

Article

Diagenetic Fluids and Multiphase Dolomitizations of Lower Paleozoic Carbonate Reservoirs, Offshore Bohai Bay Basin, Northern China

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Abstract: Dolomitization is a critical diagenetic alteration that impacts the formation of carbonate hydrocarbon reservoirs. In the offshore Bohai Bay Basin, the Lower Paleozoic carbonate reservoirs in buried hill traps, and the basement highs unconformably overlain by younger rock units, are emerging as a prospective target and predominantly occur in dolomite layers. Meanwhile, the formation mechanisms of the dolomite are not clear, which affects the understanding of the occurrence of deep dolomite reservoirs and hinders oil and gas exploration. Based on comprehensive observations of the thin sections of the carbonate samples, the dolomite types were meticulously categorized into micritic dolostone, fine-crystalline dolostone, and saddle dolomite. Then, carbon, oxygen, and strontium isotope and trace elements were examined to elucidate the dolomitization fluids and propose diagenetic models for the three kinds of dolomite formation. The mineralogical and geochemical evidence reveals that there were two kinds of dolomitization fluids, including penecontemporaneous seawater, and hydrothermal fluid. The diagenetic fluid of the micritic dolostone and fine-crystalline dolostone both involved penecontemporaneous seawater, but fine-crystalline dolostone is also affected by later burial dolomitization processes. The saddle dolomite, filling in pre-existing fractures or dissolution pore cavities, is attributed to a hydrothermal fluid associated with magmatic activities. Notably, the extensive layered fine-crystalline dolostone was the predominant reservoir rock. The initial mechanism for its formation involves penecontemporaneous seepage reflux dolomitization, which is superimposed by later burial dolomitization. The burial dolomitization enhanced porosity, subsequently facilitating the formation of a fracture-related dissolution pore cavity system, and partly filled by saddle dolomite during the Cenozoic hydrothermal events. The findings highlight that the layered fine-crystalline dolostone that underwent multiphase dolomitization is the most potential target for hydrocarbon exploration.

Keywords: diagenetic fluids; dolomitization; carbonate reservoir; Lower Paleozoic; Bohai Bay Basin



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1. Introduction

Dolomite rocks are an important kind of carbonate reservoir rock for oil and gas [1–4]. Although it has been intensively researched for over more than 200 years, the origin of dolomite remains subject to considerable controversy within the geological community and constitutes as the focal point of the carbonate reservoir formation mechanism, significantly

influencing the prediction of hydrocarbon targets [5–7]. As we know, various models of dolomite genesis were proposed, including the evaporative pumping model [8], seepage reflux model [9], mixing marine-meteoric water model [10], sabkha model [11], burial dolomitization model [12], tectonic-hydrothermal model [13], seawater dolomitization model [14], and others. Up to now, there is still confusion regarding the proposed models about the source of Mg^{2+} and the mechanism through which dolomitizing fluids undergo. Dolomitization can take place from early diagenesis to the deep burial phase. The fluid conditions of these models and their combined effects during geological evolution are different across basins and stratigraphic units, inducing the intricate nature of dolomite formation.

The exploration discoveries revealed the Lower Paleozoic carbonate rocks occurring in the buried hills, and the basement highs unconformably overlain by younger rock units, are a prospective target in the Bohai Sea, such as the BZ21-1, CFD2-1, and CFD2-2 traps. The hydrocarbon is mainly produced in dolomite reservoirs [15]. The formation mechanisms of these dolomite reservoirs are not well understood, which thereby impedes hydrocarbon exploration and predictive efforts for Lower Paleozoic carbonates of the Bohai Sea. This study conducted observations on the Lower Paleozoic dolomite coring intervals and collected core samples. Based on thin-section identification, the dolomite samples were categorized into micritic dolostone, fine-crystalline dolostone, and saddle dolomite. Saddle dolomite is characterized by its curved crystal faces and sweeping extinction, indicating formation at elevated temperatures typically related to hydrothermal activity [16]. Stable isotopes act as crucial geological tracers that enable the investigation of paleoenvironments, diagenetic processes, fluid sources, and hydrocarbon migration pathways [17–19]. Carbon, oxygen, and strontium isotope, along with trace elements, were tested to investigate the characteristics of dolomitizing fluids and the origins of the three types of dolomite formation.

2. Geological Setting

The Bohai Bay Basin is located in the eastern part of the North China Block (Figure 1A). As a typical rift basin, it ranks among the most significant oil- and gas-producing areas in China. Bohai Sea, the offshore part of Bohai Bay Basin, has emerged as the most prominent region for oil and gas discoveries in China in recent decades, holding considerable potential for future exploration. The western Bozhong Sag is the main hydrocarbon-rich tectonic unit (Figure 1B). Especially, the buried hills in the basement of the Cenozoic Basin were discovered to have significant reservoir potential, including the Archean metamorphic basement, Paleozoic carbonates, and Mesozoic volcanic rocks [20,21].

The Lower Paleozoic carbonates (Figure 1C), primarily comprising inter-bedded dolomite and limestone, underwent significant diagenetic alteration and multiple tectonic events [22]. Tectonically-induced fracture systems and fracture-related karstification are the effective reservoir spaces for hydrocarbon accumulation. The Lower Paleozoic strata are further divided into Cambrian and Ordovician stratigraphic units. From the bottom up, the Cambrian sequence comprises the Mantou, Zhangxia, Gushan, Changshan, and Fengshan formations, while the Ordovician sequence includes the Yeli, Liangjiashan, and Majiagou formations.

