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ABSTRACT

Previous studies demonstrated that melting, initiated by supercritical fluids in the 375–400 °C range, occurred as part of anthracite metamorphism in the Appalachian Basin. Based on the known behavior of vitrinite at high temperatures and, to a lesser extent, at high pressures, it was determined that the duration of the heating, melting, and resolidification event was about 1 h. In the current study, featureless vitrinite within banded maceral assemblages demonstrates the intimate association of melted and resolidified vitrinite with anthracite-rank macerals. By analogy with metamorphosed inorganic rocks, such associations represent diadysites and embrechites, i.e., cross-cutting and layered migmatites, respectively. Even though the temperature of formation of the anthracite structures is several hundred °C lower than that seen in metamorphosed inorganic rocks, anthracites are metamorphic rocks and the nomenclature for metamorphic rocks may be appropriate for coal.

1. Introduction

While there have been many attempts to correlate mineral and maceral metamorphic parameters, Kisch (1987) noted that the accuracy of the parameters and refinement of the correlations is limited by the nature of the starting materials and many other factors including, but not limited to, oxidation conditions, supply of cations, SiO_2 supersaturation, permeability and porosity of rocks, partial pressure of CO_2, changes in the composition of interstitial solutions with time, persistence of non-equilibrium mineral assemblages, and the selection of the appropriate vitrinite type for reflectance measurements. Basically, as the "coalification experiment" was run several hundred million years ago (in the case of the Pennsylvania anthracites), it cannot be replicated by experimental heating and in most, if not all cases, there are more variables than can be reliably defined. Therefore, not only is it difficult to relate structural and textural characteristics of high maturity coals to those present in metamorphic rocks, it is even more difficult to interpret textural similarities in anthracite and those in metamorphic rocks in terms of temperature and pressure conditions. Considering that organic matter is more sensitive to metamorphic processes than minerals, the question remains whether metamorphic rock terminology can be applied to high maturity coals in cases where structural and textural features resemble closely those in metamorphic rocks.

Metamorphic and tectonic structural techniques and nomenclature (after Ramsay, 1967; Spry, 1969; Bucher and Grapes, 2011) have been applied to high to low volatile bituminous coals in western Pennsylvania by Hower (1978) and Zhang and Davis (1993); to an anthracite from a mine in the Southern Anthracite field, Pennsylvania by Hower (1978) and Hower and Davis (1981); to semi-anthracite- and anthracite-rank coals in the Pennsylvania Anthracite fields by Levine and Davis (1989a); to medium and low volatile coals in the Broad Top coalfield, Pennsylvania by Levine and Davis (1989b); and to several locations in Alberta and British Columbia by Kalkreuth et al. (1990) and Langenberg and Kalkreuth (1991). At a very fine scale, the occurrence of nanotubes in anthracites and meta-anthracites with $R_{max} > 5\%$ is a reflection of the metamorphic temperatures and pressures (Silva et al., 2020). A similar mechanism was noted in anthracites intruded by igneous rocks (Li et al., 2017), but the Silva et al. (2020) discovery appears to be the first for regionally metamorphosed coals.

The theories on metamorphic events in the Pennsylvania Anthracite Fields and, indeed, elsewhere in the Appalachians and beyond, evolved from traditional views of the imposition of coal rank by folding pressures (White, 1925; Turner, 1934), a view that persisted at least to the work of Wood et al., 1969 despite Teichmüller and Teichmüller's (1966) advocacy for the influence of geothermal heating. In contrast,

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more recent works emphasize the static influence of heating at depths of several kilometers accompanying the folding (Hower, 1978; Hower and Davis, 1981; Levine, 1983, 1986; Levine and Davis, 1989a, b; Roden and Miller, 1989; Kisch and van den Kerkhof, 1991; Hulver, 1997; Faill, 1998)[1] and/or the influence of hydrothermal fluids (Gresley, 1896; Reeves, 1928; Oliver, 1986, 1992; Juster et al., 1987; Daniels and Altaner, 1990; Daniels et al., 1990, 1994, 1996; Hower et al., 1993; Hower, 1997, 2013; Hower and Gayer, 2002; Harrison et al., 2004; Ruppert et al., 2010; Ruppert et al., 2014). Higher vitrinite reflectances in the Pennsylvanian anthracites (Ruppert et al., 2010) compared to those in the underlying Devonian black shales (Repetski et al., 2008) suggest that hydrothermal fluids might have preferentially flowed through the Pennsylvanian rocks rather than the Devonian rocks. Coal, however, records higher reflectance than vitrinite in associated clastic sediments around the same intrusive body (Quaderer et al., 2016). Based on the above observations, it is hypothesized thatthe estimated peak temperature ranged from 200 °C for the semi-anthracites in the western portion of the coalfield (Levine, 1983; Nickelsen, 1983) to 260–275 °C in the higher-rank, eastern basins (Eastern Middle Field and the eastern end of the Southern Field) (Juster et al., 1987; Daniels and Altaner, 1990). Fluid inclusion (Nickelsen, 1983; Kisch and van den Kerkhof, 1991) and apatite fission-track data (Roden and Miller, 1989; Blackmer, 1992; Blackmer et al., 1994) support these temperature estimates.

