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Exploring the thermal evolution of highly- to over-mature source rocks using Raman spectroscopy and geochemical signatures

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ABSTRACT

This study investigated the heterogeneous responses of organic matter (OM) in highly- to over-mature source rocks during thermal maturation. An integrated analysis was conducted on the Raman spectroscopic and geochemical signatures of shales from the Lower Silurian Longmaxi Formation and the Lower Cambrian Qiongzhusi Formation, as well as anthracites from the Lower Permian Shanxi-Formation and the Upper Carboniferous Taiyuan Formation (collectively referred to as the Shanxi Taiyuan Formations). Additionally, burial and thermal evolution modeling was employed to support the analysis. A systematic assessment of Raman spectral parameters (e.g., the positions and intensity ratio of the D and G bands) revealed robust correlations between the thermal history patterns of source rocks and molecular structural evolution parameters. The subsequent mechanistic quantification demonstrated that the maturation state of the source rocks was subjected to the hierarchical control of three principal factors: Peak heating temperature, the duration of sustained thermal intensity, and effective maturation duration. In addition, comparative analyses demonstrated that the anthracites attained higher structural ordering under sustained thermal conditions. This contrasts with the disordered carbon matrices observed in the intermittently heated shales. Raman spectroscopy further revealed broader variations in the D and G band intensities of the Longmaxi Formation compared to the Qiongzhusi Formation. This difference is associated with their different thermal histories. The thermal burial histories confirm that shales in the Longmaxi Formation underwent thermal exposure at lower peak temperatures over a shorter duration compared to those in the Qiongzhusi Formation. Finally, this study established a maturity calibration model for over-mature source rocks through a systematic correlation between Raman peak height ratios ($R_{D/G}$) and vitrinite reflectance (R_o).

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1. Introduction

Highly- to over-mature source rocks are extensively distributed across multiple sedimentary basins in China due to the country's complex tectonic evolutionary history (Watson MP et al., 1987; He BZ et al., 2016; Zhao WZ et al., 2019). Comprehensive geochemical surveys are essential for their evaluation. This will offer critical insights into the quantity,

quality, and thermal maturity of sedimentary organic matter (OM), as well as implications for hydrocarbon generation, retention, and expulsion processes (Hackley PC and Cardott BJ, 2016; Yang SY and Horsfield B, 2020; Katz BJ and Lin F, 2021). For decades, traditional geochemical indicators, such as maximum pyrolysis temperature (T_{max}) from Rock-Eval pyrolysis, R_o , spore coloration index (SCI), solid bitumen reflectance (BR_o), and spectral micro-fluorescence, have played a significant role in assessing the thermal maturity and hydrocarbon generation of source-reservoir systems (Tissot BP 1984, Radke M et al., 1986; Zou CN et al., 2020; Cui GD et al., 2023). However, it is noteworthy that the strong heterogeneity of OM within highly mature source rocks leads to substantial variations in the physicochemical

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properties at the level of individual particles, thereby rendering individual geochemical methodologies inadequate for precise characterization of the chemical attributes of the source rocks (Roberts S et al., 1995; Henry DG et al., 2019).

In recent years, Raman spectroscopy has emerged as a widely employed technique for assessing maturity. This technique, primarily enabling the assessment of the OM aromaticity and the increasing density of aromatic moieties, has been widely used to characterize the structures of carbonaceous materials (Henry DG et al., 2019; Xu J et al., 2021; Huang BY et al., 2020; Yuan L et al., 2023; Gao ZW et al., 2022; Chen SY et al., 2022; Adsul T et al., 2023). The first-order Raman spectra of carbonaceous materials are characterized by two principal peaks: The graphitic (G) band at approximately 1600/cm and the disordered (D) band at around 1350/cm (Kelemen SR and Fang HL, 2001; Schito A et al., 2017). Thermal maturation induces systematic shifts in these peaks, with the G and D bands moving to higher and lower wavenumbers, respectively. These spectral changes allow for the use of Raman band separation (RBS; the wavenumber difference between the G and D peaks) as a robust indicator of thermal maturity (Kelemen SR and Fang HL, 2001; Xu J et al., 2021). Based on the relationship between Raman spectral parameters and OM maturity, previous researchers have proposed various equations to calculate the maturity of sedimentary OM (Liu DH et al., 2013; Zuo ZX et al., 2016; Henry DG et al., 2019; Xiao XM et al., 2020). For example, Liu DH et al. (2013) observed that a series of the Raman spectral parameters (e.g., inter-peak intervals and peak height ratios) correlate well with R_o and established two formulas applicable to coal samples with varying maturities. Wilkins RWT et al. (2014, 2015) developed a method using two regression equations based on several Raman spectral parameters to predict the equivalent R_o values (0.4%–2.5% R_o) of marine shales, demonstrating that this method can correct suppressed R_o values. While Raman spectroscopy provides straightforward indicators for assessing OM maturity, the application of these indicators to calculating the maturity of over-mature source rocks remains unclear.

Sedimentary OM (e.g., kerogen and coals), originating from distinct biogenic sources, undergoes divergent maturation pathways during geological burial. This divergence leads to systematic variations in thermal maturity estimations derived using different analytical methodologies (Kelemen SR and Fang HL, 2001; Zuo ZX et al., 2016; Henry DG et al., 2019; Xu J et al., 2021). For example, Zuo ZX et al. (2016, 2022) reported that Raman spectroscopy-derived equivalent vitrinite reflectance ($R_{mc}R_o$) values are generally consistent with those obtained from T_{max}/R_o measurements. However, they noted that the coefficient of variation (CV) of $R_{mc}R_o$ for shales is higher than that for coals. This difference is considered to be related to not only the structural characteristics of concentrated and dispersed OM but also variations in the thermal evolution history across different regions and strata. Therefore, this study examined differences

in thermal evolution parameters of over-mature shales from the Longmaxi and Qiongzhusi formations and anthracite samples from the Shanxi and Taiyuan formations using Raman spectroscopy, Rock-Eval pyrolysis, and R_o . The purpose is to identify the determinants of the maturity in highly- to over-mature source rocks, revise the existing formulas for calculating the $R_{mc}R_o$ of the source rocks, and calculate the CVs of different source rocks. These will help increase the reliability of the assessment of heterogeneous changes in over-mature OM.

