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Study on Pyrolysis Hydrocarbon Generation Kinetics and Shale Gas Formation Process of Marine Continental Transitional Rocks in Shanxi Formation, Ordos Basin

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Abstract

The marine continental transitional facies shale of $Shan_2{}^3$ sub member of Shanxi Formation in Ordos Basin was selected, and the hydrocarbon generation kinetics of Shan 23 sub member marine continental transitional facies shale was studied through open system thermal simulation experiment. The experimental results show that the pre exponential factor of Shan 23 shale is $12.92*10^{15}$/s, and the activation energy is mainly concentrated in 52~82kcal/mole. On this basis, combined with petromod basin simulation software, the burial history, thermal evolution history and hydrocarbon generation history of Shanxi Formation shale are established, and the formation process of shale gas is clarified. Finally, using the genetic method, combined with the original TOC restored by Petromod basin simulation software and the hydrocarbon generation obtained from the open system thermal simulation experiment, the resources of the study area are calculated when the hydrocarbon expulsion efficiency is 50%. The calculation results show that the shale gas resource in the study area is $12.41*10^{12}m^3$, which shows a good exploration potential of sea land transitional facies shale in the study area.

Keywords

Shan 23 Sub Segment; Sea Land Transitional Facies; Shale Gas; Hydrocarbon Generation Kinetics; Petromod Basin Simulation Software; Resources.

1. Introduction

In recent years, the exploration and development results of sea land transitional facies shale gas show its good gas bearing property and development prospect, especially in Ordos Basin, which is one of the main bodies of sea land transitional facies shale gas resources. In the coal measure strata developed in the early Permian sea land transition environment in the North China block, the dark shale has large thickness, high organic matter abundance and high degree of thermal evolution. It has always been considered as an important gas source rock for natural gas, and its shale gas exploration potential has attracted people's attention (Zhen et al.,2020). However, there is little work on its shale gas resource evaluation at present.

With the increasing attention of scholars at home and abroad to the hydrocarbon generation dynamics of source rocks, there is a deep understanding of the hydrocarbon generation mechanism and process of organic matter in source rocks, which makes it occupy an extremely important position in the evaluation of oil and gas resources in source rocks (Braun et al.,2002;

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Boreham et al.,1999; Xiong et al.,2002). The current research shows that the factors affecting hydrocarbon generation dynamics to evaluate the oil and gas resource potential of source rocks are mainly divided into three aspects: thermal simulation experiment, dynamic model and heterogeneity of source rocks (Peng et al.,2018; Cui et al.,2019; Rao et al.,2010; Wang et al.,2011). The experimental results of different systems of thermal simulation experiment, such as closed system and open system, are different. Even in the same experiment, different experimental conditions in the experiment will have a great impact on the experimental results (Huang et al.,2017). As more and more scholars study the kinetics of hydrocarbon generation (Lu et al.,1996; Behar et al.,1992; Burnham et al.,1995; Burnham,2017), At present, there are six kinds of kinetic models, among which the parallel first-order reaction model is the most widely used. At the same time, with the deepening of research, many scholars have found that there are differences in the kinetic parameters of organic matter of different parent material types. Even for the organic matter of the same parent material type in the same sedimentary basin, there are significant differences in the kinetic parameters of hydrocarbon generation (Behar et al., 2008; Peters et al.,2006). Therefore, in order to accurately obtain the hydrocarbon generation kinetic parameters of specific source rocks and simulate their hydrocarbon generation process, the corresponding source rocks must be selected for experiments.

In this study, the immature shale of Shanxi Formation in Palougou area of Ordos Basin is selected. Through the open system thermal simulation experiment, the dynamic characteristics of hydrocarbon generation (activation energy and pre exponential factor) of marine continental transitional facies shale are studied, and the experimental results are applied to Petromod basin simulation software to establish the burial history, hydrocarbon generation history and thermal history of $Shan_2^3$ sub member, so as to obtain the geological process of shale gas generation. Finally, TOC is restored by software, combined with the amount of hydrocarbon generation obtained from the open system thermal simulation experiment, and the amount of resources in the study area is calculated, which provides ideas for the evaluation of sea land transitional facies shale gas resources.

