

Original Paper

Micro-migration and accumulation of shale oil within lacustrine organic matter-rich tuffaceous shale: Insights from high-precision thermal simulation experiment on various laminasets



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ARTICLE INFO

Article history:

Received 19 December 2025

Received in revised form

5 March 2026

Accepted 15 May 2026

Available online 20 May 2026

Edited by Xiu-Fang Hu

Keywords:

Ordos Basin

Chang 7₃ sub-member

Organic-rich tuffaceous shale laminaset

Shale oil

Micromigration-accumulation mechanism

ABSTRACT

The dynamic process of generation, expulsion, retention, and accumulation of shale oil within laminated shale has attracted considerable attention because it provides important geological insights into thermal evolution. Organic-rich tuffaceous laminated shale from the Chang 7₃ sub-member of the Ordos Basin provides an ideal case for investigating the micro-migration and accumulation of shale oil, which was investigated using high-temperature and high-pressure thermal simulation experiments and a series of organic geochemistry methods. The dynamic measurement results indicate that the hydrocarbon generation and expulsion processes of shale oil can be categorized into four stages: slow hydrocarbon generation stage (Stage A), slow hydrocarbon generation-expulsion stage (Stage B), rapid increase stage of massive hydrocarbon generation-expulsion (Stage C), and rapid decrease stage of massive hydrocarbon generation-expulsion (Stage D). A pronounced disparity between hydrocarbon generation and retained hydrocarbons is evident within the same laminaset during thermal stages, accompanied by dynamic functional transformations between the hydrocarbon-generating laminaset and hydrocarbon reservoir laminaset. This indicates that Rock-Eval pyrolysis parameters can effectively identify the micro-migration direction and quantity of shale oil based on the carbon balance principle. In early stages, hydrocarbons predominantly micro-migrate through connected pore throats within intra- or inter-laminasets, whereas in middle-late stages, fractures emerge as the primary micro-migration pathways. Light shale oil preferentially migrates and adsorbs onto mineral surfaces, whereas heavier components primarily migrate and accumulate along fractures. Free hydrocarbons are mainly hosted within intergranular and intragranular dissolution pores of tuffaceous laminaset, as well as in microfractures. The micro-migration and accumulation processes of shale oil in organic-rich tuffaceous shale are the result of the synergistic and dynamic evolution of the interactions between organic matter and inorganic minerals. These results provide enhanced insights into the differential micro-migration and accumulation mechanisms of shale oil within various lithological shales.

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1. Introduction

Diverse micro- to nanometer-scale hydrocarbon reservoir spaces are facilitated by the complex laminated sedimentary

structures within organic-rich shales. These reservoir spaces are influenced by the combined effects of organic matter and inorganic minerals, as well as by their thermal evolution (Klemme and Ulmishek, 1991; Loucks et al., 2012; Wei et al., 2020). The intricate interaction between organic matter and inorganic minerals leads to significant variability in the generation, expulsion, retention, and accumulation of shale oil across different lithofacies. This variability has attracted increasing attention and constitutes a critical scientific issue in shale oil accumulation research. Micro-

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Peer review under the responsibility of China University of Petroleum (Beijing).

to nanometer-scale migration of shale oil occurs within intra- or inter-laminaset regions, whereas millimeter-scale migration takes place through microfractures, moving from organic-rich to organic-poor interlayers (Liu et al., 2012; Hu et al., 2022). Previous studies have shown that greater volumetric expansion pressure develops during the maturation stage of organic-rich shales. This increase in pressure may facilitate hydrocarbon micro-migration and promote the development of both bedding-parallel and vertical microfractures within organic-rich laminasets. These microfractures establish efficient micro-migration pathways for shale oil (Li et al., 2023; Xu et al., 2024). During the early stages of thermal evolution, generated hydrocarbons are adsorbed onto mineral surfaces. As thermal evolution progresses, the laminasets become saturated with hydrocarbons and initiate micro-migration in a free state through intergranular pores and microfractures (Xu et al., 2022; Gao et al., 2023). Further studies indicate that shale oil occurrence patterns are controlled by hydrocarbon properties and by the shape and size of reservoir spaces, which are closely related to organic matter type, inorganic mineral composition, and the stage of thermal evolution (Bai et al., 2025).

Various organic and inorganic geochemical methods, spanning from macro- to micro-scales, have been employed to elucidate the micro-migration process of shale oil. The micro-scale migration and accumulation of shale oil under static conditions have been observed and characterized (Ji et al., 2024; Hu et al., 2025). Rock pyrolysis is currently used as a qualitative or semi-quantitative method to characterize shale oil micro-migration, reflecting both the phenomenon and efficiency of micro-migration. A widely accepted consensus suggests that positive anomalies in the OSI index and PI, together with negative anomalies in $T_{\max}$, indicate shale oil enrichment. The ratio of hydrocarbon generation potential to retained hydrocarbons in individual laminae has been used to quantify migration efficiency (Jarvie, 2014; Gao et al., 2024). During the micro-migration of shale oil, geochromatographic effects occur. Therefore, a series of experimental methods commonly used to characterize macro-scale migration have also been applied. The trace element method proposes that a decreasing Mn content in minerals may indicate the hydrocarbon migration pathway, based on the stability of manganese (Mn) (Hu et al., 2024). Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS) infers micro-migration pathways by quantifying the similarity of non-hydrocarbon components in shale oil (Hou et al., 2020; Ma et al., 2023). Core-flooding extraction experiments provide supplementary evidence for compositional changes during shale oil micro-migration (Mohnhoff et al., 2016; Xie et al., 2019; Hu et al., 2025). These conventional methods primarily determine migration direction by comparing the compositional similarity and concentration of shale oil. From a microscopic perspective, variations in the spectral characteristics of light and heavy components have been used to indicate shale oil micro-migration. The underlying principle is that lighter components consist of smaller molecules and exhibit higher mobility, whereas heavier components are more polar. Laser confocal analysis identifies micro-migration direction based on the fluorescence intensity of heavy components at 600 nm to 800 nm relative to light components at 490 nm to 600 nm in shale oil (Gao et al., 2023; Hu et al., 2024). Two-dimensional nuclear magnetic resonance, known as 2D NMR, can indicate shale oil mobility. When combined with Quantitative Grain Fluorescence (QGF), it provides insights into shale oil properties and maturity (Khatibi et al., 2019; Liu et al., 2019). In addition, synchronous fluorescence spectroscopy and total synchronous fluorescence spectrograms distinguish different shale oil components according to

their spectral wavelength ranges, and analyze relative content changes to infer migration direction (Tian et al., 2022). Most of these methods face challenges such as component signal overlap, which makes clear differentiation difficult.

Numerous results regarding the micro-migration process of shale oil are influenced by the complex interplay of variations in hydrocarbon generation capacity, pore space types, mineral compositions, and oil properties (Wang et al., 2025). However, the dynamic evolution of hydrocarbon generation, expulsion, retention, and accumulation has not been thoroughly discussed. The extent of generation, expulsion, and retention of shale oil cannot be quantitatively characterized within or between laminasets. These issues are crucial for accurately understanding the micro-migration and accumulation processes of shale oil, which are key aspects of our study.

This study focuses on the organic matter-rich tuffaceous shale of the 3rd sub-member of the Yanchang Formation's seventh member (Chang 7₃ sub-member) in the Ordos Basin, with the aim of characterizing micro-migration and accumulation among laminasets during shale oil generation. This study deviates from conventional high-temperature and high-pressure thermal simulation experiments on bulk shale samples by instead utilizing shale samples with precisely defined laminaset stratifications. A systematic array of analytical techniques has been employed, including measurement of total organic carbon (TOC) content, multi-stage pyrolysis, focused ion beam-scanning electron microscopy (FIB-SEM), low-temperature nitrogen adsorption, X-ray diffraction (XRD), and spectrofluorimetry analysis. Based on the geological chromatographic effect and the carbon balance principle during hydrocarbon generation and migration, this study innovatively proposes a conceptual framework and methodology for characterizing shale oil micro-migration and accumulation processes at the lamina scale, by integrating multi-stage pyrolysis. Furthermore, by incorporating fluorescence spectroscopy, we propose qualitative parameters to investigate migration pathways and the associated compositional changes in shale oil during micro-migration. From the perspective of physical simulation in petroleum geology and organic geochemistry, this study explores the differences in the micro-migration processes and accumulation mechanisms of shale oil across various laminasets. The influence of the processes of hydrocarbon generation, expulsion, retention, and accumulation of shale oil on their differential enrichment mechanisms is expected to be thoroughly revealed. This provides valuable insights for further elucidating the mechanisms of shale oil micro-migration and accumulation in laminated lithofacies.

2. Geological setting

The Ordos Basin is a large-scale, hydrocarbon-rich cratonic basin characterized by multicyclic sedimentary processes. It is situated in the western part of the North China Block and exhibited tectonic stability during the Early to Middle Triassic (Fig. 1(a)) (Li et al., 2020). By the Late Triassic, the Qinling-Dabie Orogenic Belt had formed as a result of the collision and amalgamation of the North and South China Plates, which was accompanied by extensive volcanic activity and frequent hydrothermal events at the basin margin (Lu et al., 2021; Zhang et al., 2021; Guo et al., 2023). During the sedimentary phase of the Chang 7₃ sub-member, volcanic ash was extensively dispersed across the lacustrine basin, supplying essential nutrients such as P and Fe that stimulated the growth of aquatic plankton, significantly enhancing the basin's primary productivity (Lu et al., 2021; Liu et al., 2024).

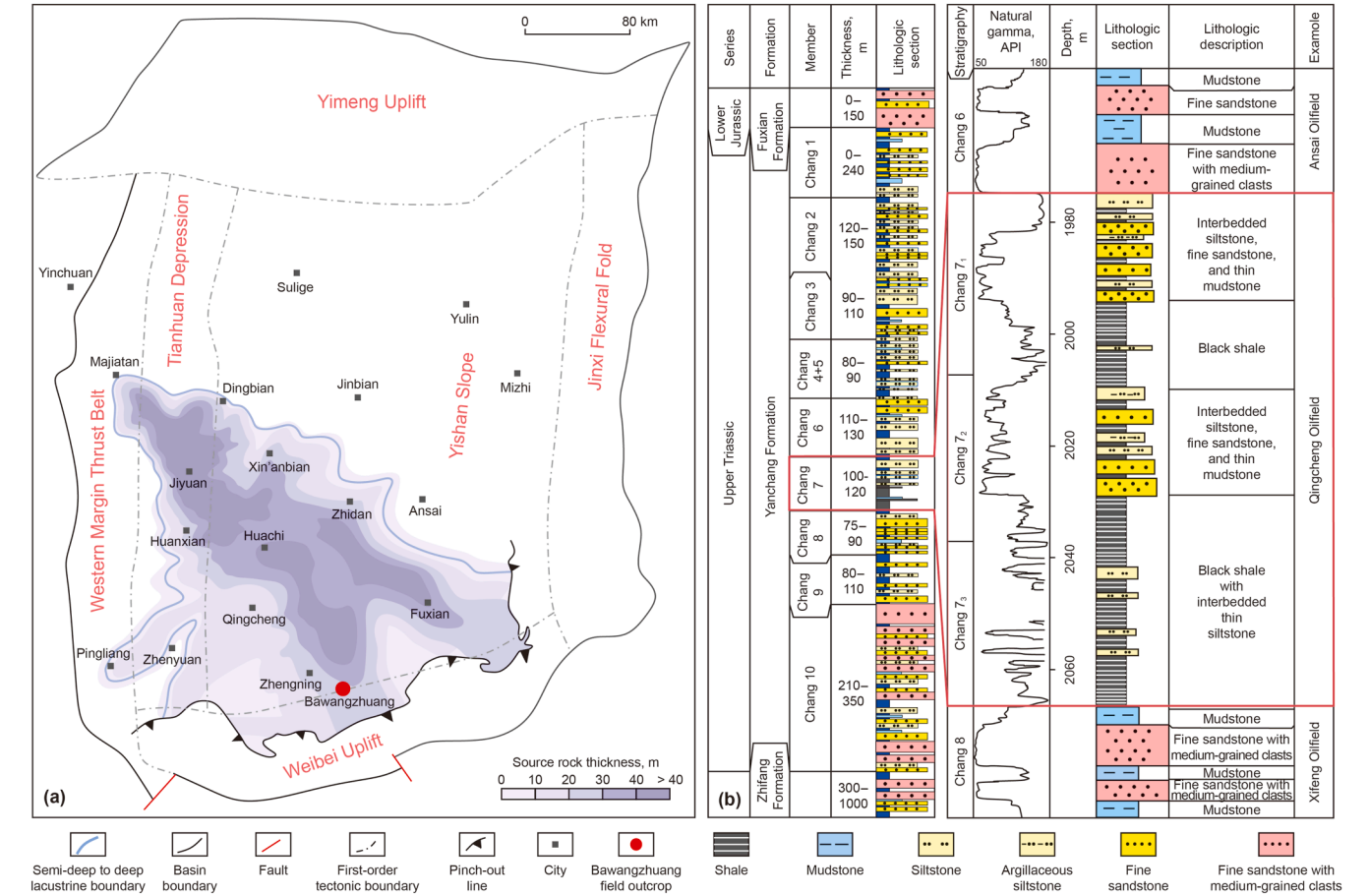


Fig. 1. (a) Tectonic units of the Ordos Basin (modified from Fu et al., 2020); (b) stratigraphic column of the Chang 73 Member in the Ordos Basin (modified from Xi et al., 2020).

