Kinetics of oil-cracking for different types of marine oils from Tahe Oilfield, Tarim Basin, NW China

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Abstract

The C1–C5 gas and carbon isotope ratios generated during the cracking of heavy, normal and high-waxy marine oils from Tahe Oilfield, Tarim Basin, NW China, using a closed-gold tube system under high pressure were presented. The three types of oil tested resulted in a similar gas-generation process. The C2–C5 range initially increased its yield with the application of pyrolysis temperature and thereafter decreased, while the C1 yield increased with the same application. High-waxy oil had the highest C1–C5 yield of 510 mg/goil, whereas heavy oil had the lowest C1–C5 yield of 316 mg/goil. The δ13C1 value was low at first, but gradually became higher as the pyrolysis temperature increased. However, the δ13C2 and δ13C3 values gradually became higher when the temperature was greater than 420 °C. The kinetic parameters of the C1–C5 gas generation for the different types of marine oils were then calculated using KINETIC software. This calculation resulted in a frequency factor of about 1.78 × 10^{14} s^{-1}, while the distribution of the activation energy of the C1–C5 gas mass generated was relatively narrow with a range from 56 to 66 kcal/mol. Among the three types of oil tested, heavy oil had the widest activation energy distribution and the lowest major frequency of activation energy. The maximum temperature at which oil could be preserved as a separate oil phase varied from about 178 °C at a slow geological heating rate to 206 °C at a fast geological heating rate. This result is based on the kinetic parameters determined and in combination with the fractional conversion (\(\phi\)) of oil to gas. Testing conducted at the volatile Middle Cambrian reservoir of well Zhongshen 1 in the Tazhong Uplift strongly supported this conclusion.

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Keywords: Oil-cracking; Kinetics; Oil stability; Marine oil; Heavy oil; Normal oil; High-waxy oil; Tarim Basin

1. Introduction

Oil stability has been one of the hottest topics in the field of geochemistry [1,2]. Earlier work suggested that, due to the relative weakness of oil stability, the generation temperature for most oils and natural gases ranged from 100 to 150 °C and 150 to 220 °C, respectively [3]. In a confined system, the oil-cracking process starts at 150 °C. Hot reservoirs were discovered in the North Sea and the Gulf of Mexico in the middle and late 1990s where temperatures ranged from 175 to 200 °C [4,5], but these oils showed little evidence of cracking. The view that large-scale cracking starts at 160 °C and completely transforms to wet gas and pyrobitumen at 200 °C has been challenged by recent discoveries of hot reservoirs.

Pepper et al. [6] proposed that the success of oil-gas cracking was overestimated and that kinetic parameters were highly dependent on the oil composition and kerogen type of the source rocks. Schenk et al. [7] investigated the kinetics of gas generation from crude oils of lacustrine, marine and fluviolacustric origin. He then suggested that, in the absence of reservoir bitumen and minerals, severe oil-to-gas cracking was very unlikely to take place at temperatures less than 160 °C, regardless of the crude-oil type or geological heating rate involved. In China, the kinetic characteristics of heavy oil and
normal oil of the Tarim Basin, NW China, were investigated [8–15], but comparisons amongst different types of oils were found to be relatively weak.

The effects of pressure on some individual compounds were also investigated [16–19]. Fabuss et al. [16] argued that for n-hexane and n-heptane, increasing pressure initially tended to speed up the oil-cracking rate from 20 to 40 bar and thereafter decreased this rate to between 100 and 800 bar. The pressure effect initially increased the thermal degradation rate of n-C_{25} from 120 to 400 bar and then decreased this degradation rate from 400 to 800 bar to its value at 120 bar [17]. Jackson et al. [18], however, proposed that in pressure ranges of 120–600 bar, the pressure always suppressed the cracking of n-C_{16}. Al Darouich et al. [19] further investigated the effect of pressure on thermal cracking of the light (C_{6}–C_{14}) aromatic fraction (LAF) of crude oil. They found that increased pressure decreased the cracking of the unstable molecular classes of LAF as well as secondary cracking of some newly formed heavier compounds (C_{15}–C_{20} and C_{20}+ polyaromatics). The simulated application of thermal cracking of light aromatics at the HP/HT reservoir of Elgin (North Sea) illustrated that pressure effects could shift thermal cracking of aromatic to higher temperatures by about 8 °C [19]. Now, it is widely accepted that high pressure suppresses oil cracking.

