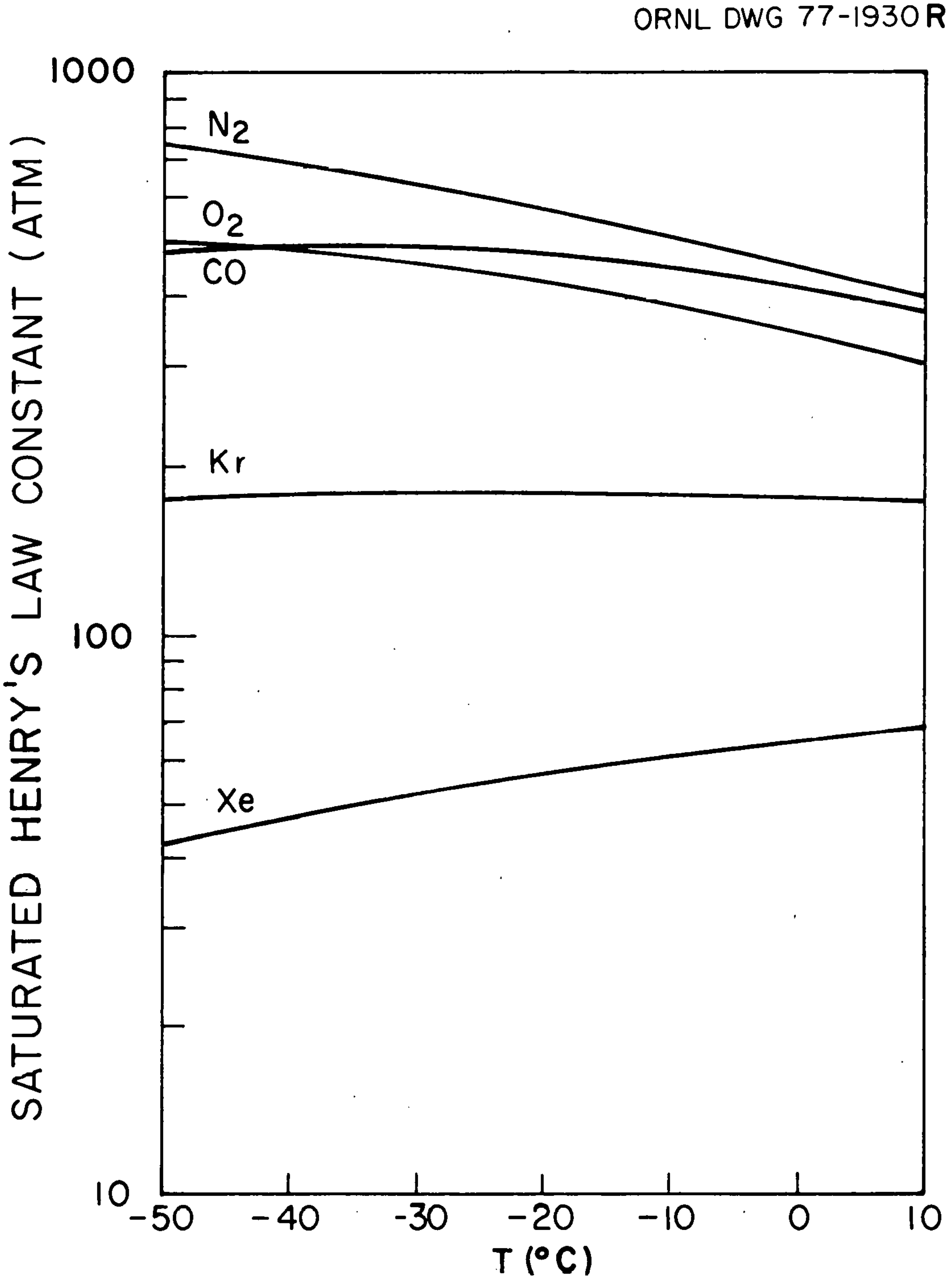

Fig. 7. Relative values of Henry's law constants for components in KALC feed gas.

liquid CO_2 for all components with an equilibrium constant greater than or equal to Xe (i.e., the higher the CF, the more soluble the component) and is defined by

$$CF_i = \frac{\text{concentration of component i in the stripper off-gas}}{\text{concentration of component i in the system feed-gas}} \ . \qquad (8)$$

The resulting data are shown in Table 7. As expected, CF(Kr) >> CF(CO) or $CF(O_2)$, which indicates that K(Kr) << K(CO) or $K(O_2)$. In several experiments CF(CO) > $CF(O_2)$, indicating that K(CO) < $K(O_2)$, which is contrary to the model predictions. Since both components (CO and O_2) are pinched in the absorption step and are of low concentrations in the stripping step, the difference in solubilities is more easily discernible in the fractionation step. Thus the fractionator DF_o was determined for these experiments and is defined:

$$DF_o(i) = \frac{\text{amount of component i in fractionator liquid feed}}{\text{amount of component i in liquids leaving fractionator reboiler}}$$

These data can be used to determine the relative values of equilibrium constants because $DF_i \propto K_i$. The results of these calculations are shown in Table 8. For eight experiments, the $DF_o(CO) < DF_o(O_2)$, indicating that for these experimental conditions $K(CO) < K(O_2)$.

These data made it apparent that a more detailed look at solubilities was warranted, and two-column experiments were performed using CO_2-O_2-N_2, CO_2-O_2-CO, CO_2-O_2-CO-N_2, and CO_2-CO-N_2 systems. Using an absorber and stripper, an attempt was made to "pinch" each component in the stripper column. These experiments utilize the fact that in stripping operations, the composition of a given component in the

Table 7. Concentration factors for three-column CO/O_2 solubility experiments

Experiment	Pump rate (cm^3/s)	Absorber L/V (mole/mole)	Fractionator L/V (mole/mole)	Stripper L/V (mole/mole)	Overall concentration factor		
					Kr	O_2	CO
C4-377	26.2	8.7	13.4	4.4	24.5	0.11	<0.03
C4-386	26.2	8.9	13.4	4.3	38.0	0.07	<0.04
C4-394	26.2	8.0	14.5	4.3	42.6	0.02	<0.03
C4-402	26.2	9.4	15.7	4.0	64.9	0.53	2.40[a]
C4-411	26.2	8.9	15.1	4.1	100.0	0.47	1.20[a]
C4-420	26.2	10.4	15.9	4.1	420.0	0.82	6.90[a]
C4-428	26.2	9.8	15.3	4.0	141.0	0.92	3.50[a]
C4-437	26.2	10.5	15.2	4.0	65.8	0.02	<0.07
C4-444	26.2	10.8	17.3	3.9	155.0	1.22	2.70[a]
C4-454	26.2	9.6	15.6	3.9	116.5	0.52	2.86[a]
C4-465	39.3	15.6	14.9	4.2	121.7	1.23	4.38[a]
C4-475	39.3	14.2	18.4	4.0	11.5	1.80	3.90[a]

[a] CF(CO) > CF(O_2).

Table 8. Fractionator overall decontamination factors for three-column CO/O_2 solubility experiments

Experiment No.	Column pressure (MPa)	Top column temperature (K)	Bottom column temperature (K)	Average column temperature (K)	Overall decontamination factor based on Kr	O_2	CO
C4-377	2.02	253.0	253.8	253.4	3	410	1410
C4-386	2.02	252.4	253.9	253.1	3	930	2040
C4-394	2.03	252.1	253.8	252.9	3	3460	2370
C4-402	2.02	249.1	253.0	251.1	3	190	60[a]
C4-411	2.03	250.3	253.7	252.0	3	360	170[a]
C4-420	2.03	249.7	252.4	251.0	3	150	40[a]
C4-428	2.03	249.5	253.4	251.4	3	260	80[a]
C4-437	2.03	253.0	253.8	253.4	4	7080	3680[a]
C4-444	2.02	250.1	252.8	251.4	9	1110	740[a]
C4-454	2.03	249.4	253.8	251.5	3	330	110[a]
C4-465	2.03	250.6	253.7	252.1	3	190	80[a]
C4-475	2.03	250.1	250.6	250.3	2	9	6[a]

[a] $DF(O_2) > DF(CO)$.

liquid feed and exiting vapor will approach or reach equilibrium (i.e., pinch) as the boilup is decreased to the point that the stripping factor for that component (i.e., VK/L) becomes <1.0.[13] The component exhibiting the least volatility (i.e., lowest equilibrium constant) will pinch first; thus, in a CO_2-O_2-CO system, if $K(O_2) > K(CO)$ then CO will pinch before O_2 (i.e., at a higher boilup).

The experimental procedure is similar to the two-column mass transfer experiments. The exception is that instead of adjusting the boilup to give liquid-to-vapor ratios in the normal range of interest to KALC, the boilup is progressively lowered until one or all of the components becomes pinched. A component was declared pinched if the concentration in the top two samplers was the same. A summary of these data (Table 9) indicated that over the range of conditions studied, $K(N_2) > K(CO)$ and $K(O_2)$, which agrees favorably with the Mullins computer model. However, the data also indicate that, as with the three-column data, $K(CO) < K(O_2)$ at temperatures >231.2 K.

Table 10 summarizes the results of the CO equilibrium constant data from both two- and three-column experiments. These data indicate that at temperatures ≤252.1 K, $K(CO) < K(O_2)$, but at temperatures ≥253.1 K, $K(CO) > K(O_2)$. Thus the reversal in solubility between CO and O_2 occurs between these two temperatures rather than at 231.2 K.

To determine the cause of the discrepancy between the experimental and model-predicted solubilities, the equilibrium data used in the model were reviewed. The model weighed equally the published CO solubility of Kaminishi[14] and Christiansen.[15] As seen in Fig. 8, there is a discrepancy between the two sets of data. Note that the shear volume

Table 9. Summary of pinched experiments

Experiment No.	Column pressure (MPa)	Top column temperature (K)	Pinched[a]		
			O_2	CO	N_2
C4-544	2.03	250.7	N	Y	NP
C4-552	2.03	250.4	N	Y	NP
C4-576	1.83	247.0	N	Y	NP
C4-584	1.83	247.5	N	Y	NP
C4-652	2.03	249.4	N	Y	N
C4-658	2.03	250.2	N	Y	N
C4-672	2.03	249.5	Y	NP	N
C4-678	2.03	249.8	Y	NP	N
C4-687	2.03	249.2	Y	NP	N
C4-716	2.03	249.6	NP	Y	N
C4-720	2.03	249.7	NP	Y	N
C4-732	2.03	249.7	NP	Y	N

[a] Y = component pinched;

N = component not pinched;

NP = component not present.