2. Samples and experimental methods

2.1. Sample collection

Before sampling highly- to over-mature hydrocarbon source rocks, this study conducted a systematic review of the distribution and geochemical characteristics of principal source rocks in China (Watson MP et al., 1987; Dai JX et al., 2016, 2021; Bao SJ et al., 2018; Zhai GY et al., 2018; Zou CN et al., 2019). Previous studies have confirmed that the organic-rich marine shales in the Longmaxi and Qiongzhusi formations in southern China and coal-bearing strata deposited during the Carboniferous-Permian transition in the Shanxi and Taiyuan formations in northern China have reached a highly- to over-mature evolutionary stage. Given their regionally extensive distribution and stratigraphic representativeness, these geological units were selected as critical targets for investigating thermal evolution processes in over-mature source rock systems. Multiple source rock samples were selected from coals and shale gas reservoirs in the Qinshui Basin, the Sichuan Basin, and northeastern Yunnan. Shales in the Lower Cambrian Qiongzhusi Formation were collected from the Niejiacun section and a borehole in Kirin District, Qujing, Yunnan, with the collected samples numbered NJC and QL, respectively. Shales in the Lower Silurian Longmaxi Formation were sampled from the Yinjiawan, Liaoyeba, and Guanyinqiao sections in Chongqing, with the acquired samples denoted by YJW, LYB, and GYQ, respectively. Anthracite samples were collected from the No. 15 coal seam of the Upper Carboniferous Taiyuan Formation in the Fenghuangshan Coal Mine and the No. 3 coal seam of the Lower Permian Shanxi Formation in the Sihe Coal Mine, with the collected samples marked by FHS and SH, respectively.

2.2. Experimental methods

2.2.1. Raman spectroscopy

Raman spectroscopy allows for the effective calibration of the thermal evolution processes of source rocks (Roberts S et al., 1995; Henry DG et al., 2019). Prior to the experiments, kerogen was separated from the shale samples, and both the isolated kerogen and coal samples were ground to pass a 200-mesh screen. Laser Raman analyses were carried out following *General specification for Raman Spectrometers* (GB/T 40219–2021) using a Bruker Senterra confocal Raman

microscope. The samples were excited using a 532 nm wavelength laser at an intensity of 5 mW. The integration time for each Raman spectrum was two seconds, with 10 accumulations per measurement. The spectral resolution ranged from 9/cm to 18/cm. Based on the Raman spectroscopy results, the characteristic spectral range of 1000–2000/cm was selected for peak fitting. The D and G peaks were fitted, and the peak separation, peak height, and full width at half maximum (FWHM) were quantitatively calculated.

2.2.2. Rock-Eval pyrolysis

Rock-Eval pyrolysis is widely used to assess the thermal maturities of kerogen and coals (Sykes R and Snowdon LR, 2002; Arysanto A et al., 2022). In this study, a Rock-Eval 6 device was used to quantify generated hydrocarbons, the hydrocarbon generation potential of kerogen (or coals), and the CO₂ content (S₃) produced during thermal cracking. Initially, the samples were held at a constant temperature of 300°C for three minutes to determine the free hydrocarbon content (S₁). The temperature was then increased from 300°C to 650°C at a rate of 25°C/min to measure the content of cracked hydrocarbons (S₂). S₃ represents the amount of CO₂ released from the decomposition of carboxyl groups and other oxygen-containing compounds in kerogen, obtained at temperatures ranging from 300°C to 500°C. All steps followed *Rock Pyrolysis Analysis* (GB/T 18602–2012).

2.2.3. Vitrinite reflectance of the source rocks

R_o serves as a critical parameter for characterizing the thermal maturity of source rocks (Mukhopadhyay PK, 1994; Liu B et al., 2020). Prior to the experiments, the samples were crushed to pass a 60-mesh screen and then embedded in epoxy resin for polishing. Following the guidelines of *Method of Determining Microscopically the Reflectance of Vitrinite in Coal* (GB 6948–1986), the mean random R_o values of seven different samples were determined using a Leica DM4500P polarization microscope and a CRAIC microphotometer. For each sample, at least 50 measurements were conducted, with calibration performed under oil immersion (refractive index N = 1.518 and R_o = 0.00% for the immersion oil).

2.2.4. Micro-thermometric analysis of fluid inclusions

Petrological observations and temperature measurements of fluid inclusions were conducted in accordance with *Test Method for Fluid Inclusion in Sedimentary Basins by Microthermometry* (SY/T 6010–2011). First, the fluid inclusions were placed on a heating-cooling stage and positioned at the center of the field of view. The inclusions were then cooled to –80°C at a rate of 10°C/min using liquid nitrogen. After cooling, the temperature was gradually increased. As the temperature approached 0°C, the heating rate was reduced to approximately 0.2°C/min for precision. Afterward, the heating was accelerated using a resistance wire. As the temperature neared the homogenization temperature, the heating rate was controlled at approximately 1°C/min. Upon reaching homogenization temperature, the

salinity was determined based on the negative correlation between homogenization temperature and solution salinity. The formation temperature at the time of fluid entrapment was then inferred (Evans MA, 1995; Nie HK et al., 2020).

3. Results and discussion

3.1. Organic geochemistry and fluid inclusion characteristics

The organic geochemical characteristics of the seven shale and coal samples were obtained using Rock-Eval pyrolysis and R_o experiments. Shale samples from the Longmaxi and Qiongzhusi formations exhibited TOC contents ranging from 0.50% to 3.0% and from 0.50% to 3.5%, respectively. The kerogen characterization plot revealed that samples from the Longmaxi Formation primarily contained Type I kerogen (i.e., sapropelic kerogen constituents) with great potential for hydrocarbon generation. In contrast, samples from the Qiongzhusi Formation predominantly exhibited types I and II₁ kerogen. Shale samples from the Longmaxi Formation showed maturity ranging from 2.2% R_o to 2.7% R_o, with minor variations observed among different samples (Fig. 1a). This result indicates that these samples had entered the mature to over-mature stage. Shale samples from the Qiongzhusi Formation exhibited a T_{max} exceeding 440°C, indicating that these samples were in the highly- to over-mature stage (Fig. 1b). To ensure the robustness of these results, this study conducted cross-validation against previous studies, yielding high consistency (Chen SB et al., 2014; Hu G et al., 2020; Shang FH et al., 2020).

