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Key Points:
- Integrated magnetic, geochemical, and petrographic study of a typical destroyed hydrocarbon reservoir was conducted
- A new link between remagnetization and destruction of hydrocarbon reservoir has been revealed
- Caution is warranted in the interpretation of paleomagnetic dates for hydrocarbon systems

Supporting Information:
- Supporting Information S1

Correspondence to:
D. Jia,
djia@nju.edu.cn

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Identifying and Dating the Destruction of Hydrocarbon Reservoirs Using Secondary Chemical Remanent Magnetization

Yong Zhang[1,2], Adrian R. Muxworthy[3], Dong Jia[1], Guoqi Wei[4], Bin Xia[2], Bin Wen[5], Maomao Wang[6], Weiliang Liu[2], and Matthew J. Brzozowski[1]

[1]State Key Laboratory for Mineral Deposits Research and Department of Earth Sciences, Nanjing University, Nanjing, China, [2]School of Marine Sciences, Sun Yet-Sen University, Guangzhou, China, [3]Natural Magnetism Group, Department of Earth Science and Engineering, Imperial College London, London, UK, [4]Research Institute of Petroleum Exploration and Development, PetroChina, Beijing, China, [5]Department of Geology and Geophysics, Yale University, New Haven, CT, USA, [6]College of Oceanography, Hohai University, Nanjing, China

Abstract Destructive processes are thought to be common in pre-Cenozoic oil-gas reservoirs. The timing, mechanism, and even identification of these processes, however, are difficult to clearly characterize, which obscures the evolution of such systems and the assessment of oil and gas reserves. Here, we reveal a new link between secondary chemical remanent magnetization acquisition and tectonically driven destruction of hydrocarbon reservoirs, which can be used to date the destructive processes and identify their tectonic controls. We performed a detailed paleomagnetic analysis of rocks from a typical destroyed reservoir (Majiang reservoir, China) and combined these data with scanning electronic microscope imaging and strontium isotope, total organic carbon, and clay analysis. We found that the Late Triassic syntilting secondary chemical remanent magnetizations of source and reservoir rocks resulted from the destructive processes driven by the Indosinian orogeny. We therefore argue that palaeomagnetic methods can be used to constrain destructive events within hydrocarbon reservoirs worldwide.

1. Introduction

Destructive processes in reservoirs are defined as those that cause the alteration or physical removal of hydrocarbons from a trap (Macgregor, 1996). They are commonly observed in oilfields and represent a key stage in the evolution of a reservoir; consequently, they are considered important when assessing hydrocarbon reserves. Postentrapment tectonism is thought to be the most significant control on hydrocarbon reservoir destruction, leading to, for example, leakage, hydrodynamic flushing, and hydrocarbon remigration (Macgregor, 1993, 1996; Nalivkin et al., 1984; Pang et al., 2018), though there are other factors not related to tectonism that can lead to destruction, for example, biodegradation (Emmerton et al., 2013; Wilhelms et al., 2001). If tectonically driven, there are many cases where it is unclear whether an oilfield has been affected by destructive processes, as evidence is rarely preserved (Macgregor, 1996). In other instances, a lack of constraints on the timing of the destructive processes makes identification of related tectonic events challenging. These uncertainties make it difficult to conduct analysis of hydrocarbon evolution and the role of determination of tectonics in hydrocarbon systems.

Paleomagnetism can be used to identify and date complex geological processes by isolating secondary chemical remanent magnetizations (CRMs) and comparing these with the apparent polar wander path for the region in question (Benthien & Elmore, 1987; Cioppa et al., 2003; De Kock et al., 2008; Elmore & Dulin, 2007; Jackson et al., 1988, 1992; Wilkinson et al., 2017; Zegers et al., 2003). A necessary condition for this technique to be successful is a link between acquisition of the secondary CRMs and geological processes. In hydrocarbon-bearing strata, the acquisition of CRM is usually linked to the maturation of organic matter and hydrocarbon migration and accumulation (Elmore et al., 1987; Blumstein et al., 2004; Emmerton et al., 2013; Abubakar et al., 2015; Manning & Elmore, 2015; Zhang et al., 2016, 2018), which are other important stages of reservoir evolution.