Concurrently, persistent sulfidation occurred under stable dysoxic conditions, where the presence of redox-sensitive elements (such as Cr, V, Ni, Cu, Zn) and H₂S further cultivated anoxic conditions at the bottom of the lacustrine basin (Yuan et al., 2021; Liu et al., 2023). As a result, the formation of super-enriched organic black shale was jointly driven by the warm and humid paleoclimatic conditions, ample accommodation space, abundant nutrients, favorable algal species, and the anoxic environment at the lake bottom during the Chang 7₃ sub-member sedimentary period (Zhang et al., 2021; Guo et al., 2023). Furthermore, high-frequency volcanic activity led to the accumulation of numerous volcanic ashes. After deposition, volcanic ash commonly undergoes rapid devitrification and alteration during early burial, forming tuffaceous laminaset and releasing Si, Al, and alkalis into pore fluids. These reactive components can promote the precipitation of authigenic quartz and the transformation of volcanic glass and feldspar into clay minerals, for example, from smectite to illite or to illite and smectite mixed layers, during progressive diagenesis (Capelli et al., 2021).

Throughout the sedimentary period of the Chang 7₃ sub-member, thin volcanic tuff laminasets alternated with black organic-rich laminasets that featured sharp boundaries (Fig. 1(b)) (Xi et al., 2020; Liang et al., 2021). These organic-rich laminasets were found to contain a significant abundance of algae, primarily *Botryococcus* and the acritarch *Leiosphaeridia* (Ji et al., 2008; Gan et al., 2023). The immature to low-mature organic-rich shales of the Chang 7₃ sub-member are predominantly classified as type I and type II₁ kerogen. The average TOC content is measured at

16.1%, with a maximum reaching 39.39%. The S₁ + S₂ values range from 10.09 mg/g to 179 mg/g, whereas the Hydrogen Index (HI) values span from 182.58 mg/g to 751.39 mg/g, categorizing them as exceptionally high-quality source rocks (Zheng et al., 2022).

3. Samples and methods

3.1. Samples

The samples were collected from the fresh outcrop section at Bawangzhuang, located in the Tongchuan area, along the southern margin of the Ordos Basin (Fig. 1(a)). These samples are part of the super-enriched organic black shale of the Chang 7₃ sub-member. The shale samples exhibit tuffaceous laminaset and organo-clay complex laminaset, with a TOC content of as high as 30.65%, an HI value of 653 mg/g, and a pyrolysis peak temperature (T_{max}) of 433 °C (Table 1). The organic matter is classified as type I,

Table 1
Sample geochemical parameters.

Sample	TOC, %	S ₁ , mg/g	S ₂ , mg/g	S ₁ +S ₂ , mg/g	HI, mg/g	T _{max} , °C
Chang 7 member	16.01	4.02	15.71	19.73	525.56	442
Chang 7 ₃ sub-member	30.65	15.92	157.70	173.62	653	433
Bawangzhuang samples						

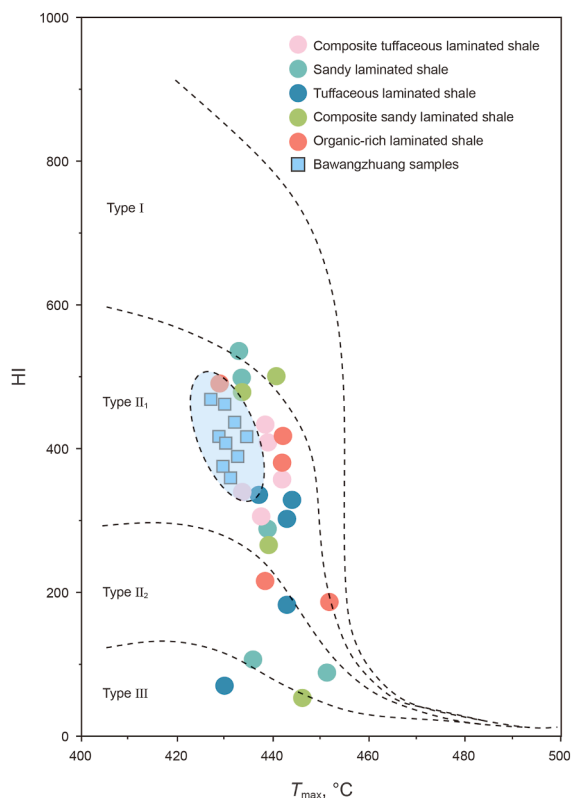


Fig. 2. HI- $T_{\max}$ plots extracted for organic matter types of various laminaset from the Chang 7₃ sub-member shale samples (modified from Xiao et al., 2025).

indicating that the samples are a low-maturity, super-enriched organic shales (Fig. 2).

3.2. Experimental design for the thermal simulation of different laminasets

The experimental apparatus employed in this study consists of a closed high-temperature and high-pressure simulation system (Fig. 3(a)). Ten identical cylindrical samples, each with a diameter of 25 mm, were extracted from a low-maturity block shale. These samples were subjected to a thermal regime, heating from room temperature (25 °C) to target temperatures of 200 °C, 250 °C, 275 °C, 300 °C, 325 °C, 350 °C, 375 °C, 400 °C, 450 °C, and 500 °C, with a duration of 12 h maintained at each level. The heating rate was set at 40 °C/h until reaching 200 °C, followed by a rate of 20 °C/h thereafter. After heating, the shale oil products generated at each specified temperature were collected for comprehensive analysis. Based on the hydrocarbon generation and expulsion curves derived from the produced samples, those heated to 300 °C, 375 °C, 400 °C, and 450 °C were selected for further investigation, which included TOC content, Rock-Eval pyrolysis, multi-stage pyrolysis, focused ion beam-scanning electron microscopy, X-ray diffraction, and low-pressure nitrogen (N₂) adsorption analysis (Fig. 3(b)).

3.3. Division, preparation, and measurement design for the laminaset samples

To conduct a comprehensive investigation of the micro-migration process of shale oil among various laminasets within the shale, samples from both pre- and post-thermal simulation experiments (total thickness: 42 mm; consisting of two distinct

laminasets: tuffaceous laminaset (TF) and organo-clay complex laminaset (OM)) were stratified into nine layers based on laminaset type. The thinnest layer is a 1.04-mm-thick OM lamina, whereas the thickest layer measures 14.32 mm in thickness (Fig. 4). The TF laminaset is dominated by felsic minerals (average 57.01%), with contents significantly higher than those of clay minerals (average 37.96%). Under plane-polarized light, TF laminaset appears light brown, and the mineral grains are uniformly dispersed. In the OM laminaset, clay mineral content reaches 41.57%, whereas felsic minerals account for 30.37%. Frequent interbedding and alternation of organic matter and pyrite bands are observed. Additionally, the OM laminaset exhibits a relatively high abundance of organic matter, with a higher density of algae observed under microscope compared to the TF laminaset. A detailed analysis of the physical and geochemical characteristics of these stratified samples (a total of 9 layers) was performed to elucidate the spatial redistribution process of shale oil as thermal evolution progresses. The investigation utilized TOC content assessment, multi-stage pyrolysis, X-ray diffraction, and low-pressure nitrogen adsorption tests, supplemented by imaging techniques, including spectrofluorimetry analysis and FIB-SEM observation.

3.3.1. Total organic carbon and Rock-Eval pyrolysis

Total organic carbon (TOC) determination is performed via a CS-230HC Carbon/Sulfur Analyzer. About 150 mg of the powdered sample is weighed and placed into a dedicated crucible, where it is treated with dilute hydrochloric acid in small aliquots to eliminate inorganic carbon. The sample is then washed repeatedly with distilled water until a neutral pH is achieved, followed by drying in a drying oven. It is subsequently loaded into the carbon/sulfur analyzer for instrumental analysis.

Rock-Eval pyrolysis analysis is conducted using a Rock-Eval pyrolysis instrument. Approximately 50 mg of the powdered sample is introduced into the pyrolysis oven, where it is heated according to a predefined temperature program. Hydrocarbon gases released from the sample at elevated temperatures are separated from the residual material using purge gas (helium) stripping and are quantitatively analyzed with a hydrogen flame ionization detector.

3.3.2. X-ray diffraction (XRD) experiment

X-ray diffraction (XRD) experiments are conducted utilizing a powder X-ray diffractometer. The test utilizes Cu-K α radiation with a step size of 0.02°. Before experimentation, shale oil is extracted from the sample, which is then dried and ground to a particle size of less than 40 μ m. The powdered sample is prepared into a test piece via the back-pressing method and is scanned at a rate of 2°/min over a range of 3° to 40°. The identification of mineral types is based on the positions of the diffraction lines observed in the resulting diffractogram. The intensity of each diffraction peak correlates with the content of its corresponding mineral. Additionally, mineral quantification is conducted using the K -value method following the integration of the diffraction peaks.

3.3.3. Low-pressure nitrogen adsorption experiment

Low-pressure nitrogen adsorption experiments are carried out via a Micromeritics ASAP-2460 fully automated surface area and porosity analyzer. Before analysis, shale sample is crushed to a particle size within the range of 60–120 mesh, then placed in a sample tube and degassed under heat and vacuum for no less than 4 h. The degassed sample is subsequently immersed in liquid nitrogen to achieve cryogenic conditions. The adsorption experiment begins with a gradual increase in relative pressure (P/P_0) after the sample reaches adsorption equilibrium. As the relative

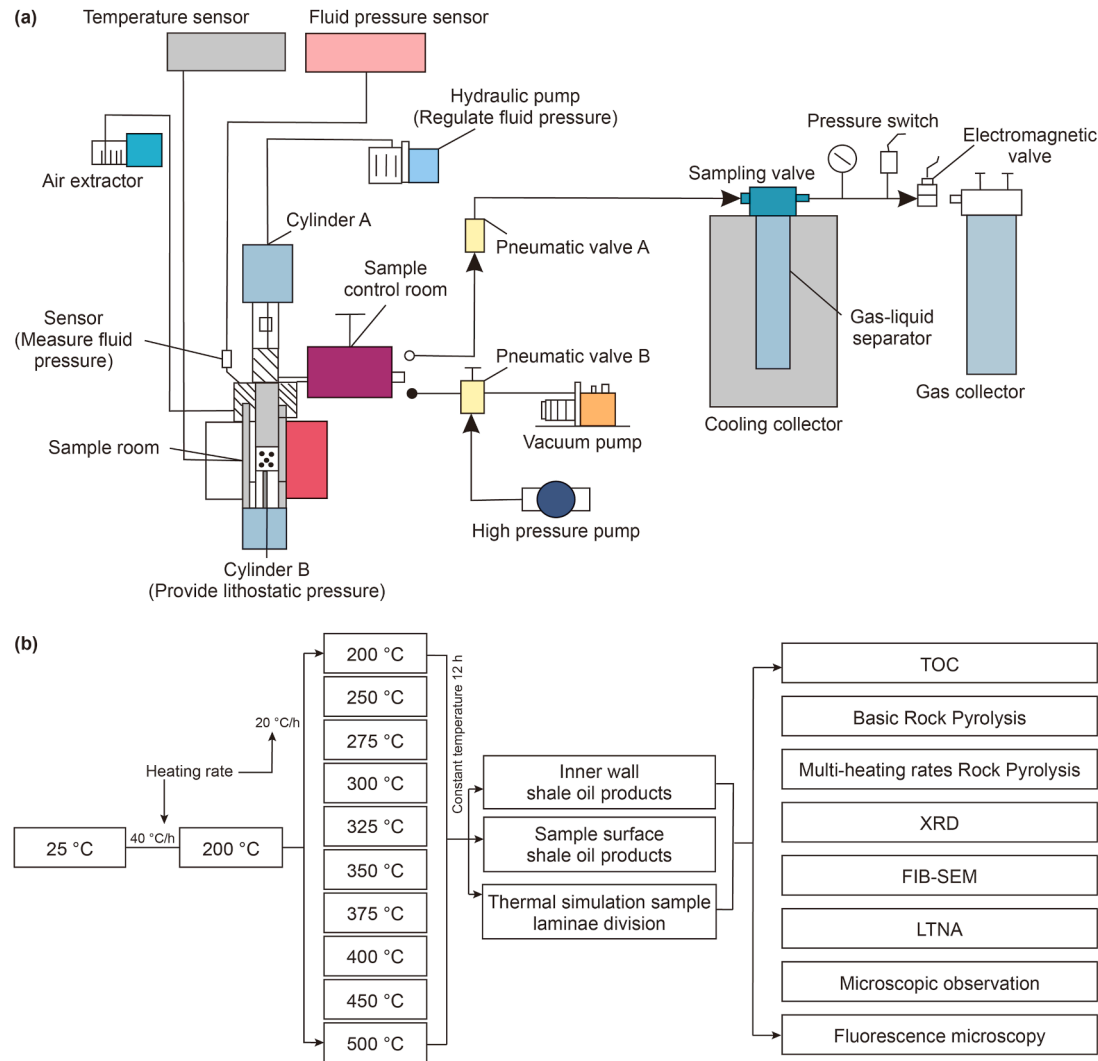


Fig. 3. (a) Schematic representation of thermal simulation equipment; (b) schematic workflow of the thermal simulation experiment.