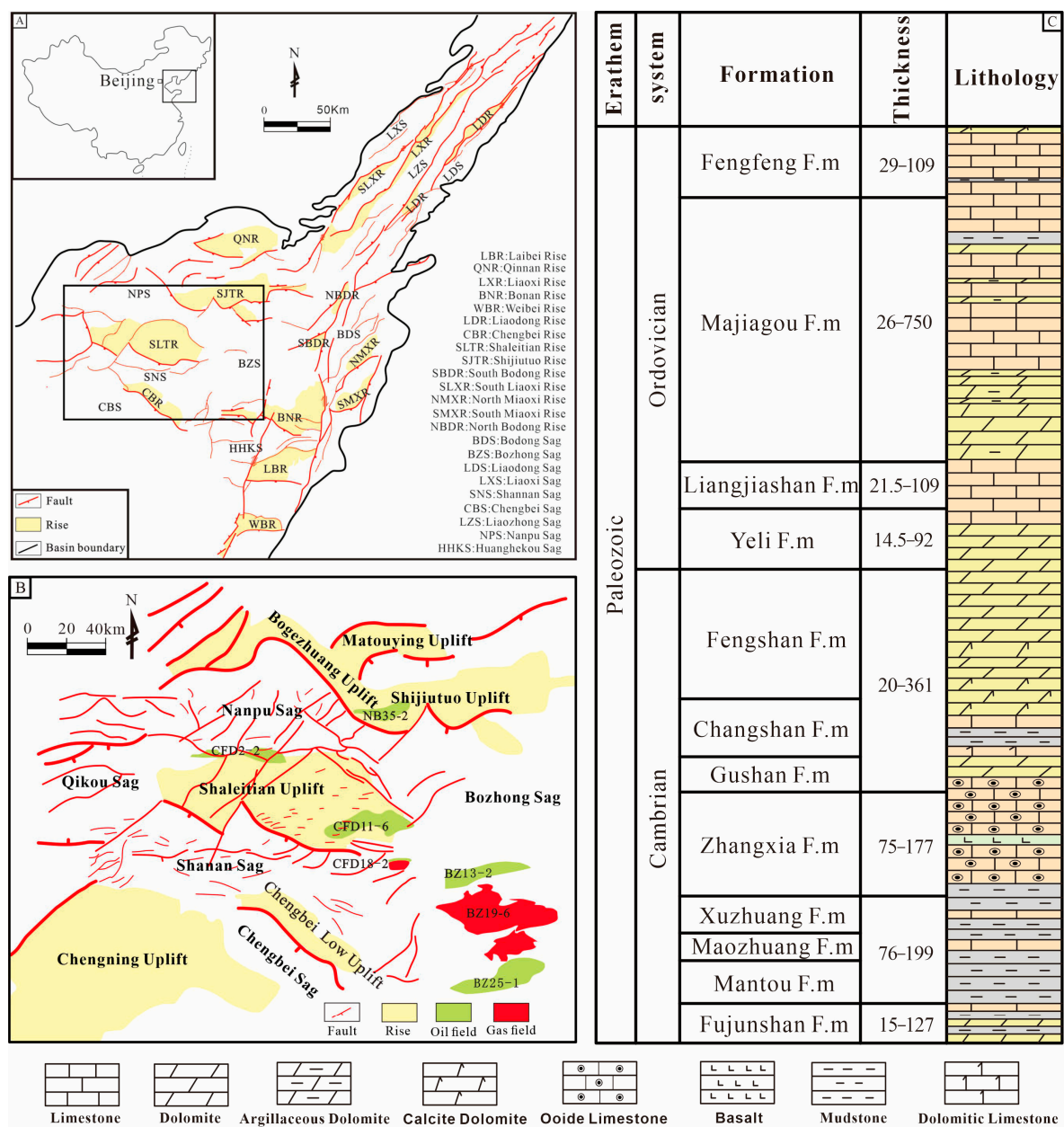


Figure 1. (A) Geological map of the offshore Bohai Bay Basin; (B) geological map of the study area; (C) stratigraphic lithology column for the Lower Paleozoic rocks.

3. Samples and Research Methods

Detailed core observation and core-photographing were conducted in the core repository of CNOOC Tianjin Branch, and 63 samples of the Lower Paleozoic cores were collected from 7 boreholes. During core observation and sampling, initial lithological assessments were made using a magnifying glass, dilute hydrochloric acid, and magnesium reagent to evaluate and document the properties of hand specimens. Bulk samples were selected for petrophysical analysis of carbonates, while smaller samples were processed into both regular and cast thin sections. The lithology, macroscopic structures, and microscopic features of the carbonate rocks were determined through a combination of hand specimen and microscopic observations, supplemented by cathodoluminescence and scanning electron microscopy techniques with detailed documentation recorded throughout. The identification of regular and cast thin sections was performed at the Polarizing Microscope Laboratory within the Institute of Sedimentary Geology at Chengdu University of Tech-

nology. Then, 52 thin sections were sent to the Cathodoluminescence Laboratory in the College of Energy at Chengdu University of Technology for cathodoluminescence analysis. The analysis was performed using a CL8200MK5 cathodoluminescence microscope under controlled conditions: ambient temperature of 20 °C and humidity of 47%. The experimental process is to expose for 5 s under the conditions of beam current 250 µA and beam voltage 15 kV after the vacuum is evacuated.

