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Research Article

Quantitative Method for Evaluating Shale Oil Resources Based on Movable Oil Content

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The light hydrocarbon content (S_1) of shale oil is lost in aboveground experimental measurements, which do not accurately reflect actual underground light hydrocarbon content and cannot meet the demands of resource quantity calculation. Based on field and laboratory experimental data from the second member of the Kongdian Formation in the Cangdong Sag, Bohai Bay Basin, the retained oil and movable oil contents in shale were calculated using a mathematical formula, and the total resources were quantified. The correction coefficient of S_1 from pyrolysis and the adsorption of oil by the total organic carbon (TOC) were determined to be 1.25 and 100 mg/g, respectively. The calculated parameter movable oil content (S_{movable}) and the corresponding calculation formula of S_1 are proposed. The lower limit of S_1 is 100 mg HC/g rock, the TOC content is 4 wt % and 6 wt %, and the corrected movable oil content of 3 mg/g and 6 mg/g, respectively, will be of great significance in shale oil geology and engineering. The optimal geological and engineering settings are divided into three categories and eight subcategories on the basis of these findings. Methods for evaluating total, movable, and recoverable oil resources from shale are discussed, which elucidate a new method for quantitative evaluation and ranking of shale oil resources. This approach is suitable for application in other shale oil exploration and development areas globally.

1. Introduction

Compared with North American shale oil, China's lacustrine oil shales are quite different in terms of reservoir and fluid properties and reservoir transformation characteristics [1, 2]. Lacustrine shales in China are mainly grayish and argillaceous shales [3]. The total organic carbon (TOC) content varies greatly from 0.7% to 28%; the vitrinite reflectance is generally lower than 1.0%; the light oil content is also relatively small; the porosity and permeability are relatively poor, and the pore throat diameter is less than 300 nm; the fluid viscosity is up to 350.0 MPa·s; the density is between 0.67 and 0.94 g/cm^3 [4–6]. It is of great significance to understand shale oil resources, especially movable resources, using current engineering technology [7]. There are three principal evaluation methods for shale oil resource evaluation: analogy, volumetric, and genetic methods [8]. The analogy method is used to estimate oil and gas resources by comparing it with oil and gas fields with similar geological characteristics. The volumetric method calculates the oil volume in a unit volume of rock and has higher requirements for data in the study area. The genetic method estimates hydrocarbon generation resources by simulating hydrocarbon generation methods, constrained by experimental objects and methods. S_1 is the key parameter for shale oil bearing evaluation, which is consistent with the concept of oil saturation in sandstone reservoirs. S_1 characterizes shale-free oil [9, 10]. Pyrolysis S_1 combined with the oil saturation index OSI (S_1/TOC) is the key parameter for shale oil bearing and mobility evaluation [11, 12]. Generally, the parameters are obtained from the wellhead cuttings, but the cuttings may be lost in the process of formation breaking from the bottom hole to the wellhead and placement. The saturation index should be the index of oil and gas mobility rather than the actual momentum. Methods for accurately correcting S_1 parameters to reflect the quantity of free oil in the underground shale reservoir and quantitatively characterizing the free oil parameters of shale oil are required. Based on the data of closed cores and cuttings, combined with field and indoor pyrolysis experiments and swelling experiments, this study discussed the calculation of the S_1 recovery coefficient, shale oil potential, and shale oil resources with the aim of establishing a suitable method for evaluating China's lacustrine shale oil resources.

2. Geological Setting

The Cangdong Sag is one of several oil-rich sag areas in the Bohai Bay Basin. It is located in the southwest of the Huanghua Depression. It is sandwiched between the Cangxian uplift in the west, Xuhei bulge in the east, Kongdian bugle in the north, and Dongguan uplift in the south (Figure 1). It is a continental faulted lake basin formed by regional extension during the Cenozoic. During the sedimentary period of the second member of the Kongdong Formation, the Cangdong Sag contained an inland freshwater to brackish water elliptical closed lake and the input of clastic materials from the four major provenances (Kongdian, Cangxian, Dongguan, and Xuhei uplifts) around the basin, the sedimentary facies showed regular changes from the edge of the lake basin to the middle of the lake basin, and the outer ring comprises extensive braided river delta deposits (yellow-orange colors in Figure 1) [13, 14]. The middle ring comprises a dense zone composed of silty mudstone, siltstone, and dolomite, and the inner ring consists of fine-grained sediments composed of thick dark shale with thin layers of siltstone and dolomite. These sediments are mainly fine-grained sedimentary rocks (>50%) with particle sizes less than 0.0625 mm and are rich in preserved organic matter, which is the source of oil in the shale (Figure 1).