Core sample	Lamination classification	Thickness, mm	Lamination	Test program						
				Rock Pyrolysis		TOC	XRD	FM	FIB-SEM	LTNA
				Basic	Multi-heating rates					
	No. 1	2.81	No. 1-TF	●	●	●	●	●		●
	No. 2	3.17	No. 2-OM	●	●	●	●	●		
	No. 3	3.83	No. 3-TF	●	●	●	●	●		
	No. 4	1.04	No. 4-OM	●	●	●	●	●		
	No. 5	5.03	No. 5-TF	●	●	●	●	●	●	
	No. 6	14.32	No. 6-OM	●	●	●	●	●	●	●
	No. 7	8.00	No. 7-TF	●	●	●	●	●		
	No. 8	1.27	No. 8-OM	●	●	●	●	●		●
	No. 9	2.53	No. 9-TF	●	●	●	●	●		

Fig. 4. Division scheme of laminasets of the selected core samples and related measurements during the thermal simulation process.

pressure nears the saturation vapor pressure ($P_0 \approx 0.101$ MPa), the pressure is carefully reduced until complete nitrogen desorption from the sample is achieved.

3.3.4. Focused ion beam-scanning electron microscope

Field emission scanning electron microscopy (FESEM) is conducted using a Zeiss Crossbeam 550 Focused Ion Beam-Scanning Electron Microscope (FIB-SEM) dual-beam system. Initially, the sample, cut to dimensions of 8 mm × 8 mm, is coarsely polished with abrasive paper. Following this, the cross-section perpendicular to the direction of the laminaset of the sample undergoes argon ion milling using a precision etching coating system (PECS II). Finally, the argon-ion-milled surface is sputter-coated with a platinum layer to ensure an artifact-free smooth surface.

4. Results

4.1. Bulk organic geochemistry characteristics of total expulsion shale oil from the whole laminaset

With increasing thermal maturity, the total expelled amount of shale oil from the entire shale column demonstrates a pattern characterized by an initial rise followed by a decline. The organic matter thermal evolution process can be segmented into four distinct stages based on the rate of change of the total hydrocarbon expulsion curve for the shale column (Fig. 5). Stage A (200–300 °C) represents the slow hydrocarbon generation phase, marked by a low expulsion rate and a relatively flat curve slope. Stage B

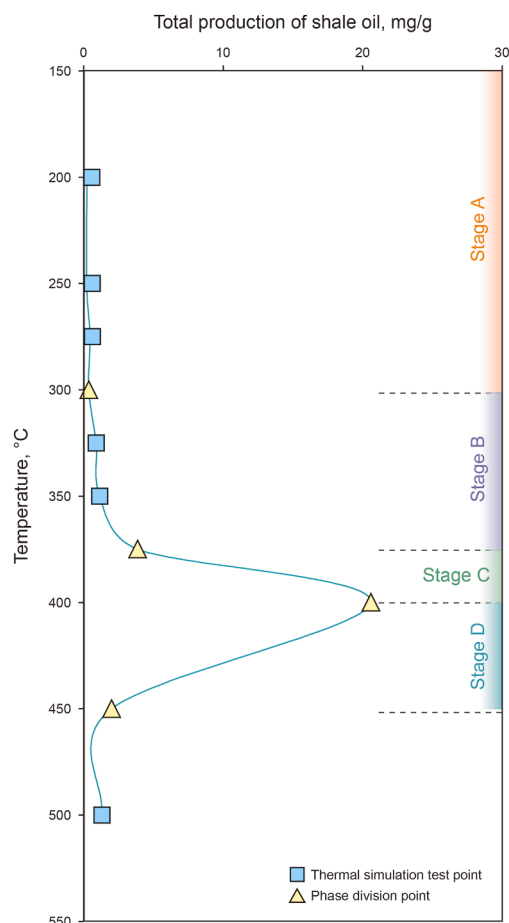


Fig. 5. Total hydrocarbon expulsion curve of the core samples.

(300–375 °C) signifies the ascent phase of hydrocarbon generation and expulsion, during which expelled volumes increase compared to Stage A, and the curve slope rises moderately. Stage C (375–400 °C) is defined by a rapid surge in the expelled oil volume, achieving peak hydrocarbon generation with the curve slope reaching its maximum value. Finally, Stage D (400–450 °C) is characterized by a sharp decline in expelled oil amount as the system transitions into the gas-prone secondary cracking phase, resulting in a significant reduction in the curve slope. It should be noted that Stages A to D are experimental thermal evolution stages defined by the inflection characteristics of the total hydrocarbon expulsion curve (Fig. 5) and the corresponding heating temperatures.

The original organic-rich shale exhibits a high overall TOC content and considerable heterogeneity. The maximum TOC value recorded is 33.5% (No. 4), whereas the minimum is at 29.4% (No. 8) (Fig. 6). As thermal maturity increases, both the TOC and hydrocarbon potential ($S_1 + S_2$) of all laminasets progressively decrease. Notably, the greatest reduction in TOC content occurs in the TF laminaset (represented by No. 1), which declines from 30.9% to 20.2%; conversely, the OM laminaset (represented by No. 2) exhibits a substantial TOC decrease of 11.5%. The free hydrocarbon content (S_1) curve initially rises and then falls with increasing maturity, peaking during Stage C. The greatest S_1 increase (from 14 mg/g to 25.87 mg/g) is observed in layers No. 2, 3, 5, and 8. Overall, there is a more significant variation in pyrolyzable hydrocarbons (S_2) within the OM laminaset than in the TF laminaset (Fig. 6).

4.2. Evolutionary characteristics of mineral composition in various laminasets

Significant mineralogical heterogeneity is observed across different laminasets, exhibiting a cyclical pattern of variation. The TF laminaset primarily consists of quartz and clay minerals, featuring a higher quartz content (51.68%) compared to clay content (37.96%), along with pyrite (6.64%) and feldspar (5.33%). Conversely, the OM laminaset shows a markedly higher clay mineral content (41.57%) relative to quartz (25.77%), while pyrite is significantly more prevalent within the OM laminaset (28.04%). The feldspar content in the OM laminaset remains similar to that in the TF laminaset, albeit low (4.6%). During the thermal evolution of organic matter, the TF laminaset experiences a minor reduction in feldspar content, declining from 5.33% to 3.19%, alongside slight increases in clay minerals (up to 44.77%) and quartz (rising to 52.29%). This is primarily attributed to the degradation of abundant organic matter, which generates organic acids that accelerate feldspar dissolution and release K^+ into the pore fluids. This process catalyzes the transformation of smectite to illite, leading to the progressive formation of illite and smectite mixed layers (Xiong et al., 2022). During the conversion of smectite to illite, the released silica precipitates as authigenic microcrystalline quartz (Cao et al., 2025). The pyrite content in the TF laminaset reduces from 6.64% to 0.54%, whereas in the OM laminaset, it plummets from 28.04% to 2.76% with increasing maturity (Fig. 7). Illitization of feldspar is not pronounced in the OM laminaset, whereas increases of varying magnitudes are observed in feldspar (from 4.6% to 5.03%), clay minerals (from 41.57% to 43.77%), and quartz (from 25.77% to 48.42%) contents (Fig. 7).

4.3. Evolutionary characteristics of pore-throat size distribution development in different laminaset

According to the IUPAC pore size classification scheme, shale pores are classified into three categories: micropores (<2 nm),

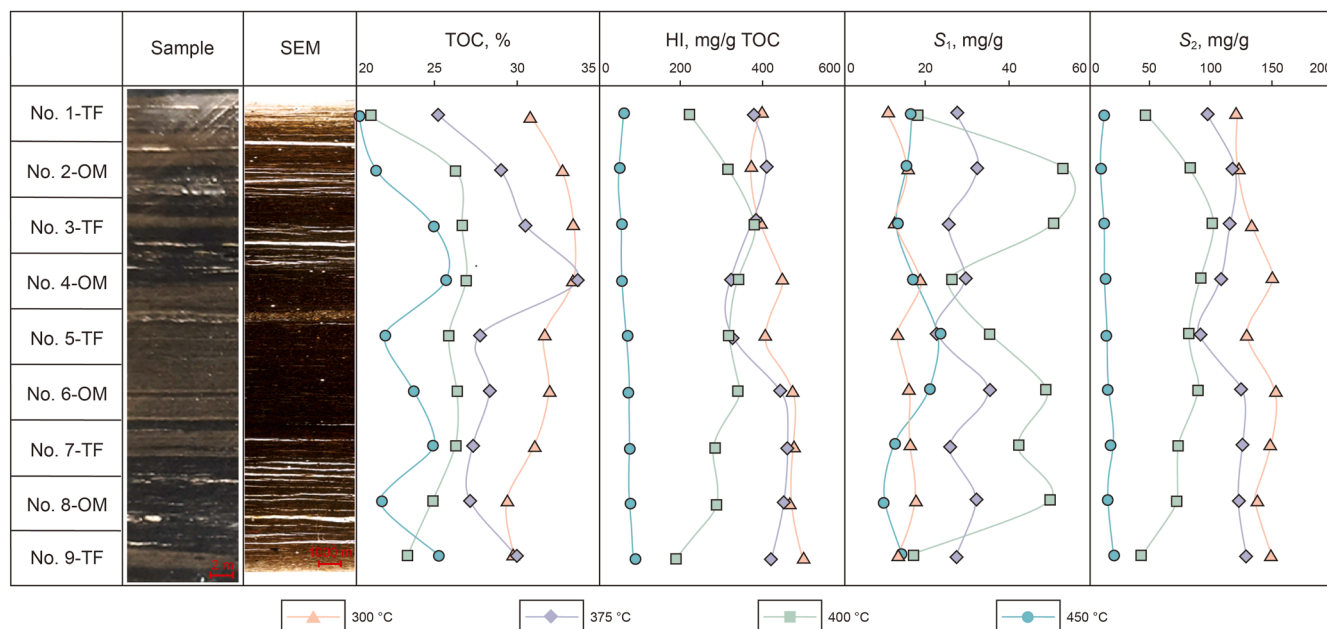


Fig. 6. Evolution plots of organic geochemical characteristics for various thermal evolution stages and laminasets.

