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Research Article

Geochemical Characteristics of Formation Water in Carbonate Reservoirs and Its Indication to Hydrocarbon Accumulation

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The migration path of formation water plays an indispensable role in hydrocarbon accumulation and preservation. The hydrodynamic field controls the content of various ions in formation water and is an important participant in hydrocarbon evolution. Formation water can basically be used to judge the preservation status of oil/gas reservoirs, especially for carbonate reservoirs; the carbonate reservoirs are a typical example in the Gaoqiao area of the Ordos Basin, China. However, it is not easy to evaluate the sealing and integrity of the gas reservoir because hydrocarbon has experienced a multistage charging process and complicated later reconstruction. The geochemical characteristics of Ordovician formation water (100 brine samples from 67 wells in the Ma5 Member) are studied, and their chemical composition is analyzed in the Ordos Basin. The results show that formation water has high overall salinity and is the original sedimentary water of the carbonate reservoir, which is the sealing reservoir and can promote the accumulation of hydrocarbons. This is also associated with stronger water-rock reactions and diagenetic transformations, such as dolomitization. The main (TDS) range is from 40 to 150 g·L^{-1}, with an average of 66.16 g·L^{-1}; the Cl$^-$ content in the formation water samples is the highest, followed by Ca^{2+}, Na$^+$, Mg^{2+}, HCO$_3^-$, and SO$_4^{2-}$. In addition, the (Cl$^-$-Na$^+$)/Mg^{2+} ratio, Na$^+$/Cl$^-$ ratio, Mg^{2+}/Ca^{2+} ratio, and SO$_4^{2-}$ × 100/Cl$^-$ ratio are closely related to gas preservation. The indication function between chemical parameters of formation water and hydrocarbon dynamics can be better understood in carbonate reservoirs by analogy study, so as to improve the accuracy of discriminating favorable hydrocarbon accumulation areas.

1. Introduction

Formation water (also known as oilfield water) can be divided into two types: general formation water and special formation water [1–3]. General formation water refers to all groundwater in an oilfield, and special formation water represents the fluid in direct contact with the hydrocarbon development layers. According to the data of large gas reservoirs in the world, formation water is generally associated with hydrocarbons and there is no obvious interface; thus, the source of formation water is a very complicated problem, which has not been unified understanding. [4, 5]. Actually, the source of formation water is believed to have the following kinds: (1) primary sedimentary water ,(2) meteoric precipitation, (3) deep fluid, and (4) convert water. In fact, formation water is usually a mixture of these [1, 6]. As an important part of fluid in the sedimentary basin, formation water dynamics are critical in hydrocarbon accumulation [7, 8]. It is not only the driving force of hydrocarbon migration but also the carrier of hydrocarbon accumulation [1]. The ionic composition and movement trajectory of formation water are closely bound up with the accumulation, preservation, and destruction of hydrocarbon reservoirs [9].

The geochemical characteristics of formation water and its connection with hydrocarbon reservoirs have always been considered by geologists, especially with high salinity in carbonate rocks [10]. Due to the strong heterogeneity, water is generally preserved in situ, so the ion geochemical parameters
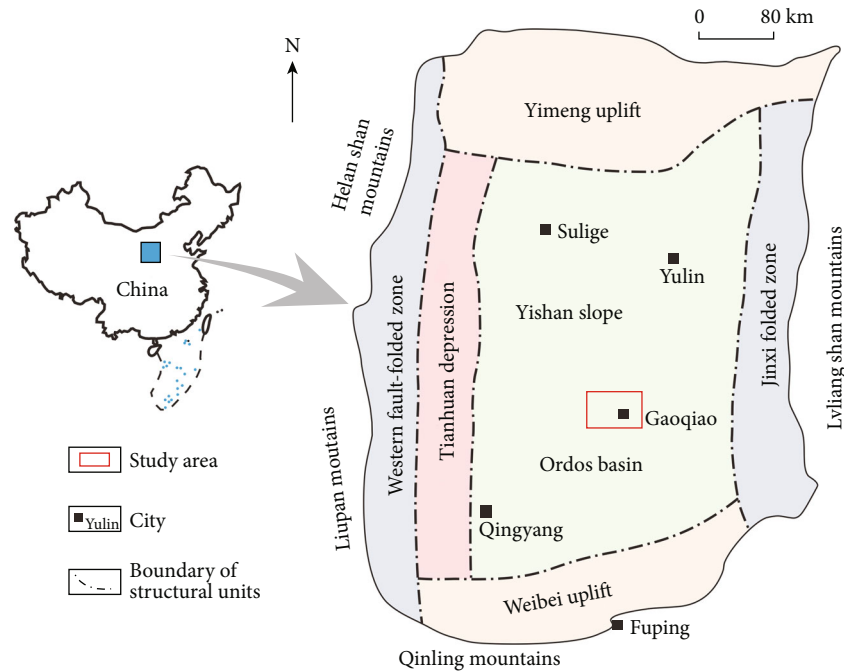
FIGURE 1: Location of the study area and tectonic units of the Ordos Basin.

