



Assessing the thermal maturity of black shales using vitrinite reflectance: Insights from Devonian black shales in the eastern United States

Bei Liu^a, Juan Teng^{b,c,*}, Maria Mastalerz^c, Juergen Schieber^a

^a Department of Earth and Atmospheric Sciences, Indiana University, Bloomington, IN 47405, USA

^b College of Energy, Chengdu University of Technology, Chengdu, Sichuan 610059, China

^c Indiana Geological and Water Survey, Indiana University, Bloomington, IN 47405-2208, USA



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ABSTRACT

Thermal maturity of source rocks indicated by vitrinite reflectance (R_o) is an important and reliable parameter to determine the petroleum potential of sedimentary basins. Originally, R_o was used to characterize the degree of coalification of coals and thus works well for coal seams, but it has limitations when used to determine the thermal maturity of black shale successions. A suite of six coal-shale pairs from the Upper Devonian black shale formations in the eastern United States (New Albany Shale, Ohio Shale, Dunkirk Shale, and Rhinestreet Shale) was selected to study the applicability of R_o in assessing the thermal maturity of black shale successions. The results show that vitrinite in the studied coal samples is dominated by collotelinite, whereas vitrinite in black shales occurs as small dispersed particles ($\sim 5 \mu\text{m}$) in the mineral matrix. When comparing the size and morphology of dispersed vitrinite particles in shales and collotelinite fragments in coals, vitrinite in shales and adjacent coals should have the same origin. The measured mean random R_o of vitrinite in coals ranges from 0.51–0.68%, and is 0.04–0.11% (average 0.07%) lower than that of dispersed vitrinite particles in enclosing shales. This observation contrasts with previously reported R_o suppression in liptinite-rich black shales. A high standard deviation of R_o measurements indicates a highly heterogeneous nature of vitrinite in black shales.

Three mechanisms can be envisioned to contribute to the higher R_o of dispersed vitrinite particles in black shales relative to enclosed coals. First, small vitrinite particles may become more oxidized because small particles are more frequently suspended during transport to the site of deposition and reworking of bottom sediments after deposition than large pieces of driftwood that will turn into enclosed coal lenses during burial diagenesis. Second, misidentification of zooclast (e.g., chitinozoan) fragments as vitrinite due to loss of diagnostic morphology. Third, various macerals in the vitrinite group (e.g., corpogelinite and collotelinite) may have had different original reflectance values. Based on the petrographic characteristics of vitrinite and vitrinite-like particles, caution should be applied when using the R_o values of dispersed vitrinite and vitrinite-like particles in black shales as indicators of thermal maturity.

1. Introduction

Vitrinite reflectance (R_o) is the most widely used proxy to indicate the thermal maturity of sedimentary organic matter (OM) in post-Silurian sediments (Buchardt and Lewan, 1990; Mukhopadhyay, 1994; Taylor et al., 1998). The reflectance of vitrinite increases with increasing thermal maturity (i.e., temperature) due to increasing degree of aromatization and condensation of the molecular structure that results in elevated ordered stacking of aromatic units (McCartney and Teichmüller, 1972). R_o has been successfully applied to reconstruct the thermal history of sedimentary basins because of this irreversible chemical reactions (e.g., Hood et al., 1975; Tissot and Welte, 1984; Tissot

et al., 1987; Mukhopadhyay, 1994), and is one of the most important parameters in source rock evaluation and shale oil/gas exploration (Tissot and Welte, 1984; Wang and Gale, 2009; Jarvie, 2012a, 2012b). Because of the scarcity and heterogeneity of vitrinite in black shales and pre-Devonian rocks, however, R_o at times fails to serve as an effective thermal maturity indicator. Therefore, documenting R_o and its equivalents of black shales accurately and precisely is of great significance for the evaluation of both conventional and unconventional petroleum systems.

Vitrinite comprises multiple macerals that are derived from the gelification of ligno-cellulosic tissues of terrestrial higher plants (Stach et al., 1982; ICCP, 1998; Taylor et al., 1998). The reflectance of one of

* Corresponding author at: College of Energy, Chengdu University of Technology, Chengdu, Sichuan 610059, China.

E-mail address: Tengjuancugb@outlook.com (J. Teng).

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these macerals (collotelinite) is generally measured and documented as the R_o of samples (ICCP, 1998). In practice, any terrestrial OM in black shales that has a reflectance higher than the liptinite group and lower than the inertinite group is likely vitrinite, and its reflectance can be measured. When vitrinite particles are very small, it is difficult to distinguish vitrinite from vitrinite-like particles (Buchardt and Lewan, 1990; Petersen et al., 2013) and solid bitumen (Hackley et al., 2015; Mählmann and Le Bayon, 2016; Wei et al., 2016; Hackley and Lewan, 2018).

Vitrinite-like particles are OM particles that resemble vitrinite in carbonaceous rocks, and their reflectance has been used to assess thermal maturity (Buchardt and Lewan, 1990; Schleicher et al., 1998; Xiao et al., 2000; Petersen et al., 2013; Reyes et al., 2018), particularly in Lower Paleozoic rocks (Buchardt and Lewan, 1990; Schleicher et al., 1998; Xiao et al., 2000; Petersen et al., 2013; Reyes et al., 2018) that lack organic debris from land-derived vascular plants (Kenrick and Crane, 1997). Petersen et al. (2013) measured the reflectance of zooclast and vitrinite-like particles in Middle Cambrian to Upper Silurian shales from central and southern Sweden and the Danish island of Bornholm (Baltic Sea), and found that the reflectance distribution of graptolite and vitrinite-like particles is almost identical. Based on this observation, they interpreted the vitrinite-like particles in these Lower Paleozoic shales to be graptolite fragments that lacked diagnostic morphological features.