Earlier work employed individual alkane compounds in cracking experiments to investigate oil stability. These cracking experiments on aromatic compounds began in the late 1980s. Compared to the alkane compound, the aromatic compound had a lower activation energy and frequency factor [20,21]. As far as the kinetics model is concerned, the component kinetics model that divided the oils into similar component classes (e.g., Kuo and Michael [22] divided oils into C_{1}, C_{2}, C_{3}–C_{6}, C_{6}–C_{14}, stable aromatic, C_{15}+ saturated, C_{15}+ resin, asphaltene, and pyrobitumen) has been widely used in the kinetics commercial software. Domine et al. [23] proposed the free-radical reaction mechanism to explain oil stability. They concluded that mature oil would be stable from 240 to 260 °C, depending on their composition, and that thermal cracking of oil to gas was not possible under reasonable basin conditions [24].

With exploration moving to deep strata, the exploration prospect of the deep Cambrian received far more attention. The well Tashen1 is the deepest well drilled by Northwest Corporation of SINOPEC in the Akekule Uplift. A little liquid hydrocarbon was obtained from the Cambrian dolomite reservoir at depths of 8406 to 8408 m using a chloroform-cooled extract and discovered an excellent dolomite reservoir at depths greater than 8000 m [25]. The exploration results conducted at the well Tashen1 encouraged both the geochemists and the geologists to focus on the exploration depth limits of deep strata [15,26]. Different types of marine oils (heavy oil, normal oil and high-wax oil) from Tahe Oilfield, NW China, were tested by using golden-tube simulation devices for this paper. Based on the kinetic parameters of gas generation calculated by KINETICS software, the temperatures for the preservation of the separate oil phases of the different types of oil were discussed.

2. Samples and methods

2.1. Samples

The Triassic normal oil from well T915, the Ordovician waxy-oil from well T901, and the Ordovician heavy oil from well T740 were selected from Tahe Oilfield, NW China. The basic geochemical data of the above three oils are listed in Table 1. The Ordovician heavy oil from well T740, with sulfur content of 2.29% (high-sulfur oil), suffered significant biodegradation. Despite the difference in the physical properties, the three oils showed great similarity in the biomarker composition.

<table>
<thead>
<tr>
<th>Well name</th>
<th>T915</th>
<th>T901</th>
<th>T740</th>
</tr>
</thead>
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<td>4603–4606</td>
<td>5820–5875</td>
<td>6260–6290</td>
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<td>(O_{2yj})</td>
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<tr>
<td>Viscosity/mPa s</td>
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<tr>
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<td>Sulfur content/%</td>
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<td>O</td>
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<td>7.10</td>
<td>41.69</td>
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<th>Aromatic fraction</th>
<th>Resin</th>
<th>Asphaltene</th>
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<th>C_{28} 20R</th>
<th>C_{29} 20R</th>
<th>C_{29} 20S(20S + 20R)</th>
<th>Ts/(Ts + Tm)</th>
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<tr>
<td>C_{27} 20R</td>
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<td>45</td>
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<td>52</td>
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<td>52</td>
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distribution, suggesting that they came from the same rock source. Differences in the physical properties of the oils resulted from the different pool-forming periods and a secondary alteration after the pool forming process.

2.2. Pyrolysis experiment

The pyrolysis device used was a closed, temperature-programmed, gold-tube system similar to that used in Refs. [8,11,27,28]. The pyrolysis temperatures were programmed from 200 °C to 620 °C. A certain amount of oil samples (10–30 mg) were sealed into the gold tubes (40 mm length, 4.5 mm diameter) by welding these in an argon environment. The gold tubes were placed in pressurized autoclaves, filled with water with a high-pressure pump, and then the autoclaves exerted pressure on the samples. A constant confining pressure of 50 MPa was maintained throughout the entire pyrolysis experiment via an adjustable pressure sensor. The autoclaves were then heated to preset temperatures using programmed heating (heating rates were 20 °C/h and 2 °C/h). The temperature and pressure of the autoclaves were controlled by a computer, with a temperature error of 0.1 °C and pressure error of less than 1 MPa.

After pyrolysis, the gold tubes were placed in a piercing device that was attached to a vacuum line equipped with a pump. The gas collection was initiated by puncturing the gold tubes in a vacuum and injecting the gas into a gas chromatograph after about a 30-s delay to achieve gas balance. The gaseous hydrocarbons (C₁–C₅) were analyzed using a HP 5890 II gas chromatograph instrument and quantified using the inner-marker method. The GC conditions applied were: Poraplot Q capillary column (30 m × 0.25 mm × 0.25 μm) with helium as the carrier gas. The gas was injected at 50 °C and maintained for two minutes, subsequently heated to 180 °C at a rate of 4 °C/min, and thereafter maintained at 180 °C for about 15 min.