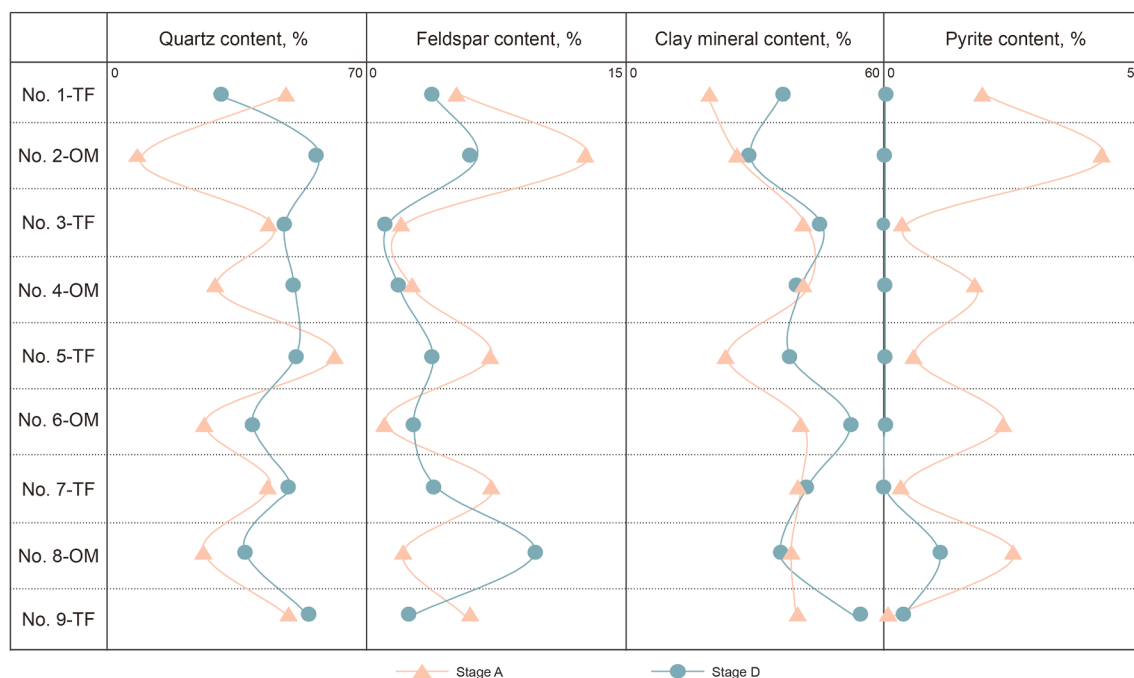


Fig. 7. Mineralogical evolution plots for various thermal maturity stages and laminasets.

mesopores (2–50 nm), and macropores (50–100 nm) (Sing, 1985). The pore size distribution curves at various stages of thermal evolution display a multimodal characteristic, indicating a broad distribution range (Fig. 8). The primary peak is predominantly found within the macropore range of 80–100 nm. Additionally, distinct proportions of pores are found across mesopore size ranges of 65–70 nm, 50–60 nm, 35–40 nm, and 25–30 nm within various laminasets. The TF laminaset shows a greater increase in pore volume compared to the OM laminaset, suggesting more developed porosity within the TF laminaset. Nitrogen adsorption analyses reveal that mesopores

and macropores are the dominant pore types across all thermal evolution stages, comprising 24.22%–51.15% and 25.9%–27.48% of the total pore volume, respectively. Micropores make up the smallest proportion, approximately 0.76%–8.1%. At similar maturity stages, the TF laminaset exhibits a lower proportion of macropores compared to the OM laminaset, while its mesopore proportion is higher. This trend is especially evident during evolutionary Stages A, B, and C. The proportion of macropores initially increases, followed by a decrease from Stage A to Stage D, exhibiting an inverse trend relative to that of mesopores (Fig. 9).

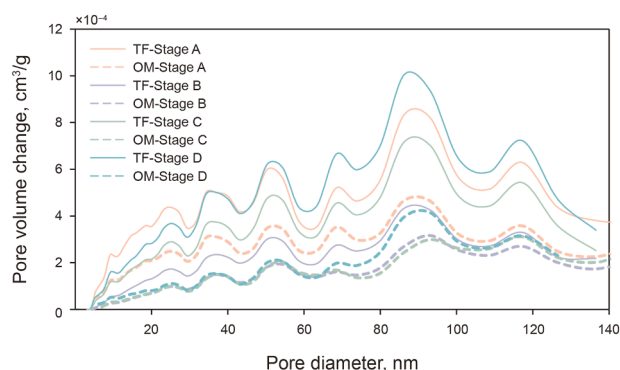


Fig. 8. Evolution plots of the pore volume change for various thermal evolution stages and laminasets.

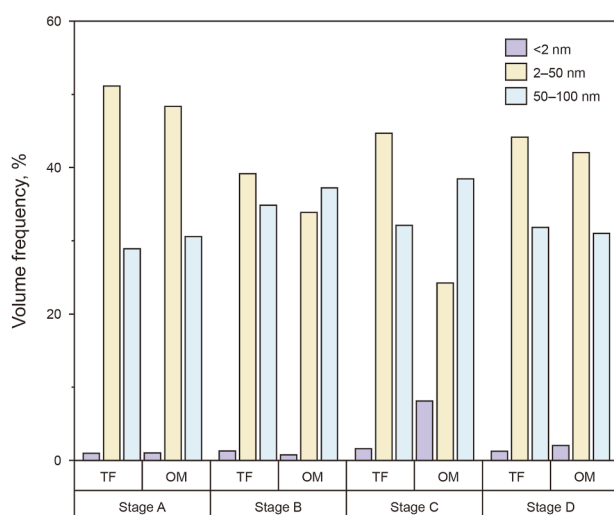


Fig. 9. Pore size distribution evolution for various thermal evolution stages and laminasets.

4.4. Thermal evolutionary characteristics of organic matter in various laminasets

The super-enriched organic shale samples from the Chang 7₃ sub-member are rich in algae, including *Botryococcus braunii*, the acritarch *Leiosphaeridia*, and minor amounts of *Parasphaeroides ovatus* (Fig. 10(j)). During Stage A, the majority of algae maintain their original structure, displaying bright yellow fluorescence under ultraviolet excitation. Initial hydrocarbon production begins, as evidenced by trace shale oil adsorbed onto organic matter surfaces observed under microscopy (Fig. 10(j)). Significant degradation of algae and organic matter occurs during Stage B, mainly characterized by a decrease in algal fluorescence intensity and a predominant yellow-green hydrocarbon covering the algal body surfaces (Fig. 10(k)). Stage C represents the peak phase of hydrocarbon generation. Organic matter is nearly completely degraded, leaving only minor yellow fluorescent remnants and releasing substantial volumes of free oil (Fig. 10(l)). Stage D signifies the end of peak oil generation, during which the oil generation rate declines rapidly. At higher thermal maturity, the organic matter is entirely degraded to a black residue (Fig. 10(m)).

In Stage A, adsorbed or locally saturated free oil exhibiting yellow-green fluorescence is present on the surfaces of felsic minerals and within connected intergranular pores–fractures in the TF laminaset (Fig. 10(a)). Concurrently, minor transverse

microfractures develop parallel to the organo-clay complex lamina. The shale oil within these microfractures contains heavier components, as indicated by yellow fluorescence (Fig. 10(e)). During Stage B, hydrocarbon fluorescence steadily intensifies. Vertical fractures preferentially form around brittle mineral grains within the TF laminaset, whereas a limited number of high-angle fractures develop concurrently within the OM laminaset. The oil in these fractures remains relatively heavy, exhibiting a greenish-yellow color (Fig. 10(b) and (f)). Stage C is mainly marked by the development of numerous vertical fractures in both laminasets, resulting in an interconnected fracture network. The fracture density is substantially higher in the TF laminaset compared to the OM laminaset. Concurrently, the shale oil content within the laminae increases, accompanied by heightened fluorescence in the fractures. The quality of shale oil becomes lighter, transitioning from yellowish-green to green coloration. Minor adsorbed oil is also observed on mineral surfaces, alongside slight dissolution of felsic minerals (Fig. 10(c) and (g)). Stage D represents the transition into the gas generation phase. The shale oil content within the laminae lessens significantly, some high-angle fractures partially close, and the intensity of green fluorescence diminishes from shale oil adsorbed on mineral surfaces and within fractures (Fig. 10(d) and (h)).

5. Discussion

5.1. Property evolution of shale oil

During the thermal evolution from Stage A to Stage D, significant dynamic changes are observed in the composition and properties of shale oil. As maturity increases, the content of light components exhibits varying degrees of enhancement across all stages, while there is a corresponding reduction in the concentration of heavy oil components (Fig. 11(g) and (h)). Notably, the heavy oil index experiences a sharp decline during Stage C, accompanied by an increase in the light oil index. This shift can be primarily attributed to the intensified cracking of heavy oil and bitumen that were previously formed from kerogen during this stage, leading to a compositional profile that is increasingly dominated by light components. Furthermore, distinct variations in peak shapes are evident in the fluorescence spectra profiles of shale oil across different thermal evolution stages and positions within the laminaset. These spectral differences serve as indicators of the relative proportion changes between light and heavy components in the shale oil (Fig. 11(a) and (b)).

Through comparative analysis of fluorescence spectra of shale oil within the laminaset at the initial maturation stage, a prevalent bimodal pattern is observed. With increasing thermal maturity, the peak wavelength of the heavy component gradually shifts toward shorter wavelengths, indicating a blue shift. The fluorescence peak of the heavy component is distributed within the emission wavelength range of 600 nm to 800 nm, whereas that of the light component lies within 490 nm to 600 nm (Gao et al., 2023). We propose the parameter K , defined on the basis of the relative positions of fluorescence peaks, to characterize the dominant component and its evolution across different thermal evolution stages. The factor K is defined as follows:

$$K = \frac{\lambda_p - \lambda_s}{|\lambda_p - \lambda_s|} \times \frac{\lambda_{l0} - \lambda_{li}}{\lambda_{h0} - \lambda_{hi}} \quad (1)$$

Here, λ_{l0} and λ_{h0} represent the maximum emission wavelength of the light and heavy components at the initial stage, respectively, and λ_{li} and λ_{hi} represent the maximum emission wavelength of the light and heavy components at the current stage, respectively. λ_p

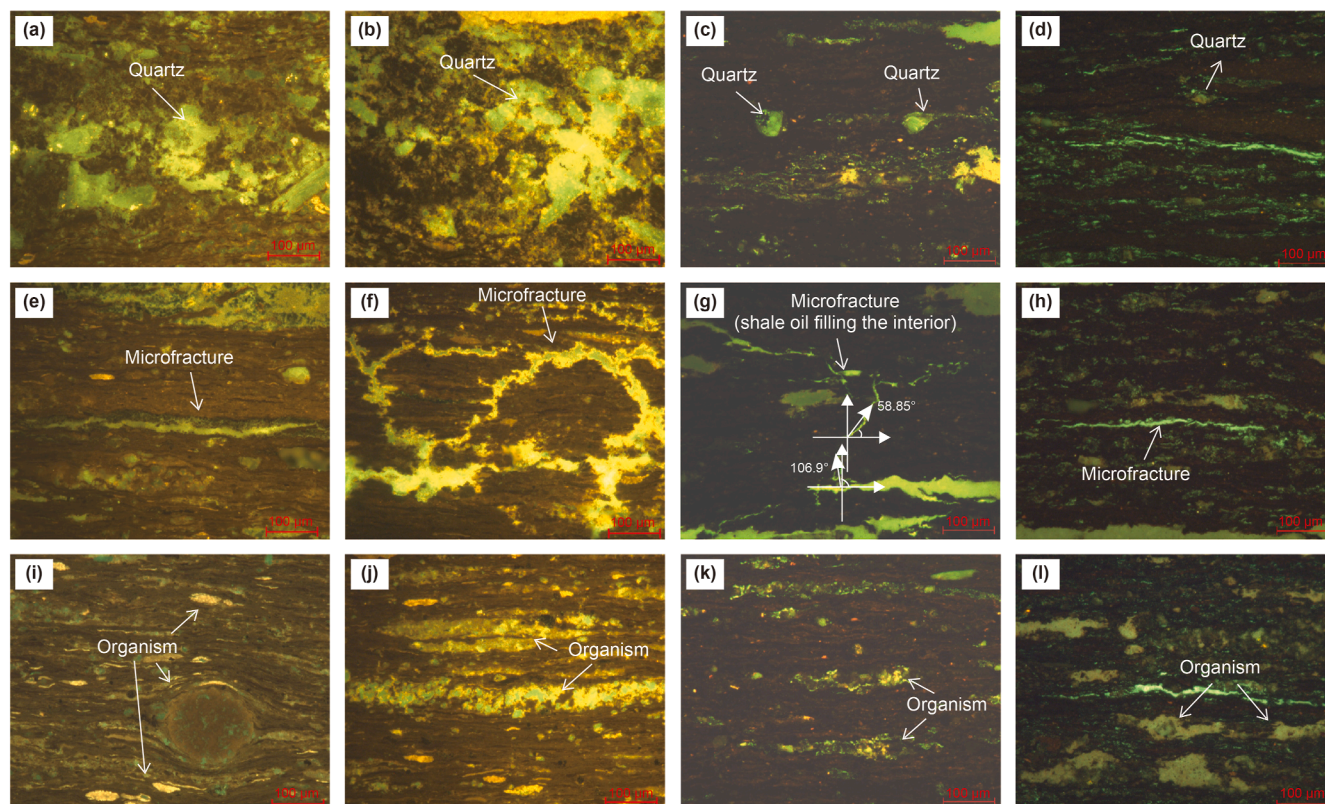


Fig. 10. Microscopic characteristics of shale oil for various thermal evolution stages. (a) Stage A: oil adsorbed on mineral surfaces; (b) Stage B: oil adsorbed on mineral surfaces; (c) Stage C: oil adsorbed on mineral surfaces; (d) Stage D: oil adsorbed on mineral surfaces; (e) Stage A: oil-bearing status in intra-layer microfractures; (f) Stage B: oil-bearing status in intra-layer microfractures; (g) Stage C: oil-bearing status in intra-layer microfractures; (h) Stage D: oil-bearing status in intra-layer microfractures; (i) Stage A: organic matter evolution status; (j) Stage B: organic matter evolution status; (k) Stage C: organic matter evolution status; (l) Stage D: organic matter evolution status.

and λ_s represent the wavelengths of the primary peak and the secondary peak at the current stage, respectively.