In Pennsylvania anthracite, based on extant maceral and resolidified thermoplastic vitrinite evidence, Hower et al. (2019) estimated that melting and resolidification of the high volatile C or B bituminous vitrinite occurred at 375 °C < T < 400 °C at a depth of at least 1-km over a time span of about an hour. Hot fluids, perhaps in a supercritical state, drove the metamorphism,emphasizing the interplay of temperature and pressure in metamorphism. The heating certainly persisted after the coal solidified, so overall, the extent of the metamorphic event would have been longer than an hour given the amount of time necessary for cooling from the high-300 °C temperature range to the ambient temperature (unknown, but certainly somewhat lower than the peak metamorphic temperature). Clearly, temperatures close to the peak metamorphic temperatures should be more influential in coal rank advancement than the subsequent lower temperatures, so the entire heating and cooling time span is less important than the time near the peak temperature (Hood et al., 1975). For example, Barker (1991) argued that the duration of heating could be a poor predictor of the vitrinite reflectance under some conditions.

Complex heating events like those discussed above often result in unusual textures in anthracites and natural cokes. In this study, the applicability of igneous/metamorphic nomenclature to coal textures is discussed by way of examples from the Pennsylvanian-age Pennsylvania Anthracite Fields (specifically the Buck Mountain coal vein[2] from Schuylkill County) and Narragansett Basin (Rhode Island) meta-anthracite fields, fire-altered Pennsylvanian coal from Alabama, and igneous intrusion-altered coals from North Carolina and Virginia (both Triassic) and KwaZulu-Natal, South Africa (Permian). It is noted that if metamorphic terminology is applied to coal textural features, it is very important to address temperature and pressure conditions responsible for their formation in comparison to the formation of similar textures in the higher temperature, about 630–800 °C, rock metamorphic regime.

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[1] In an example from outside of the Anthracite Fields, Hower and Wild (1994) noted that Jurassic coals (continental slope offshore New Jersey) had high volatile A reflectancesat current sediment depths of 3.5 to 3.9 km, emphasizing the role of tectonic history in determining the coal rank. The Jurassic coals were deposited in a rift basin, initially near a spreading ocean ridge; a passive margin, not an orogenic belt as with the Anthracite Fields.

[2] The convention in the Pennsylvania Anthracite Fields has been to refer to coal beds as veins.

2. Methods

Samples of the Pennsylvanian Buck Mountain vein from a mine in western Schuylkill County, Pennsylvania, were examined. The epoxy-bound pellets were made from the broken remnants, generally >1-mm size, of a 5 cm × 5 cm block sample that had been subjected to strength testing and from a 1-mm-thick oriented wafer prepared for thermal testing (Weisenberger et al., 2020). As such, the maceral composition and chemistry of the test pellets are not representative of the whole coal. Both pellets were prepared to a final 0.05-μm alumina slurry polish. All other samples cited in the study were previously prepared for other studies.

Reflectance measurements were taken on epoxy-bound particulate pellets using a 50×, reflected-light, oil-immersion objective utilized incoming light polarized at 45°. The reflected light passed across a grating monochrometer set at 547 nm on the path to the photomultiplier tube. Random reflectance was determined from the average of orthogonal measurements in the same area in which the maximum reflectance represents the maximum reading following a rotation of the stage.

Photographs were taken with a Diagnostics Instruments Spot Insight CMOS 5 MP camera on a LeitzOrthoplan microscope using a 50×, oil-immersion, reflected-light objective and crossed polars with and without a wavelength plate.

3. Results and discussion

3.1. Evidence from previous studies

Hower et al. (2019), based on the observed textures of the macerals in the coals, discussed melted and resolidified coal in Pennsylvania anthracites and intruded coals from Illinois, Colorado, Antarctica, and South Africa. As a summary of such textural changes, Fig. 1 shows some examples from markedly different types of coal metamorphism, all converging on some common features. Fig. 1A and B show examples of pyrolytic carbon in Pennsylvania anthracite. Much like some of the examples discussed in Hower et al. (2019), the vitrinite at the top of Fig. 1B apparently had melted. Goodarzi and Cameron (1989; their fig. 5), in an example from the Cretaceous Telkwa coalfield, British Columbia, and Suárez-Ruiz et al. (2006; their fig. 2, photo 3), in an example from a late Westphalian Peñarroya Basin anthracite, show featureless vitrinite with devolatilization vacuoles. The latter two examples (Goodarzi and Cameron, 1989; Suárez-Ruiz et al., 2006), the three images from Fig. 10 of Hower et al. (2019), and the example in Fig. 1B (and possibly Fig. 1A) of this paper are all embrechites, or layered migmatites (after Spry, 1969).

Pyrolytic carbon occurs along the edges of the brecciated fragments in Fig. 1C. Fig. 1C and D shows mosaic structures in Rhode Island meta-anthracites. Li et al. (2018, 2019) identified similar features as microcrystalline graphite in their study of a Hunan, China, Carboniferous-age coal altered by a granitic pluton. In addition, a meta-anthracite of unknown age and location, appears to have a sub-micron granularity (mosaic structure) in the vitrinite (Fig. 2; Kenyon et al., 2021 in review).