One of the problems that is commonly mentioned in connection with R_o values is the so-called R_o suppression, a common phenomenon in black shales that refers to situations where the R_o is lower than the value predicted from the R_o -depth profile (Price and Baker, 1985; Wenger and Baker, 1987; Raymond and Murchison, 1991; Goodarzi et al., 1993; Lo, 1993). Multiple mechanisms have been proposed to explain R_o suppression in black shales (McTavish, 1978, 1998; Hutton and Cook, 1980; Price and Baker, 1985; Wenger and Baker, 1987; Raymond and Murchison, 1991; Hao and Chen, 1992; Goodarzi et al., 1993; Lewan, 1993; Hao et al., 1995, 2007; Carr, 2000a, 2000b; Ujjié et al., 2004; Barker et al., 2007; Hackley and Lewan, 2018; Peters et al., 2018), such as (1) a free radical reaction mechanism by which free radicals generated from bitumen and associated volatile products contribute to termination reactions and slow the aromatization process of co-occurring vitrinite (Peters et al., 2018); (2) misidentification of solid bitumen for vitrinite (Hackley and Lewan, 2018); (3) impregnation of vitrinite with liptinite macerals, bitumen, or aliphatic compounds (Jones and Edison, 1978; Hutton and Cook, 1980; Kalkreuth, 1982; Raymond and Murchison, 1991; Petersen and Vosgerau, 1999; Carr, 2000a); (4) diagenetic enrichment of hydrogen in vitrinite (Price and Baker, 1985; Wenger and Baker, 1987; Hao and Chen, 1992; Carr, 2000a); and (5) overpressured conditions that prevent forward reactions of vitrinite maturation (McTavish, 1978, 1998; Hao et al., 1995, 2007; Carr, 2000a).

In this study, we compared the R_o of dispersed vitrinite particles in black shales with that of vitrinite from associated coals (compressed tree trunks or branches) for a suite of six coal-shale pairs from Devonian black shales in the eastern United States. Because the coal is contained within shale samples, coal and shale had the same thermal history and theoretically the R_o values of coal and vitrinite in enclosing shales should be the same. We observed distinct differences in reflectance between vitrinite in coals and enclosing shales and discussed possible reasons for these differences and implications for thermal maturity assessments of black shale successions.

2. Samples and methods

Six pairs of black shales and enclosed coal lenses were collected from the eastern United States (Fig. 1). All the black shales are from Upper Devonian formations, including the New Albany Shale of the Illinois Basin, and the Ohio Shale, Dunkirk Shale, and Rhinestreet Shale of the Appalachian Basin. The Illinois Basin and the Appalachian Basin

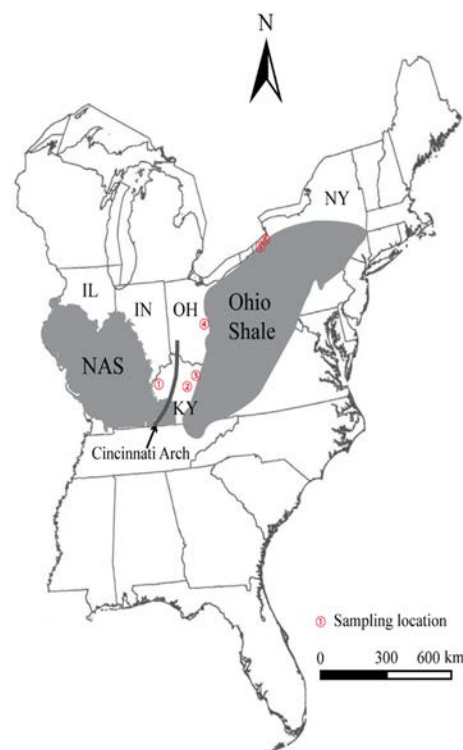


Fig. 1. Map showing the locations of sampling sites and the extent of the New Albany Shale (Illinois Basin) and Ohio Shale (Appalachian Basin). Modified from McFarlan (1943), Curtis (2002), and Mastalerz et al. (2013). NAS = New Albany Shale. ① = New Albany Shale from Shepherdsville, KY; ② = Ohio Shale from Irvine, KY; ③ = Ohio Shale from Morehead, KY; ④ = Ohio Shale from Columbus, OH; ⑤ = Dunkirk Shale from Dunkirk, NY; ⑥ = Rhinestreet Shale from Derby, NY.



Fig. 2. Photo of a thin coal layer embedded within black shales. New Albany Shale from Shepherdsville, Kentucky, USA.

are separated by the Cincinnati Arch (Buschbach and Kolata, 1990) (Fig. 1). The coal lenses embedded within black shales are about 1–2 mm thick and 10–20 cm long in outcrop (Fig. 2). Coal samples were hand-picked from the rock surface, and the directly associated shales were collected at the same time. Coal and shale samples were sealed immediately with Ziplock bags to prevent contamination.

Coal and shale samples were crushed to rock chips of < 1 mm and made into whole-rock petrographic pellets following standard coal petrography procedures (ICCP, 1963). The mean random R_o values of coal and shale samples were measured using a Zeiss Photoscope III microscope with more than 100 measurements per sample on coal

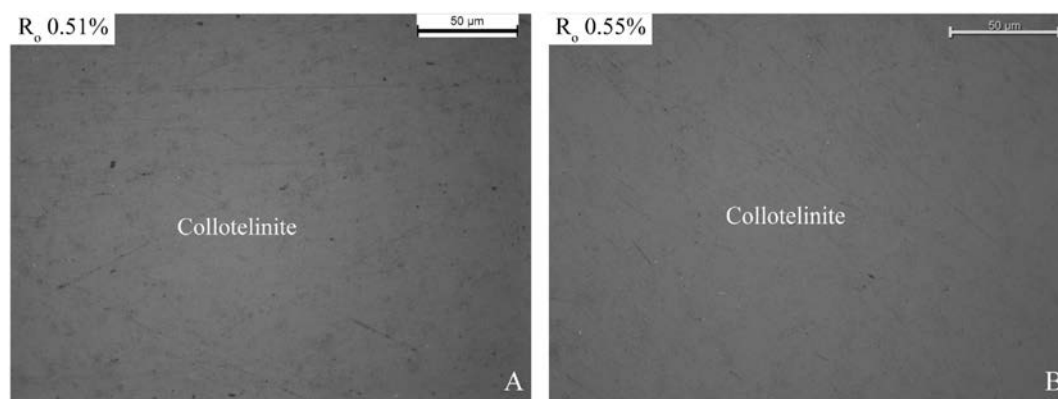


Fig. 3. Photomicrographs of collotelinite in coal samples in reflected white light and oil immersion. (A) Coal sample in New Albany Shale from Shepherdsville, KY. (B) Coal sample in the Ohio Shale from Morehead, KY.

samples and more than 25 measurements per sample on shale samples. A reflected-light microscope (Leica DM2500 P) was used to document the organic petrographic characteristics of macerals in coals and shales in reflected white light and oil immersion as well as under blue light.

