Formation Time and Material Source of Carbonate Cements in Permian Sandstone in the Ordos Basin, Western of China

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Carbonate cements are an important type of cement in Permian tight gas reservoirs in the Ordos Basin. Moreover, the distribution of carbonate cements in sandstone provides an important basis for studying the fluid mechanism and genesis of cements and a scientific basis for investigating the physical properties of reservoirs and the underground distribution of oil and gas. Based on the observations of thin sections and temperature measurements of fluid inclusions, this study concludes that the carbonate cements in the study area are dominated by calcite and ferrous calcite and divided into three phases, where the latter two phases are dominant. The material source of carbonate cements is obtained via the thermal simulations of gold tubes in source rocks, temperature of fluid inclusions in carbonate cements, composition of fluid inclusions in quartz overgrowth, and analysis of carbon and oxygen isotopes. Results show that phase I calcite may be directly precipitated from the supersaturation of pore water by Ca$^{2+}$ and CO$_3^{2-}$. The CO$_3^{2-}$ in the phase II calcite and ferrous calcite stems from the CO$_2$ generated in the hydrocarbon generation process of organic matter, while the CO$_3^{2-}$ in phase III calcite stems from the CO$_2$ generated in the decarboxylation of organic acids. Ca$^{2+}$ is derived from the dissolution of early carbonate cements in sandstone and conversion of smectite into illite in mudstone. The Permian sandstone in the Yishan Slope at the center of the basin is close to the source rock, and the carbon isotopic value of the carbonate cements, which is dominated by organic carbon, ranges between $-7.5\%$ and $-19.0\%$. However, the Permian sandstone in the Tianhuan Depression at the edge of the basin is far away from the source rock, with its carbon isotopic value ranging from $-5.9\%$ to $-8.7\%$, which is probably due to the insufficient supply of organic carbon and a small amount of early inorganic CO$_2$ remains in the reservoir.

1. Introduction

Carbonate cements are an important type of cement in sandstone reservoirs and play a vital role in sandstone reservoirs because they affect the quality of the reservoirs [1–8]. The oil availability and heterogeneous distribution of oil and gas in the tight sandstone are mainly affected by reservoir quality [9, 10]. In addition, the carbonate cement content affects the hydraulic fracturing efficiency. These parameters are important indicators for oil and gas exploration and development. It is considered that carbonate cements can be formed during both the early and late diagenetic stages but their composition, crystal shape, size, and distribution characteristics differ [3, 11]. In particular, the carbonate cements formed during the early diagenesis are favorable for the reservoirs, while those formed during the late diagenesis fill the pores and reduce the porosity [7] and permeability of sandstone, which is not conducive to the reservoirs [11, 12]. In addition, the main controlling factors of sandstone reservoir with strong heterogeneity are grain size, composition, and distribution of diagenetic facies. The uneven distribution of carbonate cements in
the sandstone reservoir also affects its heterogeneity [4]. The most important factors affecting the type and distribution of carbonate cements are the ratio of mudstone to sandstone and the distance between the sandstone reservoir and the mudstone [13].

Many previous studies on the material source of carbonate cements in sandstone have shown that carbon sources can be categorized into two types: organic and inorganic [7, 14, 15]. Organic sources of carbon in carbonate cements include dissolution of skeletal debris particles, oxidation of sedimentary organic matter by methanogens, cracking of oil, or pyrolysis of organic acids in the telodiagenesis [8, 16]. Inorganic sources of carbon in carbonate cements include supersaturation of the carbon dioxide in sea or lake water and dissolution of carbonate formed in the eodiagenesis [8]. For carbonate cements in sandstone, cation sources of the carbonate cements (Ca²⁺, Mg²⁺, and Fe²⁺) may be one of the following: evaporation of seawater or lake water [17], dissolution of early carbonate cements [18], transformation of clay minerals [13, 19], and expulsion of fluids from adjacent mudstone or shale during diagenetic periods [7, 11]. Furthermore, deep geological fluids, which are rich in carbon dioxide, provide a material source for carbonate cement formation [20, 21]. Therefore, the material sources of carbonate cements are complex.

The Ordos Basin is one of the most abundant areas of tight sandstone gas in China. Many studies have been conducted based on the Ordos Basin, mainly focusing on the charging, migration, and accumulation mechanisms of tight sandstone gas and the diagenetic evolution of tight sandstone [22–24]. Carbonate cementation plays an important role in the diagenetic evolution of tight sandstone because early carbonate cementation not only enhances the compaction resistance of tight sandstone [25] but also inhibits quartz overgrowth [26, 27]. In addition, the dissolution of early carbonate cements is conducive for the formation of high-quality reservoirs. However, few studies have focused on the carbonate cements of Permian tight sandstone in the Ordos Basin. Moreover, the material source carbon of the carbonate cements has not been studied in detail. Therefore, the distribution, the formation time, and material source of carbonate cements in the Permian tight sandstone reservoir are unclear. Nevertheless, the study of carbonate cements in Permian tight sandstone can provide a reliable basis for reservoir physical properties, fracturing efficiency, and gas distribution of Paleozoic gas reservoirs and then provide a scientific basis for exploration and development of gas reservoirs. Therefore, it is necessary to study the formation time and material source of carbonate cements of Permian tight sandstone in the Ordos Basin.

The main objectives of this paper are as follows: (1) studying the formation time, material source, and distribution of carbonate cements in the Permian tight sandstone of the Ordos Basin has important theoretical guiding significance for the prediction of reservoirs having high carbonate cements content and good physical properties and (2) determining the similarities and differences between carbon sources of carbonate cements in the sandstones of major petrolierous basins in China and establishing the relationship between the carbon sources of carbonate cements in the sandstones of different types of source rocks.

