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FILE DESCRIPTION

PHILADELPHIA FILE

SUBJECT HARRY GOLD

FILE NO. 65-4307

VOLUME NO. 1-B

SERIALS 1

thru

17

NOTICE

THE BEST COPIES OBTAINABLE ARE INCLUDED IN THE REPRODUCTION OF THE FILE. PAGES INCLUDED THAT ARE BLURRED, LIGHT OR OTHERWISE DIFFICULT TO READ ARE THE RESULT OF THE CONDITION AND OR COLOR OF THE ORIGINALS PROVIDED. THESE ARE THE BEST COPIES AVAILABLE.

Inventory Worksheet
FD-503 (2-18-77)

VOLUME 1-B

PHILADELPHIA FILES

INVENTORIED BY [illegible]

REVIEWED BY

File No: 65-4207

Re: Harry Gold

Date: 5/78 (month/year)

Serial	Date	Description (Type of communication, to, from)	No. of Pages Actual	No. of Pages Released	Exemptions used or, to whom referred (Identify statute if (b)(3) cited)
		Bulky Exhibit Inventory	1	1	
		Envelope describing contents of 1B(1)	1	1	
1B(1)	5-25-50	Bank Records (photostatic) pertaining to Harry Gold	36	36	
		Envelope describing contents of 1B(2)	1	1	
1B(2)	5-25-50	Bank Records (photostatic) pertaining to Harry Gold	8	8	
		Envelope describing contents of 1B(3)	1	1	
1B(3)	5-50	Two photos of Harry Gold w/ note	3	3	
		Envelope describing contents of 1B(4)	1	1	
1B(4)	5-22-[illegible]	Waiver of search of premises 6823 Kindred signed Harry Gold	1	1	
		Envelope describing contents of 1B(5)	1	1	
1B(5)	5-25-50	Three lists of students taking classes with Harry Gold	3	3	
		Envelope describing contents of 1B(6)	1	1	

FBI/DOJ

Inventory Worksheet
FD-503 (2-18-77)

VOLUME 1-B

PHILADELPHIA FILES

INVENTORIED BY [illegible]

REVIEWED BY _____

File No: 65-4307

Re: Harry Gold

Date: 5/78 (month/year)

Serial	Date	Description (Type of communication, to, from)	No. of Pages Actual	No. of Pages Released	Exemptions used or, to whom referred (Identify statute if (b)(3) cited)
1B(6)	—	Surveillance notes of SA's of Harry Gold, 5/11, 15, 16, 18, 19, 20, 21, 22/50	7	7	
		Envelope describing contents of 1B(7)	1	1	
1B(7)	5/12-21/50	Surveillance notes of SA's of Harry Gold, 5/10, 12, 15, 17, 18, 19, 20, 21/50	5	5	
		Envelope describing contents of 1B(8)	1	1	
1B(8)	5-29-50	Photostatic copies of articles req. for photostating by Harry Gold from Frank Gates	49	49	
		Envelope describing contents of 1B(9)	1	1	
1B(9)	5-29-50	Two photos of Leonid Korzarovich Kvasnikov front & back	1	1	
		Envelope describing contents of 1B(10)	1	1	
1B(10)	5-29-50	Two photos of Raymond Baker aka, Raph Bourrier front & back	1	1	
		Envelope describing contents of 1B(11)	1	1	
1B(11)	5-29-50	Two photos of Vladimir Nikolaevich Maguin front & back	1	1	
		Envelope describing contents of 1B(12)	1	1	

FBI/DOJ

Inventory Worksheet
FD-503 (2-18-77)

VOLUME 1-B

PHILADELPHIA FILES

INVENTORIED BY EGS

REVIEWED BY

Date: 5/78 (month/year)

File No: 65-4307

Re: Harry Gold

Serial	Date	Description (Type of communication, to, from)	No. of Pages Actual	No. of Pages Released	Exemptions used or, to whom referred (Identify statute if (b)(3) cited)
1B(12)	5-29-50	Photo of Mikhail D. Mitrofanov front + back	1	1	
		Envelope describing contents of 1B(13)	1	1	
1B(13)	5-29-50	Four photos of Pavel Petrovich Mikhailov front + back	1	1	
		Envelope describing contents of 1B(14)	1	1	
1B(14)	5-26-50	Photostatic copies of 18 checks signed by Harry Gold for his dist at Real Estate Trust Co.	8	8	Best copies available
1B(15)		Exhibit destroyed 5/3/67	—	—	refer to Bulky Exhibit Inventory
		Envelope describing contents of 1B(16)	1	1	
1B(16)	5-29-50	Advertisement Handbill for Storm Windows	1	1	
		Envelope describing contents of 1B(17)	1	1	
1B(17)	5-26-50	Photostat of original advance in salary by Pa. Alcohol + Chem Corp to Harry Gold	2	2	1 pg. - Dup

FBI/DOJ

Retained 6/27/61 aa

HARRY GOLD, WAS.
ESPIONAGE - R
65-4328-4307

THIS MATERIAL CAN BE LOCATED IN THE BULKY EXHIBIT ROOM

1. Bank records (photostatic copies) pertaining to HARRY GOLD. (Corn Exchange) (RE: Sec 1)
2. Photostatic copies of Bank records pertaining to HARRY GOLD. (PSFS) (RE: Sec 1)
3. Two photos of Harry Gold -
4. Written statement of [illegible] signed by Harry Gold
5. Photo of statute they dined with Harry Gold (see 11)
6. Surveillance note of SA Joseph A Kelly & SA Thomas Baker (see 89)
7. Note re surveillance Harry Gold as indicated (see 89)
8. Photostatic copies of letters req for photostating by Harry Gold at Franklin Institute Library
9. Two copies of photo of David Rosenovitz Greenglass
10. 2 photos of Raymond Peter Ian Rafael Boman (see
11. Two photos of Vladimir Petrovich Mazurin (see
12. Photo of Mikhail Grigorievich Molotkov (see 88)
13. Four photo of Pavel Pavlovich Mikhailov (see
14. Photostatic copies of 18 checks signed by Harry Gold for his acct at Re Real Estate Trust Co
15. Two copies of photographs shown to Harry Gold as of 5/29/50 (copy not in "B" 50
16. Advertisement Yardbird for Storm Windows (see 20)
17. Photostat copy of original document reflecting advance in salary by Am Alcohol & Chemical Corp to Harry Gold on 2-6-42

destroyed 5/3/67 es

~~and returned to exhibit room~~

Return 8/13/68 es

Return 11/18/58 es

Return 1-18/59 NCH

Return 5/3/67 es

65-4307-1-B
~~65-4328-1-B~~

FBI - PHILADELPHIA
MAY 25 1950

PE-SER 7

5/25/50

From J. ROLAND RHODES
(Name of Contributor)

CORN EXCHANGE NAT'L BANK, 20 CHESTNUT STS
(Address of Contributor)

By R. B. Lenard
(Name of Special Agent)

To Be Returned: Yes ( ) No (✓)

Description: Bank records (photostatic copies) pertaining to Harry Bell

File No. ~~65-4328-1B~~ 65-4307-1-B (1)

Photo file

Photostatic copies of Bank records pertaining to Harry Gold

Obtained on 5/25/50 by SA Robert B. Lockard

65-4307 1-B (1)

25 61687

CO-MAKER'S STATEMENT

Application No. 2-4711 Date 10/10/67

TO CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

Name of Co-maker(s)

I have agreed to become a Co-maker, with said applicant(s) for a personal loan of $ 225.00 payable in the sum of $ 12.

I have signed as a Co-maker on a promissory note which will evidence the loan if granted and I understand that by signing the note I have made myself liable for the payment thereof. In consideration of your so making the loan I make the following representations:

Print Name: MORRELL E. DOUGHERTY Age: 45 Marital Status: M Relation to Applicant(s): FRIEND

Home Address: 6 S. DUKE HEFIELD ST How Long: 10 YRS Home Phone Number: SA 7-05 39

Business Address: 4211 SPRINGFIELD AV How Long: 4 YRS How Long Have Known Applicant(s): 18 YRS

Employer: ASHESON COLLOID CORP How Long: 4 YRS Position or Trade: ENGINEER

Bank Address: 455 LAND TITLE BLDG Monthly Income: $ 500 Phone: LO 9-7113

Name of Co-maker's Bank: CORN EXCHANGE 15TH & CHESTNUT STS Savings: Special Check: Checking: Special Check:

Credit References: Name: CORN EXCHANGE Address: 15TH & CHESTNUT STS Name: ARROW STORE Address: 119 MARKET ST

List all Real Estate owned: None Mortgages $

List all automobiles owned: PLYMOUTH Make and Year 1948

Have you borrowed with your co-maker? Yes ☐ No ☐

I hereby swear or affirm that each of the above statements is true and correct.

SIGNATURE: Morrell E. Dougherty

Any connected with the proposed loan, a copy of the note, mortgage, or other instrument evidencing the loan will be furnished upon request.

Date of Application: 10/10/67

For Bank Use Only

	Check No.	Amount	Balance	Date		Check No.	Amount	Balance	Date
Appl.									
Appl.									
Appl.									

Remarks: Sent Morrell Dougherty W/ Sandra and signature cards to Mr. Thompson 11-6-67

CO-MAKER'S STATEMENT

Application No. [illegible] Date [illegible]

[illegible]/CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

[illegible] Applicant(s) [illegible] a personal loan of [illegible] for a term of [illegible] months.

[illegible] I have signed as a co-maker the promissory note which [illegible] will endorse the loan if granted and I understand that by signing the note I have such [illegible] liability for the payment thereof. [illegible] make the loan I make the following representations:

[illegible]

Relationship to Applicant(s): [illegible]

Print Name: [illegible] Married [illegible] Single [illegible] Relation to Applicant(s): [illegible]

Home Address: [illegible] How Long: [illegible] Phone Number: [illegible]

Previous Home Address: [illegible] How Long: [illegible] How Long Known Applicant(s): [illegible]

Employer: [illegible] How Long: [illegible] Position or Job Title: [illegible]

and Address: [illegible] Monthly Income: [illegible] Job or Phone No.: [illegible] Special Check: [illegible] Savings: [illegible]

Name of Co-maker's Bank: [illegible] Branch: [illegible] Regular Check: [illegible]

Credit References: Name: [illegible] Address: [illegible] Name: [illegible] Address: [illegible]

List All Real Estate Owned: [illegible] Mortgage: [illegible]

List All Automobiles Owned: [illegible] Make and Year: [illegible]

Previous dealings with you: [illegible] Co-Maker: [illegible] YES [illegible] NO

I hereby certify that each of the above [illegible] true and correct.

SIGNATURE: [illegible]

[illegible] with the proposed loan, a Certificate of Insurance [illegible] will be issued in accordance with the [illegible] provisions [illegible] in the Application for Loan.

CONSUMER CREDIT DEPARTMENT

DISCOUNT SHEET

Disb. No. _____

Acct. No. 05 66038

Terms _____ Monthly Payments _____

Int. _____ Last Payment _____

AUTHORIZED DEDUCTIONS	CHECK NO.			SETTLEMENT		
Sec. Deposits (Notes Passed)				Amount of Note		
Sec. Deposits (Equipment Rent)				Unearned Discount		
				Cr. Int.		
				Credit A/c Refinanced		
				Authorized Deductions		
	TOTAL			PROCEEDS	Check No. / Branch Settlement / Special Check No. / Regular Check No.	
				Balance		
Refinance				Late Charges		
Account No. _____				Less: Rebate		
Loan Num. _____				Credit A/c Refinance		

25 6 1681

APV. NO. _____

CONSUMER CREDIT DEPARTMENT

DISCOUNT SHEET

ACCT. NO. _____

TERMS 12 MONTHLY PAYMENT 19

DATE _____ LAST PAYMENT _____

V-23 3-48

AUTHORIZED DEDUCTIONS	CHECK NO.		SETTLEMENT		
SPEC. DEPOSITS (Notary Fees)			AMOUNT OF NOTE	448	00
SPEC. DEPOSITS (Judgment Fees)			UNEARNED DISCOUNT	15	68
			CITY TAX		15
			CREDIT A/C REFINANCED	134	99
			AUTHORIZED DEDUCTIONS		
TOTAL			PROCEEDS — CHECK NO. — BRANCH SETTLEMENT — SPECIAL CHECKING — REGULAR CHECKING	29	18

REFINANCE

ACCOUNT NO. _____

LOAN RUN _____

APPROVED _____

BALANCE	134 99
LATE CHARGES	
MISC. REBATE	3 00
CREDIT A/C REFINANCED	134 99

13.

CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY

PHILADELPHIA, PENNSYLVANIA

OCT 21 1949

FOR VALUE RECEIVED, the undersigned promise to pay within [illegible] months from the date hereof, without defeasance, to the CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA, hereinafter called "Bank", the sum of [illegible] DOLLARS in equal consecutive monthly installments of [illegible] (except the last installment, which may be unequal) beginning on [illegible] In case any monthly installment is more than 15 days in arrears, the undersigned shall pay as liquidated damages in addition thereto, a delinquency charge calculated at the rate of 1¢ for each dollar of such installment; such liquidated damages shall not exceed a total of $15.00 and only one delinquency charge shall be made on any one installment. The undersigned shall pay, in addition to all other amounts payable hereunder, (a) any fees paid to a public officer for filing, recording or releasing this or any other instrument or lien given to or obtained by the holder hereof in connection with the loan evidenced hereby, and (b) the actual expenditures, including reasonable attorney fees, for legal process or proceeding to secure or collect any installments, sums or amounts becoming due and payable hereunder.

The undersigned authorize Bank to: (1) date this note as of the date when the loan evidenced hereby is made; (2) complete any blank spaces in this note according to the terms upon which it grants the loan relative hereto; and (3) cause the signature of one or more co-makers to be added as additional undersigned to the original number of the undersigned, at any time or times and in no or with or without notice to any of the undersigned.

In the event of (a) any statement in the "Credit Statement" which was given as the inducement for this loan proving in the opinion of the holder hereof to be untrue or (b) the death of any of the undersigned; or (c) an application for the appointment of a receiver for, or (d) the making of an assignment for the benefit of creditors by or (e) any proceeding under the Bankruptcy Act by or against, or (f) any other act of insolvency, however expressed or indicated by, or (g) the issuance of an attachment or the entry of a judgment against, the undersigned, or any of them; or (h) if the undersigned fail to pay any installment or any other amount becoming due hereunder when and where due, then in any such event the full amount then remaining unpaid shall become immediately due and payable without demand or notice of any kind and the holder hereof may proceed to collect the same together with all other amounts due hereunder.

The holder hereof shall have a continuing lien upon any and all stocks, bonds, notes, moneys, balances and other property at any time in the hands of the holder belonging to the undersigned, as security for payment of this note, and also for any and all other obligations or liabilities of the undersigned to said holder, due or to become due, or that may be hereafter contracted, whether direct or contingent, whether now or hereafter held or acquired, and whether joint, several or with others, and agrees that the deficiency, if any, shall be made good from the collateral for the debt which lien shall be subject to all the provisions of this note.

The undersigned do hereby authorize and empower the Prothonotary, or Clerk or any attorney of any Court of Record of Pennsylvania or elsewhere, if this note or any installment thereof is not paid at maturity in the manner aforesaid, or upon this note maturing under any of its terms and provisions, as aforesaid, to appear for and to enter judgment against any or all of the undersigned for the above sum, with or without declaration filed, with costs of suit and release of errors, without stay of execution, and with 15% added for collection fees; the undersigned and each of them waive the right of inquisition on any real estate that may be levied on to collect this note and do hereby voluntarily condemn the same and authorize the Prothonotary or Clerk to enter upon the [illegible] for said voluntary condemnation, and agree that said real estate may be sold on a [illegible] fa. and hereby waive and release all relief from any and all appraisement laws and all exemption laws of the United States and of this state. The undersigned and all endorsers hereby waive protest of this note.

Any failure by the holder to exercise any right hereunder shall not be construed as a waiver of the right to exercise the same or any other right at any time and from time to time thereafter. The term "undersigned" as used herein, if there be more than one, shall denote joint and several liability hereunder, and shall mean all the undersigned, whether one or more, and all of the obligations of the undersigned hereunder shall be joint and several.

[illegible]

[illegible]

[illegible]

[illegible]

10 CENTS 10

5 CENTS 5

CONSUMER CREDIT DEPARTMENT

DISCOUNT SHEET

20 68930

Harry Dold
Phila General Hospital
34th St & Curie Ave
Phila Pa 12/20/49 35 yr

20 1890
3.00
345.00
20th

ACCT. NO.
TERMS 15 MONTHLY PAYMENTS 23
DATE LAST PAYMENT

AUTHORIZED DEDUCTIONS	CHECK NO.		SETTLEMENT	
SEC. DEPOSITS (Money Order)			AMOUNT OF NOTE	345
SEC. DEPOSITS (Judgment Fees)			UNEARNED DISCOUNT	24
			CITY TAX	
			CREDIT A/C REFINANCED	166
			AUTHORIZED DEDUCTIONS	
			CHECK NO. 75334	
TOTAL			PROCEEDS BRANCH SETTLEMENT / SPECIAL CHECKING / REGULAR CHECKING	154

REFINANCE
ACCOUNT NO.
PAID OUT
APPROVED

BALANCE 168
LATE CHARGES
LOAN TERMINATE
CREDIT A/C REFINANCED 116.03

DUE DATE		DUE DATE	CLASS	A/C NUMBER		AMT. OF NOTE	NO. OF MOS.	AMT. OF MONTHLY PAYMENT	
29	61687	25		61687	EPL	228.00	12	19.00	NOV

NAME & ADDRESS
DOLD Harry
6823 Kindred St
Phila Pa 19

HOME PHONE
10/21/49 yr
GR 5 8492

EMPLOYER
Phila General Hospital
POSITION
Nurse
INCOME
34.00a

ADDRESS
34th & Curie Ave
BUS. PHONE

DEALER OR COMMERCIAL CUSTOMER
DOUGHERTY Morrell E
PHONE

COLLATERAL | YR. | MAKE & MODEL | MFG. | SERIAL NO. | MOTOR NO.

Jm

Merrell E. Dougherty
Dorothy K. Dougherty
5517 Kingsland St.
Phila 43 Pa.

05105226
10-17
2/2/46-0 18
$21.00
2nd

MONTH DUE	BALANCE	DATE PAID
	INITIAL DEPOSIT	
MAR	327.0	MAR 6 1946
APR	308.83	APR 2 1946
MAY	290.66	MAY 1 1946
JUN	272.49	JUN 4 1946
JUL	254.32	JUL 10 1946
AUG	236.15	AUG 6 1946
SEP	217.98	SEP 10 1946
OCT	199.81	SEP 21 1946
NOV	181.64	SEP 21 1946
DEC	163.47	SEP 21 1946
JAN	145.30	SEP 21 1946
FEB	127.13	SEP 21 1946
MAR	108.96	SEP 21 1946
APR	90.79	SEP 21 1946
MAY	72.62	SEP 21 1946
JUN	54.45	SEP 21 1946
JUL	36.28	SEP 21 1946
AUG	18.11	SEP 21 1946

NAMES AND ADDRESSES OF CO-MAKERS
Barry Gold 6825 Kindred
Regina M. Lookebaugh 1841 S. Alden St.

V-107 MERRILL ATHENS, O. 45701

CLOSING ENTRIES

UNEARNED DISCOUNT 11.50
DISCOUNT EARNED 5.38
REBATE 6.12
REBATE
LATE CHARGES DUE
LATE CHARGES PAID
WRITE OFF
REFUND
CODE AF
NOT RETURNED SEP 26 1946
RETURNED BY MAIL TO
DATE PAID

CREDIT MEMO (CREDIT CLEARING)

COLLECTION SECTION MEMO

REF. APPL. PENDING NO. DATE
REF. APPL. PENDING NO. DATE RESULT

PRE.	NUM.	SECURITY	TOTAL VALUE	DATE	WORK

REQUEST for APPLICATION

CONSUMER CREDIT DEPARTMENT

COIN EXCHANGE NATIONAL BANK AND TRUST COMPANY

Philadelphia

Name (Style of A/C) [illegible]

Amount of Loan Requested $ [illegible]

Home Address [illegible]

Home Telephone No. [illegible]

Employed by (Firm Name) [illegible]

Annual Salary $ [illegible]

Business Address [illegible]

Business Telephone No. [illegible]

Position [illegible]

Length of Service to Present [illegible]

Purpose of Loan [illegible]

[illegible]

Source [illegible]

How identified

Identification approved by

Interviewed by

Office

61687 [illegible] 61687 [illegible] 28.00 [illegible] 79.9.00 [illegible]

[illegible]OLD Harry

[illegible]6823 Kindred St

[illegible]hila Pa #5

[illegible] 10/21/79 [illegible] 84.92

[illegible]hila General Hospital [illegible]search [illegible]

[illegible]ghth & Tucio Ave

[illegible] DOUGHERTY Harry [illegible]

If employed, we will write the follow

Position

How Long

Lodge

No.

Employer

Year Title

Weekly

Monthly

Address

Income $

We represent that each of the above statements is true and correct.

It is understood that if this application is approved, the first payment will be due from me/us one month from the exact date on which you mail your check to me/us pursuant thereto. The undersigned prefer the

of the month as the monthly due date for repayments on account of this loan.

Send mail to:

Residence

Business

Address

Address

You are hereby authorized and directed to pay out of the proceeds of said loan, if and when made, the following amounts to the following persons and/or companies to whom I/we am/are indebted.

Name

Address

Amount

The balance of said proceeds, if any, should be paid to the undersigned.

I/We authorize any one mentioned herein to furnish to you such information as you may require in connection with this application and agree that the application will remain your property whether or not the loan is granted. I/We agree to notify you immediately upon any material change in the above statement. I/We hand you herewith the following: (1) Promissory (Note (2) Signature Card.

(Answer "Yes" or "No")

Is any of this money to be used to buy a Listed Article costing $50.00 or more?

Is any of this money to be used as a down payment on the purchase of a Listed Article?

Signature(s) of applicant(s)

State relationship to first applicant

In connection with the proposed loan, a Certificate of Insurance Protection will be issued with respect to insurance upon the life of [illegible] in an amount sufficient to pay the balance of the loan, provided that in any event the limit of insurance is fixed at $2,500 on the aggregate of all obligations of said insured to Com Exchange National Bank and Trust Company, Philadelphia.

Application for

20 54740

CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

Personal Loan Department

Application No. C7736

To CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

Date 9 15 47

I (We) the undersigned, hereby make application for a personal loan in the amount of $ [illegible]

for [illegible] months, to be used for the following purpose: [illegible]

Print Name: Harry G. [illegible]

Age [illegible] Married ☐ Single ☐

Wife's Name

Home Address: [illegible]

How Long [illegible]

Phone Number [illegible]

Landlord and Address: [illegible]

Rent Per Month [illegible]

Former Address: [illegible]

How Long [illegible]

Number of Dependents

Employer: [illegible]

Position or Title [illegible]

How Long [illegible]

Badge No.

Husband Business Address: [illegible]

Income $ 200

Weekly ☐ Monthly ☐

Former Employer and Address: [illegible]

How Long [illegible]

Other Income Received from

Weekly ☐ Monthly ☐

Special Checking ☐ Savings ☐ Regular Checking ☐

Name of Applicant's Bank: [illegible]

Branch [illegible]

List automobiles owned: Year Make

Encumbrance

Title held by

List all loans, budget and installment accounts with banks, loan companies, installment houses, department stores or others

Name of Company	Address	Account Number	Original Amount	Amount Unpaid	Amount Due Each Month

List real estate owned	Date Purchased	Purchase Price	Assessed Value	Amount of Mortgages	Mortgages held by
Located at				Bal.	
Name in which title appears				Bal.	

CREDIT REFERENCES: (Preferably ones with whom undersigned has had credit dealings)

Name	Name	Name
Address	Address	Address

Statement of Account

	Pd. No.	Amount	Balance	Int.		No.	Amount	Balance	Int.
Mo.					C.M.				
Mo.					C.M.				
Mo.					C.M.				
Mo.					C.M.				
Mo.					C.M.				

[illegible] 20-24740

[illegible] 7.66

[illegible] 12.00

[illegible] 9th

[illegible] Sold

[illegible] Brotman & Associates

[illegible] 9-28 41st Ave.

[illegible] City, N. Y. 10/19/47 12 mo

[illegible]

[illegible]

[illegible]

[illegible]

[illegible] FOR BANK USE ONLY

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

CO-MAKER'S STATEMENT

Application No. _____ Date [illegible]

To CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

I agree to become a co-maker with said applicant(s) for a personal loan of $ [illegible] for a term of [illegible] months. I have signed as a co-maker the promissory note which will evidence the loan if granted and I understand that by signing the note I am made equally liable for the payment thereof. I have read the following statements and I make the following representations:

Information is confidential with the said applicant(s) whose [illegible] accordingly.

Primary Name: MORRELL E. DOUGHERTY Age: 39 Married or Single: M Relation to Applicant(s): Friend

Home Address: 5516 LATHFIELD ST. How Long: 4 YRS. Home Number: OA 4-8183

Previous Home Address: 4911 SPRINGFIELD AV. How Long: 5 YRS. How Long Known Applicant(s): 7 YRS. Years

Employer: ACHESON COLLOIDS CORP. How Long: 2 YR. Position Title: SER. ENGINEER

Business Address: LAND TITLE BLDG. Monthly Income: $ 450.00 Business Phone: LO 7-7113 Special Check: [illegible]

Name of Co-maker's Bank: CORN EX. LAND TITLE Branch: 53 & CONCH NUT ST. BROAD & CHESTNUT ST. Savings: Regular Check: [illegible]

LOANS & BANKER

Credit References: Name: CORN EXCHANGE BANK Address: 3rd & ARCH ST. Name: LAND TITLE BRANCH Address: BROAD & CHESTNUT ST.

List all Real Estate owned: NONE Purchase Price $ _____ Mortgage $ _____

List all Automobiles owned: _____ Make and Year: BUICK 1936

Previously applied with you: YES Borrower ☐ Co-Maker ☐ Yes ☐ No ☐

I certify that the above statements are true and correct.

Date _____ SIGNATURE Morrell E. Dougherty

Co-Maker

Information provided on this statement is confidential. A copy of this statement will be mailed to co-maker upon the granted application, provided requested in the application for loan.

	Payments to the end of June								
	No.	Amount	Balance	K.K.		No.	Amount	Balance	K.K.
Appl.					Cal.				
Appl.					Cal.				
Appl.					Cal.				

Type [illegible]

CONSUMER CREDIT DEPARTMENT

ACCOUNT SHEET

Harry Gold — 20-54918

W. Brotman & Associates — 87.66

29-28 31st Ave. — 212.00

L.I. City, N.Y. 9/19/47 12 pp — 9th

TERMS — MONTHLY PAYMENTS

DATE — LAST PAYMENT

AUTHORIZED DEDUCTIONS	CHECK NO.		SETTLEMENT	
DEPOSITS (Motor Week)			AMOUNT OF NOTE	212
DEPOSITS (Budget Week)			UNEARNED DISCOUNT	
			CITY TAX	
			AUTHORIZED DEDUCTIONS	
			CREDIT A/C REFINANCED	
TOTAL			CHECK NO. 6935 BRANCH SETTLEMENT	
			PROCEEDS SPECIAL CHECKING	
			REGULAR CHECKING	
REFINANCE			BALANCE	
			LATE CHARGES	
			LOSS REBATE	
			CREDIT A/C REFINANCED	

UNT NO.

RUN

OVED

If employed, we will state the following:

Employer _____ Position or Title _____ How Long _____ Badge No. _____

Address _____ Income $ _____ Weekly ☐ Monthly ☐

I/We represent that each of the above statements is true and correct.

It is understood that, if this application is approved, the first payment will be due from me/us one month from the exact date on which you mail your check to me/us pursuant hereto. The undersigned prefers the _____ of the month as the monthly due date for repayments on account of the loan.

Send Mail to: Residence Address ☐ Business Address ☐

You are hereby authorized and directed to pay out of the proceeds of said loan, if and when made, the following amounts, to the following persons and/or companies to whom I/we am/are indebted:

Name	Address	Amount
3-77		$81.50

The balance of said proceeds, if any, should be paid to the undersigned.

I/We authorize any one mentioned herein to furnish to you such information as you may require in connection with this application and agree that the application will remain your property whether or not the loan is granted. I/We agree to notify you immediately upon any material change in the above statement. I/We hand you herewith the following: (1) Promissory Note (2) Signature Card.

Signature(s) of applicant(s) _____

Relationship to first applicant _____

In connection with the proposed loan, a Certificate of Insurance Protection will be issued with respect to insurance upon the life of _____ in an amount sufficient to pay the balance of the loan, provided, that in no event the limit of insurance is fixed at $7,500 on the total of all obligations of said insured to Corn Exchange National Bank and Trust Company, Philadelphia.

Property sold
J. L. Brothers & Associates
20-28 31st Ave.
L.I.C., N.Y. 7-5/5/88 15 or

05-86033
21.000
315.000
5th

FOR BANK USE ONLY

REFINANCE

Application For Loan

CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

Personal Loan Department

Application No.

To CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

REFINANCE

Date

I/We, the undersigned, request a personal loan in the amount of

for months, to be secured by the following chattels:

Name

Age

Married ☐ Single ☐

Wife's Name

Home Address

How Long

Phone Number

Landlord and Address

Rent Per Month $

Former Address

How Long

Number of Dependents

Employer and Business Address

Position or Title

How Long

Badge No.

Income $

Weekly ☐ Monthly ☐

Former Employer and Address

How Long

Other Income Received from

$

Weekly ☐ Monthly ☐

Name of Applicant's Bank

Branch

Special Checking ☐ Savings ☐ Regular Checking ☐

List automobiles owned:

Year

Make

Encumbrance $

Title held by

List all loans, budget and installment accounts with banks, loan companies, installment houses, department stores or others:

Name of Company	Address	Account Number	Original Amount	Balance Unpaid	Amount Due Each Month

List real estate owned:

Located at

Name in which title appears

Date Purchased	Purchase Price	Assessed Value	Amount of Mortgage	Mortgages held by

CREDIT REFERENCES: (Preferably those with whom undersigned has had credit dealings)

Name	Address	Name	Address	Name	Address

Reserved for Use of Bank

	No.	Amount	Balance	Mat.		No.	Amount	Balance	Mat.
Appl.					CM.				
Appl.					CM.				
Appl.					CM.				
Appl.					CM.				

If employed, I/we will state the following:

Employer _____ Position or Title _____ How Long _____ Badge No. _____

Address _____ Income $ _____ Weekly ☐ Monthly ☐

I/We represent that each of the above statements is true and correct.

It is understood that if this application is approved, the first payment will be due from me/us one month from the exact date on which you mail your check to me/us pursuant hereto. The undersigned prefer the _____ of the month as the monthly due date for repayments on account of the loan.

Send Mail to: Residence Address ☐ Business Address ☑

You are hereby authorized and directed to pay out of the proceeds of said loan, if and when made, the following amounts, to the following persons and/or companies to whom I/we am/are indebted.

Name	Address	Amount
[illegible]		[illegible]

The balance of said proceeds, if any, should be paid to the undersigned.

I/We authorize any one mentioned herein to furnish to you such information as you may require in connection with this application and agree that the application will remain your property whether or not the loan is granted. I/We agree to notify you immediately upon any material change in the above statement. I/We hand you herewith the following: (1) Promissory Note (2) Signature Card.

(Answer "Yes" or "No")

If any of this money is to be used to buy a _____ Yes

If any of this money is to be used as a down payment on the purchase of a Listed Article _____ Yes

Signature(s) of applicant(s) _____

State relationship to first applicant _____

In connection with the proposed loan, a Certificate of Insurance Protection will be issued with respect to insurance upon the life of _____ in an amount sufficient to pay the balance of the loan, provided, that in any event the limit of insurance is fixed at $2,500 on the total of all obligations of said insured to Girard Exchange National Bank and Trust Company, Philadelphia.

[illegible]

Mary Gold
St. Pauls General Hospital
3120 Curlo Ave
CO MAKER 12/10/48 [illegible]

FOR BANK USE ONLY

Applicants Received

Approved

REFERENCE

Application For Loan

CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

Personal Loan Department

Application No.

