

THE APPLICATION OF FREE ENERGY EQUATIONS TO THE STUDY OF THE SYNTHESIS OF HYDROCARBONS FROM CARBON MON-OXIDE AND HYDROGEN.*

By W. W. MYDDLETON, D.Sc., F.R.I.C., F.Inst.Pet.

SYNOPSIS.

Equations have been derived relating free energy of reaction to temperature in the synthesis of a series of gaseous hydrocarbons from carbon monoxide and hydrogen.

In building up these equations molecular heat-temperature curves were drawn for each reactant and product and molecular heat-temperature equations were fitted over the range 25–250° C., existing equations being generally unsatisfactory for the temperature range covering synthesis. Some use has been made of molecular heat and entropy values calculated from statistical and spectroscopic data.

The suppression of reactions in which carbon dioxide is formed, in spite of free energy changes favourable to them, is attributed to special conditions prevailing at the catalyst surface.

The appearance of traces of ethylene amongst the products of synthesis instead of appreciable quantities predicted by the free energy relationships is accounted for by the incorporation of ethylene actually formed into the building up of hydrocarbon chains.

The possibility of increasing the yields of *isobutane* and *isobutene* is considered worthy of attention. The formation of aromatic hydrocarbons requires a special orientation of reactants on the surface of the catalyst, and the probability of the necessary conditions occurring is slight.

THE GASEOUS HYDROCARBONS.

The symbols employed are in accordance with the Joint Report of the Chemical, Faraday, and Physical Societies, 1937.

THE FREE ENERGY EQUATIONS AND THEIR USES.

In a study of the thermal characteristics of a reaction such as $n\text{CO} + (2n + 1)\text{H}_2 = \text{C}_n\text{H}_{(2n + 2)} + n\text{H}_2\text{O}$ the thermal data available may be assembled in the form

$$\Delta H_T = \Delta H_0 + AT + BT^2 + CT^3 \dots \dots (1)$$

where ΔH_T is the change in heat content during reaction at T° K. and ΔH_0 may be regarded as the change in heat content in hypothetical reaction at absolute zero. A, B and C are constants for a reaction producing any one hydrocarbon.

The application of equation (1) gives information necessary for the provision of adequate means of controlling the temperature in an exothermic reaction. Its general usefulness can be extended by deriving from it an equation showing the change in free energy during the reaction.

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We thus obtain a thermodynamical approach to a series of practically important questions such as the following:—

1. What operating conditions can most profitably be explored?
2. Is any other product or any other type of reaction of the same order of thermodynamic probability as the main reaction we are considering? When this is the case there is always a danger, to be explored and provided for, of some alteration in working conditions or in the form of catalyst suddenly provoking a disturbance and perhaps producing undesirable products.
3. Can the non-occurrence of equally probable reactions be accounted for by a different mechanism at the catalyst surface, such that the conditions necessary for its accomplishment cannot be fulfilled in the prevailing circumstances? The answer to this question may serve to evaluate the risk of undesirable complications dealt with in the previous question.
4. Do thermodynamical considerations show that some of the more desirable products can be formed in larger proportion?

Such questions as these will be discussed with reference to the gaseous hydrocarbons which normally accompany liquid and solid hydrocarbons during synthesis from carbon monoxide and hydrogen.

For the development of equation (1) the constants A, B and C are derived from molecular heat-temperature equations for all the substances involved in the reaction. For each reactant and product an equation is obtained in the form $C_p = a + bT + cT^2$. Then by algebraically summing the molecular heats of the products and subtracting the sum of the molecular heats of the reactants we obtain ΔC_p for the reaction.

Substituting in the equation $\left(\frac{\partial \Delta H}{\partial T}\right)_p = \Delta C_p$ and integrating we obtain A, B and C in equation (1). The integration constant ΔH_0 is evaluated by finding the value of ΔH_T for some fixed temperature in the range for which the molecular heat equations are valid. This is readily done by employing the data on heats of combustion of the substances involved in the reaction.

Substitution from equation (1) in the thermodynamical equation
$$\frac{d \Delta G}{dT} = -\frac{\Delta H}{T^2}$$
 followed by integration gives us the final free energy equation

$$\Delta G_T^\circ = \Delta H_0 + A'T \ln T + B'T^2 + C'T^3 + \dots + IT \quad (2)$$

The integration constant I can be evaluated in relatively few ways. The value of ΔG_T° must be found for some fixed temperature in the specified range. For this purpose the equation $\Delta G_T^\circ = -RT \ln K$ is not generally applicable to the reactions we are considering because the equilibrium constant K is not often available. The most useful procedure is to apply the relation $\Delta G_T^\circ = \Delta H_T^\circ - T\Delta S_T^\circ$ at $T = 298.1^\circ \text{K.}$, where ΔS_T° is the sum of the molecular entropies of the products at 298.1°K. less the sum of the molecular entropies of the reactants.

Previous thermodynamical treatment of the subject¹ has been marred by one or more of the following factors :—

A. Inaccurate thermal data such as heat of combustion, molecular heat at constant pressure, and entropy.

B. Lack of information on rates of reaction.

C. Lack of information on the difference between free energy change as normally calculated for conditions in which each reactant and each product is at atmospheric pressure, and, under the actual conditions of synthesis, when partial pressures are far removed from atmospheric.

These difficulties have been considered in the order given.

A.1. The Heat of Combustion of Reactants and Hydrocarbon Products.

In recent years highly accurate data on heats of combustion have been provided by Rossini, Jessup and their co-workers.

A.2. Molecular Heat at Constant Pressure.

We are concerned with the conditions in the catalyst space, and molecular heat values should therefore refer to gases and vapours. When we examine recorded experimental figures, we find very serious discrepancies, even in the case of such gases as the oxides of carbon. A critical study of the methods employed in the observations reveals that, where possible sources of error have been reduced and when special refinements in apparatus or technique have been introduced, the values obtained are in good agreement with those calculated from the characteristic vibration frequencies of the molecules by the application of the Planck-Einstein equation.²

For such gases molecular heats have been calculated from what are considered to be the most reliable vibration frequencies recorded in the literature.

Calculation is as yet unsatisfactory in the case of the more complex gases. Not all of the characteristic frequencies of a complex molecule can be derived from the infra-red and Raman spectra, the usual source of such information, and, in fact, experimental specific heat values have been employed to predict missing frequencies in ethylene³ and ethane.⁴ Another kind of difficulty is found in ethane and in general in molecules containing a C-C bond. There is undoubted evidence of a restriction in the freedom of rotation about this bond, and until its magnitude has been ascertained we cannot calculate the molecular heat due to the internal rotations of the molecule.^{4, 5}

For hydrocarbon gases other than methane, therefore, the best experimental values have been selected from the literature.

Method of Calculation.

The contribution made to the molecular heat at constant volume by

each mode of vibration of the molecule was found by applying the Planck-Einstein equation in the form

$$C_{vib} = R \left[\frac{c^{\frac{\theta}{T}} \left(\frac{\theta}{T} \right)^2}{\left(e^{\frac{\theta}{T}} - 1 \right)^2} \right] \times (\text{Degrees of freedom})$$

where R is the molecular gas constant, taken here as 1.9863 cal.; c is the base of natural logarithms; θ is the characteristic temperature of the vibration = $1.4327 \times (\text{Number of wave-lengths per centimetre})$.

The selection of characteristic frequencies from the spectra and the assignment to them of their appropriate degrees of freedom has become a highly specialized branch of science. An extensive examination of the data recorded in the literature has been made.

The total molecular heat at constant volume was obtained by adding the effects due to translation, rotation and vibration.

<i>Type of molecule.</i>	$C_v = C_{trans.} + C_{rot.} + C_{vib.}$
Diatomic.	$C_v = \frac{3}{2}R + R + C_{vib.}$
Triatomic linear.	
CO ₂ type.	$C_v = \frac{3}{2}R + R + C_{vib.}$
Triatomic non-linear.	
H ₂ O type.	$C_v = \frac{3}{2}R + \frac{3}{2}R + C_{vib.}$
Polyatomic. CH ₄ type.	$C_v = \frac{3}{2}R + \frac{3}{2}R + C_{vib.}$

Calculation of C_p at atmospheric pressure.

Conversion of C_v to C_p at infinite dilution was obtained from the equation $C_p = C_v + R$.

The correction for increase in pressure to 1 atmosphere was made in the usual manner by assuming that the gas obeys Berthelot's equation of state. Then if P_c and T_c are the critical pressure and temperature

$$C_{p-1} = C_{p-0} + R \left[1 + \frac{81}{32} \times \frac{1}{P_c} \times \left(\frac{T_c}{T} \right)^3 \right].$$

A.3. Molecular Heat Equations.

Many of the molecular heat equations in common use are constructed for temperatures beyond the range in which we are interested. New equations have therefore been obtained fitting the graphs molecular heat-absolute temperature for each gas over the range 25° to 250° C.

A.4. Evaluation of the Integration Constant ΔH_0 .

The value of the integration constant ΔH_0 in the free energy equation (2) can be found from equation (1) when we find ΔH_T at some definite temperature within the specified range for which the molecular heat equations have been constructed.

Heats of combustion afford the means of doing this, and as they are usually reported for 298.1° K., this temperature is used for determining ΔH_T . Reported values account for any water formed in the liquid state.

We must therefore take into account the latent heat of vaporization of water at 298.1° K.

A.5. *Molecular Entropy.*

The experimental determination of entropy involves the measurement of specific heat down to temperatures preferably below 10° K., and thereafter a graphical extrapolation to absolute zero. The method of extrapolation has been simplified by Kelley, Parks and Huffman.⁶ The latent heat appropriate to any change in state or form between absolute zero and 298.1° K. must be measured and used in the calculation.

The difficult procedures in the determination make it desirable to compare experimental values with those calculated for the simpler types of gas from statistical and spectroscopic data.

Important discrepancies between the two sets of values in the cases of carbon monoxide, water vapour and hydrogen require investigation before we decide to adopt any particular figure.

Entropy of Carbon Monoxide.

If the assumption is made that the molecule behaves as though it were unsymmetrical in the crystalline solid, the entropy due to rotation is 11.375 entropy units, and the total entropy of the gas at 298.1° K. is 47.298 E.U. The experimental value reported by Clayton and Giauque⁷ is 46.22 E.U.

There is some independent experimental evidence that the molecule behaves as though it were symmetrical. If this were the case, the rotational entropy would be 9.998 E.U. and the total entropy at 298.1° K. would be 45.92 E.U. The value taken for the calculation of free energy is the mean value of 46.07 E.U.

Entropy of Water Vapour.

The calculated value is $S_{298.1}^{\circ} = 45.09$ E.U. Direct experimental methods according to Giauque and Stout⁸ gave $S_{298.1}^{\circ} = 44.28$ E.U. A suspicion of randomness in the ice crystals led to an indirect method in which Giauque and Archibald⁹ studied the reaction $\text{Mg}(\text{OH})_2 = \text{MgO} + \text{H}_2\text{O}$. This method gave $S_{298.1}^{\circ} = 45.10$ E.U.

Entropy of Hydrogen.

The experimental determination by Giauque¹⁰ gave $S_{298.1}^{\circ} = 29.69$ E.U. From statistical and spectroscopic data Giauque¹¹ calculated $S_{298.1}^{\circ} = 31.23$ E.U. The discrepancy has been accounted for by Moelwyn Hughes¹² as being due to a difference between the entropies of *ortho*- and *para*-hydrogen. The entropy of the equilibrium mixture is calculated to be 29.59 E.U. from statistical and spectroscopic data.

The evidence at first sight appears to favour the adoption of the lower figure. It is, however, improbable that the same equilibrium mixture is concerned in synthesis. Furthermore, we can derive a value for the entropy of hydrogen from the free energy of the reaction $\text{H}_2 + \frac{1}{2}\text{O}_2 = \text{H}_2\text{O}$ for which four different methods give $\Delta G_{298.1}^{\circ} = -54,507$ cal. as an average.¹³ Assuming for water vapour $S_{298.1}^{\circ} = 45.10$ E.U. and for oxygen $S_{298.1}^{\circ} = 49.07$ E.U. we derive for hydrogen $S_{298.1}^{\circ} = 31.79$ E.U.

The conditions under which the free energy of formation of water was determined appear to exclude the possibility of normal equilibrium mixture of *ortho*- and *para*-hydrogen being involved.

The value 31.23 E.U. has been adopted.

The entropies of gases are usually referred to standard conditions at 298.1° K. and 1 atmosphere pressure and, furthermore, the gas is assumed to be in the ideal state in which the fugacity is also 1 atmosphere. When the gas is not ideal a correction can be applied by assuming, for example, Berthelot's equation of state. A gas obeying this law has an entropy lower than that of an ideal gas¹⁴ by $R \frac{27P}{32P_c} \left(\frac{T_c}{T} \right)^3$.

The correction is negligibly small for the gases and pressure and temperature conditions we are investigating.

For hydrocarbons other than methane the best experimental entropies have been adopted.

B. Rate of Reaction.

A method has been described for the measurement of the time of contact, and hence the rate of reaction under the conditions of gas flow of practical synthesis.¹⁵ Observations will be given on the rate of formation of methane and carbon dioxide.

C. Correction to be Applied to Free Energy of Reaction for Differences in Pressure.

Free energy equations such as (2) are based upon the assumption that each gaseous product and reactant exerts a pressure of 1 atmosphere. We can estimate the approximate correction to be made for the actual conditions operating during synthesis.

For this purpose it has been assumed that blue water gas is used for synthesis and that 75 per cent. of the available carbon monoxide is converted into a single hydrocarbon at 200° C. and at a total pressure of 1 atmosphere. The partial pressures of reactants and products in the residual gas have been calculated for these conditions.

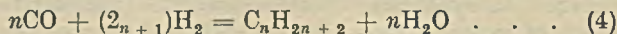
From the equation

$$G_x - G_1 = \int_1^x V dP \quad \dots \quad (3)$$

where the suffix 1 relates to the standard pressure of 1 atmosphere and x to the actual condition, we obtain

$$G_x - G_1 = RT \ln \frac{P_x}{P_1}$$

and for the general reaction



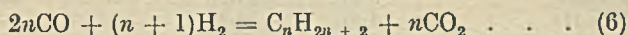
we have

$$G_x - G_1 = RT \ln \frac{p(\text{C}_n\text{H}_{2n+2}) \times p^n(\text{H}_2\text{O})}{p^n(\text{CO}) \times p^{2n+1}(\text{H}_2)} \quad \dots \quad (5)$$

For the conversion of 1 gram molecule of carbon monoxide in a reaction of this type the following values of $G_x - G_1$ have been calculated :—

Hydrocarbon formed.	Correction, calories.
Methane	2415
Ethane	2232
Propane	2255
Butanes	2264
Ethylene	2005
Propylene	2092
Butenes	2127

There is an alternative type of reaction in which carbon dioxide is formed instead of water.



For this type of reaction the corrections are :—

Hydrocarbon formed.	Correction, calories.
Methane	1413
Ethane	1093
Propane	1070
Butanes	1090
Butenes	1010

The gases have been assumed to obey the ideal gas laws, but the replacement of partial pressures by fugacities makes no significant alteration at the low pressures concerned.

Tables of Data Employed.

TABLE I.
Statistical and Spectroscopic Data.

Gas.	θ .	Degree of freedom.	Reference.
Carbon monoxide	3080	1	16
Hydrogen	6017	1	17
Carbon dioxide	956	2	18
	1914	1	
	3367	1	
	2286	1	
Water vapour	5158	1	19
	5382	1	
	1870	3	
Methane	2182	2	20
	4182	1	
	4311	3	

TABLE II.

Molecular Heat at Constant Pressure, 1 atm.
 Constants for equation $C_p = a + bT + cT^2 + dT^3$.

Gas.	a.	b × 10 ³ .	c × 10 ⁶ .	d × 10 ⁹ .	Refs. for expt. values used.
Carbon monoxide . . .	6.936	- 0.01	- 0.08	+1.625	—
Hydrogen * . . .	6.956	+ 0.07	—	—	—
Carbon dioxide . . .	5.79	+11.0	- 2.0	—	—
Water vapour † . . .	9.197	- 5.3	+ 6.36	+3.0	—
Methane † . . .	4.50	+13.3	—	—	—
Ethane . . .	3.80	+30.0	—	—	21
Propane . . .	8.512	+31.375	—	—	22
Butane (n) . . .	10.89	+40.0	+ 2.0	—	23
Butane (iso) . . .	11.95	+30.0	+20.0	—	23
Ethylene . . .	3.9	+22.0	—	—	21
Propylene . . .	3.972	+37.23	—	—	23
Butene (1) . . .	4.61	+51.3	—	—	23
Butene (iso) assumed .	4.61	+51.3	—	—	—

* In calculating the molecular heat of hydrogen the correcting term $0.000,08T$ was added.²⁴

† Wilson²⁵ has shown that correction should be made here for distortion of the molecule by adding $2\rho RT$. For water $\rho = 2.04 \times 10^{-5}$ and for methane $\rho = 1.72 \times 10^{-5}$. The corrections have been applied.

TABLE III.

Standard Molecular Entropy at 298.1° K.

Gas.	$S_{298.1}^{\circ}$ obs.	Refs.	$S_{298.1}^{\circ}$ calc.	Refs.	$S_{298.1}^{\circ}$ adopted.
Carbon monoxide	46.22 E.U.	7	45.92 E.U. 47.30 *	—	46.07 E.U.
Hydrogen . . .	29.69	10	31.23 29.59 †	11 12	31.23
Carbon dioxide . . .	51.11	26	50.83 51.07 ‡	— 27	51.00
Water vapour . . .	45.10 44.28	9 8	45.09	—	45.10
Methane § . . .	43.95 43.39 44.60 44.34	28 29 30	44.46 44.64 44.35	31 32 33	44.48
Ethane . . .	54.85	34	—	—	54.85
Propane . . .	64.70	35	—	—	64.70
Butane (n) . . .	75.80	36	—	—	75.80
Butane (iso) . . .	70.00		70.00		
Ethylene . . .	52.48	37	—	—	52.48
Propylene . . .	63.10	38	—	—	63.10
Butene (1) . . .	72.50	39	—	—	72.50
Butene (iso) . . .	69.00		69.00		

* Assuming molecule asymmetric in solid.

† Corrected for equilibrium mixture *ortho* and *para* forms.

‡ Different moment of inertia taken. More accurate method of calculation employed.

§ The first two experimental values quoted contained integration of Eastman's equation for molecular heat of methane. This equation is not accurate for the temperature range. Correction of second value gives the fourth value.

TABLE IV.

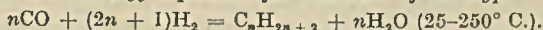
Heat of Combustion Employed, 298.1° K., 1 atmos.

Gas.	Heat of combustion, g. cal. per g. mol	Refs.
Carbon monoxide	67,636	40
Hydrogen (H ₂ O liq.)	68,318	41
Hydrogen (H ₂ O vap.)	57,877 *	42
Methane (H ₂ O liq.)	212,790	43
Ethane (H ₂ O liq.)	372,810	44
Propane (H ₂ O liq.)	530,570	45
Butane (<i>n</i>) (H ₂ O liq.)	687,940	44
Butane (<i>iso</i>) (H ₂ O liq.)	686,310	45
Ethylene (H ₂ O liq.)	337,280	46
Propylene (H ₂ O liq.)	491,820	46
Butene (1) (H ₂ O liq.)	649,660	46
Butene (<i>iso</i>) (H ₂ O liq.)	647,200	41

* From latent heat of vaporization.

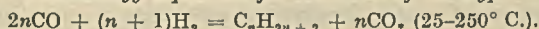
The latent heat of vaporization of water at 298.1° K. has been taken as 10,441 cal. per mol.⁴²

TABLE V.

Free Energy Equations for Reactions of the TypeGeneral equation $\Delta G_T = \Delta H_0 + AT \ln T + BT^2 + CT^3 + DT^4 \dots + IT$.

Hydrocarbon formed.	ΔH_0 $\times 10^{-3}$.	A.	B $\times 10^3$.	C $\times 10^6$.	D $\times 10^{10}$.	I.
Methane	- 45.560	14.107	- 3.90	-1.08	-1.2	-41.680
Ethane	- 76.249	26.458	- 9.54	-2.15	-2.3	-67.690
Propane	-110.13	33.397	- 7.51	-3.22	-3.4	-61.486
Butane (<i>n</i>)	-144.05	42.671	- 9.11	-4.63	-4.6	-69.913
Butane (<i>iso</i>)	-145.708	41.610	- 4.11	-7.63	-4.6	-59.081
Ethylene	- 45.212	19.402	- 5.57	-2.15	-2.3	-51.643
Propylene	- 81.547	30.981	-10.47	-3.22	-3.4	-73.165
Butene (1)	-114.702	41.994	-14.79	-4.30	-4.6	-90.010
Butene (<i>iso</i>)	-117.162	41.994	-14.79	-4.30	-4.6	-86.510

TABLE VI.

Free Energy Equations for Reactions of the Type

Hydrocarbon formed.	ΔH_0 $\times 10^{-3}$.	A.	B $\times 10^3$.	C $\times 10^6$.	D $\times 10^{10}$.	I.
Methane	- 54.955	17.494	-12.09	+0.32	+ 2.7	- 50.950
Butane (<i>n</i>)	-181.651	56.218	-41.87	+0.90	+11.0	-106.89

Relative Values of Terms Involving B, C and D.

For the equations in Tables V and VI terms in D contribute less than 0.1 per cent. to the total free energy. D and C terms together contribute less than 2 per cent. D, B and C together contribute less than 10 per cent.

TABLE VII.

Molecular Free Energy of Formation of Gaseous Hydrocarbons from Carbon Monoxide and Hydrogen.

A. Reaction in which water vapour is formed.

B. Reaction in which carbon dioxide is formed.

Hydrocarbon.	Reaction.	$G_{298.1}^{\circ}$, g.-cal.	$G_{473.1}^{\circ}$, g.-cal.	$G_{473.1}^{\circ,*}$ g.-cal.
Methane	A	- 34,401	-25,165	-25,165
Methane	B	- 41,495	-30,738	-15,369
Ethane	A	- 52,395	-33,540	-16,770
Propane	A	- 72,493	-43,938	-14,646
Butane (n)	A	- 93,355	-55,338	-13,834
Butane (n)	B	-121,730	-77,682	- 9,710
Butane (iso)	A	- 93,254	-54,373	-13,593
Ethylene	A	- 28,209	-14,595	- 7,298
Propylene	A	- 51,756	-28,582	- 9,527
Butene (1)	A	- 71,641	-38,699	- 9,675
Butene (iso)	A	- 73,059	-39,505	- 9,876

* Per g.mol. CO converted.

Discussion of Free Energy of Reaction.

The high negative values of the molecular-free energy of the reactions and the slope of the free energy *vs* temperature curve show that the lower limit of temperature will be determined by the rate of reaction. A more active form of catalyst would allow of lower working temperatures.

Formation of Carbon Dioxide.

The reaction in which carbon dioxide is formed has a higher negative free energy than the reaction producing water. It would therefore be expected to accompany the reaction in which water is formed. In the normal working of the synthetic process with nickel and cobalt catalysts the production of carbon dioxide is suppressed, but it can be initiated and allowed to replace the water-forming reaction by raising the temperature five to ten degrees above the optimum point. In the intermediate stages both carbon dioxide and water are produced, and methane appears in amount about equivalent to the carbon dioxide formed. With the entire suppression of water formation, methane is practically the only hydrocarbon to be made.

The elimination of the carbon dioxide reaction cannot be accounted for by a low rate of reaction. Aicher, Myddleton and Walker¹⁵ have shown that on a nickel catalyst at about 180° C. the rate of conversion of carbon monoxide, present in water gas, into higher hydrocarbons and water is in the region of 20 normal litres per litre of catalyst space per hour.

A quantity of the same catalyst in an aluminium tube controlled at 180° C. by an electrically heated and controlled aluminium block produced methane and carbon dioxide from the time blue water-gas was admitted for an observed period of ten days. The flow of gas was maintained at the rate of approximately 100 litres per hour for a catalyst volume of 200 c.c. The inlet and outlet gases averaged:—

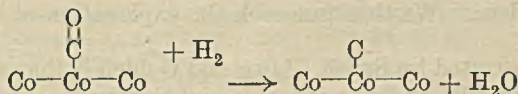
	Inlet, vol.-%.	Outlet, vol.-%.	From 100 vols. inlet.	
			Consumed, vols.	Produced, vols.
CO ₂	2.9	36.5	—	18.9
Olefines	nil	0.2	—	—
CO	40.8	1.3	40.0	—
CH ₄	2.4	36.7	—	19.5
H ₂	48.4	15.4	39.2	—
N ₂ (diff.)	5.5	9.9	—	—
	100.0	100.0		

The contraction in volume was 40.3 per cent.

One litre of catalyst space thus converted 200 litres of carbon monoxide per hour into methane and carbon dioxide.

The catalyst was found to contain about 0.5 per cent. free carbon after the reaction.

These facts point to different mechanisms in the two types of reaction. A similar conclusion was reached by Craxford⁴⁷ from a study of the rates of formation and reduction of cobalt carbide, assumed to be formed as an intermediate during synthesis with cobalt catalysts, by reaction of the metal with carbon monoxide and hydrogen.



The formation of higher hydrocarbons and water is accounted for by reaction between the carbide and molecular hydrogen. The formation of methane is assumed to be due to reaction of the carbide with chemisorbed hydrogen, while carbon dioxide is produced by the process $\text{CO} + \text{H}_2\text{O} = \text{CO}_2 + \text{H}_2$. When higher hydrocarbons and water are formed, long-chain molecules are built up from methylene groups, the immediate reduction products of the carbide, and are broken down into lower-molecular-weight hydrocarbons by chemisorbed hydrogen.

On this basis the behaviour of iron catalysts is anomalous in many ways. When they form higher hydrocarbons they produce carbon dioxide simultaneously to the exclusion of water.⁴⁸ Furthermore, the hydrocarbons have a much higher average molecular weight than those formed on nickel and cobalt catalysts.

It appears possible to account for the behaviour of all three catalysts by assuming that when carbon monoxide is adsorbed by dual attachment at adjacent atoms on the catalytic surface, water is formed by subsequent reaction with molecular hydrogen. Methylene groups are formed at the same time, and give rise to hydrocarbon chains. When carbon monoxide is adsorbed by single attachment, molecular hydrogen produces methylene groups and carbon dioxide. The methylene groups may be converted into methane by the action of chemisorbed hydrogen.

The difference in spacing of the metallic atoms in iron catalysts may

account for absence of dual attachment of carbon monoxide in this case. Experimental evidence is being sought.

The view has been expressed by H. S. Taylor⁴⁹ that at 270° C. carbon monoxide attaches itself to nickel catalysts in the form of Ni₃C and NiO. For the purpose of the present discussion it is not necessary to assume that the bond energy between carbon and nickel is sufficiently high to give rise to carbide. The carbon would be more reactive if this were not the case.

Formation of Ethylene.

On purely thermodynamical grounds we should expect much more than the traces of ethylene normally produced. Typical figures obtained during the treatment of 3000 cu. ft. of blue water-gas per hour at approximately 200° C. are:—

	Carbon converted to ethylene	---	trace
Ratio	$\frac{\text{Carbon to propylene}}{\text{Carbon to propane}}$	1.3	
Ratio	$\frac{\text{Carbon to butene}}{\text{Carbon to butane}}$	2.2	

The free energy values point to a decline in the ratio from C₄ to C₂ with an appreciable formation of ethylene. The difference in the rates of hydrogenation of propylene and ethylene is insufficient to explain the absence of ethylene. We therefore seek the explanation at the surface of the catalyst.

It was demonstrated by Smith, Hawk and Golden⁵⁰ that when ethylene is admitted along with carbon monoxide and hydrogen, a substantial part of it is used up in the formation of higher hydrocarbons. Twigg and Rideal⁵¹ showed that ethylene is adsorbed on the surface of a nickel catalyst by dual attachment on adjacent nickel atoms in the 110 and 111 planes of the face-centred cubic crystals. Herington therefore accounts for the incorporation of ethylene in the synthesis of higher hydrocarbons by assuming that synthesis normally occurs on the same planes.⁵²

Formation of Branched-chain Hydrocarbons.

The free energy relationships point to the formation of appreciable quantities of these hydrocarbons. Schaarschmidt and Marder⁵³ found 33 per cent. *iso*-paraffins in the paraffinic constituents of the synthetic gasoline fraction, and Koch and Hilberath⁵⁴ reported analyses showing presence of such compounds as 2-methyl butane, 2-methyl pentane, 3-methyl hexane, 3-methyl butene-1, 3-methyl pentene-1, 4-methyl pentene-1, 4-methyl hexene-1, and 5-methyl hexene-1. Little work has been carried out on the C₄ fraction.

As a preliminary to a systematic investigation of the C₄ fraction, samples were obtained by fractionation in a Podbielniak column. The light hydrocarbon fraction from which it was derived came from the treatment of blue water-gas on a cobalt catalyst. A C₄ sample was treated with 63 per cent. sulphuric acid according to the method of Dobryanskii.⁵⁵ The absorption, assumed due to *isobutylene*, was 14 and 16 per cent. in two

cases. According to the method of Frey and Yant,⁵⁶ in which absorption in a mixture of 2 vols. H_2SO_4 (sp. gr. 1.84) and 1 vol. water is measured, 16 and 19 per cent. *isobutylene* was found.

The possibility of formation of aromatic hydrocarbons during synthesis may be considered to require an orientation of molecules at the surface of the catalyst which is ruled out by the conditions prevailing, particularly when dual attachment of the carbon monoxide molecule is involved in the mechanism of synthesis. No such special orientation is required for the production of branched-chain hydrocarbons of the paraffin and olefine series.

In view of the importance of *isobutane* and *isobutene* as material for the production of motor spirit of high octane number and as material for chemical transformations for other purposes, a systematic investigation is planned to determine whether by modification of catalyst or in mode of operation the yield of these substances can be increased.

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FRACTIONAL DISTILLATION OF BINARY MIXTURES. SIMPLIFIED COMPUTATION OF THEORETICAL PLATES AND TRANSFER UNITS.

By A. J. V. UNDERWOOD, D.Sc., M.I.Chem.E., A.M.I.Mech.E.,
F.R.I.C., F.Inst.F., F.Inst.Pet.

SUMMARY.

Equations, in convenient form for numerical computation, are derived for the number of theoretical plates or transfer units required for the separation of binary mixtures of normal volatility. When the top or bottom product contains only a small amount of one of the components, computation can be simplified by using certain approximate equations which give satisfactory accuracy. By a simple transformation a calculation for a stripping column can be converted into one for a rectifying column.