Table 10. Summary of CO equilibrium constant data

Experiment No.	Pressure (MPa)	Average column temperature (K)	Component showing highest degree of solubility	
			O_2	CO
C4-377	2.03	253.4	X	
C4-386	2.03	253.1	X	
C4-402	2.03	251.1		X
C4-411	2.03	252.0		X
C4-420	2.03	251.0		X
C4-428	2.03	251.4		X
C4-444	2.03	251.4		X
C4-454	2.03	251.5		X
C4-465	2.03	252.1		X
C4-475	2.03	250.3		X
C4-544	2.03	250.7		X
C4-552	2.03	250.4		X
C4-576	1.83	247.0		X
C4-584	1.83	247.5		X
C4-652	2.03	249.4		X
C4-658	2.03	250.2		X

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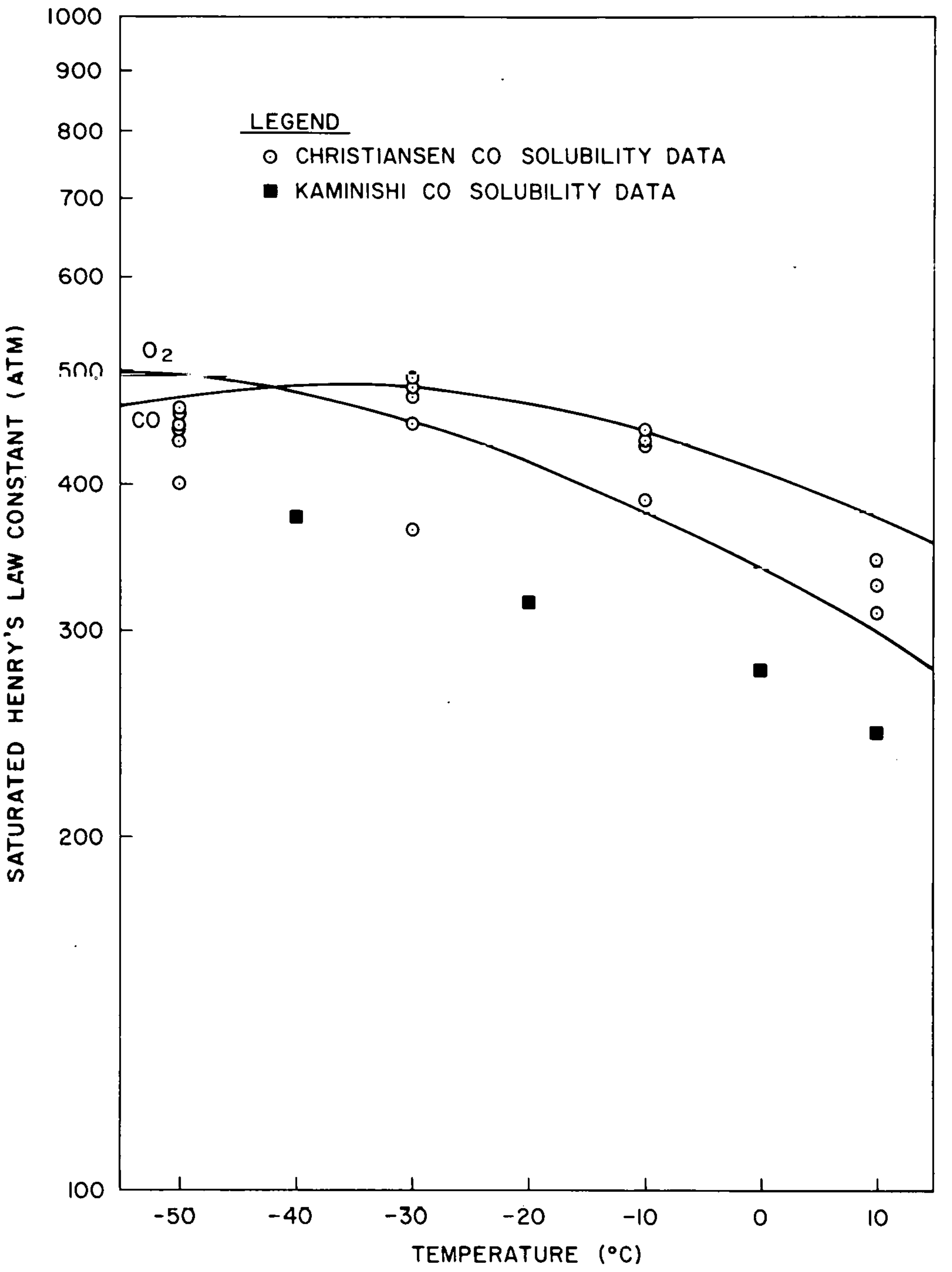

Fig. 8. Literature values for CO Henry's law constants and curves used for CO and O_2 in the computer model.

of the Christiansen data (vs that of Kaminishi) will tend to reduce the effect of the discrepancy in any model that weights the data sets equally. In fact, during the modeling the Kaminishi data were disregarded. The Campaign IV equilibrium constant data indicate that the Christiansen and Kaminishi data form upper and lower bounds for the true values. Thus, for modeling applications, some type of weighing factor will have to be determined for each of the two data sets.

5. CONCLUSIONS

The data collected during Campaign IV provided information that is applicable to a commercial-scale KALC-type process. The presence of N_2 or CO does not appreciably alter the mass transfer characteristics of O_2 and Kr as exhibited in the basic CO_2-O_2-Kr system. Thus the system decontamination factor (based on krypton) is independent of the light-gas composition in the system feed. The maximum allowable light-gas concentration in the system feed will therefore be restricted to that which overloads the column [i.e., introduces sufficient light gas (i.e., uncondensibles) to cause a light-gas block in the column, or loss of recycle CO_2 due to vaporization of the liquid].

It was determined that stripper-reboiler heat loads sufficient to remove Kr from the liquid scrub are insufficient to remove Xe, which limits the fraction of Xe absorbed from the incoming feed gas. Thus the majority of Xe exits via the absorber off-gas, which would not be expected from the relative values of Henry's constants. This presents no environmental hazard, because the proposed 150-day cooling of spent fuel prior to reprocessing will allow the complete decay of all radioactive

Xe. The experiments confirm that by increasing the stripper boilup, Xe is concentrated in the stripper off-gas along with the Kr. This concentration of the Xe also provides a Xe-free liquid scrub which results in the absorber removing essentially all of the Xe from the incoming system feed gas. The Xe can then be separated from the concentrated stripper off-gas via molecular sieves.[16] However, this method of operating seems unlikely due to the limited commercial use of Xe (it is used mainly as a light source for specialized applications).[17] The optimum Kr concentration for final long-term storage has not yet been determined. It is possible that the concentration of Xe could be used as a dilutant in this final storage.

Experiments also indicated that contrary to previously published reports, CO is more soluble in liquid CO_2 than is O_2 at temperatures below 252.2 K. This means that during typical KALC operations, CO will be concentrated along with O_2 in the fractionator off-gas. The explosive nature of CO-O_2 mixtures makes this an undesirable situation. Consequently, it will probably be necessary to remove CO from the KALC feed via catalytic conversion to CO_2.

Campaign IV is the last experimental work planned in the EES-ODF. A limited amount of future work will be carried out to improve the Mullins computer model so it will agree more closely with the experimental results. The computer model could then be used as an aid in optimizing the KALC configuration and operating conditions.

6. APPENDIXES

6.1 Appendix A: Mass Transfer Studies

To aid in the data analysis, a schematic of the two- and three-column systems are shown in Figs. A.1 and A.2 respectively. The streams are defined as follows:

①	System feed gas
②	Absorber off-gas
③	Stripper off-gas
④	Liquid leaving absorber-level tank (feed to stripper during two-column experiments; feed to fractionator during three-column experiments)
⑤	Liquid leaving stripper reboiler
⑥	Liquid entering stripper packing
⑦	Liquid leaving stripper packing
⑧	Vapor leaving stripper packing
⑨	Vapor entering stripper packing
⑩	Fractionator off-gas
⑪	Liquid entering fractionator packing
⑫	Liquid leaving fractionator packing
⑬	Vapor leaving fractionator packing
⑭	Vapor entering fractionator packing
⑮	Liquid leaving fractionator (feed to stripper during three-column experiments).

During the experiments, only the column off-gas and system feed streams could be identified as to flow rate (via calibrated rotameters) and composition (via sampling and analysis). Once the composition and

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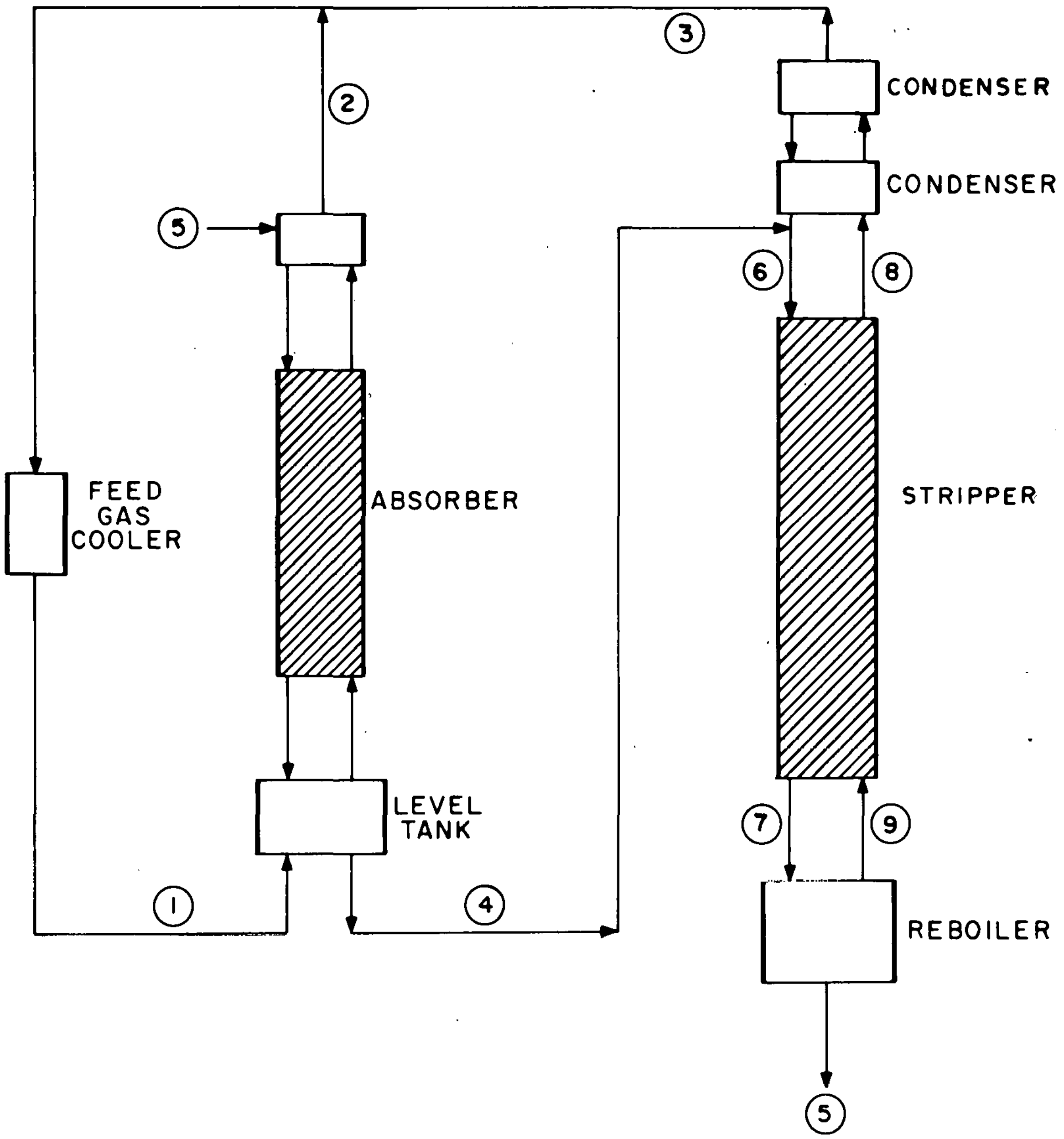

Fig. A.1. Schematic diagram of two-column system used in data analysis.