LOAN APPLICATION

TO CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

Date

I/We, the undersigned, hereby make application for a personal loan in the amount of $

for months, to be used for the following purpose:

Print Name

Age

Married ☐ Single ☐

Wife's Name

Home Address

How Long

Phone Number

Landlord and Address

Rent Per Month $

Former Address

How Long

Number of Dependents

Employer and Business Address

Position

Title

How Long

Income

Weekly ☐ Monthly ☐

Former Employer and Address

How Long

Other Income Received from

Weekly ☐ Monthly ☐

Name of Applicant's Bank

Branch

Special Checking ☐ Savings ☐ Regular Checking ☐

List automobiles owned:

Encumbrance

Title held by

Year

Make

List all loans, budget and installment accounts with banks, loan companies, installment houses, department stores, or others:

Name of Company	Address	Account Number	Original Amount	Amount Unpaid	Amount Due Each Month

List real estate owned:

	Date Purchased	Purchase Price	Amount of Mortgage	Mortgages held by

Located at

Name in which title appears

CREDIT REFERENCES: (Preferably ones with whom mortgages and bank checking)

Name	Name	Name
Address	Address	Address

Bank

Date	Check No.	Amount	Balance	Date			

If employed, wife will state the following:

Employer _____ Position or Title _____ How Long _____ Badge No. _____

Address _____ Income $ _____ Weekly ☐ Monthly ☐

I/We represent that each of the above statements is true and correct.

It is understood that if this application is approved, the first payment will be due from me/us one month from the exact date on which you mail your check to me/us pursuant hereto, and the undersigned prefers the _____ day of the month as the monthly due date for repayments on account of the loan.

Send Mail to: Residence Address ☐ Business Address ☐

You are hereby authorized and directed to pay out of the proceeds of said loan, if and when made, the following amounts to the following persons and/or companies to whom I/we am/are indebted.

Name	Address	Amount
3-11		1138.00

The balance of said proceeds, if any, should be paid to the undersigned.

I/We authorize any one mentioned hereon to furnish to you such information as you may acquire in connection with this application and agree that the application will remain your property whether or not the loan is granted. I/We agree to notify you immediately upon any material change in the above statement. I/We hand you herewith the following: (1) Promissory Note (2) Signature Card.

Any of this money to be used to buy _____

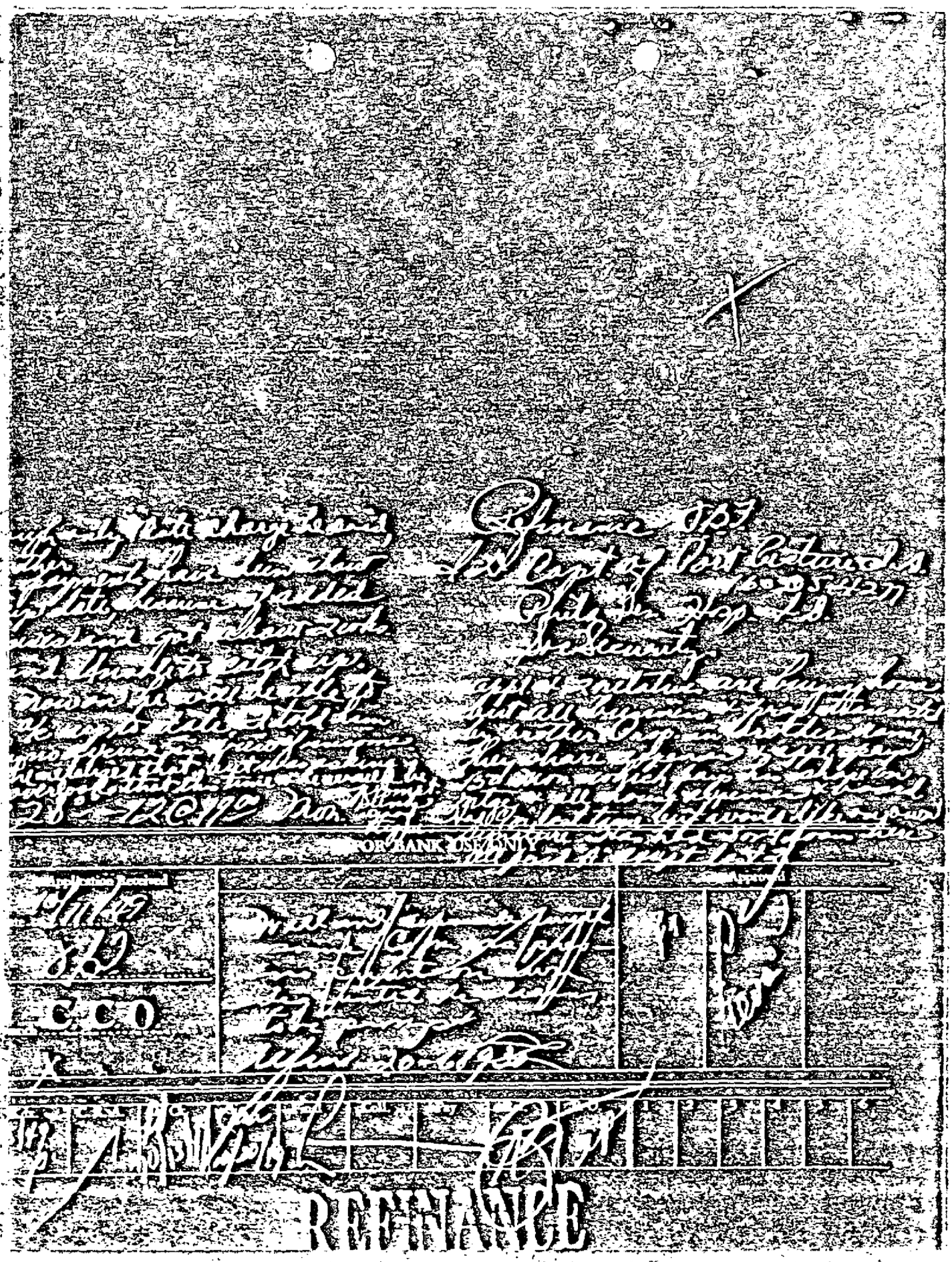
Any of this money to be used as a down payment on the purchase of a Used Auto? _____

_____ Signature(s) of applicant(s)

_____ State relationship to first applicant

Upon approval with the proposed term, a Certificate of Insurance Protection will be issued with respect to insurance upon the life of _____ in an amount sufficient to pay the balance of the loan, provided that in no event the limit of insurance is fixed at $2,500 on the total of all obligations of said insured to Gem Exchange National Bank and Trust Company, Philadelphia.

FILE


NOT BANK OF ENGLAND
RETINANCE

Application for Loan

25 61687

CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

Personal Loan Department

Application No. [illegible]

Date [illegible]

To CORN EXCHANGE NATIONAL BANK AND TRUST COMPANY, PHILADELPHIA

I/We hereby make application for a personal loan in the amount of $ [illegible]

for the following purpose: [illegible]

Print Name HARRY GOLD

Age 39

Married ☐ Single ☒

Wife's Name

Home Address 6723 Kindred St. (2d)

How Long 8 yrs

Phone Number [illegible]

Landlord and Address [illegible]

Rent Per Month $ [illegible]

Former Address 3232 Kensington Ave.

How Long [illegible]

Number of Dependents [illegible]

Employer [illegible]

Position or Title [illegible]

How Long [illegible]

Badge No.

Business Address [illegible]

Salary Per Week $ [illegible]

Former Employer and Address [illegible]

How Long 2 yrs

Other Income Received from None

Weekly ☐ Monthly ☐

Name of Applicant's Bank OLD REPUBLIC

Branch

Special Checking ☐ Savings ☐ Regular Checking ☐

List automobiles owned: Year Make None

Title held by

List all loans, budget and installment accounts with banks, loan companies, installment houses, department stores and others:

Name of Company	Address	Account Number	Original Amount	Balance Unpaid	Amount Due Each Month
3-18		10-68930	276	158	23

List real estate owned: None

Date Purchased	Purchase Price	Present Value	Amount of Mortgages	Mortgage held by

Located at

Name in which title appears

CREDIT REFERENCES: (Preferably those with whom undersigned has had credit dealings)

Name	Name	Name
Address	Address	Address

Reserved for Use of Bank

	No.	Amount	Balance	Check		No.	Amount	Balance	Check
Appl.					C.N.				
Appl.					C.N.				
Appl.					C.N.				

ORIGINAL DISCOUNT

UNEARNED DISCOUNT

DISCOUNT EARNED

REBATE

CODE

DATE CHECK NO

FIELD CARD

CREDIT MEMO (CREDIT CLEARING)

COLLECTION SECTION MEMO

Harry Gold

Philia General Hospital

45th St & Curie Ave

Philadelphia Pa 27/20/48 15 yr

PO 18930

23.00

345.00

MONTH DUE	BALANCE	DATE PAID	DATE PAID
JAN	345.00	JAN 4 1949	
FEB	322.00	MAR 3 1949	
MAR	299.00	MAR 31 1949	
APR	276.00	MAY 4 1949	
MAY	253.00	JUL 6 1949	
JUN	230.00	JUN 12 1949	
JUL	207.00	AUG 2 1949	
AUG	184.00	SEP 8 1949	
SEP	161.00	OCT 3 1949	
OCT	138.00	OCT 24 1949	
NOV	115.00	OCT 24 1949	
DEC	92.00	OCT 24 1949	
JAN	69.00	OCT 24 1949	
FEB	46.00	OCT 24 1949	
MAR	23.00	OCT 24 1949	

PAID

OCT 1949

CONSUMER FINANCE

COLLATERAL OR CO

Harry Gold

Philia General Hospital

DATE	APPLICATION	LOAN	AMT. AS MAKER	AMT. AS CO-MAKER	
3/17/44	28447		102 00		

Chemist Penna. Sugar Co. 34

Gold Harry 6823 KINDRED St. Sec. Boulevard Sta

DATE	APPLICATION	LOAN	AMT. AS MAKER	AMT. AS CO-MAKER	
		SEE OT	ERICARD		

Chemist Chemist Prochemist & Associates

Gold Harry 6823 Kindred St. Phila. 24, Pa.

DATE	APPLICATION	LOAN	AMT. AS MAKER	AMT. AS CO-MAKER	
1/10/58	1384	901168	416	PATP	

Pa. Sugar Co. (Chemist) 26 Dsy. Dr. Phila. 20.

Gold, Harry

DUE DATE	CLASS	A/C NUMBER	AMT. OF NOTE	NO. OF MOS.	MONTHLY PAYMENT
2		61587	228.00	12	19.00

NAME & ADDRESS

Cohen Barry
6423 Kindred St
Phila Pa 19138

10/21/49

HOME PHONE 215 8[illegible]

EMPLOYER	POSITION	INCOME
Phila General Hospital	Research	3400a

ADDRESS

[illegible]

BUS. PHONE

DEALER OR COMAKER & ADDRESS

Pennsylvania Power [illegible]

PHONE

COLLATERAL	YR.	MAKE & MODEL		SERIAL NO.	MOTOR NO.

AUTO	OWNS	RENTS	OTHER INCOME

AUTO INS.	PREV. LOAN NO.	AMOUNT	PAID

LATE CHARGES DUE	LATE CHARGES PAID	NO. MONTHS DUE	BALANCE	DATE PAID	BALANCE DUE	DATE PAID

ORIGINAL AMOUNT

UNEARNED DISCOUNT

DISCOUNT EARNED

REBATE

KOZ

CODE

NOTE

CHECK NO.

FIELD

CARD

CREDIT MEMO (CREDIT CLEARINGS):

COLLECTION SECTION MEMO:

COLLATERAL REGISTER

CLASS	ACCOUNT NO.	TOTAL DUE	PAYMENTS
	61587	228.00	12 19.00

NAME & ADDRESS
Gold, Harry
6623 Kirkwood St
Phila Pa

HOME PHONE: 215 Flu

EMPLOYER: Phila General Hospital
POSITION: Research
INCOME: [illegible]
ADDRESS: 5th & Cecil Ave
BUS. PHONE:
SALES OR COMAKER'S ADDRESS: [illegible]
PHONE:
COLLATERAL: [illegible]
MODEL & MODEL: [illegible]
SERIAL NO.:
MOTOR NO.:

DELINQUENT MONTHS	LATE CHARGE	MONTH	BALANCE	DATE PAID	NO. MO. DELINQUENT	DATE PAID
		NOV	228.00	NOV 29 1949		
		DEC	209.00	JAN 5 50		
		JAN	190.00	FEB 5 50		
		FEB	171.00	MAR 10 50		
		MAR	152.00	APR 5 50		
		APR	133.00	MAY 12 50		
		MAY	114.00			
		JUN	95.00			
		JUL	76.00			
		AUG	57.00			
		SEP	38.00			
		OCT	19.00			

COPY

October 15, 1945

Mr. Harry Gold
Gold Manufacturing Co.
Dallas, 24, Texas

Dear Mr. Gold:

[illegible] Chestnut Street
[illegible], Montana.

Mr. Herman:

Committee will consider with brother Joseph Gold, [illegible] pre-maker. Payments have been slow on this current account.

Photo enclosed.

CTL:ACL

[illegible] E. Kercher

Contact Gold & the [illegible]
[illegible] Contribution [illegible]
[illegible]
[illegible]

[illegible] 1943

[illegible] Henry Gold
[illegible] Abbott St.
[illegible] 24, Pa.

[illegible] Gold

[illegible] 110 Chestnut St.
[illegible] Moore

[illegible] Mr. Mathews:

[illegible] committee will consider with Morrell Dougherty as co-maker.

[illegible] Sgt. R. Kercher

Cathode Alternator + Fuse
[illegible] Small Tube [illegible] all small
[illegible] to [illegible]

B. J.

C4591

October 13, 1949

Mr. Harry Gold
6323 Kindred St.
Phila., 24, Pa.

Dear Mr. Gold:

510 Chestnut St.
Phila., Penna.

Mr. Thomas:

Will only consider with same go-maker as previous.
No credit on Owen signature until he learns to be
prompt.

Note enclosed.

GTL

J. Hagenkotter

Appl. wrtg. Owen Mon. A. M. & pt
Mt. & Tu. 3:30

CLOSING ENTRIES

UNEARNED DISCOUNT

DISCOUNT EARNED

REBATE

LATE CHARGES DUE

LATE CHARGES PAID MAY 5 1948

NOTE CODE 112 NOTE RETURNED

CREDIT MEMO (CREDIT CLEARING)

COLLECTION SECTION MEMO

REF. APPL. PENDING NO. DATE RESULT

REF. APPL. PENDING NO. DATE RESULT

DEP.	CHK.	SECURITY	TOTAL VALUE	DATE WDL.

20-64740

Harry Gold
5 A. Brotman & Associates
20-28 41st Ave.

312.00

	DATE DUE	AMOUNT	DATE PAID		DATE PAID
	OCT	8 18.00	OCT 23 1947		
	NOV	1 1 . 7 0	NOV 24 1947		
	DEC	1 2 1 . 0 0	DEC 23 1947		
	JAN	1 2 0 . 0 2	JAN 10 1948		
	FEB	1 0 1 . 7 0	FEB 24 1948		
	MAR	1 0 2 . 7 0	MAR 23 1948		
	APR	1 0 0 . 0 1	APR 23 1948		
	MAY	0 0 . 7 0	MAY 5 1948		
	JUN	7 0 . 7 2	MAY 5 1948		
	JUL	1 2 . 0 0	MAY 5 1948		
	AUG	3 5 . 4 0	MAY 5 1948		
	SEP	1 7 . 7 6	MAY 5 1948		

PAID

MAY 5 1948

CONSUMER CREDIT DEP.

NAME AND ADDRESS OF CO-MAKER PHONE

Morris E. Pomerantz 6517 Lindenfield St. Phi

Harry Gold 9/28/48 05-66035
A. Brothman & Associates 21.00
29-28 41st Ave. 315.00
L. I. City, N. Y. 5/5/48 15 or 5th

Pilgrim
5-8492

REPORT: (LIST ALL CONTACTS)

MONTH DUE	BALANCE	DATE PAID
Aug	273.—	9/17/48
Sep	[illegible]	[illegible]
Oct	[illegible]	[illegible]
Nov	[illegible]	[illegible]

CLOSING ENTRIES

UNEARNED DISCOUNT — S. D. A. BAL.

DISCOUNT EARNED — REBATE

REBATE — INT. ON S. D. A.

LATE CHARGES DUE — LOAN

LATE CHARGES PAID — REFUND

KDX CODE AB — NOTE RETURNED FEB 19 1947 — BY MAIL TO M — FIELD CARD

CREDIT MEMO (CREDIT CLEARINGS)

COLLECTION MEMO

Harry Gold 70-05033
44.16
1/26/46-6 MAKER 530.00
26th

MONTH DUE	BALANCE	DATE PAID
TLB	INITIAL DEPOSIT 530.00	14 28 1946
	485.74	APR 1946
MAY	441.48	MAY 7 1946
JUN	397.22	JUN 17 1946
JUL	353.26	JUL 20 1946
AUG	309.26	AUG 10 194
SEP	265.00	SEP 10 1946
OCT	220.96	OCT 16 194
NOV	176.72	NOV 18 194
DEC	132.56	DEC 14 1946
JAN	88.40	JAN 8 194
FEB	44.24	FEB 14 1947

PAID

NAMES AND ADDRESSES OF CO-MAKERS

Wornell F. Dougherty 5517 Stonefield St. Phila.
Regina M. Lookabaugh 281 So. Alden St. Phila.

Macy's Gold
% A. Brotman & Associates
29-28 41st Ave.

05-66033

21.00
315.00

DUE	PAID	MONTH DUE	BALANCE	DATE PAID		DATE PAID
		JUN	315.00	JUN 8 1948		
✓		JUL	294.00	JUL 21 1948		
		AUG	273.00	AUG 17 1948		
		SEP	252.00	SEP 28 1948		
		OCT	231.00	OCT 8 1948		
✓		NOV	210.00	NOV 25 1948		
		DEC	189.00	DEC 8 1948		
		JAN	168.00	DEC 21 1948		
		FEB	147.00	DEC 21 1948		
		MAR	126.00	DEC 21 1948		
		APR	105.00	DEC 21 1948		
		MAY	84.00	DEC 21 1948		
		JUN	63.00	DEC 21 1948		
		JUL	42.00	DEC 21 1948		
		AUG	21.00	DEC 21 1948		

PAID
DEC 20 1948

COLLATERAL RECEIVED

ORIGINAL DISCOUNT
UNEARNED DISCOUNT
DISCOUNT EARNED
REBATE

CODE
ENTER NO.
TO
CARD

CREDIT MEMO (CREDIT CLEARINGS)

REF. APPL. PENDING NO.
DATE
RESULT

COLLECTION SECTION MEMO

RESERVE

Date Received 5/25/50

From R. J. ARBEITER
(Name of Contributor)

PHILA. SAVING FUND SOCIETY, 7th & WALNUT STS., PHILA.
(Address of Contributor)

By R. B. Leonard
(Name of Special Agent)

To be Returned Yes ( )
No (✓)

Description: Photostatic copies of Bank records pertaining to Harry Gold

File No. ~~65-4328-1-B-2~~

65-4367-1-B (2)

PSFS
R.J. ARBEITER

Material obtained from
R. J. Arbeiter
accounting Dept.
Phila. Saving Fund Society
7th & Walnut Sts
Obtained 5/25/50 by SA Robert B. Conard

Photostatic copies of Bank Records
pertaining to Harry Gold

~~65-43028~~
65-430748(2)

3 6

NO. E 155,425

HARRY GOLD

PAYMENTS	DEPOSITS	INTEREST WITHDRAWN	BALANCES DATE	INTEREST TO DEC. 31	PRINCIPAL
249.00			JUL 15 '48		4,000.58
20.00			JUL 21 '48		3,980.58
300.00			AUG 5 '48		3,680.58
5.00			AUG 16 '48		3,355.58
150.00			SEP 10 '48		3,205.58
75.00			SEP 16 '48	11.92	3,130.58
25.00		20	NOV 5 '48	11.64	3,105.58
25.00		1	NOV 19 '48	11.51	3,080.58
25.00		9	DEC 7 '48	11.42	3,055.58
10.00		4	DEC 8 '48	11.38	3,045.58
	1,138.11	2.2	DEC 8 '48	22.64	4,183.69
	2,654.11	27.10	DEC 9		6,837.80

ACCOUNT WITH

THE PHILADELPHIA SAVING FUND SOCIETY

NO. E 155.425

HARRY GOLD

PAYMENTS	DEPOSITS	INTEREST ITEM	BALANCES DATE	INTEREST TO DEC. 31	PRINCIPAL
	30.00		2	6.1	1,755
	18.1		JAN 1	2.82	1,916.9
	34.00	51	JAN 24	2.43	1,225.69
	10.45	50	FEB 2	2.93	1,266.14
	45.00	67	FEB 14	2.60	1,311.14
	12.00	18	FEB 21	2.57.8	1,323.14
	55.00	97	FEB 28	2.6.75	1,378.14
	35.00	52	MAR 8	2.7.27	1,413.14
	185.00	1.85	MAY 5	29.12	1,608.14
	94.00	94	JUN 5	30.06	1,702.14
300.00		12.11	JUN 12	1.79.5	1,402.14
	20.00	20	JUN 16	1.8.15	1,422.14
	50.00	25	JUL 10	1.8.40	1,472.14
	64.00	32	AUG 1	1.8.72	1,536.14
	33.86	31	SEP 13	1.9.03	1,599.80
	80.00	10	OCT	1.9.13	1,679.80
	132.00		NOV 13		1,811.80
	70.00		DEC		1,881.80
	24.00		DEC		1,905.80
	12.20		DEC 18		1,918.00
	19.43		JAN		1,937.43
50.00			JAN	45	1,887.43
	50.00		FEB 24	45	1,937.43
	32.00		MAR	45	1,969.43
	200.00		MAY 31	45	1,769.43
	150.00		MAY 31	45	1,919.43
	50.00		AUG	45	1,969.43

IN ACCOUNT WITH THE PHILADELPHIA SAVING FUND SOCIETY

NO. E 15 54 25

Harry Feldman

PAYMENTS	DEPOSITS	INTEREST ITEMS	BALANCE DATE	INTEREST TO DEC. 31	PRINCIPAL
	220.00		SEP 12 '45	7.96	227.96
	275.00		OCT 3 '45		2,294.43
	18.00		NOV 13 '45		2,312.43
	100.00		DEC 2 '45		2,412.43
	26.96		DEC		2,439.39
	90.00		JAN 1 '46		2,529.39
75.00			FEB 25 '46		2,454.39
125.00			MAR 20 '46		2,329.39
200.00			APR 1 '46		2,129.39
75.00			APR 3 '46		2,054.39
200.00			APR 25 '46		1,854.39
75.00			MAY 6 '46		1,779.39
75.00			JUN 5 '46		1,704.39
	18.00		SEP 17 '46		1,722.39
250.00			NOV 2 '46	27.32	1,472.39
	50.00		NOV 15 '46		1,522.39
	27.32		DEC 31 '46		1,549.71
	95.00				1,644.71
	50.00		JAN 23 '47		1,694.71
75.00			MAY 21 '47		1,619.71
50.00			JUN 13 '47		1,569.71
7.00			SEP 29 '47		1,562.71
230.00			SEP 29 '47	220.00	1,332.71
50.00			DEC 26 '47	21.87	1,332.71
	21.87		DEC 31 '47		1,354.58
55.00			JAN 7 '48		1,299.58
40.00			JAN 30 '48		1,259.58
10.00			FEB 2 '48		1,249.58

TO CARD 3

IN ACCOUNT WITH THE PHILADELPHIA SAVING FUND SOCIETY

No. E 155,425

HARRY GOLD

PAYMENTS	DEPOSITS	INTEREST ITEMS	BALANCES DATES	INTEREST TO DEC. 31	PRINCIPAL
120.00		1.80	APR 8 '42	1.66	121.48
	15.00	.17	JUN 3 '42	1.83	59.48
	60.00	.50	JUN 3 '42	2.33	119.48
	25.00	.21	JUL 20 '42	2.54	144.48
	25.00	.21	AUG 5 '42	2.75	169.48
	15.00	.10	AUG 26 '42	2.85	184.48
	50.00	.33	SEPT 2 '42	3.18	234.48
	30.00	.15	SEPT 14 '42	3.33	264.48
	35.00	.17	SEPT 28 '42	3.50	299.48
	20.00	.07	OCT 7 '42	3.57	319.48
	150.00	.50	OCT 15 '42	4.07	469.48
	50.00	.23	DEC 14 '42	4.07	519.48
	4.07 INT		DEC 31 '42	10.14	523.55
	81.00	.78	JAN 11 '43	11.19	604.55
	30.00	.15	JAN 22 '43	12.39	634.55
	20.00	.30	APR 3 '43	12.69	654.55
	40.00	.53	MAY 1 '43	13.22	694.55
	30.00	.35	MAY 22 '43	13.57	724.55
	65.00	.65	JUN 19 '43	14.22	789.55
	60.00	.61	JUL 3 '43	14.83	850.55
	10.00	.08	JUL 19 '43	14.91	860.55
	140.00	.80	AUG 1 '43	15.71	980.55
	20.00	.13	SEPT 4 '43	15.84	1000.55
	35.00	.12	OCT 16 '43	15.96	1035.55
	35.00	.06	NOV 20 '43	16.02	1070.55
	75.00	.12	NOV 26 '43	16.14	1145.55

IN ACCOUNT WITH THE PHILADELPHIA SAVING FUND SOCIETY

NO. E 155,425

HARRY GOLD

WITHDRAWALS	DEPOSITS	INTEREST WRITTEN	BALANCES DATE	INTEREST TO DEC. 31	PRINCIPAL
	50.00	25	AUG 8 '40	25	50.00
	10.00	7	AUG 20 '40	32	0.00
	5.00	7	SEP 6 '40	39	55.00
	10.00	13	OCT 5 '40	52	65.00
15.00		12	DEC 5 '40	50	50.00
		3/31/41	3/31/41	1.00	51.50
	15.00	25	FEB 21 '41	1.85	55.50
	10.00	15	MAR 11 '41	2.00	65.50
	5.00	7	MAR 26 '41	2.07	70.50
	10.00	15	APR 5 '41	2.22	80.50
	10.00	18	APR 28 '41	2.35	90.50
	10.00	15	MAY 5 '41	2.48	100.50
	10.00	10	JUN 19 '41	2.58	110.50
15.00		15	JUL 26 '41	2.43	95.50
	15.00	18	AUG 2 '41	2.55	110.50
	50.00	20	SEP 4 '41	2.75	160.50
	50.00	10	OCT 7 '41	2.85	210.50
	15.00	5	OCT 22 '41	2.90	225.50
	15.00	5	NOV 5 '41	2.95	240.50
	20.00		NOV 19 '41	2.98	260.50
	10.00		DEC 8 '41	2.98	270.50
			12/31/42	3.46	273.48
10.00		2.01	FEB 5 '42	3.45	263.48
	1.00	6.8	FEB 9 '42	3.3	264.48
10.00		7	MAR 5 '42	3.46	264.48

OVER

ON ACCOUNT WITH THE PHILADELPHIA SAVING FUND SOCIETY

PLEASE PREPARE YOUR OWN

DEPOSIT TICKET

THE PHILADELPHIA

SAVING FUND SOCIETY

ACCOUNT NUMBER

NAME

(DEPOSITOR'S NAME PLEASE SIGN AS ON DEPOSIT BOOK)

DATE

DEPOSIT BOOK MUST ACCOMPANY THIS DEPOSIT

CASH

CHECKS

PLEASE LIST SEPARATELY

TOTAL

LESS CASH RECEIVED

NET DEPOSIT $

1738 12/41

174800 155425

PLEASE PREPARE YOUR OWN
DEPOSIT TICKET
THE PHILADELPHIA
SAVING FUND SOCIETY
MEMBER FEDERAL DEPOSIT INSURANCE CORPORATION

ACCOUNT NUMBER 875425

NAME Mary [illegible]
(DEPOSITOR WILL PLEASE SIGN AS ON DEPOSIT BOOK)

DATE 9 [illegible] 11 1941

DEPOSIT BOOK MUST ACCOMPANY THIS DEPOSIT

CASH			
CHECKS (PLACE EACH SEPARATELY)	34	43	66
TOTAL	$ 43	66	
SPACE BELOW RESERVED	TOTAL AMOUNT OF CHANGE RECEIVED FROM TELLER		
31	NET DEPOSIT	$ 53	66

[illegible] 1941

7 22 99980 ED 155425 E

SAFE DEPOSIT BOXES FOR RENT

PLEASE PREPARE YOUR OWN DEPOSIT SLIP
DEPOSITED IN
THE PHILADELPHIA SAVING FUND SOCIETY
MEMBER FEDERAL DEPOSIT INSURANCE CORPORATION

BOOK No. E155425

NAME Mary [illegible]
(DEPOSITOR WILL PLEASE SIGN AS ON DEPOSIT BOOK)

4 EUCH 15 46

DATE [illegible] 1941

DEPOSIT BOOK MUST ACCOMPANY THIS DEPOSIT

	DOLLARS	CENTS
CASH		
CHECKS	43	66
(PLACE EACH CHECK SEPARATELY)		
On Bill Due This Bank		
50		
TOTAL AMOUNT OF CHANGE RECEIVED FROM TELLER		
NET DEPOSIT	50	
SPACE BELOW RESERVED FOR TELLER'S USE		

[illegible] 1941

7 69 98 ED 155425 E

SAFE DEPOSIT BOXES FOR RENT

E155425

I AGREE TO CONFORM TO THE BY-LAWS AND REGULATIONS OF THE PHILADELPHIA SAVING FUND SOCIETY AND ANY AMENDMENTS THERETO.

WRITE NAME IN FULL [illegible]

RESIDENCE 329 Lincoln Street Phila Pa

MOTHER'S MAIDEN NAME [illegible]
FATHER'S NAME [illegible]
WIFE'S NAME
HUSBAND'S NAME

OCCUPATION [illegible]

MAIDEN NAME [illegible] NATIONALITY [illegible]

BORN AUG 28 1940 [illegible] U.S. CITIZEN

Received Mar 1960

from [illegible] B.I.

(Name of Contributor)

(Address of Contributor)

By

(Name of Agent)

To Be Returned: Yes ( ) No ( )

Description:

Two photos of Harry Gold

File No. 65-43007-1 B-3

One print of each
given to J. W. Casel
of Daily News.

WH

Date Received 5/22/50

From Harry Golf
(Name of Contributor)

6823 Kindred
(Address of Contributor)

By T. Scott Miller Jr
(Name of Special Agent)

To Be Returned Yes ( )
No (✓)

Description: Waiver of search for premises at 6823 Kindred signed by Harry Golf

File No. 65-4302-1B (4)

65-4307-1-B(4)

FD-26
(7-29-44)

May 22, 1950

I, Harry Gold, having been informed of my constitutional right not to have a search made of the premises hereinafter mentioned without a search warrant and of my right to refuse to consent to such a search, hereby authorize Richard E. Brennan, and G. Scott Miller, Jr, Special Agents of the Federal Bureau of Investigation, United States Department of Justice, to conduct a complete search of my residence located at 6823 Kindred Ave, Phila., Penn.

These agents are authorized by me to take from my residence any letters, papers, materials or other property which they may desire.

This written permission is being given by me to the above named Special Agents voluntarily and without threats or promises of any kind.

(SIGNED)
Harry Gold

WITNESSES:

G. Scott Miller, Jr.
Special Agent - F.B.I.

Richard E. Brennan
Special Agent, F.B.I.

