Origin and Distribution of Large Asphaltite in South China
Guangyou Zhu,* Meng Wang, Jingfei Li, Kun Zhao, Chao Li, Zhiyong Chen, Lei Zhou, and Zhenghui Wu

ABSTRACT: Large quantities of Triassic solid asphaltite were discovered in the Guangyuan area, northwest Sichuan. The asphaltite is formed in layers with a vertical thickness between 0.3 and 2.8 m and is stably distributed with intrusive contact with surrounding rocks. This study aims to investigate the genesis and distribution of asphaltite through trace element, biomarker, and Re-Os isotope analyses. Trace element analysis shows the enrichment of V and Cr in the asphaltite, indicating that it is derived from relatively deep hydrocarbon sources. The carbon isotope and biomarker results suggest that the asphaltite originates from Cambrian paleo reservoir. The Re-Os isotope analysis determines a formation age of 220 ± 6 Ma, which corresponds to the late Triassic, indicating the cracking of paleo reservoirs in late Triassic. Therefore, the origin of asphaltite is epigenetic-reservoir asphaltite. The generation of oil from Cambrian source rocks began at the end of Silurian and ended after Caledonian orogeny. At the end of Permian, the fracture system was well developed due to the influence of the Hercynian movement, which provided favorable conditions for the migration of Cambrian oil. By the end of Triassic, hydrocarbons generated from Cambrian source rocks were mainly distributed in fractures and reservoirs, thus forming paleo oil reservoirs. Afterward, the paleo reservoirs were adjusted to the surface or near the surface during the Indosinian movement and thus have cracked into asphaltite. The distribution of asphaltite is closely related to the tectonic activities, and the asphaltite is preferentially stored in the anticline axes, fissures, and some interlayer fracture zones.

1. INTRODUCTION
Asphaltite is crude oil produced from source rocks that has evolved and deposited hundreds of millions of years and in contact with air and water. Combined action of thermal cracking, oxidation, and bacteria caused the removal of water and light oil, while heavy hydrocarbons were retained that formed solid organic matter with stable properties. According to the morphology, appearance, color, and formation stage of asphaltite, asphaltite can be divided into four categories, namely primary-same-layer asphaltite, epigenetic-reservoir asphaltite, magmatic metamorphic asphaltite, and superficial-shallow oxidation and degassing asphaltite.

Asphaltite is the paleo reservoir after the geological structure of the strong destruction. The crude oil in the paleo oil reservoir was squeezed into the fracture or remained in the reservoir, and after thermal cracking and oxidation, it became veins or solid bitumen, and then enriched. As a kind of non-metallic mineral resources, asphaltite has a considerable utilization value. As a derivative of petroleum, asphaltite produced by it can be used as fossil fuel. In addition, it can also be used as a modified material to improve the strength of ordinary petroleum asphaltite and the life of roads, which is favored by all countries in the world.

Asphaltites are widely distributed around the world and are found in different layers. Bitumen deposits have been discovered in the Lower Cretaceous and the Late Triassic in Qingchuan County, Guangyuan, Sichuan in Karamay Wuerhe areas, Xinjiang, China. In addition, asphaltites have also been discovered in the United States in Utah, Lake Trada in Venezuela, Gard in France, Chatel in Val de Traverse in Switzerland, and Bouton Island in southeastern Indonesia. The source and origin of many large asphaltite reservoirs...
worldwide have been proposed through geochemical and geological analyses, and they can either form large accumulations or even form tars that can serve as local seals. Recently, in the course of field exploration in the Guangyuan area, several sets of high-quality asphaltites were found, which were produced in layers with a vertical thickness between 0.3 and 2.8 m, stable distribution, and intrusive contact with surrounding rocks, indicating that large-scale reservoir cracking had occurred in the Guangyuan area. The current research on asphaltite mainly uses elements, solid carbon isotopes, saturated hydrocarbon chromatography, biomarker compounds, and aromatic hydrocarbon chromatography–mass spectrometry biomarker extraction to determine the origin of asphaltite. However, the accumulation process and destruction mechanism of paleo oil reservoirs indicated by asphaltite mines are still unclear. In this paper, the relationship between asphaltite formation and reservoir destruction is discussed by studying the age of rhenium osmium and trace elements in asphaltite.