The anthracite samples exhibited a vitrinite content ranging from 79.3% to 93.4% (average: 82.1%), an inertinite content from 7.2% to 34.6% (average: 17.1%), and a minimal exinite content from 0.6% to 15.7% (average: 0.8%). Type III kerogen was identified in these samples (Fig. 1), consistent with the terrestrial origin of the OM. These samples showed R_o values varying from 3.12% to 3.52% (average: 3.3%), confirming that they were in a highly- to over-mature stage in their thermal evolution.

Fluid inclusions, formed by various geological processes, serve as valuable indicators of the physicochemical conditions prevailing in varying stages of these processes (Goldstein RH, 2001; Randive KR et al., 2014). This study conducted fluid inclusion analysis of shales in the Longmaxi Formation in northeastern Yunnan Province (Fig. 2). As a result, the key parameters of fluid inclusions in the shales were determined, including their types, gas-to-liquid ratios, homogenization temperature, freezing temperature, salinity, and density. These parameters provide critical insights into factors controlling fluid formation, such as fluid temperature, pressure, and fluid sources. Furthermore, they contribute to the reconstruction of the thermal evolution history related to hydrocarbon accumulation, including the timing of hydrocarbon charging and the paleo-temperature conditions during reservoir formation.

Shale samples collected from northeastern Yunnan were black, exhibiting calcite veins, well-developed cleavage, and

grain sizes exceeding 0.3 mm. Fluid inclusions predominantly occurring in the calcite were dominated by saline inclusions, with no hydrocarbon inclusions discovered (Fig. 2a). Among them, single-phase saline inclusions, consisting mainly of pure brine, accounted for about 90% of the total inclusions (Fig. 2a), with sizes ranging from 3 μm to 16 μm . In contrast, two-phase inclusions, mainly comprising brine solution and gas, represented only 10%, with sizes ranging from 2 μm to 6 μm . These inclusions were elliptic, circular, and irregular in shape, showing symbiotic relationships with individual liquid-phase brine inclusions. Among them, eight inclusions exhibited homogenization temperatures ranging from 140°C to 216°C, freezing temperatures from -0.8°C to -1.8°C , and salinity from 1.39% to 3.05% (Fig. 2a).

The fluid inclusions mainly occurring in sparry calcite in

the shale samples were dominated by saline inclusions, with no hydrocarbon inclusions found (Figs. 2b, c). The sparry calcite exhibited grain size ranging from 0.25 mm to 1 mm. The fluid inclusions hosted in the sparry calcite were elliptic, circular, and elongated in shape, unevenly distributed in calcite minerals, with a few being zoned along cracks. Among these fluid inclusions, two-phase inclusions with brine solution accounted for about 30%, with sizes ranging from 5 μm to 52 μm . These inclusions were elliptic, circular, or irregular in shape, distributed in calcite minerals along cracks. They generally showed symbiotic relationships with single-phase brine inclusions. Among them, eight inclusions exhibited homogenization temperatures ranging from 90°C to 128°C, freezing point temperatures from -0.8°C to -4.7°C , and salinity from 1.39% to 7.44% (Figs. 2b, c).

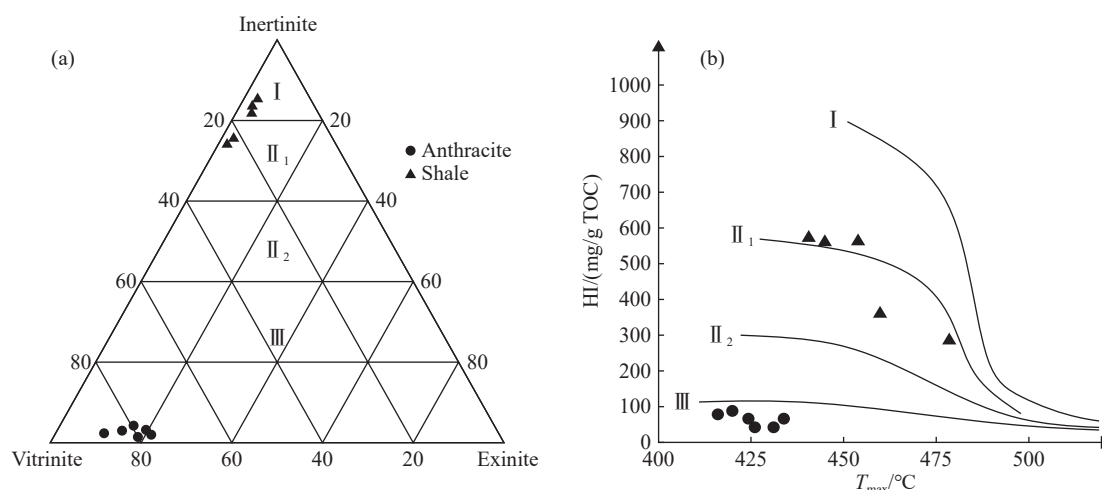


Fig. 1. a–Ternary diagram of organic macerals and b–HI vs. T_{max} plot. I–Type I source rock; II₁–Type II₁ source rock; II₂–Type II₂ source rock; III–Type III source rock.