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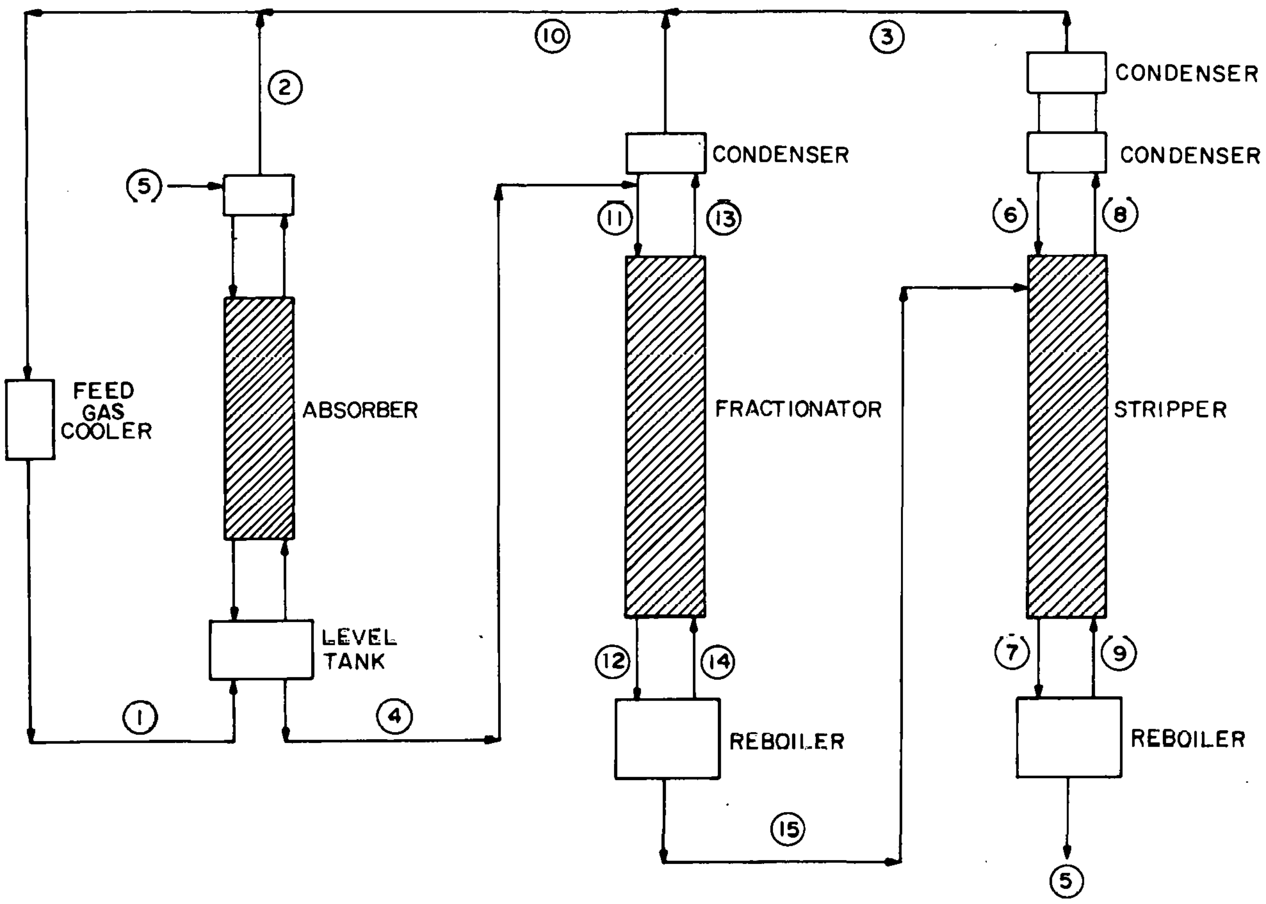

Fig. A.2. Schematic diagram of three-column system used in data analysis.

flow rates of these streams are known, a component material balance can be made. For two-column experiments this becomes

$$\beta_i = \frac{Q_2 y_2 + Q_3 y_3}{Q_1 y_1} , \qquad (A.1)$$

where

β_i = balance factor of component i,
Q_2 = flow rate of stream 2, std cm^3/s,
y_2 = concentration of component i in stream 2,
Q_3 = flow rate of stream 3, std cm^3/s,
y_3 = concentration of component i in stream 3,
Q_1 = flow rate of stream 1, std cm^3/s, and
y_1 = concentration of component i in stream 1.

For three-column experiments

$$\beta_i = \frac{Q_2 y_2 + Q_{10} y_{10} + Q_3 y_3}{Q_1 y_1} , \qquad (A.2)$$

where

Q_{10} = flow rate of stream 10, std cm^3/s, and
y_{10} = concentration of component i in stream 10.

The results of these calculations are shown in Tables A.1 through A.5.

The determination of the composition of a stream involves both sampling and analysis. Light-gas concentrations are determined using an AERO-VAC mass spectrometer,[12] Teledyne electrolytic cell,[18] and Lira Infrared Analyzer,[18] whereas the ^{85}Kr and ^{133}Xe concentrations were

Table A.1. Off-gas balance for two-column N_2/CO absorber experiments

Experiment No.	System feed gas					Absorber off-gas					Stripper off-gas					Balance factors			
	Flow rate (std cm^3/s)[a]	Composition				Flow rate (std cm^3/s)[a]	Composition				Flow rate (std cm^3/s)[a]	Composition							
		O_2 (%)	CO (%)	N_2 (%)	Kr (ppb)		O_2 (%)	CO (%)	N_2 (%)	Kr (ppb)		O_2 (%)	CO (%)	N_2 (%)	Kr (ppb)	O_2	CO	N_2	Kr
C4-35	2511	3.1		9.4	7.86	1817	1.9		5.8	0.0032	694	7.9		12.2	29.64	1.15		0.81	1.04
C4-41	2926	3.7		9.1	8.08	2142	1.4		3.7	0.04	783	7.2		18.2	31.46	0.80		0.83	1.05
C4-48	3289	1.8		6.3	4.73	2643	1.1		4.8	0.097	647	5.9		21.4	22.75	1.14		1.28	0.96
C4-87	2533	5.9		9.8	1.92	1888	1.8		2.1	0.0091	746								
C4-94	2171	4.1		4.5	12.4	1468	0.9		1.5	0.0014	689	7.9		7.0	37.1	0.76		0.72	0.92
C4-304	2303	4.1	1.8	8.9	8.92	1619	3.1	0.8	6.5	0.163	689	8.3	4.5	11.5	29.26	1.14	0.80	0.90	0.99
C4-310	2275	4.2	0.8	6.8	6.42	1614	1.7	0.1	3.3	0.100	665	4.6	2.5	10.4	21.36	0.61	1.00	0.79	0.98

[a] STP = 0.1 MPa, 294.3 K.

Table A.2. Gas stream values for two-column O_2 absorber experiments

Experiment No.	System feed gas			Absorber off-gas			Fractionator off-gas			Stripper off-gas			Balance factors	
		Concentrations			Concentrations			Concentrations			Concentrations			
	Flow rate (std cm^3/s)[a]	O_2 (%)	Kr (ppb)	Flow rate (std cm^3/s)[a]	O_2 (%)	Kr (ppb)	Flow rate (std cm^3/s)[a]	O_2 (%)	Kr (ppb)	Flow rate (std cm^3/s)[a]	O_2 (%)	Kr (ppb)	O_2	Kr
C4-1236	2463	10.05	18.99	1449	2.45	0.00063	873	22.4	33.6	90	0.097	181.8	0.93	0.98
C4-1246	2813	9.3	18.4	2001	3.6	0.38	727	24.6	47.4	94	0.33	187.2	0.96	1.01
C4-1254	2657	8.64	16.25	1638	3.38	0.0022	864	18.52	27.3	90	0.0024	209.9	0.94	0.98
C4-1264	2761	6.3	15.8	1798	5.9	0.009	864	14.8	28.6	94	0.004	203.3	1.16	1.00
C4-1272	2841	10.6	13.8	1850	5.7	0.007	864	21.9	23.9	94	0.007	211.6	0.98	1.03
C4-1280	3015	11.2	13.8	2015	4.8	0.02	868	22.2	26.9	90	0.008	197.3	0.86	0.99
C4-1290	3124	9.4	13.2	2322	6.2	0.069	736	19.7	31.4	94	0.007	193.8	0.98	1.00
C4-1300	3096	9.3	11.4	2444	4.9	0.11	628	25.0	29.3	90	0.040	194.8	0.95	1.03
C4-1310	2921	9.9	12.9	2242	5.1	0.09	628	26.9	33.5	94	0.08	190.3	0.98	1.03
C4-1318	2690	8.7	13.5	1987	4.2	0.01	623	24.1	32.2	94	0.01	189.6	1.00	1.04
C4-1326	2586	8.8	13.4	1888	3.8	0.017	618	25.8	31.9	94	0.04	171.1	1.01	1.03

[a] STP = 0.1 MPa, 294.3 K.

Table A.3. Gas stream values for two-column N_2/CO fractionator experiments

Experiment No.	System feed gas					Absorber off-gas					Stripper off-gas					Balance factors			
	Flow rate (std $cm^3/s)^a$	Composition				Flow rate (std $cm^3/s)^a$					Flow rate (std $cm^3/s)^a$	Composition							
		O_2 (%)	CO (%)	N_2 (%)	Kr (ppb)		O_2 (%)	CO (%)	N_2 (%)	Kr (ppb)		O_2 (%)	CO (%)	N_2 (%)	Kr (ppb)	O_2	CO	N_2	Kr
C4-124	2407	4.09		1.73	4.93	1694	1.25		0.88	1.43	694	10.6		3.98	13.6	0.96		1.02	1.00
C4-130	2298	5.24		2.73	3.96	1595	1.60		1.39	1.15	708	13.59		6.27	10.91	1.01		1.06	1.05
C4-135	2256	4.34		3.06	3.61	1562	1.33		1.56	1.05	708	11.25		7.05	9.95	1.03		1.08	1.07
C4-139	2308	4.56		2.91	2.89	1604	1.39		1.48	0.84	703	11.83		6.71	7.97	1.00		1.06	1.04
C4-143	2355	3.74		1.97	2.80	1737	1.14		1.00	0.81	637	9.70		4.53	7.73	0.93		1.00	0.96
C4-148	2308	6.63		3.36	2.31	2166	2.03		1.71	0.67	713	17.19		7.72	6.37	1.09		1.19	1.12
C4-316	2166	1.43	1.25	3.43	4.3	1505	0.87	0.4	2.89	2.3	656	6.13	3.5	5.16	8.7	1.72	1.07	1.04	0.98
C4-323	2208	2.33	1.76	2.5	4.4	1548	1.68	0.84	2.77	2.19	675	5.16	4.16	4.52	9.49	1.18	1.06	1.28	1.01
C4-329	2237	5.9	1.05	4.27	3.15	1557	3.93	0.5	3.11	1.56	634	10.81	2.30	4.82	8.41	1.02	1.15	0.85	1.16

[a] STP = 0.1 MPa, 294.3 K.