3. Samples and Experiments

All samples were obtained from the Kong 2 member in the Cangdong Sag. To determine the amount of oil adsorbed by kerogens with different maturities, 12 samples were taken from the core of G19 well for the swelling adsorption experiment, which is described below. To determine the rate of loss of light hydrocarbons, 46 pieces of cuttings from the G25 well closed coring section and corresponding depth wellhead cuttings were subjected to thermal extraction. Experiments were conducted to analyze the distribution of light hydrocarbon content and calculate the movable light hydrocarbon content. A total of 808 cutting samples were taken, including 192 at 3939 m depth in horizontal well GY1-3-1, 274 at 3714 m depth in horizontal well GY2-1-1, 207 at 3702 m depth in horizontal well GY2-1-2, and 135 at 3777 m depth in horizontal well GY2-1-4 (Table 1; wells are marked in Figure 1).

3.1. Swelling Adsorption Experiment. The inner diameter of the screen tube was 60 mm, and the height of the screen tube was 60 mm. In the experimental process, the sample was crushed and ground to 120 mesh and dried in an oven; 200 mg of sample was placed into the test tube and centrifuged for 30 min at 8000 rpm, and the height (H_1) of the sample was measured. The glass tube with a sieve hole at the lower end was placed into a centrifuge tube containing an organic solvent, and the sample was centrifuged for 30 min. The kerogen was soaked at 30°C for 24 h and then centrifuged under the same conditions. The height after swelling (H_2) was measured, and the swelling ratio ($Q_v = H_2/H_1$) was calculated. The total hydrocarbon retention and swelling ratio of kerogen can be converted according to the following formula [15]:

$$M = \frac{(Q_v - 1.0)\rho_1}{\rho_2}, \tag{1}$$

where M is the amount of hydrocarbon retained in unit kerogen (g/g); Q_v is the swelling ratio; ρ_1 is the weighted average density of n-tetradecane, o-xylene, acetic acid, isopropanol, and ethanol retained in kerogen (0.9 g/cm^3; and ρ_2 is the density of kerogen (1.28–1.35 g/cm^3), corresponding to a vitrinite reflectance (Ro) 0.5–1.3%.

3.2. TOC, Thermal Extraction, and Pyrolysis. The TOC values were measured using a CS-600 organic carbon analyzer, following the Chinese National Standards GB/T 19145–2003 [3] after the removal of carbonate minerals with acid.

A Vinci Technologies Rock-Eval 6 instrument was used for thermal extraction (S_1) and pyrolysis (S_2) following a procedure similar to that of Lin et al. [16] and Hood et al. [17]. For S_1, the oven was kept isothermally at 300°C, and for S_2, the oven temperature was increased to 550°C at a rate of 25°C/min. The experiment was completed at the Wuxi Institute of Petroleum Geology, Research Institute of Petroleum Exploration and Development, Sinopec. The steps are as follows: Soxhlet extraction to remove soluble organic matter from source rocks, acid dissolution to remove inorganic minerals, ultrasound for 30 min to fully saturate adsorption, and high-speed centrifugation for 30 min to remove the nonadsorbed part and yield the saturated adsorption capacity.

4. Results

4.1. Thermal Extraction and Pyrolysis. There were 46 samples in total; the TOC values were evenly distributed, the minimum value was 0.67% wt, the maximum value was 7.29% wt, and the average value was 3.49% wt. The minimum pyrolysis S_1 value measured by closed coring well samples was 0.34 mg HC/g rock, and the maximum pyrolysis S_1 value was 6.84 mg HC/g rock, with an average of 1.19 mg HC/g rock. Corresponding to the depth of cutting samples, the minimum pyrolysis S_1 value is 0.30 mg HC/g rock, and the maximum pyrolysis S_1 value is 4.89 mg HC/g rock, with an average of 0.91 mg HC/g rock. The S_1 loss value of the sample was 0.04 mg HC/g rock, the maximum was 2.14 mg HC/g rock, and the average was 0.39 mg HC/g rock. The minimum loss rate was 11.3%, the maximum was 52.7%, and the average loss rate was 29.4% (Table 2).

4.2. TOC and S_1^* Features. In general, the TOC value was positively correlated with S_1. When TOC is between 1 and 3 wt %,