can better reflect the key information of hydrocarbon migration and preservation [11–13]. In Houston's study [14], it was found that the concentration of calcium and sodium in formation water is the most critical chemical index to define reservoir sealing, reflecting the material balance and ion exchange of the rock salt layer. Méndez-Ortiz et al. studied the cretaceous fluids in the Mexico Basin and found that the water body in the high-yielding strata had a complex evolution [15], especially that the mixture of the original formation water with seawater and the water-rock interaction with the dolomite were more intense than those in other nonproductive strata. Bagheri et al. found that the concentration of sodium, calcium [16], and chlorine in basin fluids shows a mathematical transformation and linear relationship that can be used to explain albitization of calcium-rich plagioclase and can also be used to indicate oil-gas enrichment areas. By comparing the main ions and stable isotopes of formation water in different areas of the North Sea oil field, Younger et al. concluded that the formation water with high salinity is caused via high-temperature water-rock displacement about radioactive hot granite [17], which may be related to the Cenozoic tectonic uplift, thus facilitating the further preservation of the gas reservoir. The main geochemical processes of saline formation water are (1) evaporation of seawater or lake water, (2) dissolution of saline minerals, (3) metasomatism, such as albitization of feldspar and dolomitization of calcite, and (4) mineral transport by deep hydrothermal fluids [18–20]. The above reactions are bound to the salinity and ion content of formation water and can reflect the migration and preservation of hydrocarbon reservoirs to a certain extent [7, 21].

The Gaoqiao gas field is adjacent to the southern part of the Jingbian gas reservoir, one of the large proven gas fields in China [22], which is identified as a future natural gas

replacement development area in the Ordos Basin. However, complicated accumulation evolution and gas-water relationship limit the further development process of the Gaoqiao area [23–25]. Although there have been a number of studies on formation water and reservoir diagenesis characteristics [26, 27], the influence of formation water on hydrocarbon accumulation and indication of favorable areas are is clear. Therefore, this paper analyzes the chemical composition of formation water of Ma5 Member of Ordovician Majiagou Formation, a typical carbonate reservoir, evaluates formation water geochemical characteristics and distribution status, and makes a comparative study of geochemical characteristics and gas reservoir distribution. This work is expected to offer a new guidance for the target selection of carbonate gas reservoirs in the Ordos Basin.

2. Geological Setting

The Ordos Basin is the second largest sedimentary basin in China (Figure 1), covering a total area of 370000 km^2, located in the west of Lvliang Shan Mountains and east of the Helan Shan Mountains [28, 29]. Based on the tectonic morphology and evolution history of the basin, the basin can be divided into six different tectonic units: the Yimeng Uplift, the Western fault-folded zone, the Tianhuan Depression, the Weibei Uplift, the Jinxi folded zone, and the Yishan Slope [30]. Most of the natural gas is concentrated in the Yishan Slope and Tianhuan Depression. The Gaoqiao gas field is located in the middle of the Yishan Slope and is the most hydrocarbon-rich region [31].

During the Ordovician sedimentary circle, the Ordos Basin experienced multiple transgressions and regressions, accompanied by multiple secondary cycles during the rise
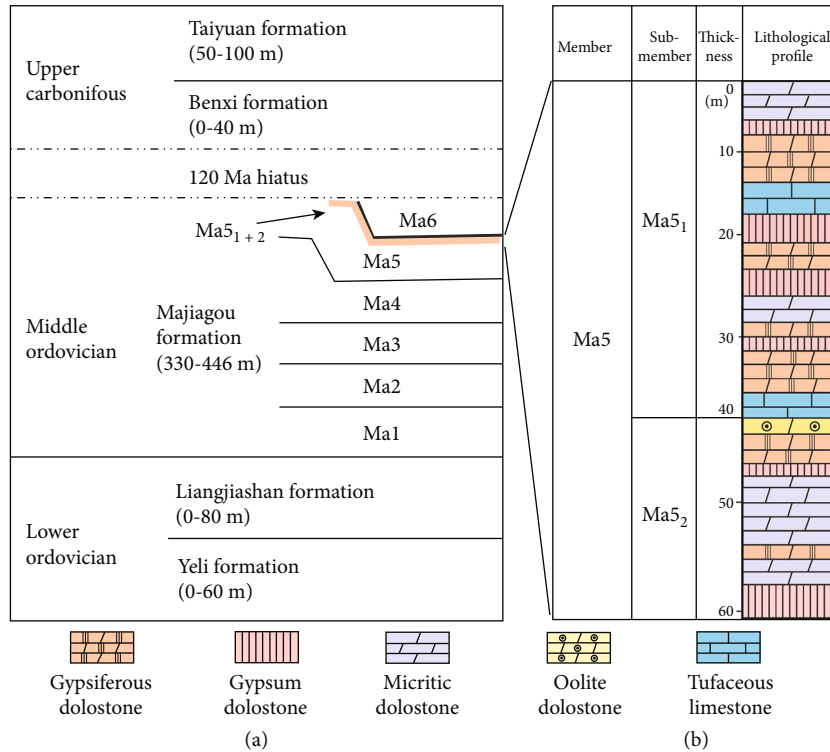
FIGURE 2: Stratigraphic development map of the Gaoqiao gas field. The studied Majiagou Formation (Ma) 5th Member (Ma5) is highlighted (Figure 2(b)).

and fall of the sea level [32]. After the Early Ordovician, Caledonian movement uplifted the basin completely and the Silurian to the Lower Carboniferous strata was missing [33, 34]. The top of the Ordovician suffered weathering and denudation for nearly 120 million years, forming the Ordovician weathering crust which is of great significance for gas accumulation [35].

Majiagou Formation is in unconformable with the overlying and underlying strata and deposited continuously, forming a set of evaporative layers dominated by carbonate rocks [36]. Majiagou Formation can be divided as the Ma1 to Ma6 Member from the bottom to the top (Figure 2). Ma5 strata are mainly composed of sea retreat and are divided into ten substrata from the bottom to the top [37, 38]. Gypsum dolomite, gypsum dolomite, and karst breccia are mainly developed in the $Ma5_{1+2}$ submember. In the Middle and Late Carboniferous, seawater intruded from the east and west at the same time and deposited a set of carboniferous and Permian coal-bearing strata alternately [39], which is the effective cap rocks of the Gaoqiao gas field.

