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Generation Time and Accumulation of Lower Paleozoic Petroleum in Sichuan and Tarim Basins Determined by Re–Os Isotopic Dating

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Abstract: With the targets of petroleum exploration transferred to the deep and ancient strata, abundant oil and gas resources have been found in Lower Paleozoic and older strata in central and western China. Due to complex evolutionary processes including multiple episodes of hydrocarbon accumulation and ubiquitously accompanied by secondary alterations, significant uncertainties remain concerning the generation time and accumulation processes of these revealed petroleum sources. In this paper, relative pure Re and Os elements existing in the asphaltene fractions of Lower Cambrian solid bitumen collected from the Guangyuan area, western Sichuan Basin, SW China and Middle–Lower Ordovician heavy oils in the Aiding area of the Tahe oilfield in the Tarim Basin, NW China were successfully obtained by sample pretreatments, and Re–Os isotopic analysis was subsequently carried out for the dating of these. The Re–Os isotopic composition indicates a generation time of Guangyuan bitumen to between 572 Ma and 559 Ma, corresponding to the late Sinian period of the Neoproterozoic era. By the means of Re–Os isochron aging, initial $^{187}Os/^{188}Os$ ratios, and carbon isotopic compositions, the Lower Cambrian bitumen is supposed to originate from source rocks of the Doushantuo Formation in the Sinian strata and subsequently migrated into the reservoirs of the Dengying Formation. This previously reserved petroleum was transformed into its present bitumen state by the destruction of reservoirs caused by tectonic uplift. The Re–Os dating results of Middle–Lower Ordovician heavy oil of Tarim Basin suggest that it was formed between 450 Ma to 436 Ma, corresponding to the Late Ordovician–Early Silurian system, and the generated petroleum likely migrate into the Middle–Lower Ordovician karst reservoirs to form early oil reservoirs. With tectonic uplift, these oil reservoirs were degraded and reformed to the heavy-oil reservoirs of today.

Keywords: Re–Os isotopic chronology; pretreatment means; petroleum generation time; Lower Paleozoic petroleum; western part of China

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1. Introduction

Abundant oil and gas resources that have been discovered in the deep Lower Paleozoic and older strata in the past two decades [1–7] have become more important for petroleum exploration in China. The thermal maturity is generally high—over stage in the Middle Neoproterozoic and even the Lower Paleozoic strata [8,9]—which is unfavorable to petroleum preservation, leading to most of them being gas reservoirs [5,10]. However, a large number of oil seepages or large-scale bitumen veins found in some areas indicate that these ancient strata are likely potential sources of petroleum [11,12]. The typical superimposed basins in western China, such as the Sichuan Basin and Tarim Basin, contain abundant ancient petroleum in the Lower Paleozoic and even the Precambrian [2,3,7] strata, indicated by the bitumen veins of the Lower Cambrian in the northern section of the Longmen Mountain thrust belt in Sichuan Basin [11] and the Middle–Lower Ordovician Tahe oil field [5] in the Tarim Basin. Source correlation and generation time of these petroleums to their potential sources have been widely carried out, but uncertainties and controversies still exist [11,13–16].

Determining the geological time of hydrocarbon accumulation is relatively difficult, although it is valuable for the understanding of formation mechanisms of oil and gas reservoirs. In the early stage of reservoir development, hydrocarbon accumulation time is qualitatively inferred by the major stages of hydrocarbon generation and expulsion of the source rock, the formation time of traps, hydrocarbon–water interface tracing, and reservoir saturation pressure/dew-point pressure. These methods are involved in the indirect dating of accumulation [17]. Since the 1990s, reservoir geochemistry, fluid inclusion analysis, and other inversion methods have been used to indirectly determine the oil and gas charging time on the microscale [17–20]. However, these methods do not directly measure the duration of each stage of hydrocarbon reservoir formation, which can be measured by indirect, qualitative, or semiquantitative methods to determine the relative time of hydrocarbon accumulation. The direct dating of hydrocarbon reservoirs (containing solid bitumen, crude oil, oil sand, etc.) is the inevitable trend of hydrocarbon accumulation geochronology from indirect and qualitative research to direct and quantitative research. With the development of laser microscale purification systems, mass spectrometry detection, and improved isotope chemical purification and separation methods, it is now possible for radioactive isotope dating to directly determine the age of hydrocarbon generation, including the isotopic dating methods of K(^{39}Ar)–^{40}Ar, Re–Os, U–Pb, Rb–Sr, and Sm–Nd. The use of radioisotope dating to determine the formation time of geological bodies and metal deposits has been shown to be effective [21–23]. However, the application of radioisotope dating in the geochronology of hydrocarbon accumulation is relatively recent, from the late 1980s, when scholars began to use isotope isochronal methods to determine hydrocarbon-generation time [24,25].