It is the aim of this study to test whether it is possible to identify a link between a secondary CRM and destructive processes of hydrocarbon reservoirs. In addition, as the orogenic fluids during the tectonic

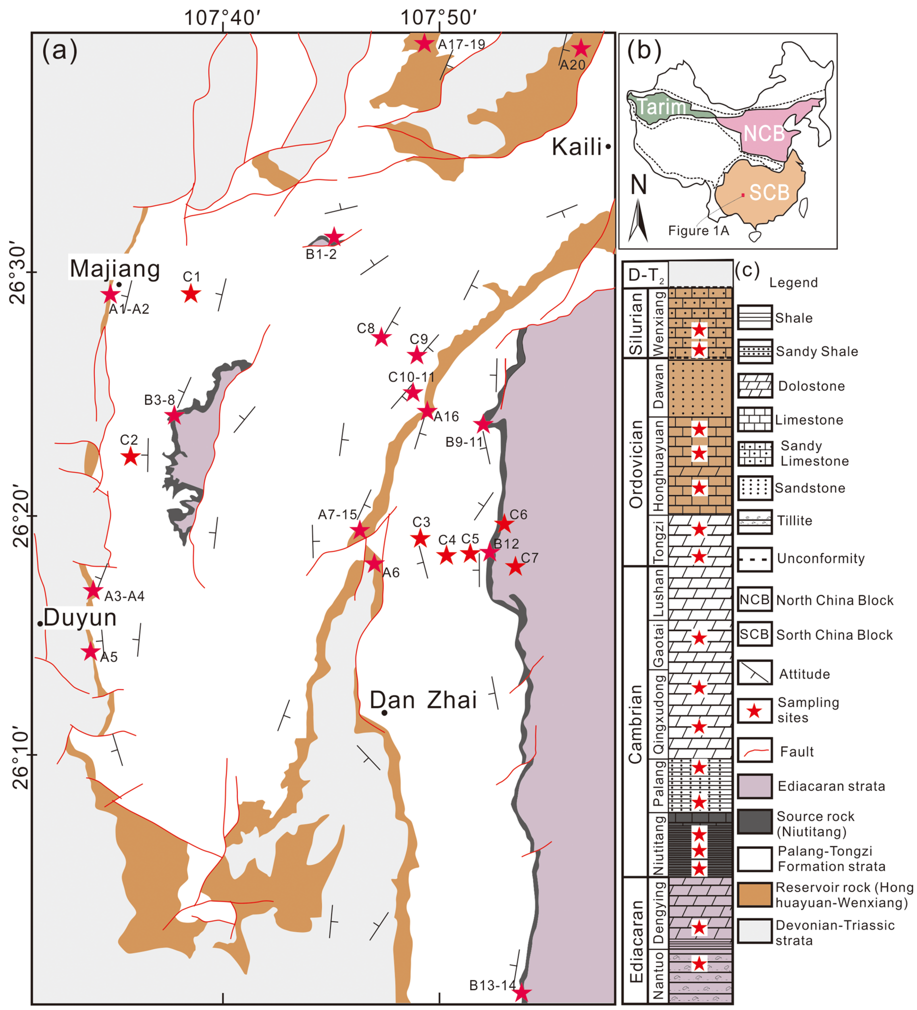

Figure 1. (a, b) Simplified geological map of the Majiang paleoreservoir (South China) showing the distribution of source and reservoir rocks, as well as sampling locations used in this study. (c) Ediacaran-Silurian stratigraphic column for the Majiang paleoreservoir.

events could have widely altered the units in the reservoir, we also want to know the relationship between the remagnetization, tectonism and wide alteration by orogenic fluids in these units. The Majiang paleoreservoir in south China (Figure 1) is ideal for this purpose as it represents a typical, destroyed reservoir with well-exposed source rocks and reservoir rocks. The reservoir was also affected by multiple tectonic events, which likely had significant impacts on the destructive processes (Ge et al., 2016; Wu, 1989).