Furthermore, core samples were obtained from three structural belts within the study area: BZ21-1, CFD2-1, and CFD2-2. These selected samples underwent carbon, oxygen, and strontium isotope analysis and detailed trace element analysis [23–25]. At the Isotope Testing Center within the Department of Geochemistry at Chengdu University of Technology, $\delta^{18}\text{O}/\delta^{13}\text{C}$ values were determined for 13 selected samples. The initial step involved cleaning the hand specimens with distilled water to thoroughly remove surface contaminants. Sampling points on the matrix were carefully selected from unweathered surfaces to ensure purity. For vein samples, fine powder was precisely obtained by drilling using a handheld micro-drill. Subsequently, the powder samples were dried in an oven at 105 °C for 2 h and then stored in a desiccator for subsequent use. A 100% phosphoric acid solution was prepared and mixed with the powder samples in a constant temperature water bath maintained at 25 °C for 36 h to facilitate CO_2 collection and purification. Then, the CO_2 was analyzed for C and O isotopes using an MAT251 mass spectrometer with an analysis error less than 0.02‰ according to the VPDB standard, following the method outlined by McCrea [26].

The strontium isotope analysis of the 13 powder samples was performed at the National Key Laboratory of Oil and Gas Reservoir Geology and Exploitation (Chengdu University of Technology). The procedure involved dissolving the powder samples in 1 mol/L hydrochloric acid for 2 h. After the step of dissolution, the supernatant was separated and purified using AG50W $\times$ 8 (h+) cation exchange columns; subsequently, the $^{87}\text{Sr}/^{86}\text{Sr}$ values were determined using a Triton Plus thermal ionization isotope mass spectrometer. To ensure both precision and accuracy, the analysis was validated through repeated measurements of the international standard sample NBS-987, with a reported standard error (2σ) of 1.218×10^{-5} .

The trace and rare earth elements of the dolomite sample were tested by the in situ LA-ICP-MS at Wuhan SampleSolution Analytical Technology Co., Ltd., Wuhan, China. Detailed instrument parameters and analytical procedures were described in the prior study [27]. The GeolasPro laser ablation system comprises a COMPexPro 102 ArF 193 nm excimer laser and a MicroLas optical system, using an Agilent 7700e ICP-MS. Helium served as the carrier gas, while argon was used as the makeup gas to adjust sensitivity during the laser ablation process; these two gases were combined via a T-connector before entering the ICP [28]. In this analysis, a laser spot size of 40 µm and a frequency of 10 Hz were utilized. For trace element analysis in individual minerals, external calibration using external standards only was performed with glass standards BHVO-2G, BCR-2G, and BIR-1G [27]. Each time-resolved analysis included approximately 20–30 s of blank signal acquisition followed by 50 s of sample signal collection. Offline processing of analytical data, including selection of sample and blank signals, correction for instrument sensitivity drift, and calculation of elemental concentrations, was performed using ICPMSDataCal software (Version 10.6) [27].

4. Results

4.1. Petrographic Characteristics

The Lower Paleozoic rocks exhibit a diverse range of lithologies, consisting mainly of dolostone and limestone. The dolomite rocks predominantly manifest in substantial

strata, with typical thicknesses ranging from 0.3 to 2 m. Petrographic analysis reveals that dolostone layers comprise up to 95% of the dolomite, suggesting extensive dolomitization in the study area. The observations of the dolostone samples through a microscopic thin section demonstrate that dolomite displays variability in four distinct types (Figure 2), including micritic dolostone, granular dolostone, fine-crystalline dolostone, and saddle dolomite, and the granular dolostone is less common. Both core and thin section examinations indicate that dolostone layers experienced extensive meteoric dissolution, leading to the formation of numerous fracture-related dissolution pores. These pores act as effective reservoir spaces for natural gas storage. In drill cores and thin sections, diagenetic fractures can be observed at a small scale with irregular morphology present in various rock types. The characteristics of these fractures reveal the structural and hydrogeological properties of dolomitic reservoirs and indicate the formation mechanisms of fractures and their impact on fluid flow. Specifically, many fractures are formed by pressure fracturing, often presenting an irregular network or dendritic distribution that cuts across grains, with bead-like vugs along the fracture surfaces due to dissolution, further emphasizing the importance of pressure and dissolution processes in fracture development.