The stable carbon isotope analysis of gaseous hydrocarbons was performed using the Isochrom II spectrometer coupled to a HP 5890 gas chromatograph. The GC conditions applied were: Poraplot Q capillary column (30 m × 0.32 mm × 0.25 μm) with helium as the carrier gas. The gas was injected at 50 °C and maintained for three minutes, subsequently heated to 150 °C at a rate of 4 °C/min, then maintained at 150 °C for about eight minutes. Each sample was analyzed at least two or three times, and the standard deviation of the replicates was less than ±0.3‰ (PDB).

2.3. Modeling of kinetic parameters

The kinetic parameters of cumulative C₁–C₅ gaseous hydrocarbons in masses of different types of marine oil were determined and computed using the KINETICS software developed by the Lawrence Livermore National Laboratory (LLNL). The discrete model was adopted in this study because of its good fit with the closed system experimental data [8–10,28].

3. Results

3.1. Yield of gas generation from different types of marine oils

Fig. 1 presents the yields of C₁–C₅, methane, and C₂–C₅ gas generation in mass with pyrolysis temperatures under two different heating rates 2 °C/h and 20 °C/h.

The yields of gas generation in mass from the three types of oil were closely related to the pyrolysis temperature and heating rate. In general, the yield of methane generation gradually increased in mass as the pyrolysis temperature increased (or the thermal maturity increased). The yield of methane generation under the slow heating rate (2 °C/h) was greater than that under a fast heating rate (20 °C/h).

The three types of marine oils had a high yield of gas generation. Among the three types, the Ordovician high-waxy oil from well T901 showed the best gas generation, with a yield of 464 mg/goil of methane at 602 °C under the slow heating rate (2 °C/h). The Triassic normal oil from well T915 had a similar yield of methane generation to that of well T901, with a cumulative value of 434 mg/goil at 603 °C (Fig. 1b). The heavy oil generated the lowest yield of methane generation with a value of 316 mg/goil at a temperature of 605 °C (Fig. 1c).

A certain amount of C₂–C₅ was generated from the three oils, with a maximum value obtained in the temperature range of 460–500 °C. The Ordovician high-waxy oil from well T901 had the highest yield of C₂–C₅ gas generation with a value of 388 mg/goil. The Triassic normal oil from well T915 had a lower yield of C₂–C₅ gas generation with a value of 346 mg/goil, and the Ordovician heavy oil had the lowest yield of C₂–C₅ gas generation with a value of 212 mg/goil.

3.2. Stable carbon isotope of the C₁–C₅ gas generation

Fig. 2 shows the stable carbon isotopes of methane, ethane, and propane generated during the oil pyrolysis of oil from Tahe Oilfield, NW China.

The value of the stable carbon isotopes of methane, ethane, and propane gas generated from the pyrolysis of all three oils tested showed a close relationship between the pyrolysis temperature and the heating rate. Taking the stable carbon isotope of methane as an example, when the temperature is lower than 400–420 °C, the value of stable carbon isotopes of methane under the fast heating rate (20 °C/h) was 0.3‰–2‰ higher than that obtained under the slow heating rate (2 °C/h). When the temperature was greater than 420 °C, however, the value of the stable carbon isotope of methane collected at a fast heating rate was 1‰–3‰ lower than that obtained at a slow heating rate.

Under the same heating rate and temperature, regardless of the oil type, the gas from the same oil sample showed a positive stable carbon isotope distribution (i.e., δ¹³C₁ < δ¹³C₂ < δ¹³C₃).

The variation of the stable methane carbon isotope of the Ordovician heavy oil-cracking gas was the smallest, with a
Fig. 1. Characteristics of gaseous hydrocarbons pyrolyzed from oils from Tahe Oilfield, Tarim Basin.

Fig. 2. Carbon isotopic characteristics of methane, ethane, and propane pyrolyzed from oil from Tahe Oilfield, Tarim Basin.
range from $-49.10\%$ to $-39.12\%$, whereas the range of the stable methane carbon isotope of the Ordovician high-waxy oil-cracking gas was the widest with a value from $-56.45\%$ to $-39.54\%$. In general, as the pyrolysis temperature increased (or the thermal maturity increased), the stable methane carbon isotope of the gas from the three types of oil cracking decreased at first, then increased with the maximum temperature of 400–420 °C at the reversion point of the isotope corresponding to a Ro of $1.5\%$–$1.7\%$ measured in the simulation experiment. This result was in accordance with the changes of the carbon isotope of the methane reported at home and abroad. Compared with the carbon isotope of methane derived from kerogen-cracking in Tarin Basin (temperature of 371 °C at 2 °C/h heating rate) at the reversion point of the isotope, the temperature of the reversion point of the isotope from oil cracking was higher than that of the kerogen cracking.