2. Geological Setting

The Majiang paleoreservoir is one of the largest hydrocarbon reservoirs in South China to display evidence for destruction (Deng et al., 2014). The source rock is a lower Cambrian black shale, and the reservoir comprises sandy limestone of the middle Silurian Wenxiang Group and limestone of the Lower Ordovician Honghuayuan Formation. The Wenxiang Group unconformably overlies the Honghuayuan Formation. These hydrocarbon-bearing strata show a linear distribution along the flank of the folds, while the oil-free sandy shale of the Cambrian Palang Formation and dolostone of Qinxudong-Tongzi Formation are distributed at the core of the folds (Figure 1a). Other exposed oil-free units around the reservoir include Ediacaran and Devonian-Middle Triassic sedimentary rocks (Figure 1).

The reservoir is located within a foreland basin belt along the western margin of the Xuefeng Uplift in the mid-Yangtze Block, which has undergone multiple tectonic events including the Paleozoic Caledonian,

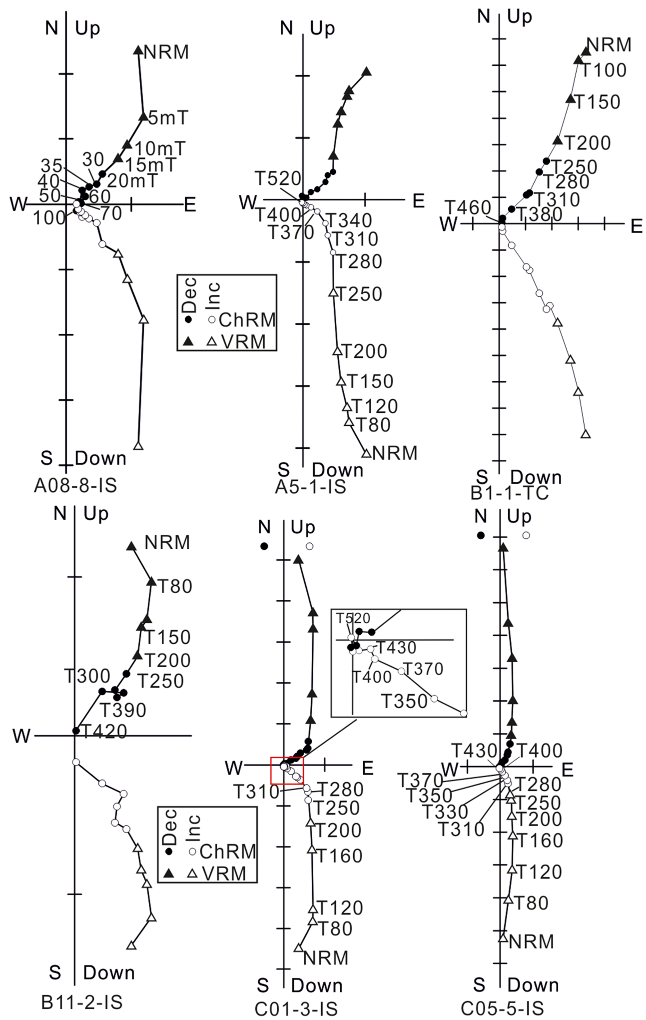

Figure 2. Progressive alternating field and thermal demagnetization plots for representative samples. ChRM and VRM were identified as shown in the figure. VRM = viscous remanent magnetization; NRM = natural remanent magnetization.

Triassic Indosinian, and Cretaceous Yanshan orogenies (Ge et al., 2016). These events likely had significant impacts on the evolution of petroleum systems in the region, and numerous tectonic models and age constraints therefore have been proposed (Han et al., 1982; Gao et al., 2012; Ge et al., 2016; Tang & Cui, 2011; Wu, 1989; Xiang et al., 2008). Based on tectonic events and basin modeling, previous studies have been unable to clearly identify the tectonic event that lead to the destruction of the hydrocarbon reservoir (e.g., Xiang et al., 2008).

3. Sampling and Methods

To investigate the characteristic remanent magnetization (ChRM) in both source and reservoir rocks, three groups of samples were collected from the Majiang paleoreservoir, over an area of ~42,000 km^2 (Figure 1). Group A samples were collected from 20 sites and comprise limestone and sandy limestone reservoir rocks of the Honghuayuan Formation and Wenxiang Group, respectively. Group B samples were collected from 14 sites within the source rocks (lower Cambrian shale). Group C samples were collected from various lithologies, which lack evidence of hydrocarbon or bitumen impregnation. These samples were primarily Ediacaran tillite (C6) and limestone (C7), lower Cambrian mudstone (C1, C5), middle Cambrian shale (C4), and dolomite (C2, C3, and C8), and upper Cambrian carbonate (C9 and C11).