Lee et al. (1985) [24] first used the authigenic illite dating method to determine the formation time of the Rotliegendes sandstone gas reservoirs in the southern part of the North Sea, which revealed the accumulation time of the oil field in the North Sea [25,26]. Parnell and Swainbank (1990) [23] first reported the accurate Pb–Pb age of uranium-bearing bitumen veins in a Wales copper mine, and time of hydrocarbon migration into the vein beds was determined. Mossman et al. (1993) [27] determined the U–Pb isotopic age of early Proterozoic shale in uranium ores in Elliot Lake, Canada. According to the theory and practice of isotopic chronology and geochemistry of mineral deposits, methods for separation and enrichment of samples and solid isotope analysis have been well established. The solid bitumen formation and petroleum migration ages in Tarim Basin, Junggar Basin, Southern China, Liaohe Oilfield, and other regions were determined by measuring the isotopic compositions of K–Ar, U–Pb, Pb–Pb, Rb–Sr, and Sm–Nd of authigenic illite, bitumen, crude oil [28–31]. Wang et al. (1997) [32] and Zhang et al. (2004) [28] first studied authigenic illite K–Ar dating in oil and gas fields in China and obtained good results for the hydrocarbon accumulation age in Tarim, Songliao, Turpan–Hami, and other basins [33]. The ^{40}Ar–^{39}Ar method was recently introduced to study hydrocarbon accumulation, effectively solving the influences of detrital illite on the dating results and strongly promoting the development of hydrocarbon accumulation chronology [34–36]. The petroleum and solid bitumen formation time and petroleum migration time in Tarim Basin, Junggar Basin, southern China, Liaohe Oilfield and other areas were studied with radioisotope systems, such as U–Pb, Rb–Sr, and Sm–Nd [37–39], which effectively constrained the absolute geological age of hydrocarbon accumulation [40].

Recently, Re–Os isotope dating methods are shown to be an effective method to access hydrocarbon accumulation age [41–43]. These methods can directly date hydrocarbon source rock, solid bitumen, crude oil, and oil sands and can acquire the oil and gas generation and migration age related to hydrocarbon accumulation. The Re–Os isotope dating method is based on the variation of the isotopic composition of osmium with time, caused by the β-decay of radioactive ^{187}Re into ^{187}Os. Re and Os have siderophile, chalcophile, and organophilic properties. Re and Os can be dissolved in water under oxidative conditions, but are not easily dissolved under reductive conditions and often accumulate in sedimentary rocks that generate oil and gas on a large scale under anoxic-reduction conditions, and are also enriched in crude oil, solid bitumen, oil sands, and kerogen. The content of Re and Os in the organic matter system has an obviously positive correlation with the abundance of organic matter. Previous studies showed that Re and Os can also exist in bitumen, kerogen, crude oil, and other organic matter in the form of organic complexes for a long time ($T < 350$ °C) [42,44] without the interference of radioactive elements in migration pathway rocks or reservoir rocks and without the influence of late modification. They can also maintain a good closed system [20]. This provides an important theoretical premise for the Re and Os isotopic dating method. The geological clock of the Re and Os isotope system in crude oil begins after the source rocks generate oil, and the isotopic composition of Re and Os in solid bitumen and crude oil reflects that of the source rocks when the petroleum is formed; therefore, the Re–Os isotopic dating system determines the hydrocarbon-generation time [20,41,43]. At the same time, the initial ratios of $^{187}Os/^{188}Os$ in solid bitumen and crude oil can also effectively trace hydrocarbon source rocks. The aim of the present study was to carry out Re–Os isotopic dating to reveal the petroleum generation time of the Lower Cambrian Guangyuan bitumen of Sichuan Basin and Middle–Lower Ordovician oil of Tarim Basin.

2. Geological Settings

2.1. Lower Cambrian Bitumen in Guangyuan Area, Western Sichuan Basin

The bitumen veins and oil seepages in the northern part of Longmen Mountain belt in western Sichuan Basin are widely distributed, mainly in the Nianziba and Kuangshanliang anticlinal structures in the Guangyuan area. The Nianziba nose-like structure and Kuangshanliang anticlinal structure are in the southeastern margin of the Longmen Mountain thrust belt in the western part of Sichuan Basin. According to statistics, the bitumen veins on Kuangshanliang and Nianziba structures are well developed (Figure 1). There are 137 veins with 37 developed in the Nianziba structure and 100 in the Kuangshanliang anticline that are distributed in the Lower Paleozoic strata, the oldest bitumen veins in the world [11]. Some bitumen veins are relatively larger, with a width of ~8 m. In the late 1960s, a 15.3 m-thick bitumen vein was discovered at a depth of 149 m to 164.3 m, and 30 L crude oil was produced from a depth of 333 m and 335.5 m in the Lower Cambrian in Tian 1 well in the Nianziba anticline structure [11]. In the Nianziba structure, the bitumen veins are mainly distributed in the middle–lower strata of the Changjianggou Formation in the Lower Cambrian at the south and north margin and middle parts of the structure. In the Kuangshanliang structure, the bitumen veins are mainly distributed in the upper–middle strata of the Changjianggou Formation in the Lower Cambrian over the whole structure, and controlled by the cracks and faults formed in the structure. The Changjianggou Formation is mainly composed of shales, sandy mudstones, and siltstones of shallow marine facies. The middle and upper parts are mainly sandstones and argillaceous siltstones where asphalt veins are produced (Figure 2). The wide distribution of bitumen veins and the discovery of oil in the Tian 1 well demonstrates the existence of Sinian–Cambrian reservoirs in these structures and indicates that there is a good prospect for finding Sinian and Cambrian reservoirs in the eastern margin of the Northern Longmen Mountain thrust belt.