Table A.4. Gas stream values for two-column CO/N_2 stripping experiments

Experiment No.	System feed gas					Absorber off-gas					Stripper off-gas					Balance factors			
	Flow rate (std cm^3/s)[a]	Composition				Flow rate (std cm^3/s)[a]	Composition				Flow rate (std cm^3/s)[a]	Composition							
		O_2 (%)	CO (%)	N_2 (%)	Kr (ppb)		O_2 (%)	CO (%)	N_2 (%)	Kr (ppb)		O_2 (%)	CO (%)	N_2 (%)	Kr (ppb)	O_2	CO	N_2	Kr
C4-100	2213	3.51		3.71	11.43	1524	1.19		1.76	0.0037	680	7.01		5.70	37.22	0.85		0.80	1.00
C4-106	2369	3.41		3.39	10.48	1623	1.07		1.59	0.015	680	6.03		5.36	35.19	0.72		0.77	0.96
C4-112	2213	2.27		2.27	9.19	1520	0.62		1.12	0.0045	680	5.45		4.11	30.64	0.92		0.89	1.02
C4-118	2482	7.69	1.8	12.43	6.69	1784	2.77		3.83	0.16	694	11.01		5.12	23.48	0.66		0.34	1.00
C4-304	2260	4.09	0.75	8.9	8.92	1604	3.1	0.8	6.46	0.16	651	8.3	4.5	11.5	29.26	1.12	1.04	0.89	0.96
C4-310	2275	4.20		6.8	6.42	1614	1.65	0.1	3.28	0.10	665	4.58	2.5	10.38	21.36	0.60	1.07	1.28	0.98

[a] STP = 0.1 MPa, 294.3 K.

Table A.5. Gas-stream values for three-column Xe experiments

Experiment No.	System feed gas			Absorber off-gas			Fractionator off-gas			Stripper off-gas			Balance factor	
	Flow rate (std cm^3/s)[a]	Composition		Flow rate (std cm^3/s)[a]	Composition		Flow rate (std cm^3/s)[a]	Composition		Flow rate (std cm^3/s)[a]	Composition		O_2	Xe
		O_2 (%)	Xe (ppb)		O_2 (%)	Xe (ppb)		O_2 (%)	Xe (ppb)		O_2 (%)	Xe (ppb)		
C4-838	1656	7.9	4.55	595	0.3	2.23	958	14.8	5.58	57	13.2	59.34	1.13	0.91
C4-845	1652	5.4	2.51	595	0.3	1.12	949	1.72	1.31	57	6.9	75.25	0.21	0.54
C4-852	1704	12.0	9.37	632	<1.0	3.71	967	18.8	10.90	47	2.2	542.5	<0.92	0.92
C4-859	1704	10.15	9.33	628	<1.0	4.23	967	18.0	12.44	57	< 1.0	196.8	<1.04	0.94
C4-869	1822	9.6	0.52	590	0.095	0.00097	1123	12.8	0.28	61	0.042	108.4	0.83	0.87
C4-881	1935	6.60	18.07	609	0.35	2.09	1123	11.3	12.33	99	0.0375	215.1	1.03	1.04

[a] STP = 0.1 MPa, 294.3 K.

determined by a beta detector.[12] Experience has proved that the determination of Kr concentrations is more reliable than that of the light gas; thus the Kr balance factor makes it possible to determine the validity of an experiment.

Although gas flow rates readily lend themselves to units of std cm^3/s, liquid flow rates do not. Hence, stream 5 must be expressed in terms of "equivalent std cm^3/s." Stream 5 is the only flow rate of concern at this point, since all other flow rates will be determined by material balance and will naturally be expressed in units of std cm^3/s. Stream 5 is calculated by:

$$Q_5 = \frac{(\rho_L^*)(Q_5^1)}{(\rho_G^s)} , \qquad (A.3)$$

where

Q_5 = liquid flow rate to the absorber column, std cm^3/s,
ρ_L^* = density of saturated liquid CO_2 at fluid temperature, g/cm^3
Q_5^1 = liquid flow rate to the absorber column, cm^3/s, and
ρ_G^s = density of CO_2 gas at standard conditions of 0.1 MPa and 294.3 K, g/cm^3.

Tables A.6 through A.9 present the results of these conversions. At this point all information that is common to the absorption, fractionation, and stripping studies have been calculated.

6.1.1 Absorber studies with N_2 and CO present

The absorber overall decontamination factor (DF_o) is defined as:

$$DF_o = \frac{Q_1 y_1}{Q_2 y_2} , \qquad (A.4)$$

Table A.6. Equivalent pump rate for absorber experiments

Experiment No.	Pump rate (cm^3/s)	Temperature (K)	Equivalent pump rate (std cm^3/s)[a]
C4-35	34.9	246.2	21,141
C4-41	34.9	245.7	20,245
C4-48	34.9	247.4	20,103
C4-87	34.9	246.0	20,245
C4-94	34.9	245.4	20,292
C4-304	26.2	243.0	15,384
C4-310	26.2	243.4	15,337
C4-377	26.2	244.4	15,290
C4-386	26.2	244.6	15,242
C4-394	26.2	244.5	15,290
C4-402	26.2	244.0	15,290
C4-411	26.2	244.5	15,290
C4-420	26.2	244.7	15,242
C4-428	26.2	244.4	15,290
C4-437	26.2	244.7	15,242
C4-444	26.2	243.7	15,337
C4-454	26.2	244.2	15,290
C4-465	39.3	245.5	22,793
C4-475	39.3	244.9	22,840
C4-1236	39.3	246.5	22,698
C4-1246	39.3	246.9	22,651
C4-1254	39.3	245.7	22,793
C4-1264	39.3	245.4	22,793
C4-1272	39.3	245.3	22,840
C4-1280	39.3	244.7	22,887
C4-1290	39.3	245.1	22,840
C4-1300	39.3	245.5	22,793
C4-1310	39.3	245.2	22,840
C4-1318	39.3	245.4	22,793
C4-1326	39.3	245.5	22,793

[a] STP = 0.1 MPa, 294.3 K.

Table A.7. Equivalent pump rate for fractionator experiments

Experiment No.	Pump rate (cm^3/s)	Temperature (K)	Equivalent pump rate (std cm^3/s)[a]
C4-124	34.9	246.0	20,245
C4-130	34.9	246.4	20,197
C4-135	34.9	246.0	20,245
C4-139	34.9	246.0	20,197
C4-143	34.9	246.4	20,197
C4-148	34.9	246.8	20,150
C4-316	26.2	243.6	15,337
C4-323	26.2	243.5	15,337
C4-329	26.2	243.4	15,337

[a] STP = 0.1 MPa, 294.3 K.

Table A.8. Equivalent pump rate for stripping experiments

Experiment No.	Pump rate (cm^3/s)	Liquid temperature (K)	Pump rate (std cm^3/s)[a]
C4-100	34.9	246.0	20,221
C4-106	34.9	245.4	20,273
C4-112	34.9	245.8	20,240
C4-118	34.9	245.8	20,240
C4-304	26.2	243.0	15,365
C4-310	26.2	243.4	15,337

[a] STP = 0.1 MPa, 294.3 K.

Table A.9. Equivalent pump rate for xenon experiments

Experiment No.	Pump rate (cm^3/s)	Liquid temperature (K)	Liquid rate (std cm^3/s)[a]
C4-838	39.3	244.7	22,878
C4-845	39.3	244.5	22,901
C4-852	39.3	244.8	22,868
C4-859	39.3	244.9	22,859
C4-869	39.3	243.7	22,977
C4-881	39.3	245.8	22,774

[a] STP = 0.1 MPa, 294.3 K.

where y refers to the Kr concentration of the indicated stream. The absorber liquid-to-vapor ratio is an "operating ratio" and does not necessarily equal the actual column liquid-to-vapor ratio. The liquid-to-vapor ratio (L/V) is defined as:

$$L/V = Q_5/Q_1, \text{ mole/mole.} \tag{A.5}$$

The column absorption factor (A) is then defined as

$$A = L/VK, \tag{A.6}$$

where K is the Kr equilibrium constant[*] at column pressure and average column temperature.

The resulting data showing the CO and N_2 effect on Kr absorption are shown in Table A.10. Of these, two (C4-94 and C4-1236) were deleted due to the poor counting statistics of the ^{85}Kr in the absorber off-gas. In the analysis of the absorber data, the measured Kr concentrations of the system feed-gas and absorber off-gas are of extreme importance, because they are the basis of the mass transfer variables of interest (DF_υ). A statistical analysis of the measured count rate proved to be the most convenient test of reliability. The standard deviation for any sample is defined as:[19]

$$\sigma_N = \sqrt{{\sigma_c}^2 + {\sigma_\beta}^2}\ , \tag{A.7}$$

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[*] All equilibrium values used in the report are calculated using the Glass model.[11]

Table A.10. Absorber mass transfer data

Experiment No.	Column pressure (psig)	Average column temp (°C)	L/V (mole/mole)	L/VK	Overall Kr decontamination factor
C4-35[a]	2.42	257.2	8.05	1.04	3620
C4-41[a]	2.41	256.9	6.93	0.89	280
C4-48[a]	2.36	256.5	6.11	0.76	60
C4-87[a]	2.36	255.4	7.68	0.99	290
C4-94[a]	2.35	256.1	9.34	1.16	13100
C4-304[b]	2.52	254.3	6.67	0.89	80
C4-310[b]	2.42	254.6	6.74	0.90	90
C4-377[c]	2.67	256.1	5.72	0.80	730
C4-386[c]	2.61	256.0	5.72	0.79	1040
C4-394[c]	2.64	255.8	5.43	0.76	440
C4-402[c]	2.51	255.8	6.04	0.80	870
C4-411[c]	2.56	255.9	5.62	0.76	700
C4-420[c]	2.47	256.0	6.39	0.84	2370
C4-428[c]	2.43	255.3	6.15	0.79	1910
C4-437[c]	2.59	257.5	6.52	0.89	3260
C4-444[c]	2.34	255.5	6.65	0.82	450
C4-454[c]	2.47	257.0	6.15	0.79	330
C4-465[c]	2.38	256.9	9.70	1.21	6520
C4-475[c]	2.46	257.1	8.87	1.12	8010
C4-1246[d]	2.32	254.3	8.05	0.99	70
C4-1264[d]	2.26	253.4	8.26	0.98	2690
C4-1272[d]	2.32	253.8	8.04	0.98	3070
C4-1280[d]	2.33	253.7	7.59	0.94	1030
C4-1290[d]	2.30	253.3	7.31	0.89	260
C4-1300[d]	2.25	252.9	7.36	0.88	130
C4-1310[d]	2.27	253.2	7.82	0.93	180
C4-1318[d]	2.31	253.4	8.47	1.03	1790
C4-1326[d]	2.28	253.7	8.81	1.06	1080
C4-1236[d]	2.40	256.0	9.20	1.17	51170
C4-1254[d]	2.25	253.4	8.60	1.01	11980

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[a] CO_2-O_2-N_2-Kr system.