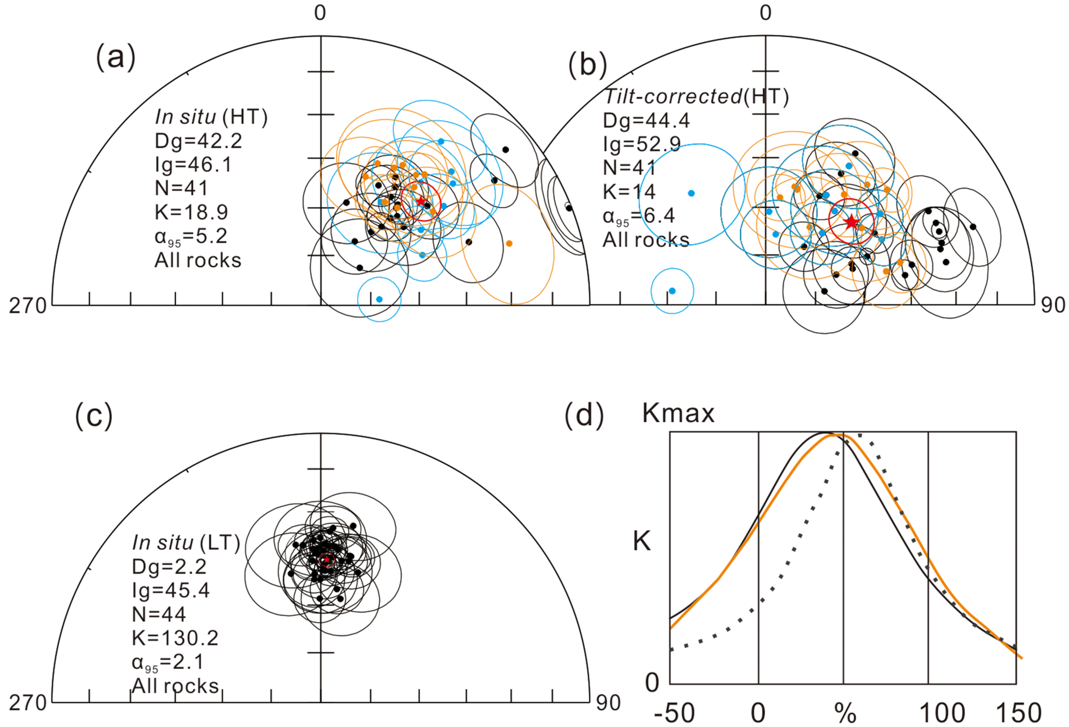

Figure 3. ChRM directions in (a) geographic and (b) stratigraphic coordinates. Black circles = reservoir rock; blue circles = source rock; brown circles = oil-free rock; red star = mean directions (c) Low-temperature components shown in geographic coordinates. (d) Fold test results. Optimum grouping of the data occurs at 38 ± 13%, 58 ± 13%, and 43.4 ± 40% unfolding for all samples (black solid line), Group A samples (dotted line) and Group C samples (brown solid line), respectively. Abbreviations: Dg, Ig, Ds, and Is = declination and inclination in geographic and stratigraphic coordinates, respectively; α_{95} = 95% confidence cone; K the precision parameter, K_{max} = maximum value of the precision parameter and N the number of specimens used to determine the statistics.

In a magnetically shielded room, samples were demagnetized through heating and alternating field demagnetization and then measured using a 2G Enterprises cryogenic rock magnetometer. To determine the magnetic carriers, isothermal remanent magnetization acquisition and demagnetization experiments were also conducted. To further characterize the magnetic carriers and diagenetic processes, fresh fragments and polished sections were prepared for scanning electron microscopy (SEM) analysis. To constrain the nature of fluids in the reservoir, seven Group A samples were selected for strontium (Sr) isotopic analysis. To quantify hydrocarbon presence and migration, we measured the total organic carbon (TOC) and extractable organic matter (EOM). To assess the role remagnetization and smectite-illite transitions, we conducted X-ray diffraction analysis.