In a previous paper¹ equations were derived for the number of transfer units required for the fractionation of a binary mixture, in which the components follow Raoult's law and the relative volatility is constant. It was also shown there that these equations, as well as a modification of the equation published by Smoker² for the number of theoretical plates, can be used to convert any separation with finite reflux into an equivalent separation with total reflux. Computation of a numerical example is however rather laborious when using these equations either for transfer units or theoretical plates. This is particularly the case for separations in which the top or bottom product contains only a small amount of one of the components. It will be shown that equations can be derived which offer considerable advantage in these respects.

NUMBER OF THEORETICAL PLATES

By making use of a property of the rectangular hyperbole, an equation for the number of theoretical plates can be derived quite simply. In Fig. 1 is shown part of a rectangular hyperbola, $y = \frac{Ax}{B+x}$, which passes through the origin 0 and has asymptotes $x = -B$ and $y = A$ parallel to the axes of co-ordinates. A line through the origin, $y = mx$, intersects the hyperbola again at L . A line parallel to OX intersects the hyperbola at P and the line OL at Q . The co-ordinates of P are (x_1, y_1) and those of Q are (x_0, y_0) .

Then
$$mx_0 = y_1 = \frac{Ax_1}{B+x_1} \dots \dots \dots (1)$$

If the abscissa of L is l , then

$$ml = \frac{Al}{B+l} \dots \dots \dots (2)$$

Dividing equation (1) by equation (2),

$$\frac{x_0}{l} = \frac{x_1(B+l)}{l(B+x_1)}$$

and

$$\frac{x_0}{l-x_0} = \frac{x_1(B+l)}{B(l-x_1)}$$

and

$$\frac{\frac{x_0}{l-x_0}}{\frac{x_1}{l-x_1}} = \frac{B+l}{B} \dots \dots \dots (3)$$

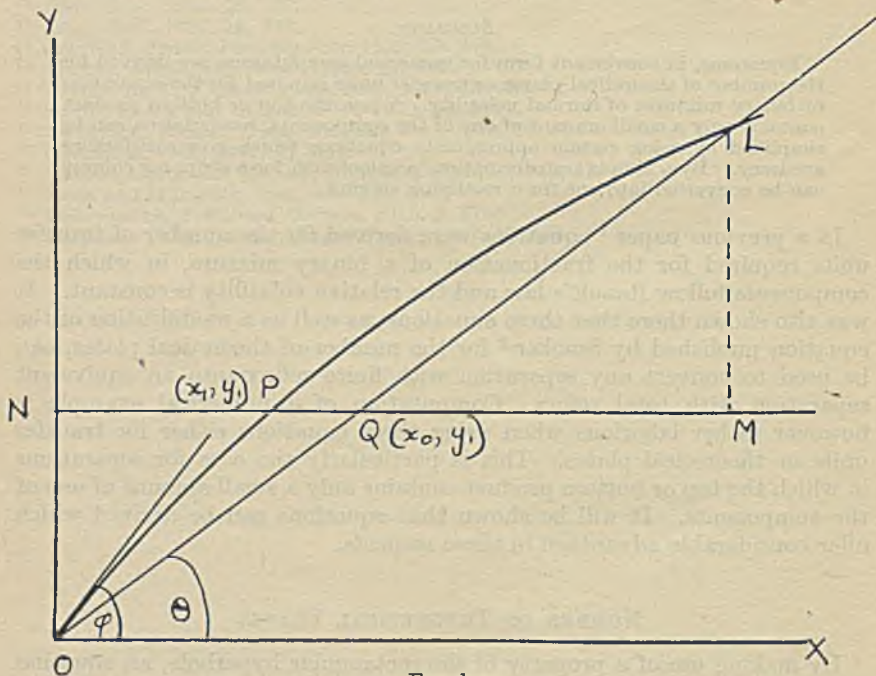


FIG. 1.

This ratio is therefore constant for any pair of points such as *P* and *Q* determined by any horizontal line such as *NM*. From equation (2),

$$\frac{B+l}{B} = \frac{A}{Bm}$$

The slope of the tangent at a point on the hyperbola is given by

$$\frac{dy}{dx} = \frac{AB}{(B+x)^2} \quad \text{and} \quad \frac{dy}{dx} = \frac{A}{B}$$

when $x = 0$. The constant $\frac{A}{Bm}$ is therefore equal to $\frac{\tan \phi}{\tan \theta}$, where ϕ is the angle between the tangent to the hyperbola at O and the x axis and θ is the angle between the line OL and the x axis.

This is also evident from the following derivation :

$$\frac{\frac{x_0}{l-x_0}}{\frac{x_1}{l-x_1}} = \frac{y_1}{x_1} \cdot \frac{(l-x_1)}{(l-x_0)}$$

As the line NM approaches the x axis and the points P and Q approach O , x_1 and x_0 become small and $\frac{l-x_1}{l-x_0}$ becomes unity in the limit. $\frac{y_1}{x_1}$ is the tangent of the angle between the line OP and the x axis. When P and O coincide, the line OP becomes the tangent to the hyperbola at O . $\frac{y_1}{x_0}$ is the tangent of the angle between OL and OX . As the ratio

$$\frac{\frac{x_0}{l-x_0}}{\frac{x_1}{l-x_1}}$$

is constant for any two points such as P and Q , it is equal to the value when P and Q coincide with O —i.e., $\frac{\tan \phi}{\tan \theta}$.

Equation (3) may therefore be written :

$$\frac{\frac{x_0}{l-x_0}}{\frac{x_1}{l-x_1}} = \frac{NQ}{PM} = \frac{QM}{NP} = \frac{\tan \phi}{\tan \theta} \dots \dots \dots (4)$$

Fig. 2 shows the usual equilibrium curve $y = \frac{\alpha x}{1 + (\alpha - 1)x}$ (which is a rectangular hyperbola with asymptotes parallel to the axes) and an operating line $y = mx + b$ cutting the equilibrium curve at O' and L , the abscissæ of which are k_1 and k_2 respectively. P_0 and P are points representing the compositions on two adjacent plates with abscissæ x_0 and x_1 respectively. All co-ordinates now refer to the usual origin O .

It has been shown that

$$\frac{\frac{NQ}{QM}}{\frac{NP}{PM}} = \frac{\tan \phi}{\tan \theta}$$

$$\frac{x_0 - k_1}{k_2 - x_0} = \frac{\tan \phi}{\tan \theta}$$

i.e.,

ϕ is the angle between the tangent to the equilibrium curve at O' and the x axis. Now

$$\frac{dy}{dx} = \frac{\alpha}{\{1 + (\alpha - 1)x\}^2}$$

and for $x = k_1$,

$$\tan \phi = \frac{\alpha}{\{1 + (\alpha - 1)k_1\}^2} = \frac{\alpha}{c^2},$$

where $c = 1 + (\alpha - 1)k_1$.

Also $\tan \theta = m$.

$$\frac{x_0 - k_1}{k_2 - x_0} = \frac{\alpha}{mc^2} \dots \dots \dots (4a)$$

$$\frac{x_1 - k_1}{k_2 - x_1}$$

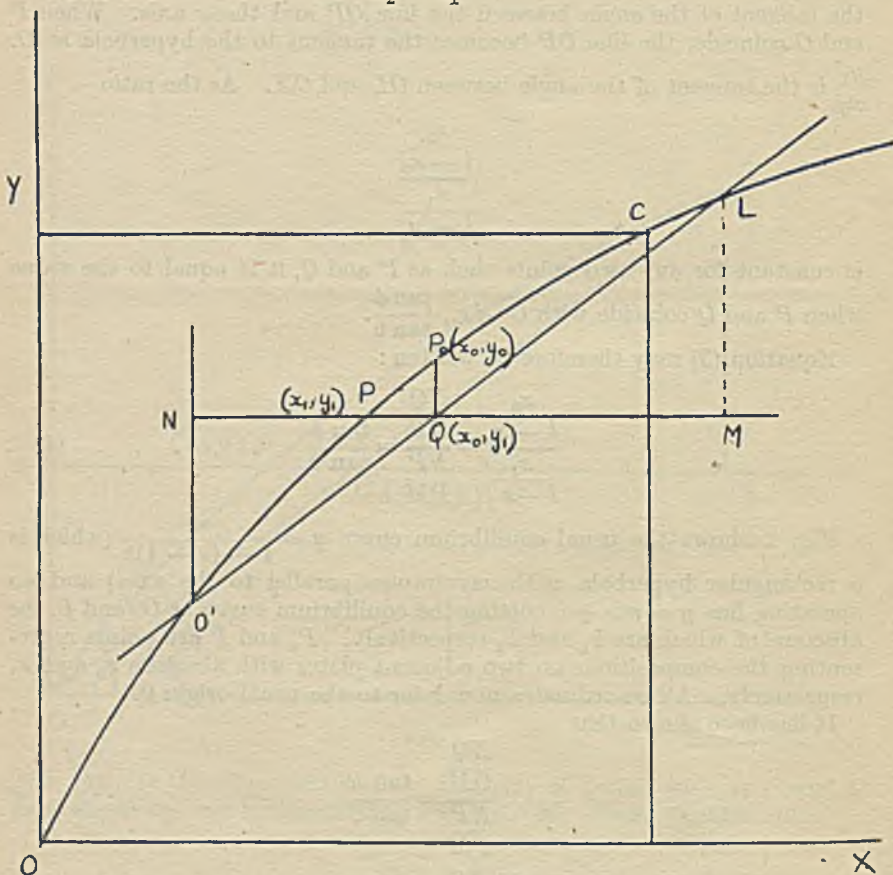


FIG. 2.

If $x_0, x_1, x_2 \dots x_n$ represent compositions on successive plates in the column, then similarly

$$\frac{x_1 - k_1}{k_2 - x_1} = \frac{x_2 - k_1}{k_2 - x_2} = \dots = \frac{\alpha}{mc^2}$$

Therefore, by successive multiplication :

$$\frac{\frac{x_0 - k_1}{k_2 - x_0}}{\frac{x_n - k_1}{k_2 - x_n}} = \left(\frac{\alpha}{mc^2}\right)^n$$

or
$$\left(\frac{\alpha}{mc^2}\right)^n = \frac{(x_0 - k_1)(k_2 - x_n)}{(x_n - k_1)(k_2 - x_0)} \dots \dots \dots (5)$$

which gives the number of theoretical plates required to pass from x_0 to x_n . For total reflux, $k_1 = 0, k_2 = 1, c = 1$ and $m = 1$ and equation (5) reduces to the well-known form. n becomes infinite when $x_n = k_1$ or $x_0 = k_2$, representing the intersection of the operating line with the equilibrium curve for a rectifying column or a stripping column respectively.

In the previous paper ¹ the equation derived by Smoker ² was converted into a form which may be written :

$$\left(\frac{\alpha}{mc^2}\right) = \frac{x_0''(1 - x_n'')}{x_n''(1 - x_0'')} \dots \dots \dots (6)$$

where
$$x_0'' = \frac{x_0 - k_1}{k_2 - k_1} \quad \text{and} \quad x_n'' = \frac{x_n - k_1}{k_2 - k_1}$$

Substituting these values in equation (6) then gives equation (5), which may also be written :

$$n \cdot \log \frac{\alpha}{mc^2} = \log \left\{ \frac{(x_0 - k_1)(k_2 - x_n)}{(x_n - k_1)(k_2 - x_0)} \right\} \dots \dots \dots (7)$$

This is a convenient form for computation. k_1 and k_2 must first be found either graphically by drawing the operating line and noting its intersections with the equilibrium curve, or analytically by solving for the abscissæ of the points of intersection from the equation :

$$mk + b = \frac{\alpha k}{1 + (\alpha - 1)k}$$

or
$$m(\alpha - 1)k^2 + \{m + b(\alpha - 1) - \alpha\}k + b = 0 \dots \dots (8)$$

which is the equation originally given by Smoker.²

It should be noted that k_1 is the lesser root of equation (8) and k_2 is the greater root. An operating line for a rectifying column will give a value of k_1 between 0 and 1, while k_2 will be greater than 1. For a stripping column k_1 will be negative and k_2 will lie between 0 and 1. It should be noted that this convention is different from that used by Smoker, who does not use the root k_2 , and denotes as k that root of equation (8) which lies between 0 and 1 for both a rectifying and a stripping column.

There is another simple proof of the relation given by equation (4a), which can be written in the form :

$$\frac{(x_0 - k_1)(k_2 - x_1)}{(x_1 - k_1)(k_2 - x_0)} = d,$$

where d is a constant. The relation between x_0 and x_1 , as derived from the equilibrium curve and the operating line is :

$$mx_0 + b = \frac{\alpha x_1}{1 + (\alpha - 1)x_1}$$

The constants k_1 , k_2 and d can be chosen so that the two equations are identical. The two equations can be written :

$$\begin{aligned} x_1 x_0 (d - 1) + x_1 (k_1 - dk_2) + x_0 (k_2 - dk_1) + k_1 k_2 (d - 1) &= 0 \\ \text{and} \quad x_1 x_0 m (\alpha - 1) + x_1 \{b(\alpha - 1) - \alpha\} + mx_0 + b &= 0. \end{aligned}$$

The two are identical if

$$\frac{d - 1}{m(\alpha - 1)} = \frac{k_1 - dk_2}{b(\alpha - 1) - \alpha} = \frac{k_2 - dk_1}{m} = \frac{k_1 k_2 (d - 1)}{b}$$

These relations give three equations for determining k_1 , k_2 , and d . From the first, second, and third relations it is seen that :

$$\frac{d - 1}{m(\alpha - 1)} = \frac{(k_1 + k_2)(1 - d)}{m + b(\alpha - 1) - \alpha}$$

so that

$$-(k_1 + k_2) = \frac{m + b(\alpha - 1) - \alpha}{m(\alpha - 1)}$$

Also

$$k_1 k_2 = \frac{b}{m(\alpha - 1)}$$

Consequently k_1 and k_2 are the roots of equation (8).

Also

$$(d - 1) = (\alpha - 1)(k_2 - dk_1)$$

\therefore

$$d = \frac{1 + (\alpha - 1)k_2}{1 + (\alpha - 1)k_1}$$

Now

$$\begin{aligned} \{1 + (\alpha - 1)k_1\}\{1 + (\alpha - 1)k_2\} &= 1 + (\alpha - 1)(k_1 + k_2) + (\alpha - 1)^2 k_1 k_2 \\ &= 1 - \frac{\{m + b(\alpha - 1) - \alpha\}}{m} + \frac{b(\alpha - 1)}{m} \\ &= \frac{\alpha}{m} \end{aligned}$$

Thus

$$d = \frac{\alpha}{m\{1 + (\alpha - 1)k_1\}^2} = \frac{\alpha}{mc^2}$$

Although equation (5) or (7) is simple in form, its application to separations where the top or bottom product contains only a small amount of one of the components necessitates a high degree of accuracy in computation. Usually, for a rectifying column, $x_0 = x_D$ and $x_n = x_F$, and for a stripping column, $x_0 = x_D$ and $x_n = x_W$. When the top product contains only a small amount of the heavier component, the point L is close to the point C in Fig. 2 and k_2 is very nearly equal to 1. Also x_0 is very nearly equal to 1, so that the term $(k_2 - x_0)$ in equation (7) is the difference of two nearly equal quantities, which must therefore be computed with a high

degree of accuracy to avoid excessive error in the difference. Similarly, for a stripping column producing a bottom product containing a small amount of the lighter component, the point O' will be close to O . k_1 will be small (and negative), and x_n will also be small, so that the term $(x_n - k_1)$ will be small. Also for reflux ratios close to the minimum reflux, it will be clear that, for a rectifying column, the term $(x_n - k_1)$ may be small, while for a stripping column the term $(k_2 - x_0)$ may be small. Exactly the same practical difficulty in computation arises if use is made of equation (6) or of the equation originally given by Smoker², namely :

$$\left(\frac{\alpha}{mc^2}\right)^n = \frac{x_0' \left\{ 1 - \frac{mc(\alpha - 1) \cdot x_n'}{\alpha - mc^2} \right\}}{x_n' \left\{ 1 - \frac{mc(\alpha - 1) \cdot x_0'}{\alpha - mc^2} \right\}} \quad (9)$$

where $x' = x - k_1$.

The difficulty also arises when a graphical method such as that of McCabe and Thiele³ is used, and necessitates drawing on a large scale those parts of the diagram where the points representing plates are close together. The nomograph published by Bisesi⁴ will also give insufficiently accurate results unless constructed on a large scale.

For example, when a mixture of two components with relative volatility 2.5 is fractionated with a reflux ratio of 3 to give a top product of composition $x_D = 0.99$, the value of k_2 is 1.0071 and the difference $k_2 - x_D = k_2 - x_n = 0.0171$. For three significant figures in the difference, five significant figures are required for the value of k_2 .

The particular cases of a top product containing only a small amount of heavy component and a bottom product containing only a small amount of light component are of frequent occurrence in practical problems. For these cases simplified approximate equations can be derived which give results of satisfactory accuracy with less laborious computation than the exact equations (7) or (9). Thomson and Beatty⁵ have made a careful study of various methods of calculating the number of theoretical plates for such conditions. They have shown that, by assuming the equilibrium curve to be linear near the ends of the x, y diagram, reasonably accurate results are obtained so long as the calculation is limited to moderate concentration ranges in these regions. For other cases, where there is a wide range between the feed concentration and the top or bottom product concentration, they do not recommend the use of that method. The simplified equations derived in the following are applicable to these conditions.

NUMBER OF THEORETICAL PLATES—RECTIFYING COLUMN.

Put $x_0 = x_D = 1 - \delta$, where δ is a small quantity.

Now
$$m = \frac{R}{R + 1} \quad \text{and} \quad b = \frac{x_D}{R + 1} = (1 - m)(1 - \delta).$$

Substituting this value of b in equation (8) gives :

$$m(\alpha - 1)k^2 + \{2m - m\alpha - 1 - (\alpha - 1)(1 - m)\delta\}k + (1 - m)(1 - \delta) = 0 \quad (10)$$

Solving this equation, neglecting powers of δ higher than the first, gives :

$$k_2 = 1 + \frac{\alpha(1-m)\delta}{m\alpha-1} \quad . \quad . \quad . \quad (11)$$

and

$$k_1 = \frac{1-m}{m(\alpha-1)} - \frac{(1-m)\delta}{m(\alpha-1)} = \frac{1-m}{m(\alpha-1)} \left\{ 1 - \frac{(\alpha-1)\delta}{m\alpha-1} \right\} \quad (12)$$

Also
$$k_2 - x_0 = k_2 - 1 + \delta = \frac{(\alpha-1)\delta}{m\alpha-1} \quad . \quad . \quad . \quad (13)$$

which gives the small term $(k_2 - x_0)$ in equation (7) with satisfactory accuracy by a slide-rule calculation.

Equation (10) can also be readily solved by noting that $k = 1$ is an approximate solution, since it is satisfied by $k = 1$ when $\delta = 0$. A second approximation to a solution is obtained by putting $k = 1 + \psi$, where ψ is small, and neglecting small quantities of the second order, which gives :

$$\psi = \frac{\alpha(1-m)\delta}{m\alpha-1} \text{ and } k_2 = 1 + \frac{\alpha(1-m)\delta}{m\alpha-1} \text{ as before.}$$

Also, since k_1 and k_2 are the roots of equation (10),

$$k_1 k_2 = \frac{(1-m)(1-\delta)}{m(\alpha-1)}$$

and
$$k_1 = \frac{(1-m)(1-\delta-\psi)}{m(\alpha-1)} = \frac{1-m}{m(\alpha-1)} \left\{ 1 - \frac{(\alpha-1)\delta}{m\alpha-1} \right\} \text{ as before.}$$

Of the four terms on the right-hand side of equation (7), $(k_2 - x_0)$ or $(k_2 - x_D)$ is given by equation (13). Also $x_n - k_1 = x_F - k_1$ (where x_F is the feed composition) and is obtained from equation (12). If the feed is not composed of liquid at its boiling point, x_F should really be replaced by

$$\frac{(R+1)x_F + (q-1)x_D}{R+q},$$

where q is the heat required to vaporize 1 mol of feed divided by the molal heat of vaporization. In many cases this will be an unnecessary refinement unless q is quite small.

The term $(x_0 - k_1)$ is equal to $(x_D - k_1)$ and the term $(k_2 - x_n)$ is equal to $(k_2 - x_F)$.

It may be noted that

$$x_D - k_1 = \frac{1-m}{m(\alpha-1)} - \delta - k_1 = \frac{m\alpha-1}{m(\alpha-1)} - \frac{(m^2\alpha-1)\delta}{m(\alpha-1)} \quad . \quad . \quad (14)$$

In many cases no substantial error will be involved if the second term containing δ is neglected.

Also
$$k_2 - x_F = 1 - x_F + \frac{\alpha(1-m)\delta}{m\alpha-1} \quad . \quad . \quad . \quad (15)$$

and in many cases the term involving δ can be neglected.

An approximate value of $\frac{\alpha}{mc^2}$ can be obtained to facilitate calculation.

$$\frac{\alpha}{mc^2} = \frac{\alpha}{m\{1 + (\alpha - 1)k_1\}^2} = m\alpha \left\{ 1 + \frac{2(1 - m)(\alpha - 1)\delta}{m\alpha - 1} \right\} \quad (16)$$

Here again the term involving δ will be negligible in many cases. It will be found that the use of equations (11) to (16) greatly facilitates numerical computation and avoids the use of five-figure logarithms which are generally necessary when using equations (6), (7), or (9).

From the value of $(k_2 - x_0)$ given by equation (13), it will be seen by substituting it in equation (7) that n is approximately a linear function of $\log \delta$ when m and α are constant.

A comparison of the results given by the exact and the approximate methods is shown in Table I for various reflux ratios and top-product compositions, with $\alpha = 2.5$ and $x_F = 0.4$. It will be seen that the agreement is very satisfactory.

A less accurate equation can be obtained by neglecting the terms involving δ in equations (12), (14), (15), and (16), giving, by substitution in equation (7):

$$n \cdot \log m\alpha = \log \left[\frac{(m\alpha - 1)^2(1 - x_F)}{m(\alpha - 1)^2 \left\{ x_F - \frac{(1 - m)}{m(\alpha - 1)} \right\} \delta} \right] \quad (17)$$

This equation will give substantial errors, of the order of perhaps 20 per cent., when used for reflux ratios near the minimum, mainly due to the error in the term $(x_F - k_1)$. It may, however, be useful for certain rough calculations.

NUMBER OF THEORETICAL PLATES—STRIPPING COLUMN.

In this case x_W is small and will be denoted by ϵ .

Since
$$m = \frac{RP + qF}{RP + qF - W} \quad \text{and} \quad b = \frac{-Wx_W}{RP + qF - W}$$

$$\therefore b = (1 - m)x_W = (1 - m)\epsilon.$$

Equation (8) then becomes:

$$m(\alpha - 1)k^2 + \{m + (\alpha - 1)(1 - m)\epsilon - \alpha\}k + (1 - m)\epsilon = 0 \quad (18)$$

When $\epsilon = 0$, one root of the equation is equal to zero, and therefore k_1 is small. Neglecting small terms of the second order in equation (18) gives:

$$k_1 = \frac{(1 - m)\epsilon}{\alpha - m} \quad (19)$$

Since
$$k_1 k_2 = \frac{(1 - m)\epsilon}{m(\alpha - 1)}$$

$$\therefore k_2 = \frac{\alpha - m}{m(\alpha - 1)} \quad (20)$$

A second approximation for k_2 is obtained by putting

$$k_2 = \frac{\alpha - m}{m(\alpha - 1)} + \psi$$

where ψ is small. Substituting in equation (18) and neglecting small terms of the second order gives :

$$k_2 = \frac{\alpha - m}{m(\alpha - 1)} - \frac{\alpha(1 - m)\epsilon}{m(\alpha - m)} = \frac{\alpha - m}{m(\alpha - 1)} \left\{ 1 - \frac{\alpha(1 - m)(\alpha - 1)\epsilon}{(\alpha - m)^2} \right\} \quad (21)$$

The values of k_1 and k_2 given by equations (19) and (21) can also be obtained by solving equation (18) in the usual way and neglecting small terms of the second order.

The approximation for $\frac{\alpha}{mc^2}$ is :

$$\frac{\alpha}{mc^2} = \frac{\alpha}{m} \left\{ 1 - \frac{2(\alpha - 1)(1 - m)\epsilon}{\alpha - m} \right\} \quad \dots \quad (22)$$

Also
$$x_n - k_1 = \epsilon - k_1 = \frac{(\alpha - 1)\epsilon}{\alpha - m} \quad \dots \quad (23)$$

Equations (19), (21), (22), and (23) are then used for computation of equation (7).

A comparison of the exact and approximate methods is given in Table II. As before, $\alpha = 2.5$ and $x_F = 0.4$. The results are given for various values of S , where S is defined by the operating line for a stripping column :

$$(S + 1)x = Sy + x_W \text{ and } m = \frac{S + 1}{S}.$$

For a stripping column on which a rectifying column is superimposed :

$$S = \frac{RP + qF - W}{W} = \frac{(R + 1)x_F - (R + q)x_W - (1 - q)x_D}{x_D - x_F}$$

As previously pointed out by the author,⁶ the quantity S may be termed the "revaporization ratio." It represents the number of mols of vapour returned by the reboiler to the stripping column per mol of bottom product withdrawn. It is thus exactly analogous to the reflux ratio in a rectifying column which represents the number of mols of condensate returned by the condenser to the rectifying column per mol of top product withdrawn. As a more convenient name for the quantity S , "reboil ratio" is suggested.

It will be seen from Table II that the agreement between the exact and the approximate methods is very satisfactory.

When equation (7) is used for a rectifying column, putting $x_0 = x_D$ and $x_n = x_F$, the value of n obtained is the number of plates in the rectifying column, including the feed-plate. For a rectifying column used alone, mounted on a still pot, the pot would be included as a plate instead of the feed-plate in the value of n . When equation (7) is used for a stripping column, putting $x_0 = x_F$ and $x_n = x_W$, the value of n obtained is the number of plates in the stripping column below the feed-plate and includes the still pot counted as one plate. For the common case of a rectifying column

superimposed on a stripping column the total number of actual plates will be the sum of the two values of n less one, the still pot being excluded.

When α is not quite constant, it will usually be desirable to follow the suggestion of Thomson and Beatty⁵ and use the value of α corresponding to the region of the equilibrium curve where the plates are closest together. This will usually be at one of the ends of the equilibrium curve.

A less accurate equation for the stripping column can be derived by using the value of k_2 given by equation (20) and neglecting ϵ in equation (22) and noting that :

$$x_0 - k_1 = x_F - k_1 = x_F \text{ approximately}$$

and $k_2 - x_n = k_2 - \epsilon = k_2 \text{ approximately.}$

Then
$$n \cdot \log \frac{\alpha}{m} = \log \left[\frac{(\alpha - m)^2 x_F}{m(\alpha - 1)^2 \left\{ \frac{\alpha - m}{m(\alpha - 1)} - x_F \right\} \epsilon} \right] \dots (24)$$

This equation is subject to the same limitations that were pointed out in connection with equation (17).

NUMBER OF TRANSFER UNITS.

It has previously been shown¹ that the number of transfer units is given by the equation :

$$N_{OG} = \frac{1}{\beta - 1} \left[\log_e \frac{x_0''}{x_n''} + \beta \cdot \log_e \left\{ \frac{1 - x_n''}{1 - x_0''} \right\} \right] \dots (25)$$

where $\beta = \frac{\alpha}{mc^2}$ and $x'' = \frac{x - k_1}{k_2 - k_1}$.

Therefore

$$N_{OG} = \frac{1}{\frac{\alpha}{mc^2} - 1} \left[\log_e \left\{ \frac{x_0 - k_1}{x_n - k_1} \right\} + \frac{\alpha}{mc^2} \cdot \log_e \left\{ \frac{k_2 - x_1}{k_2 - x_1} \right\} \right] \dots (26)$$

The equations previously derived for $k_1, k_2, \text{ etc.},$ can be used for the computation of equation (26) exactly as for equation (7).

MINIMUM REFLUX RATIO AND MINIMUM NUMBER OF PLATES—RECTIFYING COLUMN.

The minimum reflux ratio, $R_0,$ is given by

$$R_0 = \frac{x_D - y_F}{y_F - x_F}$$

which reduces to

$$R_0 = \frac{1}{(\alpha - 1)x_F} - \frac{\delta\{1 + (\alpha - 1)x_F\}}{x_F(1 - x_F)(\alpha - 1)} \dots (27)$$

As a first approximation, which is sufficient for most purposes

$$R_0 = \frac{1}{(\alpha - 1)x_F} \dots (28)$$

The minimum reflux operating line with slope $m_0 = \frac{R_0}{R_0 + 1}$ intersects the equilibrium curve at $x = x_F$, which is the corresponding value of k_1 . Then, from equation (12) :

$$x_F = \frac{1 - m_0}{m_0(\alpha - 1)} - \frac{(1 - m_0)\delta}{m_0(m_0\alpha - 1)} \dots \dots \dots (29)$$

and

$$\begin{aligned} x_F - k_1 &= \frac{1 - m_0}{m_0(\alpha - 1)} - \frac{1 - m}{m(\alpha - 1)} - \delta \left\{ \frac{1 - m_0}{m_0(m_0\alpha - 1)} - \frac{1 - m}{m(m\alpha - 1)} \right\} \\ &= \frac{1 - m_0}{m_0(\alpha - 1)} - \frac{1 - m}{m(\alpha - 1)} \text{ approx. } \dots \dots \dots (30) \end{aligned}$$

From equations (11) and (29) :

$$\begin{aligned} k_2 - x_F &= \frac{m_0\alpha - 1}{m_0(\alpha - 1)} + \delta \left\{ \frac{\alpha(1 - m)}{m\alpha - 1} + \frac{1 - m_0}{m_0(m_0\alpha - 1)} \right\} \\ &= \frac{m_0\alpha - 1}{m_0(\alpha - 1)} \text{ approx. } \dots \dots \dots (31) \end{aligned}$$

Also, from equation (12) :

$$\begin{aligned} x_D - k_1 &= \frac{m\alpha - 1}{m(\alpha - 1)} - \frac{(m^2\alpha - 1)\delta}{m(m\alpha - 1)} \\ &= \frac{m\alpha - 1}{m(\alpha - 1)} \text{ approx. } \dots \dots \dots (32) \end{aligned}$$

Then, substituting from equations (30), (31), (32), and (13) in equation (5) gives :

$$\left(\frac{\alpha}{m\alpha^2} \right)^n = \frac{(m\alpha - 1)^2(m_0\alpha - 1)}{(m - m_0)(\alpha - 1)^2\delta} \dots \dots \dots (33)$$

For total reflux, the minimum number of plates, n_0 , is given by

$$\begin{aligned} \alpha^{n_0} &= \frac{x_D(1 - x_F)}{x_F(1 - x_D)} = \frac{1 - x_F}{x_F \cdot \delta} \\ \text{or } \alpha^{n_0} &= \frac{(m_0\alpha - 1)}{(1 - m_0)\delta} \dots \dots \dots (34) \end{aligned}$$

From equations (33) and (34) :

$$\frac{\left(\frac{\alpha}{m\alpha^2} \right)^n}{\alpha^{n_0}} = \frac{(m\alpha - 1)^2(1 - m_0)}{(m - m_0)(\alpha - 1)^2} \dots \dots \dots (35)$$

As an approximation from equation (16),

$$\frac{\alpha}{m\alpha^2} = m\alpha.$$

Therefore
$$\frac{(m\alpha)^n}{\alpha^{n_0}} = \frac{(m\alpha - 1)^2(1 - m_0)}{(m - m_0)(\alpha - 1)^2} \dots \dots \dots (36)$$

This equation can be used for plotting a curve showing the variation of n with m or R .