[b] CO_2-O_2-N_2-CO-Kr system.

[c] CO_2-O_2-CO-Kr system.

[d] CO_2-O_2-Kr system.

where

σ_N = standard deviation of sample,

σ_c = standard deviation of total number of counts registered, and

σ_β = standard deviation of the number of background counts registered.

Statistical analysis indicates a 99.7% probability that the true count rate lies within ± 3 σ_N of the actual count rate.[19] Thus a somewhat arbitrary decision was made to accept a sample (as being reliable) whose counting statistics were such that the magnitude of 3 σ_N was 10% or less of the actual count rate.

6.1.2 Fractionation studies with N_2 and CO present

The column vapor rate is determined by the boilup rate:

$$Q_9 = (KW)(B) , \tag{A.8}$$

where

Q_9 = vapor rate entering column packing, std cm^3/s,

KW = heat supplied by the reboiler, kW, and

B = factor for conversion from kW to std cm^3/s, nominally 19.0 std cm^3/s per kW (see ref. 20).

The fractionator column data are shown in Table A.11. The column liquid rate (Q_7) is then determined by a material balance around the reboiler:

$$Q_7 = Q_9 + Q_5 . \tag{A.9}$$

Table A.11. Fractionator column data

Experiment No.	Reboiler heat load (kW)	Reboiler temperature (K)	Boilup (std cm^3/s)[a]	Column liquid outlet concentrations		Column liquid inlet concentrations	
				O_2 (%)	Kr (ppb)	O_2 (%)	Kr (ppb)
C4-124	0.73	254.0	1425	0.003	0.26	0.145	0.47
C4-130	0.64	254.2	1241	0.0055	0.36	0.142	0.39
C4-135	0.66	253.3	1288	0.0005	0.33	0.106	0.36
C4-139	0.66	254.0	1269	0.0013	0.25	0.103	0.28
C4-143	0.74	253.2	1416	0.0004	0.22	0.103	0.28
C4-148	0.71	254.2	1373	0.0008	0.20	0.229	0.22
C4-316	0.52	254.1	1005	0.0017	0.41	0.100	0.43
C4-323	0.54	253.9	1038	0.0004	0.38	0.100	0.42
C4-329	0.55	254.0	1066	0.0011	0.27	0.142	0.35

[a] STP = 0.1 MPa, 294.3 K.

The column liquid and vapor rates are assumed to be constant, therefore the column liquid-to-vapor ratio (L/V) becomes

$$L/V = Q_7/Q_9 \text{ (mole/mole)} \tag{A.10}$$

The column stripping factor (A^{-1}) is then defined as

$$A^{-1} = VK/L \tag{A.11}$$

where

K = oxygen equilibrium constant based on column pressure and average temperature.

The column decontamination factor (DF_c) is defined

$$DF_c = \frac{Q_6 x_6}{Q_7 x_7} \; ; \tag{A.12}$$

however,

$$Q_6 = Q_7 \; ; \tag{A.13}$$

thus

$$DF_c = \frac{x_6}{x_7} \; , \tag{A.14}$$

where

x_6 = oxygen concentration of stream 6, and

x_7 = oxygen concentration of stream 7.

The resulting mass transfer data are shown in Table A.12.

Table A.12. Fractionator mass transfer information

Experiment No.	Pump rate (cm^3/s)	Reboiler heat load (kW)	Column pressure (MPa)	Average column temperature (K)	L/V (mole/mole)	VK/L	O_2 decontamination factor
C4-124[a]	34.9	0.73	2.03	254.0	15.2	1.20	50
C4-130[a]	34.9	0.64	2.03	254.0	17.3	1.05	30
C4-135[a]	34.9	0.66	2.03	254.3	16.7	1.09	210
C4-139[a]	34.9	0.66	2.03	254.0	16.9	1.07	80
C4-143[a]	34.9	0.74	2.03	253.4	15.3	1.20	260
C4-148[a]	34.9	0.71	2.03	254.2	15.7	1.16	290
C4-316[b]	26.2	0.52	2.03	254.1	16.3	1.12	60
C4-323[b]	26.2	0.54	2.03	253.9	15.8	1.15	260
C4-329[b]	26.2	0.55	2.03	254.0	15.4	1.18	130

[a] CO_2-O_2-N_2-Kr system.

[b] CO_2-O_2-CO-N_2-Kr system.

6.1.3 Stripping studies with N_2 and CO present

The stripping data (shown in Tables A.13 and A.14) determination is identical to that of the fractionation data with two exceptions. The stripping factor defined in Eq. (A.11) now becomes

$$A^{-1} = VK/L \tag{A.15}$$

where

K = Kr equilibrium constant based on column pressure and average column temperature.

The column decontamination factor defined in Eq. (A.12) has been replaced by an overall decontamination factor (DF_o)

$$DF_o = \frac{Q_4 x_4}{Q_5 x_5} , \tag{A.16}$$

where

Q_4 = liquid feed to stripper, std cm^3/s,

x_4 = Kr concentration of stream 4, and

x_5 = Kr concentration of stream 5.

Now

$$Q_4 = Q_5 + Q_3 , \tag{A.17}$$

thus Eq. (A.16) becomes

$$DF_o = \frac{Q_3 x_4}{Q_5 x_5} + \frac{x_4}{x_5} \tag{A.18}$$

Table A.13. Stripper column data

Experiment No.	Reboiler heat load (kW)	Reboiler temp (K)	Boilup (std cm^3/s)[a]	Column liquid bottoms concentration Kr (ppb)	Column liquid feed concentration Kr (ppb)
C4-100	1.83	251.0	3483	0.0084	0.73
C4-106	1.49	250.7	2883	0.0042	0.71
C4-112	1.46	250.8	2765	0.0022	0.65
C4-118	1.16	250.9	2204	0.027	0.52
C4-304	1.02	250.8	1930	0.0085	0.99
C4-310	1.10	250.6	2086	0.0030	0.63

[a] STP = 0.1 MPa, 294.3 K.

Table A-14. Stripper mass transfer information

Experiment No.	Pump rate (cm^3/s)	Reboiler heat load (kW)	Column pressure (MPa)	Average column temperature (K)	L/V (mole/mole)	$\frac{VK}{L}$	Overall Kr decontamination factor
C4-100[a]	34.9	1.83	1.85	251.0	6.81	1.50	90
C4-106[a]	34.9	1.49	1.84	250.7	8.03	1.26	170
C4-112[a]	34.9	1.46	1.84	250.9	8.32	1.24	300
C4-118[a]	34.9	1.16	1.84	251.0	10.19	1.01	20
C4-304[b]	26.2	1.02	1.83	250.8	8.95	1.15	120
C4-310[b]	26.2	1.10	1.83	250.5	8.35	1.25	210

[a] CO_2-O_2-N_2-Kr system.

[b] CO_2-O_2-N_2-CO-Kr system.

6.1.4 Xenon studies

Of the six three-column experiments, only C4-845 showed an unacceptable off-gas balance (see Table A.5). The poor balance in C4-845 is caused by an insufficient amount of fractionator off-gas that was taken for proper composition analysis. This sample is not used in determining the mass transfer variables of interest; therefore, the experiment is assumed to be valid (and does, indeed, agree with the other data).

The absorber mass transfer data are shown in Table A.15. The data analysis is identical to that in Sect. A.1 with two exceptions: the concentrations used in Eq. (A.4) refer to Xe concentrations, and the equilibrium constant used in Eq. (A.6) refers to Xe.

As with the absorber experiments, the stripper data (Tables A.16 and A.17) determination is identical to that for N_2 and CO studies (see Sect. A.3), except that the concentrations used in Eq. (A.14) refer to Xe, and the equilibrium constant used in Eq. (A.15) refers to Xe.

6.2 Appendix B: Solubility Experiments

The three-column equilibrium constant data are shown in Tables B.1-B.3. The system concentration factor for component i is defined

$$CF_i = \frac{y_3}{Y_1} , \tag{B.1}$$

where y refers to the composition of component i in the indicated streams. An overall and component mass balance around the stripper column determines the flow rate (Q_{15}) and composition (x_{15}) of the stripper liquid feed (i.e., liquid leaving the fractionator reboiler).

Table A.15. Absorber mass transfer data for Xe experiments

Experiment No.	Column pressure (MPa)	Average column temperature (K)	L/V (mole/mole)	L/VK	Overall Xe decontamination factor
C4-838	2.48	258.3	13.8	4.55	6
C4-845	2.45	258.5	13.9	4.60	7
C4-852	2.53	259.1	13.4	4.49	7
C4-859	2.54	259.5	13.4	4.52	6
C4-869	2.50	259.2	12.6	4.23	1970
C4-881	2.57	260.3	11.8	4.05	30

Table A.16. Stripper column data for Xe experiments

Experiment No.	Reboiler heat load (kW)	Reboiler temp (K)	Boilup (std cm^3/s)[a]	Column liquid outlet concentration		Column liquid inlet concentration	
				O_2 (%)	Xe (ppb)	O_2 (%)	Xe (ppb)
C4-838	1.94	250.6	3,667	0	0.659	0.0001	0.644
C4-845	3.00	250.7	5,686		0.279		0.350
C4-852	3.57	250.5	6,753		1.08	0.0039	1.21
C4-859	4.02	250.6	7,607		1.17	0.0033	1.32
C4-869	6.96	250.4	13,119	0	0.000161	0.0005	0.0325
C4-881	4.63	250.6	8,763		0.659	0.0013	1.381

[a] STP = 0.1 MPa, 294.3 K.