2. GEOLOGICAL SETTINGS

The research area is located in the eastern part of the Longmen Mountain orogenic belt in the Sichuan Basin and is administratively affiliated to Guangyuan City, Sichuan Province. To the north is the Qinling fold belt, and to the east is the Songpanganzi fold belt. The mining site is located in the Indosinian fold belt with NE–SW strike. The Triassic and Jurassic strata are mainly exposed in the area, which is a set of shallow sea inland lake facies, and the lithology is a marl glutenite combination. The study area experienced six tectonic cycles. They are the Yangtze cycle, the Caledonian cycle, the Hercynian cycle, the Indochina cycle, the Yanshan cycle, and the Himalayan cycle, respectively.

The strata exposed in the mining area are mainly Cenozoic-Paleozoic, Silurian Longmaxi Formation, Carboniferous Huanglong Formation, Triassic Feixianguan Formation, Jialingjiang Formation, Leikoupo Formation and Xujiahe Formation, Jurassic Arliujing Formation, Shaximiao Formation, Suining Formation, and Penglai Formation, the fourth series sporadic emerged (Figure 1). The middle and lower parts of Lower Triassic are gray-purple and light-purple limestone with siltstone, gray limestone, dolomite limestone, and oolitic limestone interbedded with a large amount of solid asphaltite. Asphaltite veins are mainly distributed along faults and fractures and are produced in layers. The vertical thickness is between 0.3 and 2.8 m. The distribution is stable. It is in contact with the surrounding rocks and is the product of cracking of the reservoir. The upper part of the Jurassic is an interlayer of fine sandstone and mudstone, and solid asphaltite is also seen. Asphaltite intrudes into the reservoir along the cracks between the layers, forming an asphaltite layer up to a few centimeters thick.

3. SAMPLES AND METHODS

The asphaltite samples were collected from an outcrop near the Guangyuan city in the northeastern Sichuan (SW China) (Figure 2). They are in a layered form and show a certain directional distribution and have intrusive contact with the surrounding siltstone layers of the Triassic Xujiahe Formation. A total of nine fresh and pollution-free asphaltite samples were collected for further experiments, among which seven were tested for Re-Os isotope and the other two for trace elements compositions.
The asphaltite in the mining area is brittle asphaltite, which is dark brown-black, dense, and massive, with a rough shell-like fracture surface. The luster changes from dim to bright, with different hardness and light specific gravity.

3.1. Microelement Analysis. Elemental analyses for major, trace, and rare earth elements were carried out using inductively coupled plasma-mass spectrometer (ICP-MS), which is equipped with a Finnigan MAT Element I mass spectrometer (San Jose, California) at the Beijing Institute of Nuclear Industrial Geology. About 40 mg powder of each sample was digested with 0.5 mL diluted HNO3 (i.e., analytically pure HNO3 mixed with water at a volume ratio of 1:1) and 1 mL HF in a Teflon digestion vessel, and then processed by ultrasonic vibration for 10–15 min. The Teflon digestion vessel was then placed on an electrical heating plate at about 150 °C (302 °F) for 7 to 10 days to remove all silica. After that, the residue was mixed with 2.0 mL of HNO3 until it was completely dissolved using the ICP-MS.

3.2. Biomarkers Analysis. The biomarker analyses were performed at Petroleum Geology Laboratory, China University of Petroleum, Dongying, China. All of the glass vessels used for bitumen extraction were combusted at 700 °C in a muffle furnace to remove any organics and ultrasonically washed with purified water. Following this, 200 g of asphaltite powder was extracted by Soxhlet extraction with chloroform for 8 h. The extracts (chloroform bitumen “A”) were collected in beakers (100 mL) and covered with aluminum foil, until the chloroform was completely evaporated in a fume hood. Then, 25 mg of bitumen “A” was dissolved in an appropriate amount of n-hexane, and D10-anthracene, 5α-androstanone, and C24-D50 as internal standards were added. Approximately 12 h later, the asphaltite precipitate was separated by filtration, and the filtrate was performed using a Thermo Scientific TRACE GC Ultra-DSQ II mass spectrometer. The filtrate was divided into saturated hydrocarbon, aromatic hydrocarbon, and polar fractions subsequently using 10 mL hexane and 20 mL dichloromethane: n-hexane (2: 1), respectively, as elute in a silica gel glass column (200−260 mesh, activated at 200 °C for 4 h). The GC−MS analyses of the hydrocarbon fractions were performed using a Thermo Scientific TRACE GC Ultra-DSQ II mass spectrometer.