MINIMUM REBOIL RATIO AND MINIMUM NUMBER OF PLATES—
STRIPPING COLUMN.

As pointed out earlier in this paper, the slope, m_0 , of the operating line for the minimum reboil ratio, S_0 , is given by

$$m_0 = \frac{S_0 + 1}{S_0} = \frac{y_F - x_W}{x_F - x_W}$$

and

$$S_0 = \frac{x_F - x_W}{y_F - x_F}$$

$$\therefore S_0 = \frac{1 + (\alpha - 1)x_F}{(\alpha - 1)(1 - x_F)} \left(1 - \frac{\varepsilon}{x_F}\right) = \frac{1 + (\alpha - 1)x_F}{(\alpha - 1)(1 - x_F)} \text{ approx.} \quad (37)$$

Noting that x_F is the value of k_2 when $m = m_0$, the following approximate equations can be derived by the same procedure used for the rectifying column :

$$x_F = \frac{\alpha - m_0}{m_0(\alpha - 1)} - \frac{\alpha(1 - m_0)\varepsilon}{m_0(\alpha - m_0)} \quad \dots \quad (38)$$

$$x_F - k_1 = \frac{\alpha - m_0}{m_0(\alpha - 1)} \quad \dots \quad (39)$$

$$k_2 - x_F = \frac{\alpha - m}{m(\alpha - 1)} - \frac{\alpha - m_0}{m_0(\alpha - 1)} \quad \dots \quad (40)$$

$$k_2 - x_W = \frac{\alpha - m}{m(\alpha - 1)} \quad \dots \quad (41)$$

Substituting in equation (5) from these equations and equation (23) :

$$\left(\frac{\alpha}{mc^2}\right)^n = \frac{(\alpha - m_0)(\alpha - m)^2}{\alpha(\alpha - 1)^2(m_0 - m)\varepsilon} \quad \dots \quad (42)$$

For total reflux,

$$\alpha^{n_0} = \frac{x_F(1 - x_W)}{x_W(1 - x_F)} = \frac{x_F}{(1 - x_F)\varepsilon} \text{ approx.}$$

or

$$\alpha^{n_0} = \frac{\alpha - m_0}{\alpha(m_0 - 1)\varepsilon} \quad \dots \quad (43)$$

Therefore
$$\frac{\left(\frac{\alpha}{mc^2}\right)^n}{\alpha^{n_0}} = \frac{(\alpha - m)^2(m_0 - 1)}{(\alpha - 1)^2(m_0 - m)} \quad \dots \quad (44)$$

As an approximation from equation (22), $\frac{\alpha}{mc^2} = \frac{\alpha}{m}$.

Then
$$\frac{\left(\frac{\alpha}{m}\right)^n}{\alpha^{n_0}} = \frac{(\alpha - m)^2(m_0 - 1)}{(\alpha - 1)^2(m_0 - m)} \quad \dots \quad (45)$$

REFLUX RATIO AND REBOIL RATIO.

It has been pointed out that there is a close analogy between reflux ratio for a rectifying column and reboil ratio for a stripping column. It can

further be shown that, by suitable transformation, the calculation of plates for a stripping column can be made by using methods of calculation relating to a rectifying column. Fig. 3 shows an operating line for a stripping column, which cuts the equilibrium curve at D and F and the diagonal OB at E ($x = x_W$). Its inclination to the x axis is given by $m = \tan \theta = \frac{S+1}{S}$. If Fig. 3 is rotated through 180° about the diagonal

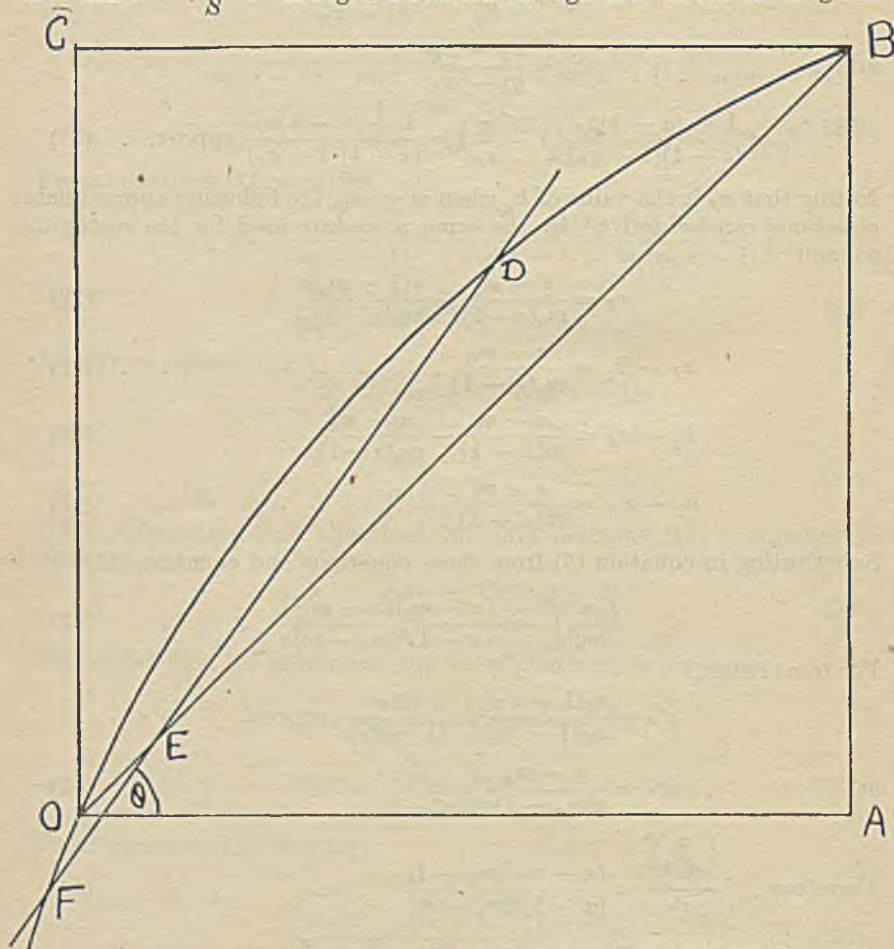


FIG. 3.

AC , the result is Fig. 4. The equilibrium curve is unchanged since it is symmetrical with respect to a line joining A and C . The stripping-column line DEF of Fig. 3 has become a rectifying-column line in Fig. 4. It now has an inclination of $(\frac{\pi}{2} - \theta)$ to the horizontal axis and its slope is

$$\tan\left(\frac{\pi}{2} - \theta\right) = \cot \theta = \frac{S}{S+1}$$

Taking B as the new origin in Fig. 4, the co-ordinates of the point E are $(1 - x_w)$. If x' and y' are the new co-ordinates in Fig. 4, the equation of the line DEF is

$$y' = \frac{S}{S+1} \cdot x' + \frac{1-x_w}{S+1} \quad \dots \quad (46)$$

This now represents an operating line for a rectifying column with a reflux ratio S and a top product $(1 - x_w)$. The calculation of plates for a stripping

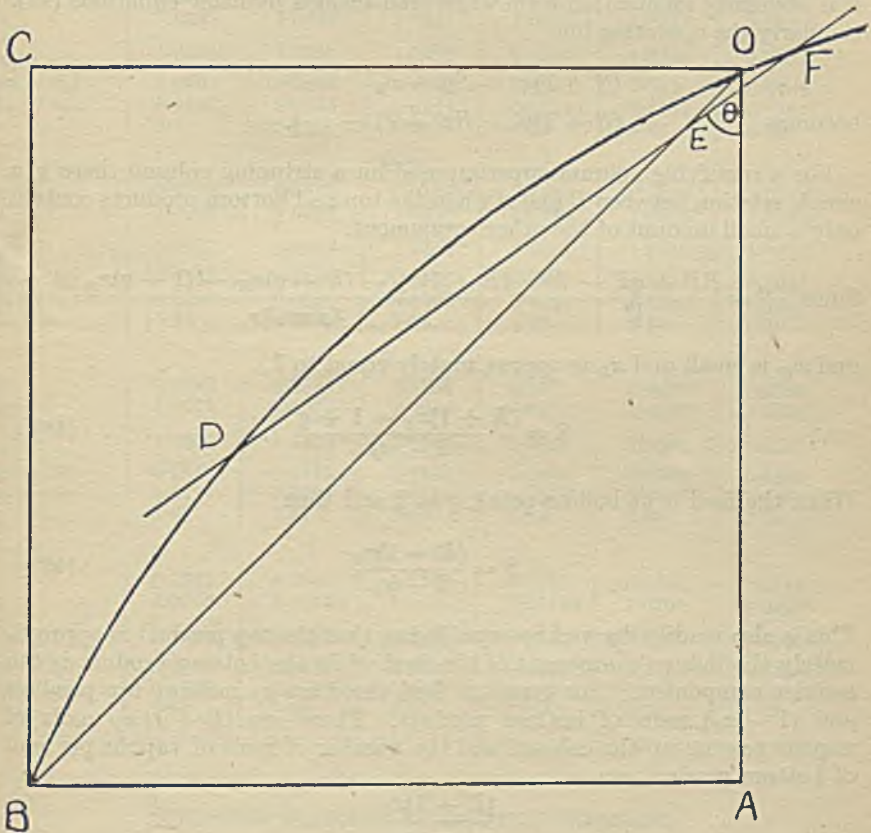


FIG. 4.

column can therefore be carried out by using the same equations as for a rectifying column, using S instead of R and $(1 - x_w)$ instead of x_D . It should be noted that if x_F is the feed composition for the stripping column in Fig. 3, the corresponding feed composition for the rectifying column in Fig. 4 is $(1 - y_F) = x'_F$.

The above results can also be demonstrated analytically. The relation between the (x, y) system of co-ordinates of Fig. 3 and the (x', y') system of co-ordinates of Fig. 4 is

$$x = 1 - y' \text{ and } y = 1 - x' \quad \dots \quad (47)$$

These equations indicate that the mol fraction of the lighter component in the liquid at any point in Fig. 3 becomes the mol fraction of the heavier component in the vapour at the corresponding point in Fig. 4. Writing the equilibrium curve in the form

$$\frac{y}{1-y} = \frac{\alpha x}{1-x}$$

it is obviously identical in form when transformed by using equations (47). Similarly the operating line

$$(S+1)x = Sy + x_W$$

becomes

$$(S+1)y' = Sx' + (1-x_W).$$

For a rectifying column superimposed on a stripping column there is a simple relation between R and S when the top and bottom products contain only a small amount of the other component.

$$\text{Since } S = \frac{RP + qF - W}{W} = \frac{(R+1)x_F - (R+q)x_W - (1-q)x_D}{x_D - x_F}$$

and x_W is small and x_D is approximately equal to 1,

$$\therefore S = \frac{(R+1)x_F - 1 + q}{1 - x_F} \quad \dots \quad (48)$$

When the feed is at boiling-point, $q = 1$ and then

$$S = \frac{(R+1)x_F}{1 - x_F} \quad \dots \quad (49)$$

This is also readily derived by considering that the top product is approximately the lighter component of the feed while the bottom product is the heavier component. For 1 mol of feed there are x_F mols of top product and $(1-x_F)$ mols of bottom product. There are $(R+1)x_F$ mols of vapour passing up the column and the number of mols of vapour per mol of bottom product are

$$\frac{(R+1)x_F}{1-x_F}$$

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RECTIFYING COLUMN—SUMMARY OF RESULTS.

	$x_D = 0.99.$		$x_D = 0.97.$		$x_D = 0.95.$	
	Appr.	Exact.	Appr.	Exact.	Appr.	Exact.
$R = 1.8.$						
k_1	0.361	0.3614	0.343	0.3447	0.324	0.3291
k_2	1.0147	1.0145	1.0441	1.0424	1.0735	1.0691
$\frac{a}{mc^2}$	1.6354	1.6353	1.6922	1.6898	1.7490	1.7431
$(x_F - k_1)$	0.039	0.0386	0.057	0.0553	0.076	0.0709
$(k_2 - x_D)$	0.0247	0.0245	0.0741	0.0724	0.1235	0.1191
n	12.20	12.23	8.70	8.75	6.85	6.89
$R = 2.$						
k_1	0.3258	0.3259	0.3108	0.3120	0.2958	0.2989
k_2	1.0125	1.0124	1.0375	1.0364	1.0625	1.0595
$\frac{a}{mc^2}$	1.6917	1.6916	1.7417	1.7401	1.7917	1.7875
$(x_F - k_1)$	0.0742	0.0741	0.0892	0.0880	0.1042	0.1011
$(k_2 - x_D)$	0.0225	0.0224	0.0675	0.0664	0.1125	0.1095
n	10.45	10.46	7.65	7.72	6.19	6.29
$R = 3.$						
k_1	0.2184	0.2184	0.2108	0.2111	0.2032	0.2040
k_2	1.0071	1.0071	1.0214	1.0211	1.0357	1.0349
$\frac{a}{mc^2}$	1.8911	1.8912	1.9232	1.9226	1.9558	1.9543
$(x_F - k_1)$	0.1816	0.1816	0.1892	0.1888	0.1968	0.1960
$(k_2 - x_D)$	0.0171	0.0171	0.0514	0.0511	0.0857	0.0849
n	7.87	7.87	5.93	5.94	4.98	5.00
$R = 4.$						
k_1	0.1642	0.1642	0.1592	0.1593	0.1542	0.1545
k_2	1.0050	1.00499	1.0150	1.01488	1.0250	1.02464
$\frac{a}{mc^2}$	2.1020	2.0119	2.0360	2.0357	2.0600	2.0595
$(x_F - k_1)$	0.2358	0.2358	0.2408	0.2407	0.2458	0.2455
$(k_2 - x_D)$	0.0150	0.01499	0.0450	0.04488	0.075	0.07464
n	7.08	7.08	5.39	5.39	4.56	4.57

STRIPPING COLUMN—SUMMARY OF RESULTS.

	$x_W = 0.01.$		$x_W = 0.03.$		$x_W = 0.05.$	
	Appr.	Exact	Appr.	Exact.	Appr.	Exact.
$S = 2.$						
k_1	-0.005	-0.00491	-0.015	-0.01423	-0.025	-0.02296
k_2	0.4527	0.4527	0.4694	0.4687	0.4861	0.4841
$\frac{a}{mc^2}$	1.6917	1.6916	1.7417	1.7402	1.7917	1.7840
$(k_2 - x_F)$	0.0527	0.05269	0.0694	0.06867	0.0861	0.08407
$(x_W - k_1)$	0.015	0.01491	0.045	0.04423	0.075	0.07296
n	10.31	10.32	7.33	7.39	5.75	5.87

STRIPPING COLUMN—SUMMARY OF RESULTS (*contd.*).

	$x_W = 0.01.$		$x_W = 0.03.$		$x_W = 0.05.$	
	Appr.	Exact.	Appr.	Exact.	Appr.	Exact.
$S = 3.$						
k_1	-0.002857	-0.002836	-0.008571	-0.008349	-0.01429	-0.01368
k_2	0.5887	0.5887	0.5994	0.5992	0.6101	0.6095
$\frac{\alpha}{mc^2}$	1.8911	1.8910	1.9232	1.9230	1.9554	1.9545
$(k_2 - x_F)$	0.1887	0.1887	0.1994	0.1992	0.2101	0.2095
$(x_W - k_1)$	0.01286	0.01284	0.03857	0.03835	0.06429	0.06368
n	7.17	7.17	5.22	5.22	4.24	4.24
$S = 4.$						
k_1	-0.002	-0.001991	-0.006	-0.005897	-0.010	-0.009714
k_2	0.6707	0.6707	0.6787	0.6786	0.6867	0.6864
$\frac{\alpha}{mc^2}$	2.012	2.0120	2.036	2.0359	2.060	2.0595
$(k_2 - x_F)$	0.2707	0.2707	0.2787	0.2786	0.2867	0.2864
$(x_W - k_1)$	0.012	0.01199	0.036	0.03590	0.060	0.05971
n	6.30	6.30	4.59	4.60	3.76	3.76

OBITUARY.

MR. HIRAM J. HALLE.

HIRAM J. HALLE, President of Universal Oil Products Company, died on 29th May at his home, Pound Ridge, New York. His health had been failing for some time.

Mr. Halle was born in Cleveland, Ohio, seventy-seven years ago. He entered petroleum refining at the time the automobile was coming into mass production, and for more than twenty-five years was an outstanding figure in the growth and development of the refining industry.

He became president of U.O.P. in 1915, and under his leadership, the Universal organization took an important part in the invention, development and servicing of processes which have revolutionized the refining industry, starting with the Dubbs cracking process and progressing through continuous research and development work to the perfecting of a number of catalytic refining processes.

CORRIGENDUM.

JULY JOURNAL.

"The Correlation of Cetane Number with other Physical Properties of Diesel Fuels."

P. 196. Table 1. Column 3. Delete "Outside" before ± 5 .

P. 197. Delete "(7)" before equation.

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Deegan, C. J., 945	Kornfeld, M. M., 953	Royds, J. S., 963, 967, 972	Worboys, W. J., 1022
Denison, R. A., 922		Rudd, H. W., 1024	Yust, H. R., 1016
Dodson, C. R., 966		Sawdon, W. A., 955, 960	Yuster, S. T., 970
la Due, J. P., 1014		Schnafsma, J. G., 1008, 1009	
Earlougher, R. C., 974			

Geology and Development.

922.* A Challenge to Geology. R. A. Denison. *Oil Gas J.*, 23.3.44, 42 (46), 66. (Annual Meeting of Amor. Assoc. Petrol. Geol. and affiliated Societies, 21st-23rd March, 1944.)—The challenge to geology, the science of the earth, is: Can its application to mineral exploration discover the new deposits needed to supply the deficit created by war and provide an ample reserve for unhampered progress in peacetime?

The development of a machine age has been possible only through an abundance of cheap power and energy, which is today obtained almost exclusively from mineral fuels. Of all the mineral fuels, the liquid and gaseous products of petroleum and natural gas are the most useful and the most universally adaptable. The winning of the war and the rebuilding in peace are dependent on adequate supplies of petroleum. Hence the challenge to geology falls heavily on petroleum geology. It can be said that the present high state of industrial development in U.S.A. is due to the acceptance and use by the petroleum industry of geology. American companies use more geologists than all the rest of the world combined. It is probably true to state that fully three-quarters of all the oil that has been produced, and that which is now known and proved, has been found by American geologists.

Over 600 members of the A.A.P.G., and possibly 1500 geologists of all classes are in the U.S. armed forces. Nearly all the college graduates in the past two years have gone directly to the armed forces. Students registered in geology now number less than 200, compared with a peace-time enrolment of 2500.

At present about 2500 petroleum geologists are called upon to find each year more petroleum than was found by the efforts of more than 3000 geologists in peace-time. Moreover, favourable sites for petroleum prospecting are becoming increasingly harder to find each year. New areas must be found in which to apply known and proved techniques; old areas and old data must be re-studied in the light of experience and new concepts. The knowledge of the relationship of oil accumulations to sedimentation must be increased to facilitate the finding of oil where there is little positive structural evidence, and where trapping is due to stratigraphic conditions. In this connection data obtained from prospecting drilling and from the structural configuration and stratigraphy of buried horizons revealed by geophysical prospecting are important and require careful consideration.

A great majority of all men who have ever worked in petroleum geology are still in the profession, and full use must be made of their accumulated experience in searching for oil.

After the war the available geological man-power will be greater than now, thus permitting accelerated exploration and the tackling of many problems. G. D. H.

923.* Wildcat Wells 17 per cent. Successful. F. H. Lahoe. *Oil Gas J.*, 23.3.44, 42 (46), 69. (Annual Meeting of Amer. Assoc. Petrol. Geol. and affiliated Societies, 21st-23rd March, 1944.)—The data on exploratory wells include wildcats (new-field and new-pool), outpost wells, and deeper-pool tests. In the deeper-pool tests only the footage drilled below the deepest known producing horizon is included.

During 1943, a total of 15,122,364 ft. was drilled in 3843 exploratory holes, and the 655 producers accounted for 2,741,303 ft. In the southern States district 1656 exploratory holes were drilled, totalling 8,227,468 ft. 287 holes were producers, and accounted for 1,546,956 ft. The average depth was 4968 ft., compared with 3935 ft. for the whole of U.S.A.

In 1943, it is estimated that 626 exploratory holes drilled on technical advice were successful, while 2616 were dry; 23 holes located for non-technical reasons were producers and 500 were dry; 6 producers and 72 dry holes were located for reasons unknown. Thus locations based on technical reasons are nearly 4.4 times as successful as those drilled without such advice.

Compared with 1942, there was a considerable increase in the exploratory effort in most parts of the country, but in Illinois, Indiana, and Nebraska there was a marked reduction both in the number of exploratory holes drilled and in their total depth. There was a decrease from 5013 ft. to 4239 ft. in the average depth of the holes drilled in California. There were marked increases in the average depth of exploratory holes in Kansas, western Kentucky, Louisiana, Mississippi, New Mexico, and Texas.

While the total number of exploratory holes, or the total footage drilled each year, has generally increased continuously since 1937, the total now proved reserves (discoveries, extensions, revisions) and the now proved reserves found during the year have declined substantially during the same period. There have been declines in the total new reserves per exploratory hole or per foot of exploratory hole, and in the newly discovered reserves per exploratory hole or per foot of exploratory hole.

G. D. H.

924.* Outline of Chinese Geology. J. M. Weller. *Oil Gas J.*, 23.3.44, 42 (46), 101. (Annual Meeting of Amer. Assoc. Petrol. Geol. and affiliated Societies, 21st-23rd March, 1944.)—Structurally, Greater China consists of three positive areas and three geosynclines. In late pre-Cambrian times and during the Palaeozoic, a thick series of dominantly marine formations accumulated in a great trough extending N.E.-S.W. across China proper, but in the north the Silurian, Devonian, and Mississippian are missing, although there was deposition in this area in the Pennsylvanian and early Permian. Important orogenic movements occurred towards the end of the Palaeozoic, and from the Middle Permian continental conditions were widespread, especially in North China and Central Asia. Eastern China was uplifted at the end of the Triassic, and subsequently it has never been subjected to important marine submergence.

During the Mesozoic and Tertiary, great thicknesses of continental sediments accumulated in a series of basins, the sites of which were changed by diastrophic movements, especially at the end of the Jurassic and of the Cretaceous, and in Middle and Late Tertiary times. There was extensive volcanism in the Late Cretaceous diastrophism. Some of the mountain chains have been raised by very recent faulting.

East-west curving mountain arcs dominate the structure of Central Asia, but in the coastal region there are N.E.-S.W.-trending structures. In most of China proper these two systems have produced a complicated interference pattern. Most parts of China are intensely folded and faulted, but there are five large sedimentary basins with moderate or gentle structures, two in Chinese Turkistan, one in Tibet and two in western and northern China. Seepages and other evidences of petroleum are known in three of the basins; the other two are practically unknown geologically. The Red Basin of Szechuan Province appears to be of greatest interest because of its stratigraphic section, type of structure and accessibility. G. D. H.

925.* Wildcat Completions and Discoveries. Anon. *Oil Gas J.*, 23.3.44, 42 (46), 167.—During the week ended 18th March, 1944, 6 oil-wells and a gas well were among the 65 wildcats completed in U.S.A. The largest discovery was a 386-brl. Cromwell sand well, some 4 ml. from the nearest oil production.

The results of wildcat completions in U.S.A. in the week ended 18th March, 1944, are tabulated by States and districts. G. D. H.

926.* Wildcat Completions and Discoveries. Anon. *Oil Gas J.*, 30.3.44, 42 (47), 139.—In the week ended 25th March, 1944, 59 wildcats were completed in U.S.A., 8 finding oil, and 1 gas. The preceding week had 65 wildcat completions.

During February, 1944, 1505 wells were completed, 912 giving oil and 161 gas. The total is 36% greater than in February 1943.

A table summarizes the results of wildcat completions in the week ended 25th March, 1944, and gives the cumulative totals for 1944 to that date. A second table gives data on all U.S.A. completions in February, 1944, including the production, footage, and numbers of wells in various depth ranges. G. D. H.

927.* Petroleum Possibilities in Szechuan Province, China. J. M. Weller. *Oil Wkly*, 3.4.44, 113 (5), 38.—The Red Basin of Szechuan Province covers about 60,000 sq. ml. The beds dip gently. Geological work on outcrops near Omei Shan and in the salt wells shows that as much as 3000 ft. of Tertiary beds may rest on the Cretaceous. There is a thick section of apparently non-marine post-Triassic beds which are red, and outcrop extensively. An important unconformity occurs at the base of the Permian. The mountains north of the basin show 15,000 ft. of Devonian beds (mainly quartzite, grading up into massive limestone) and about 1500 ft. of Silurian (greenish shale with nodular limestone). Carboniferous limestones, sandstones, and shales are reported in the south-eastern part of the province.

The Permian varies considerably on the edges of the basin, and includes basalt, limestone, shale, and coals. The distribution of Palaeozoic beds under the basin is unknown, but it is probable that the Permian Chisia limestone occurs throughout.

Some of the brine wells are 2000-3000 ft. deep, and a few give a little oil, and gas. Oil is recorded from various horizons in the Triassic, Jurassic, and Cretaceous, the best shows being in the Cretaceous (Tzuliuching formation), Jurassic (upper and lower parts of the Hsiangchi formation), and the Trias (upper and middle parts of the Chialingchiang limestone), but they are very erratic. Some of the brine wells are on anticlines, and so have served as tests for oil in post-Palaeozoic beds. On the Tzuliuching anticline brine wells have gone completely through the Chialingchiang formation without finding important amounts of oil. Elsewhere the wells have not reached the Triassic, and in the northern part of the basin they have not gone below the base of the Cretaceous. There have been few wells in the eastern part of the basin.

Several oil-seeps in the eastern part of the basin seem to be on or near steeply folded anticlines. 25 ml. south-east of Chungking a well was drilled near a seepage, apparently without especially favourable results.

The many anticlines generally trend northeast-southwest, and their seeming greater frequency in the south may be merely a question of more investigations there.

Jurassic and Cretaceous beds outcrop on the crests of some of these folds. Many folds are asymmetrical, with the south-east limb often the steeper limb. The intensity of folding increases from east to west.

The oil in post-Triassic beds, which appear to be mainly non-marine, has probably come from deeper horizons. At Tzuliuching some oil occurs in the Chialingehang limestone to which it may be indigenous.

The Mesozoic beds do not appear very favourable for oil according to the brine wells, but the Triassic limestone cannot be fully condemned. The more or less deeply buried Palaeozoic is attractive, but it has never been reached by drilling, and its distribution is unknown. The Chisia limestone (Permian) has carbonaceous and possibly bituminous beds at its outcrops in the mountains around the basin. Marine Carboniferous, Devonian, Silurian, and Ordovician beds probably underlie parts of the basin.

G. D. H.

928.* Vast Scheme of Development Projected for Middle East Oil. Anon. *World Petrol.*, April 1944, 15 (4), 35.—It appears that the Federal administration is determined to construct a pipe-line from the Persian Gulf to the eastern shore of the Mediterranean as soon as is practicable, and during the past few weeks special efforts have been made to speed up completion of the pipe-line contracts. The preliminary agreement left the location of the western terminus of the line in doubt as between Haifa and Alexandria, but it is believed that the former site is the more probable and may save as much as \$25,000,000 in the total cost.

To fulfil their obligation in respect of the billion barrel supply pledged for government use, the oil companies propose to develop as quickly as possible a known reserve of that extent. The Arabian American Company's programme calls for an expenditure of \$50,000,000 on pipe-lines, exploration, and drilling. This company proposes to replace its 3000-brl./day capacity refinery at Ras Tanura by a refinery of 50,000 brl./day capacity. A similar refinery is to be erected at the Mediterranean end of the pipe-line.

On Bahrein Island, the Bahrein Petroleum Company plans to increase its refinery capacity from 30,000 to 58,000 brl./day. The Bahrein wells give about 20,000 brl./day at present.

American entry into the search for petroleum in Saudi Arabia dates from 1933. The area of the concession was not closely defined but extended about 200 ml. inland from the eastern coast of the country. The first producing well was completed in the Dammam area in 1936, oil being found at a depth of about 2000 ft. Later oil was found below 4000 ft., and at still greater depths. When oil had been found in substantial quantities the concession was expanded to include virtually all the territory under Ibn Saud's authority. An agreement was made with the Texas Company whereby it became a joint owner with Standard of California of the companies operating in the Persian Gulf area, including the California Arabian Standard Oil Company, the title of which has now been changed to Arabian American Oil Company, Bahrein Petroleum Company, Ltd., and the California Texas Company, which is the marketing organization.

The Dammam field has 25 wells giving 15,000 brl./day, although they are capable of a much higher output. 100 ml. north-west of Dammam is the Abu Hadriya field, where one well has been drilled to 10,000 ft., and Abqaiq, which is regarded as the most promising field yet located in this region. Many other favourable structures have been found and await testing. The known reserves of Saudi Arabia are roughly estimated at 4,000,000,000–5,000,000,000 brl., and geologists believe that the area immediately around the Persian Gulf will prove to be one of the world's greatest oil-producing regions.

G. D. H.

929.* California Spurs Production to Meet War Demands. R. Winterburn. *World Petrol.*, April 1944, 15 (4), 48.—During 1943, in spite of steadily expanding activity in development and exploration, withdrawals from stocks in California indicated an average deficiency in crude oil production of about 75,000 brl./day. Various restrictions caused the completions in California in 1942 to be 569 wells, compared with 941 in 1941. 1163 wells were completed in 1943. In the absence of these restrictions the additional wells which might have been drilled would probably have made the present productive capacity about 40,000 brl./day higher than it is. Other obstacles

to development have been man-power, and material and equipment shortages, and the low price of crude. Many stripper wells were not producing during the early part of the war because of low prices, but a revision of prices in 1943 returned many marginal, low-gravity oil wells to production and stimulated the development of the undrilled parts of the same fields. A revision of prices for high-gravity crudes seems necessary.

