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MATHEMATICAL ANALYSIS OF THE EFFECT OF RETORTING PRESSURE ON OIL YIE

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Mathematical Analysis of the Effect of Retorting Pressure on Oil Yield and Rate of Oil Generation from Oil Shale

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The principal objective in the mathematical analysis presented here was to describe mathematically the oil yield, the amounts of oil degradation into coke and gas, and the rate of oil generation in the retorting of oil shale at various pressures. The results of the analysis are in good agreement with the experimental results obtained under various retorting conditions. In this analysis, the rate equations for the decomposition of organic matter, the recovery of liquid oil as oil mist or oil vapor, and the stoichiometry factors were determined from the experimental data on powdered oil shale with nitrogen as a sweep gas.

Introduction

In previous articles (Sohn and Yang, 1985; Yang and Sohn, 1985), the authors reported the results of an investigation on the effect of reduced pressure on the oil yield and the rate of oil generation from oil shale. In this paper, a mathematical analysis is presented to systematically describe the experimentally observed effects of pressure on the oil recovery rate and yield under various heating rates. The analysis was also aimed at predicting the amount and rate of coke and gas formation.

Stout et al. (1976) studied the effects of thermal histories on oil shale pyrolysis and showed that the oil yield was affected by the time–temperature history of the liberated oil and not by that of the organic matter in raw shale.

On the basis of the above result, Campbell et al. (1978) investigated the loss in oil yield due to reactions in the liberated oil. They observed that the oil degradation process occurred mainly in the liquid phase and not in the vapor phase. Same evidence was obtained by Rubel and Coburn (1982). Therefore, the present analysis considered only the oil in the liquid phase as a source of oil degradation.

It is known that not all of the organic carbons in raw shale are converted to oil and gas during retorting regardless of retorting conditions. This is in part due to the low H/C ratio of organic matter in shale and the presence of aromatic hydrocarbons in kerogen which remain relatively stable during retorting.

Miknis and Maciel (1981) observed by ^{13}C NMR that the residual carbons in retorted shale from several geologic formations mostly consisted or aromatic carbons.

Formulation of Rate Equations

The reaction mechanism formulated here consists of two important processes shown in eq 1–4; the decomposition of organic matter into oil, coke and gas, and the conversion of liquid oil into coke and gas, oil mist, and vapor oil. The oil vapor in equilibrium with liquid oil was considered as vapor oil.

$$\text{organic matter} \xrightarrow{k_1} \text{primary oil (liquid, vapor)} + \text{coke} + \text{gas} \quad (1)$$

$$\text{primary liquid oil} \xrightarrow{k_2} \text{coke} + \text{gas} \quad (2)$$

$$\xrightarrow{k_3} \text{oil mist} \quad (3)$$

$$\xrightarrow{k_4} \text{vapor oil} \quad (4)$$

This proposed mechanism is by no means meant to be a truly realistic or even a comprehensive representation of the extremely complex pyrolysis reaction. We formulated this mechanism, based on those simplified mechanisms proposed by previous investigators (Campbell et al., 1978), as the simplest version that quite successfully represents the quantities experimentally obtained in this work. Reaction 4 in the above scheme represents vapor formation by boiling. Under relatively high heating rates considered in this work, the vaporization of liquid oil component below the boiling point was neglected. Under a much lower heating rate, the latter which would depend on mass transfer rate (thus on gas flow rate and shale particle size) may have to be included.

The total amounts of coke and gas produced in reaction 1, which is referred to here as the primary reaction, are considered to be independent of retorting conditions. The stoichiometries of subsequent reactions, secondary reactions, are dependent upon retorting variables.

The rate of primary oil evolution during retorting depends only upon time–temperature history. Pressure affects not only the fractional distribution of liquid and vapor of the primary oil but also the degree of oil degradation during the secondary reaction.

As observed from the above, the primary oil undergoes two competing processes during retorting; the degradation into coke and gas, and the recovery into oil product as oil mist and vapor oil.

The vapor oil in reaction 4 is produced by physical boiling which takes place at a nonisothermal condition. In this analysis, vapor oil and oil mist are assumed to be immediately recovered out of the retort in the presence of a sufficient flow rate of an external sweep gas.

The mass balance equations are as follows

$$d\rho_1/dt = -k_1\rho_1 \quad (5)$$

$$d\rho_2/dt = k_1 f_1[1 - X(T,P)]\rho_1 - (k_2 + k_3 + k_4)\rho_2 \quad (6)$$

$$d\rho_3/dt = k_1 f_1 X(T,P)\rho_1 + k_4\rho_2 \quad (7)$$

$$d\rho_4/dt = k_1 f_2\rho_1 + k_2 f_4\rho_2 \quad (8)$$

$$d\rho_5/dt = k_1 f_3\rho_1 + k_2 f_5\rho_2 \quad (9)$$

$$d\rho_6/dt = k_3\rho_2 \quad (10)$$

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with the initial conditions; at $t = 0$, $\rho_1 = 0.1949$ (g/g of raw shale) and $\rho_2 = \rho_3 = \rho_4 = \rho_5 = \rho_6 = 0$ where $\rho_1, \rho_2, \rho_3, \rho_4, \rho_5, \rho_6$ are the amounts of organic matter, liquid oil, vapor oil, coke, gas, and oil mist, respectively.