3. Materials and Methodology

We collected 100 brine samples from 67 exploration wells in the Ma5 member. Firstly, the wellhead valve device was opened for several hours to wash pipeline residual wastewater (drilling fluid and wellbore effluent). Then, a bottom hole sampling device was used to sample the water in the Ma5. All water samples were screened through a 0.50 μm filter.
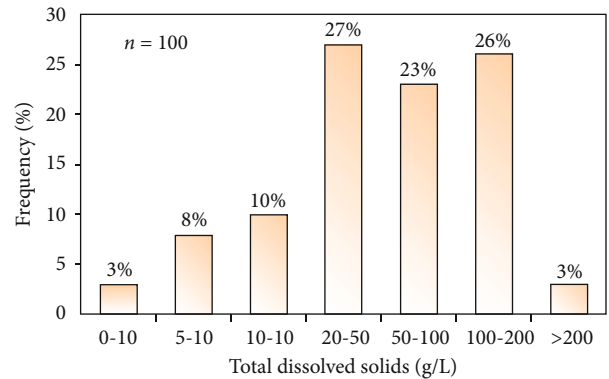

FIGURE 3: Frequency distribution map of total dissolved solids (TDS).

Saline samples were placed in 50 mL polypropylene bottles sealed with Parafilm paraffin paper until sample analysis.

The composition of the 100 brine samples was assayed at the State Key Laboratory (Northwest University, China); acid titration was suitable for the calibration of bicarbonate concentration; anions (Cl^-, SO_4^{2-}, and HCO_3^-) were measured by means of ion chromatography (PIC-10S); cations (Ca^{2+}, Mg^{2+}, Na^+, and K^+) were determined via AAS techniques [40], and the total dissolved solid (TDS) was obtained via the evaporation means. In addition, field data such as the types of exploration wells and water/gas production were provided by the Changqing Oilfield Company.
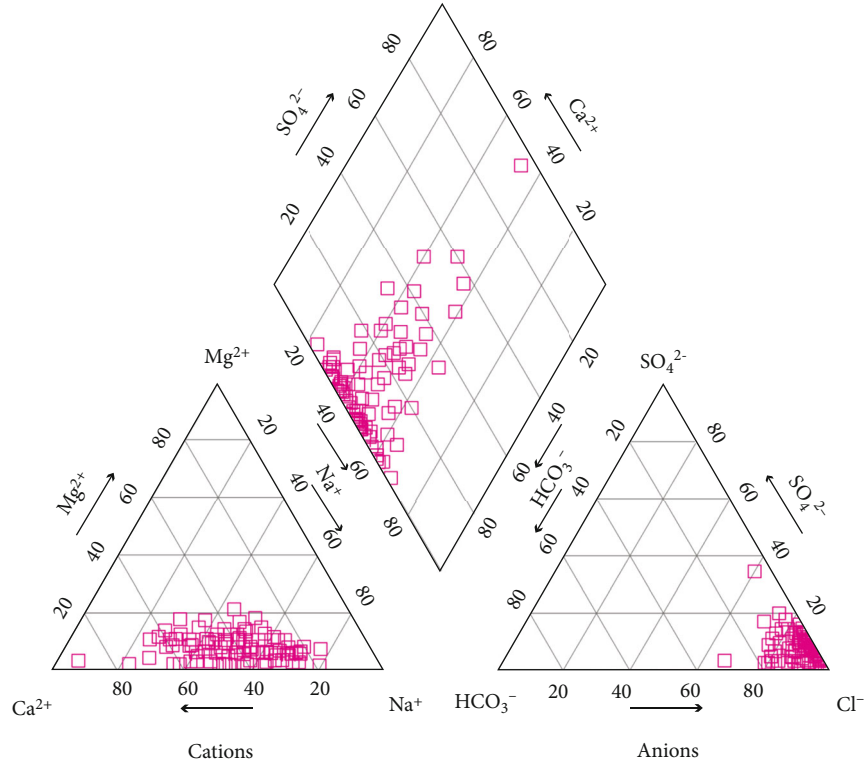
FIGURE 4: Piper diagram of formation water hydrochemical classification. This figure consists of a cation triangle (left), an anion triangle (right) and an anion-cation comprehensive map (middle).

4. Results and Discussion

4.1. Geochemical Characteristics

4.1.1. Total Dissolved Solids (TDS). The geochemical compositions of formation water in dolomite vary greatly (Figure 3), and the distribution of total dissolved solids (TDS) ranged from 4.32 to 213.74 g·L^{-1}, mainly from 40 to 150 g·L^{-1}, with an average of 66.16 g·L^{-1}. The TDS of formation water was significantly higher than that of surface water (usually about 0.1 g·L^{-1}), and 73% of formation water samples were also higher than those of sea water (30 g·L^{-1}). Highly mineralized formation water is obviously formed in a closed reductive environment with a high degree of metamorphism and evaporation [41].

The Majiagou Formation reservoir is primarily consisting of carbonate rock and karst breccia, and weathering crust karstification is very obvious. The fractures and dissolution pores are very common in dolomite and are covered by thick Carboniferous cap layers. As a result, the formation water of Majiagou Formation circulates and enriches in the deep layer, dissolving more minerals, resulting in high salinity of formation water.. Such a well-sealed hydrologic environment is conducive to gas preservation [42].