The positive and negative values of K are primarily defined by the ratio $(\lambda_p - \lambda_s)/|\lambda_p - \lambda_s|$. When $K > 0$, the fluorescence peak of the heavy component serves as the dominant peak, suggesting that the primary process is the conversion of kerogen into the heavy component. A larger K value indicates a more intensive conversion of kerogen into the heavy component in shale oil. In contrast, when $K < 0$, the light component dominates the shale oil. A smaller K value indicates a more substantial conversion of the heavy component into the light component and greater mobility of shale oil. In instances where the spectrum displays a unimodal shape at high thermal evolution stages, λ_{hi} continuously approaches the boundary between light and heavy components (600 nm). This denotes that the shale oil primarily consists of light components, with heavy components undergoing almost complete cracking.

An analysis of the fluorescence spectra of shale oil within fractures at various thermal evolution stages demonstrated that K values are positive in Stage A, where kerogen cracking predominantly generates heavy components. (Fig. 11(a)). At Stage B, thermal evolution enters the oil generation window. The K value can be either positive or negative. When the K value is positive, it indicates ongoing kerogen degradation, while the conversion of heavy components into light components is weak. In contrast, when the K value is negative, it suggests that the cracking of heavy components into light components at the measurement point gradually becomes more significant compared to kerogen cracking. During Stage C, λ_{hi} entirely corresponds to the peak wavelength of the light component, with the K value consistently

showing negative values. This indicates that the conversion of kerogen into heavy components has extensively concluded. At this stage, large-scale cracking of the heavy component occurs, leading to a significant increase in the content of the light component. In Stage D, the spectra exhibited a unimodal pattern, and the K value continued to decrease (Fig. 11(f)). At this point, the heavy components were entirely cracked, and the light component underwent secondary cracking into smaller molecular gases, further blue-shifting the primary peak wavelength.

The ratio of light to heavy components varies within fractures at different thermal evolution stages, and differences also exist between fractures and mineral pores at the same stage. Shale oil on mineral surfaces consistently exhibits lower K values than that within fractures, and these values gradually decrease as thermal evolution progresses. This indicates that mineral surfaces have a stronger tendency to adsorb the light component. The discrepancy in K values between mineral surfaces and fractures can primarily be attributed to the preferential migration of the light component, which remains within the mineral pore network.

5.2. Occurrence states of shale oil and its influencing factors

The occurrence states of shale oil primarily include free and adsorbed states. The pyrolysis parameter S_{2-1} quantifies variations in adsorbed shale oil, while S_{1-1} to S_{1-3} characterize changes in free-state shale oil (Pan et al., 2023). During the thermal evolution stage, the content of adsorbed oil initially increases, peaking at Stage C, before declining, whereas free oil content decreases continuously (Fig. 12(a) and (b)).

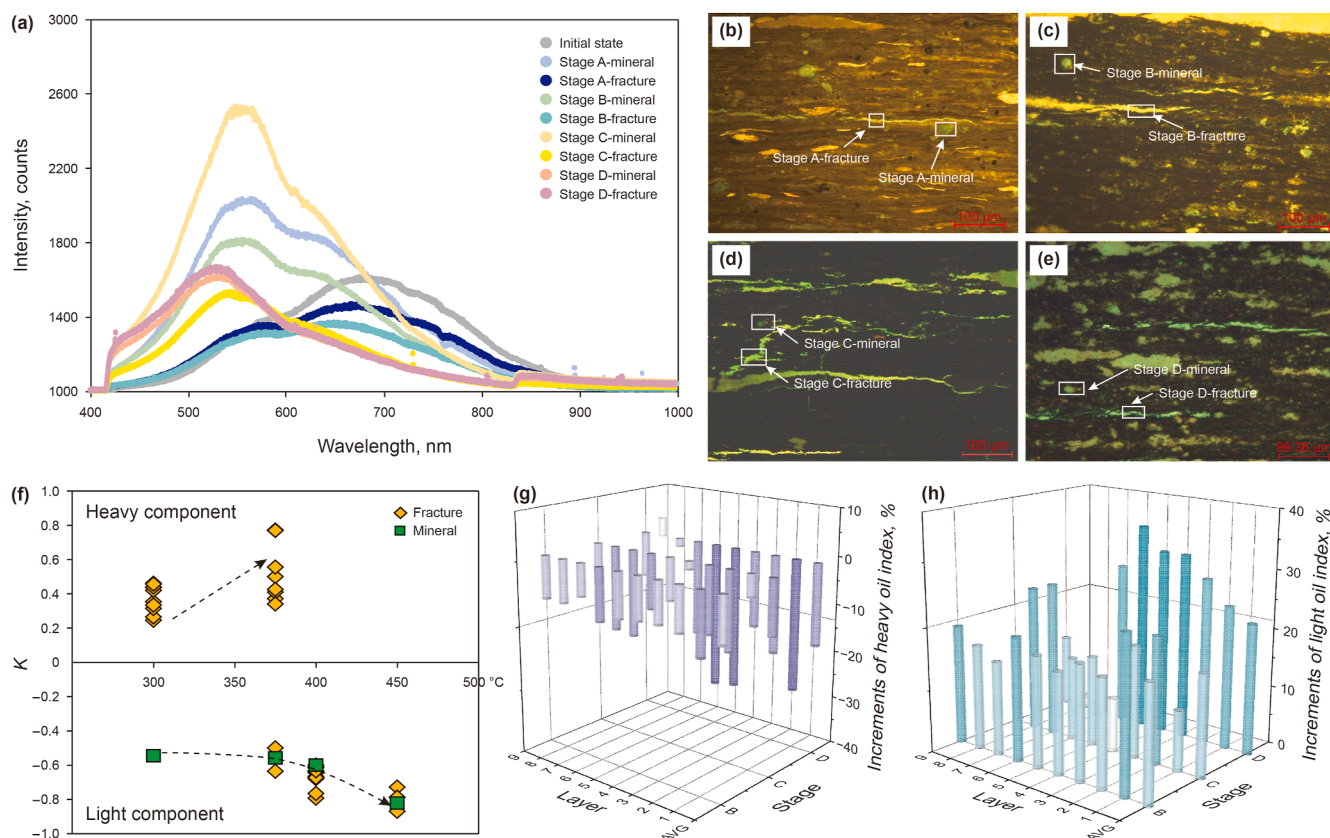


Fig. 11. Property evolution of shale oil for various thermal maturity stages and laminasets. (a) Fluorescence spectra of shale oil within cracks and on mineral surfaces for various thermal maturity stages; (b)–(e) the fluorescence spectra of Stages A, B, C and D corresponding to the position under the microscope, respectively; (f) correlation between K value and temperature; (g) change of heavy oil index in terms of the average (AVG) value and layer number; (h) change of light oil index in terms of the AVG value and layer number.

At the initial stage, the contents of shale oil in different occurrence states within the OM and TF laminasets showed clear differences. The free oil contents in the OM and TF laminasets were 15.38 mg/g and 13.35 mg/g, respectively, whereas the adsorbed oil contents were 68 mg/g and 58.27 mg/g, respectively. During Stage A, free oil begins to form at low concentrations, predominantly consisting of heavier components such as heavy hydrocarbons and bitumen, which must meet adsorption requirements before being expelled. These hydrocarbons are retained within the micropores and mesopores of the kerogen matrix, existing in adsorbed states. The TF laminaset exhibits a higher adsorbed oil content than the OM laminaset, with averages of 53.15 mg/g and 52.21 mg/g, respectively, due to its greater proportion of mesopores. As the system progresses to Stage B, organic matter reaches the threshold for hydrocarbon generation. Once kerogen achieves adsorption saturation, lighter components are preferentially expelled outward, leading to an increase in free oil content while the proportion of adsorbed oil begins to decline. In the OM laminaset, the smaller micropores and mesopores reach saturation more quickly and begin to discharge, resulting in a 15.66 mg/g increase in free oil content to a total of 32.52 mg/g, compared to a 12.72 mg/g increase to 25.75 mg/g in the TF laminaset. During Stage C, considerable kerogen degradation and cracking of macromolecular oils generate lighter oils, accelerating the conversion of adsorbed oil to free oil. Consequently, adsorbed oil content continues to decrease while free oil content reaches its peak. The TF laminaset retains more free oil due to its larger pore volume, demonstrating a 25.81 mg/g increase to 32.56 mg/g, whereas the OM laminaset records a 12.31 mg/g rise in free oil content to 44.83 mg/g. The remaining free oil is predominantly expelled through a network of nanoscale

and microscale microfractures. As the system enters Stage D, extensive cracking of free oil occurs, leading to a reduction in free hydrocarbon content, with predominantly light components expelled from the shale via the networked fractures. Average adsorbed oil contents in the TF and OM laminasets are 3.35 mg/g and 2.80 mg/g, respectively, whereas free oil content declines to 15.56 mg/g (TF) and 15.61 mg/g (OM).

The occurrence states of shale oil are influenced by various factors, including mainly the abundance of organic matter (characterized by TOC), the inorganic mineral composition, and reservoir space. Taking Stage C as an example, strong positive correlations were observed among adsorbed oil, free oil, and TOC content across different laminasets (Fig. 12(c) and (d)). Higher TOC content is associated with greater hydrocarbon-generating potential within laminaset, leading to increased hydrocarbon yields. Additionally, the substantial surface area of organic matter provides abundant adsorption sites for shale oil, while hydrocarbon-generation remnant pores serve as storage spaces for adsorbed oil. The relationship diagrams depicting free oil content in relation to distinct mineral components were also extracted and demonstrated (Fig. 12(e) and (f)). A prominent inverse relationship is noted between free oil content and the abundance of clay minerals, attributed to their exceptionally high specific surface area, which offers numerous adsorption sites for hydrocarbon molecules. Meanwhile, the small pore-throat diameters in clay-enriched intervals hinder free oil migration and increase the likelihood of throat plugging due to clay particles (Xu et al., 2022). A generally weak negative correlation is identified between free oil content and quartz mineral content. Under high TOC conditions, dissolution pores in quartz provide potential storage space for free