The stoichiometry factors f_1, f_2, f_3, f_4, and f_5 were assumed constant during retorting, and the values were determined to be 0.836, 0.100, 0.047, 0.7038, and 0.2962, respectively. The factors f_1, f_2, and f_3 were determined by extrapolating to zero pressure using the results obtained at various reduced pressures with a heating rate of 12 K/min (Yang, 1983). At zero pressure, all produced oil would be in vapor phase and thus no degradation would occur. The factors f_4 and f_5 were adopted from previous investigation by Campbell et al. (1978). They determined the molar stoichiometry factors for each product from the degradation of liquid oil. Using these values, the stoichiometry factors for coke and gas formed from unit weight of liquid oil were calculated.

The kinetics parameters determined using experimental data obtained at various pressures (Sohn and Yang, 1985) showed that the activation energy of overall first-order kinetics for retorting Colorado oil shale was relatively constant over various pressures. Only the preexponential factor varied with pressure. On the basis of this information, the rate constant k_1 for organic matter decomposition was determined by extrapolating to zero pressure, which represents the true rate constant since all of the oil produced during retorting would be in the vapor phase and thus recovered as soon as formed.

With an average activation energy 46460 cal/g-mol, the second-order least-squares method was used to determine the best-fit value of preexponential factor at zero pressure using the experimental data. The rate expression thus obtained is

$$k_1 = 4.486 \times 10^{11} \exp(-46460/RT) \text{ s}^{-1} \qquad (11)$$

The rate constant k_2 for liquid oil degradation into coke and gas was adopted from previous investigation by Campbell et al. (1978)

$$k_2 = 3.1 \times 10^{7} \exp(-35000/RT) \text{ s}^{-1} \qquad (12)$$

The rate constant k_3 for recovery of liquid oil as oil mist is not available. This rate constant was determined by a repeated trial-and-error analysis with the aid of known kinetic parameters and stoichiometry factors. The best-fit expression was obtained by using all the experimental data on the overall oil yield under various pressures and heating rates. The expression thus obtained is

$$k_3 = 0.082P^{-0.25} \times \text{heating rate s}^{-1} \qquad (13)$$

where P is in atm and heating rate is in K/s.

The rate constant k_3 may also depend on the flow rate of sweep gas, especially at low levels. The expression given here for k_3 must be considered valid for the particular sweep gas flow rate used in this work (see the following section). The effect of flow rate on k_3 was not determined in this work. This may be a worthwhile topic for further research.

The rate constant k_4 for recovery of liquid oil as oil vapor by physical evaporation process was determined as a function of temperature at different pressures. To obtain this information, simulated distillation using gas chromatography was performed on the oil product obtained under 0.05 atm at a heating rate of 9.86 K/min under a sufficiently rapid nitrogen flow rate (60 cm^3/min at 25 °C and 0.86 atm). The 0.05 atm pressure was the lowest pressure which could be obtained for batch retorting in this work because of the pressure drop necessary for maintaining the nitrogen flow. The fractional distribution of vapor and liquid compositions of oil product produced at this pressure was almost independent of heating rate. It was thus assumed that the oil produced under these conditions represent the true primary oil unaffected by subsequent degradation. Using the normal boiling point distribution obtained from the simulated distillation, the vapor–liquid distribution at different pressures and temperatures can be calculated according to the procedure of Lee and Kesler (1980) and Maxwell and Bonnell (1957). The fraction of oil vapor whose boiling point is below T thus calculated, expressed as $X(T,P)$, is given as a polynomial function of temperature under various pressures in Table I. Using these polynomial equations for $X(T,P)$, the rate constant k_4 can be determined as follows

Table I. Polynomial Equations for $X(T,P)$ at Different Pressures.[a] **($X = C_1 + C_2 \times 10^{-2}T + C_3 \times 10^{-5}T^2 + C_4 \times 10^{-8}T^3$)**

press., atm	C_1	C_2	C_3	C_4
0.05	3.74533	−1.960	3.28563	−1.66
0.1	4.03221	−1.980	3.13637	−1.506
0.5	4.31868	−2.000	2.98711	−1.35
0.86	3.62511	−1.632	2.31250	−0.97
1.0	2.98155	−1.264	1.63789	−0.59
10.0	1.00062	−0.3755	0.37304	−0.035
20.0	−0.98032	0.513	−0.89191	0.52
30.0	−0.46529	0.236	−0.40782	0.24
40.0	−0.76264	0.381	−0.63104	0.35

[a] T is in kelvins.

$$k_4 = \mathrm{d}X(T,P)/\mathrm{d}T \times (\text{heating rate in K/s}) \text{ s}^{-1} \qquad (14)$$

The mass balance equations for a linear temperature increase with time were solved simultaneously by employing the fourth-order Runge–Kutta method (Carnahan et al., 1969) with a temperature interval of 0.25 K.