A HP-5 chromatographic column was used to separate the saturated and aromatic hydrocarbon fractions. The oven temperature was initially set at 70 °C for 5 min, programmed to reach 220 °C at 4 °C/min increments, followed by 2 °C/min to 320 °C held for 20 min for the saturated hydrocarbons, and programmed from 70 °C for 5 min to reach 320 °C at 3 °C/min increments, then isothermal for 20 min for the aromatic hydrocarbons. Helium was used as a carrier gas with a constant flow rate of 1 mL/min. Both interface temperature and injection temperatures were 300 °C. The transfer line temperature was 250 °C, and the ion source temperature was 230 °C. The ion source was operated in the electron impact mode at 70 eV, and selected ion monitoring was performed.

3.3. Re-Os Isotope Analysis. First, the rock samples wrapped in the sample bag were crushed with a geological hammer, and the fresh bitumen samples were selected with toothpicks. Then, the samples were wrapped with disposable filter paper and crushed to 60−80 mesh with a geological hammer. Last, seven pieces of asphaltite samples were selected for the Re-Os isotope test.

The weighed sample is loaded in a Carius tube through a thin-neck-long funnel. After the mixture diluted, 183Re-180Os was transferred to the Carius tube frozen with liquid nitrogen with 3 mL 15 mol/L HCl, 5 mL 15 mol/L HNO3, and 1 mL 30% H2O2 were successively added, and the Carius tube was sealed with liquefied petroleum gas and oxygen flame, and heated for 24 h under the condition of 230 °C in the oven. The Os is separated by the method of direct distillation from the Carius tube for 50 min trapped in 5 mL 1:1 HBr and the method of micro distillation that is used for N-TIMS (Triton) determination of Os isotope ratio. Re was separated and enriched by acetone extraction, evaporate to dryness, and picked up in 2% HNO3 that is used for the N-TIMS determination of the Re isotope ratio.

4. RESULTS

4.1. Trace Elements of Asphaltite. Trace elements concentrations of Triassic solid asphaltite samples from the Guanyuan area are shown in the table; element V (105 to 109 μg/g), Cr (119 to 125 μg/g), and Ba (161 to 171 μg/g) were obviously enriched in the asphaltite sample, and the average concentration of all three trace elements exceeded 100 μg/g (Table 1).

| Table 1. Trace Element Concentrations (μg/g) and Related Parameters of Solid Bitumen Samples |
|---------------------------------------------|----------------|
| elements | sample 1 | sample 2 | elements | sample 1 | sample 2 |
|--------|---------|---------|--------|---------|---------|
| Li     | 40.3    | 44.4    | Ag     | 0.09    | 0.08    |
| Be     | 6.87    | 7.39    | Cd     | 1.14    | 1.21    |
| Sc     | 23.4    | 24.6    | In     | 0.09    | 0.09    |
| Ti     | 4652    | 4818    | Sn     | 2.19    | 2.32    |
| V      | 105     | 109     | Sb     | 1.72    | 1.82    |
| Cr     | 119     | 125     | Cs     | 3.05    | 3.27    |
| Mn     | 28.5    | 32.4    | Ba     | 161     | 171     |
| Co     | 5.62    | 6.46    | La     | 61.4    | 65.0    |
| Ni     | 21.8    | 23.1    | Ce     | 130     | 139     |
| Cu     | 71.0    | 75.7    | Pr     | 14.6    | 15.5    |
| Zn     | 60.0    | 58.7    | Nd     | 57.1    | 60.8    |
| Ga     | 14.6    | 15.4    | Sm     | 13.5    | 14.5    |
| Ge     | 3.95    | 4.02    | Eu     | 3.00    | 3.21    |
| As     | 3.48    | 3.24    | Gd     | 15.3    | 16.2    |
| Rb     | 36.4    | 39.7    | Tb     | 2.36    | 2.49    |
| Sr     | 205     | 219     | Dy     | 14.5    | 15.0    |
| Y      | 96.0    | 101     | Ho     | 2.83    | 2.95    |
| Zr     | 258     | 260     | Er     | 7.81    | 8.21    |
| Nb     | 17.7    | 18.5    | Tm     | 1.03    | 1.10    |
| Mo     | 2.93    | 2.84    | Yb     | 6.45    | 6.87    |
| Lu     | 0.95    | 1.00    | W      | 11.1    | 19.5    |
| Hf     | 6.31    | 6.52    | Ti     | 0.42    | 0.40    |
| Ta     | 1.81    | 2.05    | Pb     | 45.2    | 46.8    |
| Bi     | 2.31    | 2.18    | Th     | 31.8    | 33.5    |
| U      | 14.0    | 14.5    |