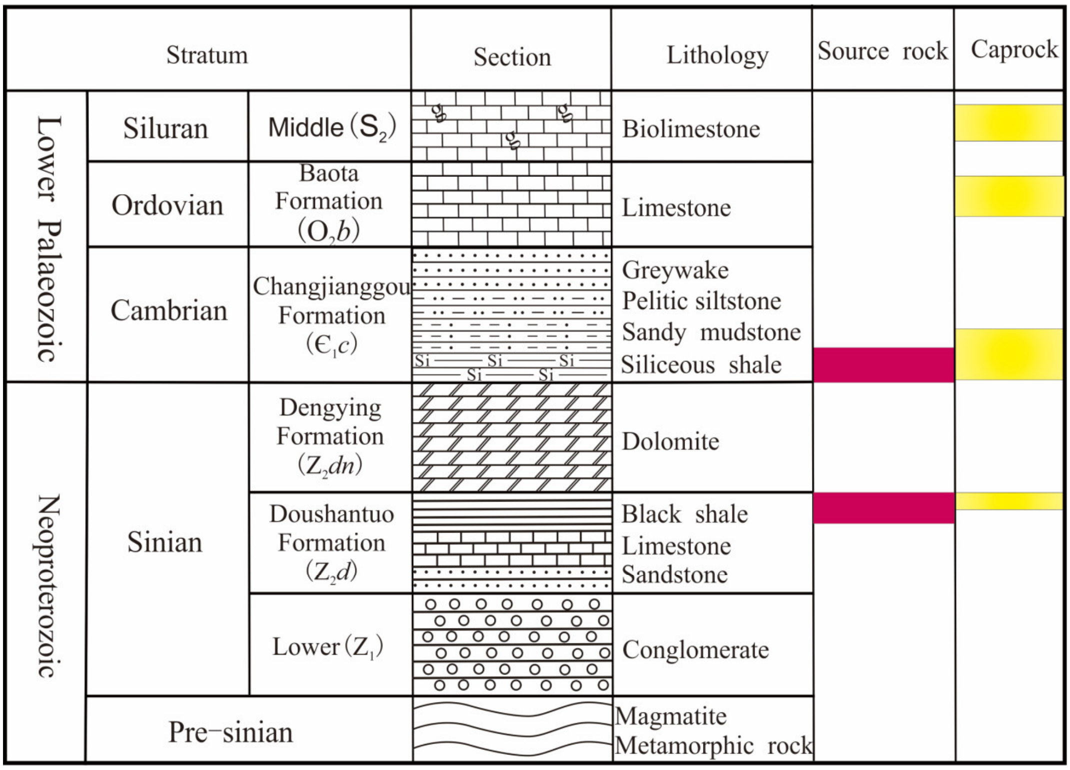

Figure 1. Strata outcrop of Kuangshanliang and Nianziba structures in Guangyuan.

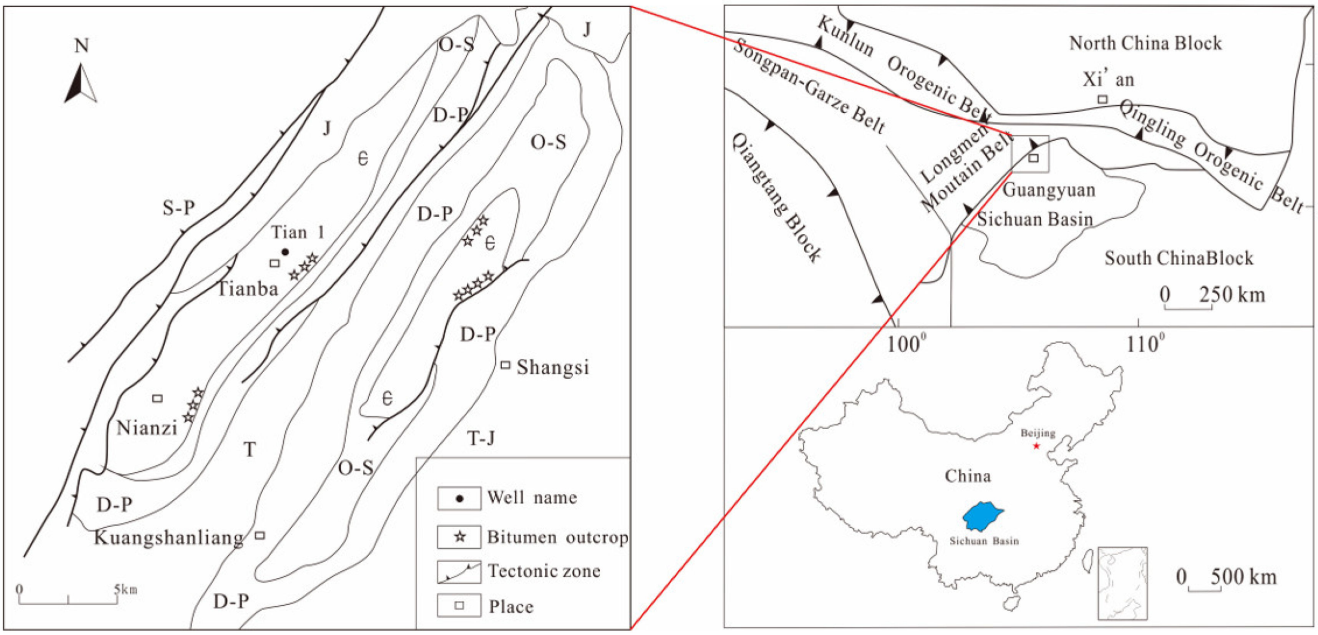

Figure 2. Stratigraphic column in north part of Longmen Mountains, western Sichuan Basin.

2.2. *Middle–Lower Ordovician Heavy Oil in Aiding Area in Tahe Oilfield, Tarim Basin*

The Aiding area is located in the downward slope direction of the northwest Akekule uplift in the Tarim Basin (Figure 3); this area had better oil and gas production in early exploratory wells, where some wells produced a small amount of heavy oil. The heavy oil was mainly distributed in the fracture–cave reservoir of the Yingshan Formation of the Middle–Lower Ordovician and Yijianfang Formation of the Middle Ordovician.