Comparison of Mathematical Analysis with Experimental Results

The experimental results were obtained from batch retorting under reduced pressures. A schematic diagram used for oil shale retorting and the experimental procedure have been reported elsewhere (Sohn and Yang, 1985).

About 70 g of oil shale sample (which came from the Anvil Points Mine in Colorado) with particle size of −8 to +48 mesh was charged in the retort and heated at heating rates from 1 to 12 K/min. The Fischer Assay grade of the shale was 36.6 ± 0.6 gal/ton. The properties of the raw shale were determined to be: organic carbon, 15.8%; hydrogen, 2.21%; nitrogen, 0.52%; and sulfur, 0.72% on weight basis of raw shale.

Nitrogen was used as a sweep gas at a flow rate of 60 cm^3/min corrected to 25 °C and 0.86 atm, which was sufficient to remove oil vapor and mist from the retort as soon as formed.

Shown in Figure 1 is the calculated and measured total oil yields as a function of pressure at different heating rates. Figure 2 shows the calculated and measured total oil yields as a function of heating rate at different pressures. As can be seen in these figures, the agreement is quite good at all the pressures and heating rates studied within 95% confidence.

Figures 3 and 4 show the comparison between the calculated and measured amounts of oil produced as a function of temperature at 0.05 and 0.86 atm, respectively, during retorting under a constant heating rate. The measured curves for oil yield were obtained using the overall first-order kinetics determined from experiments and are shown as dashed lines. The kinetics parameters

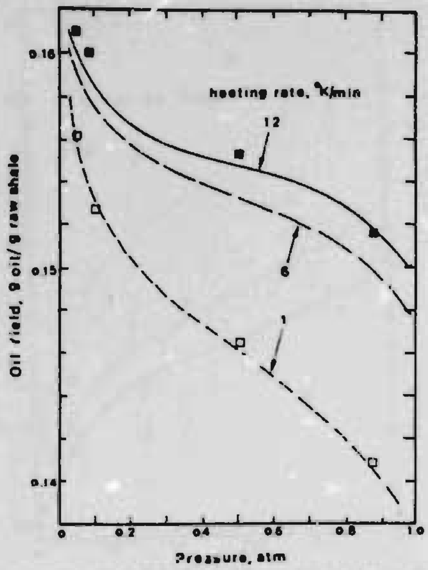

Figure 1. Comparison between calculated and observed oil yield as a function of pressure under reduced pressure. Experimental data were obtained at heating rates of 1 and 12 K/min.

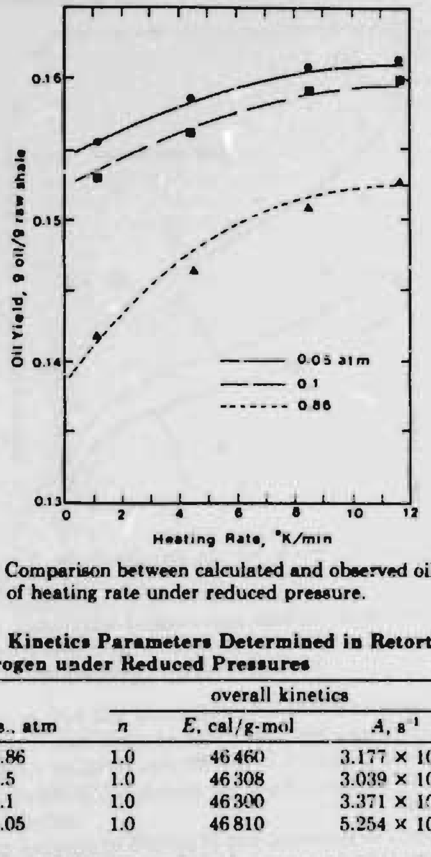

Figure 2. Comparison between calculated and observed oil yield as a function of heating rate under reduced pressure.

Table II. Kinetics Parameters Determined in Retorting with Nitrogen under Reduced Pressures

	overall kinetics		
press., atm	*n*	*E*, cal/g-mol	*A*, s^{-1}
0.86	1.0	46 460	3.177×10^{11}
0.5	1.0	46 308	3.039×10^{11}
0.1	1.0	46 300	3.371×10^{11}
0.05	1.0	46 810	5.254×10^{11}

measured at different reduced pressures have been reported elsewhere (Sohn and Yang, 1985) and are reproduced in Table II.

The calculated oil yield from the analysis represents the total amount of oil which is recovered as oil vapor and mist. From the figures, the agreement is considered to be good within 95% confidence. A close examination of Figures 3 and 4 reveals that the rate of oil generation is lower at higher pressure at both heating rates.

Figure 5 shows the calculated and measured amounts of coke produced as a function of pressure at heating rates of 1 and 12 K/min. The calculated result for a 6 K/min heating rate is also shown. All the values of coke and gas produced are expressed as weight percentage of raw shale. The amount of coke in the spent shale was determined by grinding a sample of spent shale and analyzing by a Perkin-Elmer 240 Elemental Analyzer for elemental compositions of C, H, and N. By using these values, the amount of coke produced from organic matter was determined. Although some differences are observed, the agreement can be considered to be satisfactory.