Nearly all California's current production is from fields found more than five years ago. Nineteen fields have been opened since the beginning of 1939, but most of them have thin oil-sand sections and comparatively small productive capacities. These nineteen fields gave about 38,000 brl./day in December 1943, this being some 4-8% of the State's total. Ten fields found in the preceding five-year period gave 220,000 brl./day in December 1943. Seven new fields were found in 1943, but only the Pleasant Valley Gatchell sand area seems likely to give any important increase in productive capacity or reserves.

Exploratory drilling in northern California has increased, and eight new gas-fields were opened in 1943.

Production in all restricted fields has been raised to the maximum efficient rate. 1850 idle marginal producers of heavy oil have been returned to production. Most of the new wells completed in 1943 were in the shallow heavy-oil areas of the San Joaquin Valley, on the border of the basin. 706 of the 817 new San Joaquin Valley oil-wells were in these shallow fields. Drilling activity in the Los Angeles Basin was largely concentrated in the Wilmington, Huntington Beach, and Torrance fields. In the Coastal area deep zones are being developed at Ventura Avenue, while at Santa Maria Valley there has been extension of the proved area.

There are sufficient undrilled proven locations to permit continued drilling at the present rate well beyond the end of 1944, and considerably more wells are likely to be drilled than in 1943. It is expected that the productive capacity at the end of 1944 will be 850,000 brl./day. The present daily demand of the area is 950,000 brl.

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930.* Report on Petroleum Reserves. Anon. *World Petrol.*, April 1944, 15 (4), 56.—The A.P.I.'s estimate of U.S. crude oil reserves at the end of 1943 is 20,064,152,000 brl., 18,641,000 brl. less than at the end of 1942. It refers solely to the proved or blocked-out reserves of crude oil (including condensate) known to be recoverable under existing economic and operating conditions. Details are given by States of the proved reserves at the beginning and end of 1943, the production during 1943, and the new reserves found in 1943.

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931.* Northern Alaska Naval Reserve to be Developed if Congress Grants Funds. Anon. *Oil Wkly*, 3.4.44, 113 (5), 52.—The Point Barrow area of Alaska has formations similar to those which yield oil in various parts of U.S.A. and Canada. If funds are provided, geological and geophysical investigations will be made in this area. Oil-seeps are known in this region.

The development of oil-fields in Northern Alaska will present many difficult problems.

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932.* Canadian Production for 1943 Drops Seven per cent. Under Figure for 1942. Anon. *Oil Wkly*, 3.4.44, 113 (5), 60.—During 1943, Canada produced 9,377,241 brl. of crude, compared with 10,050,544 brl. in 1942. New Brunswick gave 25,405 brl. in 1943, and the Ontario fields 138,457 brl. Alberta gave 9,213,379 brl. in 1943, and, of this, Turner Valley provided 8,991,528 brl., 710,191 brl. less than in 1942. In December 1943, Vermilion was giving 516 brl./day (29 wells), Taber 175 brl./day (4 wells), Wainwright 43 brl./day (7 wells), and Red Coulee 25 brl./day (7 wells). 27 oil-wells had been completed in the Norman Wells area up to November, 1943.

A table lists the discovery year, 1942-1943 and cumulative production, and daily production at the end of 1943 for the various Canadian oil-fields.

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the Izhma river, its capacity being reported as over 30 million barrels per year. A now 1100-ml. railroad to Khabarova gives an outlet for the mineral deposits of the North Urals and the Pechora Basin. G. D. H.

934.* Wells Completed in the United States in Week Ended April 1st, 1944. Anon. *Oil Wkly*, 3.4.44, 113 (5), 81.—315 field wells and 75 wildcats were completed in U.S.A. in the week ended 1st April, 1944. 12 old wells were deepened. A total of 5383 wells has been completed in the first three months of 1944. The corresponding figure for 1943 was 4301.

A table summarizes by States and districts the results of field and wildcat completions in the week ended 1st April, 1944, and gives cumulative totals for 1944.

G. D. H.

935.* Winkler's Three Ellenburger Pools Expanding into Major Reserves. H. S. Norman. *Oil Gas J.*, 6.4.44, 42 (48), 26.—About 1500 acres have been added to the proven productive area of the three Ellenburger fields in northern and eastern Winkler County, West Texas. Five new wells have been completed or have qualified as commercial producers in preliminary tests on the Ellenburger pay-zones.

The Wheeler field was extended nearly $\frac{1}{2}$ ml. to the north-east by a well which produces from 10,670–10,710 ft., and a $\frac{1}{2}$ -ml. south extension is assured by another well. $\frac{1}{2}$ ml. west of the Wheeler discovery, another well, after acidization, has given 50 brl./hour and 1,000,000 cu. ft. of gas/day from 10,642–10,661 ft.

The Keystone Ellenburger field has been extended 1 ml. west by a well which produced 1092 brl. in a 17-hour test, from 9610–9660 ft. 1 ml. south-east of the discovery well a further test has reached 8600 ft. The Kermit Ellenburger field has been extended $\frac{3}{4}$ ml. south by a well which produces from 10,647–10,658 ft., and a 1-ml. north-west extension is indicated by a well which has had a partially successful drill-stem test at 10,699–10,741 ft. The packer failed after 10 minutes.

This Ellenburger oil is generally of 40° to 42° A.P.I. gravity, and comparatively free from sulphur.

These deep wells are very costly, and the cherts in some areas make drilling very slow. Moreover, rigs suitable for drilling 10,000-ft. holes are not common.

G. D. H.

936.* Two Wildcat Tests Direct Attention to Oklahoma's Anadarko Basin and Florida. Anon. *Oil Gas J.*, 6.4.44, 42 (48), 26.—In Oklahoma, Carter Oil Co. 1 Cottingham in 4-7n-3w has found a new deep Wilcox sand field. The oil-sand (the Second Wilcox) is at a depth of 10,625 ft. This has given impetus to the Anadarko Basin play, and two more wells will be drilled on the structure this year. The nearest Wilcox production is at West Moore, 15 ml. to the south.

A deep well, 1 ml. west of Florida's discovery well near Sunniland, is reported in some quarters to have shown only mud in a drill-stem test at 9800 ft., while others claim that oil showed in the drilling mud.

G. D. H.

937.* Nine Tests Planned This Summer in Various Parts of South Dakota. Anon. *Oil Gas J.*, 6.4.44, 42 (48), 102.—At least 9 test wells have been planned for this summer in South Dakota. One well is to be drilled near Gustave in Harding County, and it may go to 8000 ft.

Two major tests were drilled in 1943. An 8000-ft. well in Harding County was dry, while the other well, $\frac{1}{2}$ ml. north of Ardmore in Fall River County, was completed as a gas-well. In 1940, a 7980-ft. well drilled near the Harding County test found small oil shows in the limestone, and a considerable show in the Deadwood sandstone.

There has been considerable leasing recently, and interest in western South Dakota has been stimulated by a well in the Newcastle, Wyoming, area, a few miles from the South Dakota border. This 3852-ft. well is expected to produce commercially.

G. D. H.

938.* Germany Fights for Oil. J. B. Eby. *Oil Wkly*, 17.4.44, 113 (7), 11.—Over one-third of the oil for the German war effort comes from the Ploesti area. Synthetic

oil may provide 45,000,000 brl./year, or nearly 50% of Germany's requirements, while Germany's domestic crude production is probably less than 7,000,000 brl./year.

From 1933 to 1939 there was a marked increase in Germany's oil imports, and from 1935 to 1939 the annual reserve for storage ranged 5-10 million barrels. From September 1939 to June 1941, the German war machine was using up oil faster than it was being produced, for Frodborg estimates that for a full year of offensive warfare 165,000,000 brl. of oil would be needed by Germany's armed forces. Hence it was desirable, from Germany's point of view, to secure the oil resources of the Caucasus and the Near East. Germany occupied the Maikop fields after they had been thoroughly scorched by the Russians, and it is doubtful whether she obtained any helpful quantity of oil during the occupation. The Malgobek field of the Grozny area was also occupied.

On the home front, there was now drilling in Nienhagen, Wietze, Olheim and Oborg fields, and the Nienhagen field had a notable extension. Thumon reports new fields or valuable prospects at Steimbke, Broistadt, Adolfs-Gluck, Sottorf, Tegernsee, Eicklingen-Sandlingen, Reitbrook, Worms, and Moeckefeld, most of which are in the North German plain area. The Reitbrook field near Hamburg was probably the best in promise up to 1939, according to Thumon, but there is little indication of their performance. The Heide field of Holstein produces gas. Haack mentions discoveries at Molme, Fallersloben, Rodonwald, and Fallstein within the North German basin.

In 1937 high-pressure hydrogenation of hard coal produced nearly 6,750,000 brl. of synthetic gasoline, about 90% of Germany's synthetic gasoline output that year. Figures for synthetic oil production since war began have been kept a secret, although various estimates have been made. Frodborg's consumption figures of 112,000,000 brl./year for defensive war and 165,000,000 brl./year for offensive war, if reliable, suggest that Germany's oil production from all sources must be far greater than the available data would indicate. The trend of some German propaganda on air raids may be construed as pointing to an oil shortage.

If the Roumanian fields fall into Russian hands, 35-40% of Germany's oil supply will have gone.

Tables give data on Germany's oil production, consumption, imports, and on the production of petroleum substitutes in Europe. Some maps are included. G. D. H.

939.* Efforts to Extend Poza Rica Field Highlight Drilling in Mexico. Anon. *Oil Wkly*, 10.4.44, 113 (6), 32.—The Poza Rica field is believed to have been outlined definitely east of the Cazonos river, where flank wells have shown water encroachment, and efforts have been made to define the western limits in the San Miguel Mecatepec area west of the river. In this connection a few small producers were drilled in 1943, and 2 wells, one giving 4500 brl./day, have been completed in 1944.

4 ml. north-west of the famous Dos Bocas well, Pemex Rancho Nuevo 1 has found salt water at 6600 ft. This may have been searching for a structure on echelon with the Golden Lane area. Other wildcats are under way near Guerrero, San Luis Potosi State (30 ml. south-west of Panuco), and in Veracruz State, some 15 ml. west of the Golden Lane structure, just north of the Tuxpan river.

In 1943, 16 wells were completed in Mexico, 9 being failures. The production averaged 95,600 brl./day in 1943, compared with 95,110 brl./day in 1942. Poza Rica gave 20,500,000 brl. in 1943. Its potential recovery is estimated to be 545,000,000 brl., of which about 35% has been recovered. This field has 50 wells, a few of which are closed in because of water and other troubles.

Mexico's exports were 5,350,000 brl. of oil in 1943, 25% being crude.

Tables give data on well completions in recent years, on the drilling activity at the beginning of 1944, and on the production since 1940. G. D. H.

940.* United States Completions for First Quarter Running Ahead of 1943 Record. Anon. *Oil Wkly*, 10.4.44, 113 (6), 52.—More rigs were operating in U.S.A. at the end of March than at the beginning, and in the first quarter of 1944 22% more wells were completed than in the corresponding period of 1943. Increased drilling over 1943 is shown by most States and districts except Arkansas, Illinois, North Louisiana, Texas Panhandle, and South-west Texas. The increases were above the average in California, Kentucky, Michigan, Mississippi, Montana, New Mexico, East Texas, North and West Central Texas, West Texas, Coastal Texas, and West Virginia.

At the end of March there were 2898 wells drilling and 375 rigging.

A table summarizes by States and districts the completions in March 1944 and during the first quarter of 1944, and gives some 1943 figures for comparison. G. D. H.

941.* Size of American Holdings in Near East Revealed. Anon. *Oil Wkly*, 10.4.44, 113 (6), 53.—The joint holdings of the Standard Oil Company of California and the Texas Company through the Arabian American and Bahrein Petroleum Companies are about 278,372,000 acres on the west side of the Persian Gulf, all except about 100,000 acres being in Saudi Arabia. The Gulf-Anglo-Iranian holdings in Kuwait may amount to as much as 5-6 million acres. G. D. H.

942.* Shell Acquires Large Area in South Alberta. Anon. *Oil Wkly*, 10.4.44, 113 (6), 53.—The Shell Oil Company of Canada has acquired an exploration and development lease on about 54,000 acres of Crow Indian territory in the southern foothills region of Alberta. The block is traversed by the Grease Creek in the south and by the Fallen Timber Creek in the north. G. D. H.

943.* Wildcat Activities in Colombia Are Reported. Anon. *Oil Wkly*, 10.4.44, 113 (6), 53.—Early in March, Shell's San Angel 1 was testing after reaching a depth of 9130 ft. Dificil 2, drilled to 6410 ft., was also testing. Retiro 1 is halted at 5802 ft., and electric log and formation tests have been made, although the results have not been announced.

Cantagallo 4 has been drilled to 6881 ft., penetrating the top of the Cretaceous. This is 1000 ft. below the top of the same formation in Cimitarra 1.

Casabe 16 was completed late in February for 930 brl./day, and Casabe 15 is testing. Casabe 17 is drilling. Socuavo 6, 7, 8, and 9 are in various stages of progress, and the same is true of Bocas 4 and 5.

In the Llanos, Shell's San Martin 1 is shut down awaiting the arrival of a fishing tool. G. D. H.

944.* Sixteen Wells Drilled in Peru During Year. Anon. *Oil Wkly*, 10.4.44, 113 (6), 53.—During 1943, drilling in Peru was limited to the Lobitos region, where, out of 16 completions, 3 found oil and 3 found gas; the rest were dry. At the end of the year the Peruvian production was about 38,000 brl./day. G. D. H.

945.* Completions Lag 14 per cent. Behind Quota. C. J. Deogan. *Oil Gas J.*, 13.4.44, 42 (49), 117.—4654 wells were completed in U.S.A. during the first quarter of 1944, compared with the P.A.W. quota of 5400 wells. All the P.A.W. districts fell below their quotas, the figures being 11%, 3%, 23%, 22%, and 10% for Districts 1, 2, 3, 4, and 5, respectively. In Districts 1 and 5 there was no major transportation problem, but there is a crude shortage, and the completion deficiency is attributed to lack of man-power and equipment, and to inadequate crude prices. Districts 3 and 4 lack transportation outlets, and companies operating in both Districts 3 and 2 seem to have concentrated their man-power and equipment on the latter, especially when their refineries lie in that district. Moreover, in much of District 2, 10- and 20-acre spacing is permitted. Man-power and equipment shortages and low prices have affected activities in all districts.

Drilling contractors state that the drilling time on wells is continually increasing, and on deep wells, in some cases, it is 25-50% longer than normal. Some of this increase is due to the condition of the equipment, as well as to the general lower standard of the available employees.

Statistical data on completions and quotas are given.

G. D. H.

946.* Colombian Production Drops from Wartime Peak. Anon. *Oil Gas J.*, 13.4.44, 42 (49), 124.—In January 1944, Colombia produced 1,266,034 brl. of crude, 583,947 brl. less than in December 1943. The January, 1944, natural gasoline production was 351,725 brl.

The total crude production in 1943 was 13,271,212 brl.

A table gives the production by months for 1942 and 1943.

G. D. H.

947.* **Wildcat Completions and Discoveries.** Anon. *Oil Gas J.*, 13.4.44, 42 (49), 331.—In the week ended 8th April, 1944, 67 wildcats were completed in U.S.A., nine finding oil, and one gas.

A table summarizes the results of wildcat completions by States and districts, for the week ended 8th April, 1944, and gives the cumulative totals to that date. G. D. H.

948.* **Geological Developments in the South-eastern Gulf Coast States.** H. R. Brankstone. *Oil Gas J.*, 20.4.44, 42 (50), 58.—The Amory gas-field of Mississippi was discovered in 1926. Production is from shallow wells in the Hartselle (Mississippian) sandstone. In 1930, the first well of the Jackson field was drilled. This field obtains gas from sands in the Selma Chalk (Upper Cretaceous) over an area of 7500 acres. The structure, which has 1500 ft. of closure, is believed to be of igneous origin. The Tinsley field, discovered in 1939, first produced oil from the Woodruff sand of Selma age, the producing horizon being correlated with the Jackson field's gas horizon. There are 6 pays in the Selma, Eutaw and Tuscaloosa. The structure is a faulted dome. In 1940, the Pickens field was found. Oil is obtained from the Wilburn sand of the upper Eutaw, and accumulation is controlled by faulting which interrupts the south and south-west dip of the beds. The productive area is about 2000 acres. The Carey field was discovered in 1941, oil being in a sandy limestone of Selma Chalk age. The field is on a small structure on the east flank of the Sharkey County High.

109 tests were drilled in Mississippi in 1943, 76 being considered wildcats. The Flora field was discovered, but 6 subsequent wells in it were dry. A considerable amount of geophysical work was carried out in the southern two-thirds of the State. Deep and shallow salt domes are known to occur in the Mississippi salt-dome basin. Other possible oil traps are faults interrupting the regional dip, and pinch-outs.

During 1943 Mississippi produced 18,868,000 bbl. of oil, compared with 27,545,000 bbl. in 1942.

Ten wildcats were completed in Alabama in 1943, 5 tests being in the Warrior Coal Basin, and 5 in the south-western area. The drilling has added geological information, and it is believed that the massive sand of the basal Tuscaloosa offers a reasonable chance of production. The Eutaw is present, but this is shale-chalk rather than sand. A deep well indicates that the Cotton Valley and Smackover will be present.

Four wildcats were completed in Georgia in 1943, but only one showing was reported (in Wayne County at a depth of 680 ft.). Scattered drilling in the past four years has shown that the southern third of the State may be of interest.

During 1943, 11 tests were drilled in Florida. A test in Collier County found oil showings in dolomite and limestone at 9800–9848 ft. (Lower Cretaceous), and saturated limestone at 11,600–11,626 ft., which on test gave several hundred barrels of fluid per day with 25% oil. Beds of similar age are reported to have given oil showings in Mississippi.

The surface formations of Mississippi, Alabama, and Georgia dip towards the coast and mask the subsurface structure. The Sharkey County Uplift is associated with an area of igneous activity which involves the Monroe and Richland gas fields of Northern Louisiana. Shows of oil have been located in drilling on the McHenry High, and there are excellent sands in the Eutaw and Tuscaloosa (8000–10,000 ft. deep). Commercial accumulations of oil seem likely in four horizons in the basin west of the McHenry High and south of the Jackson Uplift. The Hatchetigbee Anticline of southern Alabama has been tested without success, although beds are present which produce elsewhere. Oil and gas may be present in beds on the Ocala Uplift.

There may be 18,000 ft. of sediments in the southern part of Mississippi or Alabama. The Wilcox occurs in Mississippi, Alabama, and Georgia, and produces at Cranfield in Mississippi. The sands in this sand and shale formation are lenticular. The Selma Chalk is one of the most important pays in Mississippi. It varies in lithology and thickness. The Eutaw is predominantly sandstone and shale, the former being lenticular. The Tuscaloosa consists of sands and shales, oil occurring in the upper part. The thickness ranges 900–2000 ft., and the sands pinch out irregularly. On structure the Paluxy lies unconformably below the Tuscaloosa. Shows of oil have been reported in Mississippi. Deep drilling has shown that the Lower Glenrose may be present in Alabama and Florida, as well as in Mississippi where it has oil and gas showings.

A map and stratigraphical column are included.

G. D. H.

949.* **Wildcat Completions and Discoveries.** Anon. *Oil Gas J.*, 20.4.44, 42 (50), 109.—In the week ended 15th April, 1944, 58 wildcats were completed in U.S.A., 9 finding oil and 1 gas.

The wildcat completions are tabulated by States and districts, with cumulative totals for 1944 to the 15th April. G. D. H.

950. **Exploratory Activity for Quarter Exceeds Government's Goal.** L. J. Logan. *Oil Wkly.*, 24.4.44, 113 (8), 75.—865 exploratory holes have been completed in U.S.A. in the first quarter of 1944, 114 more than in the corresponding period of 1943. 160 of these tests have proved productive, compared with 124 in the same period of 1943. 24 important extensions have been made to established fields in the 1944 period, and 68 new oil-fields, 17 new gas-fields, and five new distillate fields have been found. 23.3% of the 270 tests completed in March were successful, and there were 52 new fields and now pays. The March exploratory drilling was nearly at the March 1941 level. In March, 1944, new reserves of oil and gas were found in Texas, Louisiana, Oklahoma, Kansas, California, Wyoming, Illinois, Ohio, and Michigan. Texas had 30 of the new fields and now pays; Kansas had 9 and Illinois 5. Two new oil-zones have been found above 4500 ft. in the Buena Vista field, Kern County, California.

The Lake St. John field, Tensas Parish, North Louisiana, has been extended, and this may stimulate increased exploration for the Tuscaloosa deep oil-sand in the region. Another seemingly important March discovery is in Winkler County, Texas, where a promising Clear Fork lime oil reserve has been found, while there are also possibilities of Ordovician production.

Tables summarize the results of exploratory drilling in U.S.A. during March and during the first quarter of 1944. A list is given of the discoveries in March 1944, with the name, place, depth producing horizon, mode of discovery, etc. G. D. H.

Geophysics.

951.* **The Radioactivity and Organic Content of Some Palaeozoic Shales.** R. F. Boor. *Oil Gas J.*, 23.3.44, 42 (46), 105. (Annual Meeting of Amer. Assoc. Petrol. Geol. and affiliated Societies, 21st-23rd March, 1944.)—Measurements of the total beta activity, the total alpha activity, and the radium content permit the determination of the radioactive substances in sedimentary rocks, the important radioactive substances being uranium, thorium, and potassium. The radioactive substances are found mainly in association with the heavy minerals of sands and sandstones; as the active potassium isotopes in evaporites, oilfield brines, in micas and other potassium-bearing minerals in clays and shales; and in the uranium and thorium content of shales, clays, impure limestones and marls, shaly sandstones, and organic sediments. Pure limestones and quartz sands have practically no measurable radioactivity, but black shales with up to 16% of organic matter have been found to carry high concentrations of the three main radioactive elements. Individual shale formations showed good correlation between the uranium content, the thorium-uranium ratio, and the carbon content. G. D. H.

952.* **A Geophysical Survey of Kuwait, Persian Gulf.** P. H. Boots. *Oil Gas J.*, 23.3.44, 42 (46), 105. (Annual Meeting of Amer. Assoc. Petrol. Geol. and affiliated Societies, 21st-23rd March, 1944.)—Prior to geophysical survey in 1936-1937 there was very little geological information available about Kuwait, which covers 12,250 sq. km. Magnetometer and gravimeter reconnaissance surveys soon revealed an anomaly which was investigated seismographically. Eventually the gravity survey covered the whole country. The seismograph survey outlined the structure which on drilling proved to be a large oil-field. G. D. H.

953.* **Subsurface Correlation by Detailed Electrolog Tabulation.** M. M. Kornfeld. *Oil Gas J.*, 23.3.44, 42 (46), 109. (Annual Meeting of Amer. Assoc. Petrol. Geol. and affiliated Societies, 21st-23rd March, 1944.)—Starting with the usual skeleton control of known stratigraphic markers or palaeontological data, further correlations proceed independently, sole reliance being made on close inspection and comparison of electric logs, assisted by superimposition on a light table. Multiple control points

are used to substantiate individual comparisons to ensure the accuracy of any one correlation. Only the most obvious correlations are recorded in the early stages. Progress is always from known to unknown, leap-frogging all obscure zones until complete positive correlations are established. Then each obscure zone is studied in detail, and further correlations are made if possible.

Differences between correlation points are not calculated until all comparative depths are completed and tabulated, thereby ensuring unprejudiced correlations free from any influence of other comparisons above or below. Once the information is tabulated and ready for study, further dependence on the electrolog strips is removed, except for checking possible errors in reading or for examining special parts for details of departure from normal.

The above method of detailed comparison provides the tabulated quantitative data of a high degree of precision. It avoids the lettering and colouring of electrolog sections. Discrepancies in scale or distortion of electrologs due to lack of uniformity in printing are minimized or eliminated by quantitative tabulation. Isopach calculations are made from the tabulations instead of from electrolog strips. G. D. H.

954.* **Reflection Seismograph Performance Along the Gulf Coast.** E. E. Rosairo. *Oil Wkly*, 10.4.44, 113 (6), 16.—At present the reflection seismograph must be considered as the dominant prospecting method, for thereby a large proportion of all prospects are evaluated and most test locations are made. Since about 1930, the reflection seismograph has dominated Gulf Coast oil prospecting, and currently this area is the scene of operation of about 25% of all the reflection crews working in U.S.A. A measure of the success is the ratio of attainment to effort, and this can be expressed in several ways: (1) the achievement ratio—number of confirmations per crew-year; (2) initial risk of discovery—crew-years per producing structure discovered (or confirmed); (3) gross reward ratio—millions of barrels of oil found per crew-year.

The term "confirmed" is used in preference to "discovered" to avoid argument as to whether full or only partial credit is to be given to the reflection seismograph. This usage is important not only in the early stages of a method (when it may contribute materially in the evaluation of prospects already indicated by other techniques), but also in the later stages when the method has only marginal anomalies to find (when another technique may contribute materially to the location of a test well).

For the purpose of this analysis, the discovered oil volumes have been credited back to the year of discovery of oil or gas in a given field.

There is a lag between prospecting effort and achievement, and an attempt has been made to determine this by trial and error. Running three- and two-year averages were tested with two- and one-year lags, and the most reasonable fit was found with running two-year averages and one-year lag. The averages were plotted as ordinates with time as abscissæ. In the activity plot the curve rises increasingly steeply after 1929, reaches a peak in 1937, and drops to a minimum in 1940, followed by a rise. The confirmation plot has a low peak in 1931, a much higher peak in 1937 after a steady rise, and then it oscillates at a high level. The discovery volume plot has a low peak in 1931, and a high peak in 1934, after which there is an irregular decreasing trend.

In plotting the three measures of success using two-year running averages and one-year lag, the measure of success may be referred to the discovery year or to the preceding year of the exploration.

The achievement ratio shows a rising trend in spite of the accumulating hazard resulting from the growing record of discoveries credited to the reflection seismograph. This rise can be attributed to the improvements of the method, to the increase in the number of the prospects as the general level of promise falls, and to increase in the amount of subsurface data available. The trend is similar for the rest of U.S.A. The initial risk is the reciprocal of the achievement ratio, and if the latter is linear the former should be hyperbolic, or approximately exponential over a limited range. On plotting the initial risk ratios on a logarithmic scale the data are representable by a straight declining line, which shows that over a period of about ten years the initial risk of finding a producing structure has fallen by 50%.

The gross-reward ratio has been found by dividing the two-year running averages of the annual discovery volumes by the two-year running averages of the activity one year earlier, and this ratio has been plotted on a logarithmic scale. There is an

exponential decrease of the gross reward, and it is halved every 2.5 years. Joint consideration of the initial risk and gross reward trends shows that if these two trends persist "successful" prospecting can hardly be anything but a losing venture, since the latter decreases more rapidly than the former.

A number of tables and figures are included.

G. D. H.

Drilling.

955.* Problems of High Pressure Drilling in India. W. A. Sawdon. *Petrol. Engr*, March 1944, **15** (6), 67-70.—Drilling practices in the Khaur field in the Punjab Province of India are described. Production from the Khaur field comes from both shallow and deep zones. Although some oil is being taken from small, old wells at very shallow depths, most of the production is from zones up to 3800 ft. in depth and from a deeper sand found at approximately 5600 ft. The extremely high pressure of a water sand at approximately 5000 ft. makes the drilling of the deeper wells somewhat difficult. Much trouble was experienced in getting through this sand while drilling the earlier of the deep zone wells, but methods and equipment have since been developed to ensure successful passage through the high-pressure zone.

The high-pressure sand made a tremendous volume of water during the drilling of the first wells, but this volume was depleted as more holes were drilled. The pressure, however, built up rapidly when the sand was penetrated, so the driller watched the hole carefully as he approached the top of the zone to observe whether the fluid increased. When the sand was reached, the circulating system was closed in, and drilling from that point continued under pressure until the high-pressure sand was drilled through and the water-string was run and cemented. While going through this zone, drilling was done either with back pressure being held on the mud returns by means of flow-beans or with the circulating system completely closed and the mud fluid returned directly to the suction end of one of the pumps. Conditions governed which method was to be used, and connections were so arranged that the driller could switch from one method to the other whenever he thought it desirable.

Casing programmes and details of mud system, pressure drilling, drill pipe, and of completion are given.

A. H. N.

956.* Oil Well Cementing. Part 2. C. A. Pitts. *Petrol. Engr*, March 1944, **15** (6), 87.—The second part of this paper deals mainly with squeeze cementing theory and practice. At the end of the paper conclusions are reached to the effect that in multiple sand-wells the most common cause of casing cementation failure is due to too little cement being run to cover all producing zones properly. This problem can be overcome by the use of data from caliper and temperature surveys. Channelling of cement slurries can be eliminated by displacing the cement into the annulus at a velocity high enough to maintain turbulent flow conditions. By running enough excess cement to bring the point of hydrostatic balance of the cement columns inside and outside the casing to a level above the top producing zone, top contamination difficulties can be overcome. Even though cement is placed in such a manner as to eliminate channelling, some formations may not be properly segregated. The characteristics of some formations may be such that they cannot be overcome by normal cementing methods, and research work is being done on this problem. The most economical and satisfactory method found to date is that of squeeze cementing through perforations after the casing cementation.

Squeeze cementing failures are invariably the result of placing insufficient cement in the well and in such a way that it will not set in the desired place. This problem can be largely overcome by squeezing at the most logical points above and below or in the producing zone. Control in placement of slurry squeezed through perforations is facilitated by reducing the number and extent of perforations to a minimum and by doing all squeeze cementing work prior to perforating the producing zone for production tests. Cement slurries thus properly placed must be given a chance to set. To ensure that the set will not be disturbed by agitation, etc., it is best to perform the squeeze job so that the cement is caused to set by dehydration. Such dehydration can be recognized by observing surface pump pressure.

A. H. N.

957. **A Method of Drilling in Deep Water.** H. E. Gross. *Petrol. Tech.*, March 1944, 7 (2), A.I.M.M.E. Tech. Pub. No. 1722, 1-8.—The waning supplies of oil in U.S.A. make it desirable to develop oil-fields in new provinces, among the most attractive of which appear to be submarine deposits, notably the Great Lakes, where the possible resources will be shallow and the water is not so rough as in the open seas. The shallower parts of the lakes could be drilled by the submersible barge of Giliasso, which is suitable for water less than 10 ft. deep, and not open, as offshore in the Gulf of Mexico or the Pacific Ocean. Conventional rigid foundations are capable of withstanding rough seas in 10-40 ft. of water, but they are unsuitable economically. The different types of rigid foundations cost \$5000-\$90,000, depending on the depth of water and type of construction. By directional drilling several wells can be drilled from a single foundation.