Date Received 5-25-50

From J. J. Malloy
(Name of Contributor)

St. Joseph College
(Address of Contributor)

By Joseph E. Spaniy
(Name of Special Agent)

To Be Returned Yes ( )
No (✓)

Description: 3 lists of students taking classes with Harry Gold

File No. ~~65-[illegible]~~ 65-4307-1-B-(6)
4307

See 65-4307-78

65-4307-1-B-(5)

C
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FERMENTATION PROCESSES

Adams, Richard S., 297-A Copley Rd., U.D. Publicker Com. Alcohol
Allen, Thomas T.M., 7966 Bayard Rd., - Publicker Com. Alchhol Co., 1429 Walnut
Alpert, Sydney, 3314 West Huntingdon St., - Publicker Com. Alc. Co.. Del.Ave. & Bigler Sts.
Beatty, John C., Jr., 1748 N. Felton St., - Penna. Sugar Co., 1037 N. Delaware Ave.
Betcher, Norbert A., 4509 Blakiston St., - Inspector of Naval Materials USN, 1600 Arch St.
Becker, Blanche M., 5608 N. 10th St., - Continental Distilling Co.,
Benischeck, Joseph J., 471 Connaroe St., -
Breitinger, Richard , 1011 Lindley Ave., - Publicker Com. Alc. Co., 1429 Walnut
Devlin, James A., 6238 N. 4th St.,
Dougherty, Morrell E., 5517 Litchfield St., - Penna. Alcohol Corp., 1021 N.Delaware Ave.
Dura, Leon J., 2729 E. Venango St.,
Gavin, Thomas F., 816 N. 63rd St., - Midvale Co.
Gold Harry, 5032 Boudinot St., Phila. 20 - Penna. Sugar Co. (Siboney Distilling Corp. 1037 N.Delaware Ave.,)
Grinnell, Nathaniel, 5409 Wynnefield Ave., U.S. Navy
Hannas, Paul V., 4935 Knox St., - Midvale Co., Nicetown, Phila.
Higgins, Samuel F., 4623 State Rd., Drexel Hill, Pa. - Midvale,Co., Nicetown, Phila. 40
Kishbaugh, Albert M., 5443 Thomas Ave., - Publicker Com. Alc.Co., Delaware Ave.& Bigler St.
Lacktman, Nathan, 238 Lecarra Drive, Lansdowne, Pa. - Publicker Com. Alc. Co.
LaMar, Felix L., Jericho Manor Apts.,119, Jenkintown, Pa.-Resinous Products & Chemical Co.
Logan, Robert B., West Valley Road Stratford Pa. - Publicker Com. Alc. Co.
Lookabaugh, Regina M., 1841 S. Alden St., Phila. 43 - Pennsylvania Sugar Co.1037 N.Del. Ave
Lucchesi, Fred J., 174 Sherbrook Ave.,-Lederle Labs, Inc., 401 N. Broad St.
Maketa, John, 234 West Queen Lane, - Midvale Co., Nicetown, Phila.
Manley, John P., 8209 Cadwalader Ave., Elkins Park, Pa., - Pennsylvania Sugar Co. 1037 N.Delaware Ave.
Mannocchi, Felix P., 636 Maris St. Publicker Com. Alc. Co., Del.Ave. and Bigler Sts.
Manzelli, Thomas A., 331 N. Felton St. Publicker Com. Alc. Co., Del.Ave.and Bigler Sts.,
McCosh, Donald D., 314 E. Meehan St., Publicker Com. Alc. Co., 1429 Walnut St.
Owen, Carl M., 301 W.Somerville Ave., Publicker Com. Alc. Co
Reihard, Donald G., Box 1639, 1421 Arch St., Publicker Com. Alc. Co. Bigler Sts.,
Ross, Sydney H., 1951 N. Patton St., - Publicker Com. Alc.Co.,Swanson and Mifflin Sts.,
Rush, Morris M., 1604 S. 5th St. - Publicker Com. Alc. Co., Del.Ave and Bigler Sts.,
Sarno, Joseph F. 119 W. Girard Ave., - Publicker Continental Co., 1429 Walnut St.
Savran, Wm. S., 208 Stoneway Lane, Merion, Pa., - School District of Phila.
Schacht, John R. 177 Marlin Ave., Lansdowne, Pa. Rohm and Haas Co.
Shapiro, Donald D., 2601 Parkway, - Publicker Com. Alc. Co.
Smith, Howard J., 37 School Lane, Springfield, Del. Co., Pa.- Publicker Com. Alc. Co. Del. Ave. and Bigler Sts.
Trombetta, Louis D., 528 Larchwood Ave., Bywood, Pa. - Continental Distilling Corp. Snyder Ave. and Swanson
Volk, Peter W., 474 E. Van Kirk St. - W.H. and F. Jordon Jr. Mfg. Co. Phila.
White, Wm. T., 318 N. York Road, Willow Grove, Pa. - Publicker Com. Alc. Co. Bigler St.

65-4307-1-B-(5

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DISTILLERY PRACTICE

1. Adams, Richard S. 297A Copley Rd., U.B. Publicker Com. Alc., Del. Ave. & Bigler Sts.
2. Allen, Thomas T.M. 7966 Bayard Rd. - Publicker Com. Alc., 1429 Walnut St.
3. Beatty, John C. 1748 N. Felton St. - Penna. Sugar Refining Co. 1037 N.Delaware Ave.
4. Becker, Blanche M., 5608 N. 10th St. - Continental Distilling Corp.
5. Beesch, Samuel C., 4922 N. 12th St. - Publicker Com. Alc.,
6. Breitinger, Richard, 1011 Lindley Ave., 41 - Publicker Com. Alc.
7. Camuso, Joseph P., 4950 Pulaski Ave. Erlanger Brewing Co., 854 N. Orianna St.
8. DeFenso, John F., 6402 Chelwynde Ave., 42 - Publicker Com. Alc.
9. Dougherty, Morrell E., 5517 Litchfield St. 43 - Penna. Alcohol Corp. 1037 N.Del. Ave.
10. Durkee, Eleanor E., 112 N. Mole St. - Publicker Com. Alc. Corp.Swanson & Mifflin Sts.
11. Fisher, Fabiran R., 513 Timburlake Rd., U.D. - Erlanger Brewing Co.
12. Fisher, Paul, 5622 Litchfield St., - Publicker Com. Alc. Co.
13. Gelb, Jacob, 5414 Wyndale Ave., - Publicker Com. Alc. Co.
14. Gold, Harry, 5032 Boudinot St., - 20 - Pennsylvania Sugar Company
15. Hannas, Paul V., 4935 Knox St., - Midvale Co., Nicetown, Phila.
16. Kishbaugh, Albert M., 5443 Thomas Ave., - Publicker Com. Alc. Co.
17. Lacktman, Nathan , 238 Lecarra Drive, Lansdowne, Pa. - Publicker Com. Alc. Co.
18. Logan, Robert B. West Valley Road, Stratford, Pa. - Publicker Com. Alc. Co.
19. Lookabaugh, Regina M., 1841 S. Alden St. 43, - Pennsylvania Sugar Company
20. Lucchesi, Fred J., 174 Sherbrook Bld., U.D. - Publicker Com. Alc. Co.
21. Maketa, John, 234 West Queen Lane, Midvale Company
22. Mannocchi, Felix P., 636 Maris St., Publicker Com. Alc. Co.
23. McCosh, Donald D., 314 E. Meehan St. - Publicker Com. Alc. Co.
24. Mercer, Robert S., 4011 Plumstead Ave., Drexel Hill - Publicker Com. Alc. Co.
25. Perrino, Rocco A., 448 Wharton St., - Publicker Com. Alc. Co.
26. Ross, Sydney H., 1951 N. Patton St., - Publicker Com. Alc. Co.
27. Sarno, Joseph F., 110 W. Girard Ave., - Publicker Continental Corp., 1429 Walnut
28. Saltarelli, Frank L. 1015 Magee St., 11 - Publicker Com. Alc. Co.
29. Savran, Wm. S., 208 Stoneway Lane, Merion, Pa. - School District of Phila.
30. Senese, Samuel J., 2546 S. Ashford St. - Publicker Com. Alc. Co.
31. Smith, Howard J., 37 School Lane, Springfield, Del.Co., Pa. - Publicker Com. Alc. Co.
32. Trombetla, Louis D., 528 Larchwood Ave., U.D. - Continental Distilling Corp.

Snyder and Swanson

Total - 32 (Female: 3)

65-4707-1-B-15

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MAINTENANCE AND CONSTRUCTION OF LABORATORY GLASSWARE I. - 3008

1. Bergstresser, Darl S.,	715 Blythe Ave, Drexel Hill	- Foote Mineral Co.,
2. Bradley, Mary R.	7476 Breyer Rd., 38	- Navy Yard
3. Brady, Howard S.	1817 Eleanor St.,	- Bendix
4. Carroll, James W.	424 Church Lane, 44	- Penna. Salt
5. Darby, Richard T.	5236 Cedar Ave.,	- Quartermaster
6. DiBerardinis, Albert	1430 W. Chew Ave.,	- Barrett Div. Allied Chemical
7. Dilworth, Catherine A.	30 Maple Ave., Upper Darby	- Penna. Salt
8. Dismukes, Douglas E. Jr.	37 W. Gravers Lane,	- Fischer and Porter
9. Dougherty, Morrell E.	5517 Litchfield St.	- Penna. Alcohol Co.
10. Erhardt, John G. Jr.	4575 Torresdale Ave, 24	- Barrett
11. Friedman, Morris	2415 N. 29th St.,	- Fischer and Porter
12. Fuhrmeister, Edw. J.	4207 N. 9th St.,	- Mastbaum Vocational School; Fischer and Porter
13. Gavin, James F.,	816 N. 63rd St., 31	- Barrett Div., Allied Chemical
14. Gold, Harry	6823 Kindred St., 24	- Pennsylvania Sugar
15. Grey, Edward A.	3816 Berkley Ave.,Drexel Hill	- Foote Mineral Co.
16. Green, Harry	2545 N. 31st St.,	- Penna. Salt
17. Holmes, George	2928 W.QQueen Lane, 29	- Lennig and Co.
18. Junker, Thomas J.	135 Grays Lane,Haverford	- Barrett Div ., Allied Chemical
19. Lio, Anthony F.,	43 Glendale Road,Upper Darby	- Navy Yard
20. Lookabaugh, Regina M.,	1841 S. Alden St., 43	- Penna. Alcohol Co.
21. McAleer, Joseph R.	Hatboro, R.F.D., Pa.	- Fischer and Porter
22. McCoy, George	69-B Sayers Ave.,Lansdowne	- Penna. Salt
23. McKnight, James J.	2715 N. 6th St.,	- Navy Yard
24. Mellner, Max	1421 Arch St., 2	- Navy Yard
25. Millar, Sam.J.	Orange Home, Hatboro, Pa.	- Fischer and Porter
26. Murphy, Mary E.	1522 Mt. Vernon St. 30	- Wm. Amer. Co.
27. Nocero, Joseph F.	8138 Germantown Ave.,	- Fischer and Porter
28. Marshall, Glenwood R.	1612 Summer St.	- Cramp's Shipyard
29. Reed, Edward L.	Lincoln Highway, Box 662, Paoli, Frazer, Pa.	- Foote Mineral Co.
30. Schmidt, Paul P.	2334 N. 4th St.,	- Barrett
31. Schwartz, Boris	1608 Godfrey Ave.,	- Quaker Chemical Production Co.
32. Sentman, Robert A.	432 Heaton Road, Hatboro,Pa.	- Fischer and Porter
33. Serianni, Michael F.	8142 Ardleigh St.,	- Fischer and Porter
34. Shapiro, Arthur M.,	5151 Sansom St. 39	- Quartermaster
35. Smith, Matthew H. Jr.	318 Ellis Road, Willow Grove Pa	- Fischer and Porter
36. Souder, John C.	569 E. Broad St.,Souderton,Pa.	- Foote Mineral Co.
37. Stalker, Robert	106 Earl Lane, Hatboro, Pa.	- Fischer and Porter
38. Willson, Lawrence G.	503 Upland Road, Manoa, Pa.	- Westinghouse, Lester, Pa.
39. Fleet, Wm. J.	189 W. Glentay Ave.,Lansdowne	- Fleet Lab

*Not on opening enrollment report.

*Kuehner, Gertrude E.,	219 S. 45th St., 4	- Precision Therm.& Instrument
*Manfredi, Frank L.	190 Burmont Road,Drexel Hill	- Navy Yard
*Wagner, Richard L.	1421 Arch St.,	- Precision Therm. & Instrument

see SW 65-4307-89

Date Received _____

From _____
(Name of Contributor)

_____
(Address of Contributor)

By Joseph S. Kelly
(Name of Special Agent)

To Be Returned Yes ( )
No (✓)

Description: Surveillance notes of SA Joseph S. Kelly and SA Theron Rushing

File No. 65-4307-1-B-(6)

5-15-50 (JGK) 65-4308-1-13-(6)

Surv. instituted 6823 Kindred at 5:29 a.m. Subj. departed res. at 6:50 a.m., proceeded by same transportation as used previous day to Phila. General Hospital, arriving there at 7:55 a.m. Surv. discontinued.

5-11-50 JGK

Surveillance instituted 6823 Kindred St 5:50 am. Subj. departed from residence 6:45 a. walked north to Longshore Avenue, west on Longshore to Castor Avenue, where boarded 59 trolley at 6:55 a. Took 59 trolley to Margaret Orthodox station transferred to Frankford Elevated 7:10 am. Travelled on el to 13th Street station where transferred to #11 trolley west bound at 7:35 a. Debarked #11 street car 36th & Woodland 7:45 a. Surveilled to entrance to Heart Station PGH where entered at 7:55 a.m. Surv. discontinued.

5-18-50 JGK 65-43088(6)

Surveillance instituted
6823 Kindred 5:45 a.m.
Subject departed residence
6:45, proceeded to Phila
General Hospital via
same transportation as on
5-11-50, arriving there
7:50 a. Surveillance
discontinued 8:30 am
Surveillance at PGH resumed
1:30 pm. Subject departed
PGH 6:00 p.m. observed
to board # 11 trolley eastbound.
Proceeded by auto to Subject's
residence while Sa Rushing
surveilled subject on public
transportation. Surv. instituted
6823 Kindred at 7:20 p
Relieved by Sa Middleton
& Searl at 7:45 p

5-16-50 JGK 65-43088-1-B(6)

Surv. instituted subject's
residence 6823 Kindred
6:30 am, discontinued
9 a.m. Enroute to Phila
General Hospital by Bureau
auto from 9 am to 10 am.
Surv. instituted PGH at
10 am, discontinued 5:10 pm

5/20/50 JGK
65-4308-1B-(6)

Surv. instituted on subject's residence 6823 Kindred at 5 am. Subj. departed residence 6:45 a.m. proceeded via same transportation as 5/11 to PGH, arriving hospital 7:50 a. Surv. at PGH from 7:50 a to 4:10 p. when discontinued

5/19/50 JGK
65-4308-1B-(6)

Surveillance instituted 6823 Kindred at 6:00 am Subj. departed residence at 6:45, proceeded by same transportation as on 5/11/50 to PGH, arriving hospital 7:50 a.m. Surv. PGH from 7:50 to 6:15. Subj. departed PGH 6:15 boarded #42 trolley east bound proceeded to FBI office, Widener Bldg. Observed to enter Widener Bldg 6:35 p. Surv. discontinued.

5-22-50 - JGK
65-43884-146

Surv. resituated subject's residence 6823 Kinsley at 6^00 a.m. Surv. discontinued at 10:40a at which time subj., SAs Brennan & Miller & Kelley, all proceeded to FBI office by Bureau auto.

5/21/50 JGK
5-21-50.
(65-43884-146)

Surv. resituated subject's residence 6823 Kinsley at 6:15a. Subject departed res at 12 noon, boarded 54 trolley, proceeded to mgt Arborday Station Frankford El. All hushing boarded El with subject. SA Kelley proceeded to PGH by Bureau auto. Surv. resituated PGH at 1:05p.p., discontinued 3:30p.

Surveillance log 5/18/50
(5-7317-1-B-6)

3^45 AM 7^40 6823 Kindred
Sur Departed 6^45 A.
Proceeded by auto X
36th & Woodland
arriving at about 7^35
Sur observed entering PGH
Grounds about 7^50 A
Surveillance Discont'd 8^30 A
Re-instituted 1^30 PM 36th
& Spruce.
Sur Departed PGH apart
6^10 P proceeded by Trolly #11
to 15th & MKT. purchased
Paper Comm 15th & MKT

Surveillance log 5/16/50
(5-7317-1-B-6)

6^30 AM Residence 6823 Kindred
9^00 A Surveillance discont'd
at 6823 Kindred
Lorette 36th and
Spruce St.
Stationary Surveillance
near Entrance PGH
from Approx 10^00 AM
until Approximately 5^10 P

65-4318 JAR

Went into Bellmont Bar
on 15th St just below
MKT. entered Subway
after emerging from Bar
in three assailants 5 & 7 mins
proceeded by Subway
& Trolley 5th St Knox &
Canton Ave — opposite
entering residence at
about 7.20 P.

65-4318 JPR

Surveillance log 5/20/50
65-4307-1-B-6

5:00 A Vic 6823 Finished
Sur. Departed 6:45 A
arr PGH 7:50 A
Stationary Surveillance
until apprx 4:00 P at
36th & Woodland

65-4318 JPR

Surveillance log 5/19/50
65-4307-1-B-6

6:00 A Vic 6823 Finished
Departed 6:45 A
arr PGH 7:50 A
Stationary Surveillance
until 6:15 P
Mobile Surveillance
PGH & the Office
arriving Office apprx
6:35 P

65-4318 JPR

Surveillance Log 5/21/50
65-4301-13(6)

6:00 A Vic 6823 Kenfield
Stationary until about
10:40 A. Surveillance
Discont'd

65-4318 PAR

Surveillance Log 5/21/50
65-4301-13(6)

6:15 A Vic 6823 Kenfield
Subj Departed 12:00 noon
Proceeded via Trolley &
Elevated Subway &
36th & Woodland - arriving
PGH about 1:05 P
Stationary Surveillance
Vic 36th & Spruce
until 3:30 P.

65-4318 PAR

See serial 65-4307-89

Date Received 5-12-56 in 21-56

From Surveillance of 5-10, 12, 15, 17, 19, 18, 20, 21-56
(Name of Contributor)

(Address of Contributor)

By [illegible]
Name of Special Agent)

To Be Returned Yes ( )
No ( )

Description:
Notes of surveillance Navy Yard as indicated —

File No. 65-4307-1-B-(7)

65-4307-1-B(7)

65-~~[illegible]~~ 4307 - Surveillance - 5-10-50

56. J Mitchell, Federal, W = White

at 6923 Kenland for Steed.

Start 7:00 A — no show

Term: 9:00 A

Sum 5-12-50 -

Charles G.

Dr. Shank Dept of Justice

Inspection at Center -

5:25 P - 6:35 for photo

No Show

KO

65 4307 5-19-50
James Mitchell + Neal Beard
Sgt. Dr. Black Officer —
Langston + Cash
6:30A took up surveillance
6:30A - 14-16ft 16mm film
exposed. B+H.
6:55A Subject in Trolley
followed by Kelly + Rainey
7:00 at PH Hosp. —
Surveillance thru 10:20A
11:20P - surveillance to home
Salerno drive Subject home
12:25A - Lights out - terminated at JMH Hrs

65-9307 5-12-50

Surveillance - PGH - 8:35 PM -

to 12:00 Mid (one alone)

JM & Hes

5-16-50.

Took over surveillance for Kelly

& Rusling at 6823 Kendal

7:20 P.

12:25 Term. L.O.

JM & Hes

5-19-50. JM & Hes

followed W.P. Jones to 6823

Kendal - Going over

at 1:45 A -

2:35 A (5-20-50) - Since Term. T/L L.O.

KOT

65-4307

5-20-50

J. M. J. Spurr V Hos

Taken to Rec - 6823 - arr at 7:00 p

Term of 12:00 A (5-21-50)

with L.O. No contrib.

5-21-50

8:00 P at PGH - by Spurr

Ambulate V clear - from Jersey

Ambulate on PTC - t 6823

Knotted, Spurr in car 49

Arrd 7:19 - arr 7:23

9:00

Term 12:00 Mid - bright out

Hos

65-4307-1-3-(7)

5/15 Mon
5/16 Tues
5/17 Wed
5/18 Thurs
5/19 Fri
5/20 Sat
5/21 Sun
5/22/50 Mon

TO

FROM

RECEIVED

DATE

SPECIAL AGENT

CLASS

FILE NUMBER

LAST SERIAL

ENTIRE FILE

CHARGE-OUT SLIP

DATE

FD-9 (12-1-49)

65-4307-1-3-(7)

5/19 Friday
5/20 Dept Secty
1:31 AM
Left Office 2:00
at home 8 Duty

6-20-50
Left Hospital 5:25 Saturday
6:45 7:30
Hold Secty at his home

5/21/50 Sunday
7:20 9:45
Left office with

TO

FROM

RECEIVED

DATE

SPECIAL AGENT

CLASS

FILE NUMBER

LAST SERIAL

ENTIRE FILE

CHARGE-OUT SLIP

DATE

FD-9 (12-1-49)

5/15/50 Mon
10:00 AM
at Office Contact
Phone Rent/Hope

5/15/50 Monday
Contact
11:30 Office Wash at 12:25 minutes

5/16/50 Tues
5/17/50 Wed
Phone
Office

5/18/50 Thursday

TO

FROM

RECEIVED

DATE

SPECIAL AGENT

CLASS

FILE NUMBER

LAST SERIAL

ENTIRE FILE

CHARGE-OUT SLIP

DATE

FD-9 (12-1-49)

65-4307-1-3-(7)

Date Received 5-29-50

From Mr Samuel J. Clevenger Jr.
(Name of Contributor)

Franklin Institute 20th & Franklin Parkway
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By A.L. Pierce
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File No. 65-43071-B (8)

(See two Memos of SA Middleton dated 5-26-50)
(and Lt. Phila Report on Gold Case B, Thermal Diffusion Research)

THE JOURNAL OF CHEMICAL PHYSICS VOLUME 12, NUMBER 8 AUGUST, 1944

Letters to the Editor

PROMPT publication of brief reports of important discoveries in physics may be secured by addressing them to this department. The closing date for this department is the third of the month. Because of the late closing date for the section no proof can be shown to authors. The Board of Editors does not hold itself responsible for the opinions expressed by the correspondents. Communications should not in general exceed 600 words in length.

Depolarization of Raman Lines

A Further Simplification of Method

GEO. GLOCKLER, JOHN F. HASKIN, AND CLAIRE C. PATTERSON
Department of Chemistry, University of Iowa, Iowa City, Iowa
June 3, 1944

GLOCKLER and Baker[1] have studied the use of Polaroid in the measurement of the depolarization factors of Raman lines. They used a mica plate in order to rotate one of the components following the method of Bitt and Yost.[2] Lately we have realized that the mica half-wave plate is unnecessary if the two Polaroid films are positioned in front of the spectrograph so that their line of contact is at 45° to the vertical axis of the spectrograph. Preliminary results seem to show that this method will work satisfactorily. This suggested arrangement is published now because further work will be delayed for an unknown period on account of the war.

[1] Geo. Glockler and H. T. Baker, J. Chem. Phys. 11, 446 (1943).
[2] F. Bitt and D. M. Yost, J. Chem. Phys. 5, 90 (1937).

Thermal Diffusion in Liquids

HOWARD CARR
Physics Department, University of South Carolina, Columbia, South Carolina
July 12, 1944

A THERMAL diffusion tube similar to that described by Korsching and Wirtz[1] has been used to observe effects obtainable by varying some of the parameters involved in thermal diffusion experiments.

The tube, formed by clamping a rectangular Pliofilm gasket between two brass strips, has been previously described.[2] Since difficulty was encountered in keeping the

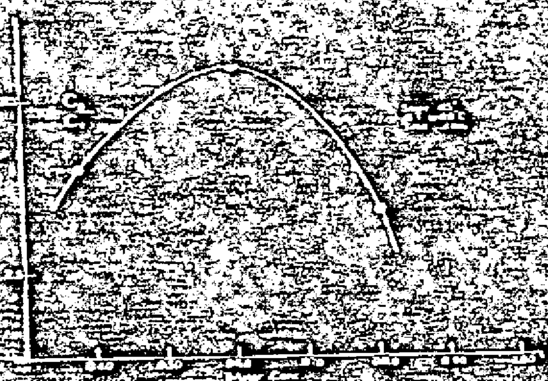

FIG. 1.

FIG. 2.

tube liquid tight with a 300-gauge Pliofilm gasket, 150-gauge Pliofilm[3] was substituted. The diffusion column thus formed was rectangular in cross section, 1.59×0.057 cm, and 1 meter long. The average temperature difference ΔT maintained between the top and bottom sides of the tube was 49°C.

Solutions of $ZnSO_4$ were used throughout the investigation since they had been previously shown to give a large effect.[2] Analyses of concentration were made by measurement of the refractive index.

The results are given in Table I. The effected separation is expressed as the ratio of the molar concentration C_B of a sample taken from the bottom of the tube to the concentration C_T of a sample from the top.

Figures 1 and 2 are graphs of data taken from Table I. They show more clearly the maxima obtained.

TABLE I.

Molar concentration of $ZnSO_4$	Time in hours	Tube slope in degrees	C_B/C_T
0.6	[illegible]	.75	1.70
0.6	[illegible]	.45	1.97
0.6	[illegible]	.5	10.8
0.6	[illegible]	.5	17.3
0.4	[illegible]	.5	3.04
0.6	[illegible]	.5	1.70
0.5	[illegible]	.5	2.25
0.6	[illegible]	0	2.55
0.6	[illegible]	.15	2.87
0.6	[illegible]	[illegible]	[illegible]
0.6	[illegible]	.45	3.70
0.6	[illegible]	.60	3.63
0.6	[illegible]	.75	2.89
0.6	[illegible]	.85	2.71

[1] Korsching and Wirtz, Naturwiss. 27, 367 (1939).
[2] H. Carr, Phys. Rev. 61, 726A (1942).
[3] Kindly supplied by the Goodyear Rubber Company, Akron, Ohio.

general conference on defense courses. The Section also participated in the formal opening of the University of Mississippi Observatory, at which D. V. Guthrie spoke on "Celestial Laboratories."

At the business meeting the election of the following officers for the year 1942–43 was announced: *Chairman*, W. L. KENNON; *Vice Chairman*, L. L. HENDREN; *Secretary*, E. SCOTT BARR; *Treasurer*, C. B. CRAWLEY; AND *Member of the Executive Committee*, A. W. DICUS.

The invitation extended by the Alabama Polytechnic Institute to hold its next meeting at Auburn, Alabama, was accepted.

E. SCOTT BARR, *Secretary*.

ABSTRACTS

2. Ultraviolet Absorption Spectra of Some Sugars. W. C. BOSCH AND E. W. ANDERSON, *Alabama University*.—The absorption spectra of aqueous solutions of ten monosaccharides and four disaccharides were studied in the region 200 to 400 mμ. The monosaccharide group included aldopentose, methyl-pentose, aldo-hexose, and keto-hexose types. The data indicate that absorption as shown by some sugars in the region of 280 mμ. As reported by other workers, sugars of the ketose type showed the most definite characteristic absorption. In general, the molecular extinction coefficients of sugars are low, being well below 10, and this suggests the possibility that absorption shown may be due to impurities. There is some evidence, however, to indicate that for certain sugars, the bands should be attributed to the sugars themselves.

(Now at Ohio State University.)

3. Photographic Effect of Zinc. PAUL S. DELAUP AND QUINTIN HOLDEMAN, *Southwestern Louisiana Institute*.—The darkening of a photographic film caused by a zinc plate placed near it was investigated. By performing the experiment in a mine far below ground it was shown that the effect was not due to cosmic rays as had been suggested by another investigator. It was also shown that the effect was not due to impact of slow-speed positive or negative particles.

4. The Infra-Red Absorption of Silver Cyanide. W. DAVID STALLCUP AND DUDLEY WILLIAMS, *University of Florida*.—The spectrum of powdered AgCN has been studied in the region 2500–1800 cm^{-1}. Intense absorption at 2178 cm^{-1} is attributed to an oscillation involving the CN group. The CN frequency for AgCN is almost 100 cm^{-1} higher than the CN frequencies of NaCN and KCN.

5. Space Group and Lead Positions in Lead Gluconate. R. SCERNSKY, *Alabama Polytechnic Institute*.—Lead gluconate, $Pb(C_6H_{11}O_7)_2$, crystallizes from water as pseudo-hexagonal plates which show Laue symmetry D_{6h}. Its and its oscillations (15°) with CuKα radiation show a hexagonal cell, with $a = 10.99$ A, $c = 17.18$ A. At the density 2.409 g/cc gives $Z = 11.99 \approx 12$ molecules/cell. Indexed diagrams reveal (0001) present only for l odd; all odd layer lines on c-axis oscillations very faint, strong intensities for every third spot along most zonal rows (h and k constant), weak third spots along remaining zonal rows, with l indices of strong or weak spots varying with particular zone-line. These variations can be accounted for only on space group $D_6^3 - C_3^2 21$ (or enantiomorphous $D_6^4 - C_3^2 21$), with lead atoms on a lattice of half the true c-periodicity. Positions for lead atoms in true cell, as determined by intensity variations and symmetry, are:

0.2, 0, ¼; 0, 0.2, ¼; −0.3, 0.3, 0;
0.7, 0, ¼; −0.5, 0.2, ¼; −0.8, 0.3, 0;
0.2, 0.5, ¼; 0, 0.7, ¼; −0.3, 0.8, 0;
0.7, 0.5, ¼; −0.5, 0.7, ¼; −0.8, 0.8, 0.

Each lead is on a twofold axis, with its two gluconate residues extended above and below along the c direction, the disposition of these residues necessitating the large cell.

The material studied was furnished through the kindness of Dr. Horace S. Isbell of the National Bureau of Standards.

6. Thermal Diffusion Tube for Liquids. H. E. CART, *University of South Carolina*.—A thermal diffusion tube has been constructed similar to that described by Korsching and Wirtz. A rectangular groove, 1 meter long, 1.59 cm wide and 0.023 cm deep was milled down the middle of two brass strips, each 41×1½×⅜ inches. These strips were clamped together, grooves facing, using No. 100 gauge Ticofilm as a gasket. Thus was formed a thin diffusion tube 1 meter long of rectangular cross section, 1.59×0.048 cm, into which liquids were introduced. A piece of channel brass, 41×1×1 inches, was soldered to the back of each of the strips to serve as water jackets. By passing hot and cold water through these jackets the top side could be heated and the bottom cooled.

With 0.75 molar solution of $ZnSO_4$, tube slope of 53° and temperature difference between slopes of approximately 55°C, the concentration at lower end was doubled in 11 hours. With 1 percent solution of sucrose increased to 26 percent at lower end in 21 hours, using a temperature difference of about 50°C and tube slope of 25°. Curves showing separation obtained as a function of time, concentration, and slope of tube will be given.

(Korsching and Wirtz, *Naturwissenschaften* 28, 35 (1940).)

7. An Interpretation of Wave Mechanics. N. RUSSELL, *University of South Carolina*.—If, in the Schrödinger wave equation for a single particle, the wave function ψ is written as Ae^{-iB}, where A and B are real functions, the equation splits up into two well-known equations. One of them has the form of the equation of continuity for a fluid of density A^2 and velocity $\nabla B/m$. The other has the form of the Hamilton-Jacobi partial differential equation for the function B, with the potential energy V replaced by $V - (\hbar^2/2m)\nabla^2 A/A$. However, these equations agree

Thermal Diffusion in Ternary Liquid Mixtures, Particularly Aqueous Solutions Containing Ferrous Chloride*

J. J. GILLISPIE AND SAMUEL BRECK
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(Received August 5, 1940)

The accumulative arrangement of Clusius and Dickel for thermal diffusion was used to study the concentration changes at the extreme ends of the apparatus that occur in various mixtures, particularly solutions of two solutes in water. The apparatus gave reasonably reproducible results. To avoid uncertainties due to creeping along the upper part of the hot wall, any apparatus intended for the study of slightly soluble substances should be designed so as to fill the apparatus used, so that it can be filled without leaving any space between hot and cold wall. The enrichment of acetic acid in water solution at the cold (and lower) end is rather small, compared with that of glycerine, or many salts or strong acids. Addition of sodium acetate does not greatly affect the enrichment of acetic acid. The enrichment of ferrous chloride in water at the cold end is considerable, but in the presence of 1 *M* HCl the enrichment occurs at the hot end. In the presence of 1 *N* H_2SO_4 the enrichment was zero. The case is opposite with zinc chloride. Here the small cold end enrichment that occurs in the presence of a trace of acid to prevent hydrolysis is considerably increased in the presence of 1 *M* HCl. In a solution of ferric chloride and 1 *N* HCl the enrichment of iron was also at the hot end. Hydrolysis takes place, but this probably works against such a result. It appears that a heavy constituent of a ternary liquid solution can migrate from cold to hot end, against the direction normally expected. A simple function of the composition is given, whose sign is the same as the sign of the enrichment at the cold end in all binary mixtures (liquid or gaseous) so far studied, but not always the same in ternary mixtures.

INTRODUCTION

THE arrangement of Clusius and Dickel[1] for producing separation by thermal diffusion works by combination of thermal diffusion along a horizontal thermal gradient between two vertical walls and convection along these walls. The arrangement used by various investigators of the Soret effect utilizes a vertical thermal gradient, the higher temperature being maintained above in order to avoid convection.

The arrangement of Clusius and Dickel may be regarded as a means for multiplying the Soret effect and offers a sensitive means of studying it. Unfortunately the factor by which the Soret effect is multiplied is not readily determinable, so that interpretation in terms of Soret effect can be only semiquantitative. Nevertheless, we have tried to investigate the behavior of some ternary mixtures containing water and two solutes, with respect primarily to the question whether the behavior of each of the solutes is independent.

APPARATUS

The arrangement was used in the form of two concentric glass tubes, steam being passed through the inner, and cooling water through the outer, the solution under investigation being in the annular space between the two tubes. Brewer and Bramley[2] have used this form. The Pyrex diffusion apparatus is shown in Fig. 1. The tubes for enclosing the annular space were selected as the best pair from about 100 samples and built into apparatus No. 1. The width of the annular space (along a radius) is about 0.75 mm, the total length about 1 meter, and the capacity of annular space plus dead space at top and bottom about 71 cc. The next best pair of tubes was built into apparatus No. 2. The width of annular space is about 1.25 mm, the length 1 meter and the capacity about 85 cc. Apparatus No. 2 gave considerably smaller separations in every solution used for comparison and only a few experiments are here reported with it.