Figure 6 shows the comparison between the calculated and determined amounts of gas produced as functions of pressure at two heating rates of 1 and 12 K/min. Calculated result for 6 K/min heating rate is also shown. The observed data on the amount of gas produced were determined from material balance of water, oil, coke, and gas with the aid of the known amount of organic matter in the raw shale. A substantial deviation is observed between the

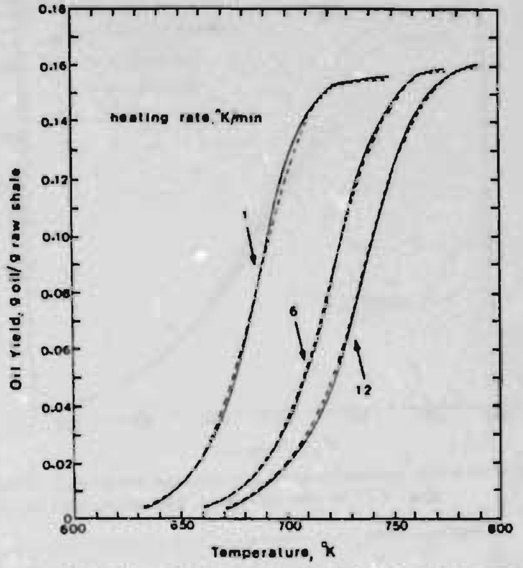

Figure 3. Comparison between calculated (—) and observed (---) oil yield as a function of temperature at different heating rates. The retorting pressure was 0.05 atm.

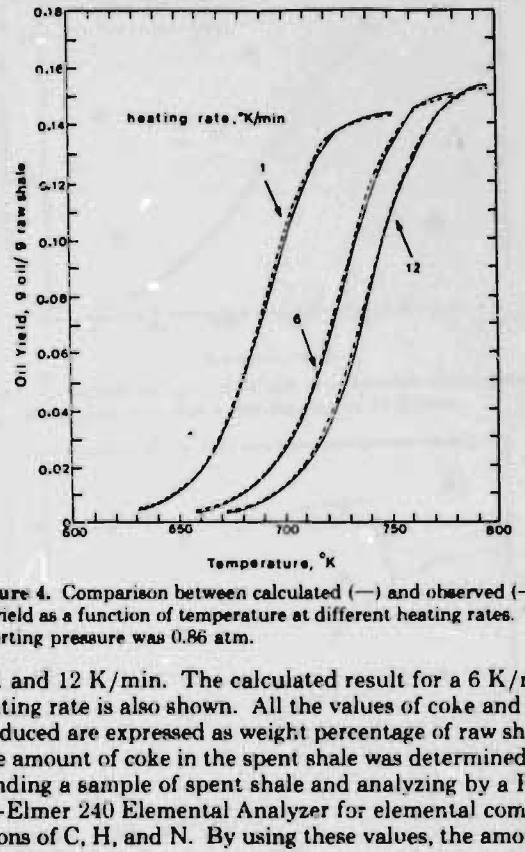

Figure 4. Comparison between calculated (—) and observed (---) oil yield as a function of temperature at different heating rates. The retorting pressure was 0.86 atm.

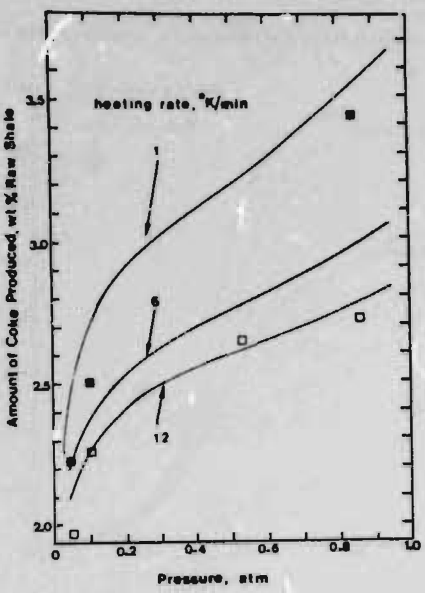

Figure 5. Comparison between calculated and observed amount of coke produced as a function of pressure. Experimental data were obtained at heating rates of 1 and 12 K/min.

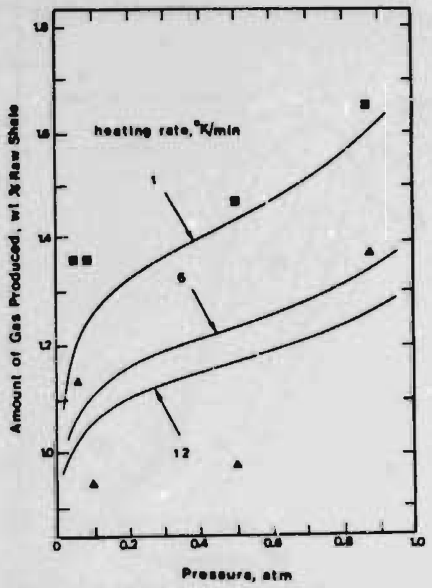

Figure 6. Comparison between calculated and observed amount of gas produced as a function of pressure. Experimental data were obtained at heating rates of 1 and 12 K/min.

two results, but the general trend is consistent.