A portable buoyant marine foundation is described which would be suitable for wells up to 3500 ft. deep. For water 30 ft. deep the cost would be about \$3000. The foundation consists of vertical buoyant logs made of casing, with buoyant horizontal compression members, and rod sway-bracing, the whole being set on stout skids with a toe, to facilitate dragging from the beach where it is assembled. Decking is placed on top to carry the derrick, and when the foundation is in position sand and gravel are dumped on the skids to prevent movement or overturn in rough weather. This type of foundation might be used in water as deep as 100 ft., and could be moved from one location to another after jotting away the sand and gravel from the skids and removing any other tie-downs.

For water up to 600 ft. deep, and where high oil recoveries or the possibility of moving the foundation to new sites would justify the cost, a more elaborate foundation is suggested. This has buoyant skids of casing, and logs mounted on them, following the log taper of a derrick, the logs being so spaced as ultimately to provide a platform of suitable width for the derrick. The first section of the foundation is built on the beach, with buoyant compression members and rod bracing. It is towed to sea until nearly submerged, whereon the second section is added. It is then towed farther out, and the third section is added, this process being continued until the drilling site is reached. Sand and gravel are placed over the skids. If the foundation is to remain permanently, the logs may be drilled through with a portable rotary for about 200 ft. into the sea-floor, and then anchorage pipe may be cemented.

An alternative foundation for water up to 100 ft. deep has a square-based pontoon with vertical sides and a central hole through which drilling can proceed. About this is built a spread footing and the lowest section of the vertical tower, the legs of which consist of angle irons which act as vertical guides for movement of the pontoon relative to the tower. After completing the first tower section, the assembly is towed to deeper water and a second section is added, this process being repeated until the well site is reached. Air-filled drums may be used to facilitate movement during the building, but these are finally removed, hydraulic anchorages are driven into the sea-floor, and sufficient water is admitted to the pontoon to sink it 10-20 ft. below water level, in which position it is secured. Sand and gravel are placed over the spread footing, and a rig platform is built. Later the water is pumped out of the pontoon to reduce the load on sections of the tower below the pontoon.

All these types of foundations offer a minimum of cross-sectional area to winds and waves. In order to avoid floe-ice trouble in the Great Lakes, gas-wells would be completed with well-head fittings just above the sea floor and with a submarine pipe-line and provision for de-waterring. An oil-field might have similar fittings, the field being flowed to depletion by an induced water-flood, started when the field is brought in.

G. D. H.

958. **Reeling Unit Extends Life of Wire Lines.** Anon. *Oil Wkly*, 27.3.44, 113 (4), 15-16.—One company, operating a number of rigs, gives its lines the best possible care in handling an ingenious semi-trailer wire-line spooler, independently powered, which requires but one man to operate, and which has ample power to permit controlled tension winding or feed off of the line. The frame of this unit utilizes the rear half of a discarded heavy-duty truck, the front section of the frame being cut off, given a high lift, and shaped to fit over the king-pin section of the trailer unit intended to provide motive power for transferring the unit through company property. The rear axle is retained, the dual wheels being required to support the anticipated weight

of rooling unit and heavy line, while the brake system provides necessary control on grades. On the foundation provided by the reconstructed truck frame is located the power unit, consisting of the engine and rear axle from a light truck, the engine being set up between the side frame members, and driving the rear axle through a shortened, conventional propeller shaft with the usual universal-joint complement retained to insure required flexibility. In place of the former wheels, sprockets are fitted, to run on the bearings which formerly served for the drive wheels of the truck. By arranging the drive through the differential so as to allow either end of the axle to be driven as desired, one sprocket may be used to power the reel, while the other end serves to actuate the reel-lifting device. The unit is photographed and detail are given.

A. H. N.

959.* **World's Deepest Well Drilled in West Texas.** C. C. Pryor. *Petrol. Engr.*, April 1944, 15 (7), 72.—The world's deepest well record was passed on 28th March, 1942, and a new record is being established in Pecos County, Texas, by the Phillips Petroleum Company Rig No. 20 with the Ada Price No. 1. The well was drilling at 15,255 ft. on 12th April, when a new core was taken. This is 251 ft. beyond the previous record-depth well of 15,004 ft. The deep wildcat was spudded on 30th June, 1942, 1980 ft. from the north and west lines, section 11, block 101, T.C. Ry. survey. The well is 35 miles south-south-east of the Apco pool and 30 miles south-east of Fort Stockton and Yates. Decision to drill on the block resulted from a combination of gravity and core drill work, the area later being surveyed by seismograph. No particular depth of the well has been announced either before or since the record depth was passed. The objective of the operators is a thorough test of the oil- and gas-bearing possibilities of the structure.

Details of formations, drilling equipment, and casing draw-works are given, and the power plant is described in some detail. The power for the main mud pump is delivered by the third diesel on the draw-works by 18 v-belts. This pump is on the ground level behind the draw-works and engine structure. The size of the main pump is 8 in. × 20 in., 425 b.-hp., and is divided-cylinder valve-pot type of construction. It has an average displacement of about 700 g.p.m. and operates at about 1500 lb. The gear ratio is 5 : 1. The suction pipe is 10 in., and the discharge, which is equipped with a 2-in. relief valve, is 4" in diameter. The pump was fitted with 6-in. liners as the well depth increased. An auxiliary, or standby, pump of the same type, 7½ in. × 16 in., was also diesel-engine driven with v-belt power transmission. It has a gear ratio of 4.77 : 1, b.-hp. of 250, and intake pipe size of 8 in. and 4 in. discharge. Average mud weight used in the well was 12.1 lb., having a viscosity of about 50. The well was drilled to 12,839 with 11.3-lb. mud and changed to 12.1. Further mud conditioning was made at 12,851 ft., 12,947 ft., and 14,088 ft. Vibrating screen-type shale shakers were used. Drilling mud was slightly gas-cut at several depths during drilling. Information on the depths of possible production, however, has not yet been released. Electric logs were run at total depths of 10,008 ft. and 15,007 ft. A geophone survey was run at 15,020 ft., and a temperature survey at the same depth with two thermometers recorded 235° and 240°. In view of the hard formations drilled in the well, little difficulty was experienced with deviation of the hole. The greatest deviation occurred at 3130 ft. and was 1½°. The average deviation of the hole was about ¼°.

A. H. N.

960.* **Portable-Derrick Drilling Units.** W. A. Sawdon. *Petrol. Engr.*, April 1944, 15 (7), 154.—Portable equipments are defined and classified and general descriptions are given of main and subsidiary parts. Different types of masts and derricks are discussed. Crown blocks are of various designs, some being made integrally with the derrick or mast. A recent development is designed to provide simple over-and-over reaving of the drilling line to eliminate cross-over of the lines and insure travelling of the block in a straight line without drift or fleet angle. The sheaves are mounted in line so that all drilling lines are parallel and in the same plane as they feed to and from the block. This feature prevents twisting and drifting of the travelling block and eliminates pull-back from the centre of the hole, permitting the block to travel in a vertical line without risk of striking the bracing of the derrick or the finger-board. This particular design is made in two types, one for use where the fast line comes down inside the derrick, and the other where it comes down to the draw-works outside the

derrick. In the latter type the fast 1 line sheave is of a larger diameter than the other sheaves and is mounted eccentrically with respect to the bearing shaft of the other sheaves. The groove of the large sheave is in line with the grooves of the other sheaves on the inside of the derrick, but extends beyond on the other side to clear completely the outside of the derrick. The fast line thus operates on a larger sheave than the other lines and reduces the speed of rotation of the fastest sheave. Some equipment, such as rotary hose, is permanently mounted in the portable derrick. Swivels are, of course, of size to fit the rig, and special light swivels with short bails have been designed for use with portable rigs. Due consideration must, of course, be given to fluid passage, which must be adequate for the operation and drilling methods contemplated.

The rotary table is sometimes a part of the drilling unit (as in the case of some self-propelled outfits), and is then designed to co-ordinate with the rest of the drive mechanism. Most types of portable rigs, however, employ special small tables that are now being supplied to facilitate operations on the confined floor of the portable derrick. For example, there is the 12½-in. light-weight machine made with 36-in. spacing between the sprocket and table centre. These small tables accommodate the drill pipe now being used with the portable equipment, as this drill pipe is seldom of diameter greater than 4½ in. Table speeds up to 250 or 300 r.p.m. are practicable with many designs. Slips, bushings, etc., are described. A. H. N.

961.* **Improved Continuous Coring of Reverse-Circulated Wells.** Anon. *Oil Wkly*, 24.4.44, 113 (8), 26.—In coring a well in West Texas the reverse circulation rig is moved on the well after the hole has been drilled, pipe set, and cemented, and the large rotary moved off. The cement plug is drilled out by reverse circulation drilling, the mud column replaced by oil and drilling carried ahead with a common rock but until the first showing of porosity or oil saturation. The same mud pit used by the large rotary is emptied of mud and filled with crude oil as soon as the cement plug is drilled. A 100-brl. tank of oil is maintained as a reserve supply. When coring is to start, the drill pipe, consisting of a string of 2-in. upset tubing which will be used in the well after it is completed, is pulled, and a 4½-in. core head is installed on the short drill collar in place of the rock-bit. The core head has a 1 11⁄16-in. opening through the body of the cutter, and no core barrel is used. A core catcher is inserted just above the core head. As the core is cut, pieces of the core are washed to the surface as they break from the formation. Cores recovered range in size from thick biscuits to pieces 12 in. or more long. Twelve to 45 ft. of lime formation is cut before the cutter becomes badly worn. However, no cutter is run for more than 36 hours before being pulled.

For each round trip the amount of core that can be cut by the conventional method of coring is limited by the length of the core barrel, whereas by the reverse circulation method the only limiting factor is the efficiency of the core cutters. In soft formation this might mean a saving of several round trips with the drill pipe in coring one well. Other saving features are discussed. During the first trials at coring continuously in West Texas one difficulty overcome was the plugging of the tubing by pieces of cores and cuttings in their upward passage. This trouble was caused by lowering velocity of the drilling fluid at each collar in the tubing string, due to the slightly enlarged area in the collars between ends of adjoining pieces of tubing. This difficulty was overcome by placing ferrules into each collar, filling in the enlarged area, and streamlining the inside of the tubing from bottom to top. The tubing still becomes plugged at times, but the bridges are removed with a "rabbit" made of two swab rubbers mounted on a solid mandrel. When a bridge forms, the circulation is stopped, the "rabbit" is placed in the rotary hose, and circulation is started down through the tubing. After the bridge is knocked out, the circulation is reversed and coring is resumed. In the early experiments the tubing had to be pulled each time it became plugged. Pieces of core were used before the swab rubber "rabbit" idea was worked out. A. H. N.

962.* **World's Deepest Well.** E. Storrett. *Oil Wkly*, 24.4.44, 113 (8), 36.—The 15,255-ft. well recently drilled in West Texas is described in detail. Spudded on 30th June, 1942, it was 40 days before the 520 ft. of surface casing was set, the derrick built, and rotary drilling begun. An interesting item in the surface-casing programme is noted in the log of the water well, drilled adjacent to the deep test. The cable-

tool water-well unit logged first water sand at 532 ft., a bare 12 ft. below the base of the surface casing. The well is cased with 2200 ft. of 13 $\frac{3}{8}$ -in. casing, and 6800 ft. of 8 $\frac{1}{2}$ -in. casing, the remainder of the depth being 7 $\frac{7}{8}$ -in. open hole. Chemically controlled mud, weighing from 12.5 to 12.8 lb., is used in conditioning and maintaining hole. Five-inch drill pipe was used to 4953 ft., with five fishing jobs on account of pipe; the 4-in. string then substituted was used to 10,008 ft., drilling being suspended at this depth to run the 6800 ft. of protection string. The 4-in. pipe showed two fishing jobs on account of pipe troubles, the succeeding 3 $\frac{1}{2}$ -in. pipe two twist-offs, at 10,800 and 12,000 ft., with a third fishing job at 11,639, caused by the drill collar being dropped due to failure of pick-up nipple. Five of the eight days intervening before successful recovery of the fish from more than 2 miles below the derrick floor, were used in repairing rotary, draw-works and drum, and in conditioning the hole and mudding to the top of the fish. The only damage to the drill collar assembly from the long drop was the loss of two cones from the bit. Out of the first 593 days charged to the well, 50 were written off as fishing time, an indication of the age and condition of the equipment at the time it was being used.

Photographs and large amount of data detail the characteristics of the well.

A. H. N.

Production.

963.* **Well Spacing—Its Effect On Recoveries and Profits.** Part 2. S. K. Clark, J. S. Royds and C. W. Tomlinson. *Petrol. Engr*, March 1944, 15 (6), 71-73.—Graphs of recoveries in brls./acre-ft. vs reciprocal of square root of acres/well are plotted for different fields, and approximately straight lines with positive slopes are obtained. Different fields are discussed. Close spacing appears to increase recovery.

A. H. N.

964.* **Derivation and Application of Material-Balance Equations to Magnolia Field.** A. B. Cook. *Petrol. Engr*, March 1944, 15 (6), 222.—The author summarizes his paper as follows: Equations based upon a material balance of fluids in petroleum reservoirs have been derived in this report to calculate the volumes of oil, gas, and encroached water in the reservoir and to indicate the changes that occur in their respective volumes during any period of time following the commencement of production. The use of equations for calculating reservoir pressures on future dates corresponding with different methods of field operations, and examples showing the application of the equations to data on the Magnolia Field, Arkansas, to indicate the changes in volumes of the oil, gas, and encroached water with time have been presented. It is shown in the report that the volumes of oil and gas initially in the reservoir or the volume of water that has entered the reservoir by a certain time can be calculated, provided any two of these three volumes and certain field and laboratory data are known. The essential field data needed in the calculations include: The initial reservoir pressure and temperature; cumulative oil, gas, and water production; the average separator pressure and temperature during their production; and the average reservoir pressure and temperature on the date corresponding to the cumulative productions. The laboratory data required are P-V-T relations for the free gas, including compressibility factors, and solubility and shrinkage data for the reservoir oil, which should be collected in such a way that the liberated gas and the shrinkage of the oil between the reservoir and the gas-oil separator can be calculated in terms of volumes of stock-tank oil. The material-balance equations given in this report can also be used to calculate the volumes of fluids in some reservoirs that initially contained oil undersaturated with gas; but the calculations are complicated, and the results obtained are not always reliable.

Material-balance equations may be used to calculate future reservoir pressures corresponding to different future rates of oil production, and to show the effect of the injection of gas or water on reservoir pressures when reliable estimates of corresponding rates of gas and water production can be made. A material-balance study of petroleum reservoirs, therefore, is an aid in determining future production rates, allowable upper limit of gas: oil ratios, and the feasibility of returning produced or extraneous water or gas to reservoirs.

A. H. N.

965. Fingering and Coning of Water and Gas in Homogeneous Oil Sand. M. G. Arthur. *Petrol. Tech.*, March 1944, 7 (2), A.I.M.M.E. Tech. Pub. No. 1723, 1-16.—The control of water production and encroachment is important in both the primary and secondary phases of oil-field exploitation, in order to produce oil and gas at the lowest cost and to achieve the greatest ultimate recovery of oil from the reservoir. An actual reservoir has many variables which influence fingering and coning of water and gas, but it appears that a study of the forces involved in the case of a homogeneous sand may indicate satisfactory means of approaching the problem of controlling water production. Accordingly a theoretical analysis has been made of fingering and coning of water and gas in homogeneous sand. First a sand is considered which is relatively thin compared with its lateral extent, and a relationship is developed between the limit of approach of a water finger to a well without being drawn into the well and the corresponding operating pressure draw-down. The second condition considered is that of water or gas entry into an oil-well as a result of coning through the bedding planes. The theory and graphical solution developed by Muskat for the limit of approach of a water or gas cone to a well without its entry into the well, and the corresponding maximum pressure draw-down, are reviewed and amplified. Charts are presented which facilitate graphical solution of the problem, and this type of graphical solution is extended to the case where gas and water coning occur simultaneously.

G. D. H.

966. Flow Into Slotted Liners and an Application of the Theory to Core Analysis. C. R. Dodson and W. T. Cardwell. *Petrol. Tech.*, March 1944, 7 (2), A.I.M.M.E. Tech. Pub. No. 1724, 1-8.—The results of theoretical and experimental studies of the effect of pre-perforated liners on well productivity are presented. The analysis concerns the rectangular type of slot, either machine or torch cut. Muskat has treated this problem in a mathematical analysis of the flow of oil or gas from natural reservoirs into both gun-perforated and slotted liners. Additional and simplified analytical considerations and conclusions for the slotted liner are presented, the solutions being applicable in the laboratory as well as in the field.

The flux through a radial-flow system the inner boundary of which is a slotted liner is shown to be approximated within 1% or 2% by the following modification of the Darcy equation for radial flow:—

$$Q = \frac{2\pi kh(p_e - p_w)}{\mu \left(\log \frac{r_e}{r_w} + \frac{2}{N} \log \frac{2}{\pi\Omega} \right)}$$

where N is the number of slot rows, and Ω is the open fraction of the pipe. The small magnitude of the effects of constricting the flow of oil, gas, or water into liner slots indicates that decisions regarding liner slot sizes should be based primarily on considerations such as sand retention and strength.

The analytical methods can be applied to permeability measurements on mounted core samples to show the effects of sealing wax or other obstructions on the end faces of the samples. In a permeability measurement on a core sample the length of which is approximately equal to its diameter or side face, a 30% covering of this face may cause only a 5% error in the result, whereas a 10% covering may cause only 1% error. These theoretical indications are confirmed by experiment

G. D. H.

967.* Effect of Well Spacing on Recoveries and Profits. S. K. Clark, C. W. Tomlinson, and J. S. Roysds. *World Petrol.*, March 1944, 15 (3), 54. See Abstracts No. 963 and 972.—This is an abridged form of the full paper.

A. H. N.

968.* Water Flow Chart for Designing Salt-Water-Disposal System. W. A. Pearce. *Oil Wkly*, 6.3.44, 113 (1), 16-17.—This chart is based on the Hazen-Williams formula which was rearranged as follows:

$$h_f = 1000 \sqrt[0.54]{\frac{v}{1.318 c^r 0.63}}$$

where h_f = loss of head due to friction in ft./1000 ft. of pipe; v = mean velocity in ft./sec.; c = Hazen-Williams coefficient of roughness; r = hydraulic radius of pipe in ft.

Although a roughness coefficient or "c" of 140-150 is claimed for asbestos-cement pipe, the material commonly used in modern salt water gathering lines, in order to allow a reasonable factor of safety this chart was constructed using a value of $c = 120$. However, conversion factors from this value of "c" to other values from 100 to 140 are given.

A. H. N.

969.* Illinois Oil Sands Found Suitable For Artificial Water Flooding. F. Squires and A. H. Bell. *Oil Wkly*, 6.3.44, 113 (1), 18.—This is a synopsis of investigation No. 89, Illinois State Geological Survey, Urbana, Illinois. In the summary it is stated that the high lights of the natural floods are these: water came from a lower formation to cause the Partlow flood. At Flat Rock, water brought in so much oil from outside the area that the drilled part of the sand produced more oil than it contained originally. The Buchanan flood is noteworthy for its record-breaking production. The Dennison territory may have been made permeable as a weathered "high" on a cross-fold. Applegate flood water moves unaccountably in the opposite direction from that of the otherwise closely related Dennison flood. Oakland City is unusual in that the flood peaks attained, and sometimes surpassed, the peak of heights of the original flush production. The Murphy flood was abandoned during the period of lag before the flood reached the pumping wells. The Dupo flood water showed the highest fluid levels at the top of the structure.

As far as this paper is concerned, natural floods have no value unless their evidence points to the probable success or failure of applied floods on other Illinois sands. It may be argued that natural floods move under their own power, and that only areas available for purposeful flooding are sands now devoid of all reservoir fluid movement. But all sand and lime oil-producing strata in Illinois are under some edge-water pressure, and edge-water encroachment differs only in speed. Given enough time, all Illinois producing sands would experience edge-water encroachment. Even Robinson sand wells, when left unpumped for a year, fill up to 200 ft. or more with fluid. Only edge-water pressure could maintain such heights of fluid column. Then how do these natural floods indicate a probable success for artificial floods? First, they demonstrate that water moves oil. Second, they demonstrate that many Illinois sands are continuously permeable over large areas. Third, when a natural flood at Sandoval was supplemented by an artificial flood introduced within it, this applied flood further increased production. It was proved that each kind of flood produced the same kind of result in increasing oil production. The efficiency of accidental floods which occur in sands under edge-water drive is further evidence that applied floods would be successful in edge-water encroachment territory. Because of these facts, it is logical to recommend that purposeful flooding in advance of natural water-drive be applied in pools that are already under edge-water pressure.

A. H. N.

970.* The Drowning and Revival of Gas Wells. Part 1. S. T. Yustor and K. J. Sonney. *Petrol. Engr*, April 1944, 15 (7), 61-66.—The revival of intentionally or accidentally drowned gas producing or injection wells is of prime interest to oil and gas producers. Water from leaky casing, excessive coning due to too rapid withdrawals, necessary drowning to control a wild well or make repairs, may all cause an almost permanent harm to a gas well by reducing its capacity. This is not only a problem confronting gas men, but is of great interest to oil producers, as gas wells play an important part in cycling operations, pressure maintenance, repressuring projects, and in secondary recovery by air and gas drive. In all cases the effect of the presence of water is to retard the flow of gas from or into the formation.

A study of the causes of the extra resistance is made with detailed explanation of Jamin action. The study shows that the problem can be solved by any of the following methods: (1) Removal of water from the vicinity of the well-bore by increasing the pressure differential between the well-bore and the formation to overcome the Jamin action and force the water ring far into the sand where its thickness and effectiveness in plugging will be greatly reduced. (2) Reducing the surface tension of the water in the sand and thereby decreasing the pressure necessary to move the water. (3) Dilution of the water in the sand with water-miscible high-vapour pressure solvent resulting in a more volatile fluid mixture that may be removed by evaporation. (4)

Combination of the above methods. With the above possibilities in mind, a series of experiments was made in the laboratory to test their practicability.

The apparatus and procedure are described and detailed results are given.

A. H. N.

971.* Determining Well Diameter. L. C. Uron. *Petrol. Engr*, April 1944, 15 (7), 119.—The diameter of a well and the casing in it will influence the cost and efficiency of operations both during the drilling of the well and its subsequent period of production. Small-diameter wells can be cased less expensively, and this is the principal advantage of the slim hole. Under favourable conditions, the cost of drilling may be less for the small hole, and it may be drilled more rapidly. This is not necessarily true, however, and if drilling conditions are difficult, the larger-diameter hole may be drilled less expensively and more rapidly. If intricate equipment must be manipulated in the well during the course of drilling, as in coring, fishing, and certain types of cementing, operations, large-diameter wells are advantageous. In the subsequent period of production, the large-diameter well is preferable from every point of view. It develops superior productive capacity, greater ultimate recovery, and it produces with lower gas-oil ratio and, therefore, more efficiently from the energy consumption standpoint. Large-diameter casings and well equipment exert less back pressure on the producing formation, permitting flow with lower pressure differential, hence with greater efficiency. A large-diameter casing permits the use of larger size of pump of greater capacity than is possible in a well of smaller diameter. When tubing has to be pulled, liners, screens, or gravel-packs placed, "shooting," acidizing, or clean-out operations conducted, the larger-diameter well is advantageous. If bottom water or edge-water has to be excluded, in a certain producing interval, or if the well has to be deepened to a lower zone, or other work-over operations performed, the large diameter well permits completing the work with greater certainty and efficiency.

The influence of diameter on drilling, production, and on proration and the practical limitations on diameters are discussed separately. It is concluded that for a given set of conditions, there is an optimum size of well and well-casing diameter that will be most advantageous. This is not necessarily the smallest practical working diameter. Small-diameter wells are more economically cased than those of larger diameter, but they may be more costly to drill. From the production and maintenance standpoint the large-diameter well has every advantage. The problem is largely an economic one and the operator must balance any possible economy in drilling and casing the smaller-diameter well against its greater maintenance and operating cost and lower production efficiency. Proration practices have a controlling influence in determining well and casing diameter under some conditions. The economy of small-diameter casing and the high production efficiency of the large-diameter well may be combined by drilling a small-diameter hole down to the cap rock and then reaming to a larger size through the reservoir rock, gravel packing a perforated liner of small size in the enlarged portion of the hole.

A. H. N.

972.* Well Spacing—Its Effect on Recoveries and Profits. Part 3. S. K. Clark, J. S. Roys, and C. W. Tomlinson. *Petrol. Engr*, April 1944, 15 (7), 124.—Further theoretical analysis of the problem and citations of practical examples are made in the third part of this paper. In summary it is concluded: (1) That well spacing does affect the recovery of oil from a reservoir; that increased well density results in increased recovery; and that within practical limits there is a definite mathematical relationship between the well spacing and the recovery. (2) That arbitrary wide spacing regulations impose definite handicaps on the adequate exploitation of small accumulations on sharp folds or against faults, and tend to increase the ratio of dry holes to producers in all cases. (3) That even from the standpoint of cost/barrel of crude recovered and of profit/dollar expended, it is not invariably true that the wider of two spacings is the more economical. For a given set of conditions there is a certain spacing that is the most economical one, and either a closer or a wider spacing will involve higher unit costs and lower unit profits. Certain other conclusions may legitimately be reached from these basic ones.

Although nothing herein is to be construed as an argument against the adoption of improved production practices in the attempt to obtain greater recoveries at lower costs, it would appear that a simple and highly effective method is being neglected

when the possibilities of close spacing are ignored. Since more wells mean increased recovery, there is a strong incentive to work towards lower development costs by utilizing every expedient to reduce the cost of the individual well rather than by widening the spacing. When wide spacing is temporarily adopted either because of material shortages or lack of information regarding the reservoir, it is highly desirable that a pattern be selected which will permit increasing the density by drilling intervening wells later. With the existing urgency for an augmented crude supply, it would seem logical to resort to the drilling of additional wells in fields now having wide spacing that are favourable to closer spacing, especially since such drilling would add potential production and reserves, with an absolute minimum of equipment as contrasted with the material requirements for exploiting new fields.

Finally, since optimum well density varies with crude prices, it would appear to be in the public interest for crude oil prices to be increased to levels that would encourage relatively close well spacing so long as the present downward trend of discoveries in relation to production continues.

A. H. N.

973.* Pilot Controlled Gas-Lift Operation. R. O. Walton. *Petrol. Engr*, April 1944, 15 (7), 166.—The system described is a unit gas-lift installation. The system, employing the pilot-actuated valves and surface controller, is unusual in that it is adaptable to any well, regardless of depth or type, if fluid is to be lifted and gas under pressure can be made available. Operation of the surface controller is familiar to operators experienced in gas injection. The surface unit is a rugged time-actuated motor valve in conjunction with an adjustable mechanical-volume control valve. By slight adjustments on the time control wheel, any frequency of on-and-off periods may be selected. After these adjustments are made, additional control is available by use of the volume control valve. The subsurface pilot-actuated valves are mounted on 3-ft. tubing subs, having full tubing openings through them. Their pilot valves are set for predetermined operating pressures before being run in a well. The construction of the valves is such that each valve will pass any volume of gas admitted to the casing annulus, after the casing pressure reaches the operating pressure of that particular valve. For example, if the pilot of one of the valves is set for an operating pressure of 300 lb., the valve will remain closed until the casing pressure increases to 300 lb.; at that point, the valve will open. If the controller is set to inject gas at the rate of 10,000 cu. ft./hr., the subsurface valve will pass gas from the casing annulus into the tubing at the same rate until the surface controller closes and allows the pressure to return to 300 lb., in turn closing the valve. The casing pressure will then remain at 300 lb. until the surface control valve opens for the next cycle of operation.

Where more than one valve is needed to unload a well from its static fluid level down to a working fluid level, the valves are set in the well so that the highest pressure valve is at the top of the string and each successively lower valve is set with a slightly lower operating pressure. Thus the valve having the lowest operating pressure is situated at the bottom of the string. This is done: First, to maintain the casing pressure lower than the operating pressures of the upper exposed valves and will keep them closed; second, the lower valves under fluid will remain open and allow fluid to bypass from the casing annulus into the tubing. After an installation of this kind is made, the gas is injected and the casing pressure is allowed to reach the operating pressure of the top valve. At this point the desired frequency of operation is set on the surface controller and the well will produce down to its proper working fluid level without additional attention. This feature of automatic work down in a well that is logged up will save many man hours to the operator.

The valve is fully described with section view illustrating its main features. Several successful installations are described.

A. H. N.

974.* Economics of Secondary Recovery. R. C. Earlougher. *World Petrol.*, April 1944, 15 (4), 42-44.—A strong plea is made to increase oil prices so that secondary recovery projects become profitable investments. The summarized conclusion is that by means of secondary recovery the country's potential oil reserves are increased by approximately 20%. The ways and means of producing this secondary oil are known and are available to the industry. The one major item that is lacking is the incentive required to induce operators to put their money into the ground with the hope of getting it back together with a substantial gain.

A. H. N.

975.* **Drilling Costs Have Risen Sharply Since 1941.** B. Mills. *World Petrol.*, April 1944, 15 (4), 45-47.—The exact increase in drilling costs of March 1944 over those of September 1941 are difficult to determine, since basic factors have changed considerably. Where shutdowns from lack of labour and materials were very rare during 1941, they were common during the past year. Next to a bad fire or blow-out, a shutdown is perhaps the most expensive time-loser a drilling contractor encounters. Shutdowns are reflected in greater operating costs in every department. In analyzing the rise in drilling costs during the past two and a half years it is not easy to separate the various elements so as to show that labour accounted for a certain specific percentage, materials another percentage, and miscellaneous items still another percentage, to make up the total increase. Increased labour costs are so tied into "losses from shutdowns" that labour actually becomes part of the shutdown loss. The fact is that all phases of drilling activity have become more expensive and have combined to bring about a substantial cost increase. Whether "labour", "shutdowns" or other reasons are to be held chiefly responsible is incidental to the main issue. Costs likewise vary with every set of conditions encountered. One drilling contractor will state that his current costs are 30% above those of late 1941; another will insist his costs are up 35% over the same period, while still a third will place his cost increase at 40%. Each contractor may be correct in his estimate, as one operator may be less seriously affected than another by a given set of factors. In many cases the increase in drilling costs apparently has been entirely too high for a drilling contractor to absorb, but one thing or another has permitted him to continue operations.