3. Results

Organic matter in the coal portion of these samples is dominated by collotelinite of the vitrinite maceral group (Fig. 3). Cellular structures can still be observed in places (Fig. 4). No inertinite or liptinite group macerals were observed.

Primary OM in black shales that enclosed the coal lenses consists of amorphous organic matter, alginite, liptodetrinite, vitrinite, and inertinite (Fig. 5), common primary macerals in Devonian black shales of the eastern United States (Robl et al., 1992; Mastalerz et al., 2012b, 2013; Hackley and Cardott, 2016; Hackley et al., 2017, 2018; Liu et al., 2017, 2019a, 2019b, 2020; Teng et al., 2020). Vitrinite occurs as small dispersed particles (~5 μm) in the mineral matrix, and is similar to the broken vitrinite fragments in coals in size and morphology (Fig. 6), suggesting that the dispersed vitrinite particles in the black shale matrix have the same source as the woody materials of the enclosed coal lenses. Alginite derived from *Tasmanites* cysts in the New Albany Shale and Ohio Shale samples shows greenish-yellow fluorescence under blue light irradiation and shows yellowish fluorescence in the Dunkirk and Rhinestreet Shale samples (Fig. 7). Solid bitumen is very rare in the New Albany Shale and Ohio Shale samples, but common in the Dunkirk and Rhinestreet Shale samples (Fig. 8) because the latter two samples have higher thermal maturities and some amorphous organic matter has been converted into hydrocarbons and solid bitumen (Hackley et al., 2018; Liu et al., 2019a).

The measured mean random R_o of vitrinite in coals ranges from 0.51–0.68%, whereas the R_o of vitrinite in enclosing shales ranges from 0.55–0.73% (Fig. 9; Table 1). Thus the R_o of vitrinite in coals is 0.04–0.11% (average 0.07%) lower than the R_o of dispersed vitrinite particles in enclosing shales. The standard deviation of R_o measurements of coals varies from 0.01–0.02, whereas the standard deviation of R_o measurements of dispersed vitrinite particles in shales ranges from 0.06–0.07, indicating that vitrinite in coals is more homogeneous than that in surrounding shales.

4. Discussion

Statistically, more than 70% of the R_o values of small dispersed vitrinite particles in shales are higher than the mean R_o of enclosed coals (Fig. 10). This can be explained by the oxidation of small dispersed vitrinite precursor particles during transport to the site of deposition and reworking of bottom sediments after deposition, because small particles can easily be suspended, compared to large driftwood pieces that will turn into enclosed coal lenses embedded within black shales during burial diagenesis. Some vitrinite particles were oxidized to such a degree that they have much higher reflectance than others, making them almost indistinguishable from semifusinite (Fig. 11). Previous studies have reported that oxidation of vitrinite can cause higher R_o (Goodarzi and Murchison, 1973; Castaño and Sparks, 1974; Buiskool Toxopeus, 1983). Wenger and Baker (1987) reported that gray shales deposited in oxic environments tend to have hydrogen-poor vitrinite that shows a higher R_o than that of nearby coals. Similarly, Hao and Chen (1992) reported that vitrinite formed in oxic environments has a lower hydrogen and a higher R_o than that formed in anoxic environments. Because dispersed vitrinite particles in black shales that are

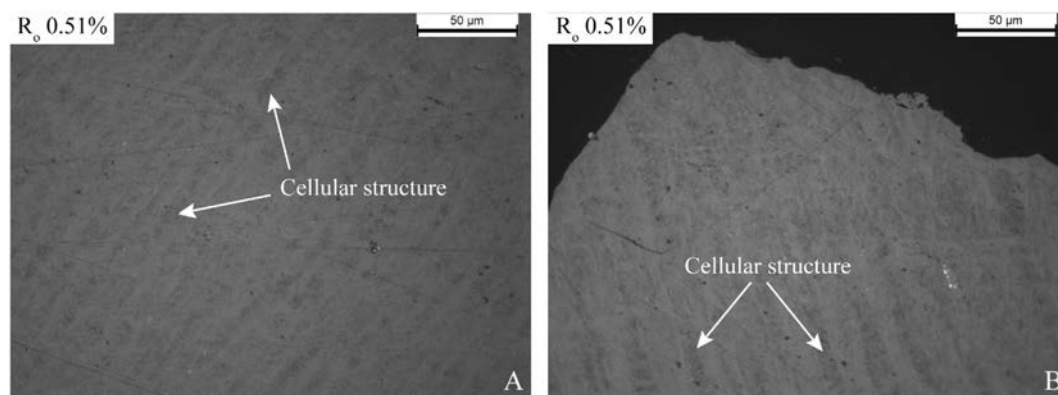


Fig. 4. Photomicrographs of vitrinite with preserved cellular structures in coal samples in reflected white light and oil immersion. Coal sample in the New Albany Shale from Shepherdsville, KY.