In July the cooling water entered (the bottom) at 20° and left at 32°, constant to about 0.5° for days. This would make the mean temperature difference of the outside surfaces of the walls 100 − 26 for 74°. In winter this difference increased to 80° and rarely 85°.

Some preliminary experiments with 10 percent (by weight) of glycerine showed with neither

* Contribution No. 463 from the Research Laboratory of Physical Chemistry. It has a principally based on the thesis presented by Mr. Breck for the B.S. degree.

[1] K. Clusius and G. Dickel, Naturw. 26, 546 (1938).

[2] A. K. Brewer and A. Bramley, Phys. Rev. 55, 590 (1939).

apparatus any difference with time from 22 to 43 hours. In later experiments a comparison of results for different lengths of time was often possible and there was no indication that more than 24 hours was required for the steady state, unless, possibly in some solutions containing three solutes.

FILLING THE APPARATUS

For all the experiments but the last three the annular space was filled so that the solution stood during the experiment at about the level of the cooling water. This left a small air-space above the liquid. The upper filling tube was closed with a glass cap and wax after the steady temperature was reached. This prevented evaporation or oxidation.

The air space allowed salts to creep up along the hot wall. There the liquid film evaporated, the solvent passing to the cold wall. With solutions containing $BaCl_2$ creeping was very considerable and the results are not stressed. In the case of the other salts (except $ZnCl_2$) creeping occurred, but to a very small extent; the deposit was estimated to amount to not more than 0.1 gram. The rate at which the fluid is circulated along the walls is very considerable and it would seem that the small loss of material should occur at the expense of both hot and cold ends of the tube, without particular prejudice to either.

Nevertheless, the results for ferrous chloride in mixtures were so unexpected that three experiments (there reported as the last three items of Table I) were done after the completion of all the other work, when we found that the apparatus can be filled so that no air space is in contact with the hot wall and creeping is thereby prevented. Since the volume of the upper dead space was considerably greater by this manner of filling, a longer time was allowed for diffusion to take place.

SAMPLING

After the diffusion, a sample of about 1 cc was taken from the top with a small capillary pipette. When cool, a portion of about 0.5 cc was accurately measured in a special pipette. A sample of about 1 cc was taken from the bottom after wasting 6 drops to clear the stopcock leads. In each of the last three experiments of Table I about 1.5 cc was taken and 1 cc used for analysis. Samples from the middle were sometimes taken with a long capillary pipette, and sometimes through the stopcock after discarding a large portion. When the transfer of component is small, the middle portion agrees in composition with the original solution used for filling and was often taken as a measure of it.

FIG. 1. Diffusion apparatus. A cork stopper was actually used in place of the higher of the two ring seals, but it requires frequent replacement because of the action of the steam.

In the case of solutions containing HCl and $FeCl_3$ that diffused for 24 days, samples from levels one-third down the tube and two-thirds down showed large changes of acidity from the original, and the original titer was used as a basis. The titers for ferrous iron from these levels gave welcome checks on the titer of the original mixture.

ANALYSIS

Measurements of refractive index were used for solutions of glycerine and of acetic acid as noted below.

In solutions containing iron, ferrous iron was determined by permanganate titration. Since chloride was present the method was checked by analysis of known mixtures. Ferric iron was determined from the titer for ferrous iron and the titer for total iron as found after reduction with SO_2 and removal of excess SO_2. In mixtures of $FeCl_3$ and HCl the acid was first determined

by titration with sodium carbonate in the presence of bromphenolblue (without any precipitation of iron) and the ferrous iron then determined by permanganate in the same sample, a stream of nitrogen being used throughout to prevent oxidation. In mixtures containing ferric iron the acidity was not determined.

In the analysis of mixtures of $ZnCl_2$ (or $BaCl_2$) and HCl the hydrogen ion was first titrated in the presence of methyl red. The

TABLE I. *Diffusion of solutions containing HCl, $FeCl_3$, or $FeCl_3$ of concentrations 0.5 N; apparatus No. 1.*

Approx. composition of solution	Time hours	Cations assay	Per cent effect	Time ratio upper/lower
1 N HCl	41	H^+	160	0.4
0.5 N $FeCl_3$	23	Fe^{+++}	24	1.29
Same	31	Fe^{+++}	21	1.30
Same	24	Fe^{+++}	25	1.32
1 N HCl, 0.5 N $FeCl_3$	29	Fe^{+++}	10	0.90
Same	23	Fe^{+++}	3	0.95
		H^+	190	0.9
0.5 N H_2SO_4, 0.5 N $FeCl_3$	23	Fe^{+++}	50	1.
		H^+	122	5.0
1 N HCl, 0.5 N $BaCl_2$	24	Ba^{++}	44	1.64
		H^+	106	3.9
1 N HCl, 0.5 N $ZnCl_2$	29	Zn^{++}	30	1.37
		H^+	100	3.7
0 of N HCl, 0.5 N $ZnCl_2$	39	Zn^{++}	4	1.04
1 N HCl, 0.5 N $FeCl_3$	1×24	Fe^{+++}	33	0.88
		H^+	180	19.
Same	1×24	Fe^{+++}	11	0.89
		H^+	94	20.
0.5 N $FeCl_3$	1×24	Fe^{+++}	22	1.27

total chloride was then determined by titration with 0.1 N $AgNO_3$ in the presence of fluorescein as adsorption indicator, and the Zn^{++} (or Ba^{++}) obtained by difference. When the method was checked by analysis of solutions of HCl a small difference of titer was found (II and III). The difference was applied as a correction factor to the results for mixtures of $ZnCl_2$ (or $BaCl_2$) and HCl in order to get the best possible relative accuracy for total chloride.

Expression of Results

It will be noted that we do not attempt to determine the total transfer of component past a horizontal plane, but only the extremes of concentration at the two ends of the apparatus. The "percent effect" we define as the titer of the sample from the lower end minus the titer from the upper end, the difference being then multiplied by 100 and divided by the titer of the original sample subjected to diffusion. When the result is negative, this means that the substance is (abnormally) enriched at the upper end. No correction was applied for changes in the average temperature (74–85°, according to the season of the year). The results which we compare were done under nearly the same conditions.

The concentration ratio lower/upper divided by upper is also of some interest and is reported in Tables I and II. It is not possible to calculate it accurately from the percent effect alone because the mean of the compositions, upper and lower, is often not equal to the original or middle composition.

Results

In apparatus No. 2 a 10 percent (by weight) solution of glycerine in water was subjected to diffusion and the solutions analyzed by the refractive index. The percent effect was 3.4. Samples at 22 and 48 hours showed no difference. Repetition of the experiment gave in 22 hours the percent effect 6.54.

In apparatus No. 1 the same solution showed an effect of 28.8 percent with no differences in the results at 20, 28 and 41 hours. A repetition of the experiment gave 35 percent in 22.5 hours.

In apparatus No. 2 a 50 percent solution of glycerine in water showed an effect of 3 percent in 18 hours.

A solution of 62 percent (by weight) acetic acid in water showed no effect by refractive index in either apparatus in 45 hours.

A more dilute solution of acetic acid (10 percent and about 1.7 mol/l) showed in apparatus No. 1 an effect of 8.1 percent in 22.5 hours and on repetition an effect of 10 percent in 20.3 hours.

Various mixtures of acetic acid and sodium acetate were prepared and subjected to diffusion in apparatus No. 1. The effect for sodium acetate was determined by the refractive index. A small correction for the refraction of the acetic acid was applied which required possible small changes in its concentration. Table II shows the composition of the original solution and the percent effect for sodium acetate. The

transfer of acetic acid was shown to be small or zero by measuring the pH of the various samples by means of a glass electrode and by then comparing the observed pH with that calculated from the measured concentration of acetate in the sample together with the concentration of acetic acid it would have were there no transfer of acetic acid. The approximate agreement between observed and calculated pH was satisfactory and showed that the transfer of acetic acid is small in the solutions containing sodium acetate.

These results are not sufficient to establish that the acid and the salt in this case diffuse independently, but it is very probable that they do so at least approximately.

More interesting results were seen when solutions containing iron chloride were subjected to diffusion. Ferric chloride in water is hydrolyzed at once along the hot wall. This is due to the extensive migration of HCl (formed by hydrolysis) from hot to cold. Ferrous chloride is not hydrolyzed in our apparatus if oxidation is entirely prevented. This is accomplished by leaving a trace of SO_2 in the solution until the apparatus is filled in a stream of nitrogen. When all the SO_2 was, on the other hand, removed by boiling in a stream of nitrogen and the filling occurred in a stream of nitrogen, a little oxidation took place and a light yellow film was deposited on the upper part of the hot wall. The results for transfer of ferrous iron were substantially the same whether this occurred or not, as is shown by the second item of Table I compared with the next two. In the case of the second item there was a slight film in the next two and in all further cases (except the last item) in which ferrous chloride was present without ferric, the trace of SO_2 prevented any film formation. In the case of the last item the SO_2 was probably too successfully removed in the apparatus by allowing the supernatant part to come momentarily to boiling under partial vacuum after the steam had been on for an hour, and a thin metallic looking film was formed along the upper third of the hot wall. The result of this experiment agrees with the others.

When solutions containing ferric chloride and HCl (1 M) are subjected to diffusion, hydrolysis still takes place on the hot wall, a crust forming on its upper part. Portions of the crust occasionally drop loose and fall to the bottom, where they perhaps redissolve in part in the strong HCl (about 2 M). The hydrolysis makes, therefore, the interpretation of the results somewhat uncertain, but any enrichment observed at the lower end is presumably too high. Thus when negative enrichments are found they are probably correct as to sign. A 28-hour experiment with a solution 1 M HCl and 0.5 M $FeCl_3$ showed −32 percent effect. Three experiments with a solution 1 M HCl, 0.5 M $FeCl_3$, and 0.5 M $FeCl_2$, running for 13, 21, and 39 hours, showed percent effects for $FeCl_3$ −26, −47, and −36, respectively, with corresponding percent effects for $FeCl_2$ −4, −17, and −12. These experiments confirm that ferric chloride can show a negative effect in mixtures and that ferrous chloride does so.

We cannot guarantee that the times were long enough for these mixtures of four components. It may be that more than one steady state of approximately the same stability is possible for such complicated mixtures.

Discussion

To account for the fact that in binary gaseous solutions the heavier species is always found to be enriched at the cold (and low) end of the gradient in sample approximate theory has been offered. The theory of Chapman also accounts for this and allows of small modification of result for molecules of widely different diameters.

TABLE II. *Transfer of sodium acetate in mixtures with chloride and in aqueous solution*

Molecules per liter	3	3	15	3	12	12	0.55	
Moles acid per liter	0.17	0.17	1.0	1.0	0.14	0.14	0.005	
Hours	20	22	23	24	35	35	77	
Concentration ratio	1.18	2.17	2.05	2.03	2.21	2.13	2.04	
Percent effect	4	66	46	46	61	54	50	

[illegible]

So far, it appears to be a fact that also in binary liquid mixtures the heavier component is enriched at the cold end, even though the heavier component has the smaller molecular weight, as in acetone-water solutions.

[1] L. Gilmour, J. Chem. Phys. 7, 830 (1939).
[2] Sydney Chapman, Phil. Trans. (London) A217, 115 (1917).
[3] K. Clusius and G. Dickel, Naturw. 27, 148 (1939).

TABLE III. *Densities of some solutions M—molar per liter of solution.*

Solution	Density at 25°
0.5 *M* in HCl and 1 *M* in $FeCl_3$	1.0312
1 *M* in $FeCl_3$	1.0210
0.5 *M* in HCl (made up at 25°)	1.0060
0.5 *M* in HCl (made up at 18°)	1.0076

In the statement of this fact we may understand the heavier component of a binary mixture to be that one which has the greater density when pure, ρ_0, and perhaps some what better we may define the heavier component to be that one which increased the density of the solution when it is added at constant temperature and pressure, i.e., that one of a binary mixture for which the function

$$f = m_1(\partial\rho/\partial m_1)_{T,p,m_2} = \rho_0(1 - \alpha_1\phi)(\partial\rho/\partial\alpha_1)_{T,p,\alpha_2/(1-\alpha_1)} \quad (1)$$

is positive, where ρ is the density of the solution, m_1 is the mass of the substance 1 and α_1 is the mass of the substance 1 divided by the total mass. The function is of course positive for that component of a binary ideal gas mixture which has the greater molecular weight.

The same definition recommends itself for the consideration of solutions containing more than two components. For any solution the sum of the functions for all the components is zero by Euler's theorem for homogeneous functions of the zero order and hence at least one component must be heavy, and at least one must be light (except for the trivial case at a point where the density at constant T and p is a maximum or a minimum).

The rule holds for all the experiments that have come to our attention, including our own on binary mixtures, that the cold-end enrichment in a binary liquid or gaseous solution has the same sign as the function f.

But from the results here reported for the chlorides of iron in HCl solution it appears that this rule does not always apply to ternary mixtures.

In the case of ferric chloride some density data of Kanitz[1] together with the data for HCl from the *International Critical Tables* indicate that the function f for ferric chloride is positive as usual (though the cold-end enrichment was found to be negative). His densities are relative to water at 25° and have been computed relatively to water at 4° for Table III.

[1] A. Kanitz, *Z. physik. Chemie* 22, 36 (1897).

The Production of Concentrated Carbon (13) by Thermal Diffusion

ALFRED O. NIER AND JOHN BARDEEN
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A thermal diffusion column 24 feet long and employing methane is being used for the concentration of C^{13}. Every 48 hours 308 milligrams of methane are removed in which the C^{13}/C^{12} ratio has been increased 11.5 times. The power consumption is 6490 watts and six liters/minute of cooling water is used. An expenditure of 156 kw-hr. of energy thus results in a transport of 1.5 mg. of C^{13} per 24 hours. The measured output is in good agreement with the value computed from the theory.

THE thermal diffusion method of Clusius and Dickel[1] has been used in a number of cases to produce methane in which the C^{13} content was increased. Two years ago a 24-foot column was constructed in this laboratory with which it was possible to obtain a satisfactory check[2] of the theory of operation proposed by Furry, Jones and Onsager.[3]

The question arose as to how this column might be extended to become useful for the production of methane in which the C^{13} content had been increased from a normal 1.1 percent to at least 10 times this value. The fact that brass tubing is made in lengths of approximately 12 feet and that there was available a vertical space of approximately 36 feet suggested the use of a 72-foot column broken into two parts. Furry, Jones and Onsager have shown that if n represents the transport of gas along the tube and if C is the concentration of $C^{13}H_4$ at any point along the tube and if the rate of removal of $C^{13}H_4$ from the lower end of the column is $C_{1/2}$, one may write

$$C = C_{1/2}\left[C_1(1-C_1)\frac{\exp[-nz/H]}{2A\,L_1} + 1\right]^{-1/2}, \quad (1)$$

where A and H can be computed in terms of various constants for the gas and the pressure, temperature and dimensions of the column.

The solution of (1) subject to the condition that C_1 is small is

$$C^0/C_1^0 = (1+\alpha)/[\alpha + \exp\{-(1+\alpha)2AL\}], \quad (2)$$

where $\alpha = z/H$ and L is the length of the column. Table I gives the computed concentration and transport under various operating conditions for a 72-foot column similar to the 24-foot column previously investigated.

In substituting in Eq. (2) $2AL$ was taken as

$$(0.58/p)(1.63+0.208/p), \quad (3)$$

where p is in atmospheres pressure. This value is three times the empirically measured value for the 24-foot section. H was computed from Eq. (30) of reference 3.

The concentrations given are all somewhat high as they are based upon the assumption that $C = C(1-C_1)$. Only in the cases of the high concentrations would this discrepancy be serious. It appears that operation at a pressure of one atmosphere with a drain of 0.150 g. of CH_4 per 24 hours would give sufficient concentration with reasonable economy. Moreover, operation at a pressure of one atmosphere or slightly lower would minimize difficulties should the system have any small leaks.

TABLE I. *Approximate operating data for a 72-foot column. Cold wall at 20°C, $C_1 = 0.01$.*

Pressure (Atmos.)	0.5		1.0		1.5		2.0	
z	C^{13} Atoms C^{12} Atoms	C^{13} Atoms C^{12} Atoms	C^{13} Atoms C^{12} Atoms	C^{13} Atoms C^{12} Atoms	C^{13} Atoms C^{12} Atoms	C^{13} Atoms C^{12} Atoms	C^{13} Atoms C^{12} Atoms	C^{13} Atoms C^{12} Atoms
$C_{1/2}$								
0	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
0.05	0.09	0.04	0.06	0.02	0.04	0.02	0.03	0.01
0.10	0.17	0.07	0.10	0.03	0.07	0.02	0.05	0.02
0.15	0.30	0.09	0.13	0.04	0.09	0.03	0.06	0.02
0.20	0.46	0.12	0.17	0.04	0.12	0.03	0.07	0.02
0.25	0.63	0.15	0.22	0.05	0.15	0.03	0.09	0.02

[1] K. Clusius and G. Dickel, Nature 140, 546 (1938); 27, 148 (1939).
[2] H. G. Thode, Nature 144, 8 (1939); H. L. Taylor and G. Glockler, J. Chem. Phys. 7, 851 (1939); 8, 843 (1940); W. V. Smith, Phys. Rev. 56, 703 (1939); 57, 899 (1940); A. O. Nier, Phys. Rev. 57, 30 (1940); J. Bardeen, Phys. Rev. 57, 35 (1940).
[3] W. H. Furry, R. C. Jones and L. Onsager, Phys. Rev. 55, 1083 (1939).

From the standpoint of maximum power economy it would be desirable to use a column for which the average circumference of the annular gas-containing space varied approximately inversely with the concentration along the column.[12] Such a column would not be practical to build, so one would reduce the size by a number of stages having progressively smaller cross sections from top to bottom. Such an arrangement would not give quite the output of a continuous large column but would require considerably less power.

The arrangement used in the present work is shown in fig. 1. Table II gives the dimensions and other pertinent data concerning the several sections. Section 1 is the original one, and section 2 was an extension added later. The combination operated at a pressure of 0.8 atmosphere with the hot walls at 800°C and produced 170 milligrams of CH_4 per 24 hours in which the C^{13} content had been increased by a factor of 6.0. In the fall of 1940 section 3 was added. The temperatures of the hot walls raised to approximately 880°C and the pressure to 1.4 atmospheres. Section 1 is connected to the 150-liter reservoir by means of two leads of ordinary ¼-inch copper refrigerator tubing. A pump P_1 circulates the gas. This consists merely of an iron cylinder which moves up and down with a period of approximately one second in a brass tube surrounded by solenoid actuated by a multivibrator oscillator. A small piece of rubber dam over a

TABLE II. *Data on the three sections of column.*

	No. 1	No. 2	No. 3
Length (cm)	124	124	100
Diameter (cm)	2.0	2.0	1.8
Volume (cc)			
O.D. of inner tube	1.0	1.0	0.75
I.D. of inner tube	0.75	0.75	0.55
O.D. of outer tube	1.00	1.00	1.00
I.D. of outer tube	0.95	0.95	0.95
Surface of hot wall (cm²)			
Surface of inner tube (cm²)	511	511	411
Surface of outer tube (cm²)	1.1	1.1	1.1
Surface of annular space (cm²)			
Nichrome Y heating wire (ft)			
Number of stages	14	14	14
Volume (cc)	1.6	1.6	1.00
Surface (cm²)	55.3	55.3	45.3
Surface (ratio)	1.00	1.00	1.40
Time for half sep. (hr)	9.00	9.00	5.00
Time for good sep. (hr)	27	27	15
$\Delta\Delta$	5.21	5.21	1.80
$\Delta\Delta$ (ratio)	1.44×10^{-4}	1.44×10^{-4}	1.26×10^{-4}
Volume of gas (cc)	150 liters		
Pressure of gas (atm)	1.0		

[12] Kramer, Ergeb. 15, 38, 1078 (1940).

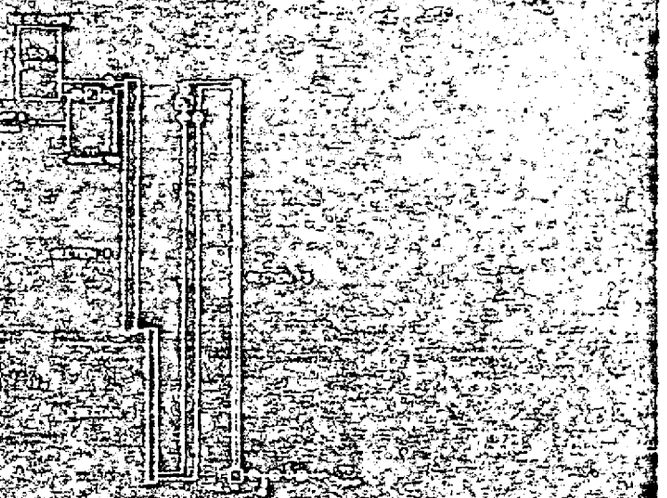

FIG. 1. Schematic drawing showing arrangement of column.

hole in the iron cylinder and a similar valve in any obstruction in the brass tube insure the unidirectional flow of gas in the tube. The pumping speed of several cc per second is sufficient to maintain equilibrium between the column and reservoir.

Section 2 is connected directly to section 1 by means of a short brass tube (length = 5 cm, diameter = 1.8 cm). Because of the limited vertical space available section 3 is set next to 1 and 2 and is connected to 2 by two leads of ¼-inch copper refrigerator tubing one of which contains a pump P_2 similar to P_1.

The construction of sections 2 and 3 was similar to that of 1 except that the steel tubing constituting the hot walls had sufficient thickness to eliminate the necessity for the extra tube used in section 1 to center the heating wire. The methane is kindly supplied by the Southern California Gas Company and is over 92 percent pure. Every 48 hours 50 cc of gas is removed from the bottom by attaching a bulb at the point marked C. Between the valves B is a small appendage which contains KOH pellets for the removal of CO_2. As the column is extremely efficient in separating heavy and purities in the methane the output is only approximately 75 percent methane. The equivalent steady drain of concentrated methane is thus 0.4 mg/hr. The C^{13}/C^{12} ratio in this is 11.8 percent. Every 3 days the contents of the upper reservoir are

changed after closing the valves between column and reservoir. The C^{D} in the reservoir is never depleted by more than 15 percent. The average C^{D}/C^{B} ratio in the reservoir is thus 0.0103. The average concentration produced by the column is hence 11.5 times.

The C^{D}/C^{B} ratio at the top of section 3 is 0.030. Sections 1 and 2 together change the concentration by a factor of 2.9 whereas section 3 produces a change of 4.0 times.

These data are not sufficient to determine empirically both the separation factor $\exp(2AL)$ and the transport (H) for the different sections of the column. It is of interest, however, to compare the observed changes in concentration with those expected from theoretical considerations. The separation factor for the 24-foot section can be estimated from the empirical expression found earlier, i.e., of (3).

This expression was obtained when the temperature difference between the walls was 300°C. Since the operating temperature difference is now about 350°, a slight change is necessary. The numerator and the first term in the denominator should be more or less independent of temperature, while the second term in the denominator should vary roughly as $(T/\Delta T)^{2}$, where T is the mean temperature and ΔT the temperature difference. The value of $2AL_1$ obtained from (3) with this correction from the change in temperature is noted in Table 1. The same expression has been used to estimate the expected values for $2AL_2$ and $2AL_3$ for the second and third sections. The value of A for the third section is taken to be the same as the A for the first section. A correction for the difference in spacing (l) was made in obtaining $2AL_2$ for the second section. These estimates assume that the correction (A_c) for asymmetry in the temperature distribution is the same for all three columns. Without further information about the operation of the columns, this is probably as good a guess as any. The transport coefficients, H, as computed from the theoretical expression of Furry, Jones and Onsager, are also listed for the three sections.

It is not difficult to compute, from these values and a combined formula similar to (2), the concentration to be expected for any given rate of drain. If methane is drawn at a steady rate of 6.4 mg/hr, the calculations yield a concentration of 12.5 times for the whole column and of 3.0 for the first two sections, as compared with the observed 11.5 and 2.9, respectively. The calculated values are thus a little higher than those obtained in practice.

The discrepancy between practice and theory could be due to a number of factors. The rate of drain is not constant but intermittent. This means that each time a sample is removed the lower end of section 3 is disturbed abruptly. This disturbance is transmitted through the entire system. Also, no doubt, due to the action of the pumps there is a certain amount of remixing at the ends of the columns, a factor which would cut down the effective over-all length. In the previous work it was shown that in section 1 there was some remixing due to some asymmetry. It might well be true that this is different for 2 and 3 than for 1. The existing line voltage fluctuation of 4 percent would also contribute to the remixing. No account has been taken in the computations of the impurities.

This apparatus has been in continuous use since the fall of 1940 and has required practically no attention aside from time spent in removing the samples from the bottom and replenishing the gas at the top. It would thus be fair to state that, including what little maintenance there is, not over 1 hours per week of a man's time was needed.

Starting from scratch, it would probably be easier to build up the column by connecting standard sections in series and parallel. The third section which is 36 feet long may be a convenient choice. With two of these sections in parallel feeding a third in series, we compute a multiplication of 11.2 times for the same rate of drain as used above. With three sections in parallel feeding one in series the factor is 14 times. Probably the values obtained in practice would be somewhat less than these theoretical values as was true in the case investigated.

In the design of any such column one would, of course, choose the heating wires of such a size that an autotransformer would not be necessary as was the case for sections 1 and 2 of our set-up.

The authors are indebted to Mr. Rudolph Thornes for his skill in constructing and assembling the columns.

[illegible] Because of the numerous approximations made, it was not worthwhile to correct H, A and K_D/K to allow for the finite construction of columns. See W. H. Furry and R. C. Jones, Phys. Rev. 57, 561 (1940).

A Confirmation of the Theory of Thermal Diffusion

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(Received July 8, 1940)

THE experiments reported by Nier[1] and Bardeen[2] demonstrated that the theory of the thermal diffusion column as given by Furry, Jones and Onsager[3] describes reasonably well the separation of the carbon isotopes using methane. Since the construction of a 40-foot column by the authors,[4] further experimental verification of this theory has been obtained.

As described previously, the column was constructed from a 2-inch pipe concentric with a 1-inch standard galvanized pipe from which the galvanizing was removed. (More favorable sizes can be obtained in steel tubes.) The difficulties originally experienced with the heating element were overcome by using four parallel loops of No. 16 Nichrome wire running to the bottom and back through 8-hole porcelain insulators. A one-quarter-inch rod running through a hole in the center of the insulators served as a method of holding them in place and facilitated removing the whole heating element when necessary. The following figures give a description of the column: width of annular space = 0.95 cm; mean circumference of annular space = 13.68 cm; area of annular space = 13.0 sq. cm; volume of annular space 14.2 liters; volume of bulb on the bottom = 12 liters; volume of reservoir on top = 24.6 liters; height of heating surface 10.9 meters.

In order to study the performance of the column, methane kindly supplied to us by the Southern California Gas Company, Los Angeles, was admitted at different pressures. Small samples were drawn from the top and bottom of the column periodically, and the ratio of the isotopes determined by analysis on a mass spectrometer. The temperature of the hot surface was maintained at an average value of 316°C., while the cold surface was that of the cold water supply, about 20°C. The average temperature difference was, therefore, 310°C. A current of 7 amperes passing through the heating element at 195 volts gave an average power consumption of about 3.25 kilowatts.

The results of several experiments at 74 cm, 50 cm, 28 cm and 19 cm of mercury are plotted in Fig. 1. The maximum concentration for a given pressure is reached after 125 to 150 hours. A pressure of 28 cm gave larger concentration than either lower or higher pressures. This is to be compared with Nier's results using a column with a spacing of 0.712 cm. The optimum pressure for that spacing was about 45 cm. A further comparison shows that the smaller spacing gives a larger separation for the same length of column. It, therefore, appears that an annular spacing of about 0.5 cm to 0.7 cm is preferable for the type of column described.

Analyses were also made on samples from the top of the column but owing to the difficulty of correcting the mass spectrographic analyses for the presence of $O^{17}H$ (mass 17 = CH_3D) they were not considered as reliable as the analyses for samples at the bottom of the column. Correc-

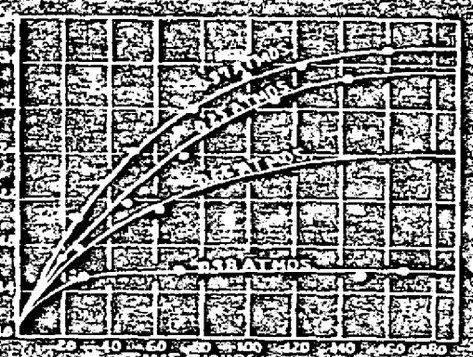

FIG. 1. Variation in the concentration of CH_3D at the bottom of the column with pressure.

[1] Nier, Phys. Rev. 57, 30 (1940).
[2] Bardeen, Phys. Rev. 57, 35 (1940).
[3] Clusius and Dickel, Naturwiss. 26, 546 (1938); 27, 148 (1939); Brewer and Bramley, Phys. Rev. 55, 590A (1939); Brewer and Bramley, J. Chem. Phys. 7, 553 (1939); Groth, Naturwiss. 27, 260 (1939); H. S. Taylor, Nature 144, 8 (1939); T. I. Taylor and Glockler, J. Chem. Phys. 7, 851 (1939); Watson, Phys. Rev. 56, 703 (1939); Cullen, J. Chem. Phys. 7, 530 (1939); Waldmann, Naturwiss. 27, 220 (1939); Nier, Phys. Rev. 56, 1009 (1939); Onsager and Watson, Phys. Rev. 56, 174 (1939); Debye, Ann. d. Physik 36, 284 (1939); Waldmann, Zeits. f. Physik 114, 53 (1939).
[4] Furry, Jones and Onsager, Phys. Rev. 55, 1083 (1939).
[5] Taylor and Glockler, J. Chem. Phys. 7, 851 (1939).

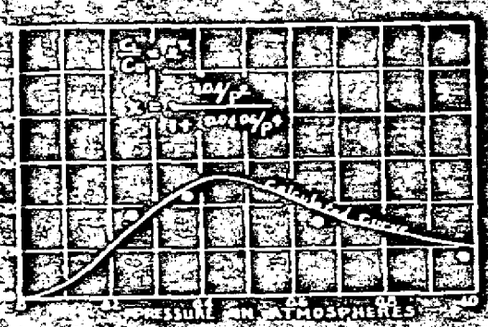

FIG. 2. Variation in the separation factor with pressure.

tions are made empirically by determining the relative heights of the peaks corresponding to masses 16, 17 and 18. The height of the $O^{16}H^{17}$ peak for water vapor varied from 25 percent to 30 percent of the height of the H_2O^{16}. Subtraction of the calculated height of the $O^{16}H^{17}$ peak from the total height at mass 17 for the sample gave the height of $C^{13}H_4$ peak. Careful regulation of the energy of the ionizing electron beam in Nier's previous analyses eliminated much of this difficulty. However, weighted mean of the values after equilibrium had been reached gave the following values for the percent of $C^{13}H_4$ at the top of the column: 74 cm = 0.70 percent, 50 cm = 0.53 percent, 28 cm = 0.40 percent, 19 cm = 0.60 percent. The corresponding ratios for the maximum concentration of $C^{13}H_4$ at the bottom divided by the concentrate at the top gives, respectively, 2.13, 1.9, 9.0, and 6.2. There was a very large separation of the impurities. The ratio of the concentration of carbon dioxide at the bottom to that at the top was often more than 1000 to 1.

The variation in the ratio of the concentration of $C^{13}H_4$ at the bottom to that at the top, C_t/C_0, can be calculated from the theoretical equations of Furry, Jones and Onsager.[3] For any given column, the dependence upon pressure can be reduced to a simplified equation of the form

$$\frac{C_t}{C_0} = e^{(a/p)(1+b/p)} = e^{\phi_0}$$

where a and b are constants and p is the pressure in atmospheres. Using the dimensions for our column and calculating the constants a and b from the theoretical equations in a manner described by Nier[4] and using his value for the coefficient of thermal diffusion, the value of ϕ_0 is given by

$$\phi_0 = \frac{1.04/p}{1+0.0106/p}$$

Figure 2 shows a comparison of the theoretical curve with experimental points. The results show reasonable agreement. The theory predicts the observed pressure for maximum separation and the manner in which the separation decreases with increasing pressure. If the observed deviations are real, they seem to indicate that the separation is less at higher pressures than would be predicted by the theory. This may easily result from mass movement of the gas or mixing due to assymmetry and irregularities in the column. This effect may also be more likely in a column with a large spacing (0.95 cm) compared to one with smaller spacing. From a study of the theoretical curves for columns of different annular space separations and upon consideration of the transport of heavy carbon, it appears that the spacing in thermal diffusion columns of this type should preferably be between 0.5 cm and 0.7 cm.