The following is the average of 10 operators' evaluations of percentage increase in drilling costs since 1941: Labour, 38%; Materials, 20.5%; Derrick, 39%; Transportation, 42.5%; Fuel, 18.5%; Shutdown frequency, 31.5%; Miscellaneous, 40%; Overall, 33%.
A. H. N.

976.* **Equivalence of Material Balance Equations For Calculating Original Oil Reservoir in Place.** S. J. Pirson. *Oil Wkly*, 3.4.44, 113 (5), 28.—It is generally believed by reservoir engineers that the various formulae which have been proposed for the calculation of oil originally in place in a reservoir are not equivalent and, in fact, sample calculations illustrating the use of the Schilthuis and Katz methods in particular cases would indicate that the two methods give results varying by as much as 20%. It is shown that, on theoretical grounds, the methods of Schilthuis and Katz are identical, and that the apparent discrepancy is due to the fact that the method of Katz includes more numerous steps in the calculation, thereby affording more chances for cumulative errors to register an appreciable effect in the final answer. In order to prove the theoretical equivalence of the Schilthuis and Katz methods, it is required to express both methods in a consistent system of notation. One of the limitations of the various articles thus far published on the subject-matter of reservoir engineering has been the lack of unity in the system of notation used. A system which has already received some acceptance in the literature, and which has the advantage of simplicity and of an actual physical meaning, is used.

One unfortunate feature of the Schilthuis material balance equation is the presence of the water encroachment term in the numerator; considerable thought and work have been expended without success by the writer and his associates in trying to eliminate this term from the equation by the simultaneous solution of various material balance equations obtained on various bases. The Schilthuis equation is essentially a gas material balance. It was thought that material balance yielding different equations could be found by writing a material balance on oil, or by writing a volumetric balance, or by establishing a density balance of the fluids, etc. When any of the above are successful, they always yield a formula equivalent to the Schilthuis equation, or resolve themselves into an identity equation from which all terms cancel out.

A. H. N.

977.* **Physical Properties of a Reservoir and of Reservoir Fluids.** E. N. Komler. *Oil Wkly*, 10.4.44, 113 (6), 26.—The study of reservoir problems usually involves the physical properties of the producing formation. The two important physical properties of the formation are its permeability and its porosity. The permeability defines the fluid-carrying capacity of the reservoir under a given set of producing conditions. Porosity defines the fluid-holding capacity of the reservoir. The porosity

is actually the amount of void space between the sand grains or in crevices or other openings in limestone formations. The amount of fluid which can be stored in the reservoir will be equal to this pore space. In addition to these two physical properties of the reservoir, it is necessary to know whether the fluid occupying the pore space is oil, water, or gas. The determination of the character of the fluid in the reservoir is known as a saturation analysis.

Porosity and permeability characteristics of formations are studied in some length. The effect of gas evolution on the characteristics of crudes is also studied, particularly the effects of shrinkage and viscosity. The effect on viscosity may be of importance when determining whether or not reconditioning work should be done. The viscosity characteristics of the crude oil in both the saturated and unsaturated conditions are important since they, together with the viscosity of the water, will determine the producing characteristics of the reservoir later in the life of the field. There are not much data available in the literature on the effect of solubility on viscosity. Evonger and Muskat have given some data on viscosity variation at pressures up to 2500 lb./in.², Hocott and Buckley also give data for a crude oil up to pressures of 725 lb./in.². In each case a variation is found. One figure shows that an oil saturated with about 525 cu. ft./barrel will have its viscosity increased from 0.6 to 1.4 centipoises when the pressure is reduced to atmospheric. Another figure shows a comparable increase on an oil having a lower solubility. If a test were made on a reservoir when new (assuming all gas stayed in solution) and then near the end of its life, a reduction of 2 to 1 in productivity index factor, due to change in viscosity alone would be indicated.

A. H. N.

978.* Effect of Physical Properties of Oil Reservoirs on Flow Performance. E. N. Kemlor. *Oil Wkly*, 17.4.44, 113 (7), 17.—The rate of production from a green well can be expressed analytically as:—

$$Q = C(P_m - P)$$

where Q = production rate in barrels/day, C = well characteristic or productivity index factor, P_m = reservoir or closed in bottom-hole pressure, P = pressure at sand face when well is producing at rate Q . This productivity index factor or well characteristic is dependent on the dimensions of the well bore, on the properties of the fluid produced, and on the size and permeability of the reservoir. This factor can be expressed analytically as follows:—

$$C^1 = \frac{2\pi kbg}{\mu \log_e \frac{r_e}{r_w}}$$

C^1 = productivity index factor in ft.³/sec./lb./ft.². Where k = sand permeability in ft.², b = sand or formation thickness, ft. μ = absolute viscosity in lb./ft.-sec. r_w = well bore radius, ft. r_e = well drainage radius, ft. g = 32.3 ft./sec.².

From this equation it can be seen that the ability of a well to produce will increase directly in proportion to the permeability k . Permeability is a measure of the resistance of the formation to the flow of fluid through it. The permeability will depend on the type of formation and on the size and method by which the pore spaces in the formation are connected. In the case of a sand reservoir it depends on the amount and location of cementing material as well as on sand grain size and packing. The higher the permeability the greater will be the capacity of any given well to produce. Permeability as considered from a production standpoint can be reduced by forcing material back into the formation which will either fill up the pore space or seal off pores immediately around the well. Such procedures are used in squeeze cementing and water shut-off work. Likewise, anything which will increase the size of the connection between pores will increase the permeability and the capacity to produce. Such practices as acidizing are to increase the size of the pore spaces immediately in the vicinity of the well bore, and, therefore to increase the effective permeability of the well. The effect of shooting, insofar as permeability is concerned, is to produce fractures of sufficient size effectively to increase the average permeability in the vicinity of the well bore.

The paper consists of a discussion on the various factors appearing in the formula for productivity index.

A. H. N.

979.* Water Flooding in the Bradford Field. A. E. Booth. *Oil Wkly*, 24.4.44, 113 (8), 30. *Paper Presented before American Petroleum Institute.*—A brief history of production of the Bradford field is given, covering the period 1870–1944, and is followed by a description of the co-operative research project on the field. This research project has been primarily responsible for much more progressive effort than that with which it is credited. It has stimulated the interest in coring and in the information thus obtained; it is responsible for publication of a brochure on treatment of water for injection purposes that is without a peer; it has aided in the ideas of wider spacing and higher pressures; it brought out the original suggestion from which developed the present method of selective shooting; it has educated operators in the study of porosity, permeabilities, oil and connate-water content of the oil-sand; and it has instigated much progressive thought and experimentation outside the project proper. Probably the most important single item, and the one with the greatest chances of substantial improvement in the economic picture of water flooding, is the increased recovery of the residual oil content of the sand. This is being approached directly through better water conditioning and higher pressures, accompanied by higher average velocities for the water front, together with coring and selective shooting. High pressures mean plant pressures of 1000–1500 lb. and sand face pressures of up to 2000 lb. Coring is done by three methods. Chip coring, cable-tool core barrel, and the diamond core. The methods are progressively more expensive in the order named and, in the same order, furnish more detailed information on the strata cored.

Several items are discussed separately—selective shooting and plugging, pumping practices, etc. A. H. N.

980.* Reservoir Data and Production Problems. E. M. Komler. *Oil Wkly*, 24.4.44, 113 (8), 72.—Previous papers in this series dealt with data and properties obtained from core analysis. These properties have, whenever possible, been expressed in terms of quantities on which it is possible to obtain information. Since the physical examination of an oil reservoir is limited to that of a few cubic inches of core, the facts obtained from core analysis must be supplemented by performance data. These performance data again are limited to production information and bottom hole pressure data. Where reasonably accurate information on these items is available, estimates of future behaviour can frequently be made. Those fields where there is little gas in solution in the oil and which are subjected to a water drive will behave reasonably well with the ideal systems. In these conditions the behaviour is similar to that of liquid flow. While formulæ as normally given are for oil wells, they will apply equally well to salt-water disposal or water drive input wells. One important application of reservoir information is to the study of the effect of completion methods on performance. Where incomplete tests are made before final producing equipment is installed, the effects of completion operations may be difficult to predict. For example, a pumping test with temporary equipment will not necessarily in itself give sufficient data to show whether or not a well should be acidized or shot. When bottom hole pressure or equivalent fluid level data are taken in connection with production data, much information can be obtained on such tests.

These tests are discussed.

A. H. N.

Transport and Storage.

981.* Protective Storage at Pipeline Terminals. H. J. D'Aragon. *Petrol. Engr*, March 1944, 15 (6), 146–150.—Attention is directed to the deterioration in quality of gasoline as well as actual physical loss due to evaporation during storage. Evaporation losses from conventional storage tanks with fixed cone roofs may be classified as those due to windage, breathing, boiling and filling. The latest information on losses, derived from actual tests covering a wide range of capacities and made under actual operating conditions, disproves the widely used assumption that evaporation loss is directly proportional to vapour pressure of product and surface area of liquid. Average standing evaporation losses, based on tank capacity, and filling losses based on throughput and vapour pressure of product, obtained as a result of tests conducted by 14 large oil companies on cone roof, gas-tight storage tanks equipped with con-

ventional pressure and vacuum valves, are provided. At pipeline terminals the greatest proportion of total loss is generally due to windage and breathing. The former can be eliminated by installation of pressure and vacuum valves and ensuring that roof seams and connections are gas tight. Breathing losses may be reduced by (a) use of variable space or expansion roofs, (b) diminishing temperature variation by suitable paints, water sprays, etc., (c) connecting the vapour spaces of several cone roof tanks to a gas reservoir or the vapour space of one tank fitted with a variable space roof. The manner in which such a system can be utilized to minimize breathing and filling losses is described and illustrated.

The method can be applied to the storage of a variety of products simultaneously; one company has manifolded tanks containing sweet and sour crudes, without evidence of contamination. Except where filling and emptying rates are greatly unbalanced, protection is thus provided with a minimum of expenditure. Floating roofs eliminate filling losses but must be applied to each tank requiring protection, and cannot be utilized in the balancing system described above.

R. A. E.

982.* Mechanical Couplings for Pipe Lines. G. E. Szekely. *Petrol. Engr.*, March 1944, **15** (6), 175-180.—A description is given of the construction, installation, and general application of the bolted positive-seal type of mechanical coupling. Improvements which have taken place in gaskets, and their protection from deterioration due to contact with oil products are outlined. The ability of such couplings to accommodate misalignment and absorb vibration, settlement, expansion, and contraction makes them useful for a variety of purposes other than pipe connections—e.g. tank connections, valves, pumps, engines, and compressors.

The couplings are also eminently suitable for repairs, replacements and insertions. Adaptations of the normal design for specific purposes are: long couplings where extensive settlement or other movement may be expected, insulated couplings for minimizing electrolytic corrosion in certain systems, reducing couplings, boltless couplings and fittings which are particularly suitable to small pipings in close quarters, e.g., for engines, pumps, compressors, etc.

Mechanically coupled pipelines no longer required can readily be picked up and relaid with a minimum expenditure of labour and material, a factor of importance at the present time.

R. A. E.

983.* Corrosion of Buried Metals and Cathodic Protection. M. C. Miller. *Petrol. Engr.*, Part I, March 1944, **15** (6), 208-212. Part II, April 1944, **15** (7), 101-110.—The fundamental causes of corrosion of buried metals are discussed, with illustrations of current flow and extent of corrosion which may be expected with various combination of metal and soil. Consideration is given to corrosion caused by dissimilar metals, dissimilar soils, and by non-uniformity of metal used for underground structures.

R. A. E.

984.* Concrete Tanks for Storage of Petroleum Products. C. G. Munger. *Petrol. Engr.*, March 1944, **15** (6), 277-278.—The pre-stressed reinforced concrete design for cylindrical tanks by the U.S. Navy Department has greatly increased the effectiveness of concrete tanks for many purposes. Such tanks require less steel than conventional concrete tanks and are practically free from cracking as the steel reinforcing is under tension and the concrete under compression. Their principal application is for petroleum products, so that coating of the tanks with a suitable material which will prevent seepage of oil through the concrete, be non-reactive to new concrete surfaces, be insoluble in oil products and eliminate all possibility of tank corrosion or product contamination, is essential. A new thermoplastic coating compound that meets these requirements and has been applied with success to a large number of oil tanks and to the interior of concrete ships, is known as Amercoat. It is manufactured by the American Pipe and Construction Company and is a compound of vinyl synthetic resins combined with solvents and non-toxic, non-reactive pigments.

The compound may be applied cold with spray or brush to give a satisfactory impermeable coating. Results of tests with a variety of products indicate the satisfactory inertness, adhesion, resistance to abrasion, etc., of the coating material.

R. A. E.

985.* Shipment of Packed Petroleum. 2. Fire Prevention and Fire Fighting. Anon. *Petroleum*, May 1944, 7 (5), 70.—This part briefly relates several warnings relative to fire prevention. Leakage from containers in ship holds and the risks therefrom are discussed. The necessity of adequate ventilation is emphasized and precautions relative to ships' ventilators are given.

The National Fire Service provide a fire-fighting unit whenever a ship loads packed petroleum and the Sea Transport Division supply all ships they charter with fire fighting equipment selected in accordance with the type of cargo being carried. This equipment is self-contained and does not have to rely on engine room pumps or fixed deck service lines. The National Fire Service train ships' crews in the operation of such equipment. Thirty illustrations from the official film on this subject are shown.

W. H. C.

Cracking.

986. Patent On Cracking. International Catalytic Oil Processes Corp. E.P. 557,207, 10.11.43 Appl. 20.11.41—During the catalytic cracking of hydrocarbon oils, such as gas oil, the oil is separated into a light and a heavy oil and a fresh or regenerated silica-magnesia catalyst is suspended in the light gas oil fraction. The suspension is continuously fed to a reaction zone operated at a temperature above 900° F. Catalyst is separated from reaction products and employed without regeneration in the cracking of the heavier gas oil fraction.

H. B. M.

Polymerization and Alkylation.

987.* Propane Refrigeration for Alkylation Units. Anon. *Oil Gas J.*, 13.4.44, 42 (49), 128.—A description is given, with flow sheet and photographs, of the operation of propane refrigeration units, of capacity 12,000 B.T.U. per hr., installed to obtain temperatures of 50° F. or below in the reaction zone of sulphuric acid alkylation plants. Make-up propane is obtained from the alkylation unit depropanizer, being treated with caustic soda to remove SO₂, and dried with calcium chloride, while air is removed by continuous venting from a small refrigerator vessel or pipe, into which is bled a small quantity of vapours from the receiver. Oil is similarly removed by bleeding a small quantity of liquid from the propane accumulator drum back to an alkylation unit surge drum, operating at a lower pressure than the propane drum. The propane flow is similar to that for any other refrigerant, the compressors being designed for a suction pressure of about 57 lb. per sq. in. when propane is evaporated at 35° F. and a discharge pressure of 175 lb. per sq. in. Water at a maximum temperature of 80° F. is used to condense the propane at 90° F. The compressors are operated on natural gas or propane if required.

C. L. G.

Refining and Refinery Plant.

988. Thiophenol in Cracked Gasoline. G. S. Windle. *Refiner*, February 1944, 23 (2), 41-45.—The paper deals with the problems which arose in the refining of Bahrein cracked naphtha due to the presence of acidic sulphur compounds, and discusses the methods of isolating, identifying, and eliminating thiophenol. The effect of thiophenol on gasoline quality and its behaviour in sweetening operations are fully explained.

In the caustic treating of naphtha for hydrogen sulphide removal, the extent of removal of thiophenol is a function both of the quantity of caustic applied and of the type of contacting equipment used. Originally the caustic scrubber at Bahrein was the conventional single-stage conflow type of equipment. No difficulty resulting from failure to remove thiophenol was observed, but the proportion of caustic applied was excessive. By installation of a two-stage counterflow caustic scrubbing unit the efficiency of the caustic in hydrogen sulphide removal was improved as expected, but the extent of thiophenol removal was reduced. Mixtures of sodium sulphide and sodium acid sulphide are not suitable for thiophenol removal. The analyses for the acid oils in typical first- and second-stage caustic reactors over an extended period showed an abrupt increase in concentration when the caustic was first used, followed by a steady decline from 1% to nil as the caustic availability was reduced from 70%

to 40% through hydrogen sulphide absorption. The non-volatile neutral oil remaining after steam distillation of the acidified caustic was constant at about 0.6% within this range, but disappeared within the range of 40 to 25% caustic availability. These variations in concentration conform to the principles of selective absorption and are consistent with the relative strengths of the acids removed by the caustic. Thus, when using two stages of contacting and rejecting caustic at 40% availability or less, hydrogen sulphide absorption in the first-stage caustic tends to reject all the acid oils back into the gasoline phase, thus throwing a larger burden of acid oil removal on the second stage of the system.

To avoid excessive reagent consumption and still obtain the desired removal of thiophenol in the caustic neutralization, the system finally adopted at Bahrein refinery comprised lime treatment of the clay-treated naphtha for hydrogen sulphide removal, followed by caustic contacting for removal of the thiophenol. A. H. N.

989. Design of Foundations For Elevated Towers. S. Garnett. *Refiner*, February 1944, 23 (2), 46-54.—When the bottom of a tower is set at an elevation of 15 feet or more above ground it is not economical to use a solid concrete pedestal as a foundation. In such cases the tower is either fitted with a skirt to bring its base to approximately ground level, or it is supported on a frame of either structural steel or reinforced concrete. This article deals with the design of a reinforced-concrete frame foundation for such conditions.

The principles of design and calculations of different loads under different loading conditions are given, and then an example is worked out in detail to illustrate the methods. A. H. N.

990. Wooden Core in Storage Reel Protects Welding Cable. Anon. *Refiner*, February 1944, 23 (2), 77.—One company welder, in attempting to determine why his cables were prematurely becoming hard and brittle, even though they were handled carefully and kept wound on small spools, discovered the cause to be associated with the fact that the spools had steel cores. Lending weight to this evidence was the observation that the inner layers of the cable, even though much less used and seldom exposed entirely to weather, were the first to show evidence of stiffening and embrittlement. Not only did the heavy rubber insulation crack, but the copper wire itself would ultimately become brittle and eventually would break within the insulation upon being used or flexed in removing it from the reel. Believing that the intense field set up by the direct current around the steel core in the centre of the spool might be creating heat and other electromagnetic conditions, causing rapid deterioration, a wooden core was turned out and mounted in the reel in place of the steel core. Almost immediately, a change was noticed. The majority of the foregoing troubles vanished, and the cable, since the cores were changed, now remains soft and pliable down to the last turn in the centre of the reel. This prolonging of the useful life of conductor cables has been the cause of substantial savings in electric welding equipment upkeep. A. H. N.

991. Maintaining Wartime Compressors At Peak Operating Capacity. B. Thiel. *Refiner*, February 1944, 23 (2), 78.—Hints are given on maintenance of compressors. A. H. N.

992. Refinery Cleaning Problems. R. W. Mitchell. Part 1. *Oil Gas J.*, 10.2.44, 42 (40), 70; Part 2. 17.2.44, 42 (41), 65.—Cleaning problems on a refinery are outlined, and the methods and materials that enable cleaning to be satisfactorily and economically accomplished are discussed. Cleaning materials should be safe to handle and to use. Caustics have poor wetting and penetrating powers, but a range of materials can be obtained which have these properties in a marked degree. They are also easily washed off after their action and are less hazardous in use. The cleaning methods should be simple and involve the least technical supervision. The cost of the cleaner relative to time and labour should be studied. A cleaner costing twice as much per pound as another can, and often does, clean more feet of surface or so many more units per pound consumed, that it is clearly most economical. Charts are given showing (1) the pH range of industrial cleaners; (2) Solutions containing

buffer solutions showing a much wider range of concentration for a given pH range than unbuffered solutions; (3) A curve showing the relationship between pH of the cleaning solution and the time required for thorough cleaning of surfaces.

The cleaners proscribed for the cleaning operations are: (a) heavy duty cleaners 8 oz./gal. or (b) 50/50 solution; (c) solvent types of soaps, 1 teaspoon/gal.; (d) inhibited acid cleaner, 33% solution; (e) compounded alkaline cleaner, 6 oz./gal. pH 12.5.

Sections dealing with the cleaning of storage tanks interiors using type (a) cleaner; exteriors, type (c); paint removal, type (a); heat exchangers, type (a); plant columns, type (a); coolers, type (d); barrels, type (a); pipes and equipment types (a) and (b); floors, type (c).

The methods of application, temperatures employed and subsequent removal of the used cleaner, are described.

W. H. C.

993. Soda-Methanol Reagent Removes Mercaptans Quantitatively. F. C. Moriarty. *Oil Gas J.*, 30.3.44, 42 (47), 82, 85.—At a refinery at Kovin, Mont., the doctor treaters have been replaced by a Unisol treating unit in which mercaptans are removed from Dubbs cracked gasoline by extraction with a concentrated solution of caustic soda containing methanol. When using doctor solution, a loss of 1 to 2 octane numbers resulted and it was necessary to add 1.1 c.c. of T.E.L. to produce 74 O.N. gasoline, and this could not be reached if any substantial quantity of straight-run gasoline were present, nor could an 80 O.N. gasoline be produced. With the Unisol process, the mercaptan content is reduced from 0.06–0.11 to 0.007%, the total sulphur being reduced correspondingly, giving a large increase in T.E.L. susceptibility, as well as an actual increase of one half O.N. It is now possible to reach 80 O.N. with 3 c.c. of T.E.L., while a blend containing 10% straight run can be brought to 75 O.N. with 0.65 c.c. of T.E.L. Treating losses have been reduced by about 1.5%. The reagents cost below 0.5 cent per brl., and labour costs are low. Apart from some corrosion, no process difficulties have been experienced even in the very low temperatures prevailing. The product has an excellent inhibitor susceptibility, 0.007% of U.O.P. inhibitor No. 4 increasing the induction period by approx. 250 minutes, while the copper dish gum content is below 10 mg. The process shows a total saving including the appreciated value of the gasoline, of \$30,000 p.a.

C. L. G.

994.* Unisol Process for Treating Gasoline. F. C. Moriarty. *Petrol. Engr.*, April 1944, 15 (7), 150–152.—A Unisol treating unit has been operating in the refinery of the Big West Oil Co., Montana, processing approximately 550 brl. a day of cracked gasoline from a Dubbs unit. Previously, doctor treatment was employed involving reduction in octane rating of the gasoline, a rather heavy treating loss, and rendering production of ethyl grade gasoline (then 80 octane number) impossible. Installation of the Unisol unit enabled reduction in mercaptan content of the gasoline from 0.06–0.11% to an average of 0.0007%; production of 80 octane gasoline with 3 mls. T.E.L. per gal. from the cracked gasoline; production of 74 octane gasoline when including all straightrun gasoline produced; a lower T.E.L. addition than previously required for the treated cracked gasoline to give 74 octane grade; reduction in treating losses. The Unisol treated gasoline usually becomes doctor sweet, or nearly so, after standing a few days in storage. There have been no process difficulties, but corrosion in the bottom of the caustic recovery section, made of carbon steel, is severe and requires considerable maintenance and repair. This will be eliminated when nickel-clad steel is available. Methanol losses are less than 2 gals. a day and total reagent costs below $\frac{1}{2}$ c. per brl. treated.

Cost of installation was \$20,000, and total savings plus appreciation in product values are estimated to be \$30,000 a year.

R. A. E.

995.* Mercapsol Process for Gasoline Treating. B. L. MacKusick and H. A. Alves. *Oil Gas J.*, 13.4.44, 42 (49), 126.—In the Mercapsol process, mercaptans are removed from gasoline by countercurrent extraction with caustic soda solution containing as solutizer, high boiling tar acids, naphthenic acids or cresols etc. Regeneration is effected by diluting and boiling to the original volume. A 5000-brl. per day Mercapsol treating unit has been in operation for 4 years with successful results, reducing mer-

captan sulphur from straight run Penna gasoline and cracked gasoline by 77% and 93% respectively, and giving a slight increase in clear O.N. and definite increase in lead response. The process removes mercaptans in one operation, using a low treating ratio, while the solution is cheap, readily available, and may be regenerated indefinitely, and is no more dangerous to handle than caustic soda. Operation is simple, and operating conditions mild, while existing doctor or caustic washing equipment can be readily converted. A flow sheet is given together with refining data on cracked gasoline from a pilot plant and on straight-run and cracked gasoline from the commercial unit.

C. L. G.

996.* **Allies Aviation Fuel Needs Supplied by More than 170 Units.** Anon. *Oil Gas J.*, 13.4.44, 42 (49), 157.—To increase the production of 100 O.N. aviation gasoline from 40,000 brl. to present requirements (estimated at considerably more than 500,000 brl. per day) the construction of over 170 units has been completed or put in hand at a cost of \$900,000,000. These include 60 catalytic crackers, fluid catalyst, T.C.C. and Houdry, producing aviation base of 80 O.N. which may be catalytically retreated to produce a more highly aromatic, more saturated product. 60 alkylation units combine *isobutane* and other *iso* paraffins with butene or *isobutene* to form *iso*-octane or alkylate of 90–95 O.N. The *isobutane* required is supplied by 31 isomerization units in addition to that from catalytic units, natural gasoline plants or distillation units. *iso*Pentane of 92–93 O.N. is obtained by concentration of natural gasoline and light fractions of crude. Cumene, toluene, and benzene are being synthesized or concentrated from suitable stocks. Selective catalytic polymerization is used to produce *iso*-octene from butenes for hydrogenation to hydrocodimer. A list is given of the 100 octane units operating or under construction, showing company and plant location, type, and completion date of units.

C. L. G.

997.* **Solvents Carry the Load in Quaker State's New Lubricating Oil Plant.** E. E. Ebner and F. T. Mertens. *Oil Gas J.*, 13.4.44, 42 (49), 173.—A description is given, with flow-sheet, operating data, and photographs, of a new furfural solvent extraction plant, an MEK-benzol solvent dewaxing plant and wax production plant at the Quaker State Oil Refining Corp. at Emlenton, Pa. The solvent dewaxing and wax recrystallization process sections are separate and may be operated in parallel or in series. Reduced wax distillate of pour point 90° F. is dewaxed to a pour point of about 0° F., yielding about 20% of slack wax of 125° F. melting point and 6% oil content, which is mixed with additional solvent and deoiled at 30° F., giving a wax of melting point 130–132° F. and an oil content of well below 1%. Alternatively, Pennsylvania cylinder stock is dewaxed to 0° F. pour point, yielding a petrolatum of 128° F. melting point with 8% oil content. This petrolatum is recrystallized at 60° F. to yield a microcrystalline wax of high melting point and oil content below 0.5%. A range of neutral and heavy oils of varying pour point and of crystalline waxes of melting point from 110° to 140° F. and microcrystalline waxes of melting point 110° to 160° F. or higher can be produced. The dewaxed oils pass to the furfural extraction unit which has a capacity of 1000–1750 brl./day, the extraction tower containing 8 nests of Raschig rings, each 5 ft. in depth, down which furfural flows at 200–300° F. Using 160% of furfural by volume, yields of 92% of neutral oil of viscosity index 113 or of 94% of cylinder stock of viscosity index 103 are obtained.

C. L. G.

998. **Patents on Refining and Refinery Plant.** J. G. Fife. E.P. 557,290, 15.11.43. Appl. 26.1.42. Contaminants can be removed from refined or used mineral oil or wax by mechanically mixing with them a solid adsorbent material. During the process the mixture is heated in a closed container under vacuum at a temperature below the flash point of the finally treated wax or oil to drive off low-boiling impurities. The adsorbent material adsorbs high boiling impurities.

Shell Development Co. E.P. 557,315, 15.11.43. Appl. 29.4.42. In the refining of sour hydrocarbon distillates containing mercaptans and stronger acids, mercaptans are absorbed into an aqueous alkaline solution containing a solutizer. A spent solution is thus obtained which is regenerated by steam stripping. An improvement is effected in the process if the hydrocarbon distillate is treated with an aqueous solution of caustic alkali free from solutizers, to remove acids stronger than mercaptans.

The resulting distillate is then treated to remove from it alkaline water-soluble impurities. Afterwards it is further treated with the aqueous caustic alkali solution and solutizer.

J. S. Highfield. E.P. 557,397, 18.11.43. Appl. 17.6.42. An apparatus is described for treating liquids with solids in suspension in order to clear or clean the liquids. It can be used for the cleaning and purification of oil. For this purpose a finely divided solid such as Fuller's earth is suspended in the oil for a considerable period during or after heating, and the oil while hot is subjected to a stripping operation. Afterwards it is cooled and filtered.

H. B. M.

Chemistry and Physics of Hydrocarbons.

999. The Viscosity of Pure Liquids. I. Non-Polymerized Fluids. R. M. Barrer. *Trans. Faraday Soc.*, 1943, **39**, 48-59.—The Arrhenius equation ($\eta = \eta_0 e^{E_A/RT}$) is used in a study of the viscosity of many pure liquids of all kinds. Variations of E_A (the energy of activation) with the nature of the liquid; and of E_A and $-\log \eta_0$ with chain length in some homologous series are discussed.

Consistent functional relations are found between $-\log \eta_0$ and E_A or $\frac{E_A}{T}$ which are independent of the nature of the liquid for liquid permanent gases, a-polar liquids, polar liquids, hydrogen and hydroxyl bonded liquids, non-polymerized ionic melts, and liquid metals. The data approximate to the general viscosity function $\eta = 3.71 \times 10^{-3} e^{0.23E_A/RT}$ poises.

These properties are satisfactorily accounted for by the zone theory of viscous flow.

C. F. M.

1000. The Viscosity of Pure Liquids. II. Polymerized Ionic Melts. R. M. Barrer. *Trans. Faraday Soc.*, 1943, **39**, 59-67.—The considerations of Part I (see previous abstract) are extended to polymerized ionic melts for which E_A may be very large and frequently depends on temperature.

The number of degrees of freedom involved in each zone and the total energy per activated degree of freedom necessary for flow to occur are estimated for typical glasses, liquid metals, simple ionic melts, a-polar and polar or hydroxyl bonded fluids.

The range of the Arrhenius energy of activation is compared for viscous flow, diffusion in solids or liquids, relaxation phenomena, and chemical reactions.

C. F. M.

1001. The Softening of Thermoplastic Polymers. Part 1—Theoretical. R. F. Tuckett. *Trans. Faraday Soc.*, 1943, **39**, 158-168.—The general deformation produced in an amorphous high polymeric material by a given stress is analysed into three components (termed ordinary elastic, highly elastic and viscous), each dependent on molecular size, structure, temperature, and duration of stress.