2. Geological Setting

The Ordos Basin is a large hydrocarbon-bearing basin and a multistage craton basin. It is located in North China with an area of 320,000 square kilometers and where several large oil and gas fields have been discovered (Figure 1) [10, 28–31]. The tectonic evolution history of the Ordos Basin can be divided into five evolutionary stages according to the five structural sequences and development characteristics of the Jinning cycle, Caledonian cycle, Indo-Chinese cycle, Yanshan cycle, and Himalayan cycles: the development stage of a continental rift in Middle and Late Proterozoic, the formation stage of a continental margin sea basin in Early Paleozoic, the formation stage of an inner craton in Late Carboniferous and Middle Triassic, the development stage of a foreland basin in Late Triassic and Early Cretaceous, and the peripheral faulted basin in Cenozoic [24, 32, 33]. The Ordos Basin is divided into six subtectonic units: the Western Thrust Belt, the Tianhuan Depression, the Yimeng Uplift, the Jinxi Fault–Fold Belt, the Weibei Uplift, and the Yishan Slope [9, 23, 31, 33].

In the Permian strata of the Ordos Basin, the strata are successively Shiqianfeng Formation, Shihzei Formation, Shangxi Formation, and Taiyuan Formation successively from top to bottom (Figure 2). The Shiqianfeng Formation is dominated by fluvial and arid lacustrine deposits [24, 32]. The Shihzei Formation is divided into eight members designated He 8 to He 1 from bottom to top. The strata from members He 1 to He 4 are named the Shangshihzei Formation, which is mainly composed of delta facies. The strata from members He 5 to He 8 belong to the Xiashihzei Formation mainly formed under braded river–braided river delta facies. The Shanxi Formation is dominated by swamp–delta deposits, while the Taiyuan Formation is predominated by tidal flat–delta deposits (Figure 2) [24, 32].

In the Permian strata, the reservoirs are dominated by tight sandstone, mainly distributed in the Shanxi Formation and the Shihzei Formation [34]. The 8th member of Shihzei Formation, which is widely distributed in the basin, contains the most developed sandstones that can be used as reservoirs [35]. The source rocks are mainly composed of coal, dark mudstone, and limestone, which are mainly distributed in the Permian Shanxi Formation and Taiyuan Formation [36, 37]. The gas of the 8th member of the Shihezi Formation mainly comes from the underlying source rocks of the Shanxi and Taiyuan Formations [38, 39]. Furthermore, the strata of the Shanxi Formation and Shihezi Formation are composed of interbedded sandstone and mudstone (Figure 2), which can form a good combination of reservoir and seal rock locally [40].

3. Samples and Methods

3.1. Samples Selection. In this study, 124 Permian sandstone samples derived from 21 wells in the Ordos Basin were employed, including core and field outcrop samples. Based
on the combination of the analysis of the collected logging data and core observations with handheld X-ray fluorescence (XRF) and X-ray diffraction (XRD) data analysis (Figure 3), 19 sandstone samples with high carbonate content located near the contact surface of sandstone and mudstone were selected to analyze the formation time, material source, and distribution of carbonate cements. The main research strata are 8th member of the Shihezi Formation and Shanxi Formation because they are the main reservoirs of the Permian in the Ordos Basin. In addition, coal samples obtained from the Shanxi and Taiyuan Formations were used to conduct simulated hydrocarbon generation experiments to analyze the relationship between the carbon isotopes of carbon dioxide produced in the hydrocarbon generation process of source rocks and carbon isotopes of carbonate cements in sandstone.

3.2. X-Ray Diffraction Analysis. An X-ray diffractometer applies the principle of diffraction to accurately determine the crystal structure of a sample and perform the phase, qualitative, and quantitative analyses of a material. In this study, the Japanese Rigaku Ultima IV was used to conduct simulated hydrocarbon generation experiments to analyze the relationship between the carbon isotopes of carbon dioxide produced in the hydrocarbon generation process of source rocks and carbon isotopes of carbonate cements in sandstone.

3.3. Handheld X-Ray Fluorescence Analysis. X-ray fluorescence analysis refers to analyzing the X-rays emitted by the analyzed sample that has been excited by being bombarded by X-rays. Because of the different elements in the analyzed sample, the energy and wavelength of the emitted characteristic X-rays differ. A qualitative analysis of elements is performed by measuring the X-ray energy or wavelengths, while a quantitative analysis of elements is performed by measuring the characteristic X-ray intensity of an element. In this study, a Thermo Scientific™ Niton XL3t handheld X-ray fluorescence (XRF) spectrometer was used for field testing of core samples from 10 selected wells. The same sample was tested twice, and the test period was 1 minute.

3.4. Gold Tube Thermal Simulation Analysis. Gold tube thermal simulation analysis models hydrocarbon generation in geological time by controlling the temperature and pressure of coal or source rock kerogen. The JGMN-1 gold tube simulator comprises a high-temperature cracking unit, a temperature–pressure control unit, a constant pressure unit, a data-acquisition and analysis, and sampling system [41].

Figure 1: Tectonic units of the Ordos Basin and location of the study area (after [23]).
| System        | Formation | Member | Lithology                        | Thickness (m) | Sedimentary facies                     | Mark layer                              |
|---------------|-----------|--------|----------------------------------|---------------|----------------------------------------|------------------------------------------|
| Triassic      | Liujiagou |        |                                  |               |                                        |                                          |
|               | Shiyanfeng|        |                                  |               |                                        |                                          |
| Permian       | He 1      |        |                                  | 22–53         |                                        | K₆ sandstone                            |
|               | He 2      |        |                                  | 26–56         |                                        | Siliceous mudstone                      |
|               | He 3      |        |                                  | 22–46         |                                        | Purple mudstone                         |
|               | He 4      |        |                                  | 17–50         |                                        |                                        |
|               | He 5      |        |                                  | 16–44         |                                        | Peach blossom mudstone                  |
|               | He 6      |        |                                  | 21–48         |                                        | Yellow-green sandstone                  |
|               | He 7      |        |                                  | 30–50         |                                        | Camwil’s neck sandstone                 |
|               | He 8      |        |                                  | 25–55         |                                        |                                        |
|               | Shan 1    |        |                                  | 30–80         |                                        | The upper coal group                    |
|               | Shan 2    |        |                                  | 26–75         |                                        | The middle coal group                   |
| Carboniferous | Tai 1     |        |                                  | 6–38          |                                        | Dongdayao limestone                     |
|               | Tai 2     |        |                                  |               |                                        | Xiedao limestone                        |
|               | Benxi     |        |                                  | 27–81         |                                        | 7# coal seam                            |
| Ordovician    | Majiagou  |        |                                  |               |                                        | Miaoqiao limestone                      |
|               |           |        |                                  |               |                                        | Miaoqiao limestone                      |
|               |           |        |                                  |               |                                        | Wujiayu limestone                       |
|               |           |        |                                  |               |                                        | Iron and aluminum layers                |
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**Figure 2:** Generalized Upper Paleozoic stratigraphy of the Ordos Basin (after [24, 32]).
The heating rate of the instrument ranges between 2 and 20°C/h, with a temperature range of 0°C–800°C and a pressure range of 0–200 MPa. The requirement for experimental samples is that their particle size range should be 80–200 mesh. Coal and kerogen samples weighed between 10 and 120 mg at each temperature point, and the source rocks weighed between 600 and 800 mg at each temperature point. The heating rate of the instrument was set at...
2°C/h, and the pressure was set at 20 MPa. The experiment was conducted at the Oil and Gas Center, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences.