2. Geological Background

The eastern structural area of Ordos Basin is located in the east of Northern Shaanxi Slope. The overall height shows a decreasing trend from east to west. The slope is less than 10m and the dip angle is less than 1°. It is a multicycle inland craton petroliferous basin with obvious stable subsidence, depression migration and torsion. The study area of this paper is Daning-Jixian area.The area of mechanism rich shale in the study area is about 5050km^2. Structurally, it is close to the Western Shanxi fold belt, showing a trend of high in the East and low in the West. According to the logging data, the study area mainly develops five *sets* of strata: Carboniferous Benxi Formation, lower Permian Taiyuan Formation and Shanxi formation, Middle Permian Shihezi Formation and Upper Permian Shiqianfeng Formation. Among them, Benxi Formation, Taiyuan Formation and Shanxi formation have deposited many sets of mechanism rich shales of sea land transition facies The most typical stratum is $Shan_2$ shale of Shanxi formation, with large thickness and high organic carbon content (Kuang et al.,2020). As a high-quality shale section in $Shan_2$, the TOC of $Shan_2^3$ sub section is generally 0.75%~3%, up to more than 10%. The organic matter type is mainly type III, containing a small part of type II_2, and the shale maturity is high, which has entered the stage of generating dry gas.

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3. Samples and Experimental Methods

3.1. Experimental Method

In this experiment, the open system Rock-Eval Ⅵ rock pyrolysis instrument of Wuxi Institute of petroleum geology, Sinopec petroleum exploration and Development Research Institute is selected to analyze the pyrolysis of $Shan_2^3$ shale sample in Ordos sea land transitional facies belt, and the kinetic simulation calculation of the pyrolysis data is carried out by using Kinetics software.

Firstly, the shale samples are divided into three groups. The three groups of samples are heated at three different heating rates (5℃/min, 15℃/min, 25℃/min). The initial temperature of the experiment is 300℃ and heated at a constant temperature for 5min to remove the adsorbed hydrocarbons. The termination temperature is set to 700℃. After the experiment, the cracked hydrocarbon S2 at different temperatures can be measured; The free hydrocarbon S1 is measured by putting each sample into a pyrolysis reaction furnace with a furnace temperature of 30℃ for constant temperature for 3min to desorb the free hydrocarbon adsorbed in the sample; S1 and S2 are detected by hydrogen flame ionization detector (FID). S3 is the content of CO2 in the pyrolysis process of organic matter, which reflects the content of organic matter and is detected by infrared detector.

The essence of oil and gas generation is the transformation of organic matter in source rocks into hydrocarbons under the action of thermal degradation. In order to obtain the chemical kinetic parameters (activation energy E and frequency factor A) of organic matter in the reaction process, we simulate the evolution process under actual conditions under high temperature and rapid conditions in the laboratory through the principle of time temperature compensation. At present, the chemical kinetic models of hydrocarbon generation of organic matter mainly include parallel reaction model (Tissot and Welte,1984; Ungerer.,1990), series reaction model (Dieckmann,2005) and total package reaction model (Allred,1966; Delvaux and Martin,1990). The first-order reaction model can be expressed as:

$$X(t) = \sum Xi(t)$$

$$Xi(t) = Xi0 * (1 - e^{-Ki(t)})$$

$$\text{By Arrhenius formula: } \mathrm{Ki} = \mathrm{Ai} * e^{-\frac{Ei}{RT}}$$

It can be seen that Xi is only a function of temperature and time.

Where: X is the total oil and gas generation at time t (dimensionless); Xi is the cumulative hydrocarbon generation amount of the i^{th} hydrocarbon generation parent at time t (dimensionless); Xi0 is the maximum hydrocarbon generation amount of Xi that can be generated by the i^{th} hydrocarbon generation parent (dimensionless); Ki is the reaction rate constant (s^{-1}); T is the time (s); Ei is the activation energy (J/mol); Ai is the frequency factor (s^{-1}); R is the gas constant, and its value is 8.314 J/(mol*k); T is the thermodynamic temperature (k).