It is shown that the various empirical softening-point tests in current use measure either the flow properties of the material or the temperature at which high elasticity develops. A typical flow-property test, the modified Kraemer-Sarnow, is analysed further, and a relation between molecular size and softening temperature is predicted and shown to hold experimentally in the case of the polyvinyl acetates.

A critical survey of several recent estimates of polymer viscosities is given, it being shown that the viscous properties of such systems have often been confused with highly elastic ones.

C. F. M.

1002. Application of Unit Operations to Fractionation and Other Vaporization Processes. Part 1. R. L. Huntington. *Refiner*, February 1944, **23** (2), 57-60.—The phase rule is defined as:—

$F + P = C + 2$, where F = the number of degrees of freedom which a system may have, such as temperature, pressure, and the composition of any component in any one phase. When a certain number of degrees of freedom has been assigned to a system, it is said that the system is fixed. P = number of phases in the system.

A phase is any homogeneous mixture, such as vapour, liquid, or solid phase. Ordinarily, there is only one vapour phase, while there may be two or more liquid or solid phases present. C = the number of components or chemical compounds which are required to make up the system. This rule applies only to systems in which the different phases are in equilibrium with each other.

The rule is illustrated by studying conditions in a steam boiler.

A. H. N.

1003. Synthesis of Gasoline from Carbon Monoxide and Hydrogen at Atmospheric Pressure. F. Fischer. *Refiner*, February 1944, 23 (2), 70-76; translated from *Brennstoff-Chemie*, 1935, 16, 1-11.—The elementary chemistry of the synthesis and of the primary reactant gases is discussed. The production of Kogasin and its properties are then studied. The utilization of Kogasins as fuels and lubricants is then very briefly indicated.

A. H. N.

Analysis and Testing.

1004.* The Constitution of Petroleum Oils. Part 2. Hydrogenation Results. A. V. Brancker. *Petroleum*, May 1944, 7 (5), 80.—For Waterman analysis the requirements are: (1) Aniline point by the I.P. method (or, for coloured oils, the special tube of Williams and Deans (*Industr. Engng Chem. (Anal. Edn.)*, 1942, 14, 63); (2) Density at 20° C. (pycnometer); (3) Refractive index at 20° C. (Abbe instrument); (4) Molecular weight (Camphor method), better results are obtained by using the modified cryoscopic equation of Brancker, Leach, and Daniels (*Nature*, 1944, 153, 407) for the depression constant K_f .

The results of work and the Waterman analyses are given on three lubricating oil fractions A , B , C , each of which is solvent extracted with SO_3 ; and also on two aromatic-free oils prepared from A and B raffinates by exhaustive acid treatment. These show there is a concentration of aromatics in the extract, and no aromatics in the acid-treated raffinates. The raffinates contained 5.4% aromatic rings and 63.3% paraffins, compared with 11.4% aromatics and 55.6% paraffins in the distillate. The naphthene content shows smaller variation in the four oils, *i.e.* the distillate, the raffinate, the extract, and the aromatic-free oils.

The extract from A was vacuum fractionated into 10 fractions. The results and the Waterman analyses are tabulated. These show that fractionation is inadequate for segregating the groups.

Another set of results on the re-extraction of extracts are given, but even this did not afford aromatics as high as would be expected. "Whatever may be the criticisms of the Waterman method, it is considered that it affords a valuable means of comparison of refining processes, and does give an indication of the comparative distribution of the three main groups of hydrocarbons."

There is a discussion on the Waterman results and how the group structures would be affected by solvent extraction and chromatographic absorption.

This leads up to the subject of ring homogeneity, which is also discussed. In accepting ring homogeneity as a basis, the obtaining of aromatic concentrates by absorption or solvent extraction can be explained by assuming that an extract consists of ring hydrocarbons which may be mono-, bi-, or polycyclic, but not a mixture of all, and that these rings have associated with them alkyl groups of different chain lengths. When paraffin chains contain sufficient numbers of C atoms, the balance is on the side of paraffinicity, and when the chains contain only a small number of C atoms, the aromatic properties of the molecule predominates. This conception may have to be modified in the future.

The raffinate figures given do not mean that the paraffins are separate compounds, the paraffinic part of the oil must be combined in some way with the cyclic hydrocarbons either as long-chains or links between rings, or a large number of alkyl groups (of few C atoms).

As no diene number was obtained with the aromatic extracts, it is suspected that conjugation in the aromatic rings is masked by alkyl groups, or evidence of its presence is prevented by steric hindrance.

Two types of hydrogenation vessels are illustrated and described, and an ingenious mounting is shown for oscillating the reaction vessel. Results of hydrogenating

naphthalene and lubricating oil A extract, in the presence of 8% Raney Ni, and 8% reduced NiO₂ at various temperatures, are shown in graphs. The results with lubricating oil A extract, completely hydrogenated after 10 consecutive runs, and other experiments, confirmed Watermans charts, and verified his constant 0.85. It is concluded from these results that the Watermans charts and constants are applicable to the petroleum oils studied, and afford a valuable method of group analysis. In all cases hydrogenation turned coloured oils to water-white products, suggesting that resonances in these oils is due to the chromophore group $\text{C}=\text{C}$ present in the aromatic nuclei, which groups become saturated on hydrogenation. W. H. C.

1005.* **Radiation, the New Petroleum Analytical Tool.** C. H. Schleseman and F. P. Hoehgesang. *Oil Gas J.*, 13.1.44, 42 (36), 41-44, 49, 72, 75, 76.—A review is given of the principles, uses, and limitations of X-ray and electron diffraction, mass spectroscopy, emission spectroscopy, Raman, ultra-violet, and infra-red spectrography for analytical purposes in the petroleum industry.

X-ray diffraction can be used to identify crystalline solids, and has been applied to the identification of inorganic deposits found on valves, bearings, oil filters, etc., and of addition agents if obtainable as crystalline solids, catalysts, etc. Its limitations are: (1) necessity for sample to be crystalline, (2) empirical nature of results requiring comparative data, (3) insensitivity to traces and unsuitability for complex mixtures. On the average, 5 hours is required for the test and 1 hour for the calculations.

Electron diffraction is used to study surface phenomena which may be too thin to be detected by X-ray diffraction, owing to the intense penetrating power of the X-rays. The maximum resolution is of the order of 0.01 micron, an important feature being the depth of focus obtained in electron micrographs. An exposure of a few minutes only is required, the necessary calculations taking about an hour.

Mass spectroscopy is used for the analysis of light hydrocarbon products, and should be useful for the analysis of surface gases in petroleum prospecting. The analysis takes 15-30 minutes, but the calculation may require several hours.

Emission spectrography is used for the identification of metals and metalloids, being capable of detecting a few parts per million of *e.g.* impurities and being particularly useful for alkali detection. Quantitative analysis can be made by comparison of the spectrograph with those from known metals. Analysis time varies from a few minutes to several hours.

Ultra-violet absorption spectroscopy can be used for the identification and estimation of molecules containing resonating groups, in particular aromatic hydrocarbons, drying oils, and resins. The location of side-chains and the presence of conjugated unsaturated groupings can be determined. The analysis requires about $\frac{1}{4}$ hour, but the calculations may take several hours.

Raman spectrography is limited to the identification of mixtures containing not more than 5 components and which do not fluoresce. The analysis takes several hours, and quantitative determinations are very complicated.

Infra-red spectrography is being applied to an increasing extent for the analysis of mixtures of light hydrocarbons which do not contain more than 4 components (owing to the complexity of the spectrograph), and for the determination of functional groups in the molecule. The analysis requires from a few minutes to an hour, while the calculation may require one hour. Difficulties arise in the obtaining of absolutely pure compounds for reference, in the measurement and amplification of the radiation, and in obtaining suitable materials for prisms and windows which will transmit infra-red radiation without selective absorption. Sodium chloride is generally used but water vapour and carbon dioxide must be excluded or corrected for. C. L. G.

1006. **An Efficient Low-Holdup Laboratory Column.** J. R. Bower, Jr., and L. M. Cooke. *Refiner*, February 1944, 23 (2), 63-66. Paper reprinted from *Ind. Engng. Chem. Ind. Ed.*, April 15, 1943.—The column described contains a packing consisting of a narrow strip of wire gauze twisted to produce a series of vertical plates at an angle of 90° to each other. A strip of 50 (or 60 × 40) mesh Monel-metal-wire gauze (1.5 times the length of the glass column) was cut from a sheet of this material with heavy scissors. The width of the gauze should be approximately two wire strands greater

than the diameter (4×6 mm.) of the tube to be packed. The two outside longitudinal strands were then removed and the projecting lateral strands bent back in one direction until the strip fitted tightly in the glass column. The gauze strip was then ready for twisting. This was accomplished by use of two pairs of round-nosed pliers, the nose of one pair of which had been ground to a thickness of about 0.15 cm. (0.06 in.) The top of the gauze strip was gripped tightly with the thin-nosed pliers, and 0.3 cm. (0.125 in.) away with the standard pliers, and the strip was then twisted through 90° with a simultaneous compression force, so that when the two pliers were at an angle of 90° they were touching each other. This resulted in the formation of two flat vortical planes at 90° to each other, separated by two horizontal, opposed quadrants. The further twisting of the gauze strip was accomplished by gripping it just below the first bend with the thin-nosed pliers and 0.3 cm. (0.125 in.) below that point with the standard pliers. The twisting (in the same direction as the first turn) and compression motions were then carried out as described above. This entire process was repeated until the whole strip had been converted into a series of about 0.3 cm. (0.125 in.) vortical gauze platelets.

The method of insertion using oleic acid as lubricant, and the operation of the column are fully described.

A. H. N.

1007.* A Laboratory Timing Clock. Note by G. F. Smith. *J. Chem. Soc.*, 1944, 113.—A simple device is described which eliminates the loss of accuracy incurred when a series of successive events are timed without stopping the clock. The seconds hand of the electric clock carries a rider which is disengaged by an electromagnet when a tapping key is depressed. After noting the reading given by the rider it is then re-engaged with the hand of the clock ready for the next measurement. It is claimed that at least four readings per minute can be made by this means with an accuracy of about $\frac{1}{4}$ sec.

C. F. M.

1008.* Use of Mass Spectrometry in Routine Analysis of Refinery Gas Samples. J. G. Schaafsma. *Oil Gas J.*, 6,444, 42 (48), 57.—A brief description of the General Petroleum Corporation's Mass Spectrometer is given. Any light hydrocarbon mixture can be analysed provided the pure constituents are available for the calibration.

n- and *iso*-pentanes and total pentanes are easily determined. Traces of C_6 hydrocarbons (0.1–1%) can be estimated with sufficient accuracy without going through the complete calculations required to separate these constituents. Oxygen and nitrogen can be accurately determined. CO_2 interferes badly with certain hydrocarbon peaks and is removed before the analysis.

The accuracy of the instrument is discussed and the results obtained by two companies, on the same samples, are shown to be highly satisfactory. Other work has shown that 0.1–0.3% of heavy ends, *i.e.* pentanes and hexanes is present in samples that were not recorded by conventional methods.

Elapsed time, man hours, samples per shift and long and short methods of analysis are discussed, and a table shows the times required for a complete analysis, and for the key components only, on samples from a gas plant and from an alkylation unit processing butenes and pentenes together. The results from the alkylation unit are compared with the times taken by the conventional low-temperature fractionation and acid absorption method.

Instrument time is usually 30 minutes for all types of samples, but calculation times differ widely. Determination of a single paraffinic constituent varies from 5 to 10 minutes, depending upon whether an isomer is present. Calculations for all components of a C_1 – C_4 paraffinic gas takes 30 minutes, the presence of *iso*- and *n*-pentanes may add another 10 minutes. A cracked C, through C_4 gas, requires about 1 hour calculating time owing to the presence of olefins, and the presence of appreciable amounts of pentenes may add from $\frac{1}{4}$ –1 hour, depending on whether they are calculated as a total or as individual pentenes.

About 1 hour per day, on an average, is spent in calibrating the instrument. A complete calibration is only needed each two weeks, but a *n*-butane check is made daily and takes less than 30 minutes. A complete analysis of a cracked gas containing no pentanes would require:—

	Elapsed time, minutes.
Introducing samples	5
Making run from Mass 10 to Mass 66	20
Making run from Mass $\frac{1}{2}$ to Mass 4	5
(H ₂ requires weaker magnetic field)	
Developing	5
Calculation for all components	60
	Total 95

For comparison, a short method for *iso*-butane-propane, from a Depropanizer (Masses 40-60) would require 25 minutes, the calculating time by inspection (included) being 5 minutes.

Short methods save much time in cases where operations can be controlled on the basis of one or two key components, *i.e.* *iso*-butane and propane from a Depropanizer. Two short cuts are explained. Calculation by inspection is extremely useful to determine up to 5% of undesirable heavy constituents caused through improper plant control.

A comparison of results of gas analyses, by combined fractionation and acid absorption, and by the mass spectrometer, is shown. W. H. C.

1009. Use of Mass Spectrometer in Analysis of Refinery Gas Samples. J. G. Schaafsma. *Petrol. Engr.*, April 1944, 15 (7), 136-144. See Abstract No. 1008. R. A. E.

Motor Fuels.

1010. Effect of War-time Fuel Developments upon Post-War Fuel Problems. A. T. Colwell. *J. Soc. aut. Engrs.*, 1944, 52 (1), 1-12.—The author has interviewed 81 of the leading oil and automobile engineers of America to obtain their views on probable trends in post-war fuels and automobiles.

The weighted average opinion leads to the following conclusions:—

- (1) No new models will be on the market for at least 18 months after the war.
- (2) It is anticipated that a portion of present high-octane capacity—about 10% of total fuel capacity—will be available for automobiles if it can be utilized. There is no object, however, in using high-octane fuel in an engine not designed for its use. It is hoped that two grades of gasoline will eventually satisfy all demands, but immediately after the war four grades will probably be marketed—*viz.*, aviation of 100 octane and 100 octane plus; premium of 85-87 octane; regular of 75-77 octane; and third grade of 70 octane for tractors.
- (3) Compression ratios of higher than 8-1 are not expected. Although limited gains in power are possible, successive increases in compression show diminishing returns, and pre-ignition rating, as distinct from detonation rating, might be necessary.
- (4) The medium size car will aim at 25-30 m.p.g., but the very small car is not foreseen. Economy may be achieved by the use of a 4-cylinder engine in some cases. The general aim will be towards economy rather than super performance.
- (5) Higher compression pressures and temperatures will require improved lubricants with better characteristics such as higher V.I., less sludging and oxidation, and more resistance to breakdown under increased pressures and temperatures.
- (6) Supercharging is expected to be used on some heavy duty vehicles, but not on automobiles as long as economy is the dominating factor.

Considerable data in the form of tables and graphs are given to support the opinions expressed. C. H. S.

1011. Patents on Motor Fuels. Shell Development Co. E.P. 556,803, 22.10.43. Appl. 4.2.42. In the isomerization of saturated hydrocarbons containing from 4 to

9 carbon atoms, the saturated hydrocarbons are contacted under isomerization conditions with a catalyst containing a small proportion of an anhydrous aluminium halide and a major proportion of activated alumina.

Anglo-Iranian Oil Co. E.P. 556,894, 27.10.43. Appl. 2.10.40. In the production of branched-chain alkanes, normal alkanes in vapour phase and an aluminium halide catalyst are contacted with porous or sorptive substances or surfaces at a reaction temperature not greater than 250° C., and determined according to the feedstock and the desired product.

Anglo-Iranian Oil Co. E.P. 556,895, 27.10.43. Appl. 1.7.41. A process is described for the production of branched-chain alkanes by the catalytic treatment of normal hydrocarbons containing more than 3 carbon atoms in the molecule. Volatile catalysts are used, e.g. an aluminium halide, having activity in hydrocarbon isomerization reactions and having a measurable vapour pressure at or below the reaction temperature.

Anglo-Iranian Oil Co. E.P. 556,896, 27.10.43. Appl. 1.7.41. Branched-chain alkanes are produced from a normal alkane containing 4 or more carbon atoms in the molecule by employing an aluminium halide as catalyst and contacting the normal alkane at isomerization reaction temperature in the reaction vessel successively with the catalyst and porous or sorptive surfaces. The process relates particularly to the production of branched-chain alkanes from normal butane or normal pentane, including hydrocarbon mixtures containing a portion of such normal hydrocarbons.

Anglo-Iranian Oil Co. E.P. 556,897, 27.10.43. Appl. 1.7.41. A process is described for the catalytic isomerization of a normal alkane containing more than 3 carbon atoms in the molecule, in the presence of an aluminium halide as catalyst. The normal hydrocarbon feedstock containing a predetermined amount of water is contacted before isomerization with an aluminium halide under conditions in which a corresponding proportion of the halide reacts with the water of the feedstock. In this way the feedstock is dried and a corresponding proportion of hydrogen halide is produced for maintaining the activity of the catalyst.

H. B. M.

Lubricants and Lubrication.

1012. The Heat of Adsorption of Long-Chain Compounds and their Effect on Boundary Lubrication. J. J. Frewing. *Proc. Roy. Soc.*, 1944, A, 182 (990), 270.—By the application of the conventional mathematical treatment of unit process rates, it is shown that the equilibrium state of an adsorbed film of a polar body at the interface between a solution of the polar body and a metal surface would be expressed by the relationship :

$$K = \frac{k_1}{k_2} = \frac{x}{C(1-x)}$$

where K = equilibrium constant ; k_1 = the rate constant for the adsorption process ; k_2 = the rate constant for the solution process ; x = fraction of surface occupied by adsorbed molecules ; C = the concentration of the polar body in the solution.

The influence of temperature and of heat of adsorption on the value of K is deduced by the use of the integrated form of the Van't Hoff isochore, and is given by :

$$\log_e K = \frac{U}{RT} + \text{constant of integration}$$

where U = heat of adsorption (assumed independent of T over the small range in question) ; R = gas constant.

The frictional behaviour of mild steel surfaces lubricated with solutions in white oil of long-chain halides, acids, α -substituted acids, esters, cyanide, thiocyanate, and a nitro-derivative has been investigated under high loads at low speeds. Tests were carried out on a machine in which rapid fluctuations in friction are transmitted through a movable member carrying one of the bearing surfaces, a record of the instantaneous friction being made (Frewing, *Proc. Roy. Soc.*, 1942, A, 181, 23). In all cases a transition from smooth sliding to stick-slips occurs at a temperature which is

characteristic of the particular solution employed. For each substance the transition temperature increases with the concentration.

Assuming that the transition occurs at a definite value for x , *i.e.*, at $x = a$, for any one material, then it follows from the foregoing relationships that

$$2.3 \log_{10} C = -U/RT_i + \text{constant},$$

T_i being the transition temperature at concentration C . Hence $\log_{10} C$ should be linear with respect to $1/T_i$. This was found to be so in all cases, indicating that the adsorbed film is in equilibrium with the solution.

The values found for the coefficients of friction were the same as those previously found for monolayers of the same substances. The adsorbed film-solution equilibrium, and the values found for the heats of adsorption suggest that the adsorption takes place by interaction of the dipoles in the polar groups with the metal atoms at the surface, not by direct chemical combination. In the case of the esters, the results suggest that the orientation at metal surfaces is similar to that at aqueous interfaces, *i.e.* when the "alcohol" chain contains five or more C atoms, the lateral attraction between the "alcohol" and "acid" chains is sufficient to force the molecule into the "hairpin" configuration, and the obstruction of the polar group by methylene chains is reduced.

C. G. G.

1013. Patents on Lubricants and Lubrication. A. La Rue Parker. E.P. 556,834, 21.4.42. Appl. 25.10.43. A paste for sealing threaded joints between metals consists of a mineral oil having a viscosity of at least 140 secs. Saybolt at 210° F., and fused lead stearate intimately blended with the oil. Approximate proportions are 120 parts of oil to 20–35 parts of stearate.

Standard Oil Development Co. E.P. 557,177, 9.11.43. Appl. 24.12.41. Pour depressors for waxy lubricating oils are prepared by chemically condensing an aliphatic olefin having between 5 and 10 carbon atoms with an aromatic compound in the presence of a Friedel-Crafts catalyst and recovering from the reaction product a residue which is substantially non-volatile at temperatures below 600° F.

H. B. M.

Special Products.

1014. Determination of Oil Deposit on Citrus Leaves by the Steam Distillation Method. F. A. Gunther and J. P. La Due. *J. Econ. Ent.*, February 1944, 37 (1), 52.—The method of Gunther and Ebeling (*J. Econ. Ent.*, 1942, 35 (3), 333) for determining kerosino deposit on citrus leaves has been modified for use with heavier spray oils. No modification is necessary to the apparatus, but dilute phosphoric acid is now used to decompose the leaves, instead of an aqueous solution of sulphuric acid, nitric acid, and aluminium sulphate, prior to steam distillation. The method is claimed to be rapid, accurate, and reproducible, and independent of the extent of penetration of the oil into the leaf tissues.

C. L. G.

1015. Laboratory Tests of Synthetic Organic Compounds as Insecticides. M. C. Swingle, J. B. Gahan, and E. L. Mayer. *J. Econ. Ent.*, February 1944, 37 (1), 70.—The results are given of tests carried out by the Bureau of Entomology and Plant quarantine at Sanford, Fla., on the toxicity towards a variety of chewing insects of 82 organic compounds. The compounds were applied as dusts on leaves which were fed to the insects in closed petri dishes and, in the case of volatile compounds, tested as fumigants after mixing with sand. The following compounds were found moderately toxic, but none are considered to be of practical value:—2 chloro-6-nitrotoluene, 4-chloro-2-nitrotoluene, *p*-nitro phenyl acetate, *d*-bromo camphor, chloral acetamide, $\gamma\gamma$ -dipyridyl, 2-iodo-3-nitrotoluene, *p*-nitropropionanilide, phenanthrene, 2-quinolinol and 8 quinolinol.

C. L. G.

1016. Toxicity of Nitro Paraffins and Chlorinated Nitro Paraffins to California Red Scale and their Effects on Lemon Fruits.—R. A. Fulton, R. L. Busbey, and H. R. Yust. *J. Econ. Ent.*, February 1944, 37 (1), 117.—In fumigation tests against red scale on lemon fruits, no significant mortality was shown by the lower nitro paraffins,

nor were the fruits injured. Of the chlorinated nitro paraffins tested, all caused severe injury at concentrations high enough to give control, with the exception of 1 : 1-dichloro-1-nitropropane. In general the mono-chlor derivatives caused more injury than the di-chlor derivatives.

C. L. G.

1017. The Use of D-D Mixture Against *Anomala* and *Adorctus* Infesting Nursery Stock. W. Cartor. *J. Econ. Ent.*, February 1944, **37** (1), 117.—A saturated solution in water (0.2%) of D-D mixture (1 : 3-dichloropropylene and 1 : 2-dichloropropane) diluted with 3 vols. of water has been shown to kill larvæ of ground beetle in nursery stock, higher concentrations injuring the stock.

C. L. G.

1018. Tests Conducted by the Bureau of Entomology and Plant Quarantine to Appraise the Usefulness of D.D.T. as an Insecticide. P. N. Annand et al. *J. Econ. Ent.*, February 1944, **37** (1), 125-159.—A series of papers gives the results of tests carried out by the above Bureau to determine the toxicity of dichlorodiphenyl-trichloroethane ("Gesarol," "Neocid," D.D.T., or G.N.B.-A.) towards a wide variety of insect pests. It was first prepared in 1874 by the reaction of chloral and chlorobenzene in the presence of concentrated sulphuric acid, and is a crystalline solid, practically colourless, almost odourless, and rather stable, insoluble in water, but soluble in most organic reagents, and of low volatility. It acts as a nerve poison when injected or absorbed into the skin.

When used in the form of dusts, solutions, or emulsions, effective control of the following insects has been obtained: Human and animal lice, bed-bugs, ticks on vegetation and dogs and cattle, chiggers, house-flies, stable flies, manure-flies, fleas, roaches, cereal pests, and a wide variety of agricultural and horticultural pests including the codling moth, cabbage caterpillar, aphids, weevils, etc. Generally it appears to be excellent against human, animal, domestic, and warehouse pests, and without ill effects, the outstanding feature being its persistence. Thus garments treated with a D.D.T. solution will withstand 3-6 washings and still remain effective against lice, while flies are still knocked down several hundred days after the spray is applied, though the initial knockdown is much slower than that of other insecticides. Against roaches, sprays gave better control than dusts, but the residual effect is less. The control of codling moth is more effective than with lead arsenate or nicotine, but there was some leaf damage and an increase in red spider population. It has been established that D.D.T. is a stomach poison for honey bees at 0.05% and a contact poison at 1.0% concentration.

C. L. G.

1019.* Liquid Fuels and Organic Chemicals from Coal and Home-Refined Petroleum. H. Moore. *J. Inst. Fuel*, February 1944, **17** (94), 65-74.—A review is given of the pre-war position in the production of derivatives from coal and petroleum from technical and economic aspects, and suggestions made for the lines on which post-war policy should be based in order to integrate the two industries, taking into account probable restriction of imports and recent developments in the production of chemicals from petroleum.

In 1938, 38.2 million tons of coal were carbonized in coke-ovens and gasworks, and about 0.5 million tons in low-temperature carbonization plants, no difficulty being experienced in the disposal of by-products, though creosote and pitch were hard to market. Recent developments, however, enable these products to be satisfactorily used as fuel. Hydrogenation is considered too expensive for large-scale production of petroleum substitutes, and the Fischer-Tropsch process of economic value only for the production of special products, e.g. waxes, fatty acids, and alcohols, though the availability of a cheap source of hydrogen might alter the picture. Acetylene from coke is another useful raw material for chemical production, but the cost is at present too high. Petroleum refining in this country is largely based on asphalt production, by reason of the difficulty of transporting it by sea and of the preferential tariff it enjoys. Lubricating-oil production and general refining have met with moderate success, but cracking plants have not been too successful, owing to the production of large quantities of gas, of little or no value at that time.

To allow of greater quantities of coal being carbonized, a 50% increase being suggested, encouragement will have to be given to the greater use of gas and coke and

to the exclusion of coal in domestic and industrial use. This will lead to a lower coal production, but an increase in the carbonizing and allied industries which will make up for the increase in unemployment underground. A reduction in atmospheric pollution can also be expected, and there will be a big increase in derivatives for the fuel and chemical industries. The use of creosote and pitch, the only by-products which may be difficult to dispose of, could be encouraged by increasing the tax on imported fuel. Much of the present tar-distillation equipment will require overhaul and replacement and modernizing to handle the additional quantities of tar. Research should be expanded on the hydrogenation and Fischer-Tropsch processes and on the use of producer gas and liquid methane as fuel for road vehicles, powdered coal, and oil for external combustion and steam engines for road vehicles.

Petroleum is a particularly useful source of chemical products by reason of its low price and wide availability, high hydrocarbon content, and the variety of products available from modern refinery processes. Home refining should be encouraged, not only to provide the basis of a petroleum chemical industry, but because it is cheaper to transport crude than its products and local requirements are best met by local refining. Refining in the consumption centre is also a more permanent proposition, and does not depend on the availability of crude from any particular field. It is estimated that the home refining of 12 millions tons of petroleum would require an outlay of £36,000,000, but a similar sum should be saved per annum in adverse trade balance by reason of the price differential between crude oil and its products. An annual turnover of 10-15 million lb. of products for the chemical industry is estimated. It is proposed that 7 refineries should be constructed near the large centres of population, the products being handled by a national organization similar to the Petroleum Board. Considerable modifications of the present method of taxation of petroleum products would be called for, and some form of preferential duty applied to home-refined products.

C. L. G.

1020.* Toluene Recovery Plant Employs Azeotropic Distillation. A. L. Foster. *Oil Gas J.*, 13.4.44, 42 (49), 130.—The Beaumont Texas Refinery of the Magnolia Petroleum Co. is recovering and concentrating 350-400 brl. per day of toluene from the thermally cracked naphthas from polyform, Cross, and combination units. The distillate is fractionated into tops of end point about 200° F., a toluene concentrate of 215-245° F. boiling range, and bottoms. The concentrates from polyform distillate contain 23% toluene, and those from Cross and combination units 8%, an overall recovery of 80% being obtained. Methyl alcohol is added to the toluene concentrate and charged to the first azeotropic tower to produce a bottom toluene/alcohol mixture containing 50-70% toluene and a top hydrocarbon/alcohol mixture containing 55% alcohol. The bottoms are mixed with additional alcohol and charged to a second tower producing a bottom fraction containing 96-98% toluene and a top fraction containing 69% alcohol. The crude toluene is contacted with 6-10 lb. per brl. of sulphuric acid, filtered through gravel, water washed, caustic washed, filtered, hydrolyzed with 20° Bé caustic for 1 hour at 300-325° F. to remove sulphates and sulphonates, and redistilled. The nitration grade of toluene produced has the following properties: Paraffin content, 0.5%; distillation range, 110.4° C.-110.8° C.; specific gravity, 0.869; cloud below, 30° C., and acid wash colour, 1-2. The aviation grade has the following properties: gravity, 31.6° A.P.I.; sulphur, 0.03%; colour, 30 Saybolt; copper dish gum, 2.5 mg./100 ml.; copper dish corrosion, nil.; accelerated gum, 4 mg./100 ml.; accelerated gum residue, 0.1 gm./100 ml.; A.S.T.M. distillation range, 227-233° F. The methanol in the azeotropic overheads is recovered by water extraction and distillation producing 99.5% alcohol and 99% water. The alcohol-free gasoline is returned to the refinery for motor gasoline blending.

C. L. G.

1021. Action of Inert Dusts on Insects. V. B. Wigglesworth. *Nature*, 22.4.44, 153, 493.—The toxic action of chemically inert dusts on insects has been attributed to their increasing the rate of evaporation of water through the cuticle, possibly by adsorption of the epicuticle fat film, thus interrupting the continuity of this film over submicroscopic areas. It has now been shown from observations, mainly on the bug *Rhodnius*, that abrasion of this film by the dust is an important factor. If alumina dust is applied to the insect suspended in mid-air, no desiccation takes place, but if it is allowed to run over the dust, it is desiccated within 24 hours. If abrasion

is prevented by applying a wax coating to the abdomen, which normally touches the ground, the desiccation is much reduced. Polyphenols present in the insect cuticle do not reduce ammoniacal silver hydroxide, owing to the protective coating, but, if this is abraded, reaction can be shown to take place in these areas. In a moist atmosphere the impermeability to water of insects rubbed with the dust is largely restored by secretion of a wax film, but if the dust is allowed to remain on the insect, by immobilization, no recovery takes place. Insects first rubbed with alumina dust will succumb to a 2% nicotine solution within 20 minutes, but will otherwise withstand it for 24 hours, rotenone giving similar results though less quickly. It is suggested that the addition of abrasives to insecticidal dusts may be worth consideration.