3.5. Analysis of Carbon and Oxygen Isotopes. The carbon and oxygen isotopes of carbonate cements were determined using the GasBench II continuous flow method with a 253 Plus mass spectrometer for 19 sandstone samples with high carbonate content from 10 wells. A small number of sandstone samples containing carbonate cements were loaded manually into a 12 mL reaction flask. High-purity helium gas (99.999%) was used for 600 s, emptying at a flow rate of 100 mL/min. Then, 4–6 drops of phosphoric acid were deposited in each exetainer. The exetainers were placed in an aluminum tray kept at 72°C for 4 h for calcite. Subsequently, the generated CO₂ gas was separated from other impurity gases through a fused silicon capillary column (Poraplot Q, 25 m × 0.32 mm) and then transported to a mass spectrometer for measurement. Normally, 18 standard samples can be simultaneously measured during the measurement process. The measurement accuracy of δ¹³C and δ¹⁸O is higher than 0.1‰. Analyses were performed at the State Key Laboratory of Isotope Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences. Moreover, the carbon isotope values in coal samples were measured. In addition, the carbon isotopes of the CO₂ gas generated by the thermal simulation of the gold tube were measured on MAT271 of the Oil and Gas Center, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences.

3.6. Fluid Inclusion Analysis

3.6.1. Homogenization Temperature of Fluid Inclusion in Carbonate Cements. Fluid inclusion analysis is mainly used to observe homogenization temperature, freezing temperature, and fluorescence of fluid inclusions in thin sections of rocks using a combination of a three-channel advanced fluorescence microscope, hot and cold platform, and a fiber optic spectrometer. Temperature measurements ranged between −196°C and 600°C, with an accuracy of ±0.1°C. The gas–liquid inclusions were measured using a Nikon 80i microscope and a THMGS 600 hot and cold platform at the Oil and Gas Center, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences. In general, the rate of the initial heating or cooling process was 5°C/min and was reduced to 0.2°C/min near homogenization or complete ice melting. The homogenization temperature (Th) and freezing-point temperature were recorded when the inclusions were completely homogenized and melted. In this paper, the homogenization temperature of fluid inclusions in carbonate cements was measured for nine sandstone samples.

3.6.2. Laser Confocal Raman of Fluid Inclusion Analysis. Raman spectroscopy is used to investigate the composition, structure, and relative content of molecules. In this study, Raman spectroscopy was mainly used to determine the composition of fluid inclusions in quartz overgrowth for 19 sandstone samples. The Horiba-JY Xplora compact multifunctional microlaser Raman spectrometer was used to analyze the fluid composition in the fluid inclusion at the State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences. The instrument has three lasers emitting at 532, 638, and 785 nm, and the spectral range is 100–4000 cm⁻¹. The 532 nm laser was used to determine the composition of the fluid inclusion.

3.7. Total Organic Carbon Content Analysis. The analysis of the total content of organic carbon involves setting pretreated samples into a combustion furnace. Carbon dioxide is generated through high-temperature oxidation and then processed in a catalytic furnace, generating CO₂. The total organic carbon (TOC) content is then measured using an infrared detector [42, 43]. The sample pretreatment process is as follows: about 0.1 g of a sample less than 80 mesh is weighed, and the weight data are accurately recorded (accuracy of 0.001 g). The weighed samples are placed into a crucible that was permeable to water. An excess of 5% hydrochloric acid is added to completely dissolve the carbonates in the samples. The samples in the crucible are washed with distilled water to make them neutral. Then, the crucible and samples are placed in an oven at 40–60°C for 12 hours and then taken out for cooling. Then, the samples are tested on the carbon sulfur analyzer. The sensitivity of the instrument is 0.1 µg/g, and the accuracy of the measured organic carbon is less than 0.1%. The experiment was conducted at the Oil and Gas Center, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences.
4. Results

4.1. Petrology. The 8th member of the Shihezi Formation and Shanxi Formation are the main research horizons in the study of carbonate cements of the Permian sandstone in the Ordos Basin. According to the observation of the thin sections of these sandstone rocks, the contents of quartz, feldspar, and rock fragments are 67%–97% (ave. 87%), 0%–3% (ave. 1%), and 2%–32% (ave. 12%), respectively. Based on the Folk classification scheme, the samples are mainly sublitharenite, while some samples are quartz arenite and litharenite (Figure 4). The XRD analysis of the samples (Table 1) and observations of the rock thin sections (Figure 5) show that the quartz particle content is the highest. In addition, the carbonate cement content is relatively high (0%–33.5%) (Figures 5(a)–5(c)), with an average of 12.9% (Table 1). Some samples with lower amounts of carbonate were used for genetic analysis of the same well.