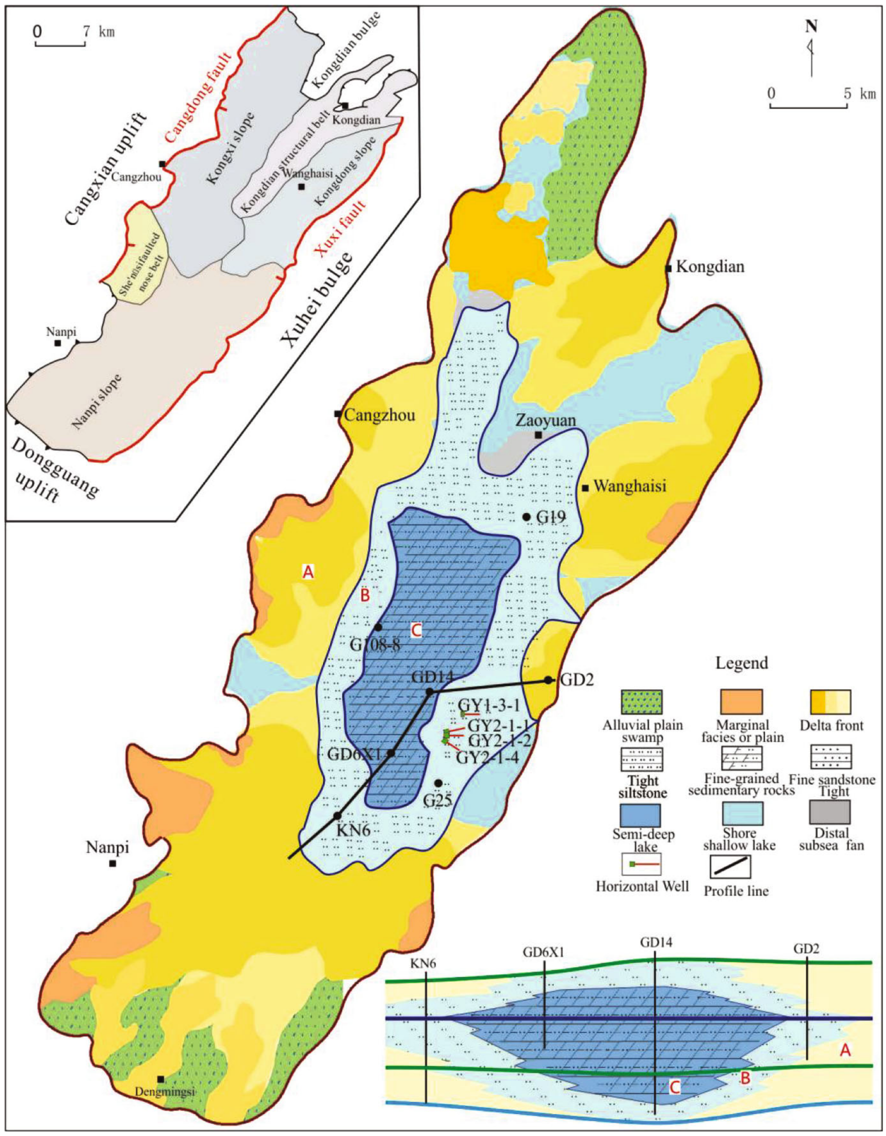

Figure 1: Structural subdivision and sedimentary facies map of the Kong 2 member in Cangdong Sag, Bohai Bay Basin.

Table 1: Experimental sample details.

Well	Sample		Experiment	
	Type	Number	Name	Result
G19	Core	12	Swelling adsorption	Adsorbed hydrocarbon
G25	Core	23	TOC, thermal extraction and pyrolysis	TOC, S_1
G25	Cuttings	23	TOC, thermal extraction and pyrolysis	TOC, S_1
GY1-3-1	Cuttings	192	TOC, thermal extraction and pyrolysis	TOC, S_1
GY2-1-1	Cuttings	274	TOC, thermal extraction and pyrolysis	TOC, S_1
GY2-1-2	Cuttings	207	TOC, thermal extraction and pyrolysis	TOC, S_1
GY2-1-4	Cuttings	135	TOC, thermal extraction and pyrolysis	TOC, S_1

the high-value part of S_1 increases rapidly with the increase in TOC. When TOC is greater than 3 wt %, the high-value part of S_1 changes or disappears in a horizontal shape, and there is an obvious upper envelope. As TOC increases, S_1 increases. When TOC is less than 1% wt, S_1 does not exceed 4 mg/g. When TOC is greater than 3% wt, S_1 approaches the maximum value, generally 12–14 mg/g, and GY2-1-2 reaches 20 mg/g (Figure 2).

Table 2: Original hydrocarbon content, loss amount, and loss rate with conventional pyrolysis of closed core and wellhead cuttings.

No.	1	2	3	4	5	6	7	8	9	10	11	12
Well	G25	G25	G25	G25	G25	G25	G25	G25	G25	G25	G25	G25
Depth (m)	3365.9	3353.7	3359.3	3355.7	3351.0	3342.5	3376.4	3339.5	3347.0	3346.0	3344.5	3352.7
TOC (%)	1.02	0.67	1.71	2.28	1.84	2.98	2.40	5.66	3.25	2.06	2.46	3.84
Core (mg/g)	0.34	0.60	0.84	0.91	1.14	1.21	1.22	1.24	1.24	1.63	1.67	1.79
Cutting (mg/g)	0.30	0.38	0.57	0.68	0.95	0.68	1.08	0.92	0.87	0.92	1.27	1.03
Loss (mg/g)	0.04	0.22	0.27	0.23	0.19	0.53	0.14	0.32	0.37	0.71	0.40	0.76
Loss ratio (%)	11.8	35.9	32.6	25.2	16.0	44.1	11.3	26.0	29.8	43.6	24.1	42.3
N.O	13	14	15	16	17	18	19	20	21	22	23	Average
Well	G25	G25	G25	G25	G25	G25	G25	G25	G25	G25	G25	
Depth (m)	3374.4	3367.9	3369.9	3351.6	3377.4	3375.4	3363.3	3366.9	3361.3	3356.7	3345.0	
TOC (%)	4.69	1.85	3.29	7.29	2.96	3.46	3.53	4.66	6.56	5.97	5.72	
Core (mg/g)	1.87	1.92	2.73	2.98	3.12	3.38	3.56	3.75	4.48	4.52	6.84	2.30
Cutting (mg/g)	1.02	0.91	2.33	2.43	1.77	2.31	2.67	3.16	3.97	2.38	4.89	1.63
Loss (mg/g)	0.85	1.01	1.40	1.55	1.35	1.07	1.89	0.59	0.51	2.14	1.95	0.67
Loss ratio (%)	45.4	52.7	14.6	18.4	43.3	31.7	24.9	15.9	11.3	47.3	28.5	29.4