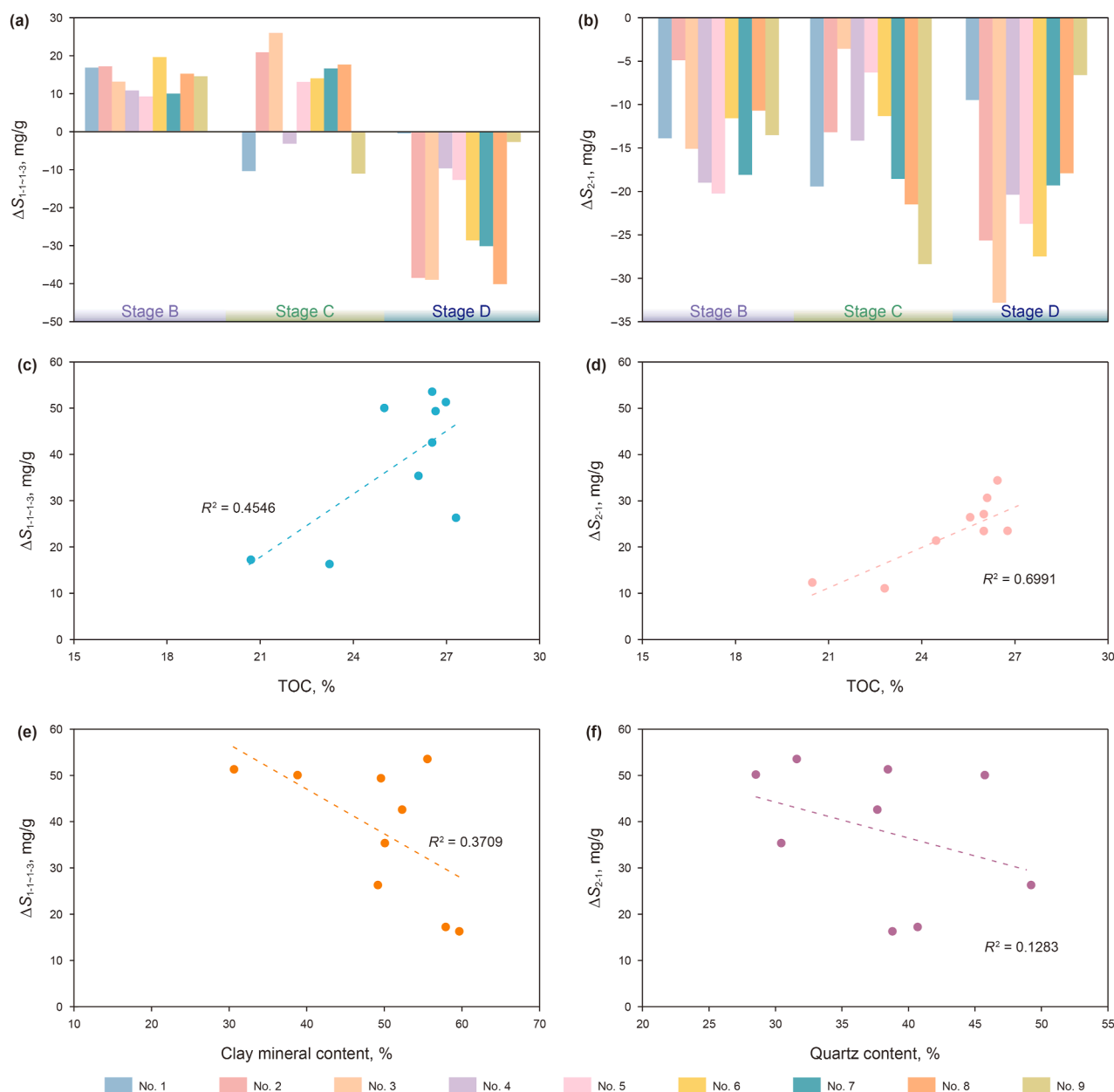


Fig. 12. Evolution of the contents of both the free hydrocarbons ($S_{1-1-1-3}$) and adsorbed hydrocarbons (S_{2-1}) and controlling factors on the shale oil occurrence states for various thermal maturity stages and laminasets. (a) Evolution contents of free hydrocarbons ($S_{1-1-1-3}$); (b) evolution contents of adsorbed hydrocarbons (S_{2-1}); (c) correlation between free hydrocarbons and TOC content; (d) correlation between adsorbed hydrocarbons and TOC content; (e) correlation between free hydrocarbons and clay minerals content; (f) correlation between adsorbed hydrocarbons and quartz content.

oil. However, pressure build-up from hydrocarbon generation leads to the formation of more macroscopic fractures, facilitating the expulsion of free hydrocarbons from the laminaset.

5.3. Micro-migration and accumulation of shale oil characterized by multi-stage pyrolysis parameters based on the carbon balance principle

The hydrocarbon generation capacity and yield of distinct laminasets conform to the carbon balance principle, in which kerogen degradation (indicating the original generative potential) is equivalent to hydrocarbon yield. In the absence of inter-laminaset material transfer, the amount of kerogen degradation within laminaset (S_{2-2}) would match its retained hydrocarbon

content ($S_{1-1-2-1}$). However, in subsurface environments, the hydrocarbons generated within a laminaset migrate to adjacent laminasets after meeting internal adsorption requirements. As a result, the degree of kerogen degradation significantly surpasses the retained oil content within the laminaset that is expelling hydrocarbons. Conversely, in a laminaset receiving hydrocarbon influx, the kerogen degradation is typically less than the retained oil content. Variations in multi-stage pyrolysis parameters during micro-migration suggest the parameter $\Delta S_{2-2} - \Delta S_{1-1-2-1}$ as a means to characterize hydrocarbon micro-migration among laminasets. A positive $\Delta S_{2-2} - \Delta S_{1-1-2-1}$ value indicates hydrocarbon expulsion from the laminaset, while a negative value denotes hydrocarbon influx into the laminaset. It is notable that during this stage, kerogen degradation occurs with increasing maturity,

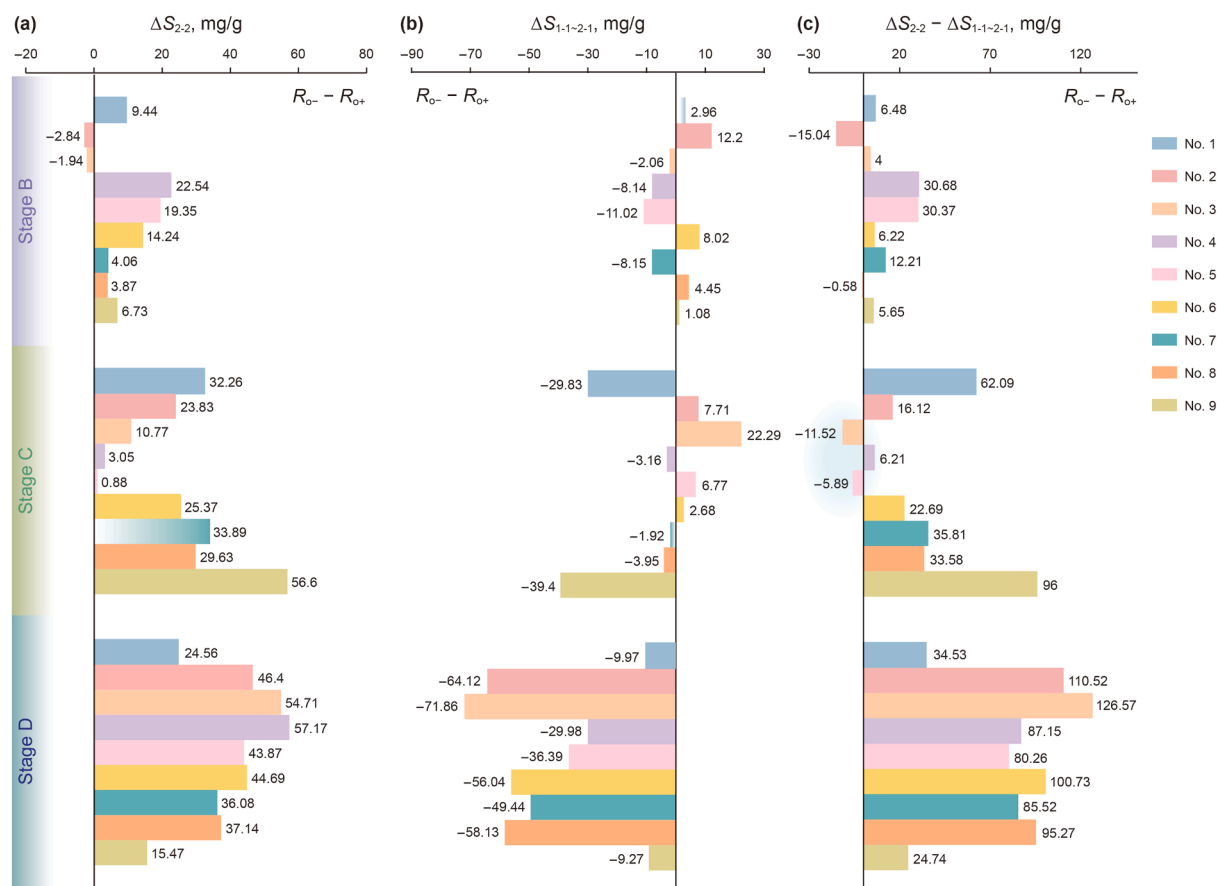


Fig. 13. Evolution graphs of retained hydrocarbon and expelled hydrocarbon content for various thermal evolution stages and laminasets. (a) Evolution graphs of the hydrocarbon generation change for various thermal evolution stages and laminasets; (b) evolution graphs of the retained hydrocarbon variation for various thermal evolution stages and laminasets; (c) evolution graphs of the migrating hydrocarbon variation for various thermal evolution stages and laminasets.

leading to a positive $\Delta S_{1-1-2-1}$ (retained hydrocarbon variation). A negative $\Delta S_{1-1-2-1}$ signifies the extensive expulsion of hydrocarbons generated within the laminaset.

Within the OM and TF laminaset of the Chang 7₃ sub-member, the hydrocarbon generation change (ΔS_{2-2}) began to rise during Stage B (Fig. 13(a)). Aside from some minor kerogen swelling observed in No. 2, kerogen degradation predominated in other laminae, with an average degradation of 24.03 mg/g and a maximum of 33.89 mg/g. Laminae No. 4, No. 5, and No. 6 emerged as the primary hydrocarbon-generating layers during this stage. At Stage C, $\Delta S_{2-2} - \Delta S_{1-1-2-1} < 0$ was observed in Laminae No. 3 and No. 5, indicating that the retained hydrocarbon content in these laminae exceeded the hydrocarbon generation change. This suggests hydrocarbon influx from adjacent laminae, in addition to in-situ generation in laminae No. 3 and No. 5, highlighting a dynamic transformation between hydrocarbon-generating laminaset and hydrocarbon reservoir laminaset. In contrast, $\Delta S_{2-2} - \Delta S_{1-1-2-1} > 0$ in other laminae signifies predominant hydrocarbon expulsion, demonstrating a clear inter-laminae hydrocarbon redistribution process (Fig. 13(c)). During Stage D, significant organic matter degradation was noted, resulting in an average ΔS_{2-2} degradation of 40.01 mg/g. This stage, corresponding to the oil-cracking gas-generation phase, showed that $\Delta S_{2-2} - \Delta S_{1-1-2-1} > 0$ occurred throughout the laminaset. This pattern implies potential inter-laminae hydrocarbon migration and further indicates that hydrocarbons cracked within the laminaset were primarily expelled as gas.

It is important to acknowledge that the experimental factors such as heating rate, isothermal holding time, and confining pressure can significantly influence kerogen cracking kinetics and the phase behavior of generated hydrocarbons, thereby affecting both the yield and composition of expulsion products (Wang et al., 2018; Han et al., 2026). Consequently, the calculated $\Delta S_{2-2} - \Delta S_{1-1-2-1}$ values should be regarded as semi-quantitative indicators of micro-migration potential rather than absolute quantifiers of migration flux. To minimize these uncertainties, all experiments in this study were conducted under standardized conditions, thereby ensuring internal comparability across different thermal stages and laminasets. Furthermore, the validity of shale oil migration interpretations is corroborated through cross-validation with independent petrophysical and geochemical datasets. Specifically, FIB-SEM observations directly reveal the development of microfractures and pore networks that spatially correspond to the inferred expulsion and accumulation zones. Fluorescence spectroscopy data exhibit systematic compositional variations in hydrocarbons along potential migration pathways, which are consistent with the redistribution patterns inferred from the $\Delta S_{2-2} - \Delta S_{1-1-2-1}$ parameter. Furthermore, pore size distribution analyses derived from nitrogen adsorption experiments display distinct evolutionary pathways across different laminasets. Collectively, this multi-proxy consistency substantiates the robustness of our conclusions, notwithstanding the inherently semi-quantitative nature of the threshold-based approach.

5.4. Micro-migration pathway and occurrence space of shale oil within various laminasets

Various pore types, including organic matter pores, clay mineral intercrystalline pores, pyrite intergranular pores, felsic grain intergranular pores, and intragranular pores, are developed in the black shale of the Chang 7₃ sub-member. The microscopic pathways for shale oil migration and their occurrence patterns are influenced by the contribution ratios of different pore types, which fluctuate due to shale oil generation, diagenetic alterations of inorganic minerals, and organic–inorganic interactions throughout the thermal evolution process. The development of interconnected intergranular pores, intra-/inter-laminaset microfractures, and cross-laminaset microfractures collectively forms the fracture-pore network system within the shale.