The Mulga, Alabama, coal, collected from a burning coal-beneficiation-reject pile, shows structures associated with melting and coking (Fig. 3A and B; from unpublished work by Hower). The Triassic natural cokes from North Carolina (Fig. 3C) and Virginia (Fig. 3D) show the mosaic structure associated with the repolymerization of a melted vitrinite into a semicoke (Gray and DeVanney, 1986; Hower and Lloyd, 1999). Fig. 3D, from the Triassic Richmond (Virginia) Basin, illustrates an example of a coke in close association with anthracite-rank vitrinite; in this case, the liquid vitroplast migrated through fractures and/or void spaces, perhaps associated with the fusinite, and repolymerized adjacent to vitrinite. Based on the nature of the coke, the pre-heating vitrinite was likely high volatile bituminous rank. Coke adjacent to inertinite and, possibly, vitrinite is shown in examples from Permian KwaZulu-Natal, South Africa, coal (Fig. 3 E,F). Vitrinite
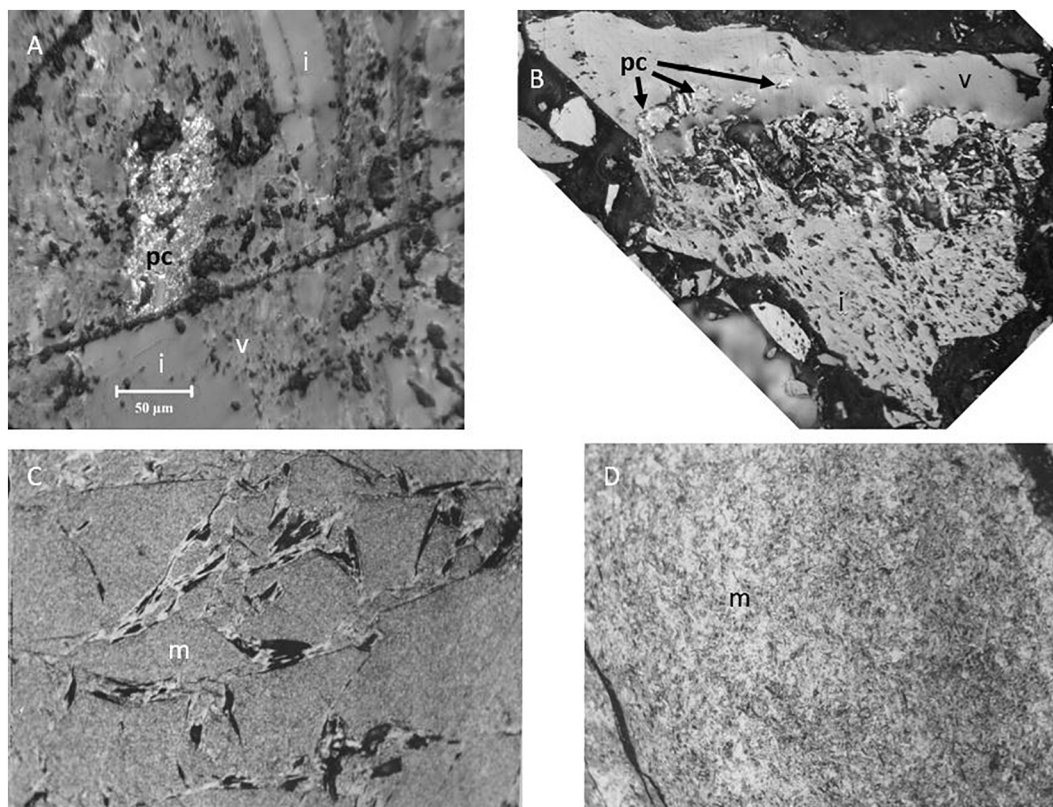
Fig. 1. Examples of thermoplastic and coke-like structures in coals: (A) Pyrolytic carbon (pc), vitrinite (v), and inertinite (i) in Schuylkill County, Pennsylvania, Pennsylvanian anthracite (sample supplied by Blaschak Coal; Hower et al., 2017). Image 94000 01. Scale = 50 μm. (B) Pyrolytic carbon (pc) with vitrinite (v) and inertinite (i) in Luzerne County, Pennsylvania, Pennsylvanian anthracite (sample supplied by Blaschak Coal). Image 94037 15. Scale = 50 μm. (C) Fine mosaic (m) structure in Cranston, Rhode Island, Pennsylvanian meta-anthracite. Scanned image (10,260 Cranston) from picture donated to Hower by Ralph Gray in conjunction with the writing of Hower et al. (1993). Scale not known. The coal rank of the Cranston meta-anthracite was discussed by Quinn and Glass (1958) and Kisch (1974c). (D) Mosaic structure (m) in Rhode Island Pennsylvanian meta-anthracite. Scanned image (9350 6 Rhode Island) from picture donated to Hower by Ralph Gray in conjunction with the writing of Hower et al. (1993). Scale not known. Skehan et al. (1982) summarized the geologic setting of this sample and the Fig. 1C sample.