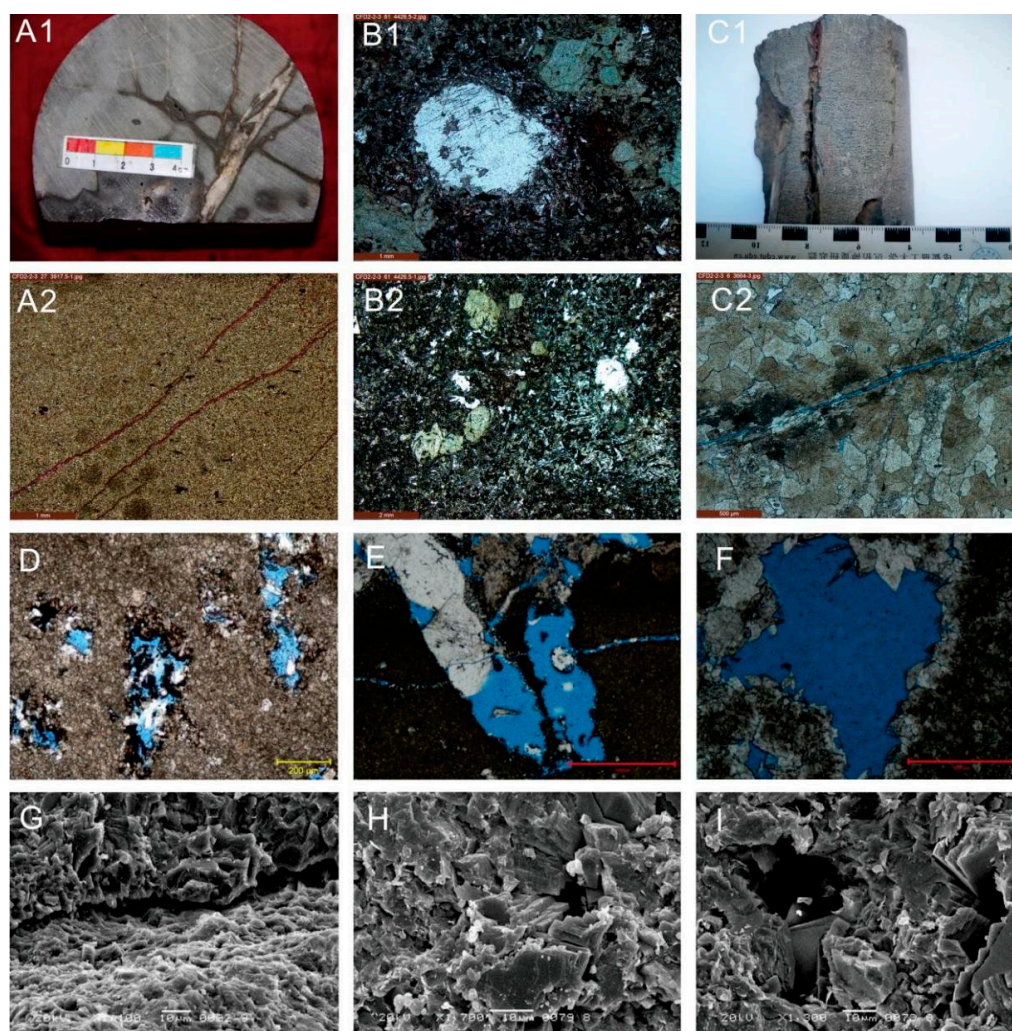


Figure 2. Microscopic and core characteristics of the Lower Paleozoic dolostones. (A1,A2): Micritic dolostone; (B1,B2): granular dolostone; (C1,C2): crystalline dolostone; (D): dissolution pore in fine-crystalline dolostone filled with dolomite, CFD 2-1-2, 3704.9 m; (E): dissolution pore in fine-crystalline dolostone with residual saddle dolomite, CFD 2-1-2, 3707.8 m; (F): residual intergranular dissolution pore with serrated edge dolomite crystals, CFD 2-1-2, 3709.8 m; (G–I): dolostones with fractures and dissolution pores in SEM.

Micritic dolostone (Figure 2(A1,A2)): This type is distinguished by its micritic texture, primarily consisting of micritic dolomite, argillaceous material, and bioclastic fragments. The argillaceous content exhibits evidence of recrystallization and a preferred directional distribution. The core appears gray-white, containing numerous calcite-filled fractures. Under SEM, the crystals are observed to be closely interlocked. The mineral content is primarily dolomite (over 80%), while calcite and clay minerals together make up less than 10%. Structurally, micrite comprises between 85% and 95%, with bioclastic fragments and argillaceous material accounting for less than 10% and 5%, respectively. Textures, like laminated micritic dolostone with preserved fabrics, generally consist of crystals less than 20 μm in size.

Granular dolostone (Figure 2(B1,B2)): Appearing in shades of light gray to gray, this rock type is characterized by a particle content exceeding 50%. These grains may include bioclasts, intraclasts, ooids, peloids, or oncoids, either singularly or in combination. The interstitial material between these grains is typically a blend of micritic matrix and sparry cement. Textures, such as very fine to fine crystalline nonplanar-a to planar-s dolomite, typically have crystal sizes ranging from 20 to 60 μm .

Crystalline dolostone (Figure 2(C1,C2),D): This type of dolomite rock predominantly exhibits euhedral to subhedral grain shapes, characterized by well-formed crystal morphologies. SEM reveals that the crystals display point or line contacts, suggesting interlocking crystal arrangements. This rock shows unfilled fractures, intergranular pores, and dissolution pores, characterized by significant development of intergranular microfractures. This texture is characterized by well-formed crystals of dolomite. Crystalline dolomite can vary widely, with fine-grained types having crystal sizes around 18 to 22 μm and medium to coarse-grained types exceeding 250 μm .

Saddle dolomite (Figure 2D,E): These are defined by their irregular shapes, exhibiting step-like or saddle-shaped crystal faces. The saddle crystalline does not occur as a layered rock, but just as the fillings in dissolution pores or caves of matrix dolostone. Saddle dolomite, characterized by non-planar anhedral to subhedral crystals with undulate extinction, often has crystal sizes ranging from 1000 to 5000 μm .