3.2. Samples

The organic matter types of shales in the sea land transitional facies belt of Shanxi Formation in Ordos Basin are mainly type Ⅲ and $Ⅱ_2$. Due to the relatively deep burial depth, the vitrinite reflectance (Ro) of shale samples is generally high. In order to objectively reflect the evolution characteristics of organic matter in source rocks, immature or low maturity samples should be selected in the experiment. Therefore, the samples of Shanxi Formation outcrop located in Palougou Section (Fig.1) in Baode County, northeast edge of Ordos Basin are selected in this experiment. Shales in Shan 23 member are mainly black in color, with well-developed foliation. Table name of geochemical analysis (table 1). The average content of total organic carbon (TOC) in outcrop samples is 2.41%, and the average hydrogen index (HI) is 137.

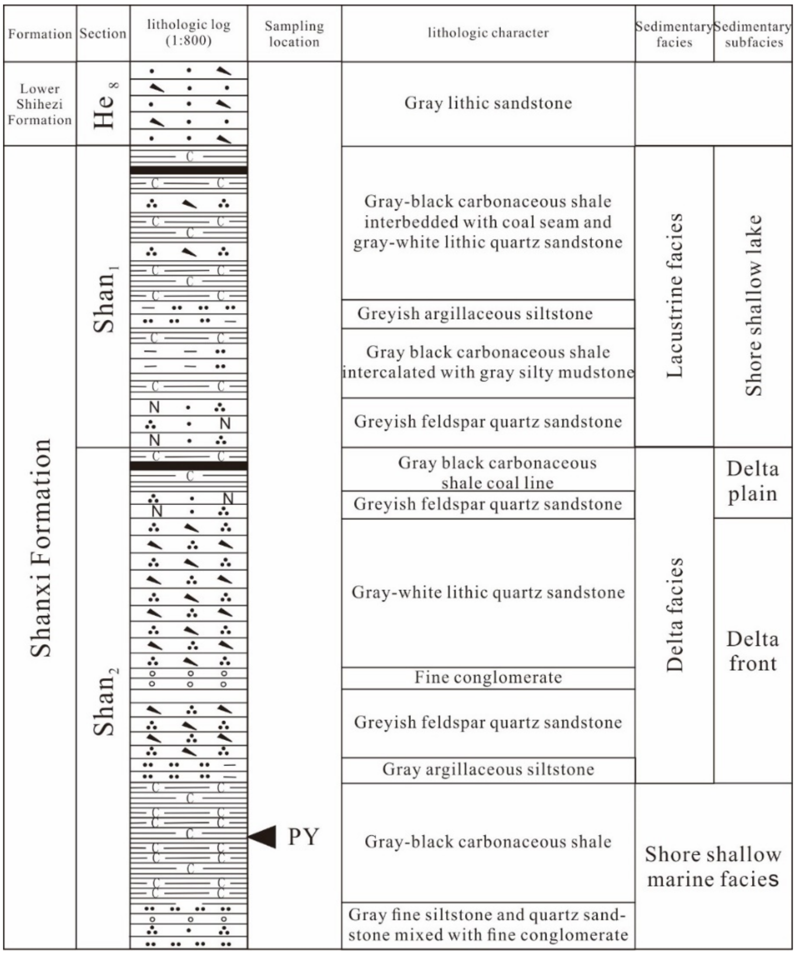

Figure 1. Comprehensive histogram of Shanxi formation of baluogou section

Table 1. Geochemical characteristics of shale samples from open system thermal simulation experiment

Sample	Horizon	Lithology	Kerogen type	S1(mg/g)	S2(mg/g)	Tmax(℃)	TOC(%)	HI
PY	Shanxi Formation	Black shale	Type III	0.12	3.26	438	2.50	130
PY	Shanxi Formation	Black shale	Type III	0.11	3.27	432	2.36	139
PY	Shanxi Formation	Black shale	Type III	0.09	3.38	435	2.38	142