Figures 7 and 8 show the coke and gas formation as functions of temperature at four different pressures. The general shape of the curves for the formation of coke and gas are similar.

As can be seen in Figure 9, the amounts of coke and gas produced at 0.05 atm with a heating rate of 12 K/min are much smaller than those with a heating rate of 1 K/min. This is because greater oil degradation occurs at a lower heating rate. The experimental data points of total amounts of coke and gas produced are also shown in the figure for comparison.

To show the amount of oil recovered as oil mist, Figure 10 was plotted using the data obtained from the analysis. About 80% of total oil produced was in the form of oil mist under 0.86 atm and 1 K/min heating rate, and about 60% under 0.1 atm and the same heating rate. These values are slightly higher than in the case of oil generation from tar sands (Oblad, 1983).

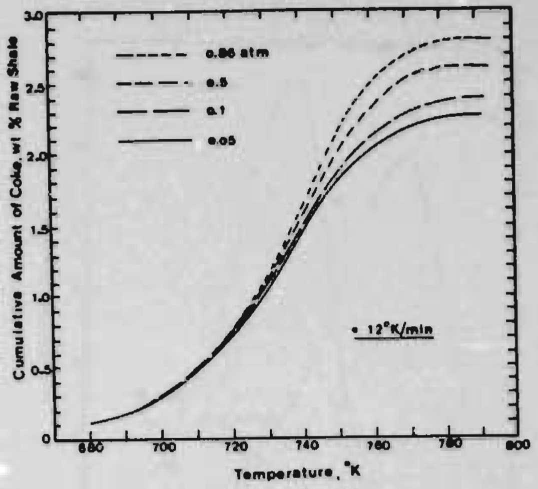

Figure 7. Calculated amount of coke as a function of temperature at different pressures with a heating rate of 12 K/min.

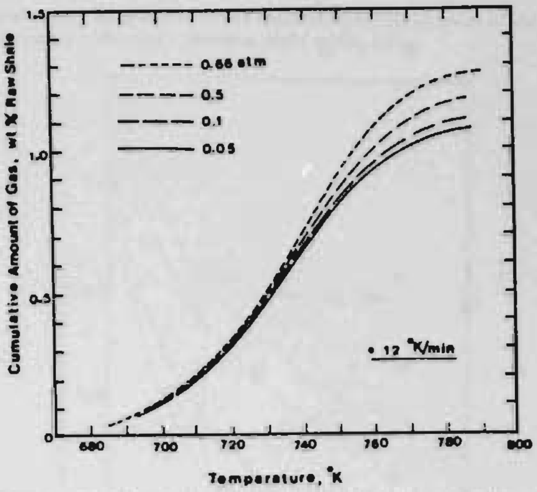

Figure 8. Calculated amount of gas as a function of temperature at different pressures with a heating rate of 12 K/min.

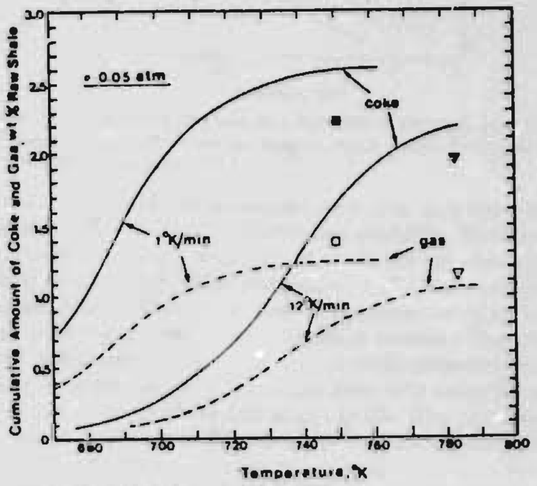

Figure 9. Calculated amounts of coke and gas produced as functions of temperature under 0.05 atm at two heating rates. The solid square and triangle, and open square and triangle represent the total percentages of coke and gas based on weight of raw shale, respectively.

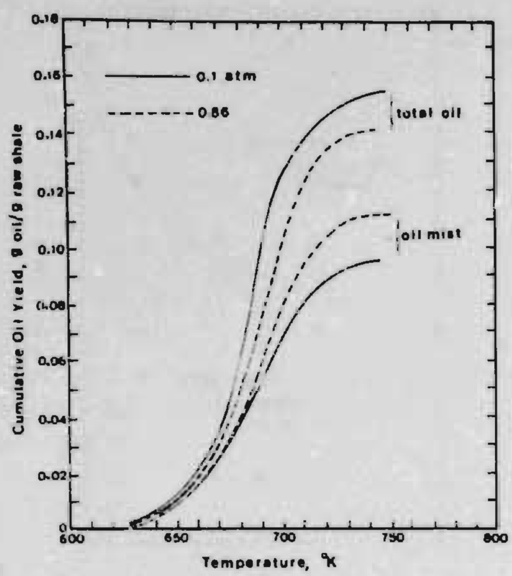

Figure 10. Comparison of total oil yield and oil recovery as oil mist as functions of temperature at different pressures. The heating rate was 1 K/min.