4.2. Characteristics of the Carbonate Cement. In thin sections of the cast stained with alizarin red and potassium ferricyanide, red indicates calcite, purplish red indicates ferrous calcite, and blue indicates pores (Figure 5). In this study, the carbonate cements of sandstone are mainly calcite and ferrous calcite, where calcite is the main substance, accounting for approximately 92%, and ferrous calcite accounts for 8%.

4.2.1. Calcite. Calcite is the main carbonate cement type in sandstone reservoirs. Calcite is distributed in intergranular pores in the form of a continuous crystal and is a pore-type cement with point contact between particles (Figures 5(a) and 5(c)). Calcite is also mainly filled in various dissolution pores and intergranular micropores of authigenic minerals, such as dissolved pores of rock fragment (Figures 5(b) and 5(e)), quartz-dissolved pores (Figure 5(i)), and intergranular micropores of authigenic kaolinite (Figure 5(f)), and there is linear contact–suture contact between particles (Figures 5(e), 5(h), and 5(l)). In addition, calcite can fill fractures (Figure 5(g)). The calcite was filled in the dissolution pores of particles (Figures 5(j) and 5(k)).

4.2.2. Ferrous Calcite. Ferrous calcite is present only in member He 8 of certain wells, not in all samples. It is filled in primary or secondary pores, such as in the primary intergranular and dissolved pores of lithic particles (Figures 5(b) and 5(d)). Furthermore, the filling of carbonate cements is purplish red to red from the edge to the center (Figure 5(d)).

4.3. Distribution of Carbonate Cements. Based on core observation and handheld XRF data analysis, we conclude that the carbonate cemented zones in the Permian sandstone in the Ordos Basin are mainly distributed near the contact surface of sandstone and mudstone (Figure 3). Concerning sandstone reservoirs, the content of carbonate cements in coarse sandstones is higher than that in medium-sandstone reservoirs. The carbonate cement content is higher in the latter than in fine-sandstone reservoirs (Figure 3). In addition, there is continuity in the lateral distribution of carbonate cements in adjacent wells, but the layers with high contents of carbonate cements have different thicknesses due to the different sand/mud ratios (Figure 6). When the sand/mud ratio is relatively high, that is, there are more sandstones than mudstones, the content of carbonate...
Figure 5: The representative carbonate cements occurrence in the cast section, in the Permian of the Ordos Basin. (a) Calcites fill the intergranular pores and metasomatism the framework grains, SH114, 2149.5 m, Shihezi Formation 8th member (He 8), plane-polarized light (PPL). (b) Two stages of carbonate cement: one stage is purple red ferrous calcite filling in the intergranular pores, and the other stage is red calcite replacing mineral particles, SH168, 2533.4 m, He 8, PPL. (c) Chlorite film develops along the grain edge first, and then calcite develops in the intergranular pores, T38, 2489.9 m, Shanxi formation, PPL. (d) Ferrous calcite and red calcite are filled in the pores of particle dissolution, and ferrous calcite precedes calcite, T38, 2513 m, He 8, PPL. (e) Calcite is filled between grains and in the dissolved pores of debris, S19-31, 3017.22 m, He 8, PPL. (f) Chlorite forms along the grain edge, and kaolinite is filled between the grains, and the micropores between kaolinite are filled with calcite, Y105, 2284.29 m, He 8, PPL. (g) The fractures are filled with calcite, L50, 3835.6 m, He 8, PPL. (h, i) Calcite is filled between the grains and in the estuar-like solution pores of the quartz grains, Y105, 2269.3 m, He 8, PPL, cross-polarized light (XPL). (j, k) The calcite is partially dissolved after replacement of the detrital particles. (j) L50, 3835.6 m, He 8, PPL; (k) SH114, 2147 m, He 8, PPL. (l) The contact relationship of particles, L40, 4122.3 m, Shanxi Formation, XPL. Red represents calcite, purple represents ferrous calcite, and blue represents pores. Ca: calcite; Fc: ferrous calcite; K: kaolinite; Ch: chlorite.
cements in the coarse-grained sandstones close to the mudstones is higher, and the content of carbonate cements in the other sandstones is lower. The thickness of the strata where the carbonate cements reside is thicker. When the sand/mud ratio is relatively low, that is, there are less sandstones than mudstones, the thickness of the carbonate cemented layer in the sandstone is relatively thinner, but the carbonate cement content is higher (Figure 3).

4.4. Aqueous Fluid Inclusions in Carbonate Cements. The homogenization temperature of the aqueous fluid inclusions in carbonate cements reflects the temperature at which the carbonate cements were formed. The fluid inclusions captured during the formation of carbonate cements in a given geological period change from a homogeneous liquid phase to a gas–liquid phase owing to the changes in temperature and pressure changes at present. The inclusions can be rectangular, circular, elliptic, or irregular in shape, with a long axis range of 3–6 μm. The homogenization temperature of aqueous fluid inclusions in carbonate cements measured by temperature rise is given in Table 2. The homogenization temperature ranges from 67.9°C to 147.2°C, with a wide distribution range.

4.5. Simulation of Hydrocarbon Generation via Pyrolysis of Source Rocks. The TOC content and carbon isotope value of coal samples simulated with hydrocarbon generation were analyzed. The TOC content of the Shanxi and Taiyuan Formations was found to be 64.8% and 55.9%, respectively, and the carbon isotope value was found to be −23.0‰ and −22.7‰, respectively (Table 3). Thermal simulation results modeling the coal in the Permian Shanxi and Taiyuan Formations in the Ordos Basin show that carbon dioxide can be generated during the heating process of source rocks. The amount of gas generated at different simulated temperatures is different (Table 3). In the experiments of gold tube pyrolysis for hydrocarbon generation from coal in the Shanxi and Taiyuan Formations, the carbon dioxide yield of coal in the Shanxi Formation varied from 1.2 to 59.5 mL/g and that in the Taiyuan Formation varied from 1.7 to 55.6 mL/g.