4.1.2. Ion Concentration. From the perspective of chemical reaction, the carbonate gas reservoir is essentially a mixed system of brine, dolomite, and natural gas and many chemical reactions have taken place in the geological period [43].
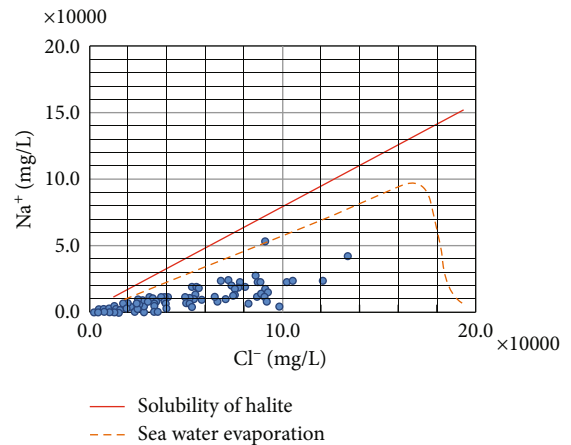

FIGURE 5: Comparison of Na$^+$ vs Cl$^-$ crossdiagram and theoretical evaporative dissolution curve.

During the hydrogeological development stages, such as deposition, burial, and water-rock reaction, the water concentration and ion characteristics change constantly, which can reflect the key information of hydrocarbon evolutionary history. The Cl$^-$ content in the formation brine samples is the highest, followed by Ca^{2+}, Na$^+$, Mg^{2+}, HCO$_3^-$, and SO$_4^{2-}$ (Figure 4).

The content of Cl$^-$ ion was in the range of 2589~133582 mg·L^{-1}, which accounted for 86% of the total anions on average, and the content of HCO$_3^-$ ion was in
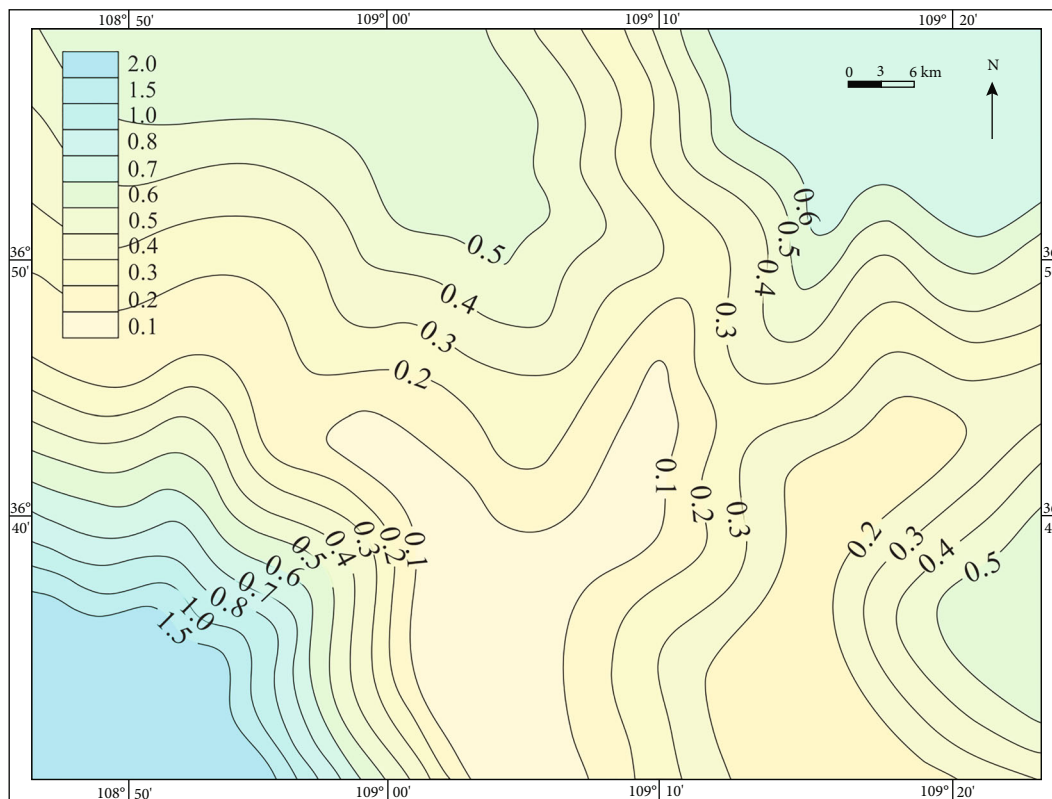
FIGURE 6: Distribution map of Na$^+$/Cl$^-$ ratios; the contour line represents specific numerical values.

the range of 12~1407 mg·L^{-1}, with an average of 481 mg·L^{-1}, which accounted for 2.35% of the total anions on average. The content of SO$_4^{2-}$ ion was also low, reflecting that the water environment is closed and anoxic. The main cations are Ca^{2+}, Na$^+$, and Mg^{2+}, and the Ca^{2+} content is between 913 and 186444 mg·L^{-1} with an average of 16190 mg·L^{-1}, while the Mg^{2+} ion content is low with an average of 722 mg·L^{-1}.

This combination of ion concentration may be the result of multiple factors during the burial and evolution of carbonate formation water in the Majiagou Formation. The Majiagou Formation is a typical paleo-karstic formation, and the superiority of Ca^{2+} concentration indicates the existence of a typical carbonate formation deposit; in addition, high K$^+$, Na$^+$, and Mg^{2+} concentrations also indicate intense chemical substitution reaction (e.g., deoinment dissolution, dolomitization, or illitization). The predominance of Cl$^-$ among the anions is principally because of the dominance of NaCl in Paleo-ocean and the lower solubility of sulfate (CaSO$_4$) and bicarbonate minerals (such as calcite and dolomite). Formation water is almost isolated formation water (CaCl$_2$), which has undergone strong metamorphism and belongs to original sedimentary water.