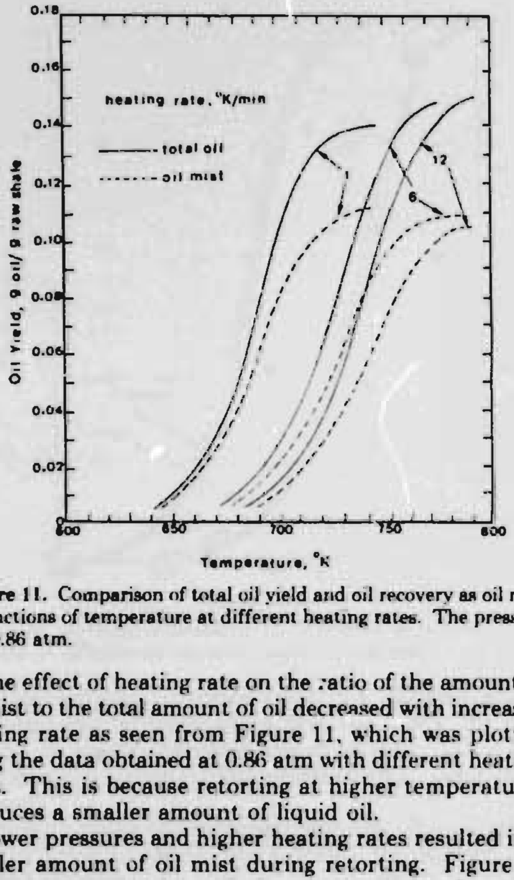

Figure 11. Comparison of total oil yield and oil recovery as oil mist as functions of temperature at different heating rates. The pressure was 0.86 atm.

The effect of heating rate on the ratio of the amount of oil mist to the total amount of oil decreased with increased heating rate as seen from Figure 11, which was plotted using the data obtained at 0.86 atm with different heating rates. This is because retorting at higher temperatures produces a smaller amount of liquid oil.

Lower pressures and higher heating rates resulted in a smaller amount of oil mist during retorting. Figure 12 shows the amount of liquid oil per unit weight of organic matter in raw shale as a function of temperature at different heating rates and pressures.

The retorting mechanism and values of parameters determined by using the experimental data obtained under reduced pressure were also used for the prediction of oil yield and rate of oil generation under elevated pressures.

Figure 13 shows the calculated oil yields under elevated pressure. The oil yield decreases significantly as pressure increases and heating rate decreases. For comparison between calculated and measured oil yields, only one data point for nonisothermal condition was available. Burnham and Singleton (1982) obtained 78 wt % FA oil yield at a heating rate of 1.8 K/min, a pressure of 27 atm, a pressed pellet of 3.2-cm diameters, and in the absence of an external sweep gas. For the comparison between the predicted and measured oil yield, the oil yield measured was corrected to 60 cm^3/min sweep gas flow rate using information reported by Campbell et al. (1978). The corrected data point is shown in Figure 13 as a black circle. The agreement can be considered to be good.

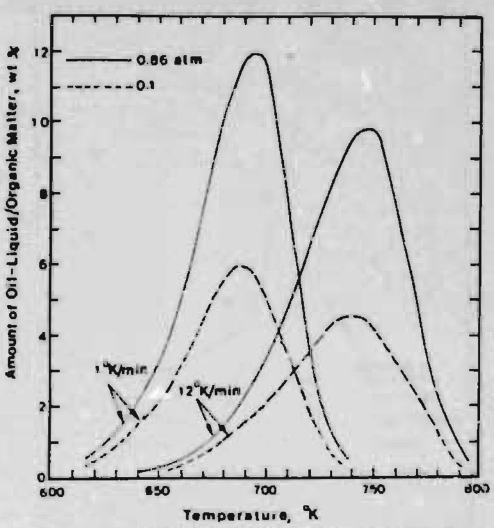

Figure 12. Comparison of the amount of oil liquid present during retorting at different pressures and heating rates.

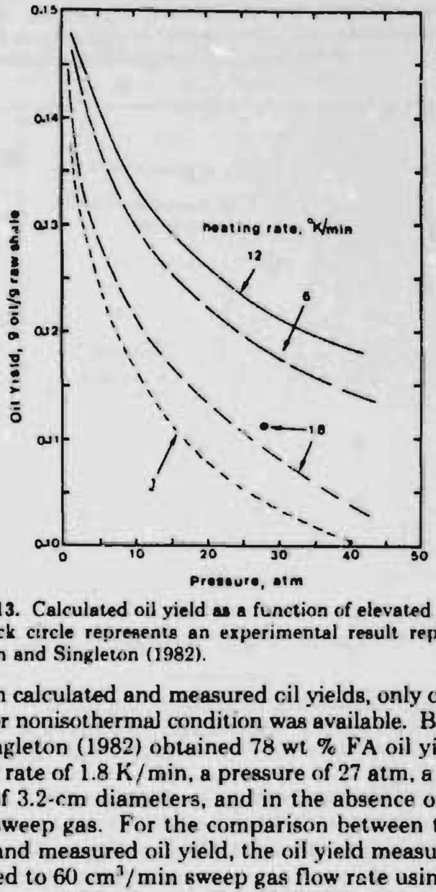

Figure 13. Calculated oil yield as a function of elevated pressure. The black circle represents an experimental result reported by Burnham and Singleton (1982).