4.3. Adsorbed Hydrocarbon Content. The content of light hydrocarbons adsorbed by low-maturity shale was high. With an increase in maturity, the amount of hydrocarbon adsorbed gradually decreases. At the low maturity stage (Ro less than 0.6%), the adsorption capacity is more than 150 mg HC/g TOC; at the mature stage (Ro > 0.6%), the amount of adsorbed hydrocarbon is 82–155/109 mg HC/g TOC. When Ro is greater than 0.8% (corresponding to a depth of 3300 m), the average amount of adsorbed hydrocarbon was 95 mg HC/g TOC (Figure 3).

5. Discussion

5.1. Correction Coefficient of Light Hydrocarbon Content. The pyrolysis parameter S_1 is the key parameter for characterizing the oil content of shale oil. Shale oil resources may change by 2–3 times before and after the correction of light hydrocarbon loss [18, 19]. Currently, most correction coefficients are obtained by indoor pyrolysis and gold tube experiments (Li et al. 2012). However, owing to the influence of core storage conditions, experimental test and analysis technology, and swelling effects, there is a large error between the number of S_1 recovery and the actual underground value [12]. According to the analysis of chromatographic results of crude oils with different properties under different storage conditions, Cooler et al. [20] and Hunt et al. [21] suggested that light hydrocarbons account for nearly 1/3 of the total oil, and almost all are lost. Zhang et al. [8] compared the results of direct pyrolysis with those of pyrolysis after different storage times; it was concluded that the longer the storage time, the more light hydrocarbon was lost. The light hydrocarbon recovery was based on 50% loss. These experiments provide a lot of good references for us to understand light hydrocarbon recovery, but there are always differences between field application and experimental analysis to provide the applicability of the method. In this study, the recovery coefficient of light hydrocarbon content was determined by comparing the pyrolysis S_1 value of wellhead cuttings with that of closed coring at the same depth. The correction coefficient was 1.42, based on the loss of pyrolysis S_1 value of 29.4%. It is worth noting that the coefficient is closely related to the maturity of organic matter, which is suitable for this study area, but corresponding experiments should be conducted in other maturity sections or other study areas.

5.2. Calculation Method of Movable Hydrocarbon. At present, the commonly used characterization index of the movable hydrocarbon content in shale oil is the oil saturation index (S_1/TOC). When the saturation index is greater than 100 mg/g, the movable hydrocarbon content is greater, and the larger the value, the greater the movable hydrocarbon content [12, 22]. According to S_1/TOC, the index reflects the movable efficiency rather than the actual momentum. To quantitatively analyze the real amount of mobile hydrocarbons, a method for subtracting the adsorption amount from the total amount of light hydrocarbon is proposed. When the maturity is 0.5–1.0%, the amount of hydrocarbon adsorbed by shale in the study area is 95 mg/g (Figure 4(a)), the total hydrocarbon amount is the experimental data, and the absolute amount of real movable hydrocarbon is the distance between the total hydrocarbon amount and the adsorption baseline (Figure 4(a)).

The derivation process is as follows.

M is the value of TOC and S_1^* at any point, the straight line $S_1^* = a * \mathrm{TOC}$ is the lower limit of the movable oil, and a is the coefficient of organic matter adsorption movable oil content. The movable oil content can be expressed as the distance from any point m to the straight line $S_1^* = a * \mathrm{TOC}$ (Figure 4(b)).

$$D = \frac{S_1{}^* - A * \mathrm{TOC}}{\sqrt{1^2 + A^2}} = \frac{S_{\mathrm{movable}}}{\sqrt{1^2 + A^2}}, \tag{2}$$