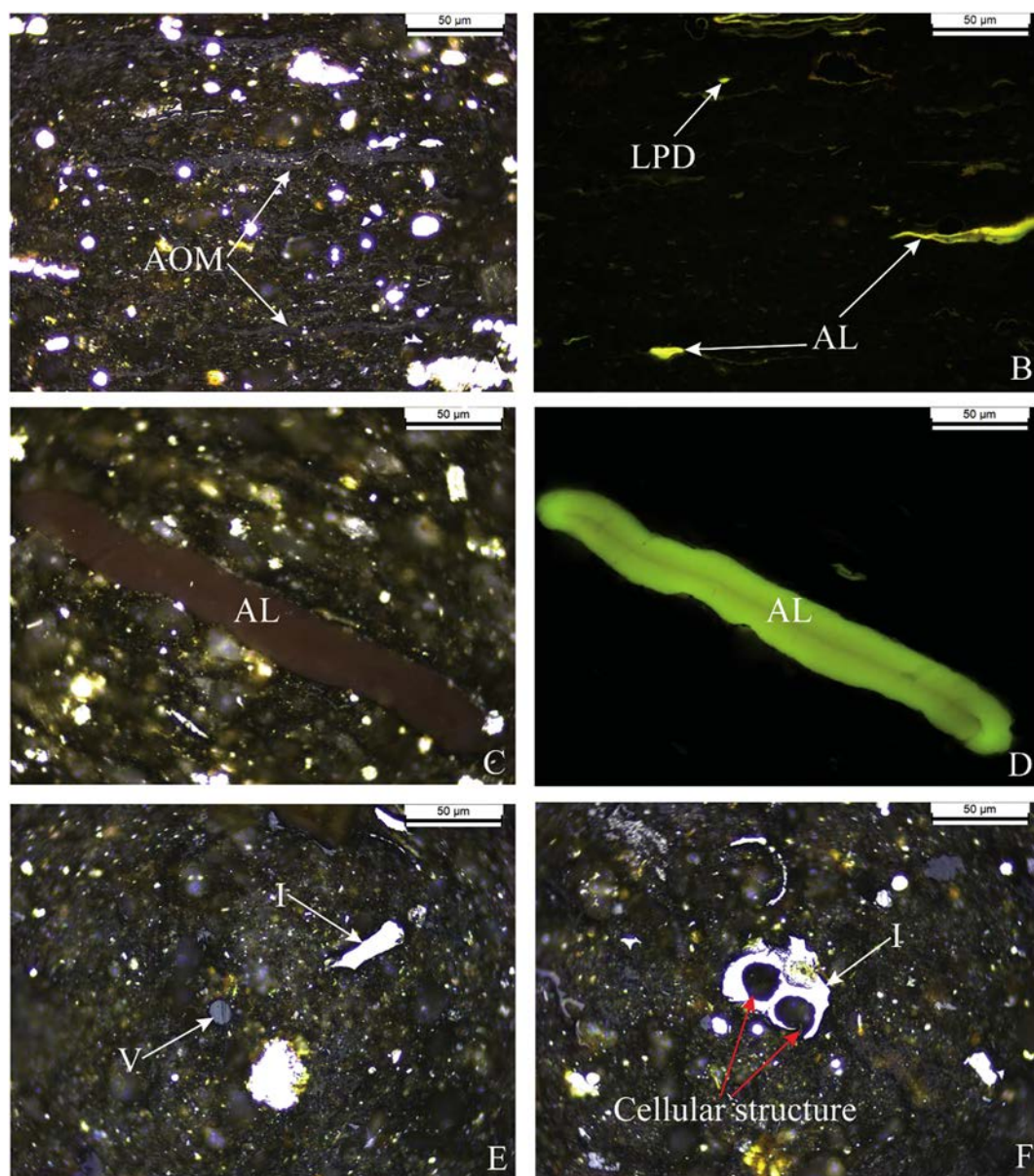


Fig. 5. Photomicrographs of primary macerals in black shales in reflected white light and oil immersion (A, C, E, F) and in fluorescence mode (B, D). Panels B and D show the same field of view as panels A and C in fluorescence mode, respectively. Note that the cellular pores in inertinite in panel F are filled with mineral matter, probably diagenetic silica. New Albany Shale sample from Shepherdsville, KY. AOM = amorphous organic matter; AL = alginite; LPD = liptodetrinite; V = vitrinite; I = inertinite.

used to measure reflectance are typically very small (Mastalerz et al., 2018), caution should be applied when measuring the R_o of small dispersed vitrinite particles in shales. Researchers should try to include as many measurements as possible (25 at least) to get an average value, as recommended by ASTM (2014) and Barker and Pawlewicz (1993).

The reflectance of zooclasts such as graptolites (Bustin et al., 1989; Goodarzi and Norford, 1985, 1987, 1989; Goodarzi et al., 1992; Petersen et al., 2013; Luo et al., 2017, 2018, 2020; Wang et al., 2019) and chitinozoans (Goodarzi, 1985; Bertrand and Héroux, 1987; Bertrand, 1990; Tricker et al., 1992; Reyes et al., 2018) also increases with progressive heating and has been successfully used to determine the thermal maturity of vitrinite-poor source rocks. Graptolite fragments without any recognizable morphology can resemble vitrinite particles (Petersen et al., 2013; Cardott and Curtis, 2018) and could be mistaken for vitrinite. For example, the vitrinite-like particles (interpreted as graptolite fragments) in the Middle Cambrian to Lower Ordovician Alum Shale (Petersen et al., 2013; their Fig. 8C1) and

nongranular graptolite fragments in the Upper Ordovician Wufeng Formation and the Lower Silurian Longmaxi Formation shales (Luo et al., 2018; their Fig. 5) are almost identical (in morphology) to some of the observed dispersed vitrinite particles in black shales in this study (Figs. 5E, 6B). However, graptolites are very rare in late Devonian and at best could make a very minimal contribution to the population of vitrinite-like particles in the studied shale samples. In contrast, chitinozoans are common, although not abundant, in the studied Upper Devonian black shales (Fig. 12). Because the reflectance of graptolites and chitinozoans is generally higher than that of vitrinite subjected to the same thermal history (Tricker et al., 1992; Petersen et al., 2013), misidentification of fragments of chitinozoan derived zooclasts as dispersed vitrinite particles could possibly cause the higher R_o of shales relative to enclosed coals (Fig. 9; Table 1).