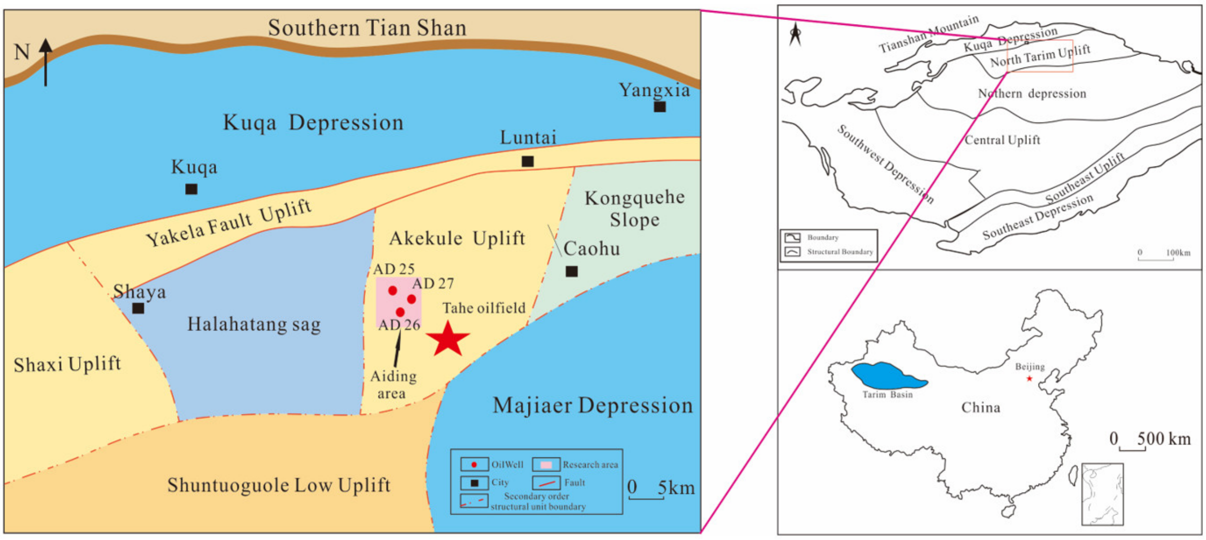

Figure 3. Structural location of Aiding area in Tahe Oilfield.

3. Experimental

3.1. Samples

3.1.1. Lower Cambrian Bitumen in Guangyuan Area, Sichuan Basin

There are two types of macroscopic characteristics of solid bitumen in the Guangyuan area. One is along a crack or fracture in the form of vein and is a purer and softer black bitumen with obvious grease luster and generally developed in the area. The bitumen has a strong smell of oil after tapping and can be ignited, being mostly clastic with pores and joints (Figure 4, Type I). This type of solid bitumen is referred to as type I in this paper. Six samples of type I solid bitumen were collected in Kuangshanliang structure. The other type of bitumen is similar to early diagenetic mudstone, with a dim surface and rough structure (Figure 4, Type II), thin and uneven thickness, mostly massive, and mostly appearing in the upper section of the Changjianggou Formation. This type of solid bitumen is referred to as type II in this paper. In this paper, 5 samples of type II solid bitumen were collected in the Nianziba structure.

Figure 4. Two types of solid bitumen veins in Guangyuan area in the Western Sichuan Basin.

Bitumen reflectance (%Rb) of type I solid bitumen is distributed between 0.38% and 0.50%, which is highly consistent with that previously introduced by Liu et al. (2003) [14]. According to the vitrinite reflectance (%Ro) conversion formula proposed by Jacob [45], %Ro values vary from 0.6% to 0.7% (Table 1), indicate a low degree of maturation. The %Rb values of type II solid bitumen distributed between 0.40% and 0.41% with %Ro values being ~0.65% indicate maturity comparable to type I bitumen. The low degree of maturation of these two types of solid bitumen indicates that the solid bitumen was derived from the thick oil generated by the Sinian source rocks during the early–low mature stage.

Table 1. Thermal maturity of solid bitumen in Guangyuan area in the Western Sichuan Basin.

No.	Sample No.	Sample Type	Section	Formation	R_b (%)	Ro (%)
1	GY–1	Type I bitumen	Bitumen pit in Kuangshanliang structure	$\in_1$ch	0.50	0.71
2	GY–3	Type I bitumen	Bitumen pit in Kuangshanliang structure	$\in_1$ch	0.48	0.70
3	GY–5	Type I bitumen	Bitumen pit in Kuangshanliang structure	$\in_1$ch	0.38	0.63
4	GY–6	Type I bitumen	Bitumen pit in Kuangshanliang structure	$\in_1$ch	0.45	0.68
5	JF–1	Type II bitumen	Bitumen pit in Nianziba structure	$\in_1$ch	0.40	0.65
6	JF–4	Type II bitumen	Bitumen pit in Nianziba structure	$\in_1$ch	0.41	0.65

3.1.2. Middle–Lower Ordovician Oil in Aiding Area in Tahe Oilfield, Tarim Basin

The oil density in Aiding area was between 1.01 and 1.06 g/cm^3, indicating super-heaviness for these oils. The distribution of saturated hydrocarbon fractions of crude oil as a whole shows the dominant characteristics of light hydrocarbons. The odd–even dominance ratio and the carbon dominance index of maturity parameters were 1.32 and 1.26, respectively, indicating that crude oil is in the mature stage with an equivalent vitrinite reflectance of 0.8 to 1.0% [46]. In this paper, three crude oil samples were collected in Aiding area, namely, well Aiding 25(O_2yj), Aiding 26($O_{1\text{-}2}$y) and Aiding 27($O_{1\text{-}2}$y).