4. Results

4.1. Paleomagnetic Results

The samples were demagnetized and their ChRM directions determined (see Table S1 in the supporting information). Demagnetization of most samples from Group A revealed a clear ChRM direction (mean value: Dg = 44.3°, Ig = 46.6°, and α_{95} = 7.9°; Figures 2, 3, and S1). To test for the age of the ChRM relative to the timing of folding, we applied the direction-correction fold test (Enkin, 2003). This yielded a maximum concentration of ChRM directions at 58 ± 13 % unfolding for reservoir rocks (Figures 3 and S2), with northeasterly declinations and moderate-steep upward inclinations. The fold test is indeterminate, which suggests a synfolding acquisition. Approximately 55% of shale samples from Group B and 79% of the Group C samples yield statistically indistinguishable ChRM directions compared to the reservoir rocks (Group B: Dg = 46.0°, Inc = 46.5°, and α_{95} = 10°; Group C: Dg = 38.0°, Inc = 44.3°, and α_{95} = 8°; Figure 3), indicating that the ChRM acquisition was coeval. The fold test for oil free samples yielded a maximum concentration of

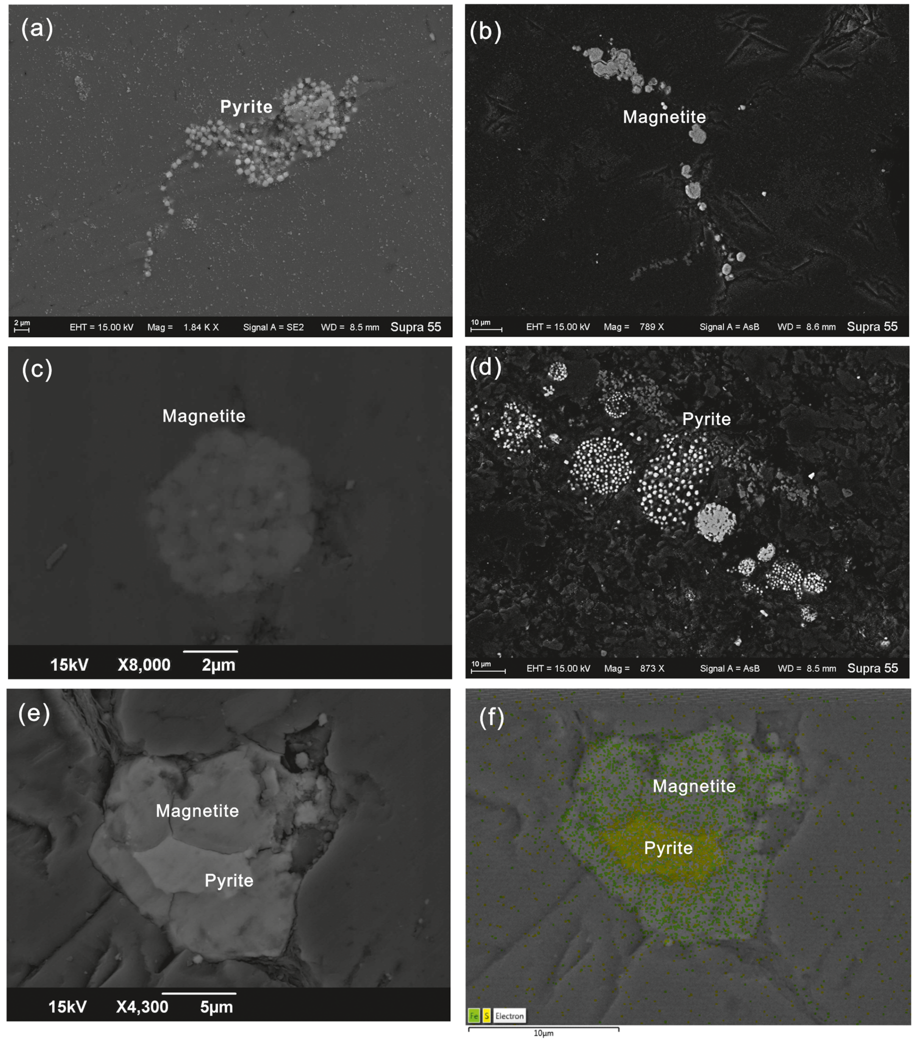

Figure 4. (a, b) Disseminated pyrite and magnetite grains distributed along linear features in fractures and pores. (c, d) Framboidal clusters of pyrite and magnetite in source and reservoir rocks. (e) Pyrite grains rimmed by magnetite, with residual pyrite preserved in the cores of the grains. (f) energy-dispersive spectroscopy map of the same area presented in Figure 4e illustrating the distribution of S (yellow) and Fe (green).