The value of stable methane carbon isotope showed a good relationship with the pyrolysis temperature (or equivalent Ro) after reaching a temperature greater than 400–420 °C. The stable carbon isotopes of ethane and propane increased with the temperature when the temperature was greater than 420 °C. Comparatively, the stable carbon isotope of ethane showed a better relationship with the pyrolysis temperature.

Compared with the stable methane carbon isotope of the gas generated from kerogen cracking, the value of the stable methane carbon isotope of gas derived from oil cracking was far lower under the same pyrolysis temperature. Using a heating rate of 2 °C/h as an example, after the reversion of the isotopes, the range of stable methane carbon isotope from oil cracking ranged from $-56.45\%$ to $-37.69\%$, whereas the value of the stable methane carbon isotope generated from kerogen cracking ranged from $-44.69\%$ to $-29.72\%$. One explanation of this phenomenon is that during the transforming process from kerogen to oil, the isotope suffered fractionation to some degree. In addition, because most of methane derived from the cracking of the heavy gaseous hydrocarbon (i.e., $C_2$–$C_3$) occurred in the cracking stage, the value of the carbon isotopes of the heavy hydrocarbons $C_2$–$C_3$ are heavier than those of the whole oil, leading to the methane having a light stable carbon isotope.

**3.3. Kinetic parameters**

Bulk kinetic parameters of the gaseous hydrocarbons in mass ($C_1$–$C_5$) obtained from experiments were different from those obtained in an open system. However, they appeared effective in evaluating the gas generated from oil cracking [29]. Using KINETICS software, the kinetic parameters, including activation energy distribution and frequency factor of $C_1$–$C_5$ generation in mass, were analyzed for the marine oils. The gas yields in mass were normalized before the kinetic calculation. The best fit between the experimental and calculated data and the results of the discrete activation energy distribution (Ea) and frequency factors (A) from the different types of marine oil are shown in Fig. 2. On the premise of frequency factor $A$ of $1.78 \times 10^{14}$ s$^{-1}$ proposed by Waples [30], the range of activation energy distribution of the three types of oil were relatively narrow with values of 56–66 kcal/mol (Fig. 3). The range of activation energy distribution of the Triassic normal oil, the Ordovician waxy-oil, and the Ordovician heavy oil were 59–61 kcal/mol, 60–62 kcal/mol, and 56–66 kcal/mol, respectively. Among the three types of marine oils, heavy oil had the widest range of energy activation distribution and the lowest main frequency energy activation. The above kinetic parameters can be well-fitted with the experimental results.

**4. The maximum temperature for preservation of oil as a separate phase in Tarin Basin and geological evidence**

**4.1. Maximum temperature for preservation of oil as a separate phase**

The thermal stability of crude oil has two implications. One is the disappearance of the separate phase oil, i.e., the maximum temperature at which a pure oil pool can preserve, and the other is the temperature at which all the moveable liquid hydrocarbons almost disappear. Claypool & Mancini [31] suggested an equation between the thermal destruction of crude oil and the gas oil ratio (GOR).

$$C = \frac{GOR}{(GOR + 3000)}$$

where $C$ is the fraction of original oil that has been destroyed and GOR is in scf/bbl.

The value 3000 is an assumed average. The actual value depends on the density and molecular weight of the oil. Furthermore, different oils produce different contents of gas in oil cracking [7,29,32,33], different PVT conditions would result in differences in mutual solubility [6], gas may leak away when it is generated, or gas may originate from other sources besides oil cracking. All of the above reasons can make the value deviate from 3000 [30,34].

McCain and Bridge [35] proposed that a GOR of about 3200 scf/bbl (i.e., 570 m$^3$/m$^3$) is the maximum at which a free oil phase can exist in a reservoir. Under the above conditions, the percentage of the oil cracking is about 51%. Hunt [36] suggested that the transition from oil to gas in a given reservoir occurs when the GOR is greater than 5000 scf/bbl (i.e., 891 m$^3$/m$^3$). At this level of oil cracking, C would be about 62.5%. It is noteworthy that according to the above definitions, a large number of oil-like molecules can exist after the separate liquid phase we call “oil” has disappeared [6]. When C is greater than 62.5%, a separate liquid phase cannot exist in a reservoir. In such circumstances, cracking involves changes in the proportion of condensate in a gas, while the heavy hydrocarbons in the gas phase are progressively transformed into gas-size molecules.