We wish to thank M. B. Mallhere, Technical Supervisor of the Southern California Gas Company, Los Angeles, for the supply of methane gas, and we are indebted to Dr. A. O. Nier for carrying out the mass-spectrographic analyses.

PRACTICAL EXAMINATION OF LUBRICATING GREASES

4. STABILITY

By M. W. WEBBER

The three preceding articles of this series dealt with the analysis of greases (pp. 18-20), the various methods for the determination of melting points (pp. 42-44), and with consistency (pp. 95-98). In the concluding instalment corrosion, low-temperature performance, water resistance and load-carrying capacity will be discussed.

THE important property of stability must be considered from several points of view, with equal consideration both in storage and in use. We have to determine and control any tendency of the grease to change its consistency, to oxidise, or to separation of its constituents. Another important aspect which is receiving increasing attention is stability of structure, especially in the case of so-called smooth or short-fibre greases for ball and roller bearings.

Stability of Consistency

In Storage.—In the first place it is essential that the grease should not harden or soften during storage. This does not call for a special method of test, but the grease should be kept under observation in typical storage conditions and check penetration tests made at suitable intervals. Normally observation over a period of three months is sufficient, but extension to six or twelve months is desirable for a new product. The author suggests the making of the first four tests at weekly intervals and then the checking of the penetration at the end of each calendar month. The curve obtained by plotting penetration against time of storage will show clearly any tendency to change of consistency.

In the case of greases designed for ball and roller bearings a useful procedure is to fill a bearing with the grease and keep it for extended periods of time both at high and low temperatures. The stability of the lubricant is determined either by the ratio of the penetration figures obtained before and after working or by comparison of the torque required to rotate the bearing at the beginning and end of the test run.

Chemical Tests in Relation to Stability

Tests carried out for other purposes are of value in predicting and controlling the consistency stability of greases. Thus excessive free fatty acid will frequently cause softening of the grease in storage and when subjected to heat or working. Greases containing free alkali are usually stable in storage and when worked at normal temperatures, but they are liable to hardening followed by separation when heated. Control of the water content is also necessary to help ensure stability.

Resistance to Oxidation

Tests to determine the stability of greases as regards their resistance to oxidation may be divided into three groups.

The first group comprises the tests involving the exposure of the grease to the atmosphere in controlled conditions of temperature and, if desired, humidity and for definite periods of time. After exposure the grease is examined to determine any change of consistency or addition to measuring oxidation by the fall in pressure of oxygen the sample of grease can be examined after any desired exposure to determine any change of consistency or development of acidity.

The third group of tests is concerned with the resistance to oxidation of greases when in ball and roller bearings both in storage and service conditions. The static form of test is similar to that described above on a test for stability of consistency. The test bearings are filled with the grease and the starting torque determined after storage. An accelerated test of this type is to be found in Appendix III to M.A.P. (Ministry of Aircraft Production) specification D.T.D. 143C, where the bearing is subjected to a temperature of 150° C. for two hours. The dynamic test consists of heating the grease when heated in a cup by means of a ball or roller bearing and afterwards examining the grease for change of consistency or development of acidity.

Stability of Structure.—The above test is also used to determine stability of structure of the grease. The importance of this is well illustrated by the following extracts from the "Agitation Tests" of the Murlin-Rockwell company as reported by Klemgard.

Change in Structure.—This condition is usually accompanied by a change in consistency and next to permanent separation it is the most undesirable characteristic a lubricant can possess. In the case of some fibre greases the change is so drastic and takes place so suddenly as to be somewhat startling to the observer. This is particularly true of most greases classified as short fibre in structure. At a certain point in the test this type of grease will suddenly turn to a longer fibre and assume the consistency of soft wax, balling up around the shaft, leaving the bearing as dry as though no grease had ever been applied. Buttery greases often change to fibre greases, and *vice versa*, under the combined action of heat and agitation.

In many cases the action of heat alone is sufficient to change the structure of a grease. A useful procedure is to heat the grease for two or three hours at a temperature say

the grease and to store them in the usual wrappings. In this method one of the freshly filled bearings is mounted in a pulley and fixed on a spindle, so that by means of a thread wound round the pulley, a determination can be made of the minimum weight or torque required to commence rotation of the bearing. Alternatively, the time required for the first complete rotation can be determined using a definite weight. In most cases it is, however, more useful to determine the minimum "unsticking" weight. Then similar tests are made with samples of the stored bearings at intervals as suggested above. Again plotting the starting weight or torque (or time of rotation) against time of storage will give extremely useful information.

This test is not concerned solely with change of consistency but also with the effects of corrosion, oxidation, and syneresis.

To obtain consistent results it is advisable to rotate the freshly filled bearings, which should each contain the same weight of grease, so as to distribute the lubricant evenly, and the various check tests must all be made at the same temperature. Further, the temperature at which the grease is stored should approximate to that expected in practice, especially if the grease is intended for use in a hot climate.

Thixotropy.—A quite different aspect of stability of consistency is resistance to mechanical breakdown on working. Greases can be quite stable in storage, but when subjected to shearing stress in the bearing they may break down either to an excessively soft grease, or to a fluid oily condition or even separate into oil and soap. In Part 10 of this article (Petrol. Lubr., 1944, 7, 96) reference was made to the desirability of extending the period of working in the standard methods for the determination of worked penetration, preferably by the use of a motorized worker, in order to eliminate manual fatigue and also the personal element.

Reference was also made to the development of breakdown tests using ball and roller bearings. Such tests consist of working or beating

In the second group we have accelerated tests where the grease is exposed in an enclosed vessel to an atmosphere of oxygen, usually at fairly high temperatures and pressures. The apparatus most commonly in use is that developed by the Norma-Hoffman Bearings Corporation, and this method of test has been included in official specifications.

The apparatus consists of a stainless steel bomb which is fitted with a suitable valve to allow flushing and filling with oxygen, and also a pressure gauge. Twenty grams of the grease are spread in thin uniform layers in five shallow dishes which are made of glass or metal similar to that with which the grease is expected to be in contact during use. The dishes are placed on the racks of a stainless steel holder which fits neatly in the bomb. The lid, which carries the valve and pressure gauge, is bolted on to the bomb, using a lead gasket to ensure pressure tightness. The bomb is purged of air by filling with oxygen to 100 lb. per sq. in., releasing and refilling two or three times. Then the bomb filled with oxygen at 100 lb. per sq. in. is immersed in a constant temperature bath at 210° F. and the oxygen bled off until a constant pressure of 110 lb. per sq. in. is reached in about two hours. Usually the test is continued for a period of 100 hours with the bomb immersed in the bath at 210° F. Oxidation of the grease is measured by the resultant fall in the pressure registered by the gauge. A typical specification requirement is that the fall in pressure after 100 hours shall not be greater than 5 lb. per sq. in. Useful information can also be gained by noting the pressure as recorded every two hours against the total time of immersion in the bath.

With nearly all greases it will be observed that the rate of fall of pressure is at first relatively low and uniform, and that after a fairly definite time there is a marked increase in the rate of pressure drop. The originators of the test have termed the first stage the "induction period" and measurement of this is of great value for research purposes. In

while heating as well as after cooling. In addition to the reversal of fibre structure mentioned above it will be found that smooth sodium greases made by milling block greases will frequently revert to the hard block consistency, especially if the soap flocs include resins.

The question of mechanical breakdown of structure is equally important, as those fibrous greases which shear stress reduces to soft buttery consistency will frequently give trouble with leakage from ball and roller bearings, universal joints, etc., and excessively high consumption in other applications. In many cases any tendency to breakdown of structure will be indicated by the ordinary working of the grease in the standard worker, especially if the test is prolonged beyond the specified 60 double strokes.

Resistance to Separation

Separation of the constituents of a soap base grease can occur either during storage or when the material is subjected to heat, pressure and/or mechanical stress. The latter will be determined during such tests as those mentioned above for mechanical breakdown of consistency and structure.

Stability in Storage.—Separation in storage is usually confined to syneresis or bleeding of oil from the grease. Examination of sample containers is usually a hole is scooped in the surface of the grease. Should the grease not be stable it will be found after storage, especially in hot situations, that free oil can be observed in the hole and in some cases poured off and measured, although the test can hardly be regarded as quantitative. Many specifications now include a test for resistance to bleeding. The procedure is to weigh say 10 grammes of the grease into a cone made of wire gauze, usually 60 mesh. A suitable cone can be made out of a semicircle of about 2½ inches radius of the gauze; this is folded into a cone and the seam soldered.

The grease should be carefully packed into the apex of the cone, finishing with a convex upper surface so as to avoid trapping any separated oil. The cone is supported in a weighed beaker and placed in a constant temperature oven. A suitable test temperature is 100° C. and the duration of the test is, in various specifications, anything from eight hours to one week. The shorter time is usually sufficient for soft greases made with extremely light oils, and the longer time is used for greases of high soap content and made with viscous oils. After heating, cone and beaker are removed from the oven, allowed to cool and the beaker is reweighed. The weight of oil collected in the beaker gives a fairly accurate quantitative measure of the liability of the grease to bleed.

Stability to Heat

Stability to heat at moderate working temperatures — say, 50-80° C. — is usually rather a question of syneresis rather than of ordinary separation, and is covered by such tests as that described above. A concerns the stability of calcium soap greases when heated at temperatures above their melting point, with consequent dehydration. The standard test in this country is as follows: 10 grammes of the grease are packed in the bottom of a tall 40 cu. cm. glass beaker, which is placed in the constant temperature oven for one hour at 120° C. The molten grease is then poured on to a clock glass and allowed to cool to room temperature. It is usually specified that the grease shall remain undisturbed for 24 hours before examination, but in nearly all cases separation is immediately apparent. Even if the grease does not show separation it is desirable that it should not harden excessively after heating and cooling. This test is a modification of that developed by British Timken Ltd., which requires more manipulation. In the Timken test 25 grammes of the grease are heated for 30 minutes in a flushing oil at 350° F. (176° C.). The grease is stirred for alternate minutes, which assists dehydration. In the shorter period of heating. After heating for the specified time the molten grease is poured into a glass tube 3 in. long by ½ in. diameter, one end of which has previously been sealed by means of a cork. After solidification of the grease the cork is removed and the grease carrying tube is supported by two pieces of flattened wire wound vertically round the tube, which is then suspended over a suitable receiver for 48 hours. Any separated oil is calculated as a percentage of the grease, and if this should exceed 6% the lubricant is considered as failing to pass the test.

Heating at 120° C. does not convey information concerning soap greases, but it is more valuable to make the test at 10° or 20° C. below the drop point of the grease. A convenient method of test is that described in Appendix V to M.A.R. specification D.T.D. 410. The grease is placed in a metal dish which is fitted with a close-fitting lid of which the top is made of perforated metal of approximately 81 holes to the square inch. After heating — for 3 hours at 150° C. in the case of greases to D.T.D. 410 — and cooling the lid is placed on the dish and the assembly is weighed. The dish is then inverted on a wad of filter paper and allowed to remain for one week. After this any adhering oil is wiped from the lid and the assembly is again weighed. The loss in weight is taken as a measure of the stability of the grease and is most valuable in the specification in question to exceed the similar tests can be used for block greases and they are of value in predicting service performance.

Stability to Pressure

Owing to their wet structure, soap base greases are liable to separation of oil if subjected to pressure, and it is necessary to control this characteristic. Most methods in use are similar to that devised by Herschel. The grease is subjected to pressure by means of a gun and the grease filtered through an Alundum thimble. The separated oil can be collected and measured. Difficulties arise owing to the clogging of the filter, and so far as the author is aware there is no method of test for stability to pressure either embodied in official specifications or otherwise in vogue.

Standard Methods

The need for standard methods of determining stability is obvious, but there is great difficulty in developing methods which will give reproducible results and which will give a true indication of the results to be expected in actual storage and service. Also it will be observed from the comments made above that the different aspects of stability are closely interwoven, and it is not easy to say in each case which property is being determined.

REFERENCES

[illegible]

LEADED FUELS (Continued from page 11)

Another method, which does not involve dismantling the engine, and which has the additional advantage of affording the same protection to the cylinder walls and piston head, is to use an upper cylinder lubricant in the petrol, carrying an suitable proportion of colloidal graphite dispersed in oil. Oiling has been investigated in considerable detail by visual exposure trials, gravimetric tests, and by the electric potential method. All have shown that the film of graphite developed under these conditions definitely does not encourage corrosion. But on the other hand it is small protection against [illegible] which relate to the corrosion [illegible]

A graphite film is manufactured [illegible] cleaned from dust, oxides, etc., and its adsorbed gas layer driven off in *vacuum*. It is still very likely to be inhomogeneous in a polycrystalline material the surface will probably consist of patches belonging to different crystal faces, and as different crystal faces have different hardness, solubilities, electro-chemical potentials, etc., all these properties will vary from

reconditioned engines this may be added to the petrol in proportion of up to 1 fluid ounce per gallon. After the running-in period is completed the proportion may be reduced or it may be colloidal graphite upper cylinder lubricant employed.

A road test has just been completed on a 6-ton lorry which has used this upper cylinder lubricant for 32,000 miles in conjunction with the Swarfine leaded petrol. On dismantling the engine, the cylinder walls, valve seats and valves were found to be in excellent condition, only one of the valves needed replacement and that for reasons not connected with the use of leaded petrol.

An extended series of tests on a 4-cylinder petrol motor on the test bench showed how beneficial this type of lubrication is. When the engine was dismantled after the tests it was found that a fine graphoidal film had developed on the cylinder walls and piston heads. The lubricating properties of this film, especially on cold starting, when the supply of lubricating oil to the cylinder walls is very meagre, was clearly shown by the dynamometer reports.

A prolonged run was made with this engine using a petrol carrying a very much higher graphite content than would be used in practice, with the object of ascertaining whether the deposit of graphite showed any tendency to build up to inconvenient proportions. No such tendency was indicated: the piston heads, cylinder walls, combustion chamber, valves and valve stems were all examined and found to be in excellent condition after the test.

Graphite v. Corrosion

The question is sometimes raised whether the presence of this graphite film on ferrous metals is likely to encourage corrosion, especially in the presence of moisture condensed from the exhaust gases. The contact surfaces involved, and thus has the advantage over oil or grease, whether viewed as a lubricant or as a means of preventing the ingress of the foreign substances.

It may be mentioned, however, that in so far as the problem under discussion is concerned, we have here a case of prevention that cure. If the fuel compounds have been allowed to form a deposit on the valves and other parts, a graphite film will not remove them, but it would have prevented adhesion if the film had been there in the first place. With low friction produced, the loss of the film by wear and tear is very small, and it can be maintained by the continued use of a graphited upper cylinder lubricant in concentrations lower than those which may be profitably employed in the initial [illegible]

REFERENCES

[illegible]

MECHANISM OF LUBRICATION

(Continued)

[illegible] Neville states: "There appears to be no metal which, when polished and examined even under low powers of the microscope, will present a surface of uniform appearance even approximately approaching that of well-polished glass. The metal surface invariably contained pits or non-metallic inclusions, iron carbide or traces of manganese in the form of grey areas with well-defined but irregular outlines."

Electron diffraction patterns of polished cast iron showed that, when covered with graphite, the brittle constituent of the metal are pulverised and removed, and only the constituent in contact with the rubbing surface of the specimen.

Finally, even if the surface is not quite [illegible] tions, but it ought to be borne in mind when these theories of friction and lubrication are scrutinised which [illegible]

(To be continued)

REFERENCES

[illegible]

О ДЕЙСТВИИ ХЛОРНОВАТИСТОЙ КИСЛОТЫ НА ТЕТРАМЕТИЛ-1,1,4,4-БУТИН-2-ДИОЛ-1,4

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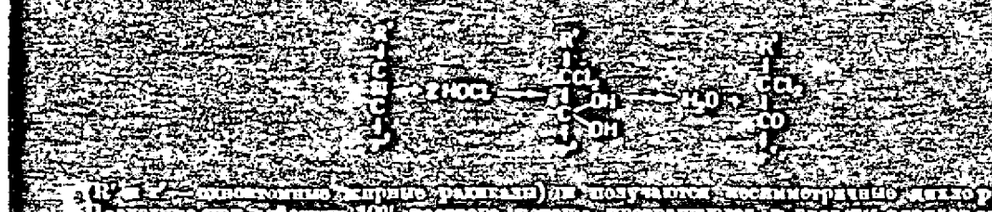
Лаборатория высокомолекулярных соединений

Настоящая работа является продолжением исследования превращений в области метилированных гликолей при действии различных реагентов.

Известно, что при действии хлорноватистой кислоты $HOCl$, $HOBr$ на третичные гликоли в 50% растворе, получаются:

В ацетиленовом ряду действие хлорноватистой и бромноватистой кислот идет преимущественно на тройной связи.

Так, А. Якорский [1] показал, что при действии хлорноватистой кислоты на бутинилированные углеводороды идет присоединение по алкилирующей схеме:

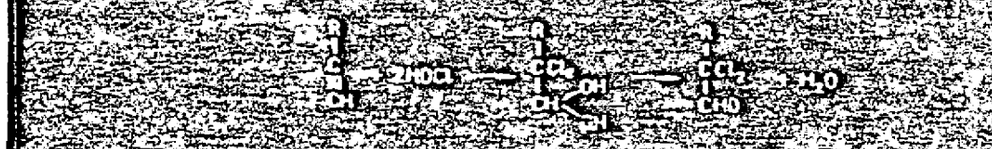


(R — алкилированный фенил, радикал) и получается хлорпиролидон.

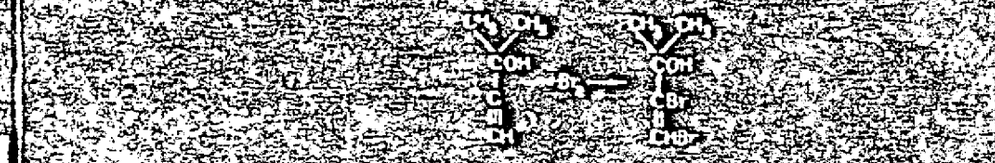
Позднее при действии 10% раствора, гликоли превращаются в кислоты жирного ряда:

(R — остаток при реакции двухатомного фенола).

Затем Ваторф [2] показал, что при действии гипохлоритной кислоты на полиметилированные углеводороды присоединение происходит по схеме:



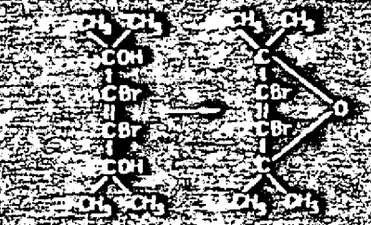
Согласно фактам из литературы, Кристаевский и Петрунин [3] и несколько позднее (Поляк [4], Дзюра [5], Кристаевский [6], Жугаев [7]) показали, что фрагментация гликолей состоит в следующем:



ному также в первую очередь бромоспирты и бромгликоли, содержащие в молекуле формально одну этиленовую связь, но являющиеся по отношению к галоидам вполне насыщенными.

В случае бромоспиртов на примере диметилэтиленгликоля [9] замещение действие брома направлено в сторону замещения следующим образом:

В случае же гликолей шестичленного при бромировании образование воды с бромгликолем в непредельных бромоокисях [1].

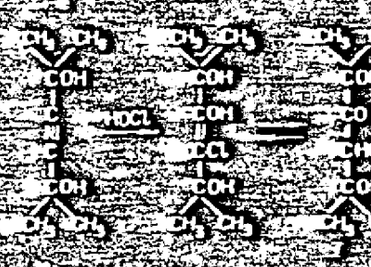


При кипячении получаются исключительно циклоокиси [7].

Таким образом шестичленовая связь (в спиртах и гликолях) в соединении с третичными группами приобретает некоторую ненасыщенность и может сама как этиленовая связь.

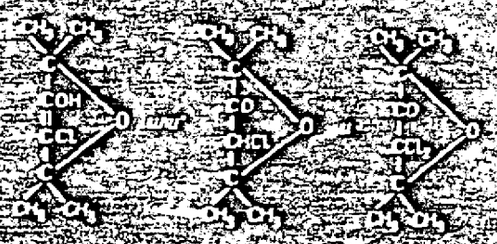
Учитывая это обстоятельство, возможно, что при действии галоидоводородных кислот на диметилэтиленовые гликоли реакция в ряде случаев не пойдет, что мало вероятно, или же будет присоединяться к гликолям одна молекула или две молекулы галоидоводородной кислоты по тройной связи.

При присоединении к тетраметилбутандиолу одной молекулы хлорноватистой кислоты следует ожидать получения изомерной смеси следующего состава:

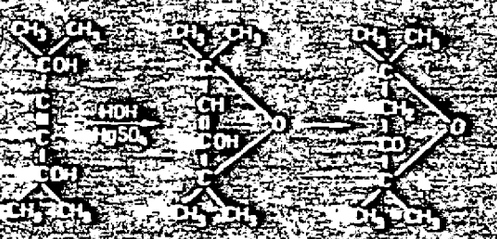


Присоединение второй молекулы хлорноватистой кислоты приведет к получению хлоркетонового строения:

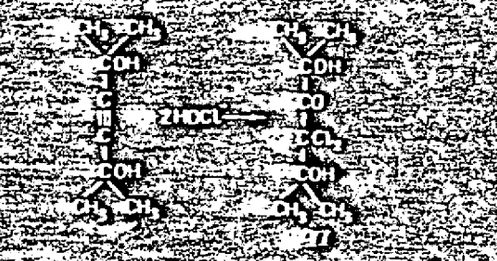
Но если присоединение хлорноватистой кислоты будет происходить с одновременным выделением воды, то как в первом случае, так и во втором могут образоваться фурановые группировки:



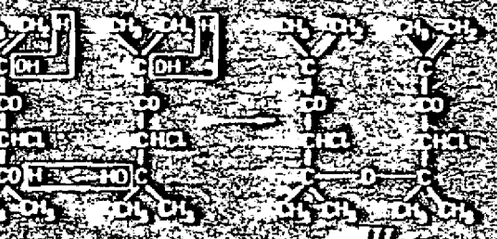
Малоучтное превращение, т. е. образование фурановых колец было наблюдено Лоуном [1] при действии серноокислой окиси трутилгликоля:



Произведенное нами исследование показало, что действительно, при действии хлорноватистой кислоты на метаметилбутинол $(CH_3)_2COH-C\equiv C-H$ присоединение идет по тройной связи с образованием двух структур монохлоркетона состава C_5H_7OCl (I) и дихлоркетона (II):



состава $C_5H_6OCl_2$ — также непредельного продукта с т. пл. 122—123°, который первоначально мы принят нами за непредельную монохлоркислоту состава $C_5H_8O_3Cl_2$. Более подробное его изучение показало, что полученный продукт бимолекулярен, отвечает формуле $C_{10}H_{12}O_2Cl_2$ (III) и образование его может быть представлено следующей схемой:



III

Указанная формула хорошо согласуется со следующими свойствами. Молекулярный вес найден равным 321, 320, 335, 316. Вычислено 335. Вещество содержит две этиленовые связи, согласно данным, полученным при гидрировании и при действии гидроперекиси бензоила, окрашивается тетранитрометаном в желтый цвет, раскисляет щелочный раствор перманганата, хлор содержит в согласии с указанной формулой, дает молекулярную рефракцию (в бензоле), хорошо согласующуюся с предложенной формулой, принимая две этиленовых связи, два карбонильных кислорода и один окисный: найдено MR_D 87.16, вычислено для $C_{11}H_{20}O_3OCl_2$ 86.66, EM_D 0.51 (вкладывания объясняется сопряжением этиленовых связей с карбонильными группами. Реакция Перевалинова на гидроксильные группы — отрицательная. Реакции на кетонные группы (с семикарбазидом, гидроксиламином, п-нитрофенилгидразином) в обычных условиях не получаются, что может иметь место при близком расположении кетогруппы к этиленовым связям.

Однако при стоянии пробы с семикарбазидом в течение шести месяцев при обыкновенной температуре выпадает кристаллический семикарбазон с т. пл. 233°.

Также следует отметить пассивность данного вещества к брому в хлорированном растворе (метод по Перлину) и к иодноватистой кислоте (определение иодного числа по Маргошесу).

Механизм образования этого бимолекулярного продукта из монохлоркетона несовсем ясен. Трудно допустить, чтобы он представлял собою продукт первичный. Так как при очистке получаемых продуктов реакции хлоргидрина с хлорноватистой кислотой была применена фракционированная перегонка, можно думать, что при повышенной температуре создались благоприятные условия для образования данного соединения из первоначально образовавшегося монохлоркетона. В крайне незначительном количестве все же удалось выделить из масла после первых перегонок кристаллический продукт в виде бесцветных иголочек с т. пл. 85°. При его исследовании качественно определено наличие хлора, гидроксилов, кетонной группы (семикарбазон с т. пл. 233°), отсутствие двойной связи, а молекулярный вес, определенный по Расту, дал цифру 210 вместо вычисленного 194.5 для монохлоркетона C_4H_7OCl.

Необходимо отметить, что сырой продукт реакции хлорноватистой кислоты с тиконом, жидкое, представляет собою масло желтоватого цвета, значительно растворимое в воде. При стоянии на него выпадает в незначительном количестве кристаллы дихлоркетона с т. пл. 103°, а при очень долгом стоянии (10) при начальном монохлоркетона с т. пл. 85°. При перегонке в вакууме продукт претерпевает какие-то изменения. Уже при незначительном нагревании (40—60°) он краснеет, кипит в широких пределах, а вышекипящие фракции дают красиво флуоресцирующие красным и зеленым цветом масла, из которых выделяются кристаллы сначала с т. пл. 103—104°, затем 122—123°.

Значительные количества жидких фракций представляют собою сложные хлорсодержащие смеси, не поддающиеся разделению перегонкой без разложения.

При перегревании этих жидких фракций получается смолообразное вещество, из которого выделяются в очень незначительном количестве розоватые кристаллы с т. пл. 172—175°. Этот продукт не содержит гидроксилов, нерастворим, содержит хлор и имеет молекулярный вес равный 301. Полностью охарактеризовать данное соединение не удалось.

Экспериментальная часть

Диметилиодуцинол получался через магний-органическое соединение по Иошану [7], т. пл. 94—95°.

Хлорноватистая кислота готовилась хлорированием мочевины с последующим разложением монохлормочевины водой по Лотеру [8].

Взаимодействие тетраметилбутиндиола с хлорноватистой кислотой

Для того, чтобы иметь обратного возвращения диолов при реакции взаимодействия тетраметилбутиндиола с хлорноватистой кислотой, поступали в работе, отлично от предыдущих опытов.

40 г тетраметилбутиндиола помещались в бутыль, куда же прибавлялся 10% раствор монохлорноватистой (682 г вместо 236 г). Реакционная смесь помещалась на качалку и встряхивалась в течение 30 часов.

Образовавшееся на дно бутыли масло отделялось и промывалось водой. Промывные воды и реакционная смесь после отделения масла подщелачивались раствором соды и взбалтывались эфиром. Эфирные вытяжки вместе с жидкой массой продукта реакции сушились плавленой углекислой солью. После отгонки эфира сырой продукт в количестве 60 г подвергался фракционированию в вакууме (1—2 мм). При первых перегонных жидких фракций выделяются кристаллы дихлорметина с т. пл. 103—104°. При последующих разгонках жидких фракций образуются кристаллы с т. пл. 122—123°.

Наряду с этими кристаллическими продуктами получается ряд жидких маслообразных фракций, из которых при продолжительном стоянии (до нескольких недель) тоже выделяются кристаллы с т. пл. 122—123°.

При перегревании этих жидких фракций получаются темнокрасные смолообразные остатки, из которых выделяются розоватые кристаллы с т. пл. 172—175° в незначительном количестве.

Таким образом в результате всех опытов получен ряд кристаллических и жидких продуктов, которые мы подвергались всестороннему исследованию. Кристаллы очищались многократной перекристаллизацией из различных растворителей, в результате чего выделены две совершенно чистые фракции с т. пл. 103—104° и 122—123°.

Для кристаллических продуктов было определено содержание углерода и водорода, молекулярный вес, молекулярная рефракция, количество гидроксильных групп, хлора, присутствие кетонной группы по получению оксима, наличие двойных связей. Для жидких продуктов определялось содержание хлора. Ниже приведены результаты, указывающие на наличие смеси продуктов и разложения при повторных перегонках.

Исследование продуктов реакции взаимодействия тетраметилбутиндиола с хлорноватистой кислотой

1. Исследование кристаллического продукта с т. пл. 103—104°

Дихлорметин т. пл. 103—104° (из бензола, спирта). Белый кристаллический продукт в виде тонких на легких иголочек или листочек, хорошо растворимый в воде, спирте, эфире, ледяной уксусной кислоте, хлороформе. При кипячении с аммиачным раствором окиси серебра дает прекрасное серебро.

0.0738 г дают 0.1141 г CO_2, 0.0400 г H_2O.
0.1172 г дают 0.2268 г CO_2, 0.0663 г H_2O.
Найдено %: 42.31, 42.99 и 6.07, 6.00.
$C_8H_{12}O_2Cl_2$. Вычислено %: 42.11, 6.11.

Определение хлора по Шенигеру:

0.0100 г, 0.0082 г дают 0.1888 г, 0.0831 г AgCl.
Найдено %: 46.39, 46.91.
$C_8H_{12}O_2Cl_2$. Вычислено %: 46.00.

Определение гидроксильных групп по методу Чугаева-Цересвитинова

0,1832 г вещ.: 36,7 мм СН4 (16°, 721 мм).

Найдено % OH 11,31

$C_{11}H_{10}O_4$. Вычислено % OH 11,36

Определение молекулярного веса по Расту

0,009 г вещ.: 0,061 г камф., Δt 16°.

Найдено M 318

$C_{11}H_{10}O_4$. Вычислено M 234

Определение молекулярной рефракции в растворе спирта

% содержания исследуемого вещества в растворе 8,015

% содержания растворителя 91,985, плотность спирта

0,79135, плотность раствора d 0,81625

Для спирта n_D 1,36211

Для раствора n_D 1,36817

Найдено MR 51,08

$C_{11}H_{10}O_4$. Вычислено MR 51,08

Для доказательства строения 2-хлорбензоина был получен диоксим (по Мейеру и Янг [11]). На одну частицу 2-хлорбензоина взято 5 частиц гидрохлорного гидроксиламина в водном 10% растворе и около 2 г сверх прибавлено теоретическое количество соды. Вес вещества 0,5 г, гидроксиламин солянокислый 0,901 г, соды 0,69 г, воды 8,13 г. Вещество растворялось в спирте. Реакционная смесь нагревалась на водяной бане с обратным холодильником в течение нескольких часов. По окончании реакции спирт отогнался, раствор подщелачивался и брались эфирные вытяжки. Оставшееся по испарении эфира кристаллическое вещество было перекристаллизовано из спирта. Вещество трудно растворимо в воде, лучше в эфире и хорошо в спирте. Т. пл. 145—146°.

Определение азота по методу Кельдаля (по Гинзбургу)

Для поглощения аммиака брали точно 20 мл 0,09 н. H2SO4. По титрованию избыток серной кислоты 13 мл 0,09 н. NaOH.

Найдено % N 8,01

$C_{11}H_{10}O_4$. Вычислено % N 8,55

Высказывалось, что синтезированное вещество представляет собой смесь 1,1,4,4-диоксо-1,4-бутиен-2,3-дифенилена с 1,2-диоксо-1,2-бутиен-2,3-дифенилена по следующим формулам:

2. Исследование кристаллического продукта с т. пл. 122—123°

Кристаллы белого цвета в виде квадратиков с т. пл. 122—123°. Они совсем нерастворимы в воде, трудно в спирте, лучше в эфире и хорошо в бензоле.

0,0621 г вещ.: 0,128 г CO_2; 0,0[illegible] г H_2O.

0,0[illegible] г вещ.: 0,1[illegible] г CO_2; 0,0[illegible] г H_2O.

Найдено % C 5[illegible],[illegible]; H 7,[illegible]

$C_{11}H_{10}O_4$. Вычислено % C 57,3; H 7,[illegible]

0,1704 г, 0,1706 г вещ.: 0,1[illegible] г, 0,1[illegible] г AgJ (Цейзель).