The coal samples of the Shanxi and the Taiyuan Formations were analyzed through the simulation experiment of the gold tube thermal hydrocarbon generation, and the CO₂ obtained from the experiment was analyzed for carbon isotopes (Table 3). The carbon isotope distribution range of carbon dioxide generated by coal thermal simulation in the Shanxi Formation ranges from −17.4‰ to −19.7‰, with an average value of −18.2‰. Moreover, the carbon isotope distribution range of carbon dioxide generated by coal thermal simulation in the Taiyuan Formation ranges from −12.4‰ to −16.2‰, with an average value of −14.8‰. The carbon isotopes of CO₂ obtained from the hydrocarbon generation simulation of the Taiyuan Formation are heavier than those of the Shanxi Formation.

4.6. C and O Isotopes of Carbonate Cements. Carbon and oxygen isotopes of carbonate cements were measured in 19 Permian sandstone samples in the Ordos Basin (Table 4). The carbon isotope distribution of the Shanxi Formation ranges from −5.9‰ to −15.9‰, with an average of −12.3‰, and the oxygen isotope distribution ranges from
Table 2: Homogenization temperature of inclusions in carbonate cements of Permian sandstone in the Ordos Basin.

| No. | Well | Depth (m) | Horizon | Homogenizing temperature range (°C) | Homogenizing temperature (°C) |
|-----|------|-----------|---------|------------------------------------|-------------------------------|
| 1   | L26  | 4297.8    | He 8    | 125.3-126.4                        | 125.8                         |
| 2   | L26  | 4297.8    | He 8    | 146.9-147.6                        | 147.2                         |
| 3   | L50  | 3835.6    | He 8    | 123.0-123.3                        | 123.2                         |
| 4   | L50  | 3835.6    | He 8    | 139.6-140.6                        | 140.1                         |
| 5   | M105 | 2574.9    | Shanxi  | 98.5-99.6                          | 98.9                          |
| 6   | M105 | 2574.9    | Shanxi  | 120.0-125.9                        | 123.5                         |
| 7   | SD19 | 3024.82   | He 8    | 97.2-102.5                         | 99.85                         |
| 8   | SD19 | 3024.82   | He 8    | 123.2-125.8                        | 124.40                        |
| 9   | SH114| 2147      | He 8    | 66.8-68.3                          | 67.90                         |
| 10  | SH114| 2149.5    | He 8    | 108.8                              | 108.80                        |
| 11  | SH114| 2149.5    | He 8    | 126.5-128.2                        | 127.30                        |
| 12  | Y100 | 2526.3    | He 8    | 98.1-103.9                         | 101.00                        |
| 13  | Y100 | 2526.3    | He 8    | 126.4-127.3                        | 126.80                        |
| 14  | Y100 | 2553.6    | He 8    | 88.4-95.2                          | 91.80                         |
| 15  | Y100 | 2553.6    | He 8    | 128.7-128.9                        | 128.80                        |
| 16  | Y105 | 2269.3    | He 8    | 103.6-105.7                        | 104.90                        |
| 17  | Y105 | 2269.3    | He 8    | 131-136.6                          | 134.03                        |

Table 3: Thermal simulation data of Permian coal in the Ordos Basin.

| No. | Sample name | Sample type | Ro (%) | TOC (%) | $\delta^{13}C_{\text{coal}}$ (%) | Horizon | Simulate the temperature (°C) | Carbon dioxide production (mL/g) | $\delta^{13}C_{\text{CO}_2}$ (%) |
|-----|-------------|-------------|--------|---------|---------------------------------|---------|-------------------------------|----------------------------------|-------------------------------|
| 1   | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 350                           | 1.2                              | -17.5                         |
| 2   | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 375                           | 10.8                             | -18.0                         |
| 3   | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 400                           | 24.7                             | -17.9                         |
| 4   | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 420                           | 28.8                             | -19.4                         |
| 5   | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 440                           | 19.5                             | -17.8                         |
| 6   | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 460                           | 14.8                             | -17.4                         |
| 7   | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 480                           | 41.4                             | -18.2                         |
| 8   | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 500                           | 39.4                             | -17.8                         |
| 9   | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 520                           | 46.0                             | -17.9                         |
| 10  | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 540                           | 48.8                             | -18.1                         |
| 11  | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 560                           | 51.5                             | -18.6                         |
| 12  | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 580                           | 59.5                             | -19.7                         |
| 13  | L           | Coal        | 0.51   | 64.8    | -23.0                           | Shanxi  | 600                           | 58.0                             | -18.2                         |
| 14  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 350                           | 1.7                              | -12.4                         |
| 15  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 375                           | 4.1                              | -13.7                         |
| 16  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 400                           | 20.3                             | -14.8                         |
| 17  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 420                           | 27.7                             | -13.9                         |
| 18  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 440                           | 38.3                             | -15.8                         |
| 19  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 460                           | 35.2                             | -14.7                         |
| 20  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 480                           | 31.9                             | -15.1                         |
| 21  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 500                           | 15.6                             | -15.7                         |
| 22  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 520                           | 4.6                              | -14.9                         |
| 23  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 540                           | 46.4                             | -14.7                         |
| 24  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 560                           | 45.2                             | -15.1                         |
| 25  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 580                           | 55.6                             | -16.0                         |
| 26  | HC          | Coal        | 0.52   | 55.9    | -22.7                           | Taiyuan | 600                           | 45.9                             | -16.2                         |
Table 4: Carbon and oxygen isotopic composition of carbonate cements from Permian sandstone in the Ordos Basin.