3.2. Pretreatment of Bitumen and Oil Samples

The Re–Os pretreatment of the geological sample was carried out in Sinopec Key Laboratory of Petroleum Accumulation Mechanisms. For both crude oil samples and solid bitumen samples, carbon isotope composition characteristics and biomarkers were used to determine whether they come from the same source rocks and were generated in the same period. If so, the following experimental procedures were followed.

For oil samples, around 10 mg was diluted by mixture solvent of n-pentane in a volume ratio of 1:4 in a 100 mL glass bottle, stirred and left for approximately 12 h to ensure it was fully mixed. The mixed solution was transferred to a tube and centrifuged at 2000 rpm for 15 min to ensure the insoluble asphaltene and soluble components were completely separated. The separated asphaltene was dried at 60 °C in an oven and then subjected to the following analysis. Solid bitumen samples were first milled into 4–5 mm particles in an agate bowl, and then 0.1–0.2 g asphaltene or solid bitumen was weighed into a Carius tube with inverse aqua regia (2 mL 12 mol/L HC1 and 6 mL l6 mol/L HNO_3) and 2 mL 30% H_2O_2 solution, with a certain amount of dilution agent added (30 ng^{185}Re and 300 pg^{190}Os). After the Carius tube was sealed, the mixed solution was dissolved by stage heating to 120 °C and stabilized for 1 h, then to 160 °C and stabilized for 2 h, and finally to 200 °C and stabilized for 24 h. These process reduced the probability of tube explosion during the sample-dissolving process and made the samples dissolve better. The Carius tubes were frozen and cut open, and the Os of the solution was separated and purified by in situ direct distillation and microdistillation. The remaining solution was added to 5 mol/L NaOH and Re was extracted with acetone. The acetone was evaporated at low temperature and nitric acid and hydrogen peroxide were added to break the organic phase for separation of Re.

3.3. Instrumental Analysis

The purified Re and Os points in the Pt belt and were determined by negative thermal ion mass spectrometry (N–TIMS). For Re, $^{185}ReO_4$ and $^{187}ReO_4$ were simultaneously determined by static Faraday mode. For Os, $^{186}OsO_3$, $^{187}OsO_3$, $^{188}OsO_3$, $^{190}OsO_3$, $^{192}OsO_3$ were determined by CDD multi-receiver mode, and $^{185}ReO_3$ was determined to deduce the influence of $^{187}ReO_3$ on $^{187}OsO_3$. $^{185}Re/^{187}Re$ = 0.59738 of common Re was used as the external standard for Re isotopic fractionation correction, and $^{192}Os/^{188}Os$ = 3.0827 was used as the internal standard iterative method for Os isotopic fractionation correction [47].

The blank Re of the whole analysis process was 3 pg/g, the blank Os was 0.2 pg/g, and the blank ^{187}Os was approximately 0.05 pg/g. The content of Re and Os in the background of the whole analysis process was negligible compared with that in the asphaltene. To verify the reliability of the pretreatment method, Re and Os contents of molybdenite (GBW04436) were determined using this pretreatment process, and this indicated that the final data were in accordance with the error requirement. Re and Os isotopic data obtained from mass spectrometry were processed by Isoplot software and the age was calculated from the slope of the isochronal line. Initial values, errors, and the weighted mean variance were also obtained.

4. Results and Discussion

4.1. Petroleum Generation Time and Origin of the Lower Cambrian Bitumen in the Guangyuan Area, Western Sichuan Basin

4.1.1. Petroleum Generation Time of the Solid Bitumen

Some exploratory studies on the source of solid bitumen in Lower Cambrian have been carried out by the means of carbon isotope and biomarker analysis, and the solid bitumen is supposed to mainly originate from Sinian–Lower Cambrian source rocks, most likely from black shale of Doushantuo Formation in Sinian [4,11,16,34]. It can also originate from the relatively immature Permian or Lower Triassic source rocks [14,48]. However, previous researchers have not yet carried out a systematic study on the hydrocarbon-generating time of the solid bitumen veins. The Re-Os isotope dating method is used here to explore the hydrocarbon-generation time and indirectly identify the source of the solid bitumen veins of the Lower Cambrian in Guangyuan area, western Sichuan Basin.

The Re–Os isotopic data of two types of solid bitumen in the Guangyuan area are shown in Table 2. Type I solid bitumen in the Kuangshanliang structure has $^{187}Re/^{188}Os$ ratios of 232.6–355.2, and the $^{187}Os/^{188}Os$ ratios ranged from 2.82–4.04 (Figure 5). According to the isochron slope, the Re–Os age of type I solid bitumen was calculated to be 572 $\pm$ 12 Ma in the Kuangshanliang structure. Type II solid bitumen distributed in the Nianziba structure has $^{187}Re/^{188}Os$ ratios of 384.4–534.3 and $^{187}Os/^{188}Os$ ratios of 4.15–5.52 (Figure 5). The Re–Os age of type II solid bitumen was calculated to be 559 $\pm$ 15 Ma in the Nianziba structure. These results comprehensively show that the hydrocarbon-generation time of the Lower Cambrian solid bitumen veins in the Guanyuan area varies from 572 Ma to 559 Ma and may originate from the ancient source rocks and correspond to the Late Sinian.

Table 2. Re–Os data of solid bitumen from Lower Cambrian in Guangyuan area in the Western Sichuan Basin.