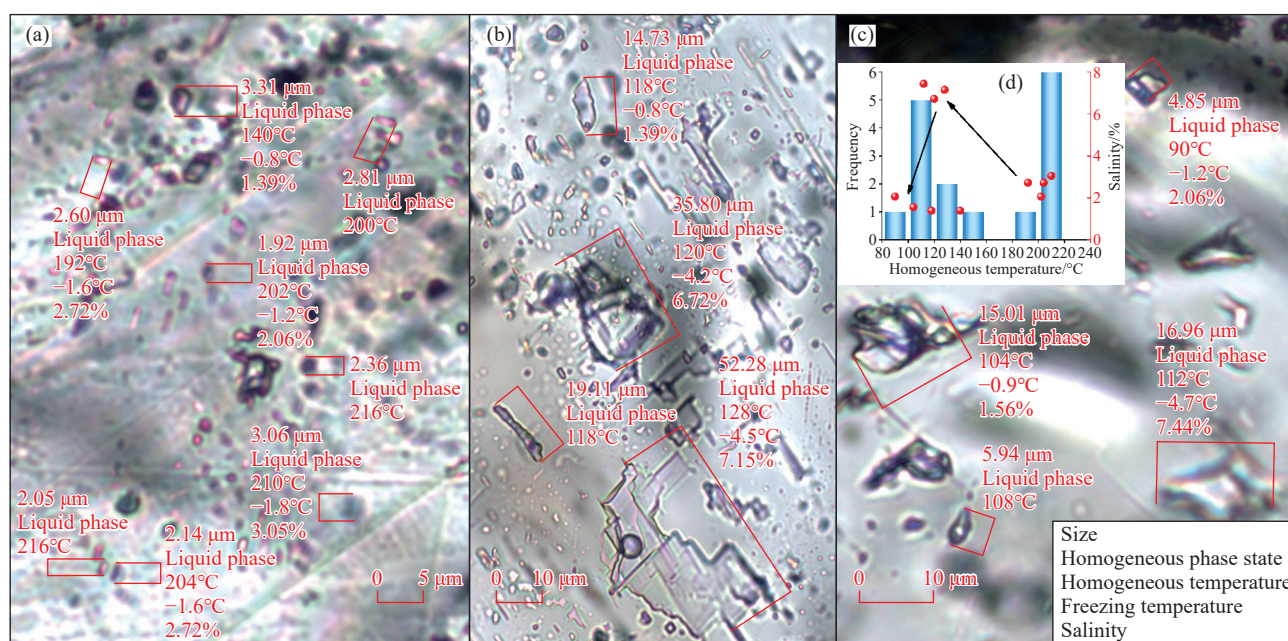


Fig. 2. Micrographs and homogenization temperature histogram of fluid inclusions in shale samples from the Longmaxi Formation. a–c–Fluid inclusions in calcite vein bodies; d–histogram of the homogenization temperature distribution of all fluid inclusions and its relationship with salinity.

Critical constraints on the sealing capacity and thermal evolution history of strata include natural fracture networks, paleo-fluid activity signatures, and geochemical indicators preserved in minerals, which facilitate the reliable assessment of the hydrocarbon preservation conditions of reservoirs (Yang W et al., 2021; Wang DD et al., 2024). The frequency distribution analysis of the fluid inclusions' homogenization temperatures reveals two distinct thermal populations (90°C–150°C and 190°C–220°C; Fig. 2d), indicating two distinct phases of hydrothermal activity of all the samples. A complementary analysis of the calculated salinity data (Fig. 2d) shows a bimodal distribution pattern (1%–4% and 6%–8% NaCl equivalent). The correlation between the salinity ranges and the corresponding temperature populations suggests that gas reservoirs in the Longmaxi Formation underwent a complex evolutionary sequence: poor preservation conditions (weak sealing), improved preservation conditions (effective sealing), and compromised preservation conditions sequentially.

3.2. Thermal maturation history of OM

The temporal evolution of OM maturity can be quantitatively modeled by integrating the geothermal history, paleo-heat flow reconstruction, and chemical kinetics-based hydrocarbon generation simulations. Accordingly, a coupled thermal-hydrocarbon evolution history can be established (Chen SB et al., 2014, 2017; Zhao L et al., 2023). The Qiongzhusi Formation, deposited during the Early Cambrian–Caledonian transition (Figs. 3a, d), experienced significant

thermal perturbations from the Middle-Late Permian Emeishan mantle plume activity. The thermal evolution of this formation occurred in two pathways: (1) Following the peak oil generation stage (i.e., the oil window), hydrocarbons evolved from the wet gas phase into the dry gas phase, with no significant hydrocarbon generation identified beyond these maturation stages, and (2) commencing in the Caledonian, the Qiongzhusi Formation entered the hydrocarbon generation evolution stage. Then, this formation gradually experienced the Hercynian–Yanshanian tectono-thermal episodes while sustaining thermal maturation, followed by continuous hydrocarbon generation until the end of the Cretaceous (e.g., QL and NJC). Moreover, maturity parameter $R_{mc}R_o$, derived from Raman spectra, reveals that the OM in the Qiongzhusi Formation is in the highly- to over-mature stage (Table 1). This finding is consistent with the pyrolysis results (Fig. 1).

The Longmaxi Formation commenced deposition during the Ordovician–Silurian transition, undergoing progressive hydrocarbon generation as the burial depth increased (Figs. 3b, d). Then, the organic maturation of this formation almost stagnated due to the subsequent Hercynian–Indosinian tectono-thermal quiescence. This is followed by the resurgence of thermal evolution during Yanshanian tectonic activation. Diagnostic analyses of fluid inclusions reveal two distinct hydrothermal pulses in the Longmaxi Formation: (1) The Early Cretaceous event (ca. 100 Ma), rendering the maturation process to reach the initial hydrocarbon generation threshold, and (2) the Paleogene thermal perturbation (ca. 50 Ma), and leading to maturation termination under uplift-driven thermal resetting. The thermal profiling reveals that the