During Stage A, lower thermal maturity leads to limited hydrocarbon generation and minimal organic acid production, preserving the morphological integrity of mineral particles (Fig. 14(a)). The nitrogen adsorption-desorption curves indicate that the pore types present in Stage A are predominantly plate-like slit structures and wedge-shaped pores (Fig. 15). The TF laminaset exhibits a higher proportion of mesopores compared to the OM laminaset (Fig. 9), with intergranular pores among brittle minerals (e.g., quartz) being the primary contributors to mesoporosity. In contrast, the OM laminaset features a greater proportion of

macroporosity, primarily due to interlaminar microfractures in clay minerals that significantly contribute to macropore development (Figs. 9 and 14(e)). The partial light components migrate through percolation via intra- or inter-laminaset pathways in Stage A (Fig. 16(a), (b), (e) and (f)). At this stage, the density of microfractures within the OM laminaset is extremely low, with relatively low fracture angles. In contrast, due to differences in mineral composition and fabric, the density of bedding-parallel fractures within the TF laminaset is relatively higher (Fig. 17).

In Stage B, the generation of organic acids, accompanied by a small volume of liquid hydrocarbons, results in the formation of partial dissolution pores around and within felsic mineral grains (Fig. 14(b)). The interparticle and intraparticle pores within quartz minerals in the OM laminaset are partially filled with free hydrocarbons (Fig. 14(f)), whereas pyrite surfaces become coated with oil films composed of adsorbed hydrocarbons. Hydrocarbon-generation-induced pressure initiates the development of localized high-angle fractures within the OM laminaset, which serve as conduits for organic acids migrating from the OM to the TF laminaset (Qi et al., 2025). The number of microfractures within the laminaset begins to increase, and the fracture angles gradually become larger (Fig. 17). During Stage B, dissolution pores provide significant storage space, while shale oil migration occurs through two distinct mechanisms: inter-pore percolation and fracture-facilitated transport. Based on the analysis of fluorescence

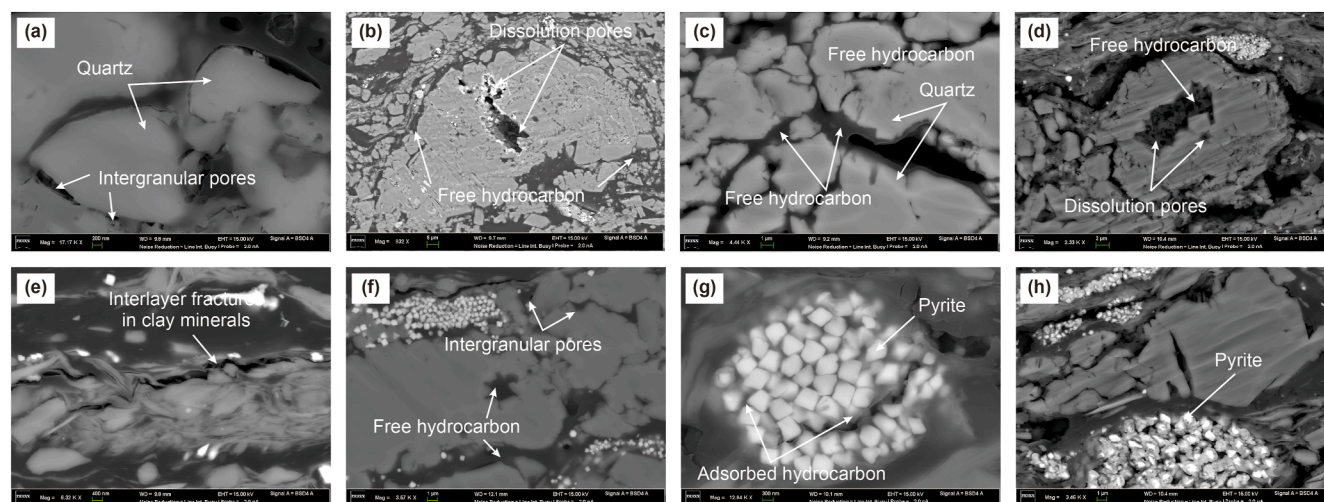


Fig. 14. Occurrence spaces of shale oil for various thermal evolution stages and laminasets. (a) TF-Stage A; (b) TF-Stage B; (c) TF-Stage C; (d) TF-Stage D; (e) OM-Stage A; (f) OM-Stage B; (g) OM-Stage C; (h) OM-Stage D.

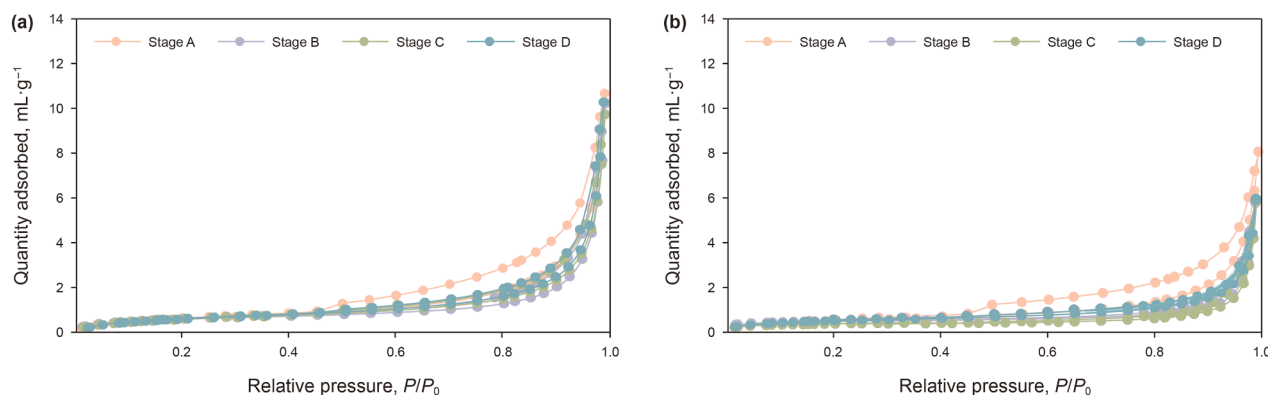


Fig. 15. Nitrogen adsorption-desorption curves for various thermal evolution stages and laminasets. (a) Nitrogen adsorption-desorption curves of TF laminaset for various thermal evolution stages; (b) nitrogen adsorption-desorption curves of OM laminaset for various thermal evolution stages.

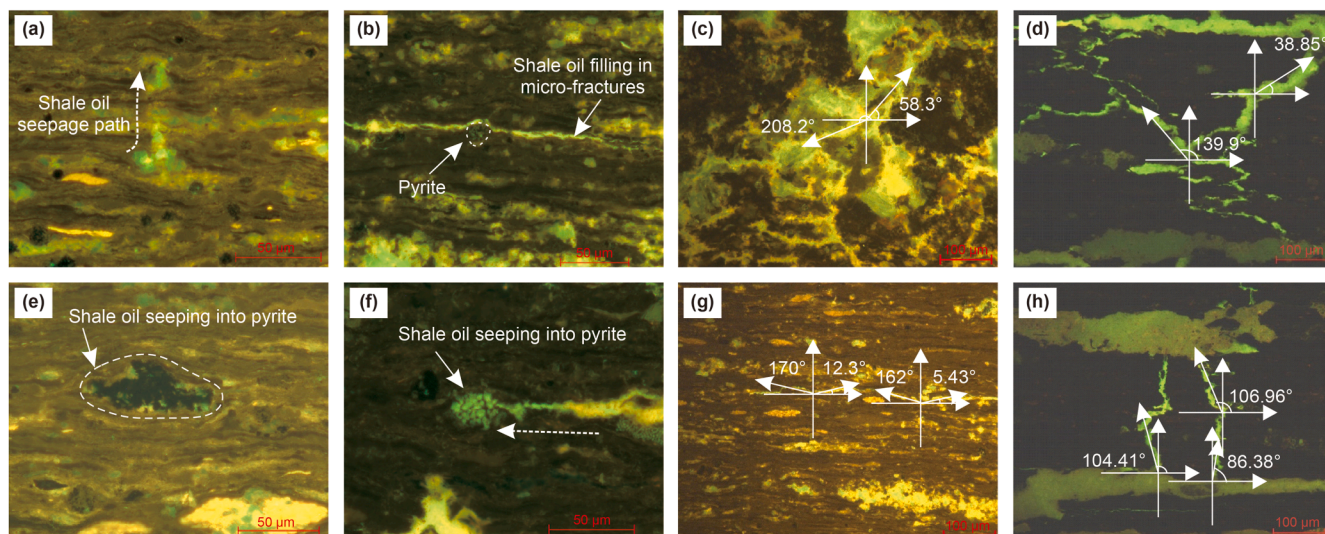


Fig. 16. Migration pathways spaces of shale oil for various thermal evolution stages and laminasets. (a) TF-Stage A shale oil seepage; (b) TF-Stage B shale oil micro-migration; (c) TF-Stage B microfractures; (d) TF-Stage C microfractures; (e) OM-Stage A shale oil seepage; (f) OM-Stage B shale oil seepage; (g) OM-Stage B microfractures; (h) OM-Stage C microfractures.

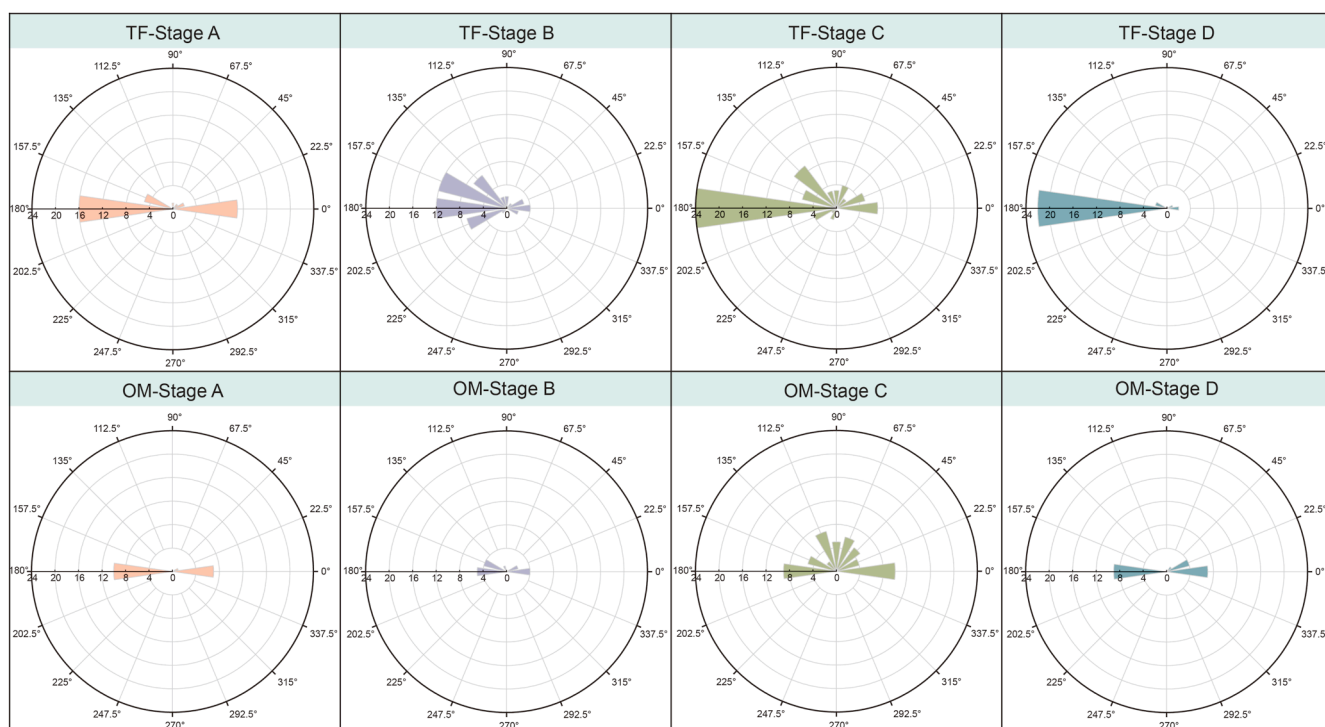


Fig. 17. Evolution diagrams of both the fracture quantity and angle for various thermal evolution stages and laminasets.

spectra, light components migrate preferentially, and the K value shows a gradual increase along the shale oil migration pathway. This trend is attributed to the higher mobility of light components.

By Stage C, oil films remain on the surfaces of pyrite particles and around localized felsic mineral grains (Fig. 14(g)). Large-scale liquid hydrocarbon charging, driven by hydrocarbon-generation-induced pressure, results in the formation of abundant high-angle fractures that facilitate hydrocarbon expulsion. Felsic minerals, subjected to compression-induced deformation, fracture, with hydrocarbons subsequently filling these fractures (Fig. 16(c)).