Table A.17. Stripper mass transfer data for Xe experiments

Experiment No.	Pump rate (cm^3/s)	Reboiler heat load (kW)	Column pressure (MPa)	Average column temperature (K)	L/V (mole/mole)	$\frac{VK}{L}$	Xe decontamination factor
C4-838	39.3	1.94	1.83	250.7	7.24	0.49	1
C4-845	39.3	3.00	1.83	250.7	5.03	0.70	1
C4-852	39.3	3.57	1.83	250.5	4.39	0.80	1
C4-859	39.3	4.02	1.83	250.6	4.00	0.88	1
C4-869	39.3	6.96	1.83	250.3	2.75	1.29	200
C4-881	39.3	4.63	1.83	250.6	3.60	0.98	2

Table B.1. Gas stream values for three-column CO/O_2 solubility experiments

Experiment No.	System feed gas				Absorber off-gas				Fractionator off-gas				Stripper off-gas				Material balance		
	Flow	Concentrations			Flow	Concentrations			Flow	Concentrations			Flow	Concentrations					
	rate (std cm^3/s)[a]	O_2 (%)	CO (%)	Kr (ppb)	rate (std cm^3/s)[a]	O_2 (%)	CO (%)	Kr (ppb)	rate (std cm^3/s)[a]	O_2 (%)	CO (%)	Kr (ppb)	rate (std cm^3/s)[a]	O_2 (%)	CO (%)	Kr (ppb)	O_2 (%)	CO (%)	Kr (%)
C4-377	2690	10.3	2.9	7.4	1369	7.6	1.8	0.02	1227	12.4	4.2	10.7	33	1.1	0.1	181.0	92	97	96
C4-386	2690	9.6	2.6	5.1	1321	7.5	1.6	0.01	1227	11.6	3.9	6.8	24	0.7	<0.1	192.7	94	99	95
C4-394	2831	9.4	3.4	4.7	1510	7.4	2.2	0.02	1227	10.6	4.8	7.0	25	0.2	<0.1	200.1	90	95	99
C4-402	2548	9.6	1.8	4.2	1227	7.3	0.7	0.01	1180	11.6	3.0	5.1	14	5.1	4.3	272.5	83	97	93
C4-411	2737	9.0	2.4	3.4	1321	8.0	1.4	0.01	1321	10.4	3.4	4.1	9	4.2	2.8	339.8	98	97	92
C4-420	2407	8.0	1.0	3.2	1085	5.9	0.2	0.003	1227	9.1	2.2	7.4	14	6.5	6.9	1345.2	92	121	97
C4-428	2501	7.1	2.0	1.8	1130	6.0	1.0	0.002	1227	9.1	3.0	2.3	5	6.5	7.0	252.7	102	98	102
C4-437	2360	9.5	1.6	0.6	1035	5.4	0.4	0.0004	1180	13.1	2.9	0.8	9	0.2	<0.1	39.5	96	104	95
C4-444	2312	6.6	0.5	0.1	1038	4.3	0.1	0.0005	1180	8.9	1.0	0.1	1	8.1	1.4	12.4	99	112	68
C4-454	2501	10.6	1.8	4.8	1227	5.5	0.6	0.03	1180	10.2	3.0	5.9	5	5.5	5.0	559.0	71	98	90
C4-465	2360	7.1	1.3	3.0	1035	0.5	<0.1	0.001	1180	10.1	2.8	3.4	9	8.7	5.7	365.4	78	111	94
C4-475	2595	9.7	1.2	18.2	1130	5.1	0.2	0.005	1180	13.5	2.4	19.6	140	16.9	4.7	210.0	98	118	103

[a] STP = 0.1 MPa, 294.3 K.

Table B.2. Equivalent pump rate for CO/O_2 three-column solubility experiments

Experiment No.	Temperature (K)	Pump rate (cm^3/s)	Pump rate (std cm^3/s)[a]
C4-377	244.4	26.2	15,273
C4-386	244.6	26.2	15,259
C4-394	244.5	26.2	15,266
C4-402	244.0	26.2	15,299
C4-411	244.5	26.2	15,266
C4-420	244.7	26.2	15,253
C4-428	244.4	26.2	15,273
C4-437	244.7	26.2	15,253
C4-444	243.7	26.2	15,318
C4-454	244.2	26.2	15,286
C4-465	245.5	39.3	22,801
C4-475	244.9	39.3	22,861

[a] STP = 0.1 MPa and 294.3 K.

Table B.3. Boilup rates for three-column O_2/CO solubility experiments

	Fractionator reboiler			Stripper reboiler		
Experiment No.	Temperature (K)	Boilup (kW)	Boilup (std cm^3/s)[a]	Temperature (K)	Boilup (kW)	Boilup (std cm^3/s)[a]
C4-377	254.1	0.6412	1241	250.6	2.37	4474
C4-386	254.0	0.6414	1241	250.6	2.43	4601
C4-394	253.9	0.5870	1137	250.5	2.43	4601
C4-402	253.9	0.5403	1048	250.6	2.70	5111
C4-411	253.9	0.5642	1095	250.6	2.64	4988
C4-420	253.9	0.5317	1029	250.7	2.60	4931
C4-428	253.9	0.553	1076	250.6	2.75	5205
C4-437	253.9	0.558	1076	250.7	2.72	5153
C4-444	253.7	0.4890	944	250.5	2.83	5356
C4-454	253.9	0.5446	1057	250.7	2.80	5309
C4-465	254.0	0.8519	1652	250.8	3.72	7050
C4-475	253.5	0.6849	1321	250.7	4.09	7734

[a] STP = 0.1 MPa, 294.3 K.

Thus

$$Q_{15} = Q_5 + Q_3 \tag{B.2}$$

and

$$x_{15} = \frac{Q_5 x_5 + Q_3 y_3}{Q_{15}} \, . \tag{B.3}$$

Similarly, an overall and component mass balance around the fractionator determines the liquid feed to the fractionator.

$$Q_4 = Q_{10} + Q_{15} \tag{B.4}$$

and

$$x_4 = \frac{Q_{10} y_{10} + Q_{15} x_{15}}{Q_4} \, , \tag{B.5}$$

where

Q_4 = flow rate of stream 4, std cm^3/s, and

x_4 = stream 4 composition of component i.

The results of these calculations are shown in Table B.4. Utilizing this information, an overall fractionator decontamination factor can be calculated:

$$DF_o = \frac{Q_4 x_4}{Q_{15} x_{15}} \, , \tag{B.6}$$

where

DF_o = overall decontamination factor of component i,

y_4 = stream 4 concentration of component i, and

x_{15} = stream 15 concentration of component i.

Table B.4. Fractionator and stripper feed determination for three-column O_2/CO solubility experiments

	Fractionator liquid feed				Stripper liquid feed			
		Composition				Composition		
Experiment number	Flow rate (std cm^3/s)[a]	O_2 (%)	CO (%)	Kr (ppb)	Flow rate (std cm^3/s)[a]	O_2 (%)	CO (%)	Kr (ppb)
C4-377	16,611	0.91	0.31	1.15	15,384	0.002	<0.0002	0.39
C4-386	16,611	0.86	0.29	0.78	15,384	0.001	<0.0002	0.29
C4-394	16,564	0.77	0.35	0.80	15,384	0.0002	<0.0002	0.32
C4-402	16,611	0.83	0.22	0.61	15,431	0.0048	0.004	0.26
C4-411	16,705	0.83	0.27	0.51	15,384	0.0025	0.0017	0.20
C4-420	16,564	0.67	0.16	0.45	15,337	0.0052	0.0055	0.19
C4-428	16,611	0.67	0.22	0.27	15,384	0.0028	0.003	0.11
C4-437	16,517	0.92	0.20	0.08	15,337	0.0001	<0.0002	0.02
C4-444	16,564	0.64	0.07	0.01	15,384	0.0006	<0.0002	0.001
C4-454	16,564	0.74	0.22	0.65	15,384	0.0024	0.0022	0.24
C4-465	24,067	0.50	0.14	0.28	22,887	0.0028	0.0018	0.12
C4-475	24,256	0.75	0.14	2.00	23,029	0.087	0.024	1.09

[a] STP = 0.1 MPa, 294.3 K.

The two-column pinched data are tabulated in Table B.5. Since the data of interest were in the internal stripper column concentrations, such factors as overall gas balance, equivalent pump rate, and column liquid-to-vapor ratios were not calculated. It should be noted that sample 19 is the top stripper column sample point and sample 16 is one-third of the way down the column (∿1.17 m).

6.3 Appendix C: Flammability of CO-O_2 Mixtures

In the presence of oxygen, certain concentrations of CO form flammable mixtures. Sources of potential combustion in a typical KALC facility are sparks from electrical equipment, heat generated by the compressor, a fire in the facility, and heat generated by the fission products. The flammable limits of CO in air at 0.1 MPa and room temperature are 12.5 to 74 vol % CO with an autoignition temperature of 883.2 K.[20] The range of conditions in a KALC-type facility will cover temperatures from >373 through 233 K, pressures up to 4 MPa, and a varying mixture of CO_2-O_2-CO-N_2. Of interest are the effects on CO flammable limits of pressure, temperature, and system composition.

Experiments conducted on the flammability limits of CO in air and pure oxygen indicate that although O_2 composition greatly effects the upper limit, it changes the lower limit only ∿10%.[21] Thus the changing O_2 content in the KALC system should have little effect on the lower flammability limit of 12.5% CO.

White[22] studied the effect of temperature on flammability limits and found that raising the temperature of a CO-air mixture from 293 to 373 K reduced the lower flammability limit by 10%. This would indicate