4. Result

4.1. Shale Activation Energy and Pre Exponential Factor

Kinetic 2015 software was used to process and calculate the experimental data to obtain the kinetic parameters of the sample (Fig.2 and table 2). The experimental results show that the pre exponential factor of the sample is $12.92*10^{15}$/s, and the activation energy of the sample is mainly concentrated in the range of 52~82kcal / mole, which fully reflects a relatively slow process of hydrocarbon generation and explains the heterogeneity of kerogen parent material. Taking the kerogen hydrocarbon generation conversion rate of 10%~90% as the effective hydrocarbon generation interval of kerogen, the hydrocarbon generation energy span of the sample is also large, which reflects that the hydrocarbon generation process of the sample is accompanied by the whole thermal evolution process of Shanxi formation.

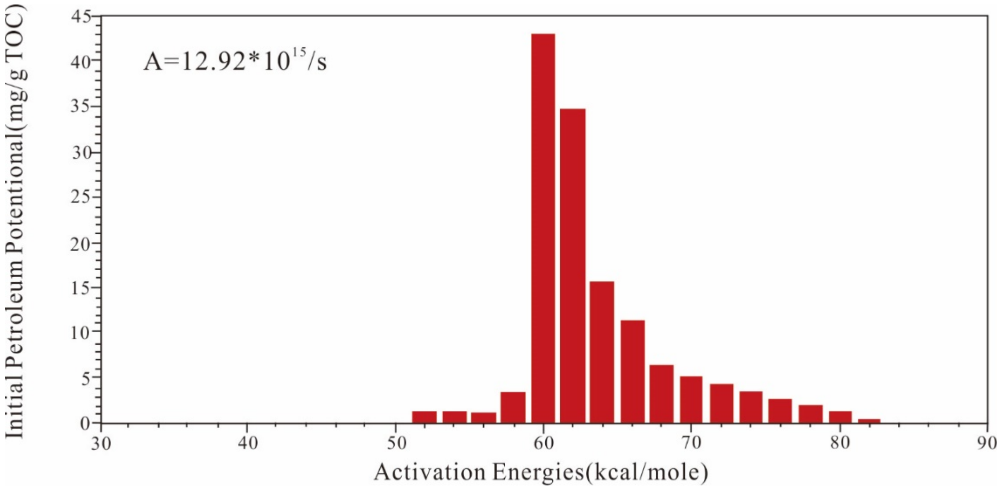

Figure 2. Distribution characteristics of shale activation energy and frequency factor

Table 2. Shale activation energy and pre exponential factor

Activation Energy (kcal/mole)	Initial Petrolrum Potentional (mg/g TOC)	(%)	Arrhenius Constant (s^{-1})
50	0.10	0.05	$12.920*10^{15}$
52	1.30	0.90	$12.920*10^{15}$
54	1.30	1.00	$12.920*10^{15}$
56	1.10	0.80	$12.920*10^{15}$
58	3.10	2.30	$12.920*10^{15}$
60	43.20	31.50	$12.920*10^{15}$
62	34.70	25.30	$12.920*10^{15}$
64	15.40	11.30	$12.920*10^{15}$
66	11.30	8.30	$12.920*10^{15}$
68	6.30	4.60	$12.920*10^{15}$
70	5.10	3.70	$12.920*10^{15}$
72	4.40	3.20	$12.920*10^{15}$
74	3.40	2.50	$12.920*10^{15}$
76	2.60	1.90	$12.920*10^{15}$
78	1.90	1.40	$12.920*10^{15}$
80	1.30	0.90	$12.920*10^{15}$
82	0.40	0.30	$12.920*10^{15}$
84	0.10	0.05	$12.920*10^{15}$