Vitrinite in the studied shale samples appears highly heterogeneous, as suggested by the high standard deviation of R_o (Table 1). Some vitrinite particles have sharp edges, suggesting that they could be

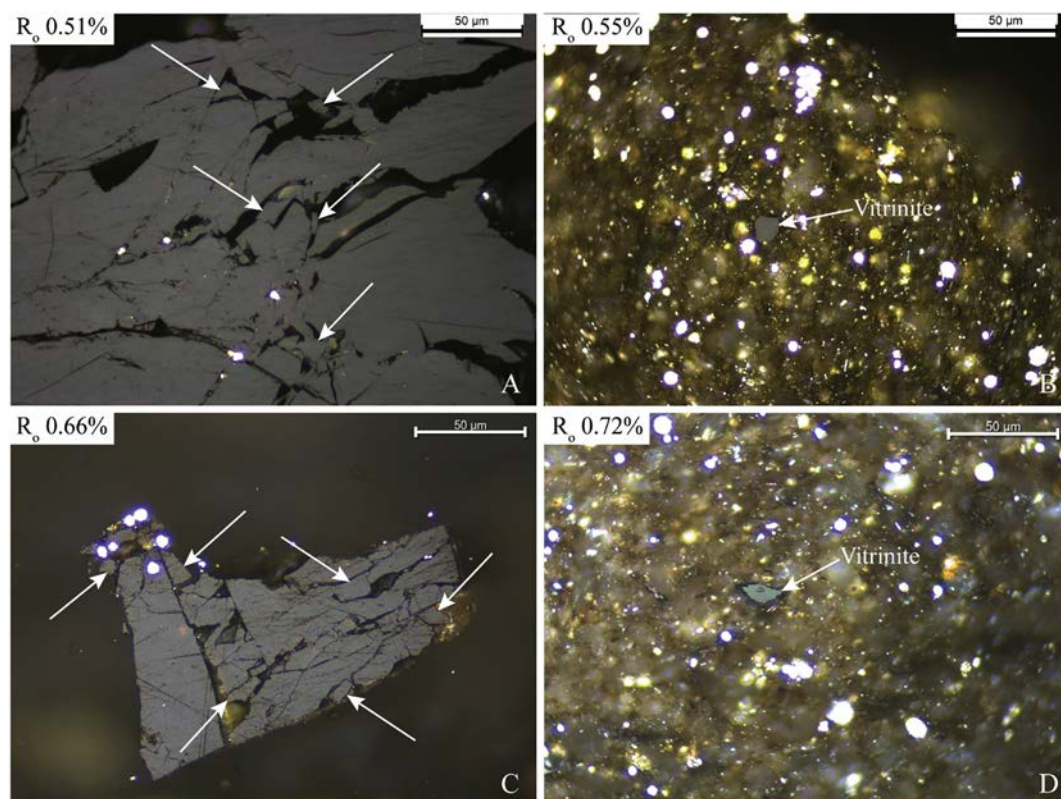


Fig. 6. Photomicrographs of vitrinite fragments (white arrows) in coals (A, C) and dispersed vitrinite particles in shales (B, D) that enclosed the coal samples in reflected white light and oil immersion. (A – B) Coal and shale samples from the New Albany Shale from Shepherdsville, KY. (C – D) Coal and shale samples from the Dunkirk Shale from Dunkirk, NY.

fragments of collotelinite, whereas others are rounded and could possibly be corpogelinite (Fig. 13). Corpogelinite commonly occurs as discrete bodies that fill cellular pores of plant tissues (ICCP, 1998), and looks very similar to the rounded vitrinite particles observed in shales in this study (Fig. 13A). Corpogelinite typically has a higher and wider range of R_o measurements than co-occurring collotelinite (ICCP, 1998), and therefore averaging R_o measurements from collotelinite and corpogelinite particles in black shales may be another reason why the R_o values of shales are higher than those of enclosed coals.

Previous studies reported that the R_o of black shales is often lower than that of interbedded coals (Wenger and Baker, 1987; Raymond and Murchison, 1991; Goodarzi et al., 1993), which contrasts with the findings from this study. Impregnation of bitumen and aliphatic compounds within vitrinite was previously proposed to explain R_o suppression in black shales, because liptinite macerals in black shales are

often oil-prone. For example, Hutton and Cook (1980) reported an inverse relationship between mean maximum R_o and alginite content in the Joadja oil shale and interbedded coals. However, there is some evidence against the influence of bitumen saturation on R_o suppression. Specifically, Barker et al. (2007) compared the R_o values of black shale (including the New Albany Shale) and coal samples before and after organic solvent extraction and found that the R_o values were almost identical. Indeed, it is difficult for large bitumen molecules to enter vitrinite particles (surface coating is possible) in black shales that do not show detectable pores under SEM (Liu et al., 2017). However, we do not exclude the possibility that light hydrocarbons impregnate vitrinite particles initially and solidify later through thermal or microbial degradation, because vitrinite hosts large amounts of micropores (< 2 nm) and small mesopores (2–50 nm) (Rouquerol et al., 1994) detected by low-pressure CO_2 and N_2 adsorption analysis (Mastalerz

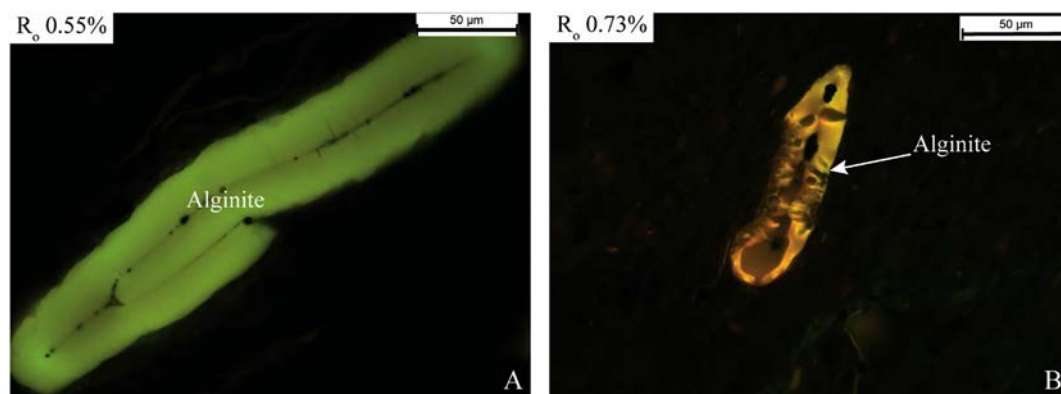


Fig. 7. Photomicrographs of alginite derived from *Tasmanites* cysts in the New Albany Shale (A) and the Rhinestreet Shale (B) in fluorescence mode.