Metal elements are generally combined with organic compounds and thus accumulated during sedimentation. With the increase of burial depth and temperature, organic matter undergoes pyrolysis—polymerization reaction under heat. For example, some active metals such as V, Ni, Mo, Hg, Sb, U, etc. are transformed, and with the migration and leaching of organic matter, the other part remains in the source rock in the form of sulfide or adsorption on the surface of sulfide. Therefore, in the initial hydrocarbon generation stage, the chemical properties of the elements determine the initial abundance of each element in crude oil. Active elements
are transferred from the source rock to oil during the maturation process while the inactive ones are preferentially enriched in the source rock and less likely transferred by migration or leaching.17

V is the trace element with the highest abundance in crude oil and asphaltite.15,16 It is well known that V occurs mainly in the form of metalloporphyrin and non-porphyrin complexes in crude oil and asphaltite.17 Compared with other organic metal complexes, porphyrin compounds of V have higher thermal stability.19 In the process of oil generation, migration, and degradation, V will be accumulated very stably in crude oil or asphaltite. In addition, the enrichment of V in crude oil and asphaltite also reflects the enrichment of V in the organic matter of source rocks. The concentration of V element is higher than 100 μg/g, so it can be inferred that the source rock of asphaltite should also be enriched with V. The enrichment of Ba element may be related to the symbiotic barite in this area.

In asphaltite, the siderophile elements (Ti, V, Cr, Mn, Fe, Co, and Ni), which are closely related to the deep matter are enriched. This combination of elements reflects the close relationship between asphaltite and deep source fluids. It shows that the formation of asphaltite may be related to the deep source fluid.

4.2. Asphaltite Isotope. The asphaltite samples are distinctly characterized with isotopically light carbon isotopes. The δ13C value of asphaltite A is −35.8‰, while the δ13C value of each group is −36.1‰ to −34.3‰ (Table 2). The study found that the oil reservoir is correlated with the crude δ13C, which indicates that the older the formation age is, the lower the δ13C value is.20 By comparison with δ13C of the same time or adjacent times, the δ13C values of lower Cambrian and upper Ordovician-lower Silurian in Moxi area of the Sichuan Basin are −36.8‰ to −31.2‰ and −31‰ to −31.2‰.21 The Cambrian is in a reducing environment. At that time, the atmospheric oxygen content was very low, only 1% of the current atmospheric content.21,22 Meanwhile, prokaryotes and cyanobacteria in the ocean will preferentially absorb 12C in CO2 during photosynthesis, resulting in 12C enrichment.23 The low δ13C value of asphaltite should reflect the crude oil derived from lower Cambrian.

4.3. Asphaltite Biomarker. Previous studies have found that steranes and terpenes are not detected in Cambrian oil source rocks in some areas and the reason is inconclusive.23 The distribution of steranes and terpenes in the asphaltite veins was normal. As shown in Figure 3, the pregnane (C29 and C31) constitutes two strong peaks, therefore the peaks of regular sterane are suppressed lower, indicating that the thermal evolution of the asphaltite is quite severe. The regular sterane series of the asphaltite show roughly “V” shape distribution with the contents of each regular sterane as: C27 accounts for 35%, C28 accounts for 30%, and C29 accounts for 45%. The considerable content of C28 sterane indicates that blue-green algae conquered land and evolved to terrestrial higher plants, similar sterane distribution has been reported in Neoproterozoic and early Cambrian sediments from the Centralian Superbasin (Australia)27 and Cambrian oil/sources in the Tarim Basin (China).29 The content of C28 regular sterane is not very low, indicating that it is related to algae organisms.28 In addition, the relatively low content of rearranged sterane and the low ratio of rearranged/normal sterane of 0.15 jointly indicate that the environment was partially reductive at the time.29,30

Generally speaking, saturated hydrocarbons are characterized by low maturity period, while aromatic hydrocarbons reflect the characteristics of high maturity period. As a marker of eukaryotic input, monoaromatic steranes have more parent characteristics than steranes,23 and the distribution of C27–C28–C29 monoaromatic steranes in aromatic fractions does not change significantly in the whole oil window.23 The Cambrian source rocks are characterized with a predominance of C28 triaryl sterane and a high content of C29 sterane as suggested by previous studies on Cambrian marine source rocks in the Tarim Basin.29 The solid asphaltites present have a high content of C28 triaryl sterane, and C29 sterane and the ratio of C26/27/C28 triaryl sterane value is 0.6, indicating the typical characteristics of Cambrian marine sources.