Найдено % OCH3 20,[illegible]; 20,[illegible]

$C_{11}H_{10}O_4$. Вычислено % OCH3 21,2

Определение [illegible] по Гаусу

0,008 г, 0,009 г, [illegible] 0,008 г, 0,011 г, [illegible] 0,115 [illegible]

Найдено: [illegible] 21,81

С.Н. 0,05 [illegible] 1,8851

Определение [illegible] по Гаусу

0,017 г, 0,18 г, [illegible] 175 г, [illegible] 0,016,00

Найдено: [illegible] 31,56

С.Н. 0,05 [illegible] 1,8851

Определение [illegible] при рефракции в растворе белка

[illegible] содержание исследуемого вещества в растворе 3%

[illegible] содержание растворителя, [illegible] плотность [illegible] при 20° [illegible] 0,8[illegible] г/см³

[illegible] плотность растворителя при 20° [illegible] 0,9[illegible] г/см³

[illegible] (белка) [illegible] 1,0[illegible]

[illegible] растворителя [illegible] 1,0[illegible]

[illegible] С.Н. 0,05 [illegible] Найдено: [illegible] 5,10 [illegible] 0,01

[illegible] С.Н. 0,05 [illegible] Найдено: [illegible] 8,[illegible]

Количественное определение [illegible] (титрованию по АСА [illegible])

[illegible] 0,110 [illegible]

Пробирка [illegible] № 1 [illegible] [illegible] № 1 [illegible] (1 мр — [illegible])

[illegible] 20 мл [illegible] 0,03 мл)

[illegible] [illegible] [illegible]

Погонность мотора 18,15 мл

[illegible] 19 мл [illegible] 2 [illegible] 8,8 мл [illegible]

[illegible] 0,0303 [illegible]

Пробирка № 1 г, [illegible] [illegible] № [illegible] [illegible] — [illegible] 0,1 [illegible]

19,9 — 7,8 мл)

Погонность мотора 12,22 мл

Найдено: [illegible] 5,20 [illegible] 0,1 [illegible] 7,0,0 мл

Опыт № [illegible] (по [illegible])

Хлорфенолкрасный

0,1016 г, 0,001 [illegible]

Прибор [illegible] 10 мл [illegible] 0,116 [illegible] [illegible] 12 мл II [illegible] [illegible] Концентрация [illegible] 0,5%

Опытные [illegible] при комнатной [illegible] [illegible] 20 суток. Результаты [illegible] [illegible] 1.

Таблица 1

[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]

Выводы: [illegible]

Цвет хлорфенола, кристаллы в электронных [illegible] Хорошо растворим в воде, спирте, эфире, уксусной кислоте.

На [illegible] реакции с тетранитрометаном отрицательная, на алифатическую группу по Церевитинову — положительная, на кетонную группу положительная — получен комплексон [illegible] 213).

Определение хлора по Степанову

0.0700 г, 0.0315 г вещ.: 0.0512 г, 0.0150 г AgCl.
Найдено %: Cl 18.07, 17.61.
$C_8H_{15}O_3Cl$. Вычислено %: Cl 18.23.

Определение молекулярного веса по Расту

0.00586 г вещ.: 0.0591 г камфоры, Δt 19.7°.
Найдено: M 203.4.
$C_8H_{15}O_3Cl$. Вычислено: M 194.5.

Анализ жидких фракций

В результате многократной фракционировки масла получен ряд фракций, содержание хлора в которых колеблется от 30 до 42%.

Для монохлорида $C_8H_{13}O_3Cl$ вычислено 18.90;
Для дихлорида $C_8H_{14}O_3Cl_2$ „ 31.00;
Для трихлорида $C_8H_{13}O_3Cl_3$ „ 43.04.

Выводы

Данная работа показала, что реакция взаимодействия хлорноватистой кислоты с тетраметилбутиндиолом протекает в двух направлениях:

1. Происходит присоединение двух молекул хлорноватистой кислоты по тройной связи с образованием дихлоркетона состава $C_8H_{14}O_3Cl_2$ с т. пл. 103—104°.

2. Присоединение одной молекулы хлорноватистой кислоты с образованием первоначально монохлоркетона, который почти нацело, во время фракционированной разгонки продуктов реакции, реагирует со второй молекулой монохлоркетона с выделением трех молекул воды и образованием продукта состава $C_{16}H_{24}O_3Cl_2$ с т. пл. 122—123°.

ЛИТЕРАТУРА

[1] А. Фаворский, ЖРФХО, 26, 559—603 (1894). — [2] Витторф, ЖРФХО, 32, 88—117 (1900). — [3] Крестинский и Перованцова, Проток. Лгр. хим. общ., [illegible] 1935. — [4] Иоцич, ЖРФХО, 35, 430 (1903). — [5] Дюпон, Ann. Chim. [8], 30, 518—524 (1913). — [6] Крестинский, ЖРФХО, 58, 1067 (1926). — [7] Кругляков, ЖОХ, 6, 925—931 (1936). — [8] Дюпон, Ann. Chim. [8], 30, 536 (1913). — [9] Иоцич, ЖРФХО, 34, 242 (1902). — [10] Легоен, C. R., 155, 681—683 (1912); Zbl., 1922, III, 40. — [11] Мейер и Ним, Ber., 75, 1164. — [12] Прилежаев, ЖРФХО, 42, 1387 (1910).

Поступило в Редакцию
[illegible]5 декабря 1939 г.

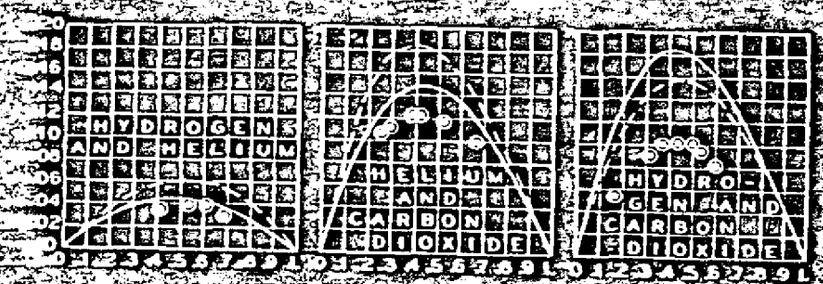


Fig. 1. Equimolecular fraction of the heavier gas in the cold chamber plotted against the mole fraction of the heavier gas in the cold chamber. Data of Elliott and Masson, temperatures about 0° and 500°. Solid curves from Eq. (7), broken curves from Eq. (1).

$(1 - \alpha)\, d \ln \gamma / d \ln T$). In the limit, when the bombardment is not accompanied by a return flow, $\beta = \alpha\gamma/T_1$, the factor becomes zero, and thermal separation vanishes.

Equation (12) can be applied to find the theoretical effect of foreign gases on the separation of two gases of kinds 1 and 2. No experiments seem to have been made of this kind.

We have for gas 2

$$d \ln x_2/d \ln T = (1 - m_2/\bar{m}_{1,2}\alpha), \quad (13)$$

from which together with Eq. (12)

$$d \ln (x_1/x_2)/d \ln T = m_2/\bar{m}_{1,2}\alpha - m_1/\bar{m}_{1,2}\alpha$$

$$d \ln \gamma/d \ln T = \bar{m}_{1,2}\alpha. \quad (14)$$

According to this equation, the relative separation of two gases by thermal diffusion, defined as the left-hand side of Eq. (14), is inversely proportional to the mean root molecular weight of the mixture and it can therefore be increased by addition of a third gas of low molecular weight to decrease this mean. The limit of the mean for addition of gas 3 is m_3 approached as the composition approaches that of pure gas 3, and at the limit the effect of adding gas 3 would be to multiply the relative separation by the ratio $(m_3 - \bar{m}_{1,2})/m_3$, where x_1 and x_2 are the mole fractions before addition of the third gas. For the separation of the chlorine isotopes 35 and 37 by thermal diffusion of hydrogen chloride the separation would be multiplied in the limit a little better than four times by the use of hydrogen as third gas. This consequence of Eq. (14) has recently been pointed out by the author.[10]

[10] J. L. Gaunt, J. Chem. Phys. 7, 748 (1939).

Comparison with Experimental Results

Figure 1 shows the separations found by Elliott and Masson[11] for three mixtures of gases over practically the same temperature intervals, about 0° to 500°. The separations in percent are plotted against the percent of heavy gas in the cold end, though average percent of heavy gas is more convenient for theoretical purposes. The continuous line is that given by the integrated Eq. (7) and the broken line is that given by Chapman & approximate Eq. (1) with approximate integration.

The agreement of Eq. (7) in the first two cases (H_2 and He, He and CO_2) is impressive, but the disagreement in the third case (H_2 and CO_2) is not to be denied, as the indications are that the data are not too low, but rather somewhat too high. When other data are taken into account it is found that the observed separations are usually less than those calculated from Eq. (5) or (7), so that Eq. (5) is good enough with approximate integration and comparisons are best made with the aid of the fraction which the observed effect is of the calculated. When Eq. (1) is used this fraction is denoted by R. When Eq. (5) or (7) is used this will be denoted by C.

Now either Eq. (1) or (5) ought to be valid for the purpose of comparing one investigator's work with that of another when the temperatures and compositions are not too different. Yet the work of different investigators does not agree very well when compared in this way, as can sometimes be seen with little or no calculation. Furthermore Ibbs and Walkemann[12] have pointed

[11] T. J. Ibbs and A. C. G. Walkemann, Proc. Roy. Soc. (London) A134, 513 (1932).

TABLE I. *Values of 100 C over a range of temperature.*

Lower temperature	−41	−51	−58	−78	−119	−179
100 C	84	87	85	75	61	47

TABLE II. *100 times C for various mixtures. C equals ratio of observed separation over that calculated by Eq. (5).*

	H_2O	SO_2	CO_2	H_2	CO_2	CO_2	Mean
He	88	95	78	87		83	86
Ne	95		100	74			90
A	96	100		44	81		76
H	87	74	44		54	60	62
N_2			11	34		88	74
CO_2	83			80	88		77
H_2O				52			52

but that θ ($=dx/d\ln T$) is not really determined by the composition as both equations indicate. Thus for 47 percent CO_2 with H, they find $R_t=0.42$ for temperatures below 144° and 0.60 between 144 and 300°. Also at very low temperatures θ, and therefore C decrease very considerably. Thus for the experiment of Ibbs, Grew, and Hirst with 46 percent N_2 and argon, higher temperature −16°, the values of C are as given in Table I. The upper limit is 85. Similar results hold for other mixtures investigated by them including mixtures of monatomic gases.

A table of C values cannot therefore be very exact. In the Table II the results for extremely low temperatures have been omitted (or the upper limit found) and other results averaged. It is not presented as a critical summary of data, but only to illustrate the general degree of applicability of Eq. (5).

The values of $100C$ range from 100 down to 44, the mean for hydrogen being especially low. In certain cases R_t (the corresponding ratio for the Chapman approximate equation) has been calculated and Table III gives a comparison for nine cases. When C is not the same in Tables II and III it is because in Table III the value is for the same experiment for which R_t had been calculated, whereas for Table II some attempt at averaging was made.

TABLE III. *Comparison of C and R_t.*

Gases	H_2O	SO_2	H_2O	CO_2	H_2O	H_2	SO_2	CO_2	N_2	SO_2	Mean
100 C	95	96	87	83	100	74	50	62	38	80.5
100 R_t	75	78	72	78	83	51	34	63	11	63.1

The mean difference is favor of C is 15. The highest figure for R_t is 78, whereas C exceeds this in six cases. It seems clear that Eq. (5) is not only simpler to apply than Eq. (1) but gives considerably better results. Furthermore the results of Eq. (5) are sometimes excellent.

General Discussion

One may expect that the observed separations will not exceed those calculated by the new Eq. (5) and will seldom (except at extremely low temperatures) fall below 50 percent of the calculated values.

It may be, however, that the proviso should be made that the experimental value be obtained by apparatus not too different in character from the various arrangements already used. For, the equations purport to determine the separation by means of only thermodynamic variables. Now the problem does not look like a thermodynamic one and nonthermodynamic variables should be expected to enter. But, perhaps because they do not enter the approximate equation of Chapman, the experimenters often report only thermodynamic variables, omitting any detailed description of apparatus dimensions, and they do not appear to have studied the possible effect of any variables other than the pressure, composition and the two extreme temperatures. There is however enough agreement between different experiments with different apparatus to indicate that thermodynamic variables are at least sufficient to nearly determine the degree of separation in ordinary practice.

Separation by Vertical Parallel Walls at Different Temperatures

If the quantity C is calculated for the experiments of Clusius and Dickel, values are found of a different order of magnitude. Thus for their separation of the isotopes of chlorine $100C$ is found to be 5000. This indicates that their apparatus is acting as an ensemble of 50 units in

cascade. As the total length was 290 cm, the height of each equivalent unit was 5.8 cm.

For the separation of oxygen from air they used apparatus of two lengths, 1 meter and 2.9 m. The heights of the equivalent units as calculated from Eq. (7), the nitrogen being treated as pure, are 3.55 and 3.58 cm.

In the case of their separation of carbon dioxide and hydrogen they reported 40 percent CO_2 at the hot end and 100 percent at the cold end of a 1-meter tube. Using the integrated Eq. (7) with a value of $C=0.60$ it is found that even 27 units in cascade would produce 99.94 percent CO_2 at the cold end.

IMPROVEMENT OF THE SIMPLE EQUATIONS

If the layers of gas in the vertical thermal gradient were strictly isothermal, differing progressively by dT in temperature, the resulting equations ought to be about as accurate as the calculation of the partial pressure by similar methods, i.e., within one percent even for mixtures containing carbon dioxide except when one temperature is extremely low.

There is no mathematical difficulty here. The same equations also result if we call the temperature $T_1-dT/2$ and $T_1+dT/2$ at three successive planes serving to bound two layers and write $T_1-dT/2=T_2-dT_2/2$, etc. for the average values in the lower layer and expressions like $T_2+dT/2$ for the average in the upper layer. But there is a physical difficulty. For it does not seem probable that the different species come across the plane of reference from the same average distance. Let the species s_1 come from below from the average distance L_1 where the temperature is $T_1-L_1/L\,dT$, the density $n_1-L_1/L\,dn_1$, etc., and come from above from the average distance L_1 (or L plus a differential, but does not matter) where the temperature is $T_1+L_1/L\,dT$, etc. Over the small interval the functions can be supposed linear; the quantity L is a grand average distance over all species.

By this procedure all the preceding equations are obtained again with the sole difference that every root molecular weight m_i is now divided by L_i.

As the equations are homogeneous in m_i the L_i's cancel if assumed equal. It appears that the best we can do at present is to put the quantities L_i equal (or ignore them). If we put them equal to the mean free paths, as calculated for mixtures from any ordinary values of molecular diameters, the calculated separations diverge for the most part worse from the data. This can be seen from the equation for binary mixtures in the form

$$\frac{d\ln x_1/x_2}{d\ln T}=\frac{(m_2/m_1)(L_1/L_2)-1}{2x_1+(m_2/m_1)(L_1/L_2)x_2} \tag{15}$$

Now the right-hand side already comes out too great when $L_1/L_2=1$, particularly when m_2/m_1 is large, as in the case of carbon dioxide (2) and hydrogen (1). But as the ordinary free path of hydrogen in a 50 percent mixture is about three times that of carbon dioxide, this is equivalent to taking in the simple equation a threefold greater ratio of m_2/m_1, which results in a larger calculated separation. Except in the case of helium, L_2/L_1 usually exceeds 1 as estimated in this way when m_2/m_1 is taken to exceed unity.

But in dealing with gas mixtures, particularly when the identity of the molecule crossing a plane is of important, the mean free path that must be used for L_i must take account of the persistence of velocity. In the case of carbon dioxide and hydrogen, the average distance that a molecule of carbon dioxide goes before being much influenced in its progress by collision may well exceed the corresponding distance for a hydrogen molecule. According to the present body of data it appears impossible to obtain an agreement by means even of an arbitrary (but "smooth") persistence function of the ratios of the molecular weights. We may suppose that the persistence is determined in part by the nature of the intermolecular forces, an which case the ordinary mean free path is useless and the simple equations obtained above are probably the best working equations that can be obtained at present.

It is of interest that Chapman's approximate Eq. (1) for these two gases can only fit data if the diameter of carbon dioxide is assumed smaller than that of hydrogen (about one third).

rapidly as the temperature rises, since $(\gamma_1 - \gamma_2)$ (Fig. 1) decreases very rapidly.

The determination of the energy relations of gaseous films requires apparatus of high sensitivity and considerable accuracy. It is apparent from the equations that as the area of a gaseous film increases the increment of heat content (h) approaches zero for spreading, while for expansion it approaches h_s or about .16 erg cm^{-2} at ordinary temperatures, and q_v approaches q_s or about .41 erg cm^{-2}. It is obvious that in this case (ΔH_s) and (Q_s), which refer to one mole of the substance of the monolayer, lose their significance, but this is not true of h_s and q_s.

It is often forgotten that a film, such as that of an amphipathic (polar-nonpolar) organic substance on water, includes all of the region of the surface which contributes to the surface energy, that is all of the energy associated with the surface which is not accounted for by three-dimensional thermodynamic treatment which neglects the extent of the surface. The contribution of the first layer of water molecules to this energy is very important, whether it is covered by the organic layer completely or incompletely, and while the second and third, etc., layers rapidly become less important, it is obvious that the first layer of water molecules, though it contributes by far the greatest share of the surface energy due to the water in the surface region, is not the only one involved. Thus in the utilization of the results of this paper in connection with the problem of intermolecular attraction in the monolayer or the film, the energy of spreading and of extension are both of fundamental importance. It has been considered advisable to limit the present paper to the thermodynamics of films. Any applications to molecular theory has been deferred to later papers.

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Effect of Gravitational Field on the Thermal Diffusion Separation Method

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A test of the effect of gravitational field on the operation of the Clusius-Dickel thermal diffusion method for gas separation has been made by use of a hollow disk, with one hot surface, [illegible] increased in both the equilibrium separations and the rates of separation with increasing centrifugal force were obtained for speeds below a certain maximum value. Above this both the rate and equilibrium separations fell rapidly. H_2-CO_2 mixtures were used. The results indicate that the standard column would be improved if it were placed in a larger field up to ten times that caused by gravity. Theories for the action of the thermal diffusion column should show this. The apparatus cannot compete with the standard column type for isotopic separations but may have some application to industrial [illegible] problems.

CLUSIUS and Dickel[1] have shown that the counter current flow obtained in the gas between two vertical concentric cylinders, the inner one of which is heated, operates to increase the effect of the thermal diffusion separation of gases. The effects of pressure, dimensions of the apparatus, temperature gradient, and character of the gaseous components all have been investigated in some detail,[2-11] both theoretically

[1] K. Clusius and G. Dickel, Naturwiss. 26, 546 (1938).
[2] S. Chapman, Proc. Roy. Soc. London A93, 1 (1917); Phil. Trans. Roy. Soc. London 217, 157 (1917); 181 (1918).
[3] D. Enskog, Zeits. f. Physik 12, 533 (1911); Zeits. f. Physik 2, 537 (1911).
[4] F. L. Hicks, Proc. Roy. Soc. London A99, 385 (1921); 107, 170 (1925).
[5] A. K. Brewer and A. Brimley, Phys. Rev. 55, 500 (1939).
[6] K. Clusius and G. Dickel, Zeits. f. physik. Chemie, B44, 397 (1939).
[7] A. O. Nier, Phys. Rev. 57, 90 (1940).
[8] W. Furry, R. Jones and L. Onsager, Phys. Rev. 55, 1083 (1939).
[9] J. Sawkommen, Transactions 27, 210 (1939); Zeits. f. Physik 114, 53 (1939).
[10] P. Debye, Ann. d. Physik 40, 284 (1939).
[11] W. Bertsch, Phys. Rev. 57, 85 (1940).

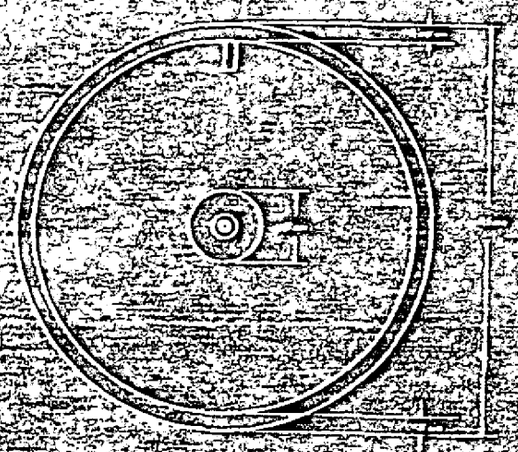

FIG. 1A. Side diagram of the centrifugal thermal diffusion apparatus (drawn to scale).

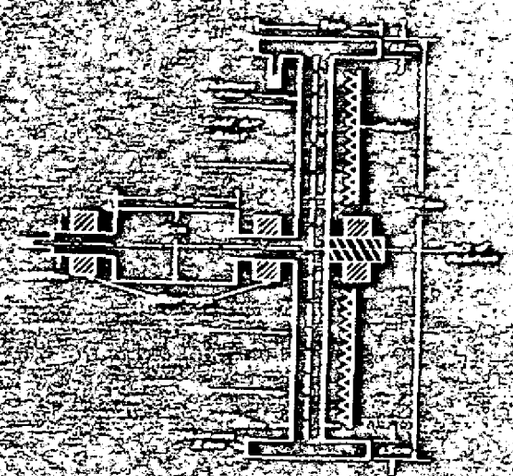

FIG. 1B. Cross-sectional diagram of apparatus (drawn to scale).

and experimentally. The authors have attempted to investigate the effects of variation of the gravitational field by use of a rather drastically modified form of the apparatus.

APPARATUS

Since the use of centrifugal force as the only way of varying the gravitational field the apparatus was designed so the necessary counter current would occur perpendicular to an axis of rotation. The standard tube arrangement was replaced by a rotating hollow disk, one surface of which was heated and the other cooled. Reservoirs for the gas were provided at the rim and on the axis of rotation as shown in Figs. 1A and 1B.

The whole apparatus consisted of brass approximately 1 mm thick. The hot plate was heated by radiation from a Nichrome wire heater, wound on transite sheet and mounted approximately 1 cm from the brass surface, while the opposite face was cooled with an air blast from a large electric fan. The rim reservoir was in the shape of a hollow rectangular tire with a volume of 780 cc. A metal cock for sampling the rim gas was located on the inner surface of the rim as shown. A reservoir of 200 cc volume was located on the axis of rotation with a sampling cock attached. The wheel was rotated by a direct-current motor of variable speed by means of a belt running over a wooden pulley mounted around the cylindrical reservoir on the rotational axis. Three ball bearings were mounted on iron brackets on a sturdy platform for support of the wheel. The speeds of rotation were measured with a standard stroboscope to about one percent accuracy. The speed of rotation was constant to one or two percent at each speed, though a small amount of angular vibration set in at the highest speeds of approximately 1000 r.p.m.

The surface temperatures were measured by placing Chromel-Alumel thermocouples in direct contact with the surfaces. Various segments of the heater coil were provided with separate leads so that the area of the heated surface might be varied in both magnitude and distance from the axis of rotation.

The inner surfaces of the plates were 7 mm apart and the volume of the interplate region was 600 cc.

The gas analyses were made on 10-cc samples taken by stopping the wheel and using the rim and axis cocks. It was shown by sampling on the axis during rotation and after stopping that stopping the wheel did not lead to appreciable mixing in the two or three minutes required to take the samples. For most of the work, in which CO_2 and He were being separated, the analysis was accomplished by freezing out the CO_2 in a liquid air trap and reading the pressure change on a capillary manometer. By using capillary tubing throughout the system and taking reasonable care to make the correction accurately for

the decrease in pressure of the He caused by the cooling, it was possible to reproduce the analyses to within one percent.

The He—O_2 gas used was mixed in a pressure cylinder at several atmospheres pressure and introduced into the apparatus at atmospheric pressure. The air was removed by flushing with excess of the gas. It was not possible to evacuate the wheel because the walls were too thin to support atmospheric pressure.

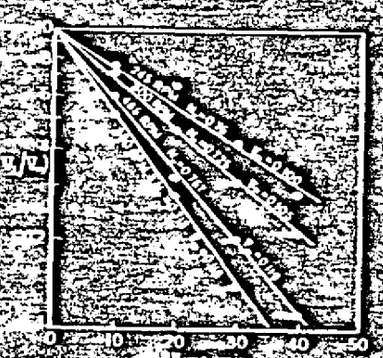

FIG. 2. Separation at time t at various speeds of rotation. $T = 200°C$, heater area = 278 cm².

RESULTS

Separation vs. time

Figure 2 presents data obtained with the hot plate at 40°C and the cold plate at 50°C, the wheel rotating at various speeds and the initial composition of the gas having various values. The ordinates are logarithms of one minus the ratio of the separation at any given time to that for the equilibrium state. The separations, Δ, were calculated as the difference between the mole fraction, N_c, of He at the rim or the axis and the initial mole fraction, N_0. A typical set of data is given in Table 1. The data seem to justify approximately the exponential expression

$$\Delta = \Delta_e(1 - e^{-\lambda t}) \qquad (1)$$

TABLE 1. Separation vs. time; 33 r.p.m., $T = 200°$, N_0 (center) = 0.117, N_e = 0.757.

Time (min)				
Δ	0.60	0.83	1.20	0.11

for the dependence of the separation on the time, Δ being the separation at any given time, Δ_e the equilibrium separation, and λ a constant measuring the rate of separation. Figure 3 presents the data for the effect of variation of the temperature gradient on the rate of separation, λ. Figure 4 shows the variation of the rate, λ, with r.p.m. A rather sudden break occurs at about 400 r.p.m. The main point about these data is the approximately linear rise in the rate with the square of the speed of rotation, ω^2, until the break occurs.

Figure 5 shows a similar set of data for the variation of the equilibrium separation, Δ_e (center), with ω^2. Here also an approximately linear rise occurs till ω^2 becomes approximately 400 r.p.m. Figure 6 presents the data for the variation of Δ_e (center) with the temperature difference. Figure 7 shows the Δ_e (center) vs. ω^2 of two heaters, the larger having an area of 335 cm² and the smaller having 278 cm², proving that the heater area had no serious effect on the speed at which the linear rise in Δ_e (center)

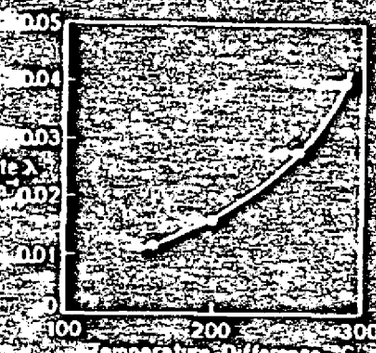

FIG. 3. Rate of separation vs. temperature difference.

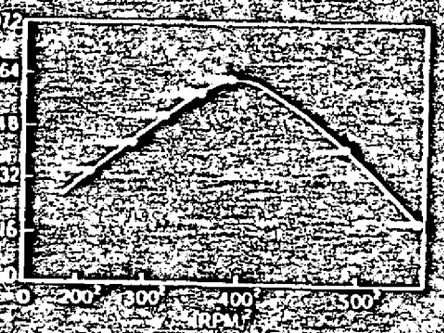

FIG. 4. Rate of separation vs. speed of rotation.

FIG. 5. Equilibrium separation vs. speed of rotation. $\Delta T = 290°C$, $\Delta V_c = 0.65$, heater area = 278 cm², time for each run = 2 hr.

breaks. Figure 8 gives the dependence of Δ_c (center) on the heater area, the open circles referring to variations obtained by changing the radius of the inner edge of the heated region, while the solid circles belong to data obtained by varying the radius of the outer edge. Apparently there is no large difference between these two sets of data.

DISCUSSION

The gas flow which occurs is a combination of a general movement of the gas near the hot surface inward toward the axis with a transfer flow of part of this hot gas across to the outward moving cold stream as illustrated in Fig. 2. The hot gas flows across to the cold stream at a rate which is constant for each unit of area of plate surface. The truth of this statement is apparent if one notices that both the force acting to push the gas toward the rim and the area across which it moves in flowing toward the rim increase directly with the distance ω from the axis of rotation. To maintain approximately constant pressure throughout the gas it is necessary that, instead of compression occurring as the hot gas stream moves inward, a partial movement across to the cold stream must occur to compensate for the decrease in volume available. This also keeps the outward moving stream from suffering a pressure decrease. For a given movement inward from ω to $\omega - d\omega$ the decrease in total rate of flow is proportional to $\omega d\omega$ because the area across which the flow occurs is proportional to ω and the decrease in the force causing the flow is proportional to $d\omega$. This

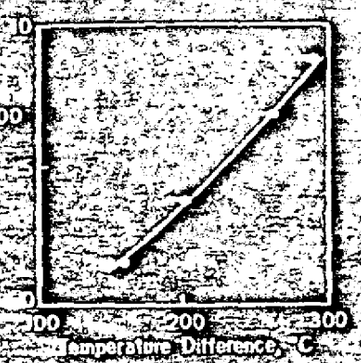

FIG. 6. Equilibrium separation vs. temperature difference.

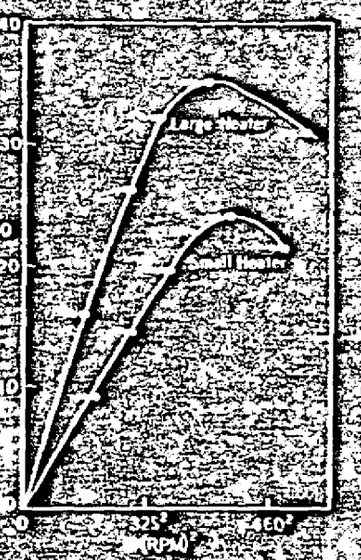

FIG. 7. Effect of heater area on equilibrium separation for large heater, $\Delta T = 290°C$ heater area = 435 cm², $\Delta V_c = 0.62$. For small heater, $\Delta T = 290°C$ heater area = 278 cm², $\Delta V_c = 0.65$.

decrease must be equal to the flow to the cold stream. The area of the plane parallel to the wheel surfaces across which this flow occurs is obviously as proportional to $\omega d\omega$ also, so the ratio of the flow from hot to cold stream per unit area of wheel surface is constant over the whole wheel.

Using the dimensions of the apparatus, the viscosity of the gas, and approximate values for the temperature gradient the velocity of flow is calculated to be about 150 cm/sec at the inner edge of the heater and 500 cm/sec at the rim at 275 r.p.m. The hot to cold stream flow is about 25 cm/sec under these conditions. At other

speeds the values should be higher or lower in proportion to ω^2.

The data show that in the neighborhood of 400 r.p.m. the increases of both the rate and equilibrium separation with increasing ω^2 break rather sharply into decreases. Both the suddenness of the breaks and the gas velocities at which they occur indicate that turbulence sets in under these conditions.

The Chapman-Enskog thermal diffusion process, which concentrates the light constituent near the hot side, must be established rapidly with respect to the rates of flow below those for 380 r.p.m. or the linear rates shown in Figs. 4 and 5 for A and A_s (center) at ω^2 probably would not exist. In order that the counter current action should increase directly with the rate of flow, equilibrium must be nearly completely established for the whole range of speeds. It is conceivable that the breaks in Figs. 4 and 5 are caused by failure to establish equilibrium rather than by turbulence. The chief argument against this interpretation is the sharpness with which the breaks occur.

The linear increase of A_s and A with the area of the heated surface indicates strongly that the most important part of the current flow is the transfer of the hot gas to the outward moving cold stream which has been shown above to occur at a constant rate per unit area of heated surface independently of the distance from the axis of rotation. It seems reasonable that this

Fig. 8. Equilibrium separation vs. heater area ($\omega^2 = 340$ r.p.m. $\Delta T = 290$ °C).

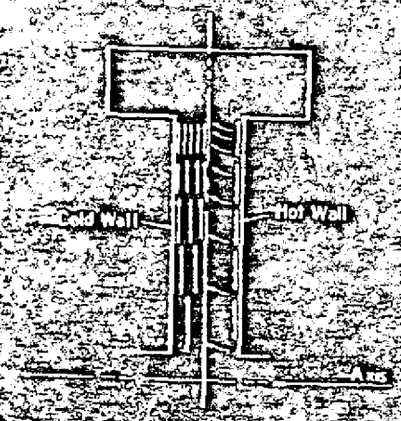

Fig. 9. Type of flow taking place while apparatus is in operation.

should be the case. The gas moving to the cold stream carries an excess of the heavy component with it leaving the inward moving hot stream somewhat purified.

It seems certain also that turbulence should set in for this type of flow at somewhat lower velocities than for those existing in the standard cylindrical thermal diffusion apparatus. On the other hand, it may be true that the condition that equilibrium be established rapidly with respect to the counter current velocity may be less easily violated in the wheel apparatus because of the intimate nature of the contact between the inward moving hot gas and that part of it which is just leaving to join the cold stream. However, since the rises were observed to continue till the main part of the gas was subjected to a field approximately 20 times that due to gravity, it is relatively certain that the ordinary Clusius-Dickel-Brewer-Bramley column would be much improved if it were possible to increase the gravitational field to some ten times the normal value. We should expect all theories for the actions of the columns to show this possibility.

The wheel method seems to offer little promise as a competitor with the columnar type of apparatus. Our apparatus was about equivalent in action to an average column five or six feet long. It is possible, however, that the use of wheels of much larger radii with surfaces heated by flames or superheated steam instead of electricity would afford a less expensive method of separating certain gases of industrial importance.