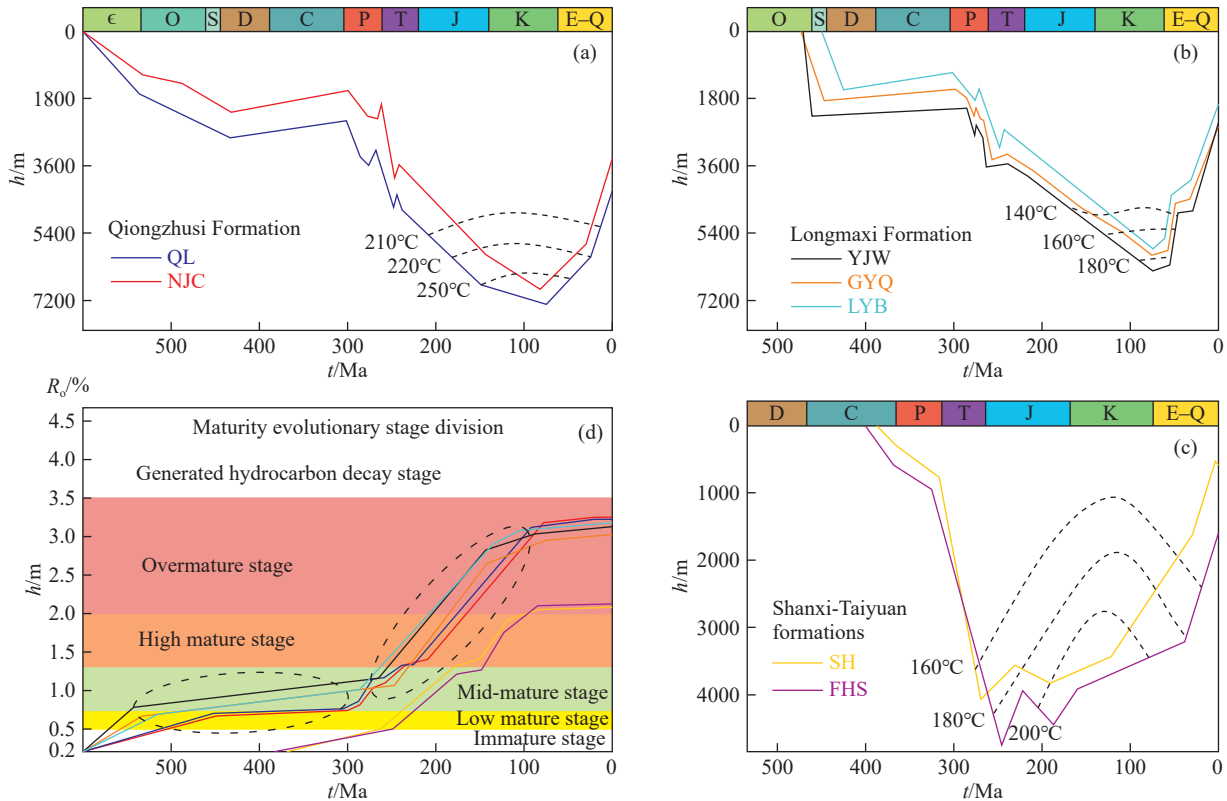


Fig. 3. Burial and thermal histories of the sampling areas.

Table 1. Raman spectral parameters and $R_{mc}R_o$.

Sample No.	Peak D /cm ⁻¹		Peak G /cm ⁻¹		Ratio of peak height $R_{D/G}$	Peak position difference $d(G-D)$	$R_{mc}R_o$ /%
	Position	Height	Position	Height			
FHS	1332.7403	3796.8021	1603.7776	6270.8567	0.6055	271.0373	3.4647
SH	1252.1066	428.62562	1604.4942	1555.8236	0.2755	352.3876	3.0800
NJC	1339.3432	361.57123	1603.6632	395.20666	0.9149	264.3200	3.8255
QL	1336.6175	628.09536	1601.5174	800.24278	0.7849	264.8999	3.6739
GYQ	1348.3763	506.98024	1604.1902	1177.0932	0.4307	255.8139	3.2610
YJW	1332.9564	338.9424	1604.1873	578.66837	0.5857	271.2309	3.4417
LYB	1334.1679	704.56474	1604.6487	1277.1078	0.5517	270.4808	3.4020

Longmaxi Formation features a subdued geothermal regime (peak temperature: < 200°C; effective heating duration: 300 Ma) compared to the Qiongzhusi Formation, which has an intensive thermal history (peak temperature: > 250°C; sustained maturation duration: 500 Ma). The $R_{mc}R_o$ values indicate that the OM in the Longmaxi Formation is in the mature stage (Table 1). This result is consistent with the pyrolysis results (Fig. 1).

The Shanxi–Taiyuan formations were continuously deposited from the Carboniferous through the Late Triassic, with a maximum burial depth of about 5000 m (Figs. 3c, d). Progressive maturation transformed OM in these formations from low to moderate maturity. During the Middle Jurassic evolution of the Qinshui Basin, the thermal maturation rates of anthracite samples from the Shanxi–Taiyuan formations remained subdued under the Indosinian tectono-thermal modulation. The subsequent Yanshanian orogeny, coupled with crustal-scale magmatic perturbations, induced accelerated maturation kinetics, culminating in terminal over-maturation by the end of the Early Cretaceous. Vitrinite reflectance thermometry indicates advanced thermal maturation in the Shanxi–Taiyuan formations, with measured R_o values ranging from 3.12% to 3.52%. This indicates that the anthracite samples from the Shanxi–Taiyuan formations were in the over-mature stage. This result demonstrates the methodologic consistency between thermal maturation signatures and independent pyrolysis indices (Fig. 1).