Table B.5. Summary of pinched experiments

Experiment No.	Pump rate (cm^3/s)	Reboiler heat load (kW)	Column pressure (MPa)	Top column temperature (K)	1 down from top column temperature (K)	O_2 concentration (%) Sample 19	O_2 concentration (%) Sample 16	CO concentration (%) Sample 19	CO concentration (%) Sample 16	N_2 concentration (%) Sample 19	N_2 concentration (%) Sample 16	Pinched[a] O_2	Pinched[a] CO	Pinched[a] N_2
C4-504	39.3	0.78	2.05	252.2	252.2	5.8	5.3	1.6	1.6	0	0	?	Y	NP
C4-507	39.3	0.78	2.05	252.6	252.6	5.3	5.2	1.1	1.3	0	0	Y	Y	NP
C4-526B	39.3	0.75	2.04	250.0	250.0	6.6	6.7	3.1	3.2	0	0	Y	Y	NP
C4-538	39.3	0.77	2.03	250.2	250.2	6.7	6.5	2.9	2.8	0	0	Y	Y	NP
C4-544	39.3	0.78	2.03	250.7	250.8	4.4	3.9	2.8	2.8	0	0	N	Y	NP
C4-552	39.3	0.85	2.03	250.4	250.6	3.2	2.6	3.2	3.0	0	0	N	Y	NP
C4-558	39.3	0.87	2.03	250.0	250.7	6.6	3.9	4.0	2.1	0	0	N	N	NP
C4-564	39.3	0.79	1.83	248.3	249.2	2.0	0.5	2.8	0.4	0	0	N	N	NP
C4-570	39.3	0.72	1.83	247.8	248.2	4.0	1.9	2.2	1.2	0	0	N	N	NP
C4-576	39.3	0.67	1.83	247.0	247.1	5.2	4.1	2.6	2.4	0	0	N	Y	NP
C4-584	39.3	0.65	1.83	247.5	247.5	6.2	4.6	2.4	2.4	0	0	N	Y	NP
C4-606	39.3	0.57	1.83	247.2	247.2	2.6	2.6	3.2	3.1	0	0	Y	Y	NP
C4-612	39.3	0.63	1.82	246.9	246.9	3.8	3.8	3.1	3.0	0	0	N	N	NP
C4-634	39.3	0.86	2.03	250.3	250.6	1.3	0.5	2.5	1.7					
C4-652	39.3	0.80	2.03	249.6	249.8	1.6	1.0	3.5	3.5	2.1	0.5	N	Y	N
C4-658	39.3	0.81	2.03	250.2	250.4	1.4	1.1	3.5	3.5	1.6	0.6	N	Y	N
C4-664	39.3	0.75	2.03	249.9	250.1	3.7	3.9	1.8	2.2	1.3	0.7	Y	Y	N
C4-668	39.3	0.71	2.03	250.5	250.7	1.8	1.5	0	0	2.4	1.5	?	NP	N
C4-672	39.3	0.72	2.03	249.5	249.6	1.8	1.8	0	0	2.3	1.9	Y	NP	N
C4-678	39.3	0.76	2.03	249.8	249.9	3.4	3.4	0	0	2.6	2.0	Y	NP	N
C4-682	39.3	0.80	2.03	250.2	250.3	5.2	4.0	0	0	4.2	1.3	N	NP	N
C4-687	39.3	0.69	2.03	249.2	249.3	2.8	2.9	0	0	2.4	1.9	Y	NP	N
C4-694	39.3	0.40	2.03	249.4	249.3	2.8	2.8	0	0	2.4	2.3	Y	NP	Y
C4-712	39.3	0.62	2.03	249.1	249.0	0	0	2.4	2.4	2.4	2.3	NP	Y	Y
C4-716	39.3	0.68	2.03	249.6	249.7	0	0	2.9	2.7	3.2	2.3	NP	Y	N
C4-720	39.3	0.80	2.03	249.7	251.2	0	0	2.6	3.0	2.8	0.9	NP	Y	N
C4-724	39.3	0.83	2.03	251.1	252.8	0	0	2.5	0.7	2.7	0.2	NP	N	N
C4-728	39.3	0.79	2.03	250.3	251.9	0	0	3.4	1.8	3.0	0.2	NP	N	N
C4-732	39.3	0.73	2.03	249.7	250.2	0	0	1.7	1.7	2.2	0.8	NP	Y	N

[a] Y = component pinched;
N = component not pinched;
NP = component not present.

that at the near-cryogenic operating conditions of KALC (nominally 253 K) the lower flammability limit would be somewhat higher than 12.5%, but at the elevated compressor outlet temperatures (>373 K during Campaign IV experiments), the lower limit would be slightly less than 12.5%. However, Bone[23] discovered that raising the pressure of a CO-air mixture from 0.1 to 1 MPa increased the lower limit by 20%. This indicates that in KALC operations the high operating pressures will outweigh the temperature effects and cause the lower flammability limit to be somewhat higher than 12.5% CO. No published data were found on the flammability limit effects of simultaneously varying composition, temperature, and pressure. Consequently, the effects may not be additive.

6.4 Appendix D: Equipment Performance Through Campaign IV

6.4.1 Refrigeration units

No component failures occurred during Campaign IV. Units A and B require constant monitoring and/or maintenance because of refrigerant losses through leaks. Substantial time is required to obtain proper temperatures when operating parameters are changed (i.e., light-gas concentration, pressure and/or flow alterations).

Unit A has a total of 28,835 operating hr, with 11,972 hr accumulated during Campaign IV. Unit B has a total of ∿29,000 operating hr,* with ∿12,000 hr accumulated during Campaign IV. Unit C has a total of 9300 operating hr, all accumulated during Campaign IV.

6.4.2 Liquid CO_2 pump

This pump has a total of 6189 operating hr with 3184 hr accumulated during Campaign IV. During this campaign, the pump piston loosened from

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*Timer failed at some unknown time during Campaign IV.

its drive rod (which effectively lengthened the stroke), forcing the end into the diaphragm "back-up" plate which caused the plate to fracture. This necessitated the replacement of both the diaphragm and the back-up plate.

The pump heads require external refrigeration to maintain a temperature of 233 K, and the suction pressure must not be less than 1.7 MPa to prevent gasification of liquid CO_2. The pump has also proved to be unreliable at flow rates <25 cm^3/s.

6.4.3 Gas compressors

The main compressor has a total of 8007 operating hr with 5151 hr accumulated during Campaign IV.

The auxiliary compressor has a total of 2105 operating hr, with 1713 hr accumulated during Campaign IV.

Early in Campaign IV (with ∿6850 total operating hr), the main compressor hydraulic system failed. Upon investigation, it was discovered that the oil pump was not operating due to a failure in the metal coupling of the crank shaft and the oil pump. Inspection of the auxiliary compressor crank shaft also displayed the same failure (see Fig. D.1).

It was surmised that because of the very close tolerances of the coupling, undue stress was applied to the "female" connector on the crank shaft; when the "male-end" oil pump shaft was pressed into the crank shaft, it caused complete metal fatigue under continued operating loads. The female connector was replaced by a shaft without the slot, which appeared to solve the problem.

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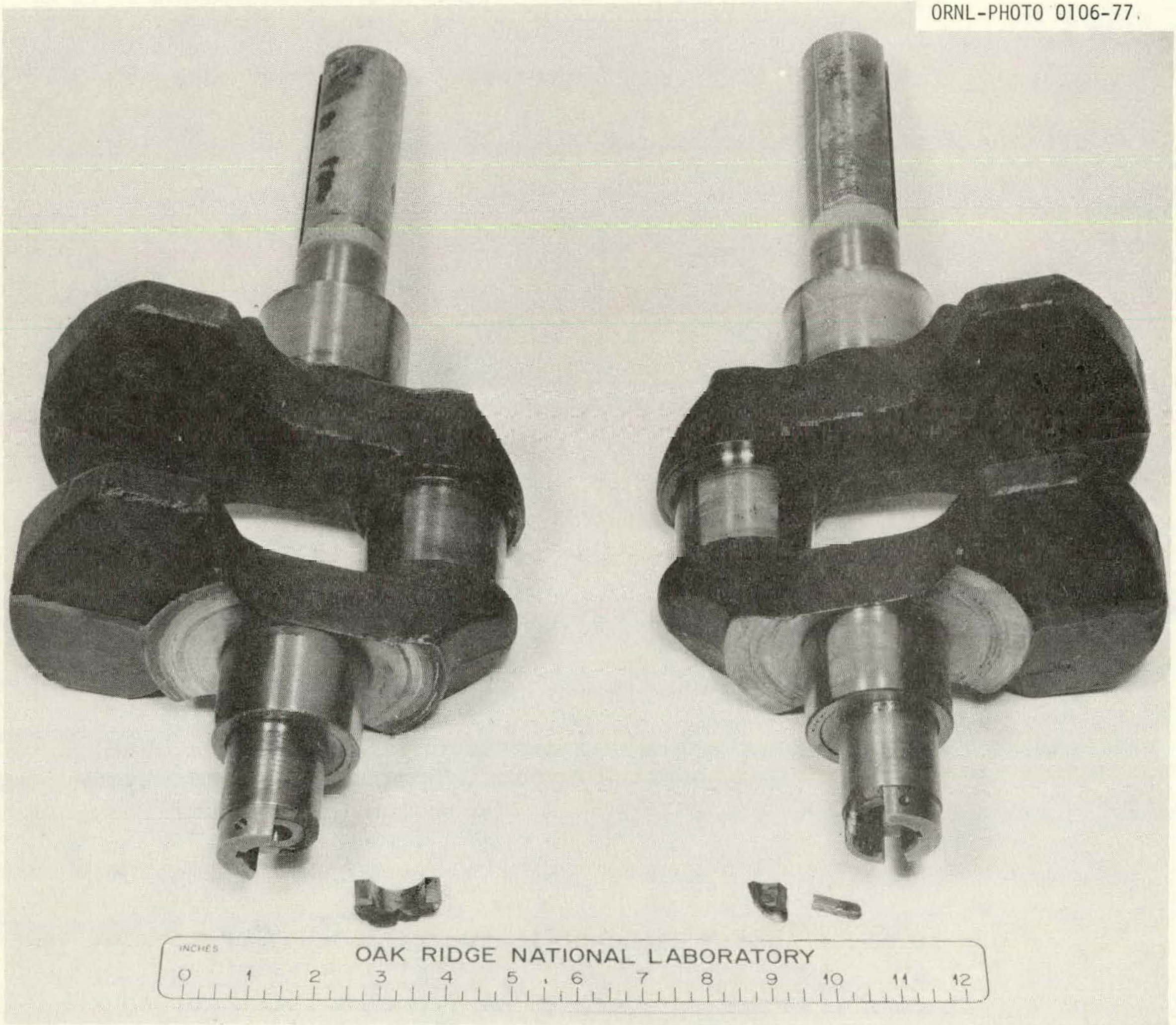

Fig. D.1. Main and auxiliary (left to right) crankshafts showing the failed couplings.

6.5 Appendix E: Chronology of KALC Campaign IV

1976

6-25 through 7-12	The system facility was being prepared for Campaign IV as follows: (1) servicing of all heavy equipment; (2) recalibration of recorders, controllers, etc.; (3) addition of piping and valves for future installation of a third column (fractionator); and (4) recharging system with CO_2 and O_2.
7-13	Start system for total system shakedown.
7-14	Corrected minor leaks. System started. Pump-head cooler kicked out causing system shutdown. Recharged pump-head cooler with refrigerant.
7-15 through 7-23	System operated daily. Fine-tuning of all condenser valves necessary to obtain proper system temperatures and pressures.
7-26	Added O_2 and 0.5 Ci of ^{85}Kr to system.
7-27	Experiment C4-5.
7-28	Refrigeration control adjustments necessary on feed gas cooler.
7-29	Experiment C4-12.
7-30	Refrigeration unit "B" kicked off because of excessive R-502 on suction heads. "Trimmed" thermal expansion valve on feed gas cooler.
8-2	Pump-head cooler off. Leaking seals on circulating pump.
8-3	Recirculating pump (seals) repaired. Seals leaked again after $\sim$2 hr of operation. Installed new thermal expansion valve (TXV) on feed gas cooler.
8-4	Replaced seals in recirculating pump on pump-head cooler unit. Bus-bar in electrical panel B-1 became loose and arced open.
8-5	Repaired electrical panel B-1.
8-6	Started system for testing and/or tuning of new TXV on feed-gas cooler, pump-head cooler forced shutdown; suspected a broken valve plate.
8-9	Valve plate on pump-head cooler O.K.; found blown head gasket and repaired it.