On the Theory of Isotope Separation by Thermal Diffusion

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AND
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We here develop the theory of the process in an apparatus with two concentric tubes, such as that used by Brewer and Bramley. In the first section we describe the process of thermal diffusion in general, and review the theoretical information on the subject. In Section II we set up the equations for the convection and diffusion processes, and obtain an expression for the net transport of a single isotope which is valid for arbitrary microscopic properties of the gas. The assumption that the viscosity and heat conductivity of the gas are proportional to the absolute temperature is usually a good approximation; in this case the expression reduces to a simple form. In Section III we apply the transport equation to questions of separation factor and speed of operation. This is done both for the case of discontinuous operation, in which the approach to equilibrium is allowed to reach a certain stage and then the contents of an end reservoir are removed, and for the case of operation with a continuous flow of gas through the tube. The advantages of the two methods are compared. In the last section we apply the formulas to a numerical example, the concentration of the C_{13} isotope by the use of methane, in an apparatus of moderate dimensions and power consumption.

I. INTRODUCTION

RECENTLY Clusius and Dickel[1] and subsequently Brewer and Bramley[2] have reported the results of experimental work on the separation of isotopes by thermal diffusion. Their results suggest that this may become an important method. In this paper we discuss theoretically the problem of applying the process of thermal diffusion to isotope separation.

The phenomenon of thermal diffusion consists in the fact that a temperature gradient in a mixture of two gases gives rise to a gradient of the relative concentration of the two constituents. If the mixture as a whole is at rest, the equilibrium concentration gradient is such that the effect of thermal diffusion is just matched by the opposing effect of ordinary diffusion. The effect is by no means small; in some cases the coefficient of thermal diffusion may be as much as half of the ordinary coefficient.[3] A similar phenomenon is known in the case of liquids, where it is known as the Soret effect.[4] The theoretical explanation of the Soret effect has been signally unsuccessful.[5]

No simple theory can give an account of thermal diffusion; neither its existence nor its sign can be derived from elementary considerations. Its presence can be understood only by a detailed consideration of the equations of transport in a gas. The phenomenon was completely overlooked by the classic workers in kinetic theory; it was first discovered theoretically by Enskog in 1911, and independently by Chapman six years later, and first demonstrated experimentally by Chapman and Dootson in 1917.[6] The theory was fully and elegantly developed by Enskog[7] and Chapman.[8] The more accessible treatment of Chapman is, however, unfortunately invalid because of algebraic errors.[9] Chapman's corrected result[10] may be shown to be identical with the result of Enskog. It is the result obtained by Enskog in his doctoral dissertation that is given in the well-known textbook of Müller-Pouillet.[11]

[1] K. Clusius and G. Dickel, Naturwiss. 26, 546 (1938).
[2] A. K. Brewer and A. Bramley, Phys. Rev. 55, 590A (1939).
[3] S. Chapman, Phil. Trans. A217, 181 (1917).
[4] Ludwig, Wien. Akad. Ber. 20, 539 (1856); Soret, Ann. Chim. Phys. 22, 293 (1881).
[5] Wierzejewski, Ann. de physique 2, 67 (1914); Porter, Trans. Faraday Soc. 23, 314 (1927); also criticized by Chapman, Phil. Mag. 7, 1 (1929).
[6] S. Chapman and F. W. Dootson, Phil. Mag. 33, 248 (1917); for a complete list of the experimental literature on thermal diffusion, see T. L. Ibbs, Phys. Rev. 4, 1133 (1937).
[7] D. Enskog, Doctoral Dissertation, Uppsala, 1917, published by Almqvist and Wiksells.
[8] D. Enskog, Arkiv f. Mat. Astron. o. Fys. 16, No. 16 (1921).
[9] S. Chapman and W. Hopworth, Phil. Mag. 48, 391 (1924); the presentation of the value of D_T in this paper does not become complete until it is stated that the factor [illegible] is [illegible].
[10] S. Chapman, Proc. Roy. Soc. A119, 159 (1928).
[11] Müller-Pouillet, Lehrbuch der Physik, eleventh edition, Vol. 3, Part 2, p. 110.

The coefficient of thermal diffusion vanishes for Maxwellian molecules (molecules which interact with a force which varies as the inverse fifth power of the separation). For molecules such that the force varies as an inverse power of the separation which is greater than five, the direction of the diffusion is such that the lighter molecules move toward the warmer part of the gas, in general, while for an inverse power less than five, the lighter molecules accumulate in the colder part of the gas. A general result of the theory is that the coefficient vanishes when the mode of interaction, the radii, and the masses of the two sets of molecules are all equal; this fact is also obvious from symmetry.

Even to the first approximation (the result comes out as an infinite determinant), the coefficient of thermal diffusion can be stated in closed form only for the case of rigid elastic spheres, in which case the effect is greatest. The experiments show a coefficient which is in general something less than half of the value predicted for elastic spheres.

The equation of diffusion, with thermal diffusion included, is

$$c_1(\mathbf{v}_1-\mathbf{v}) = -D_{12}\,\mathrm{grad}\,c_1 + D_T/T\,\mathrm{grad}\,T, \quad (1)$$

where c_1 is the relative *particle* density of the lighter constituent, with $c_1+c_2=1$, $\mathbf{v}_1$ is the *convection* velocity of the lighter constituent alone, while $\mathbf{v}$ is the convection velocity of the gas as a whole ($\mathbf{v}=c_1\mathbf{v}_1+c_2\mathbf{v}_2$), D_{12} is the coefficient of ordinary diffusion, which in the case of isotopes may be replaced by D_s, the coefficient of self-diffusion, and D_T is the previously mentioned coefficient of thermal diffusion, T is the absolute temperature. In the corresponding equation for c_2, the D_T term has the opposite sign.

It can be shown rigorously for the case of hard spheres that the ratio $D_T/D_{12}=k_T$ depends only on the relative concentrations and on microscopic quantities but not on the pressure or temperature. It is reasonable to suppose that for actual molecules this ratio does not vary with pressure or temperature to any significant extent.

For the separation of isotopes, we are interested in the case when the two sets of molecules in question are chemically identical and differ only in mass. In this case, the expression for k_T becomes greatly simplified because the modes of interaction and the radii of the two constituent molecules are then the same, since these properties are completely determined by the electronic configuration. For this special case, Enskog's result reduces to[1]

$$D_T/D_{12} = k_T = \alpha c_1 c_2, \quad (2)$$

where $\alpha=(105/118)(m_2-m_1)/(m_2+m_1)$, the m's being the relative masses of the two types of molecules. It is to be remembered that number 1 is the lighter species. This expression is the first approximation for the case of elastic spheres, in which furthermore we have retained only terms of the first order in $(m_2-m_1)/(m_2+m_1)$.

The experimental determinations of k_T for the case of *dissimilar* molecules yield values somewhat less than half of the theoretical result for elastic spheres. Until experiment yields us values for k_T in the case of isotopes, therefore, we suggest that

$$k_T = 0.35(m_2-m_1)/(m_2+m_1) \quad (3)$$

be used as a provisional value in the design of apparatus. The exact value of the numerical coefficient will vary from molecule to molecule because of the different modes of interaction.

When values for k_T in the case of isotopes are determined experimentally, they may lead to valuable conclusions about the nature of the intermolecular forces in the gas in question. Thermal diffusion should be an excellent way to obtain this information, because it is one of the few phenomena in a gas that depend essentially on the peculiar characteristics of the molecules.

The knowledge of k_T for the case of isotopes is particularly valuable, because in this case there is no mixing of the characteristics of two dissimilar molecules.

The use of thermal diffusion for separating isotopes was first suggested by Chapman.[2] Its use was discussed by Mulliken,[3] on the basis of Chapman's first treatment. Mulliken concluded that it could not compete with other methods.

[1] S. Chapman's earlier expression (reference 16) is

$$k_T = \frac{15}{4}\frac{m_2-m_1}{m_2+m_1}\frac{2.15-3.25c_1c_2}{2.15-c_1c_2}.$$

[2] S. Chapman, reference 4, p. 181; J. H. Jeans, *The Dynamical Theory of Gases*, fourth edition (Cambridge, 1925), p. 323.

[3] S. Chapman, Phil. Mag. 36, 182 (1919).

[3] R. S. Mulliken, J. Am. Chem. Soc. 44, 1033 (1922).

But he did not consider the very effective method of fractionation employed by Clusius and Dickel.[1]

Clusius and Dickel ran a heated wire up the axis of a long vertical hollow tube, into which was put the gas whose isotopic constituents were to be separated. The combination of the effect of thermal diffusion and of the convection currents set up in the tube is such as to increase greatly the rather small separation caused directly by the radial temperature gradient. The light isotope collects at the top, while the heavier isotope goes to the bottom of the tube, as we shall discuss in detail in the next section.

The procedure of Brewer and Bramley was very similar. They used two concentric tubes, of one and two centimeters diameter. The inner tube was heated.

In the following two sections we examine quantitatively what is occurring in this new method of separating isotopes. We find it possible to set up formulae showing the dependence of the performance characteristics on the dimensions of the apparatus, the temperature, the density, etc. These results should be useful in designing apparatus for isotope separation by this method.

II. THE CONVECTION FIELD AND THE TRANSPORT EQUATION

Description of apparatus and notation

The apparatus is supposed to consist of two concentric cylinders, the inner one heated and the outer one cooled, mounted in a vertical position, with reservoirs at the top and bottom communicating with the annular space between the cylinders, where convection and diffusion take place. The difference of the radii may be supposed small compared with either radius, so that we may proceed as if the convection were taking place in a thin flat slab, of breadth B equal to the mean circumference of the cylinders, thickness $2w$ equal to the difference of the radii, and length $2l$, the length of the tube.

In addition to the notation already introduced, we shall use the following: T_i, T_o = absolute temperature of the outer, inner tube, $\Delta T = T_i - T_o$; x = radial coordinate, $-w < x < w$; z = coordinate along the tube, $-l < z < l$; c_p = specific heat at constant pressure, in cal/gram-deg.; Q = heat flow, in cal/cm² sec.; λ = thermal conductivity, in cal/cm deg. sec.; p = pressure; η = viscosity; ρ = density; g = acceleration of gravity; J_1, J_2 = flux of species 1, 2 in grams/cm² sec.; σ = total upward transport, in grams/sec.; σ_1, σ_2 = upward transport of species 1, 2, in grams/sec., $\sigma = \sigma_1 + \sigma_2$; c_0 = value of c in the bottom reservoir; c_1 = value of c in the top reservoir; c_e = equilibrium value of c; with $\sigma = 0$; c_s = a specified value of c; m = mass of gas in the top reservoir, and τ = relaxation time of the equilibrium. In all considerations subsequent to Eq. (29), the temperature-dependent quantities are to be evaluated at the average temperature, $\frac{1}{2}(T_i + T_o)$.

General treatment of the convection and diffusion field

Throughout this section and the next it will be assumed that it is the lighter isotope, species 1, which one is interested in concentrating. The considerations will be completely symmetrical in respect to the interchange of 1 and 2; however, it we understand that the vertical coordinate changes sign when we apply our equations to the concentration of a heavy isotope, that is, c_1 becomes the concentration c_0 in the top reservoir, while the small collecting reservoir would be at the bottom; and so on in an obvious manner.

In the immediately following presentation, we shall develop a mathematical description of the convection and diffusion processes in the tube, and derive the fundamental relations which yield the transport of species 1 along the tube. Having obtained these relations, we then proceed in the next section to determine the behavior of the apparatus in the large, to find what separation factors can be obtained, and at what speed a mixture of given concentration may be produced.

We shall calculate the convection by using the temperature distribution as determined by conduction only. Near each end of the tube, there is not a different temperature as entering the convection; this distribution is, of course, different. On the basis of the treatment of the convection problem given below, one finds that the length over which this difference remains appreciable is of the order of

$$l_1 = (c_p \rho^2 g B w^3 \lambda)^{1/2} (T_i - T_o)/(T_i + T_o)^{3/2} \quad (1)$$

where, here as in (5), the temperature dependent

quantities ρ, η, λ are evaluated at the average temperature $(T_2+T_1)/2$. Our neglect of these regions depends on the condition $2l_c \ll l$, which will usually be strongly satisfied in practice.

The treatment of the convection flow as laminar is justified since the Reynolds number may be shown to be roughly

$$\omega^2 l^2/2\lambda\eta)(T_2-T_1)/(T_2+T_1), \qquad (5)$$

which in practical cases will be very much smaller than the values at which turbulence occurs in flow through tubes or between plates. In the region of the tube where the temperature gradient is determined by conduction alone, we have

$$\lambda Q = \lambda dT/dx;$$

$$\lambda wQ = \int_{T_1}^{T_2} \lambda dT, \qquad (6)$$

$$\lambda w/\lambda = (Q/\lambda)(\partial/\partial T).$$

We shall neglect the dependence of λ on the concentration c_1 and also the dependence on c_1 of ρ, η, and D. These dependences are not large when the fractional difference of molecular weight is small. Moreover only transverse differences have any significant effect on the performance, and the transverse difference of concentration is very small. Thus only the temperature dependences of the various quantities is of any importance.

The hydrodynamical equation of steady, viscous flow is

$$\eta(\text{div grad})\mathbf{v} = \text{grad}\, p - \rho\mathbf{g}. \qquad (7)$$

We assume that the velocity is entirely in the vertical direction, and that it is independent of z. This is a very close approximation since the pressure variation within the tube is negligible. Then we have from (6) and (7),

$$\eta = (Q/\lambda)(d/dT)(\eta/\lambda)(dv/dT) = (dp/dz) + \rho g, \qquad (8)$$

with the boundary conditions

$$v(T_1) = v(T_2) = 0. \qquad (9)$$

By (1), the equation for the flux of species 1 is

$$\mathbf{J}_1 = [c_1 + D(-\text{grad}\, c_1 - \alpha c_1 \text{grad} \ln T)]\mathbf{v}, \qquad (10)$$

and a corresponding equation holds for $\mathbf{J}_2$.

We shall base our calculation of the transport of species 1 through the tube on the assumption that the time variation of the concentration c_1 at each point in the tube is negligible—that is, that the condition is either stationary or quasi-stationary. A stationary condition exists at final equilibrium in a closed apparatus, and also when there is a uniform output of the concentrated isotope. During the approach to equilibrium the condition will be essentially stationary provided each end reservoir is sufficiently large. The volume of each reservoir must be larger than that of a length of tube in which the equilibrium concentration changes by a factor e.

Accordingly we proceed from the equations

$$\text{div}\, \mathbf{J}_1 = \text{div}\, \mathbf{J}_2 = 0. \qquad (11)$$

Eq. (10) then gives, on application of (6),

$$\partial/\partial T(\rho D/\lambda)[(\partial c_1/\partial T) - (\alpha c_1/T)]$$
$$(\lambda\rho/Q^2)[v(\partial c_1/\partial x) - D(\partial^2 c_1/\partial x^2)]. \qquad (12)$$

The term containing $(\partial^2 c_1/\partial x^2)$ is the one that introduces the effect of longitudinal diffusion. The retention of this term would make it impossible to carry out the analysis in any simple manner. Since the effect of diffusion along the tube can be calculated separately quite accurately, we shall omit the term in question, and calculate from

$$Q/\partial T(\rho D/\lambda)[(\partial c_1/\partial T) - (\alpha c_1/T)]$$
$$(\lambda\rho/Q^2)v(\partial c_1/\partial x). \qquad (12')$$

We now introduce a function $G(c_1, T)$ defined by

$$(\partial c_1/\partial x)G(c_1, T) = -(\lambda Q^2/\rho D)J_1, \qquad (13a)$$

$$Q^2[(\partial c_1/\partial T) - (\alpha c_1/T)]. \qquad (13b)$$

Then from (12'),

$$Q/\partial T(\rho D/\lambda)(\partial c_1/\partial x)G(c_1, T)$$
$$= \lambda v(\partial c_1/\partial x). \qquad (14)$$

From this point on we shall assume that the dependence of $\partial c_1/\partial x$ on T can be neglected. This is equivalent to assuming that $\partial c_1/\partial T$ is independent of x, which is physically reasonable for a quasi-stationary condition. We shall at once establish two important consequences of this assumption. First, the total transport J through the tube is zero; second, G is a function of T only.

Since div $\mathbf{J}_1$ is zero, the flow of species 1 must be constant along the tube—that is,

$$J_1 = B\int_{T_1}^{T_2} c_1 v\,dz = \text{constant},$$

$$v = (B/Q)\int_{T_1}^{T} v c_1\,dT, \tag{15}$$

$$(dv/dz) = (\partial c_1/\partial z)(B/Q)\int_{T_1}^{T_2} v\,dT$$

$$= (\partial c_1/\partial z)\,v = 0.$$

Then, since $\partial c_1/\partial z$ is not identically zero, the total transport must vanish.

By (13a), we have

$$G(z, T_1) = G(z, T_2) = 0 \tag{16}$$

since J_1 must vanish at the walls. The factor $(\partial c_1/\partial z)$ on each side of (14) cancels, and since the only remaining quantity that can be a function of z is $G(z, T)$, one finds, on integrating (14) that $(\lambda D/\Lambda)G$ can depend on z only through an additive function of z. Then, since (16) hold identically in z, we have

$$G(z, T) = G(T). \tag{17}$$

We have now, canceling the $\partial c_1/\partial z$ in (14), and solving for v,

$$v(T) = (\Lambda/\lambda Q)(d/dT)[\lambda D G(T)/\Lambda]. \tag{18}$$

Then from (18) and (16),

$$G(T_1) = G(T_2) = 0. \tag{19}$$

If we now substitute (18) in (8), and differentiate with respect to T, we obtain as the differential equation for $G(T)$

$$\frac{d}{dT}\left(\frac{\lambda}{\Lambda}\frac{d}{dT}\left(\frac{\lambda D}{\Lambda}G(T)\right)\right) = \frac{\rho}{\Lambda}\frac{dT}{dT} \tag{20}$$

with the boundary conditions

$$G(T_1) = G(T_2) = G'(T_1) = G'(T_2) = 0. \tag{21}$$

By the calculation of this single function one has, by (18), a complete description of the convection process; and, within the limits of our approximations, one has, by (13), a description of the transverse diffusion, both thermal and ordinary. By the use of this same function we can proceed to obtain an expression for the transport of a single isotope along the tube.

According to (15) and (18), the net vertical transport of species 1 is

$$\mathcal{J}_1 = (B/Q)\int_{T_1}^{T_2} c_1 (d/dT)[\lambda D G(T)/\Lambda]\,dT. \tag{22}$$

Partial integration and substitution from (13b) and (21) lead to the expression

$$\mathcal{J}_1 = H c_1 - K\,\partial c_1/\partial z, \tag{23}$$

where H and K are positive constants defined by

$$H = (B/Q)\int_{T_1}^{T_2} (\lambda D/\Lambda)\,G(T)\,dT \tag{24}$$

and

$$K = (B/Q)\int_{T_1}^{T_2} (\lambda D/\Lambda)\,G(T)\,dT. \tag{25}$$

We have removed $\partial c_1/\partial z$ from under the integral sign of (24), because $\partial c_1/\partial z$ varies only slightly with z. We could not consider c_1 as constant in (22), however, because it is only the slight variation of c_1 which prevents this integral from vanishing. In Eq. (23) the term Hc_1 gives the transport of species 1 caused by the existence, due to thermal diffusion, of a transverse flow of this isotope, which makes the concentration in the column of gas moving upward differ from that in the column moving downward. At the beginning of the process, when $\partial c_1/\partial z = 0$, this is the only cause of transport. After a longitudinal concentration gradient has been built up, the convection currents lead to a certain amount of remixing of the gas, represented by the term $-K\,\partial c_1/\partial z$. There is, however, another remixing effect which is not included in Eq. (23), because in replacing (12) by (12′) we omitted the term which corresponds to diffusion along the tube. Since the convection velocity will be small, however, the longitudinal diffusion will be practically independent of it. If we ignore the convection velocity, the transport of species 1 due to longitudinal diffusion is

$$\mathcal{J}_1 = -K_1\,\partial c_1/\partial z, \tag{26}$$

where

$$K_1 = (B/Q)\int_{T_1}^{T_2} \lambda D\,dT.$$

The total transport of species 1 is obtained by adding Γ_T to the Γ_1 given by (23).

Specialization to the Maxwellian case

The treatment so far has been general, in the sense that the quantities D, Λ and η might have an arbitrary temperature dependence. If, however, the mean free path varies as the $(\alpha-4)$th power of the temperature, and if σ_D is independent of T, the following quantities should all be constant:

$$(\eta/T^{\alpha}),\quad (\Lambda/T^{\alpha}),\quad (\rho D/T^{\alpha}),\quad (\eta/\Lambda),\quad (\rho D/\Lambda).$$

Then (20) becomes

$$(d/dT)T=(d\eta/dT)T-\eta(dG/dT)$$
$$=-T^{\alpha}(\Lambda\eta^2)/(\rho DT-\eta)\times(27)$$

The general solution of (27) has been obtained, and there appears to be no bar to the explicit evaluation of ΓT and ΓK in this manner. The formulae would be awkward in practice, however, because of heavy cancellation of terms. The writers have worked out the cases of $\alpha=\frac{1}{2}$ (hard spheres) and $\alpha=1$ (Maxwellian molecules). Since the case $\alpha=1$ is much simpler than any other, and corresponds fairly well to the temperature dependence of the properties of most gases, we suggest that the formulas for this case be used in practice. This simplification is particularly well justified because our knowledge of the all-important factor σ itself is so inexact.

For $\alpha=1$, integration of (27) and application of (21) gives

$$G(T)=-[(\Lambda\eta_0)/(24\rho_0 D)]$$
$$\times(T_0-T)^2(T_0-T)^2/(T_0+T_1).\quad(28)$$

and (6) becomes

$$\Gamma_T=\Delta T/2Q.\quad(29)$$

where in this and all following formulae, ρ, D, Λ, η and T are to be evaluated at $T=\frac{1}{2}(T_0+T_1)$. If (28) and (29) are used in (24), the result is

$$H=[(\pi^{\frac{1}{2}}\Lambda\eta B)/(90\rho_0)](\Delta T/T)^2/(\Delta T/T),\quad(30)$$

where

$$J(\Delta T/T)=30T^{-2}(\Delta T)^{-1}[(\Delta T)^2/6-T_0T_1$$
$$+(T_0T_1)^2(T\Delta T)^{-1}\ln(T_0/T_1)]$$
$$=15\sum_l(\Delta T/2T)^{2l}/[(2l+1)(2l+3)]$$
$$\times(2l+5)]$$
$$=1+(15/3\cdot5\cdot7)(\Delta T/2T)^2+\cdots$$

$J(\Delta T/T)$ has the value unity for $\Delta T=0$, and the value 5/4 for the extreme case $T_1/T_0=0$, and lies between these two values for intermediate cases. For $T_1=2T_0$, $J=1.016$, while for $T_1=3T_0$, $J=1.039$.

From (25) and (29), we have

$$\Gamma=H/2K=(63/4)$$
$$\times[(\alpha\pi D)/(\rho\eta\omega^2)]J(\Delta T/T),\quad(31)$$

and (26) gives

$$\Gamma_K/K=1890(T_0^2+T_0T_1+T_1^2)$$
$$\times[D_0/(\omega^2\rho\Delta T)]^2.\quad(32)$$

The corresponding relations for a gas consisting of hard spheres follow.

We introduce the abbreviations

$$\eta=\eta_0,\quad \omega=\eta_0,\quad \omega_0=\eta_0,$$
$$\sigma=(\omega-\omega_0)/(\omega+\omega_0),$$
$$\Phi(\sigma)=(1+\sigma)^2(3-\sigma)/(\omega-\omega_0).$$

Then we find

$$G(T)=-[(\Lambda\eta_0T)/(72\rho_0D)](\omega-\omega_0)^2[1-\Phi(\sigma)]^2$$
$$\times[(6-5\sigma^2)(3+\sigma)\Phi(\sigma)-(3+\sigma^2)(3+\sigma)-(5-\sigma^2)\sigma],$$
$$\Gamma=[(\sqrt{3}\Omega\eta_0T)/(\rho_0\omega^2)]$$
$$\times[(\omega-\omega_0)^2/(\omega+\omega_0)(3+\sigma)(5-\sigma)]\chi(\sigma),$$
$$\chi(\sigma)=1-\sigma^2/3-(\sigma+\sigma^2)$$
$$\times[\sigma^2/(1\cdot3\cdot5\cdot7)+\sigma^3/(3\cdot5\cdot7\cdot9)+\cdots],$$
$$\chi(0)=1,\quad \chi(1)=4/9,$$
$$\Gamma_K=[(\Omega\Lambda\eta_0T^2)/(63\rho_0^2D)]$$
$$\times[(\omega-\omega_0)^2(\omega+\omega_0)/(3+\sigma)^2(5-\sigma)^2]\psi(\sigma),$$
$$\psi(\sigma)=1-(692/825)\sigma^2+(146/2475)\sigma^3$$
$$+(4/75)\sigma^4+(13/7425)\sigma^5,$$
$$\psi(0)=1,\quad \psi(1)=(2044/7425),$$
$$\Gamma_K=[(\Omega\Lambda D)/(\rho_0\eta_0T)](\omega-\omega_0)^2.$$

In some cases the ratio (32) will be quite small, so that (23) gives essentially the total transport. For example, with fixed dimensions of apparatus this will be true provided the pressure is sufficiently high ($(\rho/D)\propto(\text{pressure})^2$). For the sake of simplicity we shall proceed with most of

our considerations on the assumption that this condition $(K_2/K)<1$ is satisfied. In cases where this is not true, it is only necessary to replace A in the following by

$$A' = A/[1+(K_2/K)]. \quad (33)$$

III. Separation Factor and Speed of Operation

Discontinuous operation

We shall now apply the results we have just derived to the case of *discontinuous* operation, by which is meant that one waits until the upper reservoir has approached its equilibrium concentration, and then removes all of its contents and starts over again. It is assumed that the reservoir is closed off from the tube during the removal of the gas; otherwise the mixing with gas drawn in from the tube would make it impossible to obtain the concentrations and amounts calculated here.

Our fundamental equation for these calculations is that for the total transport of species 1 up the tube. By (23) and (31) we have

$$\dot{m}_1 = H[c_1(1-c_1) - (2A)^{-1}(dc_1/dz)]. \quad (34)$$

At the final equilibrium, when the concentration in the top reservoir has ceased to increase, $\dot{m}_1 = 0$, and the solution of (34) is

$$c_1(z) = \tfrac{1}{2}\{1+\tanh A(z-z_0)\}, \quad (35)$$

where z_0 is determined by the condition that the total quantity of species 1 is conserved. If we set

$$c_1(-l) = c_1^0; \quad c_1(l) = c_2, \quad (36)$$

we find for the equilibrium *separation factor*:

$$(c_2/c_1^0)(1-c_1^0)/(1-c_2) = e^{2Al}. \quad (37)$$

If c_2 is small with respect to unity, this becomes

$$(c_2/c_1^0) \cong e^{2Al}. \quad (38)$$

When $\dot{m}_1$ is not zero, but has some constant value, that is before the final equilibrium has been established, the solution of (34) is

$$c_1(z) = \tfrac{1}{2}\{1+q\tanh qA(z-z_0)\}, \quad (39)$$

where

$$q = (1-4\dot{m}_1/H).$$

Under the boundary conditions $c_1(-l) = c_1^0$, $c_1(l) = c_1^l$ we have now

$$c_1^l = \tfrac{1}{2}[1-(q/2)\{q\tanh 2qAl - q\}]/[q\tanh 2qAl - q]\}. \quad (40)$$

where $q_0 = 1-2c_1^0$.

In practice, it will be advantageous to make the lower reservoir sufficiently large that c_1^0 does not drop appreciably during the separation, since if it should drop, one would not realize all of the separation factor. If the reservoir is sufficiently large, then c_1^0 is also the initial concentration of species 1. The constancy of c_1^0 may often be secured more conveniently by maintaining a continuous flow through the bottom reservoir. The quantity q_0 is then constant and equal to the initial value of q, and the equation for the time rate of change of c_1^l is

$$dc_1^l/dt = \dot{m}_1/m_l. \quad (41)$$

Let $t(c_1^l)$ be the time for the concentration of the top reservoir to rise from its initial value c_1^0 to some required value c_1^l. The integral of (41) may then be expressed as

$$t(c_1^l) = m_l\int_{c_1^0}^{c_1^l} dc_1^l/\dot{m}_1. \quad (42)$$

The integral must be done numerically or graphically, by calculating $\dot{m}_1$ from (40) for several values of c_1^l.

In many cases, however, a close approximation can be used, which eliminates the necessity of numerical integration. We note that

$$c_1(1-c_1) = c_1'(1-2c_1') + (c_1'-c_1)^2 \quad (43)$$

where c_1' is arbitrary. If c_1^l differs from c_1^0 by only a small amount, say not more than 0.2 or 0.3, one can select a value of c_1' and a constant to replace the last term in (43) in such a way that the resulting linear expression is a good approximation to $c_1(1-c_1)$ throughout the range in question. If in (34) the quantity $c_1(1-c_1)$ is replaced by such a linear approximation, then (34) and (41) can be integrated explicitly, and the formula for c_1^l so obtained represents a simple exponential approach to equilibrium, which can be characterized by a relaxation time. This gives us a useful way to describe the speed of operation for cases in which a linear approximation to (43) is valid. In the case that the

range of c_1 is larger than about 0.3, the approach to equilibrium is of a more complicated type, which can not be characterized even approximately by a relaxation time.

A suitable value for c_1 is the mean value of c_1' and c_1''. In the present case, the constant value inserted in place of $(c_1 - c_1')'$ will not appear in the relaxation time. Then by the procedure outlined, we find a relaxation time (time for $c_1' - c_1''$ to rise to $1 - e^{-1}$ of its equilibrium value):

$$\tau_{eq} = (m_1/H)[e^{H/(1-c_1)} - 1]/(1 - c_1). \quad (44)$$

There are several special cases of (44) worth mentioning. If c_1 is always small compared to unity,

$$\tau_{eq} = (m_1/H)(e^{H} - 1). \quad (45)$$

If the separation factor, which is e^{H} in this case, is much larger than one, we see that the relaxation time is directly proportional to the separation factor.

If, however, we are working in the middle region where both c_1 and c_2 are approximately one-half,

$$\tau_{eq} = 4(m_1/H). \quad (46)$$

While if c_1 is always close to unity,

$$\tau_{eq} = (m_1/H)(1 - e^{-H}). \quad (47)$$

It may perhaps be possible in practice to reduce the equilibrium time to some extent by a suitable variation of the pressure during operation. From (30) and (31) we see that H and K vary, respectively, as the second and fourth powers of the pressure. According to (23) the transport of species 1 is determined essentially by H during the early part of the separation process, while dc_1/dz is still small. Thus a high pressure at the beginning increases the transport. In order to reach a large concentration gradient finally, the pressure must be reduced. Ideally a proper manipulation would considerably advance the time at which the equilibrium is obtained. Such a procedure, however, meets a considerable technical difficulty, since the volume of the upper reservoir would have to be varied along with the pressure in order to prevent the loss of a great deal of gas down the tube.

Continuous operation

So far we have been discussing operation with no total transport of gas through the tube. In such operation the concentrated material is obtained by waiting until the approach to equilibrium has reached a certain stage and then removing the contents of the top reservoir. There is also available a continuous type of operation, in which a small steady flow of gas is removed from the top of the apparatus, so that there is a small total transport of gas along the tube.

The convection field with which we have been calculating, which is described by the function $G(T)$, provides no total transport along the tube. Because of the linearity of the hydrodynamical equation, however, we can superpose on the convection field an unidirectional velocity field corresponding to a very small additional pressure difference. Since the additional velocity field is unidirectional and c_1 varies only very slightly across the tube, we can omit all discussion of the details of the new field and the distribution of c_1 across the tube. We simply write for the transport of species 1 due to the new field the product of the mean value of c_1 at the given position along the tube and the total transport of gas. The *total* transport of species 1 is then

$$\Pi = c_1 \Gamma_1 - H c_1 - K(dc_1/dz) + \text{const.} \quad (48)$$

where c_1 is now constant along the tube. The quantities H and K may still be taken as defined by (30) and (31), since, with attainable speed of operation, the velocities in the superposed unidirectional field will in practice be extremely small.