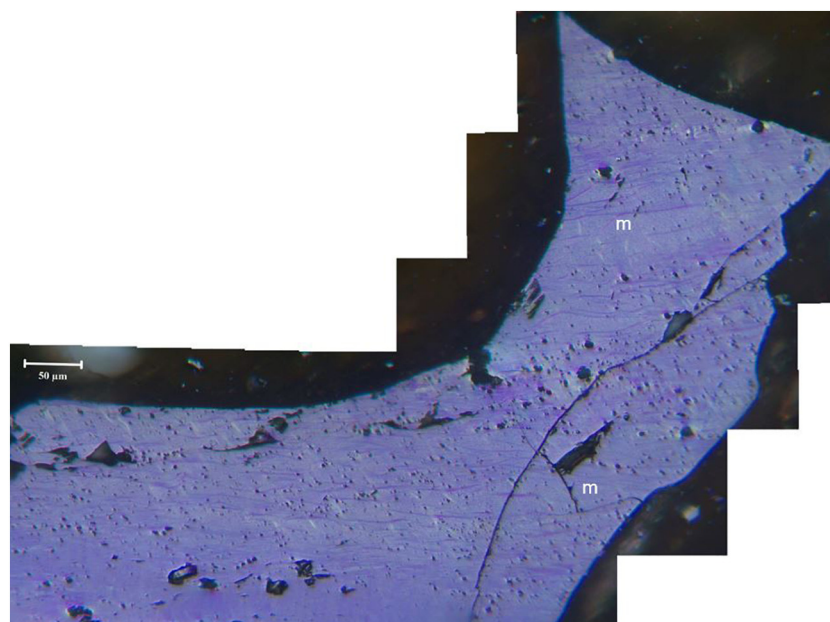
Fig. 2. Sub-micron mosaic (m) structure in vitrinite from the North Carolina site of the 1718 *Queen Anne's Revenge* shipwreck (Kenyon et al., 2021, in review). The meta-anthracite, with 7.5% R_{max}, was likely from a U.S. Navy ship in the late-19th /early-20th century, and, thus, might be from the Pennsylvania Anthracite Fields.As a domestically produced smokeless fuel, Pennsylvania anthracite was a common fuel source used by the U.S. Navy at that time; therefore, the co-location with a 1700's shipwreck is coincidental. Image CCA 04 14. Scale = 50 μm.
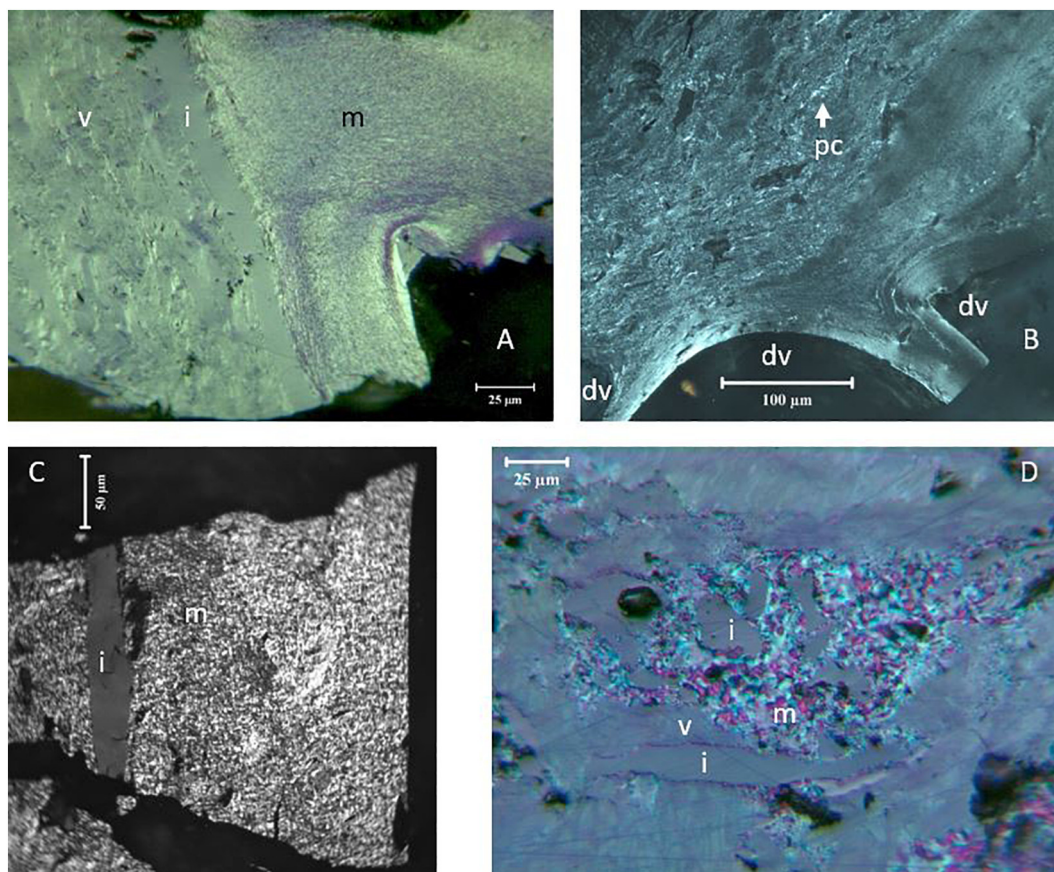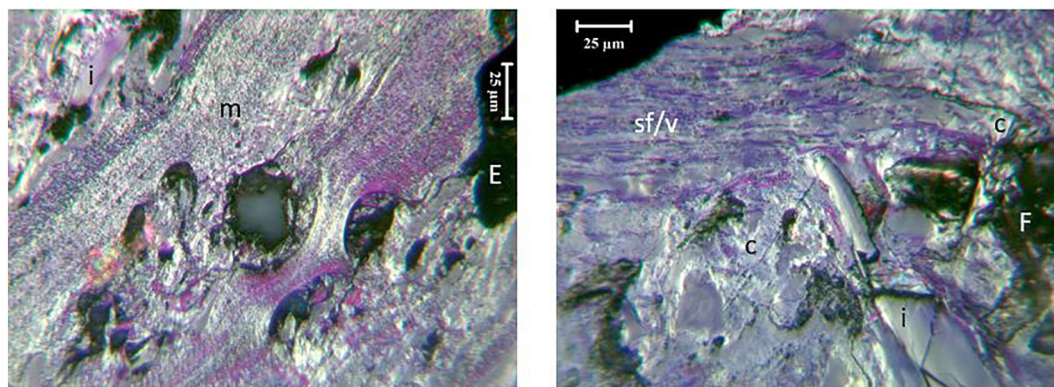
Fig. 3. (A) Fine mosaic (m) structure in heated (originally) Pennsylvanian high volatile A/medium volatile bituminous vitrinitefrom Mulga, Alabama (sample collected by Hower; site discussed by Kolker et al., 2009). Inertinite (i) and vitrinite (v) are also shown. Image Mulga 04. Scale = 25 μm. (B) Thin bands of pyrolytic carbon (pc) and devolatilization vacuoles (dv) in Pennsylvanian heated (originally) high volatile A/medium volatile bituminous vitrinite from Mulga, Alabama (sample collected by Hower; site discussed by Kolker et al., 2009). Image Mulga BW 10. Scale = 100 μm. (C) Mosaic structure (m) and inertinite (i) in natural coke from the Egypt mine, Triassic Deep River basin, Lee County, North Carolina (region discussed by McArver, 2006; also see: http://www.ncmarkers.com/Markers.aspx?sp=search&k=Markers&sv=H-41). Image Vandy 683 15. Scale = 50 μm. (D) Mosaic structure (m) after thermoplastic vitrinite adjacent to inertinite (i) and anthracite-rank vitrinite (v) in Triassic coal from Richmond Basin, Virginia (Hower, 2014). Image Vandy 675 02. Scale = 25 μm. (E) Vesiculated mosaic coke (m) with inertinite (i). KwaZulu-Natal, South Africa. Image 69763 14. Scale = 25 μm. (F) Inertinite (i) and a semifusinite/vitrinite (sf/v) with coke (c). KwaZulu-Natal, South Africa. Image 69763 18. Scale = 25 μm.