C. L. G.

1022. Raw Materials for the Plastics Industry. W. J. Worboys. *Brit. Plastics*, June 1944, 16 (181), 249.—The raw-material requirements for the range of plastics now made or which might be made in the U.K. in the future are discussed. For the present thermosetting plastics, phenol, cresol, benzene (for synthetic phenol) can be supplied at competitive prices by the coal-tar distillation industry and formaldehyde, methanol, and urea by the synthetic nitrogen industry. Acetylene, raw material for the vinyl products, requires a cheap supply of calcium carbide, previously imported from Scandinavia, and if domestic production of this material is not economic, importation of carbide or manufacture of the resins from ethylene could be considered. This can be obtained from alcohol, from molasses or waste agricultural products, or from coke-oven gas, though the latter source will require considerable technological development. Oil cracking also provides ethylene, but the yield is small unless the oil is cracked for this purpose. From propylene produced at the same time, acetone, raw material for the acrylic resins, can be obtained. Acetone is also produced in the U.K. from alcohol, and this source provides acetic acid for cellulose acetate, while a new American process manufactures it from carbon monoxide and methanol.

Some of the American plastics, made only on the small scale in the U.K., e.g., polyvinylidene chloride, polyvinyl butyral, ethyl cellulose, and cellulose acetobutyrate, require ethylene, acetylene, and acetic acid and butyl alcohol. The fermentation of molasses produces butyl alcohol, while *isobutyl* alcohol is manufactured from coal by a modification of the methane synthesis.

The newer American plastics, e.g. melamine-formaldehyde and other cross-linking resins, as well as a wide range of plasticizers, are largely derived from oil, but coal is not excluded as a source of these products, just as in the U.S., Russia, and Germany synthetic rubber is derived to a greater extent from alcohol or coal than from petroleum.

It is pointed out that cost of the raw material is only a small item in the cost of the finished product and that purity is a very important consideration. Future plastics will probably be complementary to existing types, and new fibre-forming polymers are likely to be developed, as also are modifications or new forms of existing materials.

The plastics industry should be able to make an important contribution to the country's post-war trade.

C. L. G.

1023. Dermatitis from Synthetic Resins and Plastics. C. A. R. *British Plastics*, June 1944, 16 (181), 256.—It is suggested that present hypersensitivity to dermatitis is due to a deficiency in the diet of certain unsaturated fatty acids (e.g., linoleic and linolenic) owing to the reduced amount of linseed oil cake in cattle food affecting the milk and butter produced. In one laboratory in the plastics industry, dermatitis difficulties have been cleared up by the use of a proprietary maize preparation which may be applied externally or taken internally.

C. L. G.

1024. The Influence of Metallic Salts on the Drying of Linseed Oil. H. W. Rudd. *J. Oil Colour Chem. Assoc.*, June 1944, 288, 111-118.—A study has been made of the effect of a series of alkali salts including sulphate, chromate, borate, and silicate on the drying of linseed oil containing cobalt, manganese, and lead driers, the paint being prepared in emulsion form so as to give intimate contact between oil and salt on evaporation of water. The effect of the combination of salt and drier has been correlated with the nature of the film resulting from contact of the aqueous salt solution

and the oil containing the drier. Increase in weight tests were carried out on thin films of the linseed oil-drier emulsion (using ammonium oleate-gelatin emulsifier) to which 2½% of the salt was added, the films being first dried *in vacuo* over calcium chloride to remove water. In general, the salts caused an increase in induction period and a decrease in the rate of oxygen absorption, this not being due to poisoning of the drier, as oil recovered from the emulsion dries normally. In the absence of driers, sodium sulphate and sodium phosphate have no effect, whereas chromates accelerate drying. In general, salts, except chromates, had little or no effect on oil containing cobalt drier (linoleate), but some variable inhibition effect on oils containing lead and manganese driers. Chromates, in the presence of driers, accelerate drying as a result of oxidation, except in the case of lead, where the induction period is prolonged owing to precipitation of lead chromate. A microscopic examination of the surface phenomena resulting from placing a minute drop of the oil plus drier on a thin film of salt solution revealed that combinations of salt and drier which inhibited drying in the above tests gave rigid or semi-rigid interfacial films, while those which had no effect on drying gave non-rigid films of the normal type. There can be no question of chemical action between the salt and soap, as the oil from broken emulsions which exhibit inhibition of drying dries normally, nor is there any sign of precipitation. It is suggested that a molecular complex is produced which prevents the soap from catalysing the oxidation of the oil. This complex disappears when the emulsion is broken, but when rapid evaporation of the water from a film takes place the complex remains intact in the case of combinations of sufficient stability, while with others it disappears and the oil dries normally.

C. L. G.

1025. Action of Inert Dusts on Insects. H. Kalmus. *Nature*, 10.6.44, **153**, 714.—Confirmation of the conclusions reached by Dr. Wigglesworth (see Abstract No. 1021) on the importance of abrasion and possibly adsorption of the waxy coating on insects, in causing the desiccation of insects by inert dusts, has been obtained by tests on *Drosophila* flies, etc. Dusting the insects with charcoal caused rapid death in atmospheres of low humidity, but in high humidity they survived several hours. Immobilization with carbon dioxide, to reduce abrasion, before treating with dust also prolonged their survival. Abrasions caused by charcoal were observed under the microscope on the abdomen of the cottonstainer *Dysdercus* sp., where the femora had rubbed it. Cutting the legs before dusting prevented scratching and increased the time of survival.

C. L. G.

1026. Coal Tar Paints. Anon. *Chem. Tr. J.*, 16.6.44, **114**, 620.—E.P. 560,390 granted to the South Metropolitan Gas Co. and G. H. Fuidge covers the preparation of improved coal-tar paints which do not become brittle and crack on outside exposure. Coal-tar pitch, of melting point about 80° C., is treated at 180–230° C. with air or oxygen or with 1½–3% sulphur to increase the melting point to 95–120° C. The pitch is then fluxed with 3–12% of coal-tar oil boiling above 300° C., e.g. anthracene oil, and with 5–15% of a vegetable oil and 5–15% of a suitable filler—lampblack or kieselguhr—added (quantities based on final composition before thinning). The final paint contains 30–45% of coal-tar naphtha, to which is added a small percentage of crude pyridine bases, unless already present in the thinner.

C. L. G.

1027. Activation of Pyrethrins by Sesame Oil. E. A. Parkin and A. A. Green. *Nature*, 1.7.44, **154**, 16.—The effect of lubricating oil and other adjuvants to pyrethrins in increasing the kill on mosquitos (*Aedes aegypti*) described by David and Bracey (*Nature*, 1944, **153**, 594) (see Abs. No. 915) has been examined in tests on house flies carried out in an 18 cu. ft. capacity chamber. It was found that the addition of 0.5% of a sesame oil, oleic acid, medicinal paraffin, or lubricating oil to a 0.05% pyrethrin solution in kerosine had no effect on the kill, while 0.5% of other sesame oils gave considerable increases. The former products were shown to reduce the rate of knockdown, and the latter to increase it, thus showing that activation, as far as kills on house-flies is concerned, does not depend on reducing the rate of knockdown. The addition of 0.05% sesamin gives a very large increase in toxicity, and this compound is considered to be mainly responsible for the increased kill given by some samples of sesame oil, although desesaminized sesame oil still shows some adjuvant

activity. The inactive sesame oil was found to contain only 0.3% of sesame as compared with the normal figure of 1%, but as this oil actually reduced kill and knock-down, secondary factors must also play a part in determining the effect of sesame oil on pyrethrin sprays.
C. L. G.

1028. Patent on Special Products. Standard Oil Development Co. E.P. 557,291, 15.11.43. Appl. 28.4.42. A process is described for the production of substantially pure aromatic hydrocarbons, such as benzene, toluene, mixed xylenes, etc., from certain types of hydrocarbon oils generally in the gasoline range and containing at least 30% of olefins.
H. B. M.

Detonation and Engines.

1029. Motion Analysis by Means of High Speed Photography. H. D. Jackes. *J. Soc. aut. Engrs*, 1944, 52 (1), 49-53.—The author stresses the advantages of high-speed photography in the analysis of the operation of moving parts of engines in comparison with stroboscopic devices and single or multiple flash still pictures. Film records obtained, when projected at normal speed, give a magnification of 150 times.

Many subjects which lead themselves to this form of motion analysis are discussed and photographs taken at high speed are produced.
C. H. S.

1030.* Fuel Consumption of Diesel Engines. W. F. Schaphorst. *Petrol. Engr*, March 1944, 15 (6), 98.—A table has been prepared showing the fuel consumption in lb. per h.p. hr. of efficient modern diesel engines of varying h.p. when operated at full load. To enable an estimate of consumption, based on per cent. of full load at which the engine installed may be expected to operate normally, another table shows increase in fuel consumption which may be normally expected at percentages of full load ranging from 25 to 100%.
R. A. E.

1031.* Servicing Fuel-Injection Nozzles. P. H. Schweitzer. *Oil Gas J.*, 30.3.44, 42 (47), 97.—The causes of nozzle trouble in diesel engines are briefly discussed, and recommendations made for its prevention and for the testing, diagnosing, correction, and cleaning of nozzles, the freeing of stuck needles, and the reassembling of the injectors.
C. L. G.

Coal and Shale.

1032. Shale Oil. Anon. *Chem. Tr. J.*, 30.6.44, 114 (2980), 674.—*Foreign Commerce Weekly* (U.S.) of 17th February gives a review of the shale-oil industries in the main producing areas, with figures for production and yield of products. In Scotland there are between 480 and 884 million tons of recoverable oil shale, but mining and development are difficult and expensive. There are six plants in operation, handling 5000 tons of shale daily, yielding 18-45% of crude oil. 15% of the product yield is scrubber naphtha, while the remaining 85% consists of some naphtha, kerosine, gasoil, light lubricating oil, diesel fuel, wax, fuel oil, and coke.

The Estonian deposits yield on the average 15-30% of crude oil, though some give 53-67%, the 1939 production being 1,260,000 brl. of crude oil from 1,650,000 tons of shale. In 1943 it was hoped to produce 1,274,000 brl. from the shale. The gasoline is stated to be of high specific gravity and anti-knock value.

The Swedish shale produces only 5-7% of oil, but is located at convenient centres, and much of it is in 30-ft. seams accessible by open cut mining. The shale residues can be utilized almost completely. A new process has been devised for the direct extraction of oil from shale beds by electrical heating, but requires very large amounts of electricity. Production in 1943 of crude oil was probably about 567,000 brl.

In Manchuria, the shale overlies coal which is mined by the open-pit method, and thus has to be removed, so that the cost of mining the shale is charged against the mining of coal. The yield of oil is low, about 6%, the 1937 production being stated to be 966,000 brl., but it was hoped to increase production up to 3,500,000 brl.

In the Soviet Union it was planned to produce 4,000,000 tons of shale in 1942 in the Leningrad Oblast and Volga areas. Other shale deposits are found in Nova Scotia, Quebec, and New Brunswick in Canada, in Brazil, France, New South Wales,

while extensive deposits, estimated at 400,000 million tons, from which 100,000 million bbl. of oil might be obtained, exist in the U.S., the richest deposits being in the Green River formation in Colorado, Utah and Wyoming. From these, 60-80 gallons of oil per ton of shale can be obtained. Among the by-products from shale oil are phenols, occurring to the extent of 1%, and nitrogen bases including pyridine.

C. L. G.

1033. Tar Products as Building Materials. G. H. Fuidge. *Chem. Tr. J.*, 7.7.44, 115 (2981), 5.—In a paper recently presented to the Association of Coke-Oven Managers in Sheffield, developments in the replacement of bitumen by tar in paints, roofing felts, floor and roof mastics, and cable compounds were discussed. B.S. 1070 covers two tar-base paints which will give effective protection of iron and steel exposed to weather or immersed in water, if the metal surface is properly prepared and the paint carefully applied. A seven-year life can be expected from a two-coat application, but the tar-base paint should not be applied over a fresh oil paint or a coating of oil paint subsequently applied. For pipe dipping a refined tar of 20° C. E.V.T., heated to 300° F., is used. For roofing felt saturants, the use of tar is confined to the sanded felts (with blown bitumen a sand finish is unnecessary). For laminated felt roofing, B.S. 989 specified a 40° C. melting-point pitch as the saturant and a 50° C. melting-point pitch as the adhesive. Pitch felt is also used for damp-proof courses (B.S. 1067) and for damp-proofing of basement and water-proofing of basement and brick static water-tanks. Mastics from 60° C. melting-point pitch containing coarse fillers and stone are being used to an increasing extent for oil-resistant floorings (B.S. 1093). Their use in roofing mastics, for which fine fillers are required, is being investigated. Other uses for pitch and tar include battery sealing and cable compounds, though for the latter purposes a specially refined product is required to avoid fuming troubles in hot applications.

C. L. G.

BOOK REVIEW.

Emulsionen für die Metallbearbeitung (Part 1). By H. Stäger and H. Künzler.

The choice of an emulsion for any particular type of work has till now been mainly a result of practical experience. The authors have tried through extensive experiments to give some satisfactory indications of how theoretical considerations may be applied to this choice.

In an introductory discussion it is pointed out that the two properties of cooling and lubrication, fundamental to all liquids used in metal-working, cannot be considered except as interdependent. The effect of cooling is to increase the maximum velocity at which the metal can be worked. Lubrication is not so easily measured, and in this connection the opposing tendencies of the liquid to spread over the surface ("Benetzungsfähigkeit") and to collect and run off in droplets ("Abtropffähigkeit") are mentioned and the lack of suitable methods of measurement indicated.

The structure of emulsions and the effect of stabilizers are discussed from the physical chemical standpoint in the light of recent work. The authors consider that too little attention has as yet been paid to the interfacial forces involved in the use of emulsions, and have investigated this aspect of the matter, among others.

Experiments were carried out on the stability of various emulsions, which were heated for varying lengths of time. The stabilities and rates of separation were then studied. Here some interesting micro-photographs of emulsions and interfaces are given. Practical tests of stability in different types of work at varying concentrations were also made. It was found that the stability of an emulsion decreased with increase in temperature.

Extensive work on the corrosive effects of emulsions is also described, after a review of recent developments in the theory of corrosion. The importance of the nature, *i.e.*, degree of smoothness, of the metallic surface is emphasized. A complete range of surfaces was employed, using samples of pig iron and of medium hard carbon steel. An interesting series of photographs shows the degree of corrosion after 24 hours, and demonstrates the increase of corrosion with dilution of the emulsion and roughness of the surface. With increasing concentration the action diminishes until finally even the roughest surfaces are immune. The authors conclude that the concentration of an emulsion must be chosen to suit the grade of surface it is required to produce. Examples are given of cases in which the authors successfully substituted suitable emulsions for more expensive fatty oils or mineral fatty oil mixtures, *e.g.* steel screw manufacture using the automatic lathe, thread cutting in nickel-chrome steel, tooth-cutting in carbon steel.

W. H. THOMAS.

BOOKS RECEIVED.

Iron and Steel Institute, Journal. Vol. 148, No. 2. Proceedings, 1 P to 732 P. Survey of Literature, 1 A to 206 A. Iron and Steel Institute, 4, Grosvenor Gardens, London, S.W.1.

B.S.S. No. 1168 : 1944. Non-Reinforced Diamond Dies for Wire Drawing. Pp. 15. British Standards Institution, 28, Victoria Street, London, S.W.1. Price 2s. net.



INSTITUTE NOTES.

AUGUST 1944

PERSONAL NOTES.

Dr. Gustav Egloff (Member of Council), director of research of Universal Oil Products Company, and President of the American Institute of Chemists, has received the degree of Doctor of Science, *honoris causa*, at the commencement exercises of the Philadelphia College of Pharmacy and Science.

Dr. Egloff was selected for this honour "in recognition of his outstanding services in the great field of chemistry, notably in the petroleum industry."

Mr. T. H. G. Brayfield (Fellow) has written from the Stanley Civilian Internment Camp, Hongkong, to say that he and his colleague, Mr. A. W. Black (Fellow) are both well.

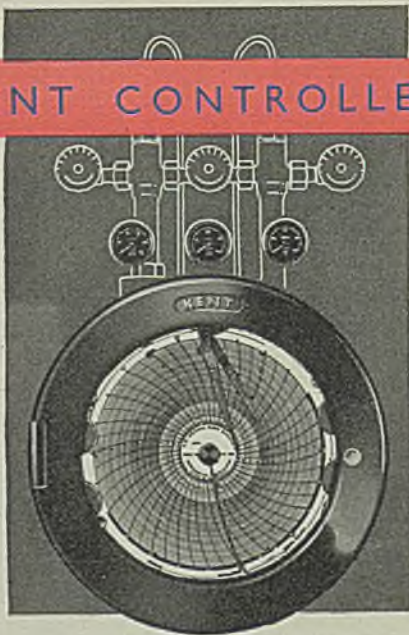
Mr. H. G. Shatwell (Member) has been appointed to the post of Professor of Industrial Chemistry, King Fuad 1st University, Cairo, and will be leaving the country as soon as the travel ban is lifted.

Mr. C. E. Keep (Fellow) is visiting this country from India.

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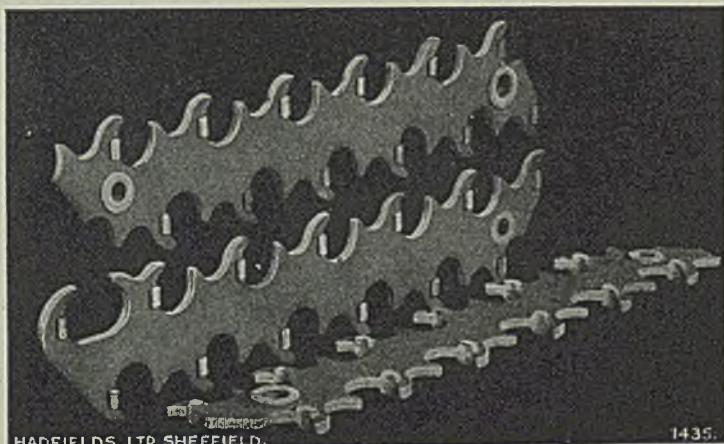
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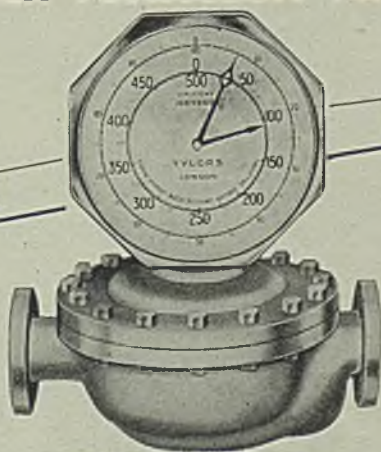
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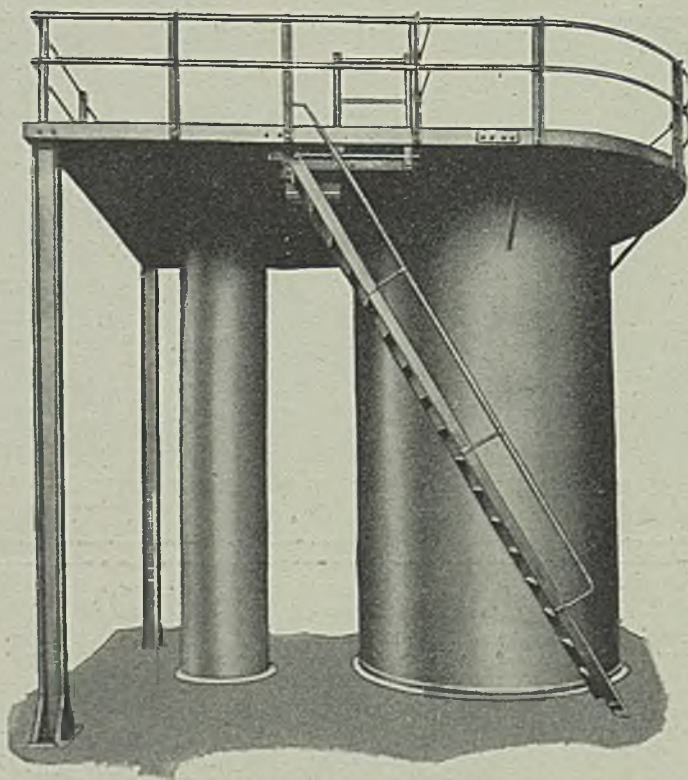
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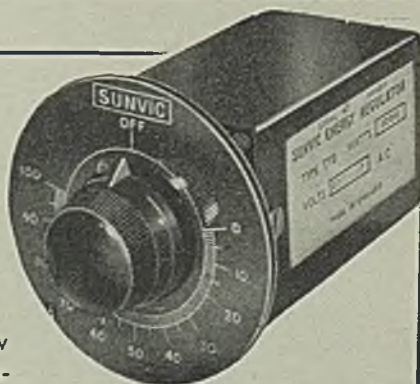
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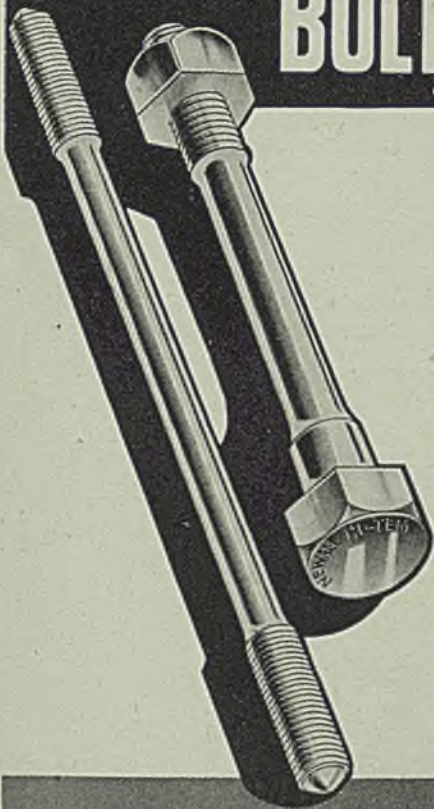
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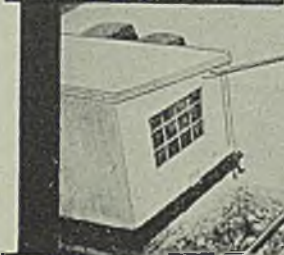
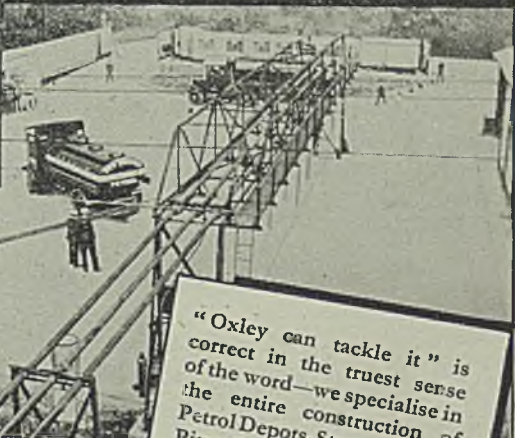
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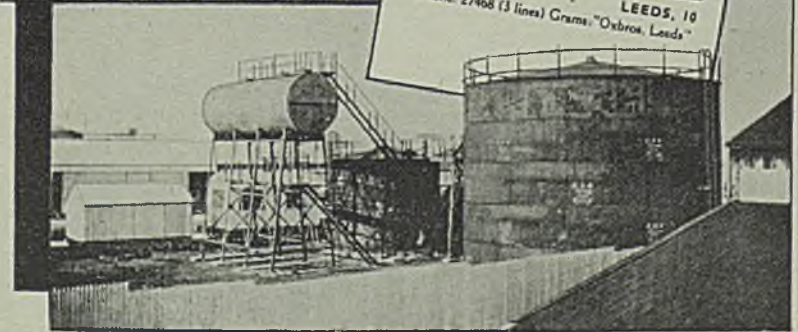
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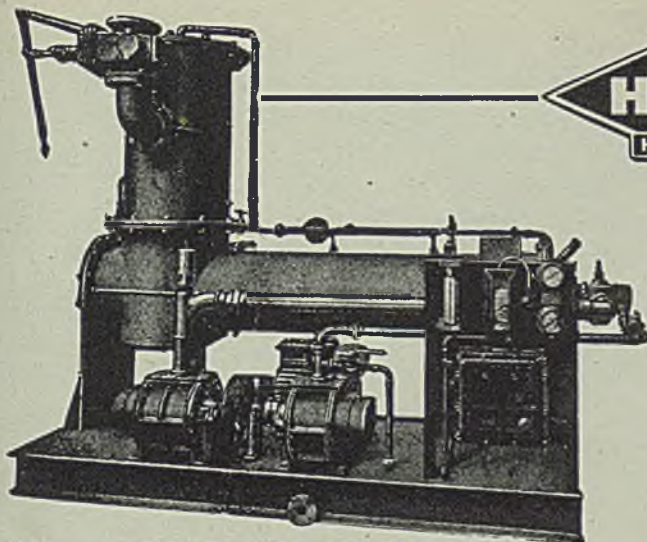
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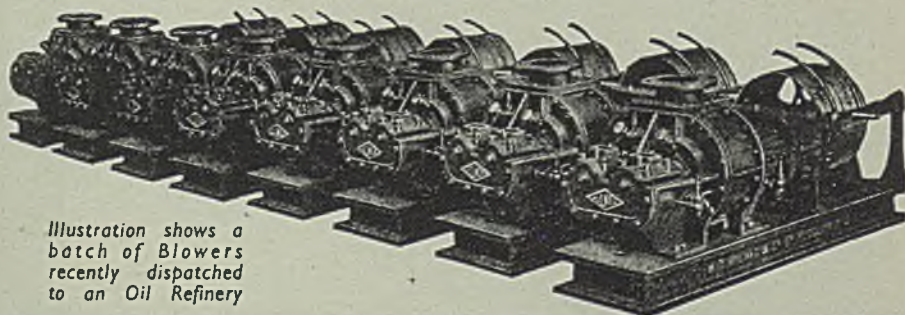


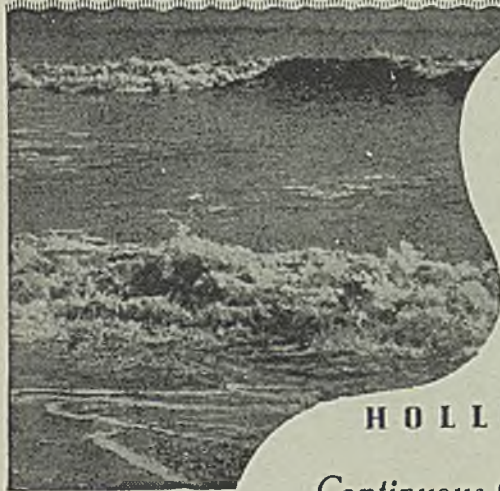
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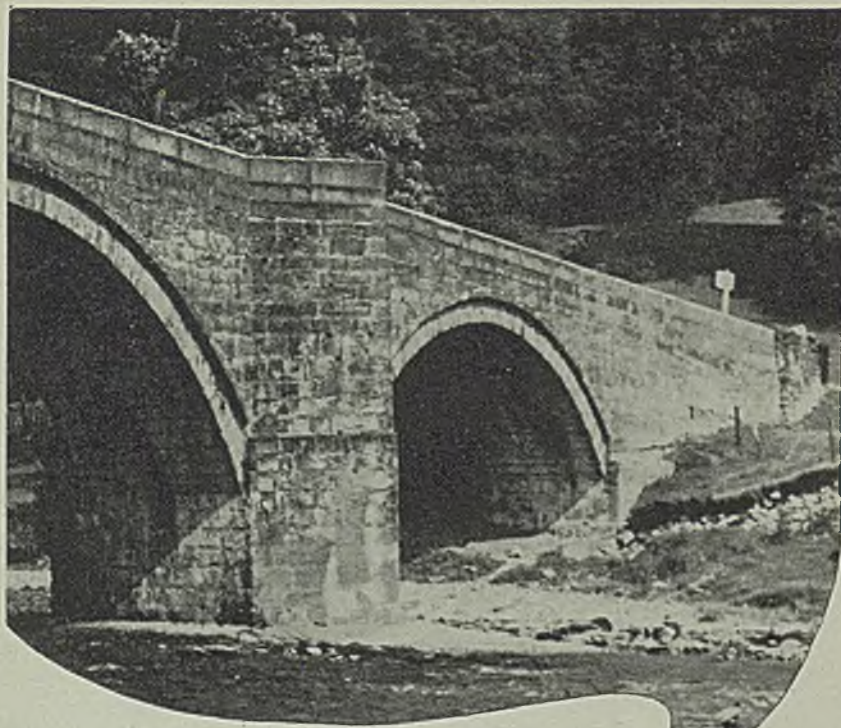
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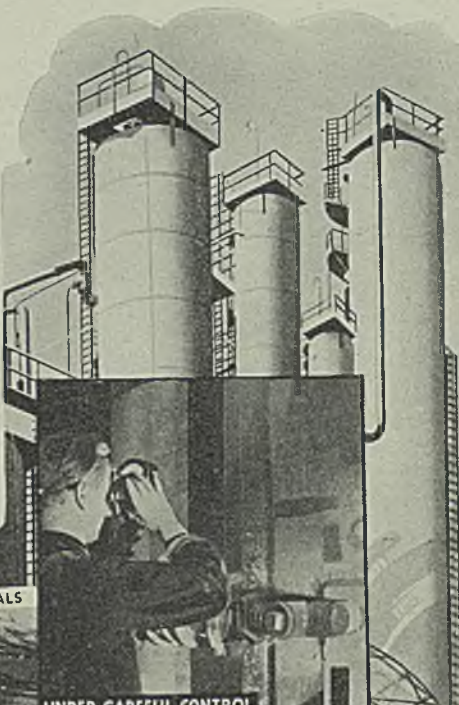
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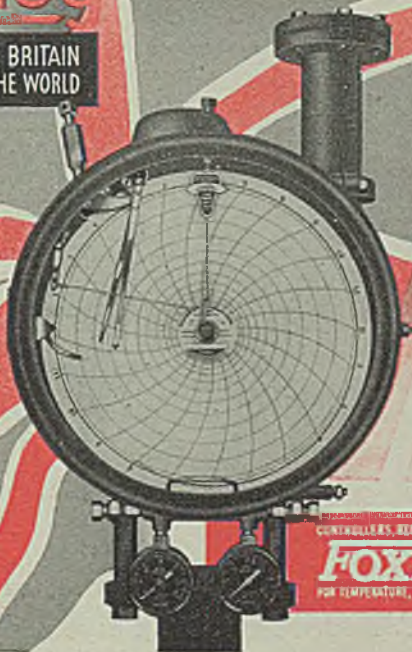
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