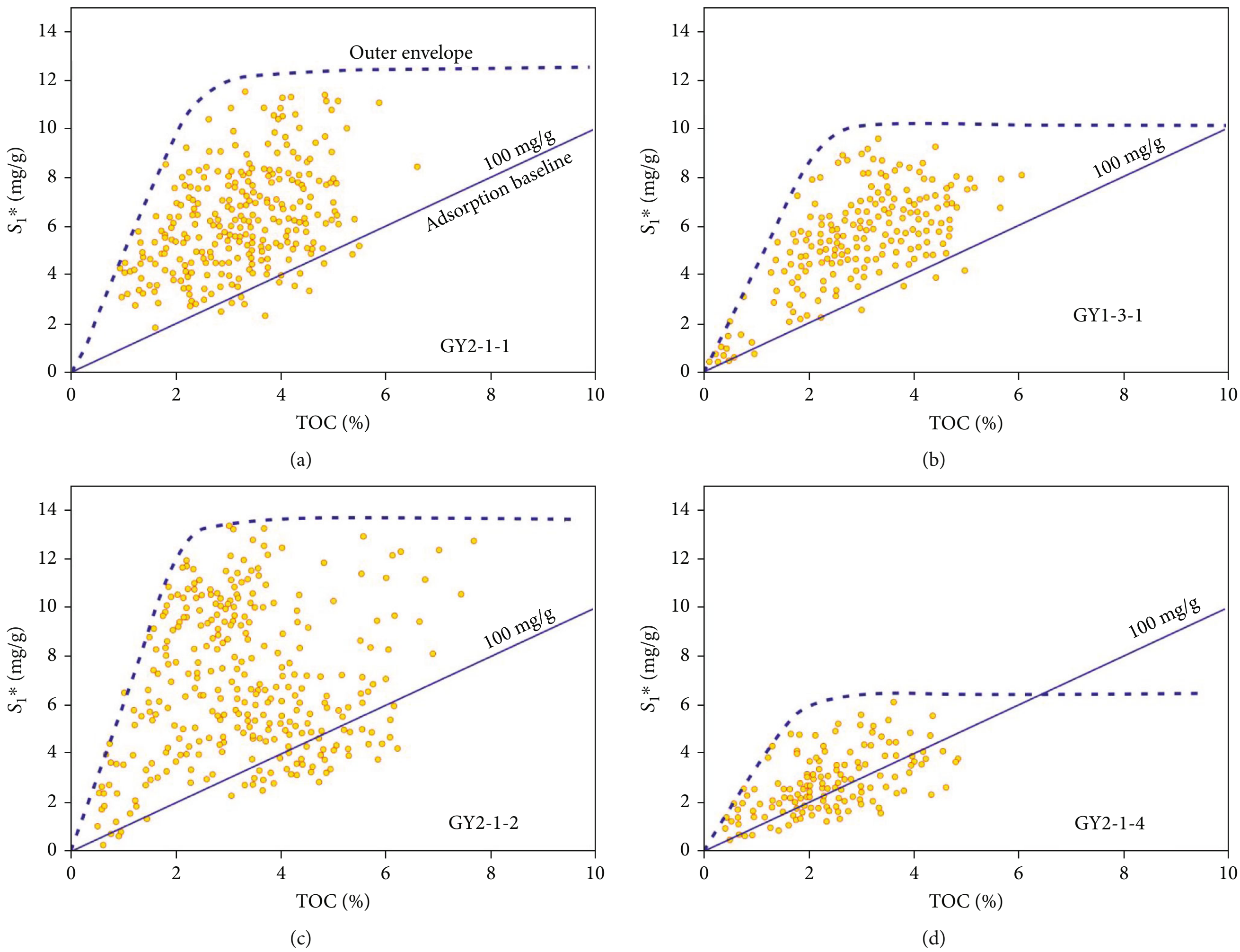

Figure 2: Distribution chart of TOC and S_1^* characteristics in the horizontal section of key wells: (a) well GY2-1-1, equivalent to a vertical depth of 3714 m; (b) well GY1-3-1, equivalent to a vertical depth of 3939 m; (c) well GY2-1-2, equivalent to a vertical depth of 3702 m; (d) well GY2-1-4, equivalent to a vertical depth of 3777 m. Samples were obtained from horizontal well cuttings of the second member, Kongdian Formation.

$$\cos\theta = \frac{1}{\sqrt{1^2 + A^2}} = \frac{D}{L}. \tag{3}$$

Substitute Equation (3) into (2):

$$S_{\mathrm{moveable}} = L. \tag{4}$$

5.3. Shale Oil Resource Evaluation

5.3.1. Resource Classification. The normal development of shale oil resources requires fracturing in addition to good geological conditions [23]. High-quality geological conditions mainly include good physical properties and oil-bearing properties [24]. Currently, the average porosity of shale is 3–8%, and the permeability is lower than 0.1 mDa. The difference in oil-bearing properties is key to the success of shale oil wells. Zhao et al. (2020) considered that the probability of high yield is high when S_1 is greater than 3 mg/g, and S_1^* increases linearly with TOC when TOC is less than 3% wt. When the TOC is approximately 3% wt, the linear distance from the corner of the outer loop to the lower adsorption limit was the largest, indicating that the mobile hydrocarbon content was large. Brittle mineral content is an important reference index for fracturing. TOC is a plastic mineral, and its content has a significant influence on brittleness (Figure 5). When the TOC content was 4 wt %, the content of brittle minerals was only 70%. When the TOC value reached 6 wt %, the brittle mineral content was close to 60%. Furthermore, the brittleness of shale with low organic matter (TOC < 4 wt %) is the best for S_1^*, followed by that of shale with high organic matter (TOC > 4 wt %). Concurrently, when TOC is greater than 4 wt %, S_1^* decreases with an increase in TOC.