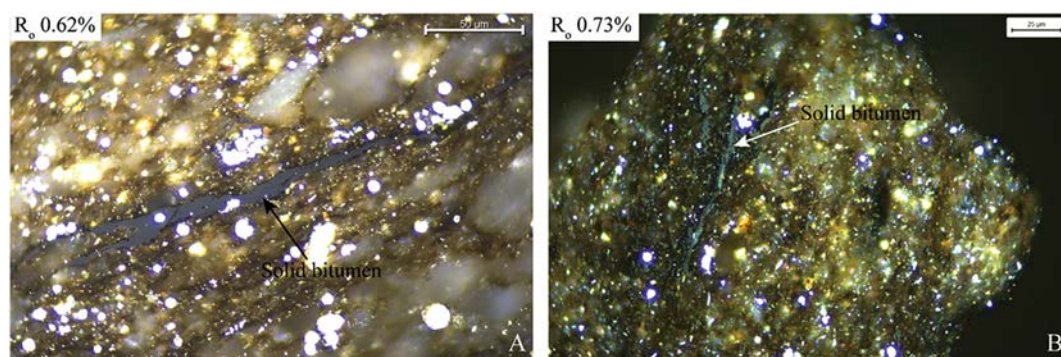


Fig. 8. Photomicrographs of solid bitumen in the Ohio Shale from Columbus (A) and the Rhinestreet Shale (B) in reflected white light and oil immersion.

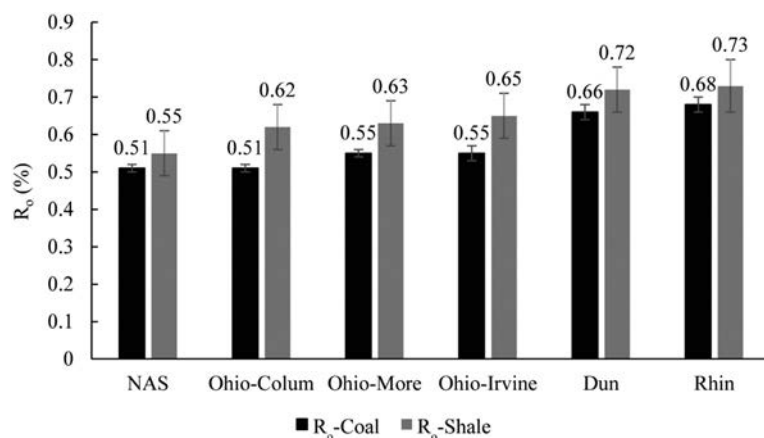


Fig. 9. Histogram of the mean random R_o of shales and enclosed coals. NAS = New Albany Shale from Shepherdsville, KY; Ohio-Colum = Ohio Shale from Columbus, OH; Ohio-More = Ohio Shale from Morehead, KY; Ohio-Irvine = Ohio Shale from Irvine, KY; Dun = Dunkirk Shale from Dunkirk, NY; Rhin = Rhinestreet Shale from Derby, NY (Table 1). Error bars show standard deviation.

Table 1

Measured vitrinite reflectance (R_o), number of measurements, standard deviation, and alginite fluorescence of the studied coal and shale samples.

Sample	Location	Formation	Lithology	R_o	N	SD	Alginite fluorescence
NAS-1	Shepherdsville, KY	New Albany Shale	Coal	0.51	203	0.01	
NAS-2			Shale	0.55	94	0.06	Greenish yellow
Ohio-1	Columbus, OH	Ohio Shale	Coal	0.51	122	0.01	
Ohio-2			Shale	0.62	64	0.06	Greenish yellow
Ohio-3	Morehead, KY	Ohio Shale	Coal	0.55	131	0.01	
Ohio-4			Shale	0.63	96	0.06	Greenish yellow
Ohio-5	Irvine, KY	Ohio Shale	Coal	0.55	123	0.02	
Ohio-6			Shale	0.65	114	0.06	Greenish yellow
Dun-1	Dunkirk, NY	Dunkirk Shale	Coal	0.66	143	0.02	
Dun-2			Shale	0.72	34	0.06	Yellow to orange yellow
Rhin-1	Derby, NY	Rhinestreet Shale	Coal	0.68	154	0.02	
Rhin-2			Shale	0.73	91	0.07	Yellow to orange yellow

N = number of measurements; SD = standard deviation.

et al., 2012a; Teng et al., 2017; Liu et al., 2018). Therefore, the impregnation of vitrinite by bitumen could potentially affect the R_o measurements of black shales, however, its influence should be negligible.

Peters et al. (2018) studied R_o suppression through hydrous pyrolysis experiments on artificial rocks composed of vitrinite-rich coal and varying contents of liptinite-rich kerogen and concluded that R_o suppression occurs in vitrinite mixed with liptinite macerals because free radicals generated from bitumen and associated volatile products contribute to termination reactions that dampen the aromatization process of co-occurring vitrinite. In this study, the coal lenses are about 1–2 mm thick and are in direct contact with enclosing black shales. Any chemical reaction that influences the maturation of vitrinite should therefore have the same effect on vitrinite in coals and shales alike. The free radical reaction mechanism is therefore unlikely to have caused the difference in R_o of shales and enclosed coals observed in this study.

Misidentification of solid bitumen for vitrinite can influence the R_o measurements of shales as well (Wei et al., 2016; Hackley and Lewan, 2018) because solid bitumen transformed from oil-prone macerals has a lower reflectance than co-occurring vitrinite below R_o 1.0% in black shales (Hackley and Lewan, 2018; Liu et al., 2019a). Hackley and Lewan (2018) conducted artificial maturation of coals and shales and found that the reflectance of solid bitumen is always lower than that of vitrinite subjected to the same temperature below R_o 1.6%. Based on a suppressed R_o trend (Lewan, 1993) and solid bitumen reflectance trend, they suggested that mistaking solid bitumen for vitrinite could explain the commonly reported R_o suppression in upper Paleozoic marine shales of early to mid-oil window maturity. Dispersed vitrinite particles in shales examined in this study are similar to vitrinite fragments in associated coals (Fig. 6), and are unlikely to be solid bitumen that fills the void spaces between mineral grains and fractures (Mastalerz et al., 2018). In addition, solid bitumen is very rare in the studied New Albany