occurs adjacent to coke in the same coal (fig. 12C of Hower et al. (2019)). Given the timing of melting of vitrinite and the repolymerization as a coke, on the order of one hour, even if the cokes illustrated above did not form *in situ*, they did not flow far from their melting point prior to resolidification.
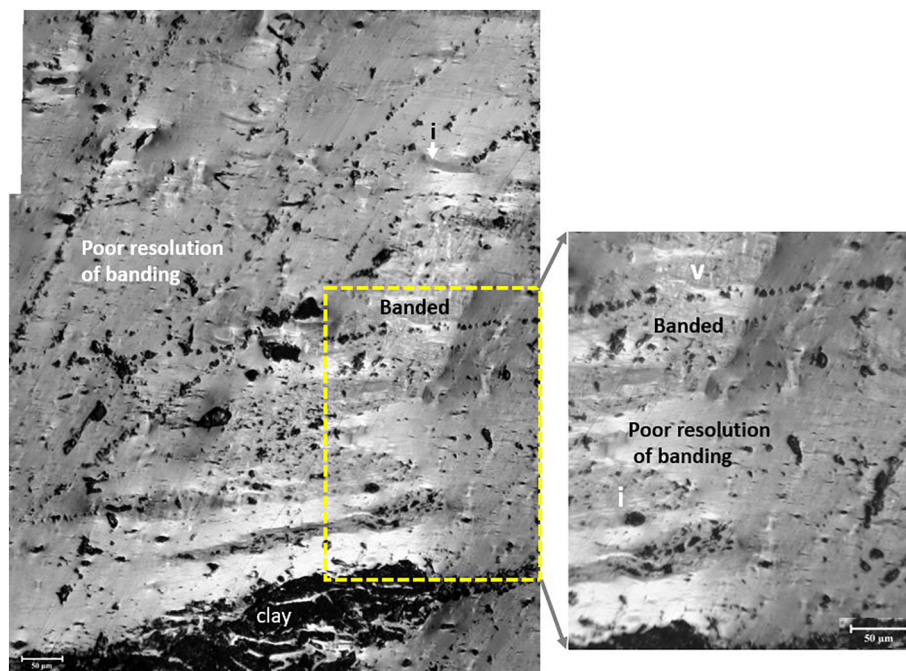
3.2. Evidence for melting of vitrinite