4.2. Geochemical Characteristics

The diagenetic fluids undergoing the replacement processes in dolostones frequently would leave behind geochemical signatures indicative of their origin. These signatures are evident in stable isotopes, trace elements, and rare earth elements. Consequently, this research utilized geochemical analysis methods to ascertain the provenance of the diagenetic fluids in the dolostones from the study area. The analysis included the examination of carbon and oxygen isotopic composition, along with strontium isotopes and rare earth element assessment. To facilitate the investigation, the samples were classified into three distinct groups: micritic dolostone, fine-crystalline dolostone, and saddle dolomite (filling in pores or caves). The comprehensive results obtained from these rigorous experiments are systematically organized in a table (Table 1).

Table 1. Isotope testing results for carbon, oxygen, and strontium of the Lower Paleozoic dolostones.

Sample Number	Lithology	$\delta^{13}\text{C}$ (‰PDB)	$\delta^{18}\text{O}$ (‰PDB)	$\text{Sr}^{87}/\text{Sr}^{86}$
SXB051-1	Micritic dolostone (D1)	1.259	−9.512	0.709190
SXB051-8	Micritic dolostone (D1)	−0.245	−7.731	0.709411
SXB052-2	Micritic dolostone (D1)	−0.950	−7.522	0.710628
SXB052-7	Micritic dolostone (D1)	1.159	−8.847	0.709706
SXB142-4	Micritic dolostone (D1)	0.652	−7.325	0.710416
SXB143-5	Fine-crystalline dolostone (D2)	0.811	−12.812	0.712473

Table 1. Cont.

Sample Number	Lithology	$\delta^{13}\text{C}$ (‰PDB)	$\delta^{18}\text{O}$ (‰PDB)	$\text{Sr}^{87}/\text{Sr}^{86}$
SXB143-5	Fine-crystalline dolostone (D2)	0.407	−11.903	0.712576
SXB143-5	Fine-crystalline dolostone (D2)	−1.113	−13.401	0.713093
SXB143-5	Fine-crystalline dolostone (D2)	0.618	−12.006	0.713403
SXB143-6	Fine-crystalline dolostone (D2)	−0.812	−12.623	0.712780
SXB051-7	Fine-crystalline dolostone (D2)	0.114	−11.312	0.713219
SXB441-5	Saddle dolomite (SD)	2.657	−15.126	0.714882
SXB421-7	Saddle dolomite (SD)	3.012	−14.726	0.715341

4.2.1. Carbon and Oxygen Isotope Characteristics

Carbon and oxygen isotope analysis is widely recognized as a crucial method for studying carbonate rocks, primarily due to its established reliability, feasibility, and global comparability of results. Notably, isotope values can provide insights into the nature of the fluids of dolomitization. Early diagenetic dolostone is typically characterized by higher carbon and oxygen isotope values than those of seawater, with values exceeding -8‰ . In contrast, the oxygen isotope values for burial dolostone are more negative compared to seawater and decrease with increasing depth and temperature.

According to the experimental results (Table 1, Figure 3), the $\delta^{13}\text{C}$ values of samples from the micritic dolostone in the study region range from -0.950‰ to 1.259‰ , with an average of 0.375‰ . Meanwhile, the $\delta^{18}\text{O}$ values range from -7.325‰ to -9.512‰ , averaging -7.187‰ . For fine-crystalline dolostone, the $\delta^{13}\text{C}$ values range from -1.113‰ to 0.811‰ , averaging -0.631‰ , while the $\delta^{18}\text{O}$ values are between -11.312‰ and -13.401‰ , averaging -12.342‰ . There are only two samples of saddle dolomite, with $\delta^{13}\text{C}$ values ranging from 2.657‰ to 3.012‰ and $\delta^{18}\text{O}$ values are between -14.726‰ and -15.126‰ . It is important to note that the $\delta^{18}\text{O}$ values from the fine-crystalline dolostone are significantly lower than those from the micritic dolostone and are considerably lower than the current oceanic value of -7‰ . Figure 3 provides a graphical representation of the carbon and oxygen isotope data for the dolostones based on these experimental findings.

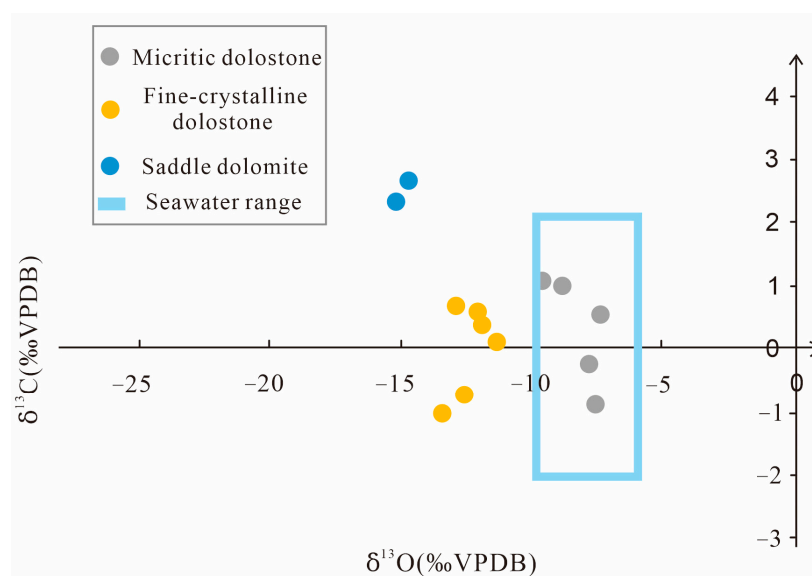


Figure 3. The cross plot of carbon and oxygen isotope composition of the Lower Paleozoic dolostones.