According to the analysis of trace elements, isotopes, and biomarkers, it is believed that the asphaltite originated from the Cambrian paleo reservoir and distributed in the Triassic strata. As a result of tectonic activities, asphaltite is moved upward by deep materials and deposits and forms minerals in fractures and reservoirs. Therefore, it can be judged that the asphaltite is epigenetic-reservoir asphaltite.

5. DISCUSSION

5.1. Re-Os Isotope. Re and Os isotope test analysis of asphaltite samples are shown in the following table (Table 3). It can be seen from the table that the Re and Os content in the asphaltite sample is low, the Re content is among 484.2 to 503.8 μg/g, the Os content is between 0.3705 to 0.5628 μg/g, the ratio of 187Re/188Os is among 4324 to 5885, and the ratio of 187Os/188Os is among 17.06 to 24.34. The ratio of Re and Os and the content of asphaltite in this study are similar to those of asphaltite samples reported by predecessors,29,31 and the same isotopic composition has been obtained for the determination of repeated samples of the same sample. Therefore, the data obtained in this study are reliable.

The Re and Os isotopic composition of asphaltite is different from that of the crust due to its high Re and Os content (crust Re < 1 to 2 μg/g, Os < 50 pg/g).29 This indicates that these elements are most likely to come from sediments, which are

Table 2. Family Compositions and Carbon Isotopes Compositions of Bitumen Veins (data from ref 23)

| compositions | bitumen A | saturate | aromatic | nonhydrocarbon | asphaltene |
|--------------|-----------|----------|----------|----------------|------------|
| family compositions/% | 6.7667 | 0.7 | 2.66 | 35.04 | 61.6 |
| carbon-isotope compositions/‰ | −35.8 | −34.3 | −36.1 | −34.9 | −36 |

![Mass chromatogram of steranes (m/z 217)](image) of asphaltite in Guangyuan.
Previous studies have suggested that in the process of oil generation, Re and Os in hydrocarbons such as black shale will inevitably partly enter the generated hydrocarbons through the hydrocarbon expulsion process, and the hydrocarbon generation and migration process are the main driving force for the homogenization of the oil/gas ratio. Ravizza et al. (1989) pointed out that most trace metals in oil are closely related to the functional groups of asphaltite components. Therefore, this study believes that Re and Os and other metal elements are discharged from the source rock through the hydrocarbon expulsion of the source rock. After the discharge, Re and Os and the generated hydrocarbon materials travel through the reservoir for a long distance. Then, due to the adjustment and destruction of multiple tectonic movements, the light components in the crude oil were eventually dispersed, and the asphaltite and heavy components remained, thereby forming the asphaltite seen today. The Re and Os isotopes may be reset. At the same time, the presence of organic matter leads to a certain degree of fractionation of the isotopes.34

Selby et al. (2005) performed Re-Os isotope dating on the asphaltite associated with the Nunavut Polaris Mississippi Valley (MVT) lead–zinc deposit in Canada to obtain a Re-Os isochron age of 374.2 Ma ± 8.6 Ma. The Rb-Sr dating and paleo-magnetic dating have good consistency within the error limit, confirming the possibility of asphaltite Re-Os isotope dating and interpreting its age significance as the time when oil and gas were damaged.

The asphaltite Re-Os isotope isochron obtained in this experiment has important geological significance and meets the conditions for the construction of an isochron. The formation of a large amount of solid asphaltite can be used as a sign of the destruction of ancient oil and gas reservoirs. According to the Re-Os isotope analysis of asphaltite in this study, it is believed that the obtained isochron represents the age of asphaltite formation and corresponds to the destruction time of the paleo reservoir, that is, it was destroyed in the late Triassic around 220.7 ± 6.1 Ma.

5.2. The Geological Process of Asphaltite Formation.

The Cambrian source rocks began to generate oil at the end of Silurian, and after Caledonian uplift and erosion, the hydrocarbon generation ended. At the end of Permian, due to the influence of the Hercynian movement, the Longmenshan fold uplifted and developed to the southeast, forming the fold belt of the eastern margin. Some faults that are basically consistent with the direction of the main axis are formed, and a good fracture system is developed, which provides convenient conditions for the migration of Cambrian crude oil. By the end of Triassic, Cambrian source rocks started secondary generation and entered the peak of oil generation at the late Triassic. The generated hydrocarbons were mainly distributed in fractures and reservoirs, forming paleo oil reservoirs. After Indosinian movement, the crust changed from an early extension movement to a squeezing
movement, which led to the uplift of the basin and the migration of the paleo reservoir to the surface or near the surface.