By using the frequency factor (A) of about $1.78 \times 10^{14}$ s$^{-1}$ and narrow Gaussian distribution of Ea (mean activation energy = 59 kcal/mol, $\sigma = 1.5$ kcal/mol), Waples [30] calculated the maximum geological temperature of the separate oil phase. Although the difference of $C$ is 11.5%, the
difference of the maximum geological temperature of the separate oil phase is only about 3–4 °C.

Fig. 4 and Table 2 show the relationship between the kinetic parameters of mass C$_1$–C$_5$ generated from the three types of oil from Tahe Oilfield and the maximum temperatures for the preservation of oil as a separate phase at various heating rates.

From Table 2, the geological temperature range for the preservation of oil as a separate phase is from 178 to 206 °C at average geological conditions. The temperature difference is only 2–6 °C according to two different C values (C = 51% and C = 62.5%). From a slow heating rate of 0.5 °C/Ma to a fast heating rate of 10 °C/Ma, the temperature range of the oil as a separate phase is from 178 to 185 °C and 199 to 206 °C, a 8–12 °C and 4–8 °C difference from the results of the Waples [30] and Tian et al. [11], respectively. The temperature difference was the result of the activation energy distribution and the values employed in the calculation. Waples [30] suggested a Paleogene-Neogene petroleum system could be preserved at a temperature range from 188 to 190 °C, and in very rapid sedimentation rates the oil phase can exist at a temperature of 200 °C.

Fig. 3. Kinetic parameters of C$_1$–C$_5$ gas generation in mass of oils from Tahe Oilfield, Tarim Basin.

Fig. 4. Temperature at which liquid oil disappears as a separate phase for a range of possible geological heating rates.
dominated by alkane gases with a methane content of 68.6% and dryness coefficient of about 0.778. The gas also has a relatively high content of CO$_2$ with a value of 10.9% and low content of N$_2$ with a value of 0.795%.

Although at present the reservoir temperature of the Middle-Lower Cambrian of well Zhongshen 1 ranges from 160 to 165 °C, in geological time the Cambrian suffered high paleo-temperature periods twice; one was in the Ordovician, whose paleo-temperature gradient was about 35–40 °C/km, and the other was in the Permian, whose gradient reached 34–38 °C/km [37]. According to the paleo-temperature of well Tacan 1 from the Tazhong Uplift, the paleo-temperature of the Middle Cambrian nearly reached 180 °C in the Late Hercynian, whereas the paleo-temperature of the Lower Cambrian was higher than 180 °C. Knowing that the Cambrian reservoir formed in the Early Caledonian era, the existence of the volatile reservoir of the Middle Cambrian from well Zhongshen 1 gives strong evidence that the Cambrian reservoir could be a separate oil phase at a temperature near 180 °C.

5. Conclusions

(1) During the oil-cracking process, three types of oil from Tahe Oilfield, NW China, generated very high contents of C$_1$–C$_5$ gaseous hydrocarbon and an abundance of C$_2$–C$_5$, of which the high-waxy oil possessed the highest yields. The different types of oil generated similar gases in the oil-cracking process, with methane generation increasing with temperature and C$_2$–C$_5$ increasing at first then decreasing with temperature.

(2) The stable carbon isotopes of the gases derived from oil cracking of the three oil types of Tahe Oilfield showed a positive isotope distribution, that is, $\delta^{13}$C$_1 < \delta^{13}$C$_2 < \delta^{13}$C$_3$. The values of stable carbon isotope of ethane and propane increased with the pyrolysis temperature when the temperature exceeded 420 °C. Among the three oils, the range of stable methane carbon isotope of the oil cracking from well T740 was the narrowest.

(3) On the premise of a frequency factor A of $1.78 \times 10^{14}$ s$^{-1}$, the range of activation energy of mass C$_1$–C$_5$ generated from the three types of oil was narrow with a value of 56–66 kcal/mol. Comparatively, heavy oil had the widest activation energy distribution and the lowest main frequent activation energy with value of 59 kcal/mol.
Using KINETIC software, the maximum temperature at which oil could be a separate phase varied from 178 to 206 °C during average geological conditions. The existence of a volatile reservoir in the Middle Cambrian of well Zhongshen 1 of the Tazhong Uplift provides strong evidence for this conclusion.

Conflict of interest

The author declare no conflict of interest.

Foundation item


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