Sample No.	Structure	ω (Re)/(ng.g^{-1})		ω (Os)/(ng.g^{-1})		ω (^{187}Os)/(ng.g^{-1})		$^{187}Re/^{188}Os$		$^{187}Os/^{188}Os$	
		Data	2 σ	Data	2 σ	Data	2 σ	Data	2 σ	Data	2 σ
GY–1	Kuangshanliang	298.9	0.9	4.571	0.015	2.124	0.009	314.9	1.4	3.559	0.019
GY–2	Kuangshanliang	345.9	1	5.24	0.016	2.475	0.01	317.8	1.3	3.619	0.018
GY–3	Kuangshanliang	510	1.8	10.556	0.048	3.893	0.019	232.6	1.3	2.825	0.019
GY–4	Kuangshanliang	534.5	1.6	10.841	0.035	4.014	0.016	237.4	1	2.836	0.014
GY–5	Kuangshanliang	358.7	1.1	5.476	0.019	2.542	0.011	315.3	1.4	3.556	0.02
GY–6	Kuangshanliang	277.8	0.8	3.765	0.044	1.988	0.019	355.2	4.2	3.985	0.02
JF–1	Nianziba	592.9	1.8	5.349	0.017	3.858	0.015	533.6	2.3	5.525	0.028
JF–2	Nianziba	545.1	1.6	5.014	0.016	3.539	0.014	523.4	2.3	5.407	0.027
JF–3	Nianziba	477.7	1.4	4.467	0.033	3.114	0.023	514.9	4.1	5.34	0.056
JF–4	Nianziba	497.6	2.9	6.232	0.053	3.374	0.027	384.4	3.9	4.147	0.048
JF–5	Nianziba	539.62	2.36	6.68	0.024	3.622	0.015	388.9	2.2	4.154	0.023

Note: The uncertainty of Re and Os content in the data include the weighting error of the sample and diluent, the isotopic composition error, the calibration error of the diluent, the fractionation correction error of the mass spectrometry, and the isotope ratio error of the sample. The uncertainty levels are 2σ.

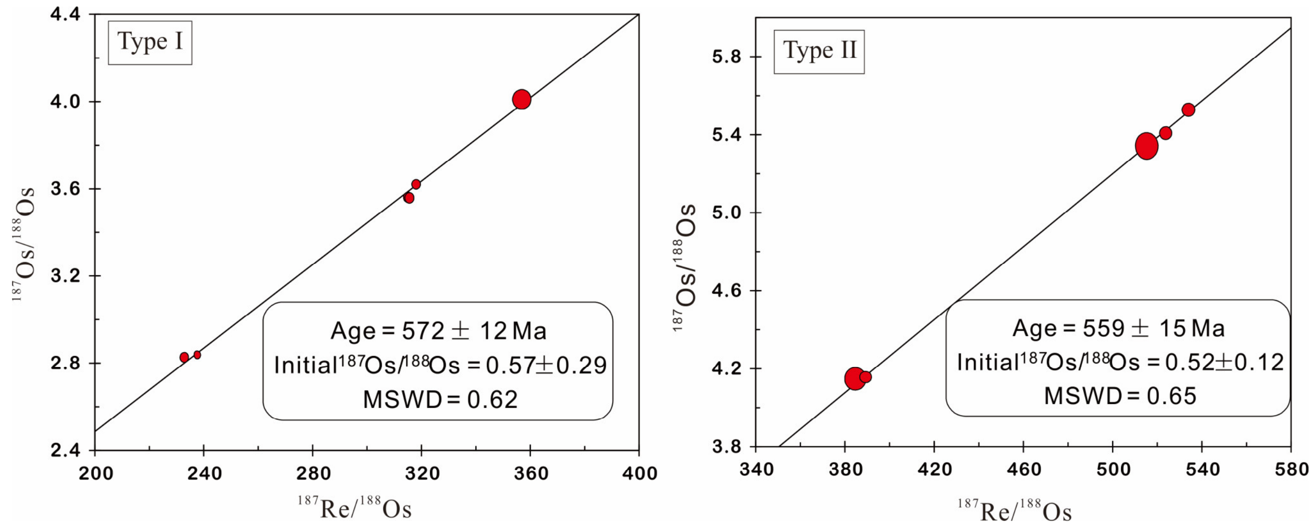

Figure 5. Correlation of $^{187}Re/^{188}Os$ and $^{187}Os/^{188}Os$ of Lower Cambrian bitumen in Guangyuan area.

4.1.2. Source Rock Correlation with the Solid Bitumen

Previous studies have proposed that the initial $^{187}Os/^{188}Os$ ratios of solid bitumen and crude oil can be used to effectively trace source rocks [49,50], providing a new method for oil–source correlation. Figure 5 shows that the two types of solid bitumen have similar initial $^{187}Os/^{188}Os$ ratios (0.57 ± 0.29 and 0.52 ± 0.12 for type I and type II, respectively), indicating that they have similar source rocks.