4.2. Product Characteristics

It can be seen from Fig. 3 that the yield of gaseous hydrocarbon in the sample (Fig.3a) starts at 340 ℃ and stops at 700 ℃ under different heating rates (5℃/min, 15℃/min, 25℃/min). Different heating rates (5℃/min, 15℃/min, 25℃/min) will not affect the final gaseous hydrocarbon yield of the sample, but when the temperature range is between 380℃~600℃, the gaseous hydrocarbon yield of the sample heated at the heating rate of 5 ℃ / min at the same temperature is significantly higher than that of the sample heated at 15 ℃ / min and 25 ℃ / min, and the higher the heating rate, the lower the gaseous hydrocarbon yield at the same temperature (Ma et al.,2017). The cumulative conversion rate of pyrolysis hydrocarbon of the sample (Fig. 3b) is also related to the thermal simulation temperature and heating rate. With the increase of thermal simulation temperature (the increase of thermal evolution degree), the cumulative conversion rate of pyrolysis hydrocarbon of kerogen increases. However, under three different heating rates, the conversion of samples with lower heating rate at the same temperature point is higher than that with higher heating rate. At the same time, this feature can also be reflected by the hydrocarbon generation rate curve (Fig.3C). When the heating rate is 5℃/min, the temperature reaches 444℃, and the hydrocarbon generation rate reaches the peak of 1.45mg/g TOC/deg.C; When the heating rate is 15℃/min, the temperature reaches 462℃, and the hydrocarbon generation rate reaches the peak of 1.52mg/g TOC/deg.C; When the heating rate is 25℃/min, the temperature reaches 474℃, and the hydrocarbon generation rate reaches the peak of 1.49 mg/g TOC/deg.C. It can be found that the faster the heating rate, the greater the temperature when reaching the peak of hydrocarbon generation rate. Therefore, the slower heating rate is more conducive to the cracking of organic matter. The fast heating curve of the same sample lags behind the slow heating curve, which reflects the complementary relationship between temperature and time in the process of chemical reaction.