Figure 14 illustrates the predicted and measured oil yields at elevated pressures. The measured oil yields are obtained by Bae (1969) under an isothermal condition with a constant temperature of 773 K and a constant residence time of nitrogen sweep gas in the retort. For comparison, the oil yield was expressed as a percentage based on the oil yield obtained at atmospheric pressure. Agreement between the two results is quite good up to about 30 atm. The increasing difference above this pressure might be due

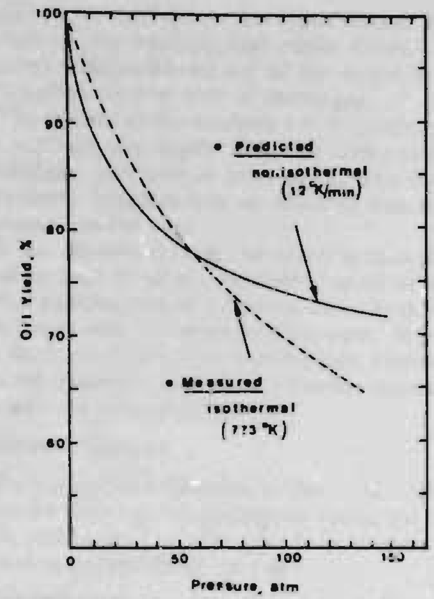

Figure 14. Comparison between calculated and observed oil yield under elevated pressure. The observed data were reported by Bae (1969).

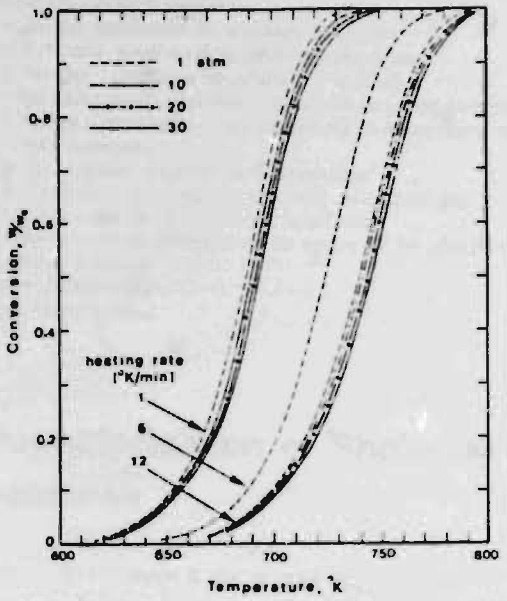

Figure 15. Calculated extent of oil conversion as a function of temperature at different elevated pressures and heating rates.

to the increasing importance of cracking of oil vapor, which is not included in this analysis, and also due to the fact the rate constant for the recovery of oil mist used in the present analysis may be less satisfactory at these high pressures.

Figure 15 shows the calculated amount oil generation as a function of temperature at different heating rates and elevated pressures.

For comparison of rate of oil generation at elevated pressure, two measured rates of oil generation are shown together with the rate of oil generation calculated by the analysis. Figure 16 shows the calculated and measured extents of oil conversion as a function of temperature. The observed data were obtained by Burnham and Singleton (1982), and retorting conditions are given in the figure. There were significant differences in the rate of oil generation. The difference in the rate of oil generation is likely

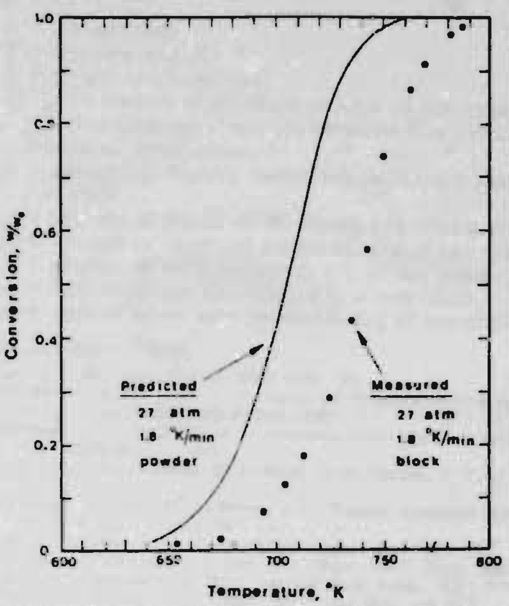

Figure 16. Comparison of calculated and observed extent of oil conversion as a function of temperature. The experimental results were reported by Burnham and Singleton (1982).