5.3. Prediction of Asphaltite Distribution. Sichuan Basin has developed five sets of source rocks, namely early Cambrian, late Ordovician-early Silurian, and middle Permian and late Permian, of which there are two sets of regional quality mud source rocks (lower Cambrian, upper Ordovician-lower Silurian) mainly composed of shale and mudstone. Middle Permian source rocks are carbonate rocks, and the upper Permian source rocks Rock are coal strata. The distribution of source rocks is well developed, which provides a good physical basis for the formation of asphaltite. The presence of asphaltite can be found in many strata in the Guangyuan area (Figure 5).

The distribution of asphaltite is closely related to the structure. The ore body is basically distributed along the fault. The asphaltite in the ore has a flowing structure. After Hercynian movement, a large number of faults and fissures were formed in the study area. Oil and gas along the faults and fractures, such as oxidation mineralization in air and water are stored in the anticline axis, fissure, and some of the formation of the interlayer fracture zone.

The destruction of paleo reservoirs is marked by the formation of a large amount of solid asphaltite. According to the Re-Os isotope analysis of asphaltite in this study, it is believed that the obtained isochron represents the age of asphaltite formation and corresponds to the destruction time of the paleo oil reservoir. It was destroyed in late Triassic around 220.7 ± 6.1 Ma.

6. CONCLUSIONS

(1) The enrichment of V and Cr in the trace elements of asphaltite indicates that the formation of asphaltite may be related to relatively deep hydrocarbon sources.

(2) The asphaltite veins are well developed and widely distributed in the Guangyuan area. They were originated from Cambrian paleo reservoir, distributed in Lower Paleozoic strata, controlled by the fault and fracture system formed during tectonic activities in Hercynian orogeny.

(3) This study suggested that the Re-Os isochron age of 220.7 ± 6.1 Ma indicates the formation time of the asphaltite, representing the time when the paleo oil reservoir was destructed, and the last time when the bitumen Re-Os isotope system was subjected to thermal transformation as well.

AUTHOR INFORMATION

Corresponding Author
Guangyou Zhu — Research Institute of Petroleum Exploration and Development, PetroChina, Beijing 100083, China; orcid.org/0000-0002-7282-6990; Phone: +86 18601309981; Email: zhuguangyou@petrochina.com.cn

Authors
Meng Wang — Research Institute of Petroleum Exploration and Development, PetroChina, Beijing 100083, China
orcid.org/0000-0002-7282-6990
Jingfei Li — Research Institute of Petroleum Exploration and Development, PetroChina, Beijing 100083, China
Kun Zhao — Research Institute of Petroleum Exploration and Development, PetroChina, Beijing 100083, China
Chao Li — National Research Center for Geoanalysis, Chinese Academy of Geological Sciences, Beijing 100037, China
Zhiyong Chen — Research Institute of Petroleum Exploration and Development, PetroChina, Beijing 100083, China
Lei Zhou — Research Institute of Petroleum Exploration and Development, PetroChina, Beijing 100083, China
Zhenghui Wu — Research Institute of Petroleum Exploration and Development, PetroChina, Beijing 100083, China
ACKNOWLEDGMENTS

This study is supported by China National Petroleum Corporation (CNPC) Scientific Research and Technology Development Project (grant no. 2019B-04 and 2018A-0102). We thank Professor Shi Quan from China University of Petroleum (Beijing), Professor Wang Lanheng from Petro-China Southwest Oil and Gas Field Company, and Professor Liang Digang from PetroChina Exploration and Development Research Institute for their guidance and help.