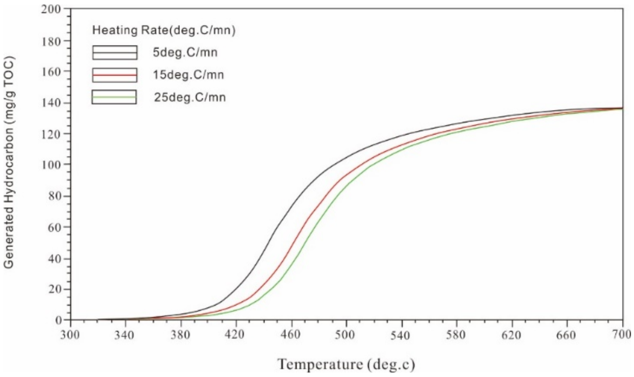

Figure 3a. Hydrocarbon generation of shale under different heating rates

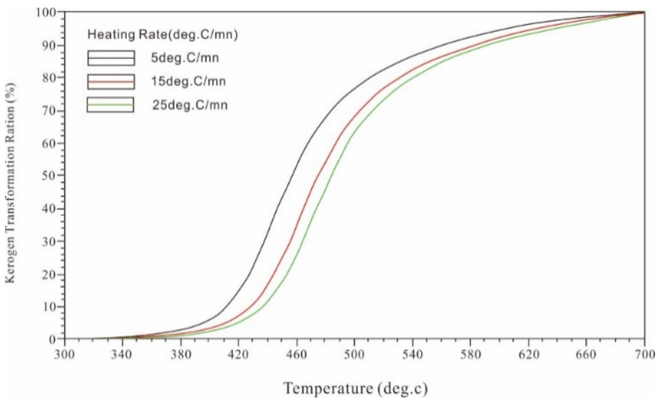

Figure 3b. Kerogen conversion at different heating rates

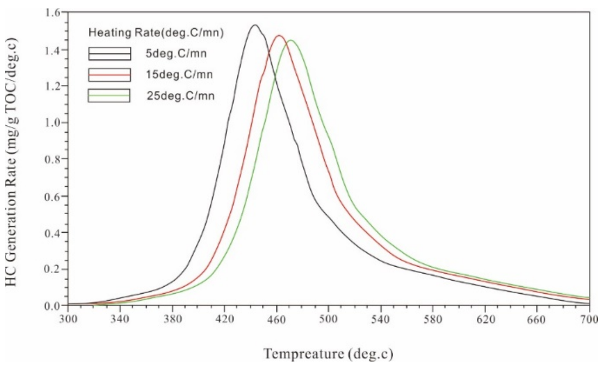

Figure 3c. Hydrocarbon generation rate of shale under different heating rates

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5. Discuss

5.1. Study on the Formation Process of Shale Gas in the Study Area

The kinetic parameters of hydrocarbon generation obtained from the open system experiment are input into Petromod software to establish the shale dynamic model of Shanxi Formation in the sea land transitional facies belt of Ordos Basin. Well MK5, a typical well in the southeast edge of Ordos Basin, is selected. According to the stratigraphic stratification data, paleoheat flow, paleowater depth and surface temperature of ponding, the buried thermal evolution history of shale in Shan 23 sub member of Shanxi Formation in this area is established by using Petromod basin simulation software (Fig.4a). It can be seen that there are four different periods of uplift and denudation in this area, which occurred in the Yanchang formation at the end of Triassic, the end of early and Middle Jurassic century, the Xianghe formation and Anding formation of Jurassic, and the end of Cretaceous century. Among the four uplift and denudation, the denudation amount at the end of the Cretaceous century is the largest. Considering the actual compaction degree of the underlying Triassic Jurassic system, the denudation amount of the stratum at the end of the Cretaceous in the eastern edge of the Ordos Basin is estimated by the acoustic time difference method (Weng et al., 2011). It is concluded that the denudation amount of well MK5 since the end of the Cretaceous is about 1570m. At the same time, it can also be found that the denudation at the end of the Cretaceous century has a particularly obvious impact on hydrocarbon generation.

According to the simulation results of typical well MK5 by petromod basin simulation software (Fig. 4b, c and d), the hydrocarbon generation of $Shan_2^3$ sub member of Shanxi Formation began in the Middle Permian (276Ma), at this time, RO reaches 0.3%. And a large amount of hydrocarbon generation began in the late Jurassic (180Ma), at this time, RO reaches 1.02%. Finally stopped at the end of the early Cretaceous (79Ma), at this time, RO reaches 2.81%. The shale of $Shan_2^3$ sub member of Shanxi formation mainly produces tertiary hydrocarbon generation, and a large amount of shale gas mainly occurs in the second and third hydrocarbon generation. The first hydrocarbon generation mainly occurs in 250Ma to 195Ma. At this time, the organic matter is cracked under the thermal action to generate oil and gas, which is mainly oil generation. Therefore, the gas production rate is low, reaching the peak value of gas generation rate of 0.0004 mgHC/g TOC/ma at 213Ma. The second hydrocarbon generation mainly occurs from 183Ma to 147Ma. At this time, the gas source is mainly generated in two ways: one is generated by organic matter, and the other is generated by oil cracking. At this time, the gas generation rate is fast, reaching the peak value of 0.0034 mgHC/g TOC/MA at 170Ma. The third hydrocarbon generation mainly occurs in 135Ma to 96Ma. The gas source is also composed of organic matter and oil cracking. The peak gas generation rate is 0.0038 mgHC/g TOC/MA at 121Ma.