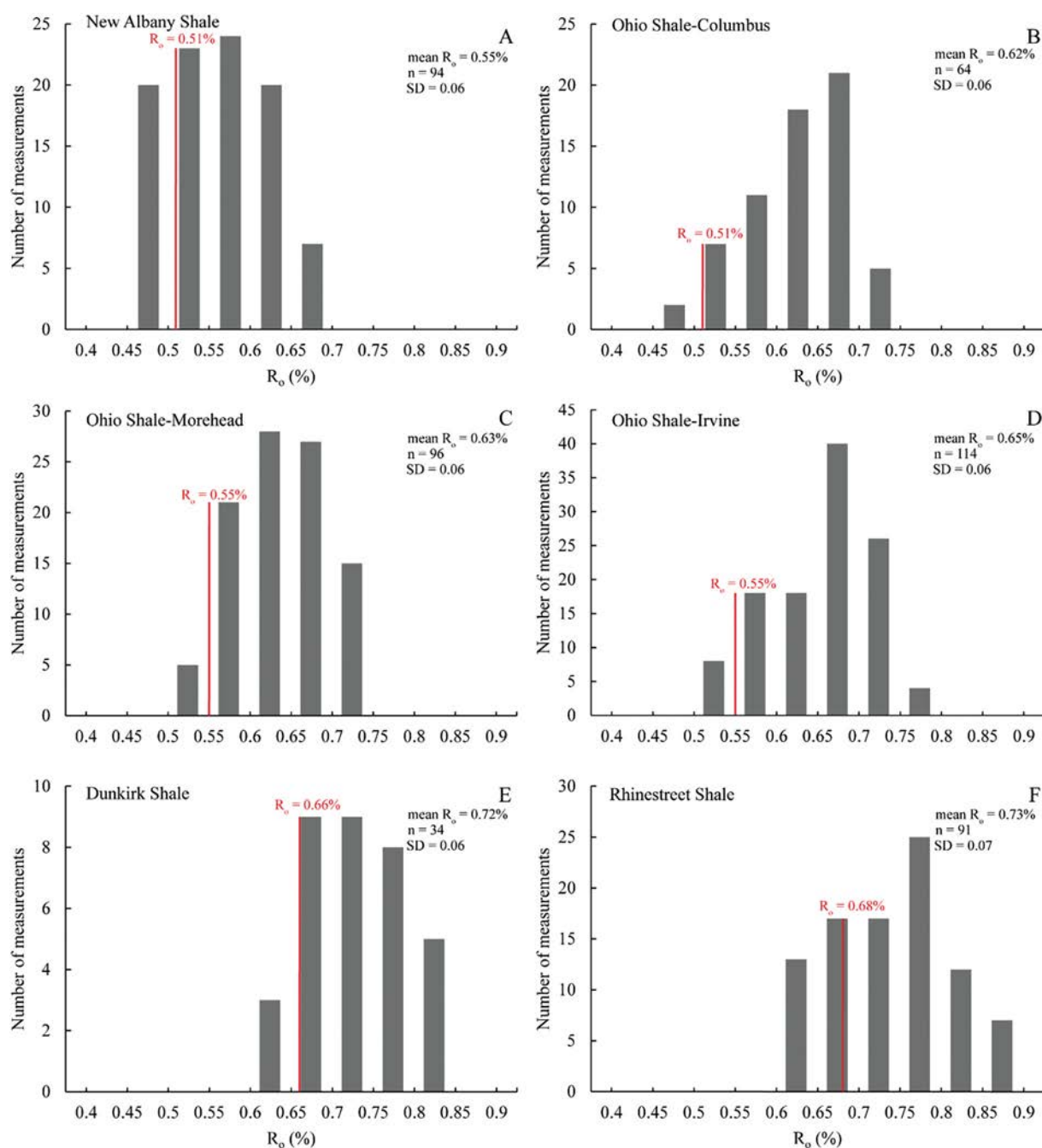


Fig. 10. Histograms of measured R_o of dispersed vitrinite particles in shales that enclose coal lenses. (A) New Albany Shale from Shepherdsville, KY; (B) Ohio Shale from Columbus, OH; (C) Ohio Shale from Morehead, KY; (D) Ohio Shale from Irvine, KY; (E) Dunkirk Shale from Dunkirk, NY; (F) Rhinestreet Shale from Derby, NY (Table 1). n = number of measurements; SD = standard deviation. Red bar indicates the mean R_o of enclosed coals. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Shale and Ohio Shale samples because of their low thermal maturities.

Thermal maturity of black shales as indicated by R_o is an important parameter in reconstructing the thermal history of sedimentary basins and in the exploration for hydrocarbon resources. In this study, the R_o of black shales is 0.04–0.11% (average 0.07%) higher than that of enclosed coals (Fig. 10). Although this R_o difference does not significantly shift the hydrocarbon generation window of the studied black shales, a reevaluation of the applicability of R_o in black shale successions is still recommended, because some vitrinite could become severely oxidized and result in erroneous evaluation of thermal maturity. In addition, oxidized vitrinite can be indistinguishable from “normal” vitrinite (unoxidized) at high maturities. Therefore, R_o should be combined with

other thermal maturation indices such as solid bitumen or zooclast reflectance, Rock-Eval T_{max} , fluorescence of liptinite macerals, or the conodont alteration index to accurately evaluate the thermal history of black shale successions (Teichmüller and Durand, 1983; Bertrand and Héroux, 1987; Tissot et al., 1987; Petersen et al., 2013; Hackley and Cardott, 2016; Mastalerz et al., 2018; Liu et al., 2019a; Schmidt et al., 2019).