4.2. Ion Ratio Parameters.
Formation water chemical ion ratio parameters can reflect the hydrogeochemical status of formation water and the scale of water-rock interaction [8, 40]. The commonly used ion ratio parameters are sodium-chloride coefficient, desulfurization coefficient, magnesium-calcium coefficient, and metamorphism coefficient. These

characteristic parameters are often used to judge the direction of fluid movement and the strength and sealing of formation water activity and have a certain genetic relationship with hydrocarbon migration and accumulation and preservation.

4.2.1. Sodium/Chloride (Na$^+$/Cl$^-$) Ratio.
The Na$^+$/Cl$^-$ ratio is an important index of formation sealing, formation water activity, and reservoir hydrogeochemical environment. The higher the (Na$^+$/Cl$^-$) ratio is, the stronger the fluids are affected by the external factors and the more adverse it is to the preservation of hydrocarbons. The smaller the (Na$^+$/Cl$^-$) ratio is, which reflects the reduced water environment, the weaker the influence of external water and the more favorable it is for the preservation of hydrocarbons.

All of the data are below the seawater evaporation curve in the figure, indicating that the loss of Na$^+$ in the fluid is independent of chlorine solubility (Figure 5). Na$^+$-Ca^{2+} substitution during the feldspar metamorphism is not sufficient [7, 44–46]; the lower Na$^+$/Cl$^-$ confirms strong water-rock reaction occurring in this region. The (Na$^+$/Cl$^-$) ratio in the southwest area is relatively high (Figure 6), which is unfavorable to hydrocarbon accumulation. The (Na$^+$/Cl$^-$) ratio in central and eastern regions is lower, indicating that formation water is less affected by external water and strata sealing is better.

4.2.2. Magnesium/Calcium (Mg^{2+}/Ca^{2+}) Ratio.
The Mg^{2+}/Ca^{2+} ratio is a key parameter reflecting the development degree of secondary pores. Dolomitization and dissolution of calcite can optimize the physical properties of the reservoir
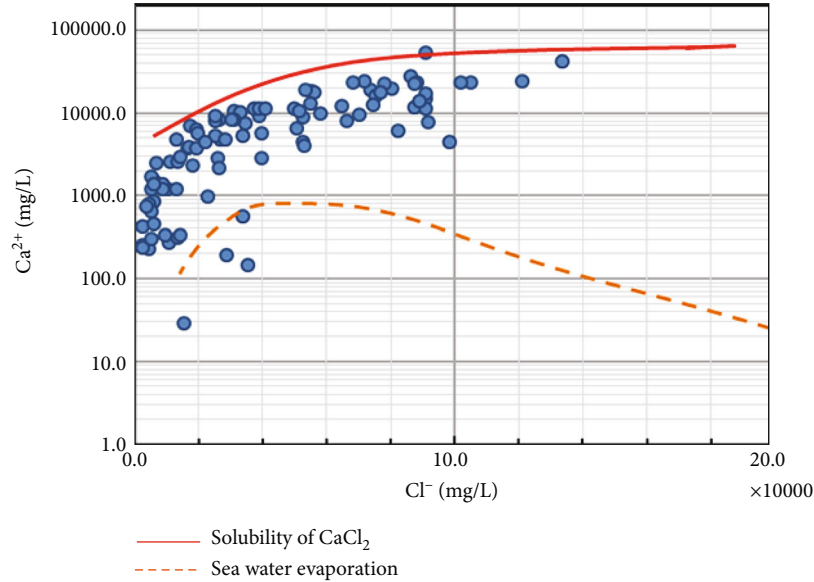
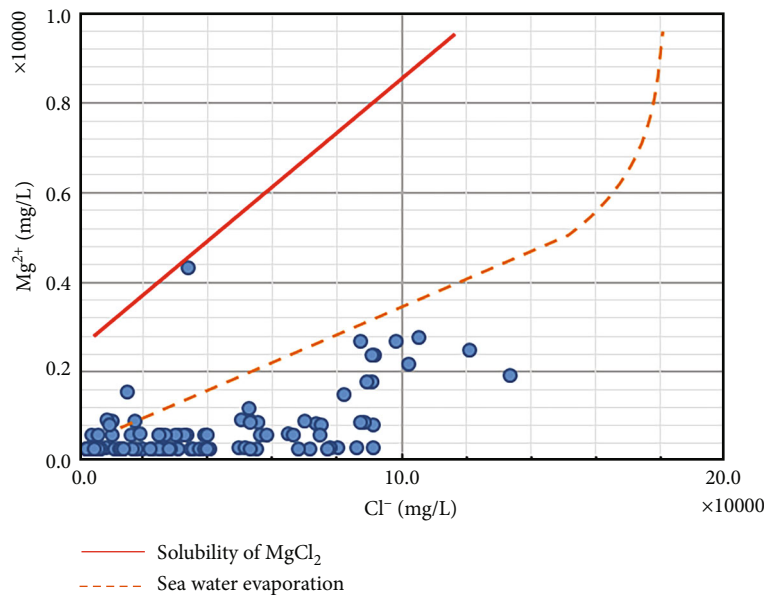Figure 7: Comparison of Ca^{2+} vs Cl^- crossdiagram and theoretical evaporative dissolution curve.