This study systematically integrated published stratigraphic datasets and multidisciplinary research findings on the burial and thermal evolution histories of the target geological units. This ensures the methodological rigor of the analytical workflow. Comparative analysis reveals that the thermal maturation histories of the target formations are roughly consistent with existing basin models. Nevertheless, notable spatiotemporal differences were identified in evolutionary pathways due to differential tectonic dynamics and lithostratigraphic heterogeneities across discrete structural domains (Xing YS and Luo HL, 1984; Yu K et al., 2018; Chen SB et al., 2021; Qiu NS et al., 2021; Tang XL et al., 2021; Chen C et al., 2022; Cheng M et al., 2024). In this study, the burial and thermal histories reveal that compared to the shale samples, the coal samples featured a reduced burial depth range, subdued geothermal regimes, and truncated thermal durations (Fig. 3). Compared to shale samples from the Qiongzhusi Formation, the anthracite samples exhibited

similar peak thermal temperatures but significantly shorter thermal evolution durations and smaller maximum burial depths (Figs. 3a, d). These findings collectively indicate that the final thermal maturity is primarily controlled by peak temperature. In contrast, the time scale of thermal activation determines the maturation rate. Notably, compared to shale samples from the Longmaxi Formation, the final thermal evolution outcomes of the anthracite samples were primarily determined by the accelerated burial-uplift process under comparable thermochronometry and thermal intensity conditions. Despite rapidly reaching the peak thermal conditions in the accelerated burial-uplift process, the anthracite samples exhibited lower ultimate maturation levels than shale samples from the Longmaxi Formation (Figs. 3b, d). This result demonstrates that truncated thermal evolution duration, attributable to rapid tectonic activity, is insufficient for complete organic structural reorganization. Furthermore, it is noteworthy that early uplift contributed significantly to the lower maturity of the coal samples by inducing premature termination of hydrocarbon generation. A comparative analysis of the Longmaxi and Qiongzhusi formations reveals consistent conclusions that the Qiongzhusi Formation exhibited markedly a retarded chronology of thermal maturation compared to the Longmaxi Formation, as evidenced by the delayed initiation and termination of the maturation process. Therefore, the temporal duration of thermal evolution governs the maturation rates of highly- to over-mature source rocks, and the peak paleo-temperature dictates their ultimate maturity thresholds, with both parameters jointly controlling their thermal evolution trajectories.

3.3. Raman spectra

The Raman spectral analysis involved standard analytical parameters, including the positions of the D and G bands, FWHM, and relative intensity ratios (Fig. 4a). For the characteristic D and G band regions (highlighted) in the raw spectra of all samples (Fig. 4b), precise spectral deconvolution was performed through Gaussian-Lorentzian peak fitting using the PeakFit software, yielding highly robust fitting results ($R^2 \geq 0.9996$). A comparative cross-sectional analysis of the D and G band parameters was conducted (Fig. 4c). The deconvolution procedure involved separating the D and G bands within their characteristic spectral domains (Fig. 4c).

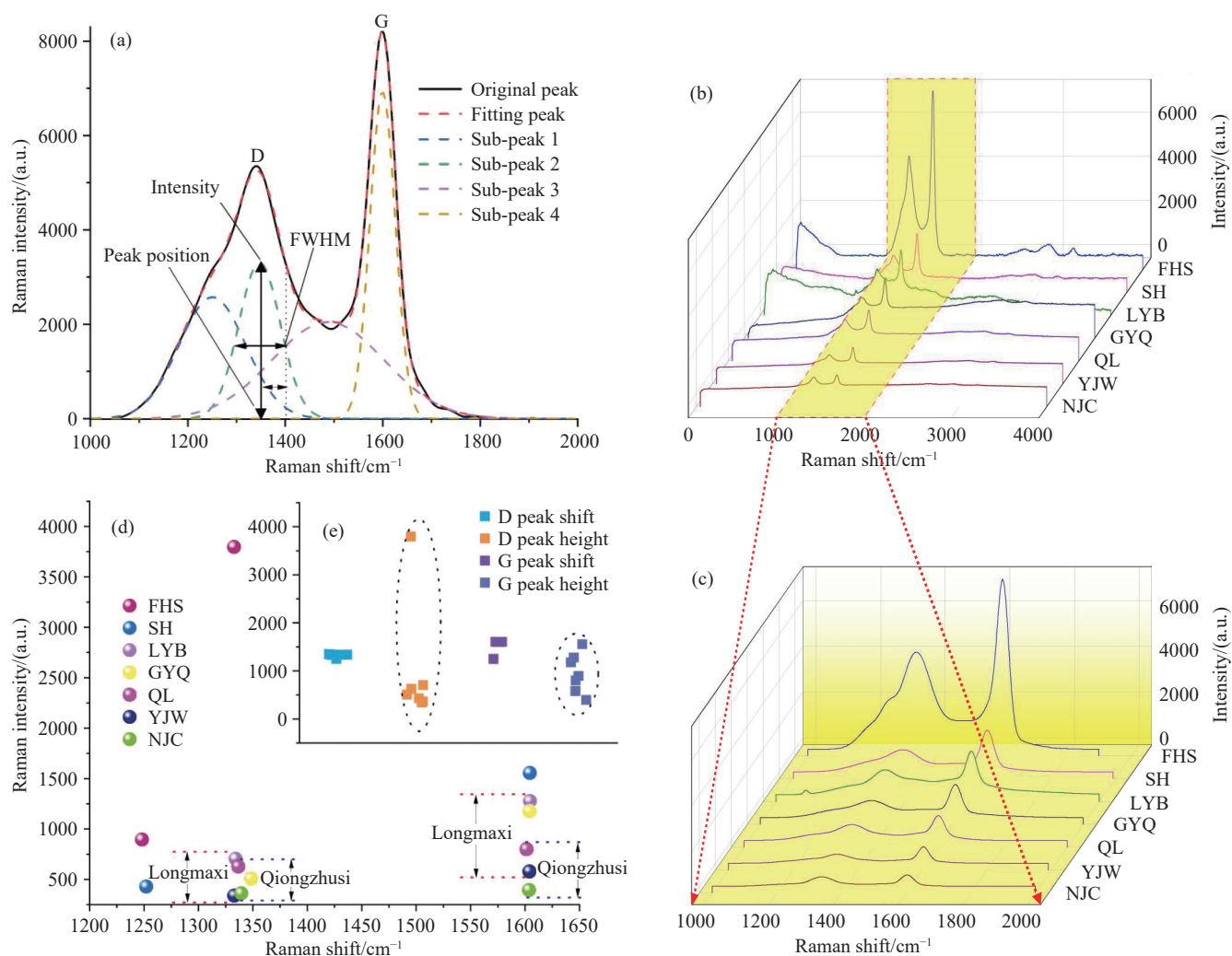


Fig. 4. Raman spectral analysis and schematic diagrams. a–Fitted peak parameters for sample FHS, illustrating the significance of each fitted component; b–raw Raman spectra obtained from tests, with the highlighted area indicating the data segment selected for fitting; c–fitted D and G peaks of all samples; d–distribution of Raman shifts and intensities of the D and G peaks; e–cross-sectional comparison of the parameters of the D and G peaks.

through multi-component fitting using the Voigt function. In this process, each primary band (e.g., the D and G bands in Fig. 4a) was resolved into discrete sub-peaks, typically comprising four components. To minimize interference from adjacent peak residuals, from which signals affect the fitting of neighboring peaks, this study optimized peak deconvolution by strategically reducing the number and spectral overlap of sub-peaks. This enhanced the fitting precision for quantifying the D and G bands.