To summarize, the geological and engineering evaluation template of shale oil was established based on these findings. The lower limit of movable oil is 100 mg HC/g rock; TOC content of 4% wt and 6% wt and corrected movable oil content of 3 mg/g and 6 mg/g divide the optimal geological and engineering settings of shale oil into three categories and eight subcategories (Figure 6).

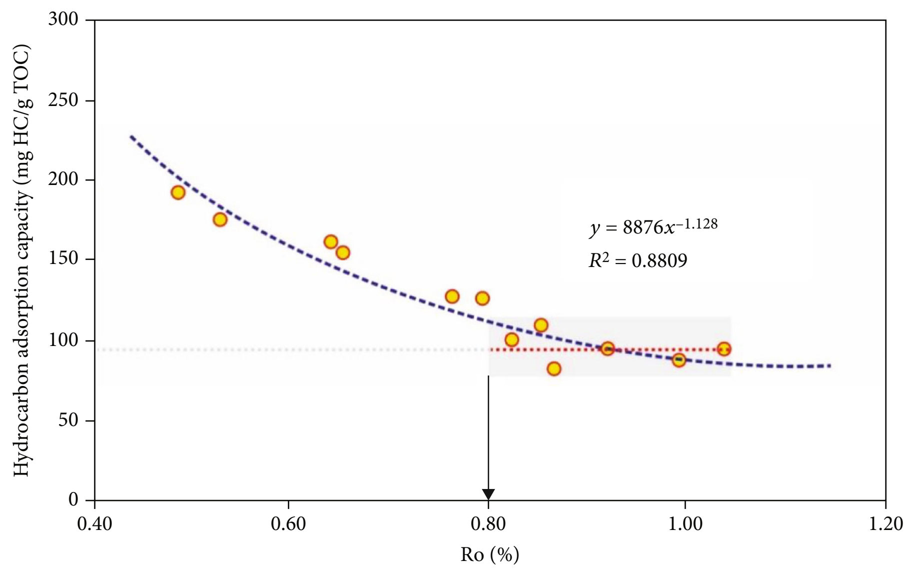

Figure 3: Quantitative relationship between shale maturity and hydrocarbon adsorption determined by swelling test.

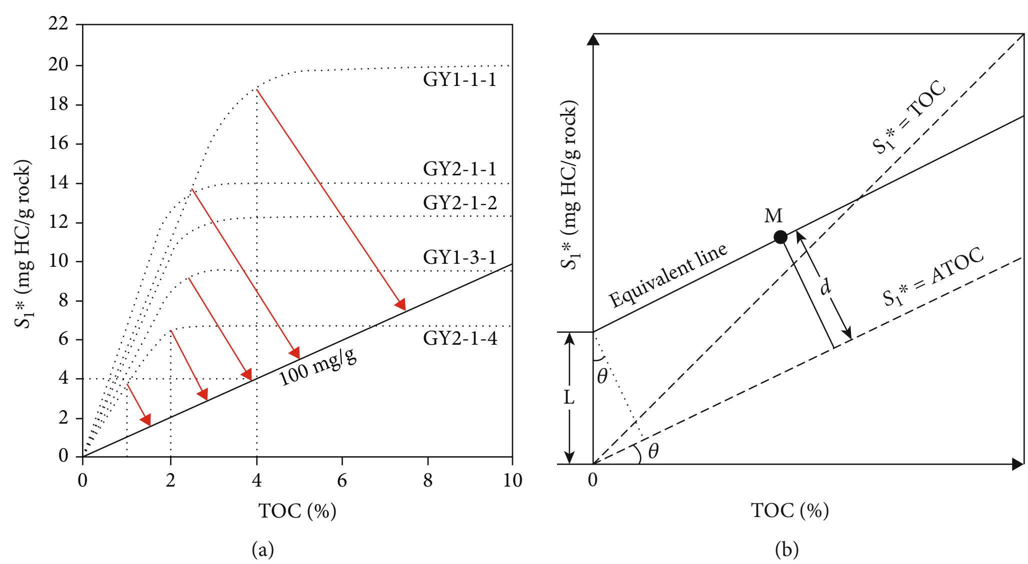

Figure 4: Characterization and formula derivation of movable hydrocarbon in shale oil: (a) schematic diagram of TOC and S_1 outer envelope in horizontal section of key well; (b) parameter diagram of shale movable hydrocarbon calculation formula.