Figure 8: Comparison of Mg^{2+} vs Cl^- crossdiagram and theoretical evaporative dissolution curve.

and reduce the Mg^{2+}/Ca^{2+} ratio of formation water. Mg^{2+} consumption is related to dolomitization (equation (1)), while Ca^{2+} consumption is mainly related to calcite and laumontite precipitation [46, 47]. Therefore, we plotted the relationship between Ca^{2+} and Cl^- content (Figure 7); most of the data points lie between the $CaCl_2$ solubility curve and seawater evaporation trajectory, indicating that evaporation is not critical. Dolomitization is crucial in Ca^{2+} and Mg^{2+} replacement reactions (equation (1), Figure 8). Mg^{2+} was clearly lower than the seawater evaporation trajectory, suggesting that Mg^{2+} was replaced by Ca^{2+} in addition to evaporation.

Dolomitization occurs mostly in closed reservoirs dominated by evaporative environments, and the Mg^{2+}/Ca^{2+} ratio can represent capping conditions [6]. Well-enclosed reser-

voirs have lower Mg^{2+}/Ca^{2+} ratios, less interference from external water, and more hydrocarbon storage, as shown in the middle region of Figure 9.

$$CaCO_3(calcite) + Mg^{2+}(brine) \rightarrow CaMg(CO_3)_2(dolomite) + Ca^{2+}.$$
$$(1)$$
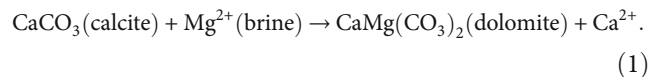
4.2.3. Desulfurization Coefficient $SO_4^{2-} \times 100/Cl^-$ Ratio. Sulphate in formation water is easy to be reduced in the anoxic reducing environment, which is conducive to the preservation of hydrocarbon, so desulfurization can be used as an environmental indicator [48]. Desulphurization usually takes place in the anoxic environment. In deep hydrocarbon
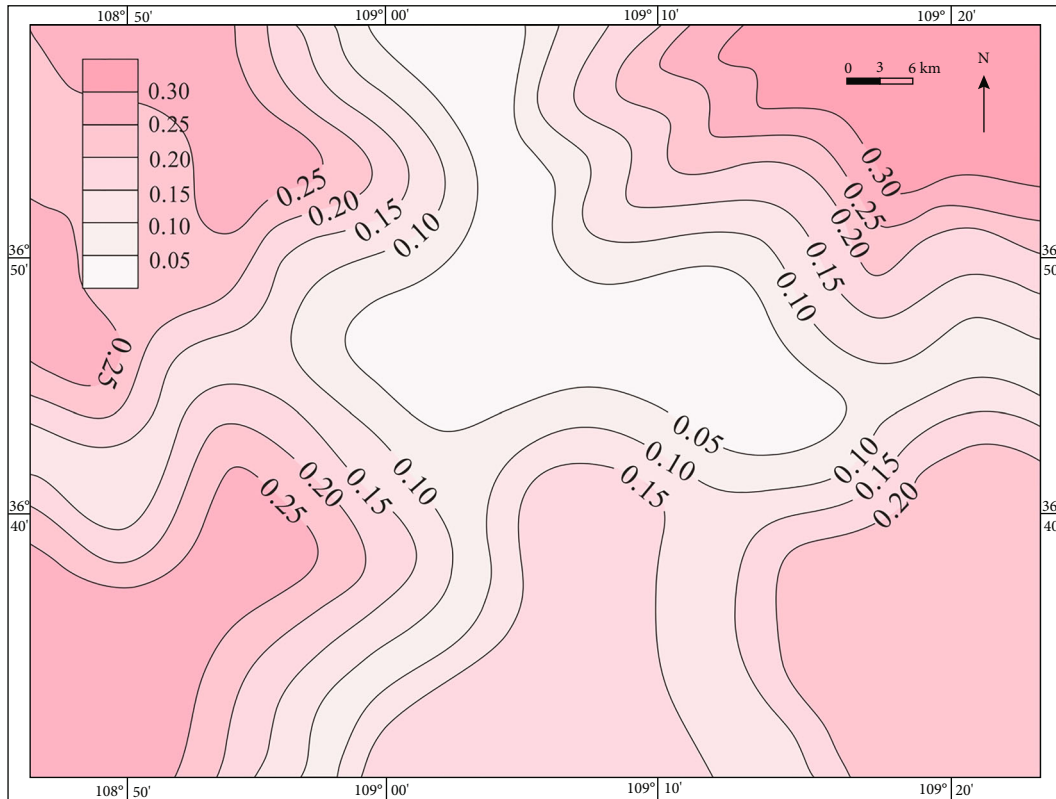
FIGURE 9: Distribution map of Mg^{2+}/Ca^{2+} ratios; the contour line represents specific numerical values.
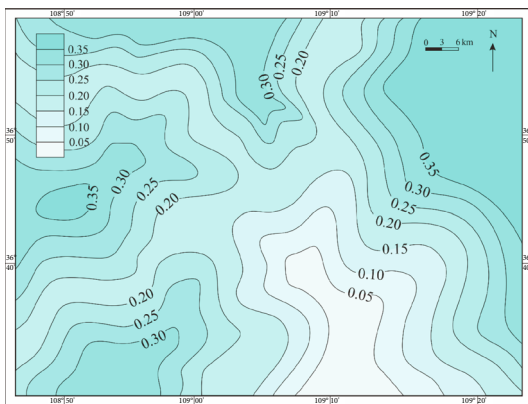