REFERENCES

(1) Qin, J.; Fu, X.; Liu, X. Solid bitumens in the marine carbonate reservoir of gas field in the northeast area of the Sichuan Basin. *Geologica Sinica* 2007, 8, 1065−1071+1161 (in Chinese with English abstract).
(2) Zhang, L.; Wei, G.; Wu, S.; Wang, Z.; et al. Distribution characters and hydrocarbon-generating potential of bitumens of sinian-lower paleozoic in Sichuan Basin. *Petroleum Geology & Experiment 2005, 27* (3), 276−298 (in Chinese with English abstract).
(3) Wei, J. Genesis discussion of natural asphalt in Guangyuan [D]. Chengdu University of Technology, 2014. (in Chinese with English abstract).
(4) Zheng, F.; Zhu, G.; Chen, Z.; Zhao, Q.; Shi, Q. Molecular composition of vanadyl porphyrins in the gilsonite. *Journal of Fuel Chemistry and Technology 2020, 48* (5), 562−567 (in Chinese with English abstract).
(5) Gong, A.; Song, X. Evaluation of geological relics and resources of bituminous ore in Wuerhe District, Karamay City, Xinjiang Province. *Journal of Xinjiang Normal University* 2009, 28 (1), 8−12 (in Chinese with English abstract).
(6) Rao, D.; Qin, J.; Teng, G.; Zhang, M. Source analysis of oil seepage and bitumen originating from marine layer strata in guangyuan area, the northwest Sichuan Basin. *Petroleum Geology & Experiment 2008, 30* (6), 596−605 (in Chinese with English abstract).
(7) Edwards, D.; McKirdy, D. M.; Summons, R. E. Enigmatic asphalts from the southern Australian margin: molecular and carbon isotopic composition. *PESA J. 1998, 26, 106−129.*
(8) Logan, G. A.; Jones, A. T.; Kennard, J. M.; Ryan, G. J.; Rollet, N. Australian offshore natural hydrocarbon seepage studies, a review and re-evaluation. *Mar. Pet. Geol. 2010, 27, 26−45.*
(9) Hao, B.; Hu, S.; Huang, S.; Hu, J.; et al. Geochemical characteristics and its significance of reservoir bitumen of Longwangmiao Formation in Moxi Area, Sichuan Basin. *Geoscience 2016, 30* (3), 614−626 (in Chinese with English abstract).
(10) Tian, X.; Hu, G.; Li, W.; et al. Geochemical characteristics and significance of sinian reservoir bitumen in leshan-langansui paleo-uplift area, Sichuan Basin. *Natural Gas Geoscience 2013, 24* (5), 982−990 (in Chinese with English abstract).
(11) Ji, K.; Guo, S.; Li, X.; Luo, Y.; et al. The nuclear magnetic resonance (NMR) characteristics of the carbonate rock reservoir which has dissolved pores and asphalt. *Natural Gas Geoscience 2017, 28* (8), 1257−1263 (in Chinese with English abstract).
(12) Tong, C. Formation mechanism of fault-fold structure in Sichuan Basin. *Natural Gas Industry[J] 1992, 12* (5), 1−6 (in Chinese with English abstract).
(13) Liu, S.; Sun, W.; Song, J.; Deng, B.; Zhong, Y.; et al. Tectonics-controlled distribution of marine petroleum accumulations in the Sichuan Basin, China. *Earth Science Frontiers 2015, 22* (3), 146−160 (in Chinese with English abstract).
(14) Socorro, J.; Maurrasse, F. J.-M. R. Continuous Accumulation of Organic Matter-Rich Sediments Associated with Oceanic Anoxic Event 1a in the El Pujal Section, Organyá Basin, Catalunya Spain and Its Relation to Episodic Dysoxia. *Cretaceous Res. 2019, 225.*
(15) Barwise, A. J. G. Role of Nickel and Vanadium in Petroleum Classification. *Energy Fuels 1990, 4, 647−652.*
(16) Powell, T. G.; Macqueen, R. W. Precipitation of Sulfide Ores and Organic Matter. Sulfate Reactions at Pine Point, Canada. *Science 1994, 224, 63−66.*
(17) Parnell, J. Metal Enrichments in Solid Bitumens: A Review. *Miner. Deposita 1988, 23, 191−199.*
(18) Cao, J.; Wu, M.; Wang, X.; et al. Advances in research of using trace elements of crude oil in oil-source correlation. *Advances in Earth Science 2012, 27* (9), 925−936 (in Chinese with English abstract).
(19) Lewan, M. D.; Maynard, J. B. Factors Controlling Enrichment of Vanadium and Nickel in the Bitumen of Organic Sedimentary Rocks. *Geochem. Cosmochim. Acta 1982, 46, 2547−2560.*