As before we introduce $A = H/2K$, and the convenient dimensionless parameter

$$\lambda = \Gamma/H. \quad (49)$$

We shall treat only the equilibrium case, where c_1 has a constant value. There is little value in treating the approach to the new equilibrium, because the apparatus presumably would be operated with $\lambda = 0$ until c_1 rose to the value intended for continuous operation. Then we have $\partial c_1/\partial z = 0$ and therefore

$$\Pi = c_1 \Gamma - H c_1. \quad (50)$$

The differential equation for c_2 is now

$$[1-c_2+(1+\alpha)c_2-\alpha c_2^2]\,dc_2=2A\,dz, \quad (51)$$

whose solution is

$$c_2(z)=\{1+\alpha+\gamma\tanh\gamma A(C-z)\}/2\alpha, \quad (52)$$

where

$$\gamma=\{(1+\alpha)^2-4\alpha c_1\}^{\frac{1}{2}}.$$

Eliminating c_2 by the condition $c_2(-l)=c_1$, $c_2(0)=c_0$, we find

$$\tanh 2\gamma Al=\gamma(c_0-c_1)\{(1-\alpha)c_1+(1+\alpha)c_0-2\alpha c_1c_0\}^{-1}. \quad (53)$$

This rather formidable equation gives the important relation between α and c_0. It must be solved in any given case by successive approximations. The procedure may usually be based on the assumption that α is small, since only for small values of α can a large separation factor be realized. If, for example, one replaces γ by unity on the left number, and by the linear approximation $1+\alpha(1-2c_1)$ in the right member, one obtains as a first approximation

$$\alpha=\{(c_0+c_1)-2c_0c_1\}\tanh 2Al-(c_0-c_1)\}$$
$$\times(c_0-c_1)^2(1+\tanh 2Al-2c_1)^{-1}. \quad (54)$$

The quantity γ can then be calculated with the value $\alpha=0$ for α, and (53) can then be solved for the second approximation $\alpha(2)$, and so on. This particular procedure converges well if c_0 is small but fails entirely if $c_0\geqslant\frac{1}{2}$.

For cases in which the range of values of c_2 to be covered is small, one can simplify the analysis by replacing c_2 in (48) by a linear expression in c_2, just as was done in obtaining (44), (47) for discontinuous operation. In the present case the resulting formula involves the parameters of the linear expression in a more complicated way than does (44). Simple formulas can be given, however, for each of the three special cases corresponding to (45)–(47). When c_2 remains fairly small throughout, we can replace c_2 by c_0 in (48), and, remembering that α must be small, we obtain

$$(c_2/c_1)=(c_0/c_1)^{1-\alpha}. \quad (55)$$

Hence, in which c_2 is throughout in a range of values of perhaps 0.3 to 0.7, we can replace c_2 by $\frac{1}{2}$, and find

$$(c_2/c_1)=(1-c_1^{\alpha/2})/\alpha. \quad (56)$$

When $(1-c_2)$ remains fairly small, we can replace c_2 by $1-c_1$ in (48), and obtain

$$(1-c_2)/(1-c_1)=(c_1^{\alpha/(1-c_1)}-c_1)/(1-c_1). \quad (57)$$

These three formulas correspond precisely to (45), (46) and (47), respectively, and involve just the same linear approximations.

Continuous operation is the only practical method if we wish to use several stages in series, and the use of multi-stage apparatus seems distinctly desirable if we wish to combine a large separation factor with large speed of production. We have seen that if c_1 is small, the quantity $\alpha=\gamma/H$ must be small compared to unity. Therefore, in any stage, the transport is effectively limited by the quantity $Hc_1(1-c_1)$, where c_1 is the concentration at the bottom of the stage in question. If, now, c_1 is small, we see that in order to obtain a transport equal to that of a later stage, H must be larger for the first stage. A large H, however, means either a large K, and then the separation will be small, or a large B. A compromise would probably be used in practice.

The multi-stage apparatus should therefore consist of a series of vertical tubes; the first tube should be of large size and will produce a small change of concentration; subsequent tubes should be made progressively smaller.

Comparison of the two types of operation in a single-stage apparatus.

In the case of small c_1, formulas (45) and (55) make possible a simple direct comparison of the separations obtained by the two methods at a given effective speed, provided that H is large enough so that γ/H can be neglected compared to unity. Eq. (55) can be written

$$(c_2/c_1)=\theta/(1+\theta)\,P^{\alpha/(1-\alpha)}, \quad (58)$$

where

$$\theta=(c_0/c_1)^{1-\alpha}/(H/\gamma)=\alpha. \quad (59)$$

θ is a parameter inversely proportional to the speed, and c_2/H is the equilibrium separation given by (38). A third quantity also obtained by

setting $\omega = 0$, or $\theta \to \infty$. In *discontinuous* operation the quantity which corresponds to γ is m/t_0, where t_0 is the time allowed for the separation to approach equilibrium before the reservoir is emptied. Then if we put

$$\gamma' = H/(\omega + 4H) \qquad (59)$$

equal values of γ and γ' correspond to equal effective speeds of production by the two methods. According to Eq. (45), the exponential approach to equilibrium now takes essentially the form

$$(\xi_0/\xi) = (1 - \gamma')^{2H/t_0} \qquad (60)$$

The expressions (58) and (60) show that, at a given effective speed, a greater fraction of the equilibrium separation ratio can be obtained by using discontinuous operation.

In the case of γ near one-half, we can introduce the parameters

$$\gamma = (\Delta t_0)^{-1} \qquad \gamma' = H_0/(\Delta t_0 m_0) \qquad (61)$$

equal values of which correspond to equal effective speeds. The equilibrium concentration difference, from (56) with $\omega = 0$ is Δt, and the realized concentration differences are, by (56) and (46),

$$(\xi_0 - \xi) = \gamma(1 - \gamma^2/3)\,\Delta t \qquad (62)$$

for continuous operation, and

$$(\xi_0 - \xi') = (1 - \gamma'/3)\,\Delta t \qquad (63)$$

When less than a fraction $(1 - \gamma')$ of the equilibrium concentration difference is to be obtained, the continuous method works slightly better; otherwise, the discontinuous is better.

In the remaining case, γ nearly equal to one, it is not possible to introduce a single parameter characterizing the speed in such a way that a simple comparison of the two methods is possible. Rough computation from (47) and (57) shows that when large ratios $(1 - \xi_0)/(1 - \xi)$ are required, continuous operation is decidedly faster.

General remarks on speed of operation

In the case of *discontinuous* operation, the quantity which characterizes the speed of operation in terms of the mass of the element with a given concentration, divided by the time required to produce it, is m/t_0, where t_0 is the fraction of the molecular weight contributed by the element in question. This characteristic quantity varies as H/t_0. In the continuous case, the corresponding quantity is $\gamma/\Delta t$, which, for a given concentration, also varies as H/t_0. Thus in either case the variation of the speed with t_0 is given by

$$\gamma = H/t_0 = (\omega/\pi)B/\rho)(\Delta T/T). \qquad (64)$$

Since Δt must have a given value, and since $D = \eta/\rho$, we see from (31) that ω must vary as

$$\omega = (\eta)^{\frac{1}{2}}(\eta/\rho)^{\frac{1}{2}}. \qquad (65)$$

If we accept (3), ω varies as η, and we have the significant relation

$$\gamma = (\pi^{\frac{1}{2}}/4)^{\frac{1}{2}}(\eta/\rho)^{\frac{1}{2}}B(\Delta T/T). \qquad (66)$$

To get a large speed at the specified concentration, it is evident that one should choose as slight an molecule as possible, because of the strong dependence on η. The factor B can be increased by using a larger tube, or several tubes in parallel. Since under ordinary conditions ρ is proportional to the pressure, the speed varies as the square root of the pressure and as the three-fourths power of the length, *provided* that r_2, the difference of the radii, is readjusted for changes of pressure and length in accordance with (65).

It is assumed in the argument leading to (66) that the ratio K_1/K is very small compared with unity, since otherwise we should not be justified in using (31) and should use instead the modified value (33).

The power expended because of conduction is by (29)

$$P = \Delta B(\Delta \lambda BQ) = 4\lambda BN\Delta BAT/\omega \qquad (67)$$

in watts, and, by (65), is proportional to $\eta^{\frac{1}{2}}\Delta BAT$; the viscosity and heat conductivity are independent of the pressure. Thus so far as conduction is concerned, the speed and power are just proportional to one another, as long as a fixed concentration difference is demanded, and a fixed temperature difference is used. The advantage of a rather large ΔT is evident, but there is little use in going to extreme temperatures, since it is the ratio of ΔT to the *mean* temperature that is involved, and also since the radiation loss will be large if T is made very

high. The power consumption due to radiation between the walls is independent of w, and for nontransparent tubes is given by

$$P_r = 2B(1-R_1)(1-R_2)\times(1-R_1R_2)^{-1}\sigma(T_2^4-T_1^4), \quad (68)$$

where R_1 and R_2 are the average reflection coefficients of the walls of the tubes, and $\sigma = 3.7\times10^{-12}$ watt/cm² deg.⁴.

Effects of Asymmetry; General Constructional Considerations

We shall here consider briefly the effect of asymmetry in the apparatus. Due to non-uniformity of heating or cooling of the tubes, it is possible that the temperature may vary around the tube. This will give rise to an additional convection, characterized by a slight upward flow at the hotter part of the annulus, and a corresponding downward flow at the colder part. For simplicity, we may suppose that the temperature field is still independent of z, but varies with the azimuthal angle, φ, in the simple manner

$$T = T(r) + (\delta T/2)\cos\varphi. \quad (69)$$

One can then show that the essential effect is to introduce another contribution to the transport given by $-K_\varphi\,dc_1/dz$, where

$$K_\varphi/K = (\delta B^2/16\sigma w^2)(\delta T/\Delta T)^2, \quad (70)$$

provided that $B \gg w$. Since K_φ/K must be kept fairly small, Eq. (70) imposes rather strong conditions on the tolerable asymmetry of the temperature in the apparatus to be discussed in the next section. If K_φ would become equal to K for $\delta T = \Delta T$.

A lack of centering of the tubes would have a considerable direct effect on performance, because H and K depend on the third and seventh powers of w. The asymmetry in temperature brought about by the lack of centering would, if appreciable, be still more harmful. To minimize it, good conduction is essential. Thus metal tubes are to be preferred on several accounts: for precision in construction, high conductivity and large reflection coefficient to reduce radiation loss.

The importance of symmetry makes it evident that advantages in speed and concentration should be sought by increases in length rather than by the use of high pressures and small values of w. Also, both for accuracy of construction and because of the factor B^2 in (70), it is better to use several tubes in parallel rather than large tubes.

To avoid lack of symmetry in the temperature distribution, the water cooling must be efficient and the heating of the inner tube must be very uniform. Heating by condensation of a vapor rather than by resistance wire seems advisable.

IV. A Numerical Example: C^{13}

In order to illustrate the application of our formulas we shall apply them to the design of a single-stage apparatus for the concentration of C^{13}. Here we are concerned with the concentration of a *heavy* isotope at the *bottom* of the tube. Our equation for A and all our equations concerned with the speed of operation apply here with no change except the replacement of c_2 by c_1. A formal justification of this rule is provided by inspection of the fundamental equations (23) and (48). In these equations we must interchange c_1 and c_2; change the sign of the term in $\partial c_2/\partial z$ since this term changes sign in rewriting (40) for species 2; and change the signs of the $\partial c_1/\partial z$ terms, because we are now concerned with transport *down* the tube. When this is done the equations are unchanged in form except for a change in sign of the term in $\partial c_1/\partial z$. This is proper, since the concentration is now increasing downward instead of upward.

We take as the specifications of our apparatus: length of tube, $2l = 800$ cm; $2B = 2.7$ cm; $T_1 = 300°K$; $T_2 = 600°K$; pressure, 1 atmosphere; and equilibrium concentration, $c_1 = 0.30$. Since the carbon isotope of mass 13 occurs with a natural abundance of one part in about 92, and therefore $c_2^0 = 0.0108$, it follows at once from (37) that we must have $\alpha H = 39.3$, or $H = 0.0023$/cm.

The required value of the difference in radii, $2w$, and the speed of operation are now determinate, once we have selected the molecule which is to carry the carbon.

Our general discussion of the speed shows

[1] Vaughan, Williams and Tate, Phys. Rev. 46, 317(A) (1934).

that one should use as slight a molecule as possible (cf. Eq. (66)). Methane, CH_4, is therefore eminently suitable. Extrapolating to 450°K the data for CH_4 given in the *International Critical Tables*, we find

$$\begin{aligned} \eta &= 1.60\times10^{-4}\ \text{poise}, \\ \rho &= 0.433\times10^{-3}\ \text{g/cm}^3, \\ D &= \eta/\rho = 0.32\ \text{cm}^2/\text{sec}, \\ k &= 1.20\times10^{-4}\ \text{cal/cm sec deg}, \end{aligned} \tag{71}$$

and by (3)

$$\kappa_0 = 0.0106.$$

The conditions on the relaxation length λ, (Eq. (4)), and the Reynolds number (5) are strongly satisfied, as we shall verify after λ_0 has been evaluated. In the case under consideration, however, it is not true that $\lambda/K \ll 1$, so that the effect of longitudinal diffusion cannot be ignored. Thus we must equate not λ, but λ/K, to the value 0.0023/cm.

Putting the values (71) in (31) and (32), we find

$$\lambda = 3.3\times10^{-3}/w^{1/2} \quad \text{and} \quad \lambda/K = 6.1\times10^{-3}/w^{1/2}.$$

Since λ/K must have the value 0.0023, we have by (33),

$$w^{1/2} - 1.5\times10^{-3}w^{1/2} + 3.1\times10^{-3} = 0.$$

The larger of the two positive real roots gives the value $w = 0.62$ cm. The ratio λ/K is about 0.58, so that it certainly could not have been neglected.

In order to discuss the question of speed, we must know the value of H. We find by (30),

$$H = 2.3\times10^{-7}/\text{sec}.$$

The range of ξ in question, 0.01 to 0.30, is just within the range of validity of the linear approximation (44). If we wait a length of time of the order of the relaxation time, we shall reach a concentration in the bottom reservoir of only about 0.19. In this case, we are concerned only with the range 0.01 to 0.19, and a suitable value of ξ is 0.10. If we suppose the bottom reservoir contains 3.0 grams of gas, or approximately 10^4 cc, the relaxation time, given by (44), is

$$\tau = (3.0/2.3\times10^{-7})\times(39.3)^{1/2} = 11/0.8$$
$$= 1.9\times10^{7}\ \text{seconds} = 7\ \text{days}.$$

The speed of production by the discontinuous method is thus 88 mg/day of methane, or about 66 mg/day of carbon, with a concentration of 19 percent C^{14}.

A rough idea of the speed obtainable in continuous operation may be obtained from Eqs. (58) and (60). These formulae really apply only in the approximation $\xi = 0$, but should be fairly reliable for purposes of comparison when a more accurate value has already been found for one method. The use of the time τ between removals in discontinuous operation corresponds to $\theta = 1$ in (60). Putting $\theta = 1$ in (58), we see that continuous operation at a speed of 88 mg of methane per day will give a concentration of roughly $\frac{1}{8}$, or about 13 percent C^{14}. To obtain an estimate of the speed at concentration 19 percent, we put

$$\theta = 7/(0.19) = 1 - e^{-\theta}$$

and obtain $\theta = 1.72$. Since θ is inversely proportional to the speed, continuous operation will give roughly $88/1.72 = 51$ mg/day of methane at 19 percent C^{14}.

The speed of production at 19 percent C^{14} can be calculated exactly from (53). The estimated value just obtained corresponds to $\alpha = 5.9\times10^{-7}/\text{sec}$, or $\alpha^{(0)} = \tau H = 0.0259$. This can be used as a first approximation in solving (53). It gives $\gamma = 1.0161$, $\tanh 2\gamma = 0.95315$. The next approximation is now obtained by solving (53) for α as it occurs explicitly. The value obtained is $\alpha^{(1)} = 0.0290$, and the value to which the approximations converge is $\alpha = 0.0279$. This corresponds to a speed of 55 mg/day.

The power consumption due to conduction, by (66), is 2450 watts, while that due to radiation, by (67), would be 7000 watts if the tube walls were perfectly black ($R_1 = R_2 = 0$). If the walls were a polished metal, however, this value might well be cut down to less than 500 watts.

The Reynolds number, by (5), is of the order of 3. Since in the case of pure pressure flow through tubes or between plates this number must become of the order of several thousand before spontaneous turbulence occurs, it is difficult to believe that there could be any turbulence here.

By (4), the length of tube at each end in which the temperature gradient differs from

65-4307-1-B (5)

that determined by conduction is of the order of 10⁻⁴ cm, and thus may certainly be neglected.

The calculations given above for C^{α} are based on the value $\sigma = 0.0106$ which is given by Eq. (3). This equation is, however, only a rough guess, still at the best one possible. It may be found by experiment that the actual values of H and K for any given gas are distinctly different from those calculated from values of σ given by (3), perhaps by as much as a factor 2. In practice, therefore, it will be necessary to do some preliminary work to find the actual value of σ. It is suggested that this be done by running the apparatus at a fairly high pressure. For instance, in the example just discussed, the use of a pressure of two atmospheres instead of one will multiply H by 4 and divide K by approximately 4, so that the separation factor will be small, but so will also be the equilibrium time. If one now measures α as a function of the time, one will obtain the values of both H and K. Either one will yield the value of σ by (30), or by (31), (32) and (33). We may now extrapolate K to the desired value by changing the pressure, since we know that H/K and K^2/K vary, respectively, as the second, inverse second, and inverse fourth power of the pressure.

Preliminary adjustments such as those just described will of course be necessary only in the first work on each gas. Once the correct value of σ is known from experiment, our equations should make it possible to design apparatus which will perform in acceptable agreement with specifications.

One of us (L. O.) is indebted to Dr. G. Akerlof and Dr. C. W. King of the Chemistry Department, and to Dr. C. C. Furnas of the Chemical Engineering Department of Yale University for advice in regard to feasible construction and operation of apparatus on laboratory and industrial scale. The others (W. H. F. and R. C. J.) are grateful to Professor K. T. Bainbridge and Dr. R. Sherr of the Physics Department of Harvard University for interesting and helpful conversations.

Calculation of the Second Virial Coefficient of Helium Gas for the Lowest Measured Temperature

LOUIS GROSSER
Department of Mathematics, Brooklyn College, Brooklyn, New York

(Received April 3, 1939)

IN RECENT articles an explicit and calculable expression has been derived for the second virial coefficient of a monatomic gas. This is the B in the equation of state

$$pV/RT = (1 + B(T)/V + C(T)/V^2 + \cdots)$$

and, assuming the Einstein-Bose statistics, is given by the formula

$$B = \frac{N_0 V}{2} - 16N_0^2 V^2 \sum_j (j+1) \times \exp[N_0 V^2 \psi(r_j)/\alpha^2]$$

$$16\pi^2 N_0^2 \sum_j (j+1) \int_0^\infty r^2 \times \exp[-N_0 V^2 \psi(r)/\alpha^2] \, dr \quad (1)$$

Here $\psi = \psi/mkT$, ψ is the phase found from the phase equation for the relative motion of two radially interacting molecules,

$$\frac{d^2\psi}{dr^2} + \left(\frac{8\pi^2 m}{h^2}\left(\frac{\alpha^2}{r^2} - \psi\right) - \frac{(j+1)}{r^2}\right)\psi = 0$$

$\alpha(j)$ are the discrete states for two such molecules, α is the Bohr radius, and ψ is the interaction potential between the two molecules.

Extensive calculations have recently been made with formula (1) by Massey and Buckingham on the inert gases, in particular, helium. These calculations are based on the use of the Slater-Kirkwood potential for ψ. The calculation of

[1] L. Holborn and G. E. Otto, Zeitschr. f. Physik, 33, 913 (1937); L. Grosser, Phys. Rev. 51, 1108 (1937).

[2] H. S. W. Massey and R. A. Buckingham, Proc. Roy. Soc. London, 168, 378 (1938).

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Wt 150 lbs.
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Raymond Baker

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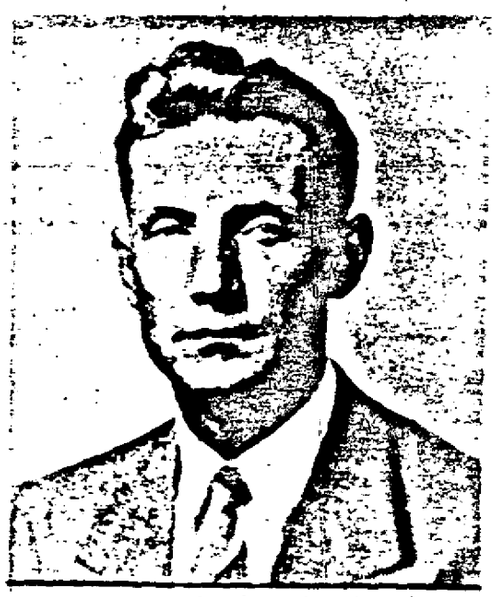

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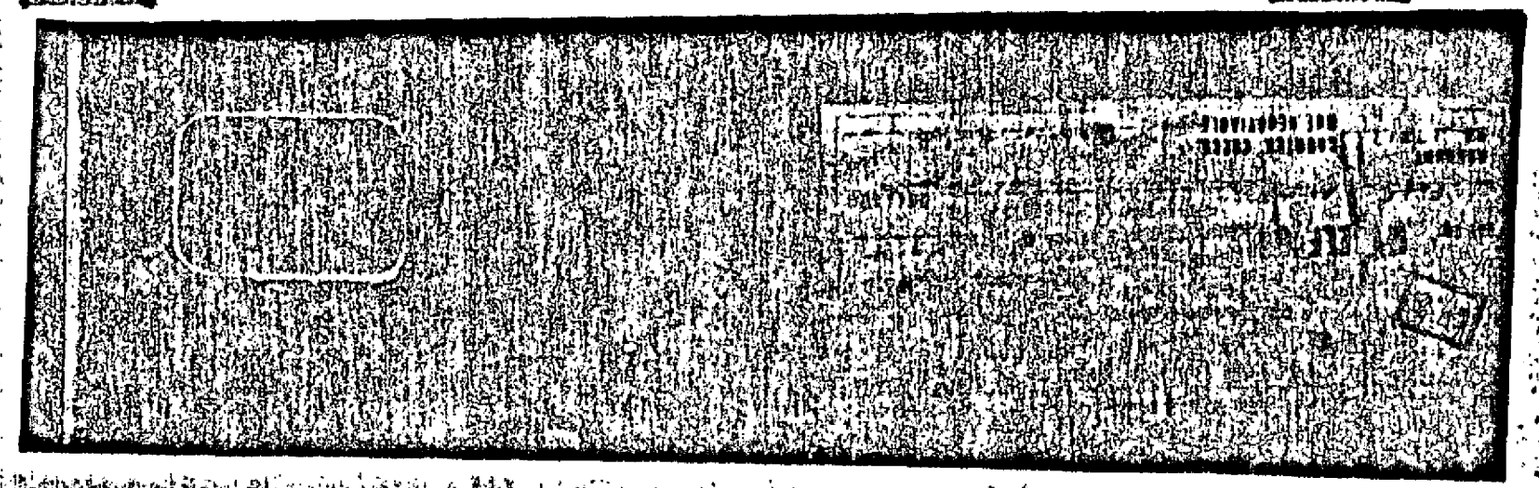

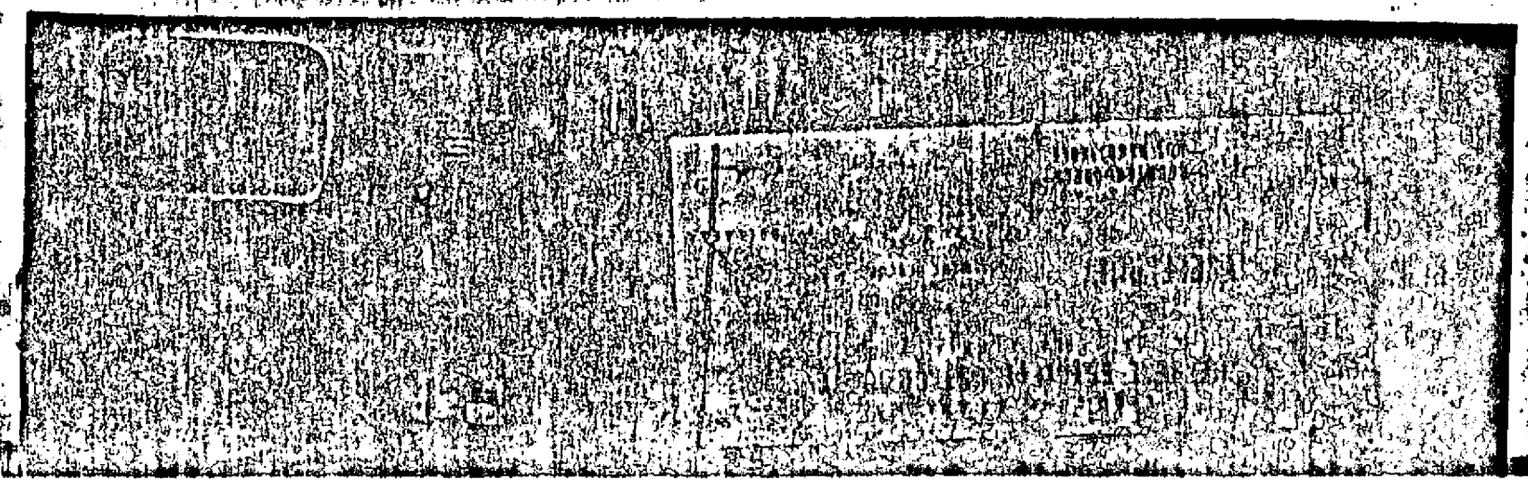

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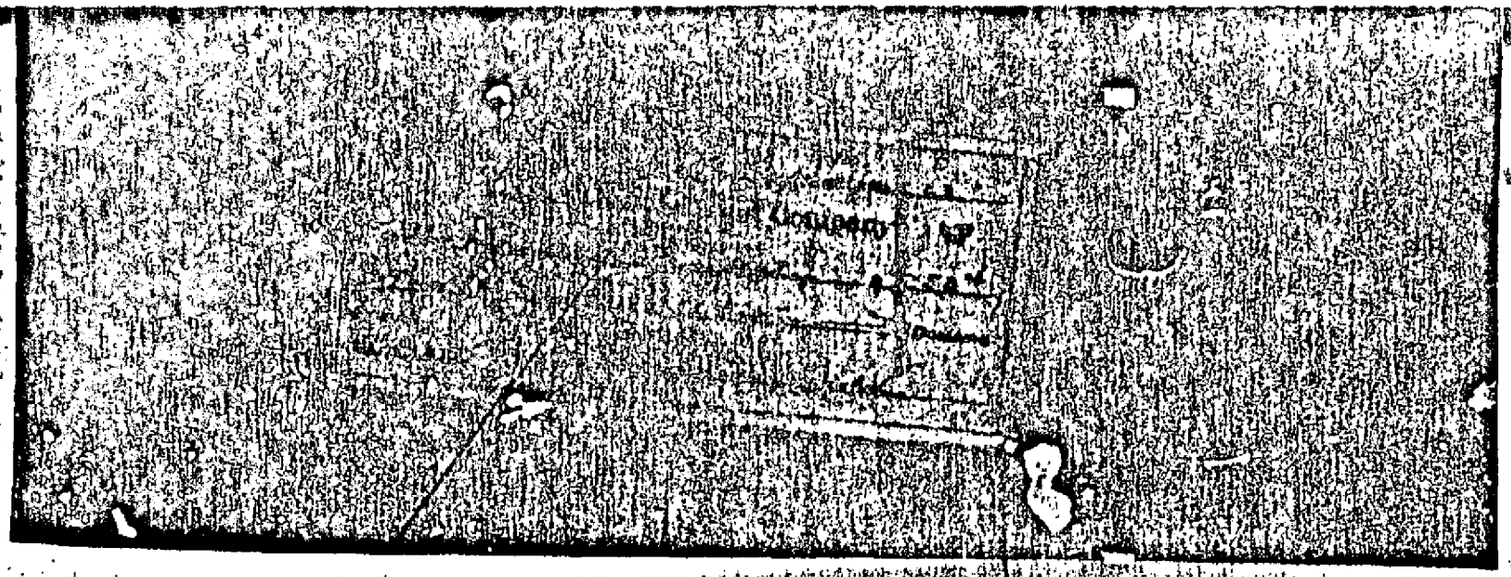

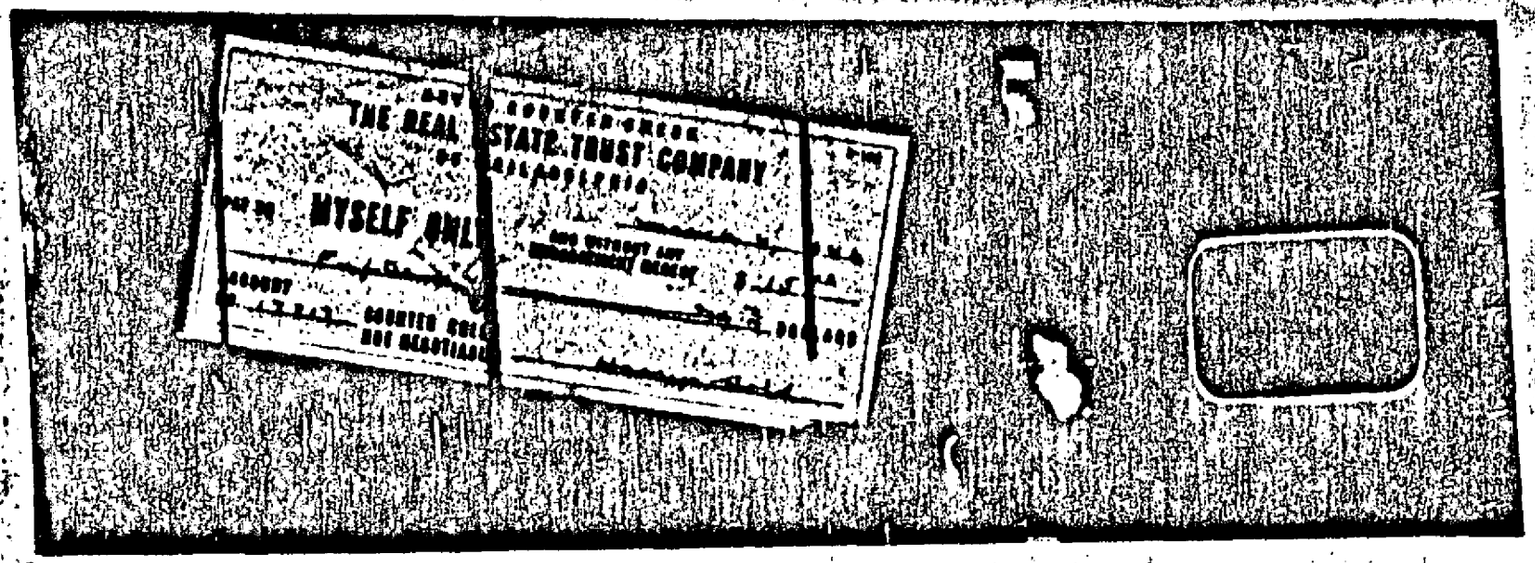
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File No. 65-4307-1-B (17)

APPLICATION FOR: Advance on Salary [X]
Employee Loan [ ]

February 8, 1945
(Date)

To: The Treasurer of Pennsylvania Alcohol & Chem. Corp. Subsidiary of Pennsylvania Sugar Company
(Company)

Amount of Advance [X] Loan [ ] desired $500.00

Reason for the application: Certain commitments were made by me, based on information given to me by an official of the Company that we had been granted a salary increase retroactive to May 8; we were never given to believe otherwise.

Name Harry Gold Department Alcohol Laboratory

Salary per week $48.48

Remarks: In order for me to obtain the full benefit of this loan, I would like to amortize at the rate of $5.00 per week for the present.

If this Advance [X] Loan [ ] is granted I agree to have $5.00 withheld from salary each week, starting with week beginning 2/16/45 until Advance [X] Loan [ ] has been repaid.

Harry Gold
(Signature)

Recommended: V. K. Hamilton
(Department Head)

Approved: F. Burke
(Pres. - Ex. V.P.)

A. J. Stocker
(Treasurer)

Board of Directors
(Board of Directors)

February 21, 1945
(Date)

65-4307-1-D (17)

Advance on salary ☐

APPLICATION FOR

Employee Loan ☐

Pennsylvania-Alcohol & Chem. Corp.

Subsidiary of

(Date)

Pennsylvania Sugar Company

(Company)

Amount of Advance ☒ Loan ☐ desired $100.00

Reason for the applications certain emergencies were made by me, namely one

information given to me by an officer of the company that we had been

granted a salary increase retroactive for May 8, we were never given the

allowe other vise

Item Hour's Gol Department/Alcohol Laboratory

Salary per week $40.40

Enclosing the order for and for obtain the milk benefit for this loans I would

like to amortize the sum rate of $2.00 per week, from the present

In the Advance ☒ Loan ☐ be granted I agree to pay $2.00 weekly from

my each week, starting with the week beginning until Advance ☐

has been repaid.

(Signature)

(Department Head)

Approved

(Treasurer)

(Board of Directors)

Board of Directors

(Date)

February 21, 1942

(Date)

(Personnel Dept.)

65-4307-1-B(17)