FIGURE 10: Distribution map of $SO_4^{2-} \times 100/Cl^-$ ratios; the contour line represents specific numerical values.
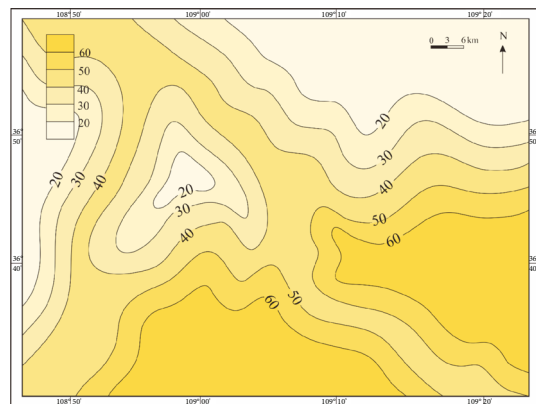

FIGURE 11: Distribution map of $(Cl^- - Na^+)/Mg^{2+}$ ratios; the contour line represents specific numerical values.

reservoirs, the smaller the desulfurization coefficient, the better the formation sealing, indicating a higher degree of formation water reduction, which is more beneficial to the preservation of large gas reservoirs [49]. The desulfurization coefficient in the middle and southern region is less than 0.20, which is obviously lower than that in the present sea and river water (this value is 12 [50]), reflecting that the formation sealing in this region is good, which is conducive to the preservation of hydrocarbons (Figure 10).

4.2.4. Metamorphic Coefficient $(Cl^- - Na^+)/Mg^{2+}$ Ratio. The metamorphic coefficient can represent the intensity of the water-rock chemical process and the degree of ion replace-

ment during the migration of formation water [51]. The longer the water-rock reaction time, the more complete the ion exchange will be under the condition of closed and deep burial. Correspondingly, the higher the formation water $(Cl^- - Na^+)/Mg^{2+}$ ratio is, the more favorable it is to hydrocarbon preservation.

The $(Cl^- - Na^+)/Mg^{2+}$ ratio of the $Ma5_{1+2}$ Member is between 13 and 65, with an average of 38.3. The metamorphic coefficient is high from east to south, and these areas are located in the tectonic highland areas such as karst landforms and hills and are weakly eroded by meteoric waters. Fluid disconnection from other open systems in the subsurface results in strong water-rock interactions in this closed
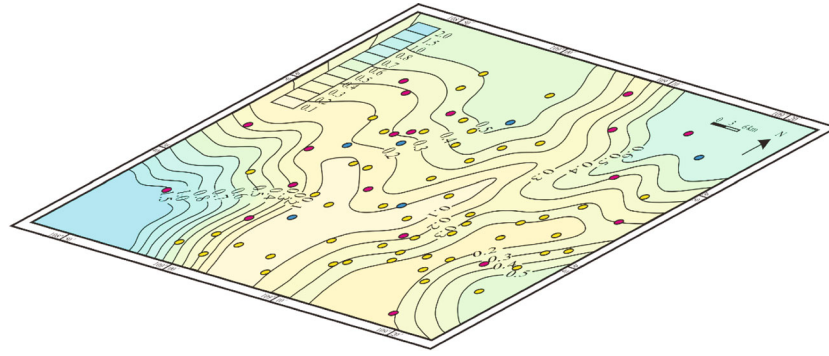
Gas well
Gas & Water well
Water well

(a) Na^+/Cl^-
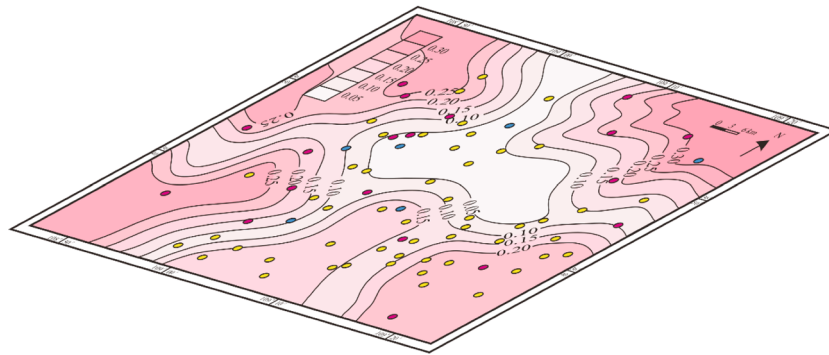
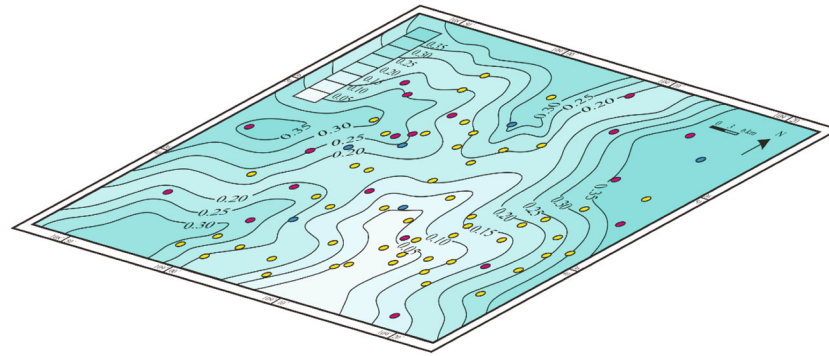Gas well
Gas & Water well
Water well