5.3.2. Calculation Method. The specific steps of quantitative calculation of shale oil resources are as follows: (1) obtain the correction coefficient of movable oil content (S_1) using an experimental method, obtain the movable oil content (S_1^*) close to the formation state, calculate the total amount of retained oil in shale, and evaluate the total shale oil resources; (2) determine the recoverable amount of shale oil by combining the correction of the pyrolysis movable oil content (S_1^*) and residual TOC content. The key parameter can be used to calculate the geological movable resources of shale oil; and (3) establish an evaluation template for shale oil geology and engineering to calculate the recoverable resources of shale oil.

(1) Total Light Hydrocarbon Resources of Shale Oil. The correction coefficient of S_1 was obtained by physical experimentation.

The free oil loss rate was determined using the free oil content of fresh samples from sealed coring and cutting samples from the corresponding depth of the wellhead, and the free oil correction coefficient (K_c) was determined.

$$Q_{total} = 0.1 \times S_{shale} \times h_{shale} \times \rho \times S_1 \times K_c. \quad (5)$$

Q_{total} is shale-free oil resource (10^4 t); S_{shale} is total shale area (km^2); h_{shale} is total shale effective thickness (m); ρ is

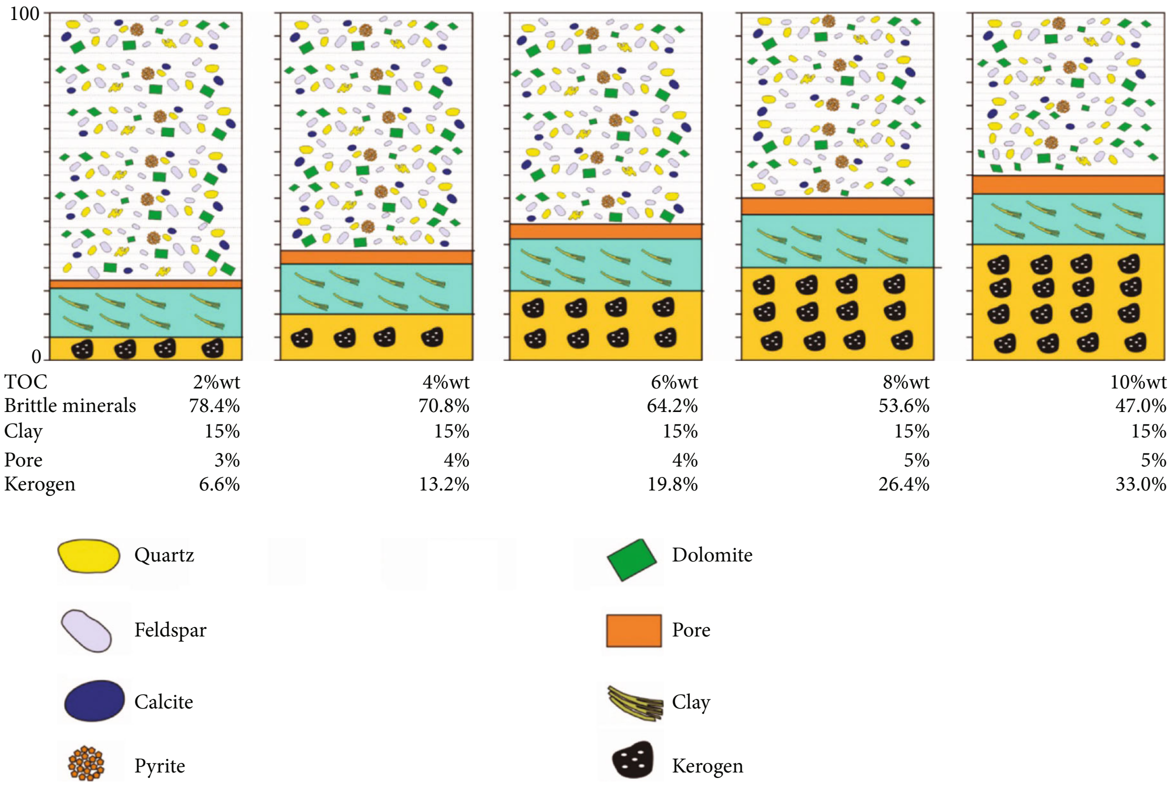

FIGURE 5: Effect of TOC on the content of brittle minerals.

shale density (2.5 g/cm^3); S_1 is light hydrocarbon content of residual oil (mg HC/g rock); and K_c is the dimensionless correction coefficient of shale free oil loss.

(2) Movable Shale Oil Resources.