ChRM directions at 42.9 ± 40% unfolding, which may also suggest a synfolding acquisition. The Group B samples were only exposed at the eastern limb of two folds, so we did not perform fold tests on this groups' data.

4.2. The Results of Rock Magnetism and SEM

Isothermal remanent magnetization and its thermal demagnetization analyses indicate that magnetite is the principal carrier of the ChRM (see supporting information Figures S3 and S4). These findings are supported by SEM observations, which identified fine-grained iron oxides and pyrite. Iron oxides partly occur as disseminated grains (<10 μm in diameter) along fractures and pores, with part of grains occurring as framboidal clusters (Figures 4 and S5). We also frequently observed pyrite grains with iron oxides rims within fractures and pores (Figures 4 and S6).

4.3. Sr Isotope Results

The reservoir samples are characterized by $^{87}Sr/^{86}Sr$ ratios that range from 0.708977 to 0.713530, which are radiogenic relative to the Sr curves of seawater in the Early Ordovician (~0.7088) and middle Silurian (~0.7081; McArthur et al., 2001; Figure S7). The $^{87}Sr/^{86}Sr$ ratios of the Ordovician samples exhibit

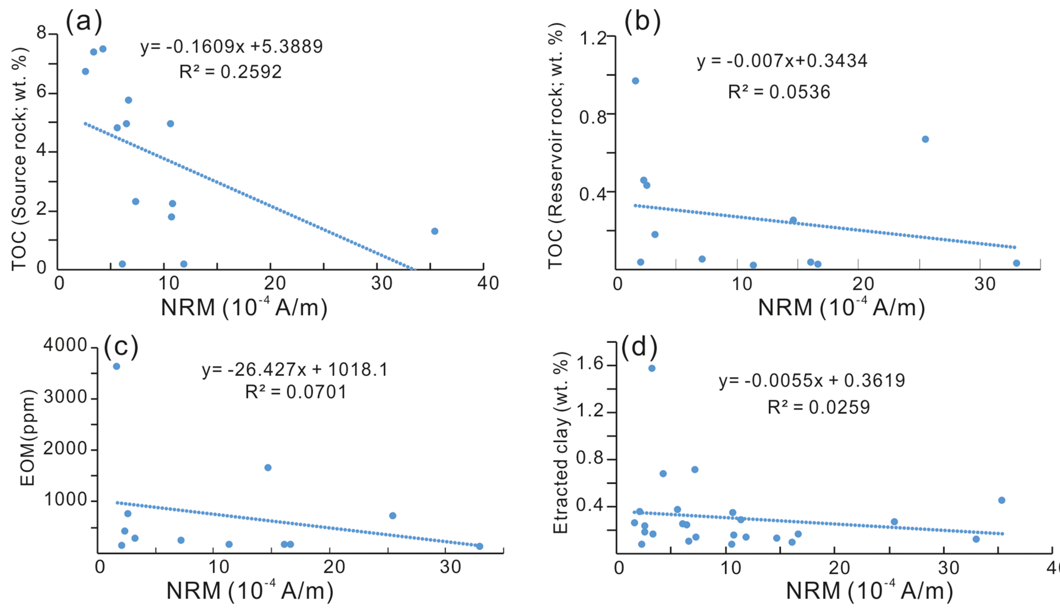

Figure 5. Binary diagram illustrating the relationship between TOC and NRM for (a) source rock and (b) reservoir rocks, (c) between the EOM and NRM for reservoir rocks, (d) between the extracted clay and NRM for representative reservoir and source rocks. EOM = extractable organic matter; NRM = natural remanent magnetization; TOC = total organic carbon.