(b) Mg^{2+}/Ca^{2+}

Gas well
Gas & Water well
Water well

(c) $SO_4^{2-} \times 100/Cl^-$

FIGURE 12: Continued.
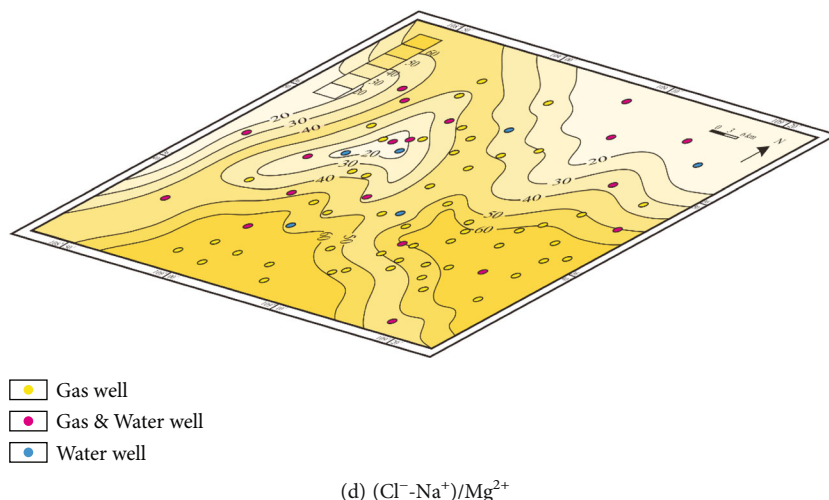
(d) $(Cl^- - Na^+)/Mg^{2+}$

FIGURE 12: Correlation between geochemical properties of formation water and gas/water well distribution.

formation. Therefore, Na^+ and Mg^{2+} are mainly replaced by Ca^{2+}, so the metamorphic coefficients are high. The north-eastern region of the study area has lower metamorphism, which is not conducive to hydrocarbon accumulation (Figure 11).

4.3. Correlation between Gas Accumulation and Formation Water Geochemical Characteristics. The formation water of the Majiagou Formation in the Gaoqiao area has undergone several hundred million years of geological transformation since the deposition of sediments. The initial marine sedimentary water has undergone the influence of various tectonic movements, including the alternation of meteoric water and surface water in the supergene denudation period, the intervention of hydrothermal fluid, and the complex interaction of water-rock and hydrocarbon in the burial period, resulting in the fundamental change of water chemical composition.

Based on the above formation water characteristic research, gas/water wells in the Gaoqiao area are projected onto the plane distribution; the correlation between water geochemical properties and gas preservation was analyzed (Figure 12). Among the 76 gas testing wells in the study area, there are 51 gas wells, 19 gas-water wells, and 6 water wells. There are more gas wells in the region where the (Na^+/Cl^-) ratio is less than 0.3, mainly concentrated in the southern region, while the number of water-producing wells in the northwest region is greater than 0.3. The number of producing water wells in the region where the (Mg^{2+}/Ca^{2+}) ratio is greater than 0.2 increases, the proportion of gas wells in the region where the $SO_4^{2-} \times 100/Cl^-$ ratio is less than 0.25 which is 85%, and the proportion of gas wells in the region where the $(Cl^- - Na^+)/Mg^{2+}$ ratio is greater than 40 which is as high as 90%, which can be a good indicator of the distribution characteristics of gas or water wells.

The results show that most gas wells are concentrated in regions with a low (Na^+/Cl^-) ratio, low (Mg^{2+}/Ca^{2+}) ratio, low $SO_4^{2-} \times 100/Cl^-$ ratio, and high $(Cl^- - Na^+)/Mg^{2+}$ ratio, while water wells are mostly concentrated in regions with a high (Na^+/Cl^-) ratio, high (Mg^{2+}/Ca^{2+}) ratio, high $SO_4^{2-} \times$ 100/Cl^- ratio, and low $(Cl^- - Na^+)/Mg^{2+}$ ratio. The preservation of natural gas in the carbonate reservoir is closely bound to the properties of formation water, which can well indicate preservation conditions of the gas reservoir, which also provides great help for the next gas reservoir exploration and development.

5. Conclusions

(1) Formation water plays a crucial role in hydrocarbon migration, accumulation, and preservation for carbonate reservoirs. This paper analyzes and interprets the geochemical characteristics of formation water about the Gaoqiao area in the Ordos Basin, China. The test results showed that the geochemical characteristics varied significantly in the Ma5 Member; the main (TDS) range is from 40 to $150\,g\cdot L^{-1}$, with an average of $66.16\,g\cdot L^{-1}$; the Cl^- content in the formation water samples is the highest, followed by Ca^{2+}, Na^+, Mg^{2+}, HCO_3^-, and SO_4^{2-}.

(2) In formation water, Na^+ and Mg^{2+} ions were depleted, while Ca^{2+} ions were enriched. The formation water is original sedimentary water in carbonate reservoirs, and its genesis is related to evaporation effect and water-rock interaction effect, having experienced intensive metamorphism effect

(3) The relationship between formation water geochemical characteristics and distribution of production well types is studied. The Na^+/Cl^- ratio, Mg^{2+}/Ca^{2+} ratio, and $SO_4^{2-} \times 100/Cl^-$ ratio are negatively correlated with the number of gas wells, while the metamorphic coefficient is positively correlated with the number of gas wells. Through this study, it can be found that the formation water geochemical parameters have a certain correlation coefficient with hydrocarbon preservation, so they can be regarded as discriminative markers for identifying favorable areas of carbonate reservoirs

Data Availability

The data used to support the findings of this study are included within the supplementary material (available here). The material is the result of the original geochemical parameters of formation water measured experimentally.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgments

Supplementary Materials

Supplementary Table 1: the ion concentrations of the formation water samples. (Supplementary Materials)

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