(20) Zhang, S.; He, K.; Hu, G.; Mi, J.; Ma, Q.; Liu, K.; Tang, Y. Unique Chemical and Isotopic Characteristics and Origins of Natural Gases in the Paleozoic Marine Formations in the Sichuan Basin, SW China: Isotope Fractionation of Deep and High Mature Carbonate Reservoir Gases. *Mar. Pet. Geol. 2018, 98, 68−82.*
(21) Ren, X. *Biological History* (Vol. 2) [M]; Science Press: Beijing, 1980.
(22) Zhang, W.; Shi, X.; Jiang, G.; Tang, D.; Wang, X. Mass-Occurrence of Oncoids at the Cambrian Series 2—Series 3 Transition: Implications for Microbial Resurgence Following an Early Cambrian Extinction. *Gondwana Res. 2015, 28, 432.*
(23) Huang, D.; Wang, L. Geochemical characteristics of bituminous dike in Kuangshanliang area of the Northwestern Sichuan Basin and its significance. *Acta Petrolei Sinica 2008, 1, 23−28* (in Chinese with English abstract).
(24) Logan, G. A.; Summons, R. E.; Hayes, J. M. An Isotopic Biogeochemical Study of Neoproterozoic and Early Cambrian Sediments from the Centralian Superbasin, Australia. *Geochem. Cosmochim. Acta 1997, 61, 5391−5409.*
(25) Yu, S.; Pan, C.; Wang, J.; Jin, X.; Jiang, L.; Liu, D.; Liu, X.; Qin, J.; Qian, Y.; Ding, Y.; Chen, H. Molecular correlation of crude oils and oil components from reservoir rocks in the Tazhong and Tabei uplifts of the Tarim Basin, China. *Org. Geochem. 2011, 42, 1241−1262.*
(26) Philp, R. P.; Gilbert, T. D. Unusual Distrubution of Biological Markers in an Australian Crude Oil. *Nature 1982, 299, 245−247.*
(27) de Leeuw, J. W.; Cox, H. C.; van Graas, G.; van de Meer, F. W.; Powell, T. G.; Baas, J. M. A.; van de Graaf, B. Limited Double Bond Isomerisation and Selective Hydrogenation of Sterenes during Early Diagenesis. *Geochem. Cosmochim. Acta 1989, 53, 903−909.*
(28) Cheng, B.; Chen, Z.; Chen, T.; Yang, C.; Wang, T-G. Biomarker Signatures of the Ediacaran – Early Cambrian Origin Petroleum from the Central Sichuan Basin , South China : Implications for Source Rock Characteristics. *Mar. Pet. Geol. 2018, 96, 577−590.*
(29) Bao, J.; Zhu, C.; Wang, Z. Typical End-Member Oil Derived from Cambrian-Lower Ordovician Source Rocks in the Tarim Basin, NW China. *Pet. Explor. Dev. 2018, 45, 1177−1188.*
(30) Selby, D.; Creaser, R. A.; Dewing, K.; Fowler, M. Evaluation of Bitumen as a 187Re,187 Os Geochronometer for Hydrocarbon Maturation and Migration: A Test Case from the Polaris MVT Deposit, Canada. *Earth Planet. Sci. Lett. 2005, 235, 1−15.*
(31) Selby, D.; Creaser, R. A. Re–Os Geochronology of Organic Rich Sediments: An Evaluation of Organic Matter Analysis Methods. *Chem. Geol. 2003, 200, 225−240.*
(32) Zhao, X.; Wang, Z.; Liu, C.; Li, C.; Jiao, P.; Zhao, Y.; Zhang, F. Characteristics and Geological Significance of Re-Os Isotopic System of Evaporites in Mbkoumassi Deposit, the Republic of Congo. *J. Afr. Earth Sci. 2018, 138, 14−21.*
(33) Ravizza, G.; Turekian, K. K. Application of the 187Re-187Os system to black shale geochronometry. *Geochemica et Cosmochimica Acta 1989, 53, 3257−3262.*
(34) Li, J.; Xie, Z.; Dai, J.; Zhang, S.; Zhu, G.; Liu, Z. Geochemistry and Origin of Sour Gas Accumulations in the Northeastern Sichuan Basin, SW China. *Org. Geochem.* 2005, 36, 1703–1716.

(35) Lomando, A. J. The Influence of Solid Reservoir Bitumen on Reservoir Quality. *AAPG Bull.* 1992, 76, 1137–1152.

(36) Cai, C.; Xiang, L.; Yuan, Y.; Xu, C.; He, W.; Tang, Y.; Borjigin, T. Sulfur and Carbon Isotopic Compositions of the Permian to Triassic TSR and Non-TSR Altered Solid Bitumen and Its Parent Source Rock in NE Sichuan Basin. *Org. Geochem.* 2017, 105, 1–12.

(37) Rogers, M. A.; McAlary, J. D.; Bailey, N. J. L. Significance of Reservoir Bitumens to Thermal-Maturation Studies, Western Canada Basin. *AAPG Bull.* 1974, 58, 1806–1824.

(38) Marsh, H.; Foster, J. M.; Hermon, G.; Iley, M. Carbonization and Liquid-Crystal (Mesophase) Development. Part 2. Co-Carbonization of Aromatic and Organic Dye Compounds, and Influence of Inerts. *Fuel* 1973, 52, 234–242.