The coal sample examined in this study, an anthracite (R_{max} 5.30% and R_{random} 4.51%) from the Buck Mountain vein, Western Middle Anthracite Field, Schuylkill County, Pennsylvania, has poor to
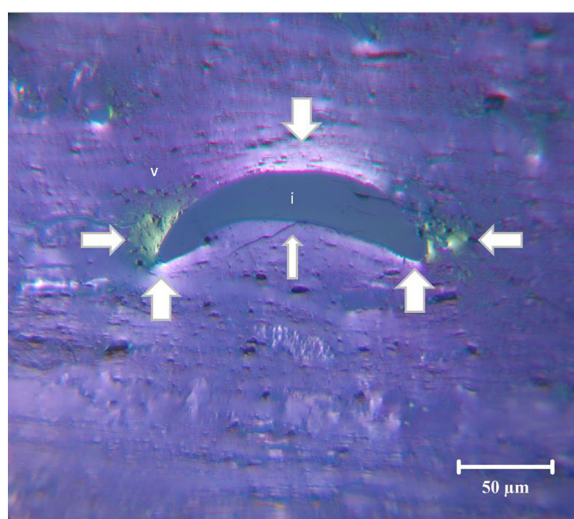

Fig. 3 (*continued*).

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Fig. 4. Portion of the Buck Mountain vein with areas of banded vitrinite (v) and inertinite (i) and contiguous areas of poor to non-existent banding. Composite image 8689 s 08 with inset image 8689 s 08d. Scale = 50 μm on both the composite and inset images.

non-existent banding flankinga region with clearly banded coal (Fig. 4). Inertinite is the best indicator of the vague traces of banding in the adjacent "poor to non-existent banding" region. In terms of migmatites, this is a diadysite structure, a cross-cutting migmatite (after Spry, 1969). Anisotropy in anthracitic vitrinite is common when the vitrinite is deformed around a mineral or harder maceral (inertinite) (see Fig. 5). While the contrast seen in Fig. 5 is a function of the deformation stress associated with the mineral band (clay) at the bottom of the composite image, this does not appear to be the case in the Fig. 4 example, as both the composite and inset views show that the anisotropy-related change
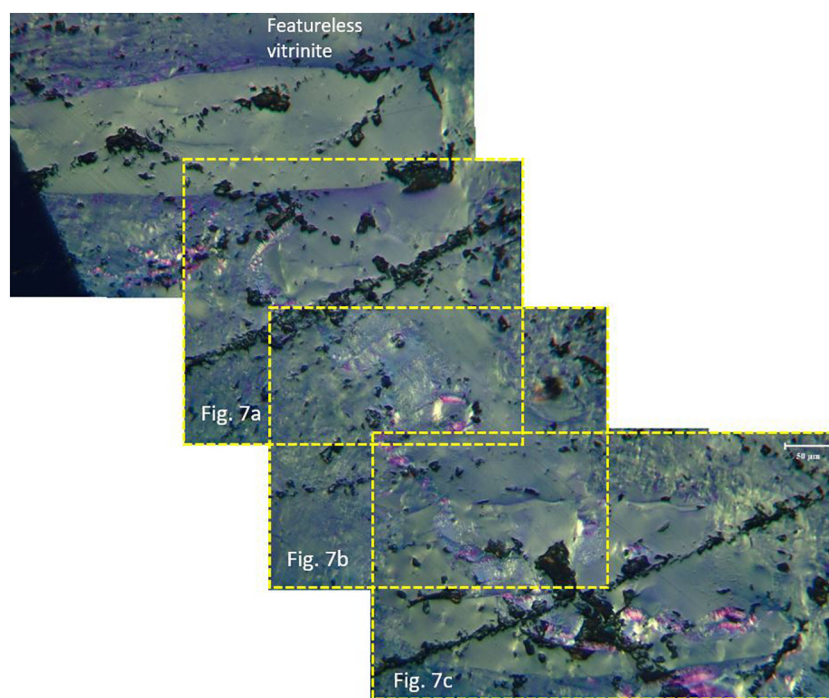
in reflectance of the vitrinite occurs within the "poor resolution of banding" area in the inset.

Fig. 6, along with the enlargements of select areas of the composite view (Fig. 7A–C), shows an area between the two inertinites with even clearer signs of the loss of banding and of the presence of pyrolytic carbon along one side of the latter feature and in one of the inertinite grains (Fig. 6).The area of interest, the proposed diadysite migmatite texture (after Spry, 1969), is a featureless vitrinite lodged between two inertinite grains about 300-μm apart (Fig. 7A–C). In this view, the left side of the featureless vitrinite has pyrolytic carbon along its edge, extending more than 100 μm into the otherwise unaltered vitrinite (in part on Fig. 7A with the entire length shown on Fig. 6). In Fig. 7C, the pyrolytic carbon extends along fractures in the inertinite. Fig. 7A–C shows the fine, micron-scale details of the pyrolytic carbon.

This isotropic coke apparently formed consequent to the heating and melting of the high volatile C/B bituminous vitrinite. As per the discussions of Hower et al. (2019), the estimation of the original rank of the melted coal is constrained by the lack of mesophase in the thermoplastic vitrinite. This is in contrast to the original higher bituminous rank, perhaps high volatile A to medium volatile, as inferred from the possible mesophase structure,of theprecursors of the Narragansett Basin vitrinite (Fig. 1C,D); the Lee County, North Carolina, coke (Fig. 3C); and the Mulga vitrinite (Fig. 3A,B). The presence of pyrolytic carbon, both in vitrinite porosity and at edge of melt zone, and in fractures in inertinite suggests a gas phase associated with the Anthracite Field melting event (after de Sousa, 1978; Goodarzi, 1985; Goodarzi et al., 1992).