| No. | Well | Depth (m) | Horizon | δ¹³C_{Carb}/PDB (‰) | δ¹⁸O_{Carb}/PDB (‰) | T (°C) |
|-----|------|-----------|---------|---------------------|---------------------|--------|
| 1   | L26  | 4297.8    | He 8    | -8.7                | -14.8               | 110.9  |
| 2   | L40  | 4122.3    | Shanxi  | -5.9                | -15.0               | 112.4  |
| 3   | L50  | 3835.6    | He 8    | -7.9                | -16.6               | 126.2  |
| 4   | M105 | 2574.9    | Shanxi  | -11.8               | -15.9               | 120.0  |
| 5   | SD19 | 3017.22   | He 8    | -15.9               | -16.0               | 121.3  |
| 6   | SD19 | 3024.82   | He 8    | -16.0               | -15.2               | 114.1  |
| 7   | SH114| 2147      | He 8    | -15.0               | -15.5               | 117.3  |
| 8   | SH114| 2149.5    | He 8    | -13.7               | -15.1               | 113.4  |
| 9   | SH168| 2526.7    | He 8    | -13.7               | -15.6               | 118.1  |
| 10  | SH168| 2533.4    | He 8    | -9.7                | -15.3               | 115.3  |
| 11  | T38  | 2451.3    | Shanxi  | -15.9               | -16.6               | 126.9  |
| 12  | T38  | 2489.9    | Shanxi  | -15.7               | -15.7               | 119.0  |
| 13  | T38  | 2513      | Shanxi  | -15.9               | -15.7               | 118.2  |
| 14  | Y100 | 2526.3    | He 8    | -7.5                | -16.8               | 128.3  |
| 15  | Y100 | 2553.6    | He 8    | -10.3               | -16.8               | 128.8  |
| 16  | Y100 | 2563      | He 8    | -11.2               | -16.8               | 127.9  |
| 17  | Y100 | 2565.7    | He 8    | -11.8               | -15.9               | 120.3  |
| 18  | Y105 | 2269.3    | He 8    | -12.1               | -16.0               | 121.2  |
| 19  | Y105 | 2284.29   | He 8    | -14.1               | -17.6               | 135.6  |

Note: oxygen isotope temperature $T (°C) = 16.45 - 4.31(\delta_1 - \delta_{w}) + 0.14(\delta_1 - \delta_{w})^2$ (65). In the formula, $\delta_1$ represents the oxygen isotope in CO$_2$ produced by the reaction of CaCO$_3$ and 100% phosphoric acid in the sandstone sample at 25°C, namely, $\delta^{18}O_{Carb}$; $\delta_{w}$ represents the oxygen isotope composition of the growth of aquatic organisms, that is, the oxygen isotope when carbon dioxide in water and carbon dioxide in air reach equilibrium at 25°C. At this time, $\delta_{w}$ is 0, which is the oxygen isotope value of present seawater.

−15.0‰ to −15.9‰, with an average of −15.6‰. The carbon isotopes of member He 8 samples range from −7.5‰ to −19.0‰, with an average of −12.4‰, and the oxygen isotopes range from −14.8‰ to −17.6‰, with an average of −16.4‰.

5. Discussion

5.1. Timing of Carbonate Cements. The homogenization temperature of aqueous fluid inclusions, oxygen isotopic data of carbonate cements, and microscopic characteristics of rock thin sections combined with the thermal and burial histories can be used to determine the formation time of carbonate cements [7, 8]. The measured homogenization temperature of the aqueous fluid inclusions in carbonate cements ranged between 67.9°C and 147.2°C (Table 2). In the Yishan Slope, the homogenization temperature distribution presents three peaks; the first peak is between 60°C and 70°C, the second peak is between 90°C and 110°C, and the third peak is between 120°C and 140°C (Figure 7). In the Tianhuan Depression, the measured homogenization temperature is mainly distributed in the latter two stages, i.e., 120–130°C and 140–150°C (Figure 7). Experimental results show that there are three phases of carbonate cements in the study area. The phase I calcite formed in the eodiagenesis may be precipitated directly from pore water. The carbonate cements of phase I are dissolved under the action of the acid fluid in the mesodiagenesis. Therefore, the carbonate cements in the sample are mainly phase II and phase III. The homogenization temperatures are consistent with the precipitation temperatures of carbonate calculated by oxygen isotopes (ranging from 110°C to 140°C) (Figure 8, Table 4). Observations in the rock thin sections also indicate that carbonate cements are dominated by phase II and phase III (Figure 5).

According to the formation temperature of the three phases of carbonate cements combined with the thermal and burial histories of the Ordos Basin [24, 44, 45], we conclude that the formation temperature of the phase I calcite in the Yishan Slope is 60–70°C and the formation time in Triassic is 217–237 Ma. The carbonate cements of phase II are mainly calcite and ferrous calcite (Figures 5(b) and 5(d)) that formed at a temperature of 90–110°C and developed at 163–191 Ma in the Jurassic, while phase III calcite was formed at a temperature of 120–140°C and developed at 123–132 Ma in the Cretaceous. In the Tianhuan Depression, the carbonate cements of phase II is mainly calcite, which formed at a temperature of 120–130°C and developed at 146–160 Ma in the Jurassic, and phase III is calcite, which formed at a temperature of 140–150°C and developed at 113–125 Ma in the Cretaceous (Figure 9).

5.2. Material Source of Carbonate Cements

5.2.1. Carbon Source. The carbon isotopic values of carbonate cements in sandstone can be used to indicate the carbon source [46, 47]. In this study, the carbon isotope values of coal samples from the Shanxi and Taiyuan Formations are −23.0‰ and −22.7‰, respectively. However, the carbon isotopic values of organic matter in the Toarcian Oceanic
**Figure 7:** Homogenization temperature distribution of inclusions in carbonate cements, Ordos Basin.

**Figure 8:** Comparison of homogenization temperature and oxygen isotope temperature of inclusions.
Anoxic Event in Yorkshire, UK recorded by previous studies ranged from $-27.0\%$ to $-33.2\%$ [48]. Compared with the latter, the carbon isotope value of the source samples in this study area is heavier, indicating that the organic matter in the coal samples of the Shanxi and Taiyuan Formations is not correlated with the Toarcian Oceanic Anoxic Event. The carbon isotopic distribution of carbonate cements in the sandstone in the area under study ranges from $-5.9\%$ to $-19.0\%$ (Table 4). The carbon isotopic value of the carbonate cements in the area under study is lighter than that of the normal carbonate cements, which indicates that the carbon source of the carbonate cements in the study area may be related to the decarboxylation of organic acids or hydrothermal fluids [8, 16, 21]. In addition, the study area covers almost the entire basin and the carbon isotopic values of the carbonate cements in the sandstone are all negative. However, in general, hydrothermal activity occurs only locally, not in the entire basin. In addition, there is no abnormally high temperature in the oxygen isotope calculation of carbonate cements and the homogenization temperature of fluid inclusions in carbonate cements and no hydrothermal minerals are observed in the thin section. Therefore, the carbon source of carbonate cements in the sandstone in the study area is not related to the hydrothermal activity but only to the decarboxylation of organic acids (Figure 10) [49].