The abundant development of brittle minerals within the tuffaceous laminaset further promotes fracture formation. Nitrogen adsorption-desorption curves indicate that Stages B, C, and D predominantly exhibit open plate-like fracture pores and dead-end pores lacking adsorption hysteresis loops (Fig. 15). This suggests that fractures primarily function as pathways for hydrocarbon migration during the mid-to-high thermal evolution stages (Fig. 16(c), (d), (g) and (h)). The density of intra-laminaset fractures increases markedly, accompanied by a sharp rise in their frequency (Fig. 17). A complex, interconnected fracture network becomes visible via microscopy (Fig. 16 (d) and (h)).

In Stage D, previously formed hydrocarbon-generation-induced pressure fractures close, leading to a significant reduction in fracture density, with only limited bedding-parallel and tectonic fractures remaining (Fig. 17). The deformation damage intensifies on quartz mineral surfaces (Fig. 15(d)), while only minor intraparticle

dissolution pores are filled by organic matter. Under high-temperature conditions, pyrite experiences severe deformation, retaining minimal hydrocarbons (Fig. 14(h)). When a balance between the micro-migration and accumulation of shale oil is achieved across different stages, pre-existing interconnected

Table 2
Nitrogen adsorption experimental data at different thermal evolution stages.

Stage	Laminaset	S_{BET} , m ² /g	V_{tot} , cm ³ /g	V_{micro} , cm ³ /g	V_{meso} , cm ³ /g	D_{avg} , nm	Hysteresis loop
A	TF	3.1949	0.0196	0.000194	0.010060	24.626	H ₄
B	TF	0.9963	0.0077	0.000100	0.003028	31.065	H ₃
C	TF	2.3453	0.0139	0.000221	0.006224	23.752	H ₃
D	TF	3.4534	0.0190	0.000235	0.008389	22.012	H ₃
A	OM	1.7964	0.0114	0.000115	0.005527	25.444	H ₄
B	OM	0.5386	0.0050	0.000039	0.001720	37.722	H ₃
C	OM	0.5744	0.0052	0.000426	0.001275	36.678	H ₃
D	OM	1.3751	0.0066	0.000136	0.002802	19.383	H ₃

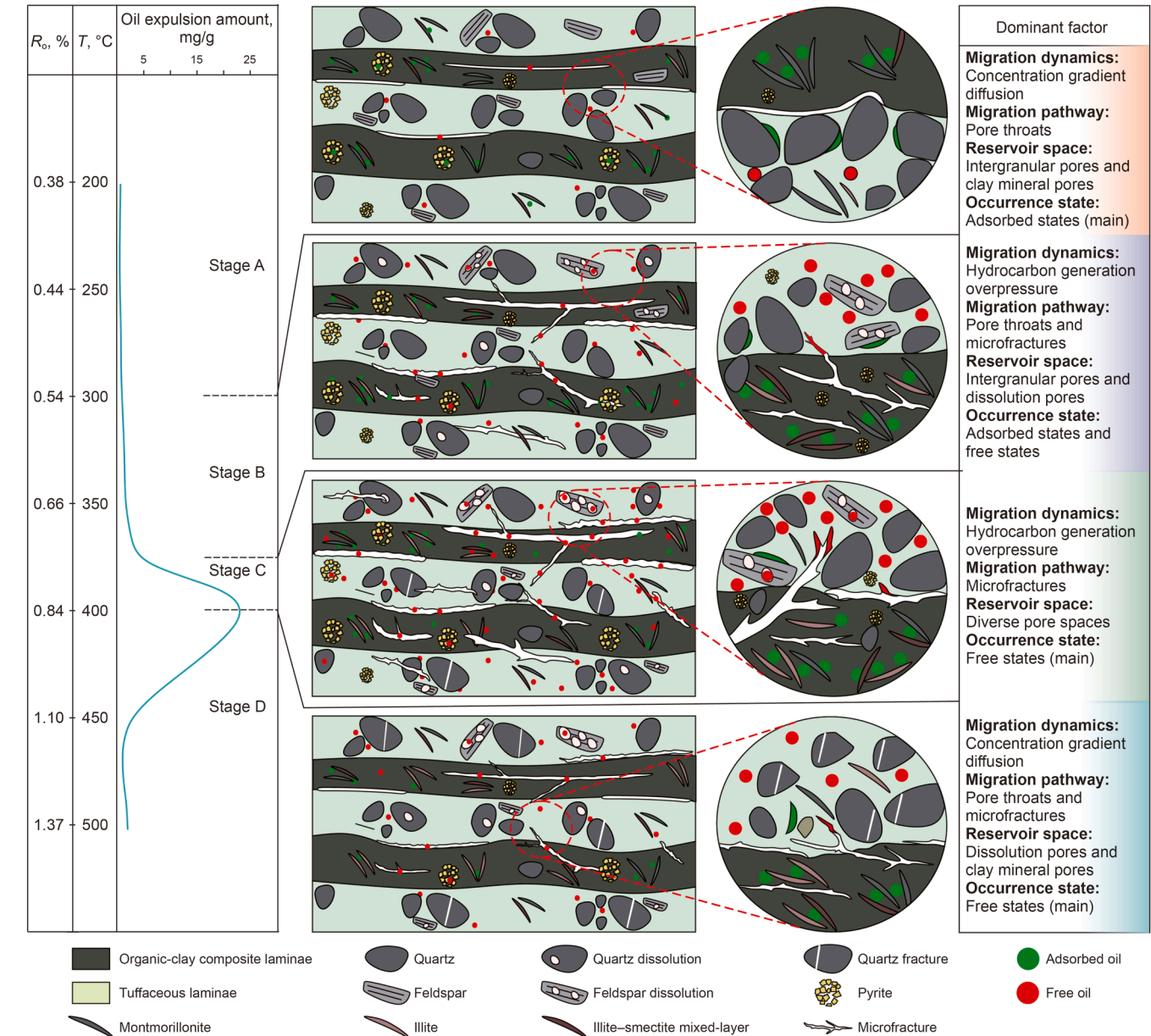


Fig. 18. Micro-migration and accumulation model of shale oil in organic-rich tuffaceous shale of Chang 7₃ sub-member.

intergranular pores, bedding fractures, and high-angle microfractures collectively serve as effective occurrence spaces. As the occurrence capacity increases, shale oil predominantly exists in a free state.

The nitrogen adsorption experiment provides quantitative constraints on the dynamic evolution of pore structures and allows us to better constrain the mechanisms of organic-inorganic interactions (Table 2). In the TF laminaset, total pore volume (V_{tot}), mesopore volume (V_{meso}) and specific surface area determined by Brunauer-Emmett-Teller method (S_{BET}) initially decrease at Stage B, followed by a continuous increase at Stages C and D, while the hysteresis loops transition from H_4 to H_3 . This trend indicates that at intermediate temperatures, organic-acid-induced mineral dissolution and early micro-fracturing contribute to the formation of slit-like or fracture-related pores, enhancing connectivity and facilitating micro-migration pathways. V_{micro} remains negligible throughout, suggesting that micropores are not the primary storage or migration medium in these laminasets. In the OM laminaset, V_{tot} and V_{meso} generally decrease at Stages B and C, suggesting that mineral transformation or precipitation processes partially fill pore space, while the hysteresis loops remain H_3 , indicating the persistence of slit-like pores formed earlier. At Stage D, a slight recovery of V_{meso} and a decrease in average pore diameter (D_{avg}) implies the combined effect of thermal cracking and secondary fracture formation, enhancing migration channels in OM laminaset. Overall, these stage-dependent trends quantitatively support the role of organic-inorganic interactions in shaping pore evolution and micro-migration pathways across both lamina types.

5.5. Micro-migration and accumulation model of shale oil in organic-rich tuffaceous shale

A comprehensive micro-migration-accumulation model has been developed to elucidate the processes underlying shale oil generation, expulsion, retention, and accumulation within the organic-rich tuffaceous shale of the Chang 7₃ sub-member (Fig. 18). This is essentially characterized by heavy components, mainly contained within kerogen in an adsorbed state, with smaller quantities adsorbing onto inorganic mineral surfaces during Stage A. In Stage B, localized preferential degradation of organic matter occurs, leading to the formation of organic acids and the concurrent illitization of feldspar minerals. This stage also witnesses the development of minor intragranular dissolution pores on the surfaces of felsic minerals, while the free oil content increases, mainly residing within intergranular and intragranular dissolution pores. Through this process, adsorbed hydrocarbons remain retained within the intercrystalline spaces of clay minerals. Stage C is mainly marked by significant shale oil generation accompanied by extensive microfracture development. These microfractures act as primary migration pathways for free hydrocarbons, predominantly located in intergranular pores, intragranular dissolution pores, and microfractures. A notable decline in heavy components is observed, with lighter components becoming predominant within the laminaset. As the thermal evolution progresses into Stage D, shale oil undergoes further cracking into gas, and previously formed hydrocarbon generation-induced pressure fractures close. This phase results in considerable deformation and destruction of felsic minerals and pyrite, with minor quantities of free oil being retained within intergranular pores (Fig. 18). The differential enrichment of shale oil is co-influenced by thermal, organic, and inorganic evolution. This integrated model offers vital theoretical insights into the enrichment mechanisms of shale oil across diverse lithofacies.

The micro-migration and accumulation model proposed herein is derived from a specific lithofacies (organic-rich tuffaceous shale of the Chang 7₃ sub-member) and therefore reflects the unique characteristics of this depositional system. The applicability of this model is greatest for shale systems characterized by similar lithological assemblages and depositional conditions, specifically those comprising volcanic ash interbeds or tuffaceous laminasets interbedded with organic-rich shales.

6. Conclusions

This study systematically elucidates the microscopic mechanisms of shale oil migration and accumulation within the organic-rich tuffaceous lithofacies of the Chang 7₃ sub-member in the Ordos Basin. This investigation employs thermal simulation experiments alongside finely divided laminaset and multi-scale organic-inorganic geochemical methodologies. The results obtained indicate that the migration and accumulation of shale oil between various laminasets result from the synergistic and dynamic evolution of organic matter-inorganic mineral interactions.

Four distinct stages are identified during the hydrocarbon generation and expulsion processes of organic matter within various laminasets. Based on the expulsion volume change rates of shale oil, four stages are delineated: the slow hydrocarbon generation stage (Stage A), the slow hydrocarbon generation-expulsion stage (Stage B), the rapid increase stage of massive hydrocarbon generation-expulsion (Stage C), and the rapid decrease stage of massive hydrocarbon generation-expulsion (Stage D).

Throughout the thermal evolution process, the tuffaceous laminaset (TF) exhibits a pronounced dynamic functional transformation between the hydrocarbon-generating laminaset and the hydrocarbon reservoir laminaset. During Stages A and B, the majority of OM and TF laminasets act as primary hydrocarbon-generating sources. By Stage C, the TF laminaset emerges as the predominant reservoir for shale oil. The parameter $\Delta S_{2-2} - S_{1-1-2-1}$, formulated based on material balance principles, quantifies hydrocarbon migration among different laminasets. Early-generated hydrocarbons migrate mainly through interconnected pore throats within and between laminasets via seepage. Light components preferentially migrate and adsorb onto mineral surfaces, interparticle pores, and edge fractures within the laminaset. In the middle to late stages, fractures become the principal migration pathways, with heavier components predominantly migrating through these fractures.

Shale oil primarily exists in two forms: free and adsorbed states. During Stage C, the TF laminaset, mainly characterized by a higher pore volume and more high-angle fractures, hosts a greater proportion of free hydrocarbons. Both types of laminasets develop abundant occurrence spaces. Free hydrocarbons are chiefly hosted within interparticle pores, intraparticle dissolution pores, and microfractures found in the TF laminaset. Conversely, adsorbed hydrocarbons are concentrated primarily within pyrite intercrystalline pores and clay mineral intercrystalline pores in the OM laminaset.

CRedit authorship contribution statement

Jin-Jun Xu: Writing – original draft, Methodology, Formal analysis, Data curation, Conceptualization. **Kai-Yue Li:** Writing – review & editing, Formal analysis, Data curation, Conceptualization. **Ying-Chang Cao:** Writing – review & editing, Formal analysis. **Xiao-Bing Niu:** Writing – review & editing, Visualization. **Ke-Lai Xi:** Writing – review & editing, Data curation. **Jie Wei:** Writing – review & editing, Investigation. **Xiu-Jian Ding:** Writing – review & editing, Data curation. **Xiu-Juan Wang:** Writing – review & editing, Formal

analysis. **Xiao Hui:** Investigation. **Yi-Ru Zhao:** Writing – review & editing, Investigation. **Jia-Yi Liu:** Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The research presented in this paper was supported by a general program from the Natural Science Foundation of Shandong Province, China (No. ZR2025MS692).

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