5. Conclusions

Coal lenses enclosed in several Upper Devonian black shale formations from the eastern United States provided a unique opportunity to

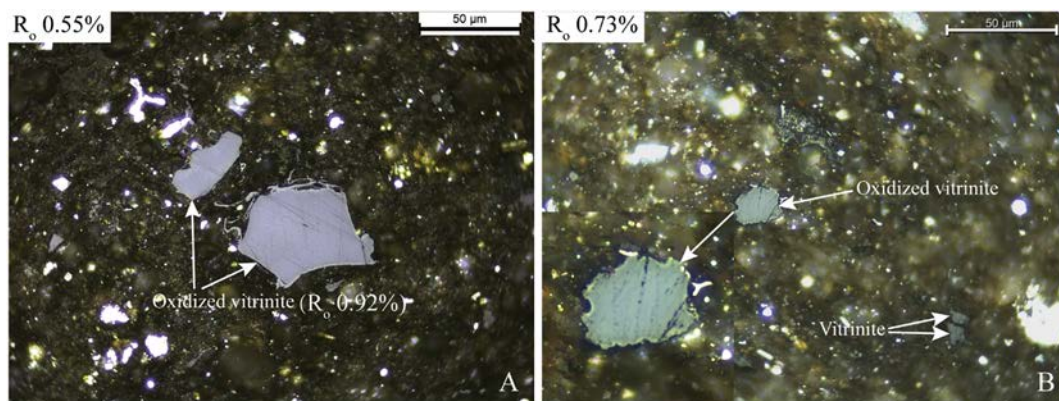


Fig. 11. Photomicrographs of oxidized vitrinite in reflected white light and oil immersion. (A) New Albany Shale from Shepherdsville, KY. Note the sharp edges of the vitrinite particles and the oxidized rim of the larger vitrinite particle. These two particles were excluded for R_o measurements. (B) Rhinestreet Shale from Derby, NY. Insert image is the close-up view of the oxidized vitrinite particle showing an oxidized rim.

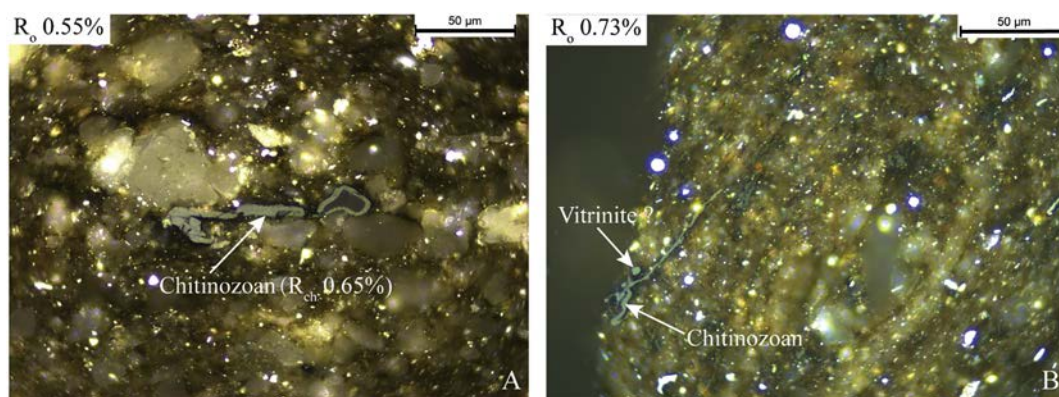


Fig. 12. Photomicrographs of chitinozoan fragments in the New Albany Shale (A) and the Rhinestreet Shale (B) in reflected white light and oil immersion. Note that the vitrinite-like particle next to a chitinozoan fragment in panel B has a comparable reflectance to the chitinozoan fragment, suggesting that it could be a chitinozoan fragment cut perpendicular to the chitinozoan. R_{ch} = chitinozoan reflectance.

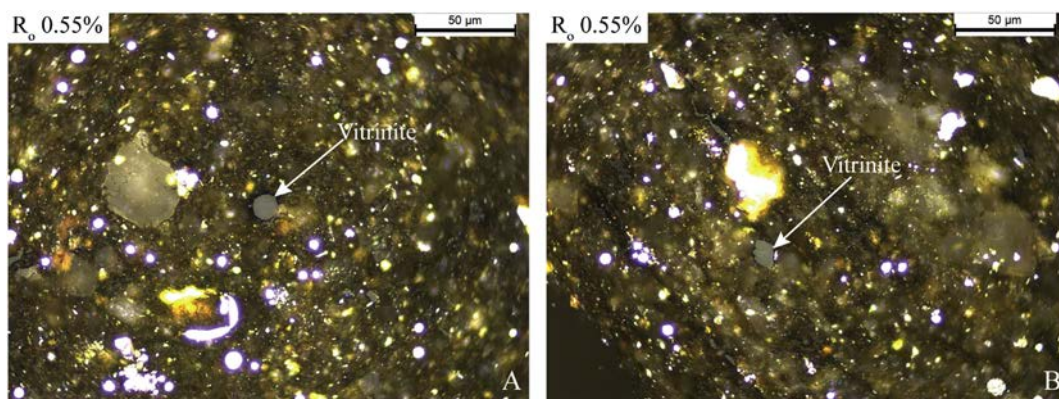


Fig. 13. Photomicrographs of dispersed vitrinite particles in shales in reflected white light and oil immersion. (A) Rounded vitrinite particles that could be cor-pogelinite. (B) Vitrinite particles with sharp edges that could be fragments of collotelinite. New Albany Shale sample from Shepherdsville, KY.

study the applicability of R_o in assessing the thermal maturity of black shale successions. Vitrinite in shales occurs as small dispersed particles ($\sim 5 \mu\text{m}$) in the mineral matrix, whereas coals embedded in shales occur as lenses about 1–2 mm thick and 10–20 cm long in outcrop and contain only vitrinite when examined under the microscope. These coal lenses could be tree trunks or branches transported from nearby land or exposed arches. The measured mean random R_o of dispersed vitrinite particles in black shales is 0.04–0.11% (average 0.07%) higher than that of enclosed coals with R_o ranging from 0.51–0.68%. The higher R_o of dispersed vitrinite particles in shales could be caused by oxidation of

small vitrinite precursor particles because small particles are more frequently suspended during transport to the site of deposition and reworking of bottom sediments after deposition than large driftwood pieces that will turn into enclosed coal lenses during burial diagenesis. Misidentification of fragments of zooclasts such as chitinozoans without recognizable morphological features for dispersed vitrinite particles and variability of phytoclasts in black shales could also contribute to the higher R_o values of shales than those of enclosed coals. When assessing the thermal maturity of black shales via R_o measurements, one should be mindful of the large heterogeneity of vitrinite in black shale

successions.

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Declaration of Competing Interests

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.

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