The inheritance effect of stable carbon isotope composition provides the theoretical basis for the comparison between source rock, crude oil, and solid bitumen. In general, the carbon isotopic shift of crude oil caused by thermal maturation is 1‰ to 3‰ smaller than that of kerogen from the corresponding source rocks, while the carbon isotopic shift of solid bitumen is 2‰ to 3‰ greater than crude oil and the $\delta^{13}C$ values of solid bitumen 1‰ to 2‰ greater than that of kerogen [51,52]. Therefore, the carbon isotopic compositions of solid bitumen and source rock can be used to correlate the hydrocarbon sources of the Lower Cambrian bitumen veins in Guangyuan area. The $\delta^{13}C$ values of type I solid bitumen are between −37.3‰ and −35.7‰, while the carbon isotopic values for type II bitumen are between −38.2‰ and −36.8‰. The similarity of carbon isotopic values of the two types of solid bitumen indicates a common source shared. Based on the carbon isotopic compositions of the Lower Cambrian solid bitumen and potential source rocks, carbon isotopic compositions of Lower Cambrian solid bitumen are significantly different from those of the Lower Cambrian, Upper Ordovician Wufeng Formation–Lower Silurian Longmaxi Formation, which is consistent with the carbon isotopic compositions of kerogen from the Doushantuo Formation in the Sinian system (Figure 6). Therefore, it can be inferred that the solid bitumen is derived from the source rocks of the Doushantuo Formation. Wang et al. (2005) [34] found that the distribution of Doushantuo Formation black shale in the Sinian was abundant and stable, the thickness from 30 m to 40 m, and TOC from 0.2% to 7.0% (generally between 1.8% and 2.5%), and belonged to sapropelic and sapropelic–humic organic matter. Wang and Han (2011) [4] inferred that the thickness of the black shale in the Doushantuo Formation ranged from 25 m to 40 m and the TOC from 1.8% to 2.5%, belonging to sapropelic organic matter. The solid bitumen veins in the Guangyuan area may be derived from source rocks of the Doushantuo Formation of the Sinian, while the organic matter abundance of the Changjianggou Formation shale was low, making it not possible for it to be the source rock of solid bitumen veins [4]. Xie et al. (2003) [16] also proposed that the organic carbon content of the Sinian in the northern part of Longmen Mountain was relatively high and found the solid bitumen in the Guangyuan area came from the black shale in the Sinian Doushantuo Formation by comparing steroid and terpenoid biomarkers. Tian (2009) [53] inferred that the solid bitumen of the Lower Cambrian in the Guangyuan area occurred as a high content of norhopanes, indicating a source rock in the Doushantuo Formation. The black shales of the Doushantuo Formation are not exposed, and no drilling has revealed the strata in the area, but their high organic matter abundance and good organic matter type can generate a large amount of hydrocarbon based on previous results.

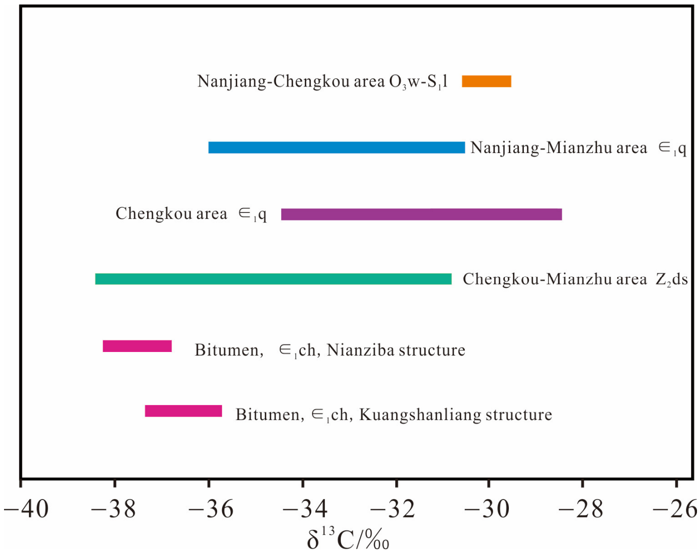

Figure 6. Carbon isotope relationship between solid bitumen in Guangyuan and kerogen of source rocks in Sichuan Basin.

According to the Re–Os isochron aging, the initial ^{187}Os/^{188}Os ratios, carbon isotopes, and biomarkers, the solid bitumen of the Lower Cambrian in the Guangyuan area originated from high-quality source rocks of the Doushantuo Formation. The source rocks with good organic matter type were in low-maturity evolution between 572 Ma and 559 Ma, and produced a certain amount of thick oil during the early hydrocarbon-generation stage. The upper part of Doushantuo Formation is shale and a good caprock. With the development of the Dengying Formation reservoir and the increase of the burial depth of the stratum, the thick oil produced by the source rocks of the Doushantuo Formation entered the Dengying Formation reservoir rocks, forming ancient thick oil reservoirs under pressure. This mechanism is supported by the widespread development of solid bitumen in the Dengying Formation dolomite rocks in the upper Sinian in the Guangyuan–Wangcang–Micang Mountain area on the eastern side of the Longmen Mountain belt [54].

In the western Sichuan Basin, tectonic uplift occurred during the Indosinian period, fractures and faults developed, the Dengying Formation paleo-reservoir was uplifted, and thick oil migrated up to the Changjianggou Formation along fractures or faults. At this time, the Changjianggou Formation strata were raised to near the surface and the thick oil was modified through water washing, oxidation, and escape during the subsequent stage [55]. Therefore, the Lower Cambrian bitumen vein is the current oxidized bitumen formed by the upwards escape of the Dengying Formation crude oil after the destruction during the Indosinian period [56].