4.2.2. Strontium Isotope Characteristics

The isotopic composition of strontium of seawater recording in carbonates, influenced predominantly by crustal and mantle-derived sources, displays a stable $\text{Sr}^{87}/\text{Sr}^{86}$ ratio

within the same geological period. This characteristic makes strontium isotopes a reliable tracer for the origins of dolomitizing fluids. Lake waters typically show $\text{Sr}^{87}/\text{Sr}^{86}$ ratios that are similar to the average freshwater ratio of 0.7119. However, coastal and fault-bounded lake basins may show lower $\text{Sr}^{87}/\text{Sr}^{86}$ ratios due to seawater and hydrothermal impacts. Historical evidence suggests that the $\text{Sr}^{87}/\text{Sr}^{86}$ ratio in seawater has remained relatively stable, with modern seawater carrying an average value of 0.70907327 [29]. In the Early Paleozoic era, seawater exhibited $\text{Sr}^{87}/\text{Sr}^{86}$ ratios ranging from 0.7073 to 0.7120. In the study area, micritic dolostone presents $\text{Sr}^{87}/\text{Sr}^{86}$ ratios between 0.7090 and 0.7120, fine-crystalline dolostone falls within a range of 0.7120 to 0.7140, while saddle dolomite reveals $\text{Sr}^{87}/\text{Sr}^{86}$ ratios higher than 0.7140 (Figure 4). These results indicate that most analyzed samples within the study area, except for micritic dolostone, exhibit higher $\text{Sr}^{87}/\text{Sr}^{86}$ ratios than those found in modern seawater.

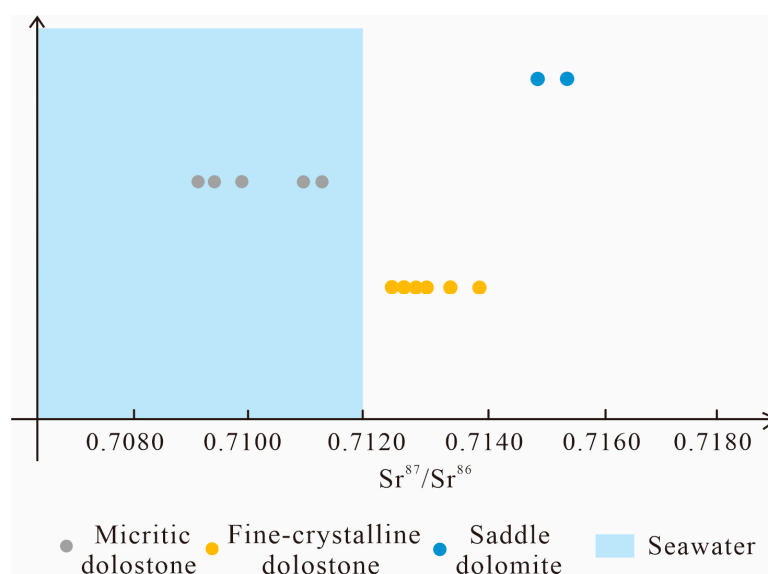


Figure 4. Distribution characteristics of strontium isotope values in Lower Paleozoic dolostones.

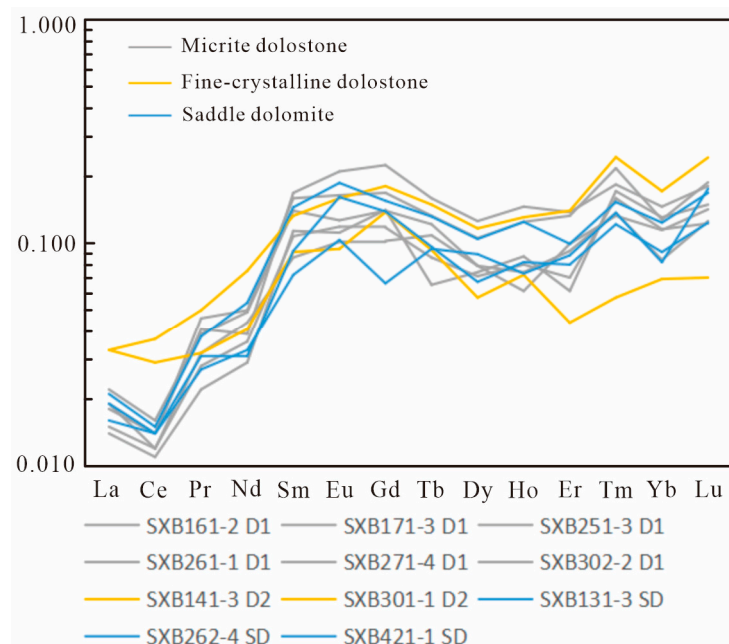
4.2.3. Rare Earth Element Characteristics

The role of trace elements, particularly rare earth elements (REEs), in understanding the origin of dolostones is crucial. REEs, due to their chemical stability, preserve their original fluid characteristics during burial, making them less susceptible to the effects of temperature and pressure. These rendered REEs are an essential tool in deciphering the characteristics of dolomitizing fluids and the diagenetic environments they were exposed to.