(1) The movable oil content (S_1^*) can be obtained by using the correction coefficient to correct the residual oil

(2) Based on the experimental data of kerogen swelling adsorption, the content of residual TOC adsorbed oil and amount of adsorbed oil per unit organic matter (K_{abs}) were determined

(3) Calculate the movable oil content ($S_{movable}$) with Equation (5)

$$S_{movable} = S_1{}^* - (K_{abs} \times \mathrm{TOC}),$$
$$S_1{}^* = S_1 \times K_c, \quad (6)$$

where $S_{movable}$ is the amount of residual oil (mg HC/g rock); S_1^* is the moveable oil content after correction, unit: mg HC/g rock; S_1 is the content of conventional thermally extractable oil, unit: 1 mg HC/g rock; and K_{abs} is the adsorption capacity of unit organic matter, mg HC/g TOC.

$$Q_{movable} = 0.1 \times S_{shale} \times h_{shale} \times \rho \times S_1{}^*, \quad (7)$$

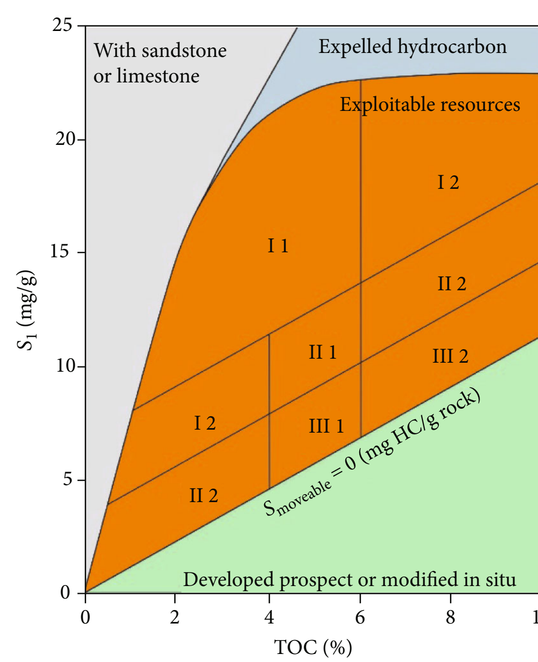

FIGURE 6: Shale oil resource evaluation template.

where $Q_{moveable}$ is the movable resources of shale-free oil (10^4 t); S_{shale} is total shale area (km^2); h_{shale} is total shale effective thickness (m); ρ is shale density (2.5 g/cm^3); and S_1^* is the content of conventional pyrolysis movable oil (mg HC/g rock).

Table 3: Shale oil movable resources of XX layer in XX block.

Type		Thickness (m)	Favorable area (km^2)	$S_{movable}$ (kg oil/t rock)	Resource (10^4 t)
I	I_1	50	23.6	10.2	3009.0
	I_2	46	24.2	8.4	2337.7
II	II_1	35	34.5	4.8	1449.0
	II_2	25	15.6	4.2	409.5
III	III_1	45	52.4	2.5	1473.8
	III_2	50	39.8	2.5	1243.8
Total					9922.7

Average thickness, favorable area, and measurable parameters are calculated by the weighted average method.

(3) Recoverable Resources.

(1) The TOC content and S_1^* chart of corrected movable oil content were established to quickly evaluate optimal geological settings
(2) Combined with the brittleness index template, the optimal engineering template settings can be evaluated
(3) Determine the lower limit of recoverable resources and determine the range of recoverable resources

The template was used to classify the study area horizontally and vertically to calculate the recoverable shale oil resources. The formula for calculating the shale-oil movable resources is as follows:

$$Q_{recoverable} = 0.1 \times S_{sweet} \times h_{sweet} \times \rho \times S_{movable}, \quad (8)$$

where $Q_{recoverable}$ is shale-free oil resource (10^4 t); S_{sweet} is shale sweet spot area (km^2); h_{sweet} is shale sweet spot thickness (m); ρ is shale density (2.5 g/cm^3); and $S_{movable}$ is the amount of residual movable oil (mg HC/g rock).

Vertically, single wells are classified by TOC content, residual moveable oil, and the brittleness index. The distribution characteristics of the plane TOC content, residual oil moveable, and brittleness index in the study area were determined by seismic inversion. Superposition classification was conducted, and shale oil moveable resources were calculated using Equation (8) (Table 3).

6. Conclusion

The light hydrocarbon recovery coefficient of shale obtained by the underground core and wellhead cutting method is more practical than other conventional methods. Affected by the TOC content and thermal maturity of organic matter, the recovery coefficient of light hydrocarbons varies in different areas and depths.

The shale oil moveable index is the index of shale oil moveable efficiency, and the moveable oil content index can be used to quantitatively characterize shale oil movability.

The corrected light hydrocarbon content was used to calculate total shale oil resources, while the movable oil content index was used to estimate movable shale oil resources. There are abundant shale oil resources in the continental basins of China. The Cangdong Sag, Bohai Bay Basin, has been distributed in 100 million tons of shale oil reserves, which requires further research and methodological development in the field of geology and engineering.

Data Availability

All data are in the form of figures and tables in the manuscript, and there is no Table 3; Table 4 should be Table 3.

Disclosure

The funders had no role in the study design; collection, analyses, or interpretation of data; writing of the manuscript; or decision to publish the results.

Conflicts of Interest

The authors declare no conflict of interest.

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