The mechanisms discussed by Hower et al. (2019) and in this paper represent ephemeral events. In coals, short-duration events such as this heating, melting, and resolidification over the course of about 1 h at a melting temperature of 375–400 °C (Hower et al., 2019), require higher temperatures than in regional metamorphism. Mineral-based temperature estimates of short-term events in coals are problematical; indeed, Kisch (1987) suggested that the "no time influence" model of Price and Barker (1985) over-estimated the metamorphic temperature inferred from the mineral assemblages (Fig. 8). This could explain why the projected temperatures exceed the mineral-based estimates by

Fig. 5. Inertinite (i) in vitrinite (v). Note the differences in brightness between points where the curved inertinite in intruding into the vitrinite (broad arrows) compared to the opposite points of no intrusion (narrow arrow) and minimal intrusion (intermediate-width arrow). For discussions of similar phenomena in non-coal metamorphic rocks, see Spry (1969). Anthracite, with 5.75% R_{max}, is from the Queen Anne's Revenge site (see discussion with Fig. 2 caption). Image CCA 03 04. Scale = 50 μm.

Fig. 6. Portion of the Buck Mountain vein with large inertinite grains, banded vitrinite, and areas of poor to non-existent banding ("featureless vitrinite") between and above the inertinite. The areas for Figs. 7a to 7C are indicated. Composite image 8689 t 04. Scale = 50 μm.
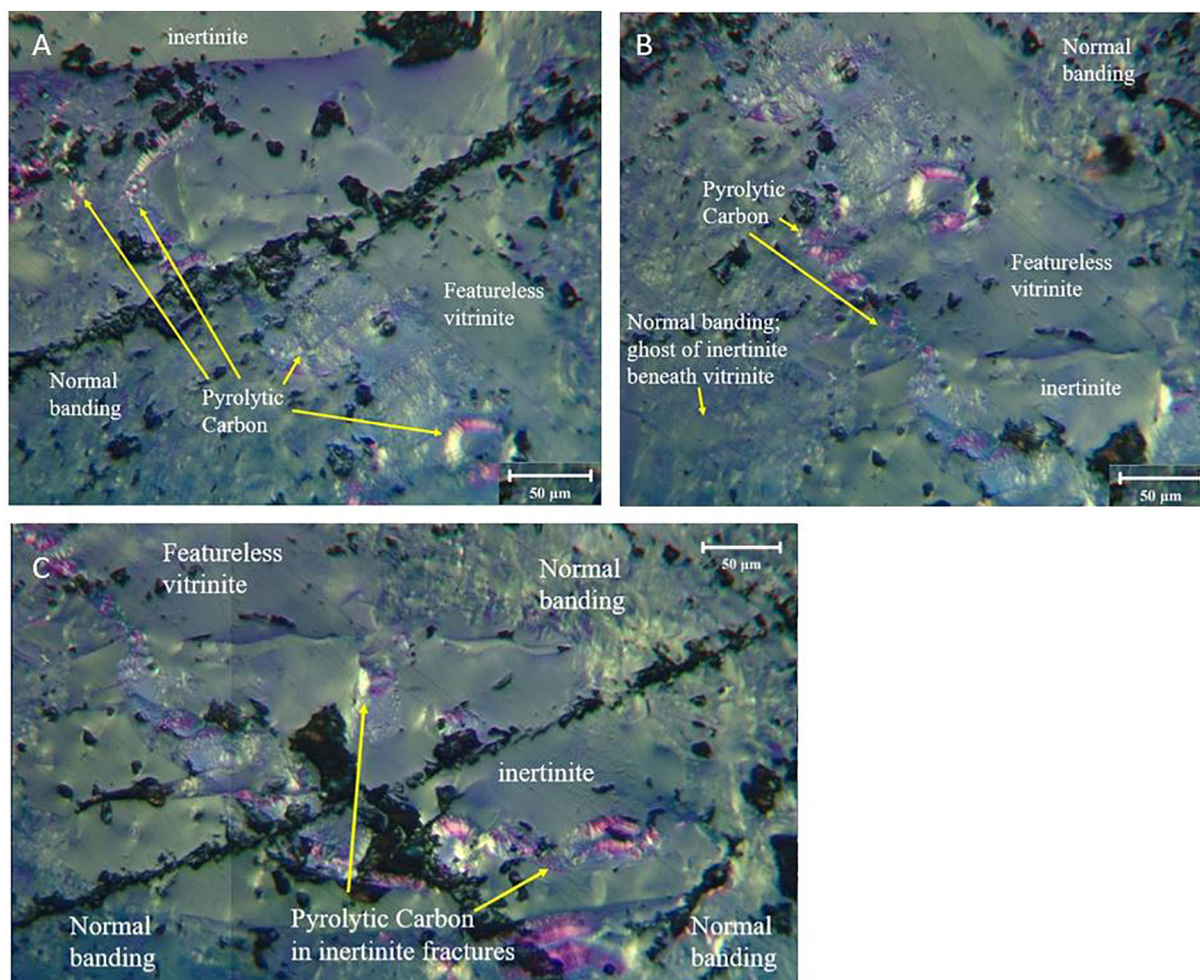
Fig. 7. (A) Inertinite, normal banding, featureless vitrinite, and pyrolytic carbon. Image 8689 t 04d. Scale = 50 μm. (B) Inertinite, normal banding, featureless vitrinite, and pyrolytic carbon. A portion of one inertinite fragment is visible beneath this veneer of vitrinite. Image 8689 t 04c. Scale = 50 μm. (C) Inertinite, normal banding, featureless vitrinite, and pyrolytic carbon. Some of the pyrolytic carbon is present in fractures in the inertinite. Composite image 8689 t 04b and 4a. Scale = 50 μm.
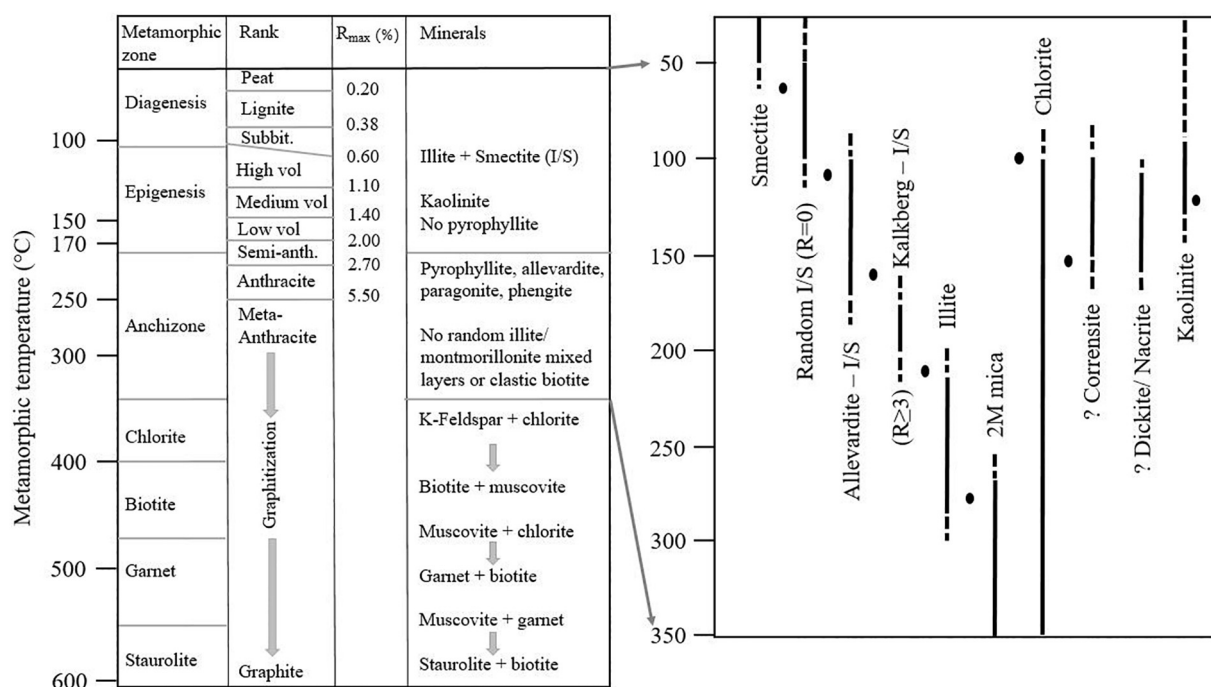
Fig. 8. Metamorphic zones, coal rank, approximate vitrinite maximum reflectance (R_{max} %), and some of the diagnostic mineral transitions and assemblages (after Kisch, 1974c, 1990). Key: Subbit. – subbituminous; High vol – high volatile bituminous; Medium vol – medium volatile bituminous; Low vol – low volatile bituminous; Semi-anth. – semi-anthracite. Mineral diagenesis diagram (on right; after Pollastro (1990) as modified after Hoffman and Hower (1979)) showing temperature ranges of common clays and I/S in diagenetic and low-grade metamorphic settings. Note: Solid ovals denote approximate phase-limiting or phase-transitional temperatures.