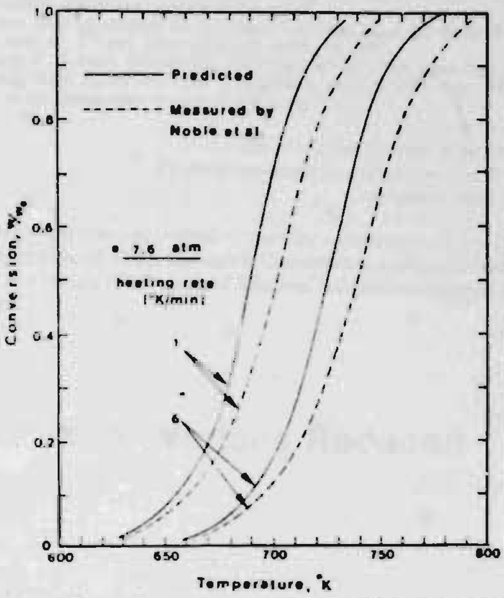

Figure 17. Comparison of calculated and observed extent of oil conversion as a function of temperature. The observed data were obtained using kinetics reported by Noble et al. (1981).

to be due to the absence of a sweep gas in their experiments, which lowers the rate of oil evaporation. This points to the significant effect of a sweep gas, especially at an elevated pressure.

The measured rate of oil generation shown in Figure 17 was obtained using the kinetics parameters determined by Noble et al. (1981). The difference between the calculated and measured rates may be due to the same reason mentioned above.

Conclusion

A mathematical analysis has been carried out to predict the oil yield, the amounts of coke and gas, and the rate of oil generation under reduced pressure. The analysis was based on a previous investigation which showed that the oil degradation into coke and gas mainly occurs in the liquid phase and not in the vapor phase. It was also assumed in this analysis that vapor oil and oil mist were recovered immediately out of the retort in the presence of a sufficient flow rate of sweep gas.

The results of the analysis are in good agreement with the experimental results obtained under various retorting conditions. However, at pressures greater than 80 atm the agreement becomes less satisfactory due to the reasons discussed in the text.

It was observed from the analysis that about 80% of total amount of oil was recovered as oil mist at 0.86 atm with a heating rate of 1 K/min, and about 60% resulted at 0.1 atm with the same heating rate. It indicates that the fraction of oil in the liquid phase decreases with decreased pressure, and thus a smaller amount of oil degraded into coke and gas.

Acknowledgment

The authors are thankful to Drs. A. G. Oblad and R. C. Aiken for their helpful discussions during the course of this work, and to Dr. S. Z. Song for help in analyzing the elemental composition of the coke.

Nomenclature

A = preexponential factor, s^{-1}
E = activation energy, cal/g-mol
f = stoichiometry factor
f_1 = for oil produced in primary reaction
f_2 = for coke produced in primary reaction
f_3 = for gas produced in primary reaction
f_4 = for coke produced from liquid oil in secondary reaction
f_5 = for gas produced from liquid oil in secondary reaction
k = rate constant
k_1 = for organic matter decomposition
k_2 = for liquid oil degradation into coke and gas
k_3 = for recovery of liquid oil as oil mist
k_4 = for recovery of liquid oil as vapor oil by physical evaporation process
n = reaction order
P = pressure, atm
R = gas constant
T = temperature, K
w = weight of oil evolved
$X(T,P)$ = fraction of initially generated oil that exists in vapor phase at pressure P and temperature T as determined by simulated distillation
ρ_1 = amount of organic matter left in the raw shale, g/g of raw shale
ρ_2 = amount of liquid oil produced, g/g of raw shale
ρ_3 = amount of vapor oil produced, g/g of raw shale
ρ_4 = amount of coke produced, g/g of raw shale
ρ_5 = amount of gas produced, g/g of raw shale
ρ_6 = amount of oil mist produced, g/g of raw shale

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Characterization of Shale Oil Produced under Various Reduced Pressures

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The crude shale oil obtained under reduced pressures by a nonisothermal technique was analyzed for physical and chemical properties. A lower pressure, higher heating rate, and higher flow rate of sweep gas resulted in a higher nitrogen content and higher values of viscosity, density, Conradson carbon, and pour point as well as a higher percentage of higher boiling distillates and a lower H/C ratio. The sulfur content remained relatively constant in the pressure range studied. As for chemical properties, a greater amount of paraffinic and aromatic compounds and increased aromaticity were observed at a lower pressure, while saturated isoprenoids decreased with decreased pressure. The olefinic fraction remained relatively constant under various pressures.

Introduction

A number of investigators (Allred, 1964; Hill et al., 1967; Bae, 1969; Weitkamp and Gutberlet, 1970; Burnham and Singleton, 1982) have studied the pressure effect on oil yield and properties produced at the conditions ranging from those encountered in in situ processing to those in Standard or Modified Fisher Assay experiments. Although many studies on the effect of pressure on oil properties have been carried out, there is little systematic study on the effect of reduced pressure.

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