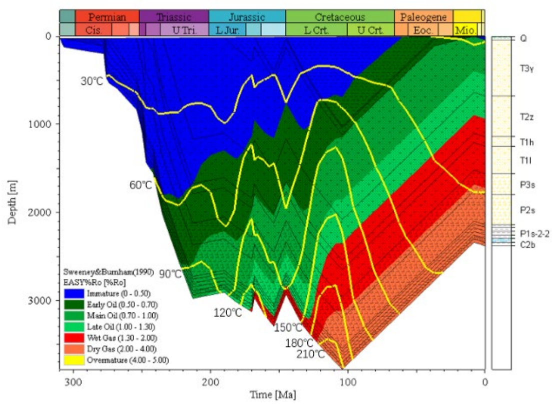

Figure 4a. Thermal evolution history of shale in Shanxi formation of Ordos Basin

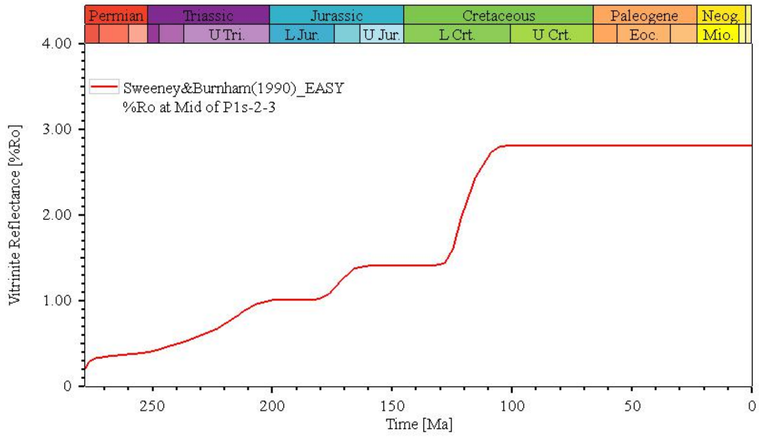

Figure 4b. Evolution history of shan$_2$^{3} vitrinite reflectance

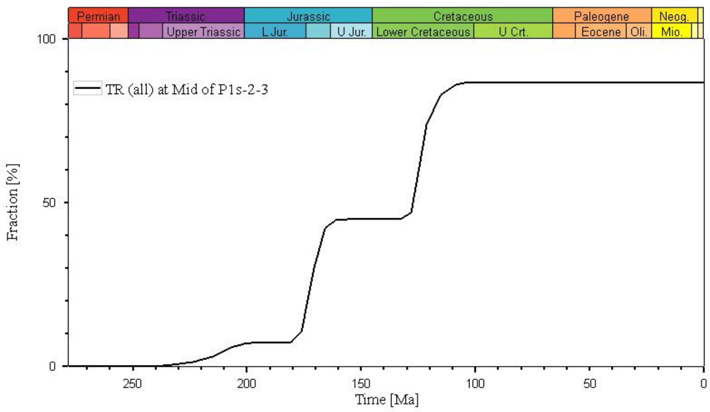

Figure 4c. Shan$_2$^{3} source rock conversion

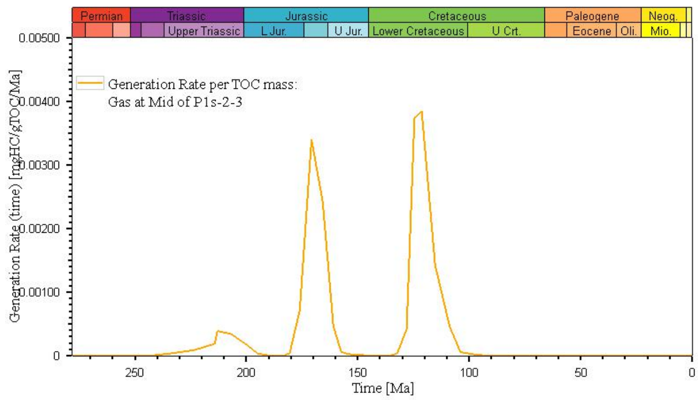

Figure 4d. Gas generation rate of shan$_2$^{3} source rock

5.2. Shale Gas Resource Evaluation

In recent years, more and more attention has been paid to the sea land transitional facies shale gas in Ordos Basin, but its resource evaluation is less. In order to calculate the shale gas

ISSN: 2710-0588

resources of sea land transitional facies in the study area, the genetic method - organic carbon method is selected for calculation.

$$Q_c = S \times h \times \rho \times C \times K_c \times X \times (1-K) \quad (1)$$

Where: Q_C: natural gas resources; S: Area of mature source rock; h: Thickness of mature source rock; ρ: Density of source rock; C: Residual organic carbon; K_c: TOC recovery coefficient; X: Hydrocarbon yield; K: Hydrocarbon expulsion coefficient.