Observations of thin sections indicate that the formation of carbonate cements occurs later than the overgrowth of quartz (Figure 11). In addition, Raman analysis of fluid inclusions in the quartz overgrowth shows that the fluid inclusions are mainly composed of carbon dioxide and methane (Figure 12). This suggests that hydrocarbon charging occurred during the quartz overgrowth, and the carbon dioxide and hydrocarbon gases (methane and ethane) in the fluid inclusions were generated by the process of hydrocarbon charging. This is consistent with the results of previous studies [24, 40, 50]. Carbon dioxide from oil and gas charging provided the carbon sources for late carbonate cements. In addition, carbon isotope tests of carbonate cements in the Permian sandstone reservoirs (Table 4) and the carbon dioxide obtained from the simulation of gold tube thermal hydrocarbon generation in Permian coal (Table 3) were conducted. The results show that the carbon isotopic characteristics of the carbonate cements in most samples are consistent with the carbon isotopic characteristics of the carbon dioxide generated during the thermal evolution of the source rocks (Figure 13). This is consistent with the Raman analysis results, indicating that the carbon source of carbonate cements is of organic origin, which is derived from CO$_2$ generated in the process of hydrocarbon generation from organic matter [16, 51].

However, the carbon isotopes of the carbonate cements in the sandstones of the Tianhuan Depression are heavier than those of the Yishan Slope (Figure 13). In addition, the CO$_2$/CH$_4$ value in the fluid inclusion of quartz overgrown in the Tianhuan Depression is lower than that in the Yishan Slope (Figure 12). This is because the source rocks and gas reservoirs mainly developed on the Yishan Slope at the center of the basin, while the Tianhuan Depression is located in the edge of the basin (Figure 1). During the migration of organic carbon dioxide from the center to the edge of the basin, part of the organic carbon dioxide is consumed [40], which makes the amount of organic carbon dioxide less and gives it the carbon isotopic characteristics of the diagenetic period. This view was also supported by temperature tests of inclusions in carbonate cements, which indicated...
that carbonate cements were formed slightly later in sandstone samples located in the Tianhuan Depression than those located in the Yishan Slope (Figures 7 and 9).

The carbon isotopes of the carbonate cements in the sandstone samples in the southeast of the study area became heavier with an increase in the distance from the Shanxi formation.

![Figure 10: Relationship of carbon and oxygen isotopes of carbonate cement, Ordos Basin. I: diagenetic carbonate; II: carbonate associated with biogas; III: carbonate associated with organic acid decarboxylation.](image)

![Figure 11: The contact relationship and inclusions distribution between quartz secondary enlargement and carbonate cements in Permian sandstone. (a, b) M105, 2574.9 m, Shanxi Formation, PPL; (c) SH114, 2149.5 m, He 8; (d) L26, 4297.8 m, He 8; (e) M105, 2574.9 m, Shanxi Formation; and (f) SH168, 2533.4 m, He 8. Q: quartz particles; Qo: quartz overgrowth; Ca: calcite.](image)
Figure 12: Raman characteristics of fluid inclusions of quartz in the middle and upper Permian sandstone of the Ordos Basin. (a) SH114, 2149.5 m, He 8; (b) Y100, 2563 m, He 8; (c) L26, 4297.8 m, He 8.
Formation source rocks (Figure 14(b)). This indicates that carbonate cement formation is influenced by the decarboxylation of organic matter in adjacent mudstones or source rocks [6, 52] and the organic CO$_2$ decreases as the distance from the source rock increases. Therefore, the carbon isotopes in this region also show a mixture of organic and inorganic isotopes (Figure 13); the farther away from the source rock, the more the amount of inorganic carbon.

5.2.2. Ca$^{2+}$ and Fe$^{2+}$ Source. Previous studies have shown that Ca$^{2+}$ and Fe$^{2+}$ can also originate from the dissolution of feldspar and the dissolution [53] and transformation of volcanic rock debris [3, 54]. They have also shown that the rock debris content is high, and the feldspar content is low in the study area. Moreover, Ca$^{2+}$ and Fe$^{2+}$ can be derived from the dissolution of carbonate cements by organic acid in mudstone [55]. Mudstones contain high clay mineral content and much lower smectite content in I/S (%S) [40]. Therefore, the cations of carbonate cements in sandstone are mainly derived from the conversion of smectite into illite in adjacent mudstones. In most cases, iron ions obtained from mudstone include Fe$^{3+}$, and the reduction of petroleum reduces Fe$^{3+}$ to Fe$^{2+}$ [50, 56]. In this study, the investigation of carbonate cements in sandstone reservoirs in the study area revealed that the Ca$^{2+}$ and Fe$^{2+}$ of the carbonate cements in the sandstone probably originate from the transformation of smectite into illite in the adjacent mudstone and the dissolution of early soluble components in sandstone [7, 53].