In the present study, a range of carbonate rock samples was analyzed, revealing that REE concentrations varied (Table 2). The ΣREE values ranged from 5.064 to 10.758, with a mean of 6.722; the light REE (LREE) values fell between 2.345 and 5.538, with an average of 3.267; and the heavy REE (HREE) values ranged from 1.420 to 2.837, with an average value of 1.973. These findings point to a relative enrichment of heavy REEs, indicating a distinct geochemical profile. Further analysis of the seabed samples using a PAAS (Post-Archean Australian Shale) normalized plot provides additional insights. A left-skewed distribution pattern was observed, suggesting reduced LREE content and relative enrichment of HREEs. The average LREE/HREE ratio indicates the fractionated behavior of REEs in the carbonate environment. The $(\text{Ce}/\text{Ce}^*)_{\text{N}}$ values typically show a negative anomaly consistent with the characteristics of seawater (Figure 5), suggesting that the REE distribution was primarily influenced by the seawater source, indicating that dolomitization occurred under seawater influence.

Table 2. Isotope testing results for rare earth elements in Lower Paleozoic dolostones.

Sample Number	Lithology	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	ΣREE
SXB051-1	Micritic dolostone (D1)	0.715	1.149	0.365	1.331	0.926	0.225	1.034	0.123	0.585	0.144	0.392	0.074	0.410	0.077	7.548
SXB051-8	Micritic dolostone (D1)	0.555	0.928	0.245	1.215	0.595	0.128	0.546	0.067	0.332	0.079	0.199	0.064	0.325	0.053	5.331
SXB052-2	Micritic dolostone (D1)	0.732	0.945	0.280	1.477	0.479	0.109	0.475	0.084	0.371	0.060	0.282	0.054	0.322	0.061	5.731
SXB052-7	Micritic dolostone (D1)	0.852	1.288	0.347	1.678	0.771	0.136	0.649	0.093	0.368	0.073	0.262	0.055	0.237	0.054	6.863
SXB142-4	Micritic dolostone (D1)	0.546	0.858	0.196	0.985	0.625	0.120	0.654	0.051	0.348	0.087	0.175	0.069	0.367	0.064	5.144
SXB302-2	Micritic dolostone (D1)	0.671	1.096	0.409	1.706	0.878	0.176	0.779	0.102	0.491	0.123	0.378	0.087	0.358	0.080	7.334
SXB143-5	Fine-crystalline dolostone (D2)	1.275	2.280	0.281	1.377	0.504	0.101	0.637	0.072	0.266	0.071	0.125	0.023	0.195	0.030	7.238
SXB143-6	Fine-crystalline dolostone (D2)	1.260	2.937	0.438	2.528	0.732	0.170	0.832	0.115	0.541	0.129	0.396	0.098	0.479	0.104	10.758
SXB421-7	Saddle dolomite (SD)	0.607	1.104	0.239	1.122	0.400	0.112	0.307	0.073	0.416	0.073	0.249	0.055	0.231	0.076	5.064
SXB421-1	Saddle dolomite (SD)	0.821	1.179	0.335	1.832	0.797	0.200	0.716	0.101	0.485	0.123	0.281	0.062	0.348	0.072	7.352
SXB441-5	Saddle dolomite (SD)	0.719	1.150	0.270	1.051	0.511	0.173	0.644	0.075	0.316	0.082	0.228	0.049	0.256	0.053	5.577
	PASS normalized values	38.20	79.60	8.83	33.90	5.55	1.08	4.66	0.77	4.68	0.99	2.85	0.41	2.82	0.43	

**Figure 5.** PAAS-normalized REE distribution patterns for Lower Paleozoic dolostones.

5. Discussion

5.1. Properties of Dolomitizing Fluids

The geochemical characteristics of marine carbonates, including their stable isotopes and trace elements, are significantly influenced by diagenetic fluids [30]. This renders dolostones a valuable tool for understanding the mechanisms of formation across different environments. The fractionation of $\delta^{13}\text{C}$ during diagenesis is typically minimal, often attributed to organic carbon oxidation and the burial of organic carbon. When the burial rate of organic carbon surpasses its oxidation rate, more ^{12}C enters the organic carbon pool, leading to more negative $\delta^{13}\text{C}$ values in contemporaneous marine carbonates [31]. However, oxygen isotopes are highly sensitive to both temperature and diagenetic alteration. Deviations in carbonate $\delta^{18}\text{O}$ values during diagenesis primarily result from changes in temperature and salinity of the diagenetic fluids. Processes such as recrystallization, mixing with continental freshwater, and increased temperatures in the burial environment may result in a negative shift in $\delta^{18}\text{O}$ values [32]. Conversely, an increase in the depositional environment's water salinity and intensified evaporation will cause a positive shift in $\delta^{18}\text{O}$ values [33]. Strontium isotopes in carbonates are capable of retaining original seawater signatures. Throughout each geological period, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in seawater has typically remained within a stable range [34]. However, during diagenesis and burial, diagenetic

fluids originating from adjacent rock layers or deep-seated magmatic sources may alter the original seawater signatures recorded in carbonates. By analyzing the distribution range of $^{87}\text{Sr}/^{86}\text{Sr}$ values, the origins of these diagenetic fluids can be inferred [35–39].