A comparative analysis of the distributions of D-band peak heights and Raman shifts (Figs. 4d, e) reveals that, except for anthracite sample FHS, the D-band intensities of the remaining samples predominantly fell below 750 arbitrary units (a.u.). All the samples exhibited D-band shifts within a range of 1250–1350/cm and G-band shifts centered around 1600/cm, indicating similar thermal maturation levels across the sample set. This uniformity is further supported by calculated $R_{mc}R_o$ values (Table 1). The D and G band positions of shale samples from the Longmaxi Formation ranged from 338/cm to 704/cm and from 578/cm to 1277/cm, respectively,

exhibiting broader spectral distributions compared to those of shale samples from the Qiongzhusi Formation (D: 361–628/cm; G: 395–800/cm). In combination with thermal maturity analysis, this disparity is interpreted as indicative of different thermal evolution durations in the burial-uplift process. The extended thermal history of the Longmaxi Formation likely enhanced the structural reorganization of OM, resulting in broader Raman spectra. Additionally, it can be observed that the Raman spectral profile of OM progressively evolved into distinct unimodal spectra with increasing thermal maturity, reflecting more orderly arrangements of carbon atoms. Overall, the comparative analysis indicates that compared to the underlying Qiongzhusi Formation, the Longmaxi Formation experienced more rapid burial-exhumation process but was subjected to lower paleo-temperatures, reduced thermal intensity, and a shorter thermal duration. These constrained thermal conditions impeded structural ordering within the organic matrix and the rearrangement of carbon atoms, ultimately resulting in broader Raman spectral peaks.

For source rocks in the highly- to over-mature stage, the RBS tends to progressively narrow rather than widening, with spectral stabilization ultimately arriving (Xiao XM et al., 2020; Hackley PC et al., 2021; Cheng NN et al., 2022). R_o exhibited a negligible correlation with RBS (Fig. 5a), while the intensity ratio between the D and G bands showed a strong positive correlation with the degree of thermal maturation (Fig. 5b). Based on the analysis of the Raman peak height ratios of the coal and shale samples at the highly to over-mature stage, this study modified the Eq. 1 for calculating $R_{mc}R_o$ under such advanced thermal conditions:

$$R_{mc}R_o = 0.85771R_{D/G} - 2.36624 \tag{1}$$

where $R_{mc}R_o$ is the Raman-derived vitrinite reflectance parameter, and $R_{D/G}$ is the peak height ratio of the D band to the G band.

This quantitative model correlates parameters of the D and G bands in Raman spectra with thermal maturity indices, establishing a classification scheme calibrated against standard R_o values. The Raman-derived formulation circumvents the reliance on vitrinite through spectral parameterization, thereby enabling a reliable assessment of thermal evolution in lithologies with low or absent vitrinite content.

3.4. Coefficient of Variation for $R_{mc}R_o$

This study quantified the OM heterogeneity using CV, calculated using the ratio of the standard deviation to the average (Table 2). For source rock samples, CV is indicative

of the differential responses of OM during its thermal evolution. A higher CV value is associated with stronger OM heterogeneity and greater variability in response to geological processes, and vice versa. A comparative analysis of CV values from equivalent stratigraphic units across different sampling locations enables the characterization of behavioral differences in lithologically comparable source rocks under varying geological conditions.

Samples from the Shanxi–Taiyuan, Qiongzhusi, and Longmaxi formations exhibited average CV values of 0.186, 0.212, and 0.208, respectively (Fig. 6). These values demonstrate that samples from the Qiongzhusi Formation showed the highest degree of OM heterogeneity and the greatest variability in its response to geological processes, followed by those from the Longmaxi Formation, which displayed the greatest absolute regional variation in CV values across sampling locations. In contrast, the anthracite samples, sourced exclusively from the Qinshui Basin, showed minimal divergence in response to geological processes, corresponding to their lower average CV. This systematic variability quantitatively confirms the differential responses of OM to geological processes with varying intensities.

Samples from the Shanxi–Taiyuan, Longmaxi, and Qiongzhusi formations exhibited distinct thermal evolution intervals of approximately 50–75 Ma, 280–300 Ma, and 250–270 Ma, respectively, progressing from low to high thermal maturity. The anthracite and shale samples had divergent overall thermal histories. Specifically, the shale samples were characterized by a prolonged stagnant phase (170–250 Ma), followed by a more condensed active maturation interval (50–80 Ma), with the latter consistent with the thermal evolution phase of the anthracite samples. A Raman spectral analysis reveals significantly higher D- and G-band intensities of the anthracite samples compared to the shale samples (Figs. 4d, e). This finding indicates a higher degree of structural ordering of the anthracite samples under continuous thermal regimes, as opposed to the disordered carbon matrices of the intermittently heated shale samples under similar thermal exposure. During the highly- to over-mature stage leading up to the cessation of thermal evolution, the three sample groups followed distinct thermal trajectories. Specifically, the anthracite samples and shale samples from the Longmaxi and Qiongzhusi formations experienced increases in temperature from approximately 180°C to over 200°C, from 120°C to above 180°C, and from 200°C to beyond 250°C, respectively. The thermal evolutionary

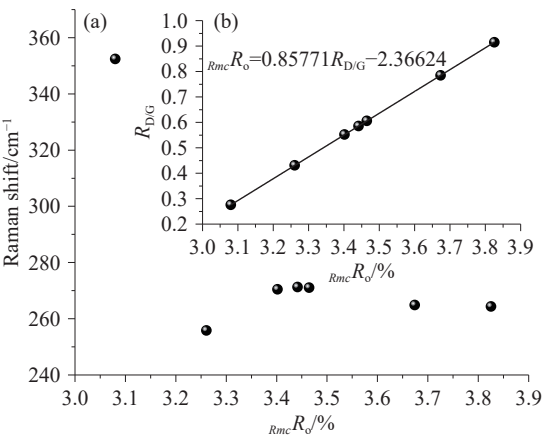


Fig. 5. Diagrams showing the relationships of the Raman parameters with vitrinite reflectivity.