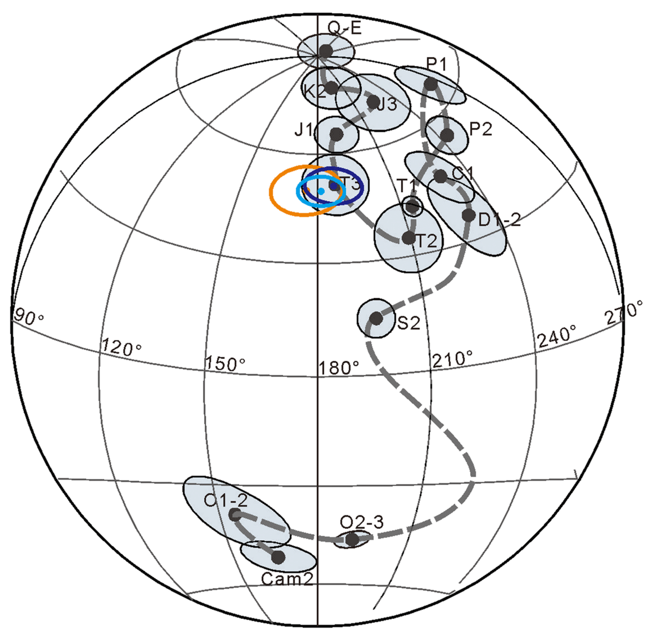

Figure 6. ChRM poles plotted with the apparent polar wander path for the south China Block. The blue oval represents the ChRM direction at K_{max}, the orange oval is the fold-corrected direction, and the purple oval represents the in situ direction.

significant scatter despite the small analytical uncertainties of the data, indicating that these rocks were altered to different degrees, by radiogenic, chemically active fluids.

4.4. The Results of TOC and EOM

A subset of 25 representative source and reservoir rock samples were selected for TOC measurement from those samples with obvious ChRM directions (Table S2). The source rock samples exhibit higher TOC values, ranging from 0.20 to 7.53 wt.%, while reservoir rocks ranging from 0.02 to 0.97 wt.%. A weak negative correlation exists between TOC and natural remanent magnetization (NRM; reflecting the magnetite concentration), with R^2 (coefficient of determination) values of 0.25 for source rock samples and 0.05 for reservoir rock samples (Figure 5). EOM was determined for 14 representative samples (including 12 reservoir rock samples) containing obvious ChRM directions, and the values ranged from 134.1 to 3,640 ppm. The correlation between the EOM and NRM is very weak with R^2 at 0.07 (Figure 5).

4.5. The Clay Analysis

Clay minerals were extracted from insoluble residue that remained after dissolution of source and reservoir rocks in weak acid and hydrogen peroxide. The proportion of insoluble residue containing clay ranged from 69.1 to 94.4 wt.% for source rocks, 3.8 to 5.9 wt.% for reservoir limestone rocks, and 23.7 to 94.3 wt.% for reservoir sandy limestone rocks (Table S2). The proportion of extracted clay (3.3–53.2 mg) also varied (0.076 to 1.57 wt.%) and exhibited no correlation with NRM ($R^2 = 0.0259$; Figure 5). Based on the X-ray diffraction analyses on the extracted clay, the clay minerals largely comprise chlorite and illite, with no smectite for all source and reservoir rocks (Figure S8).

5. Discussion and Conclusion

The reservoir, source and oil-free rocks all carry ChRMs, which are indistinguishable at a statistical level (Figure 3 and Table S1). The mean pole position (40.9°N, 181.1°E) determined for reservoir and source rocks in Majiang paleoreservoir plots close to the Late Triassic mean pole of the apparent polar wander path for south China (Yang & Besse, 2001; Zhang et al., 2015; Figure 6). This postdates the early Paleozoic depositional age of samples, indicating that the ChRM is a secondary remagnetization. We identify the ChRM as chemical remagnetization, because (1) the unblocking temperatures (up to 500 °C) are too high to be thermoviscous in origin and (2) SEM observations (Figure 4) identified fluid flow features along fractures, which is indicative of chemical remagnetization events (Elmore et al., 2012).