1976

8-10 through 8-13	System components operating properly except continued trimming of refrigeration valves was necessary.
8-16	Experiment C4-22.
8-17	Refrigeration adjustments necessary.
8-18	Experiment C4-29.
8-19	Experiment C4-35.
8-20	Experiment C4-41.
8-23	Refrigeration adjustments.
8-24	Continued refrigeration adjustments.
8-25	Experiment C4-48.
8-27	Stripper column flooding studies.
8-30	Experiment C4-56.
8-31	Experiment C4-63.
9-1	Experiment C4-69.
9-2	Experiment C4-75.
9-7	Refilled system with CO_2 and added O_2 and N_2.
9-8	Experiment C4-81.
9-9	Experiment C4-87.
9-13	Added CO_2 and 0.5 Ci of ^{85}Kr to system. Tuned the main condenser refrigeration valves in order to operate the stripper column at reduced pressure (1.8 MPa).
9-14	Experiment C4-94.
9-15	Experiment C4-100.
9-16	Experiment C4-106.
9-17 through 9-20	Reinsulated various components. Completed installation of a third column (fractionator).
9-21	Experiment C4-112.

1976

9-22	Adjustments in refrigeration controls necessary. Added O_2 and N_2 to system.
9-23	Experiment C4-118.
9-27	Experiment C4-124.
9-28	Corrected minor valve leaks. Adjusted refrigeration controls.
9-29	Experiment C4-130.
10-1	Experiment C4-135.
10-4	Experiment C4-139.
10-5	Experiment C4-143.
10-7	Added O_2 and N_2 to system.
10-8	Experiment C4-148.
10-12	Recalibrated gas chromatograph.
10-13 through 10-21	Third column (fractionator) valved into system. Correction of minor leaks. Adjustments made on fractionator condenser refrigeration controls. Calibrated liquid-level probes.
10-22 through 10-28	Fractionator column flooding studies.
11-2 through 11-3	Corrected leaking feed valves to fractionator column.
11-4	Experiment C4-171.
11-5	System emptied of liquid CO_2 and shutdown for scheduled electrical outage.
11-8	System refilled with CO_2.
11-9	Recheck of fractionator flooding data.
11-10	Experiment C4-181.
11-11	Experiment C4-188.
11-12	Experiment C4-197.

1976	
11-15	Set system for two-column operation. Refrigeration adjustments necessary.
11-16	Experiment C4-206.
11-17	Experiment C4-214.
11-18	Experiment C4-221.
11-19	Experiment C4-228.
11-22	Refrigeration adjustments necessary.
11-23	Experiment C4-237.
11-29	Experiment C4-244.
11-30	Reset system for three-column operation. Refrigeration adjustments necessary.
12-1	Experiment C4-251.
12-2	Experiment C4-259.
12-3	Experiment C4-266.
12-6	Experiment C4-273.
12-7 through 12-9	Reset system for two-column operation with necessary refrigeration adjustments. Minor piping changes made on analytical instrumentation.
12-10	Experiment C4-304.
12-13	Refrigeration adjustments necessary.
12-14	Experiment C4-310.
12-15	Refrigeration adjustments necessary.
12-16	Experiment C4-316.
12-17	Experiment C4-323.
12-20	Experiment C4-329.
12-21	Gas compressor malfunctioned due to broken crank shaft. Purged CO from system.
12-27	Fractionator column flooding studies.

1976

12-28	Fractionator column flooding studies.

1977

1-3	Refrigeration adjustments necessary.
1-5 through 1-6	Fractionator column flooding studies. Main gas compresser repairs completed.
1-7 through 1-10	Data analyses.
1-11 through 1-17	Refrigeration adjustments necessary. Minor leaks at various valves corrected.
1-18	Experiment C4-348.
1-19	Experiment C4-356.
1-21 through 1-25	Refrigeration adjustments necessary. Minor leaks at various valves corrected.
1-26	Experiment C4-365.
1-27 through 2-8	Replaced leaking bellows in two research control valves. Corrected other minor system leaks. Refrigeration adjustments necessary.
2-9	Experiment C4-377.
2-10	Experiment C4-386.
2-11	Experiment C4-394.
2-13	Serviced various items of heavy equipment.
2-14	Experiment C4-402.
2-15	Experiment C4-411.
2-16	Experiment C4-420.
2-17	Experiment C4-428.
2-18	Removed rotameters from system for calibration.
2-28	Rotameters reinstalled in system. Refrigeration adjustments necessary.

1977

3-3 through 3-7	Refrigeration adjustments necessary.
3-8	Experiment C4-437.
3-9	Experiment C4-444.
3-10	Experiment C4-454.
3-11	Experiment C4-465.
3-14	Experiment C4-475.
3-15	Experiments C4-487 and C4-493.
3-16	Gas chromatograph calibrated.
3-17	Experiment C4-502.
3-18	Experiment C4-504.
3-21	Experiment C4-507.
3-22	Recalibrated stripper reboiler heater volt and ampere indicators.
3-23 through 3-24	Purged system of all light gases in preparation for CO_2-O_2 experiments.
3-25	Experiment C4-512.
3-28	Experiment C4-516.
3-29	Experiment C4-518.
3-30	Experiment C4-522.
3-31	Experiment C4-524.
4-4 through 4-5	Refrigeration adjustments necessary.
4-6	Experiment C4-526.
4-7	Experiment C4-538.
4-11 through 4-12	Refrigeration adjustments necessary.

<u>1977</u>

4-13	Experiment C4-544.
4-14 through 4-19	Refrigeration adjustments necessary. Repairs made on mass spectrometer.
4-20	Experiment C4-552.
4-21	Experiment C4-558.
4-22	Refrigeration adjustments necessary.
4-25	Experiment C4-564.
4-26	Calibrated mass spectrometer.
4-27	Experiment C4-570.
4-28	Experiment C4-576.
4-29	Experiment C4-582.
5-2	Refrigeration adjustments necessary.
5-3	Experiment C4-588.
5-4	Experiment C4-594.
5-5	Experiment C4-598.
5-6	Experiment C4-602.
5-9	Experiment C4-603.
5-10	Experiment C4-606.
5-11	Experiment C4-612.
5-12	Experiment C4-618.
5-13	Experiment C4-623.
5-16	Refrigeration adjustments necessary. Recharge system with CO_2.
5-17	Experiment C4-628.
5-18 through 5-19	Refrigeration adjustments necessary.

1977	
5-20	Experiment C4-634.
5-23	Experiment C4-640.
5-24	Experiment C4-646.
5-25	Experiment C4-652.
5-26	Experiment C4-658.
5-27	Experiment C4-664.
5-31	System purged to remove CO. Recharge with CO_2, O_2, N_2, and ^{85}Kr.
6-1	Experiment C4-668.
6-2	Experiment C4-672.
6-3	Experiment C4-678.
6-6	Experiment C4-682.
6-7	Experiment C4-687.
6-8	Partial plug in pressure control valve in suction line to the main gas compressor. Cleaned and back in service.
6-9	Experiment C4-690.
6-10	Experiment C4-694.
6-13	System purged to remove O_2 from system. Recharged with N_2.
6-14	Experiment C4-699.
6-15	Experiment C4-704.
6-16	Refrigeration adjustments necessary. CO added to system.
6-17	Experiment C4-708.
6-20	Add R-502 to refrigration unit B to bring unit to full charge.
6-21	Experiment C4-712.
6-22	Experiment C4-716.

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6-23	Refrigeration adjustments necessary.
6-24	Experiment C4-720.
6-27	Experiment C4-724.
6-28	Experiment C4-728.
6-29	Experiment C4-732.
6-30	Refrigeration adjustments necessary.
7-1	Experiment C4-736.
7-5	Purged system and charged with CO_2 and CO.
7-6	Experiment C4-740.
7-7	Experiment C4-744.
7-8	Experiment C4-748.
7-11	Experiment C4-752.
7-12	Experiment C4-756.
7-13	Experiment C4-760.
7-14	Experiment C4-764.
7-15	Experiment C4-768.
7-18	Experiment C4-772.
7-19	Experiment C4-776.
7-20 through 7-21	Repairs necessary on temperature recorder.
7-22	Experiment C4-780.
7-25	Experiment C4-784.
7-26	Experiment C4-785.
7-27	Purged system and added O_2 and CO_2.
7-28	Refrigeration adjustments necessary.
7-29	Experiment C4-792.

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8-1	Refrigeration adjustments necessary.
8-2	Experiment C4-800.
8-3	Experiment C4-804.
8-4	Replaced trim in two research control valves.
8-5 through 8-17	All refrigeration units recharged with R-502, and necessary refrigeration adjustments made.
8-18	Experiment C4-816.
8-19	Refrigeration adjustments necessary.
8-22 through 8-23	Experiment C4-828.
8-24 through 8-26	System purged to remove ^{85}Kr and added ^{133}Xe.
8-29 through 8-30	Experiment C4-838 and C4-845.
8-31 through 9-1	Experiment C4-852 and C4-859.
9-2	Refrigeration adjustments necessary. Shutdown for vacations.
9-8 through 9-13	Refrigeration adjustments necessary.
9-14	Experiment C4-869.
9-15	Experiment- C4-881 and C4-891.
9-16	Experiments C4-901, C4-906, and C4-918.
9-19	Experiment C4-924.
9-20	Experiments C4-939 and C4-948.
9-21	Experiments C4-958, C4-968, and C4-977.
9-22 through 9-25	Refrigeration adjustments necessary.
9-26	Experiments C4-986 and C4-997.

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9-27	Refrigeration adjustments necessary.
9-28	Experiment C4-1006.
9-29	Refrigeration adjustments necessary.
9-30	Experiments C4-1016 and C4-1026.
10-3 through 10-4	Minor piping modifications made. Changed trim in a research control valve. Reinsulation of some piping.
10-5 through 10-7	Refrigeration adjustments necessary.
10-10	Experiment C4-1039.
10-11	Experiment C4-1048.
10-12	Refrigeration adjustments necessary.
10-13	Experiment C4-1055.
10-14	Experiment C4-1062.
10-17	Refrigeration adjustments necessary.
10-18	Experiment C4-1069.
10-19	Experiment C4-1078.
10-20	Experiment C4-1086.
10-21	System purged of ^{133}Xe. Krypton-85 added.
10-24	Experiment C4-1094.
10-25	Experiment C4-1102.
10-26	Experiment C4-1108.
10-27	Experiment C4-1114.
10-28	Refrigeration adjustments necessary.
10-31	Experiment C4-1120.
11-1	Experiment C4-1128.
11-2	Experiment C4-1134.

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11-3	Experiment C4-1140.
11-4	Refrigeration adjustments necessary.
11-7	Experiment C4-1146.
11-8	Experiment C4-1154.
11-9	Experiment C4-1160.
11-10	Experiment C4-1166.
11-17	Experiment C4-1202.
11-18	Refrigeration adjustments necessary.
11-21	Experiment C4-1212.
11-22	Experiment C4-1222.
11-28 through 12-2	Refrigeration adjustments necessary. Replaced back-up plates and diaphragm in liquid solvent pump.
12-5	Experiment C4-1236.
12-6	Experiment C4-1246.
12-7	Experiments C4-1254 and C4-1264.
12-8	Experiment C4-1272.
12-9	Experiments C4-1280 and C4-1290.
12-12	Experiments C4-1300 and C4-1310.
12-13	Experiments C4-1318 and C4-1326.
12-14 through 12-15	Refrigeration adjustments necessary.
12-16	Experiment C4-1334.
12-19	System shutdown in a standby condition.

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