Table 2. Calculated Coefficient of Variation for $R_{mc}R_o$.

Sample No.	Raw data from the five-point measurement method					Standard deviation	Average	CV
FHS	3.4935	3.4121	3.5982	3.5177	3.6590	0.0854	3.5361	0.242
SH	3.6017	3.6431	3.6099	3.5070	3.5644	0.0464	3.5852	0.130
NJC	3.8134	4.0867	4.0074	3.9202	3.9231	0.0919	3.9502	0.233
QL	3.7073	3.5324	3.6995	3.7123	3.5924	0.0731	3.6488	0.200
GYQ	3.4610	3.4619	3.5486	3.5222	3.3938	0.0540	3.4775	0.155
YJW	3.5206	3.6322	3.8641	3.4858	3.6168	0.1323	3.6239	0.365
LYB	3.5654	3.4692	3.4733	3.5333	3.5217	0.0367	3.5126	0.104

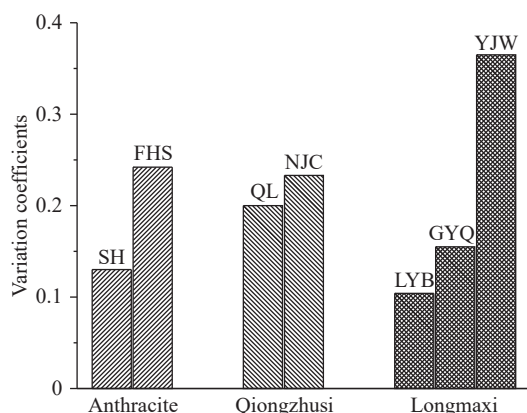


Fig. 6. Bar chart of CV.

processes of these samples comprised two distinct phases: an accelerated maturation stage followed by a maturation stabilization phase (Fig. 3d). During the accelerated phase, the maturities of the anthracite samples and samples from the Longmaxi and Qiongzhusi formations increased significantly by 0.7% R_o , 1.4%–1.6% R_o , and 1.6%–1.7% R_o over durations of approximately 100 Ma, 150 Ma, and 140 Ma, respectively. Notably, these samples exhibited varying maturation rates and thermal gradients in this phase. This quantitative relationship demonstrates that thermal intensity plays a more critical role in determining final maturity levels than heating duration under continuous thermal evolution. This result establishes an increase in temperature as the dominant factor governing hydrocarbon generation thresholds during late-stage thermal maturation. The CV values of samples LYB and GYQ, collected from the Longmaxi Formation, closely aligned with those of samples from the Qiongzhusi Formation. In contrast, sample YJW from the Longmaxi Formation displayed distinct variability. This divergence is attributed to the sampling location of YJW. This sample was collected along the basin margin, where intensified geological stresses—evident in both the burial history patterns of YJW and the CV values of the anthracite samples—exceeded that experienced by LYB and GYQ. Furthermore, a comparative analysis reveals the presence of hierarchical effects of three thermal parameters, i.e., peak heating temperature, duration of sustained thermal intensity, and heating duration, on OM (Section 3.1), with their controlling effects on the final OM maturity decreasing sequentially. Notably, the latter two temporal parameters not only modulate the maturity level of OM but also influence its structural ordering, underscoring their secondary yet important role in the thermal evolution of OM.

4. Conclusions

(i) A comparative analysis of the thermal histories of the anthracite and shale samples reveals that the heating duration governs the thermal maturation rate of source rocks, with the maximum temperature attained acting as the predominant determinant in their ultimate maturity thresholds.

(ii) An integrated analysis of the samples reveals the presence of hierarchical influence of peak heating

temperature, duration of sustained thermal intensity, and effective maturation duration on OM maturity, with their controlling effects decreasing sequentially. This result demonstrates that temperature produces a predominant effect on the final maturation outcomes. In contrast, the heating duration and thermal continuity modulate the structural ordering of OM, playing a secondary yet notable role in its molecular organization.

(iii) Compared to the Qiongzhusi Formation, the Longmaxi Formation displays broader ranges of the D and G peak intensities in the Raman spectra. This difference is primarily attributable to two factors, as revealed by an integrated analysis of CV, burial history, and thermal evolution. First, the Longmaxi Formation was subjected to reduced thermal exposure (e.g., lower heating temperatures and a shorter heating duration), which led to less ordered OM relative to the Qiongzhusi Formation. Second, intrinsic organic heterogeneity caused differential responses of OM to geological processes, ultimately inducing variations in its maturity.

(iv) By correlating thermal evolution histories with Raman peak height ratios, this study establishes a fine-scale maturity quantification model for highly- to over-mature source rocks. This model demonstrates high applicability in the assessment of unconventional hydrocarbons.

CRedit authorship contribution statement

Fu-hua Shang and Xiao-peng Sun designed the model and computational framework and analyzed the data. Fu-hua Shang and Shu-wei Ma carried out the implementation. Fu-hua Shang performed the calculations. Fu-hua Shang and Xiao-peng Sun wrote the manuscript with input from all authors. Yu-tong Pang and Guan-qun Zhou conceived the study and were in charge of overall direction and planning.

Declaration of competing interest

The authors declare no conflicts of interest.

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