5.3. Evolution Process and Distribution of Carbonate Cements. The types, crystal shapes, development locations, and distribution patterns of carbonate cements formed in sandstone at different diagenetic stages are different [57]. In the study area, carbonate cements of eodiagenesis probably precipitated from early pore water with Ca$^{2+}$ and CO$_3^{2-}$ supersaturated [57]. As the burial depth increased, the formation temperature gradually rose and the organic matter in mudstone or source rock gradually matured. During this process, organic acids were formed [8], which migrated to the sandstone and dissolved the carbonate and soluble components of rock debris in the sandstone [8]. As the dissolution proceeded, quartz overgrowth occurred, the diagenetic environment changed from acidic to alkaline [24, 58], and the quartz dissolved [30]. Then, phase I oil and gas charging occurred during 175–210 Ma [44], bringing CO$_2$ from the hydrocarbon generation process of organic matter and Ca$^{2+}$ and Fe$^{2+}$ from the montmorillonite to illite transformation, thus forming the phase II calcite or ferrous calcite. At formation temperatures above 120°C, organic acids were unstable and decomposed [8, 59]. At this time, hydrocarbon generation continued in the coal of the Shanxi and Taiyuan Formations, and phase II oil and gas charging occurred [53]. Therefore, carbon dioxide and light hydrocarbon components entered the sandstone reservoir. The dissolution of CO$_2$ reduced the pH of the diagenetic environment; thus, the phase II carbonate and rock debris were dissolved [59]. Dissolution caused a pH increase, easing the precipitation of carbonate minerals. At this time, phase III calcite was
formed, with its carbon originating from the decarboxylation of organic acids.

The formation time and carbon isotopic values of carbonate cements in the Permian sandstone in the Ordos Basin are slightly different in different regions of the study area. The carbonate cements in the Yishan Slope developed early (217–237, 203–191, and 123–132 Ma) and had light carbon isotopes (−7.5‰ to −19.0‰), which were mainly from organic sources (Figure 13). Compared with the Yishan Slope, the carbonate cements in the Tianhuan Depression developed late (224–243, 146–160, and 113–125 Ma) and had heavier carbon isotopes (−5.9‰ to −8.7‰) and were a mixed organic and inorganic sources (Figure 13). Carbonate cements have a continuous distribution in the lateral direction (Figure 6) and mainly developed in the vertical direction near the contact surface of sandstone and mudstone.
In addition, with the increase in the distance from the source rock, the carbonate content is lower and the carbon isotope is heavier (Figure 14).

5.4. Implications for the Carbon Sources of Carbonate Cements in Petroliferous Basins. Previous studies have been conducted on carbonate cements of sandstone in various petroliferous basins [10, 16, 58, 60, 61]. In this study, the carbon isotopic distribution of carbonate cements in the Ordos Basin ranged from $-5.9$‰ to $-19.0$‰, and the carbon mainly originated from organic sources. The carbon isotopic data of the carbonate cements in the Cretaceous sandstones of the Songliao Basin indicate that the carbon of the carbonate cements is organic sources [60]. However, the carbon isotopic characteristics of the carbonate cements in the Triassic sandstones in the Ordos Basin indicate that the carbon sources are mainly of mixed organic and inorganic sources (Figure 15) [10]. Moreover, the carbon isotopes in the Bohai Bay Basin, Pearl River Mouth Basin, and Junggar Basin show that the carbon mainly comes from mixed organic and inorganic sources, while the carbon isotopes in a small part of the samples originate from organic sources (Figure 15). A possible reason for this phenomenon is that a large amount of organic acids were formed during the thermal evolution of type III kerogen [62, 63], which provided a rich source of organic carbon for the late carbonate cements. However, the amount of organic acids formed by type I kerogen and type II$_1$ kerogen was lower during thermal evolution [62, 63] and the CO$_2$ formed by the dissolution of early carbonate cements could not be eliminated over time. Therefore, the carbon source of late carbonate cements was a mixture of organic and inorganic sources. In summary, the source of oil and gas in sandstone is type III kerogen and the carbon of the carbonate cements in sandstone is of organic origin. The source of oil and gas in sandstone is of type I and type II$_1$ kerogen, and the carbon of the carbonate cements in sandstone originates from mixed organic and inorganic sources.

6. Conclusion

(1) The carbonate cements of the Permian sandstone in the Ordos Basin are mainly divided into three phases, and the latter two phases are dominant. Phase I is calcite, which probably is the primary precipitate obtained directly from pore water and formed at a temperature of 60–70°C. The calcite was formed at 217–237 Ma in the Yishan Slope. Phase II is mainly composed of calcite and ferrous calcite formed at 90–110°C and developed at 163–191 Ma in the Yishan Slope. However, in the Tianhuan Depression, the formation temperature was 120–130°C and the formation time was 146–
160 Ma). Phase III is calcite, which formed at 120–140°C and 123–132 Ma in the Yishan Slope and at 140–150°C and 113–125 Ma in the Tianhuang Depression. In conclusion, the formation temperature of carbonate cements in the Tianhuang Depression at the edge of the basin is higher than that of the Yishan Slope at the center of the basin and the formation time is later than that of the center of the basin.

(2) The distribution range of isotopic carbon in carbonate cements in the study area is between −5.9‰ and −19.0‰, which indicates that the carbon source is related to the decarboxylation of organic acids. In the Yishan Slope, the carbon isotopes in the carbonate cements mainly range from −7.5‰ to −19.0‰, indicating that the carbon mainly originates from organic sources. In the Tianhuang Depression, the carbon isotopes are mainly distributed in the range of −5.9‰ to −8.7‰, indicating that the carbon mainly originates from a mixture of organic and inorganic sources.

(3) Permian carbonate cementation in the study area is mainly dominated by phases II and III. The phase II of carbonate cements is mainly filled in the dissolution pores of organic acids, and CO$_3^{2-}$ originates from the CO$_2$ generated in the maturation process of organic matter. The phase III of carbonate cements is mainly filled in the dissolution pores of carbonic acid, and CO$_3^{2-}$ originates from the decarboxylation of organic acids. In addition, Ca$^{2+}$ and Fe$^{2+}$ mainly originate from the dissolution of early carbonate particles and the transformation of smectite into illite.

(4) Carbon isotopic analysis of the late diagenetic carbonate cements in sandstones from several petrolierous basins shows that carbon isotopic compositions are relatively light. This indicates that the carbon content of carbonate cements in sandstone mainly originates from organic sources or a mixture of organic and inorganic sources.

Data Availability

The data that support the findings of this study are available on request from the corresponding author.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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