The results of open system thermal simulation experiment show that the gas production rate of the sample finally reaches 138 mg/g TOC. According to the established relationship chart between shale gas Ro and exhaust coefficient (Gan, 2015), the exhaust coefficient of shale gas in the study area is 56%. In the study area, the shale thickness of $Shan_2^3$ sub section is the largest among the three sub sections of $Shan_2$, which can reach 20~50m, with an average of 26m. The simulation results of petromod basin simulation software show that the TOC recovery coefficient of standard well MK5 in the study area is 1.06 (Fig.5). TOC takes the average value of 3.7% in the study area, the shale density is $2.65g/m^3$, and the area of mechanism rich shale in the study area is $5050km^2$. If the above parameters are brought into formula (1), the resource of the study area can be obtained as $1.155*10^{12}m^3$. Predecessors have evaluated the resources of sea land transitional facies shale gas in the eastern edge of Ordos Basin (Kuang et al., 2020). Among them, the favorable area of shiluobei Daning Jixian county is about 6000 km^2, and the amount of geological resources is (8700~13800) $*10^8m^3$. Through the previous research results and the resource calculation results, it can be found that the sea land transitional facies shale resources in Ordos Basin are large and have good exploration potential.

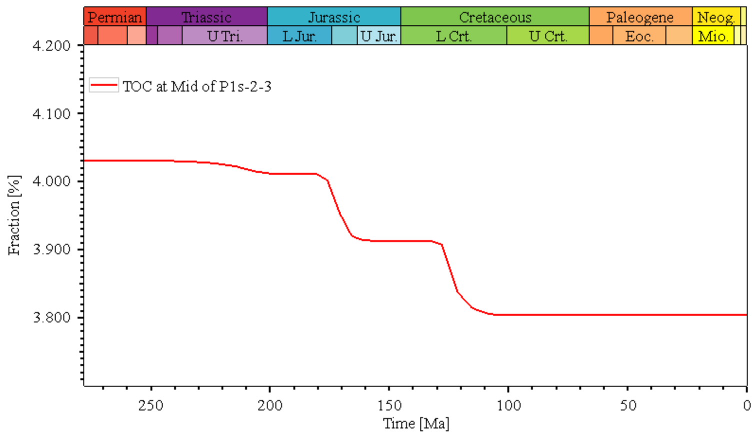

Figure 5. TOC variation curve of shale in Shanxi formation of well MK5

6. Conclusion

(1) The dynamic characteristics of shale hydrocarbon generation in Shan 23 sub member of Shanxi Formation in Ordos Basin were studied by using open system hydrocarbon generation thermal simulation experiment. The results show that the pre exponential factor is $12.92*10^{15}/s$, and the activation energy is mainly concentrated in 52~82kcal/mole.

(2) the hydrocarbon generation of $Shan_2^3$ sub member of Shanxi Formation began in the Middle Permian (276Ma), and a large amount of hydrocarbon generation began in the late Jurassic (180Ma), finally stopped at the end of the early Cretaceous (79Ma). During this period, when shale gas is generated in large quantities, it is mainly generated by organic gas and oil cracking gas at the same time.

ISSN: 2710-0588

(3) According to the genetic method - organic carbon method, the shale gas resources of $Shan_2^3$ sub member of Shanxi Formation in Ordos Basin are calculated. Considering that the hydrocarbon expulsion efficiency is 50%, the shale gas resources are $1.155*10^{12}m^3$. It can be seen that the sea land transitional facies shale gas of Shanxi formation has good exploration prospects.

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Conflict of Interest

The authors declare that they have no competing interests.

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