We conclude that the action of orogenic fluids represents a viable remagnetization mechanism for hydrocarbon-bearing strata in the Majiang paleoreservoir. The following three observations lead us to this conclusion: First, this is supported by the apparent universality of the CRM acquisition; for example, the CRM was found to be recorded in black shale with rich organic matter and clay, hydrocarbon-bearing limestone, oil-free tillite, carbonate-bearing rocks, and mudstone (Figure 1c). In the source rocks, the fluids could not have been produced during the maturation of organic matter, as there is no positive correlation between source rock TOC and NRM intensity (reflecting the magnetite concentration; Manning & Elmore, 2015; Zhang et al., 2016). The TOC and EOM values for reservoir rocks also show very weak correlations with NRM intensity, which suggests that the hydrocarbon migration is not the mechanism for CRM acquisition (Zhang et al., 2016). The lack of correlation between the proportion of extracted clay and NRM intensity for both reservoir and source rocks probably excludes clay diagenesis as the main mechanism for remagnetization. This conclusion is furtherly confirmed by the SEM observations, which indicates that the authigenic magnetite is likely formed from the altered pyrite (Figure 4; Suk et al., 1990; Brothers et al., 1996; Fruit et al., 1995) and not from a smectite-illite transition (Katz et al., 1998; Lu et al., 1991; Moreau et al., 2005). Second, Sr isotopes from reservoir rocks indicate that the altered fluids were chemically evolved, which is the general feature of orogenic fluids (Elmore et al., 1993; O'Brien et al., 2007; Zechmeister et al., 2012). Thirdly, the Late Triassic syntilting remagnetization was temporally and spatially related to the Indosinian orogeny, given that the paleoreservoir was located in the foreland basin belts.

The Majiang paleoreservoir has undergone Caledonian, Indosinian, and Yanshanian tectonic events (Ge et al., 2016; Wu, 1989). The Caledonian period has generally been accepted as the time of hydrocarbon generation and accumulation (Ge et al., 2016; Han et al., 1982; Tang & Cui, 2011; Wu, 1989), though the precise timing is controversial. During the Indosinian orogeny, our data suggest that source and reservoir rocks were strongly altered by large-scale orogenic rather than hydrocarbon fluids, indicating that the reservoir was hydrodynamically flushed and water washed, both of which are common destructive processes in foreland basins and complex tectonic zones (Dahlberg, 1995; Macgregor, 1996). Additionally, the syntilting acquisition of remagnetization indicates that new deformations of the reservoir (e.g., folds, faults, and even uplift) occurred during the alteration of orogenic fluids, and the reservoir could have been further destroyed by these deformations during the orogeny, for example, leakages, remigrations (Dahlberg, 1995; Macgregor, 1996). Therefore, it is concluded that the Late Triassic Indosinian tectonic event could have yielded an important destruction of the Majiang reservoir. During the Yanshan orogeny, it was suggested that the tectonic events led to the uplift and near-surface exposure of the reservoir, which could have completely destroyed the Majiang reservoir (Ge et al., 2016).

On a global scale, most pre-Cenozoic reservoirs have been destroyed, and destructive processes have only been documented in a limited number of well-drilled basins (Macgregor, 1996). In this study, the presented link between the chemical remagnetization and destruction of hydrocarbon reservoir provides new insights for the identification and timing of destructive processes in hydrocarbon reservoir systems. Given that most destructive processes are thought to involve the action of fluids that also lead to the formation of authigenic magnetic minerals, this provides a protocol for constraining destructive processes in future studies, by combining clear sampling strategies with paleomagnetism and other multidisciplinary techniques.

Previously paleomagnetic methods have been used to date maturation of organic matter (or hydrocarbon generation) in source rocks and hydrocarbon migration and accumulation in reservoir rocks. Our paleomagnetic dates from the Majiang paleoreservoir has not recorded hydrocarbon generation and migration but rather hydrocarbon destruction. With this in mind, we suggest that future paleomagnetic studies need to demonstrate a clear link between the hydrocarbon generation or migration processes and remagnetization in hydrocarbon-bearing strata, if the palaeomagnetic data are to be correctly interpreted.

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