4.2. Generation Time of Heavy Oil in the Middle–Lower Ordovician in the Aiding Area, Tahe Oilfield

According to the characteristics of saturated hydrocarbon distribution and carbon isotopic compositions of heavy oil in the Aiding area, three heavy-oil samples (AD25, AD26, and AD27) were selected for Re and Os purification, separation, and quantitative analysis, excluding epigenetic alteration and mixed source. Based on the slope of the isochrone line obtaining by fitting the Re and Os data, the Re–Os ages of the Ordovician heavy oil in the Aiding area of Tahe Oilfield were determined to be approximately 450 Ma to 436 Ma (Figure 7); therefore, the corresponding generation time of the Ordovician heavy oil is from

Late Ordovician to Early Silurian. The oil was formed earlier, which is consistent with the lower maturity of the crude oil. Some scholars believe that the Ordovician oil reservoir in the Aiding area may have formed before the Silurian period and demonstrate that the oil filling time is from the Late Caledonian to the Early Hercynian period because of the reservoir fluid inclusion of the Aiding 4 well [46]. These results show that the oil-generation time determined by Re–Os dating is correct.

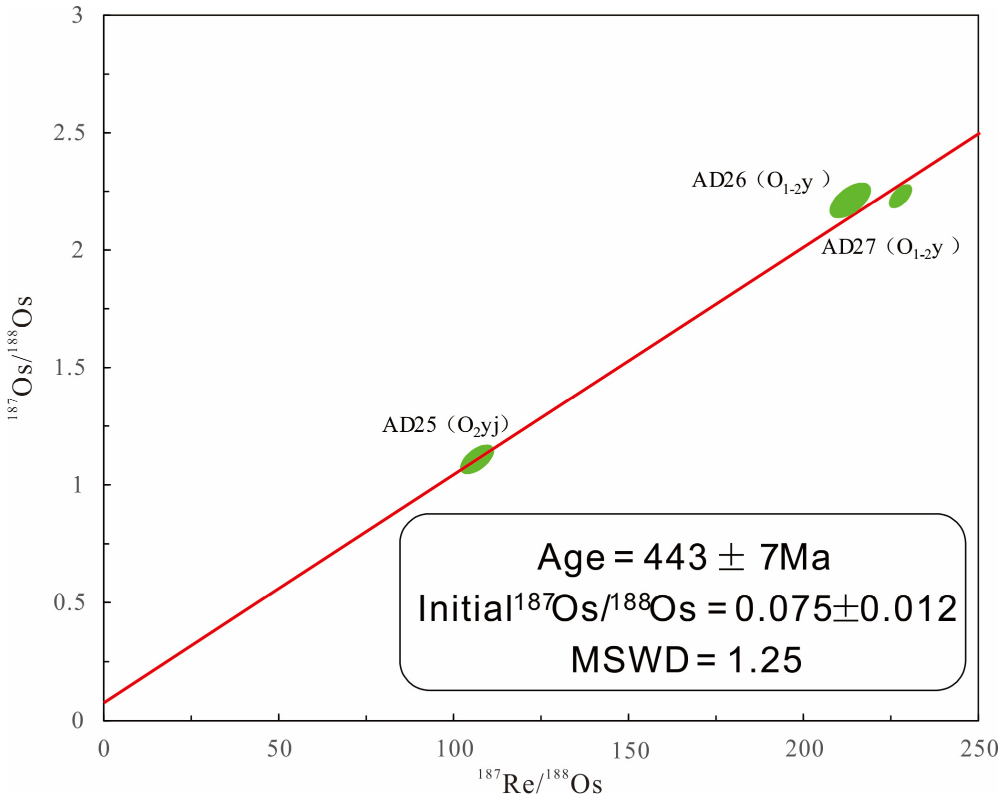

Figure 7. Relationship between ^{187}Re/^{188}Os and ^{187}Os/^{188}Os of the Ordovician heavy oil in the Aiding area of Tahe Oilfield in Tarim Basin.

In the Late Caledonian, the Cambrian source rocks in the Manjiaer depression and its slope area entered the peak of oil generation, and the Aiding area is in a high position of the structure with a favorable direction for oil and gas migration and accumulation. At the same time, as a result of the overall uplift of the first episode in the Middle Caledonian stage, the Yijianfang Formation strata of the Middle Ordovician was exposed, and consequent karstification developed widely in the Aiding, which provided sufficient space for oil and gas charging and formation of early oil and gas reservoirs. Since the Aiding area is located in the low part of the structure and the shielding effect of the Tahe axis, oil- and gas-accumulation conditions were poor during the period from the Early Hercynian to the Late Himalayan. In the early stage of the Hercynian period, the early oil reservoir was severely destroyed by water washing and oxidation with strong uplift and denudation, which formed the present heavy-oil reservoir.

5. Conclusions

Based on the pretreatment technology of the Re–Os isotope dating method of minerals, asphaltene extraction, dissolution, Re–Os purification, enrichment and separation of pretreatment technology were established. Re–Os isotopic dating and oil source rock of two types of solid bitumen veins were indicated in the Lower Cambrian in the Guangyuan area, western Sichuan. The hydrocarbon-generation time of the Lower Cambrian solid bitumen in the Guangyuan area varied from 572 Ma to 559 Ma, indicating the oil may have originated from the source rocks of the Doushantuo Formation. This source rocks were of low maturity and began to produce a certain amount of thick oil during 572 Ma and 559 Ma. Subsequently, thick oil entered Dengying reservoir rocks to form a thick paleo-oil reservoir and formed the present bitumen vein through the late uplift.

Meantime, the Re–Os dating results of Middle–Lower Ordovician heavy oil in the Aiding area in Tarim Basin suggested that it was formed between 450 Ma to 436 Ma, corresponding to the Late Ordovician–Early Silurian system, and the generated petroleum likely migrated into the Middle–Lower Ordovician karst reservoirs to form early oil reservoirs. With tectonic uplift, these oil reservoirs were degraded and reformed to the present heavy-oil reservoirs.

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