100 °C or more. Neither the mineral- or maceral-thermoplasticity-based estimate is necessarily inaccurate, they are just developed for distinctly different materials; the mineral-based changes would not have happened within the time interval of the heat pulse necessary to influence the macerals. While the melting of the vitrinite and the alteration of the megaspores (as per Hower et al., 2019) may have been initiated by supercritical fluids in the 375–400 °C range, the overall metamorphic push to anthracite rank would have been the result of the lingering thermal effect of the event (or multiple thermal events). Even at 1-km depth, the minimum necessary for the stability of supercritical fluids, the heat would have lingered long enough to significantly influence coal rank.

4. Summary

Coal, specifically the vitrinite in bituminous coals, can undergo melting from 375 °C to 400 °C and resolidification in the mid-400 °C range or in about an hour, even if the temperature does not reach the mid-400 °C's. The textures associated with this phenomenon are best described by nomenclature usually reserved for igneous and metamorphic rocks.

The textures seen in Fig. 5, and to a lesser extent in Fig. 3, are diadysites (after Spry, 1969), a crosscutting migmatite. In contrast, the Fig. 1A (this paper) and Hower et al.'s (2019) fig. 10C varieties would be embrechites (after Spry, 1969), or layered migmatites. The texture of coal versus gneiss/granite, in this case, is independent of the temperature, just as the rock type (meaning sedimentary/metamorphic/igneous) is fundamentally independent of the distinction between macerals versus minerals.

Neither the temperature of formation nor the basic composition of the rock should disqualify the adoption of an appropriate term from another geology discipline, in this case from metamorphic petrology to coal geology. Particularly since anthracites are the temperature equivalent of, at least, low-grade metamorphic rocks (Kisch, 1974a–c), metamorphic petrology nomenclature is appropriate for the description of

high-rank coals. In the settings examined here, the temperature increase not only initiated the melting of the vitrinite, but it also contributed to the increase in coal rank from high volatile C/B bituminous at the onset of the metamorphic event to the anthracite rank now seen in the coal.

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

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