The distribution of strontium isotope in mudstone-dolostone ranges from 0.709 to 0.712, with a mean of 0.710. This range matches the seawater values of the same period during the Ordovician era [40,41], suggesting that the diagenetic fluids for these dolostones were derived from contemporaneous seawater during the depositional phase. The carbon and oxygen isotope values of micritic dolostone also largely fall within the range of contemporaneous seawater [42,43]. The $\delta^{18}\text{O}$ values, which are highly sensitive to the influence of diagenetic fluids, exhibit minimal alteration. The REE (rare earth element) distribution patterns exhibit a left-skewed pattern, characterized by LREE (light rare earth elements) being depleted relative to HREE (heavy rare earth elements) $\text{LREE} < \text{HREE}$, along with negative Ce anomalies similar to those found in seawater. These characteristics collectively indicate that the dolomitizing fluids for micritic dolostone were contemporaneous seawater.

The carbon isotope composition of fine-crystalline dolostone closely resembles that of micritic dolostone. However, the $\delta^{18}\text{O}$ values are slightly more negative, likely due to oxygen isotope fractionation during burial diagenesis as temperatures increased. The REE distribution patterns in fine-crystalline dolostone also exhibit a left-skewed pattern, characterized by LREE being depleted relative to HREE, along with negative Ce anomalies similar to those found in seawater. This suggests that fine-crystalline dolostone retains much of its original seawater information [44]. Notably, the $^{87}\text{Sr}/^{86}\text{Sr}$ values in fine-crystalline dolostone are slightly higher than those of contemporaneous seawater, indicating that the dolomitizing fluids responsible for the precipitation of saddle dolomite underwent interaction with underlying basement rocks, thereby assimilating strontium from these substrates [45,46]. The hydrothermal fluid subsequently induced the recrystallization of micritic dolostone into a finely crystalline dolostone matrix, which resulted in the partial overprinting of both the original Sr isotopic compositions and stable isotopic ratios within the rock fabric [47,48].

The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of saddle dolomite, which fills the pores within the samples, are outside the range typically associated with contemporaneous seawater [49]. This suggests that the dolomitizing fluids were derived from external sources rather than syndepositional seawater. The strontium isotope values of this saddle dolomite are notably higher than those expected for contemporaneous seawater, possibly linked to hydrothermal fluids associated with crustal magmatism [50]. Furthermore, the presence of a positive Eu anomaly in the REE (rare earth element) distribution supports the hypothesis that the dolomitizing fluids were influenced by hydrothermal fluids associated with magmatic activity [51,52]. These findings collectively shed light on the complex interplay between geological processes and fluid compositions during the formation of saddle dolomite.

5.2. Origin of the Dolomites

The previous analysis of fluid properties shows that distinct fluid characteristics are associated with different types of dolomite, suggesting varied dolomitization mechanisms. Micritic dolostone retains early diagenetic seawater signatures of strontium and carbon-oxygen isotopes, exhibiting a rare earth element distribution pattern similar to seawater which is characterized by a left-skewed type. Obviously, its fluid properties are consistent with Lower Paleozoic contemporaneous seawater, indicating it belongs to an early diagenetic dolostone formation, and it could be explained by the evaporative pump and reflux dolomitization model [53]. Some collected samples display needle-like gypsum formed through evaporation, in association with micritic dolostone. This also suggests that micritic dolostone is likely formed via evaporative pump dolomitization within the

supratidal zone [54]. This type of dolostone typically occurs in thin layers, which is consistent with typical supratidal sedimentary characteristics. Sedimentological studies have demonstrated that during the Cambrian–Ordovician period, the North China craton was of a widespread shallow marine environment [55]. In this context, strong evaporation within a carbonate platform environment may have increased the Mg/Ca ratio of seawater. This allowed magnesium ions to replace some of the calcium ions in aragonite, resulting in broad dolomitization [56,57]. This finding provides valuable insights into the geological processes and environmental conditions for the formation of various types of dolostones within the study area.

Fine-crystalline dolostone occurs in thick layers in the study area. Its dolomitizing fluids were closely related to the original seawater. This suggests that fine-crystalline dolostone likely formed via reflux dolomitization during the syndepositional period [58]. Due to the Lower Paleozoic Basin of North China craton being featured as a semi-closed lagoon under strong evaporation [59], the Mg/Ca ratio of seawater could have been significantly elevated against the surrounding marine area. Driven by concentration and density gradients, high-magnesium fluids within these lagoons might have infiltrated shoreward aragonite sediments located in the intertidal zone, resulting in dolomitization via calcium ion replacement by magnesium [60]. The fine-crystalline dolostone displays clear negative Ce anomalies, and the crystals within this type of dolostone are relatively regular, suggesting they experienced a following slow-dolomitizing environment [61]. Thus, we argue that the fine-crystalline dolostone further underwent a superimposed dolomitization in a buried reductive environment after the initial reflux dolomitization.

Saddle dolomite observed within large pores or caves as pore-fillings material are proximal to fault zones, which indicates their formation is related to fractures and faults. Analysis of the fluid characteristics also show that the formation of saddle dolomite could be attributable to an external hydrothermal fluid [62–64]. The low degree of crystallinity of saddle dolomite suggests a rapid dolomitization process [65]. Consequently, the origin of this type is consistent with the hydrothermal dolomitization model [66]. The Bohai Bay Basin has been subject to multiple magmatic events since the Mesozoic era, with evidence of Cenozoic magmatic intrusions within sedimentary strata [67,68]. This indicates a geological background conducive to the presence of hydrothermal fluid sources within the study area, revealing the potential mechanisms driving the formation of saddle dolomite and its incidental role in the overall reservoir formation.

5.3. Development Model of the Dolostone Reservoirs

Globally, approximately half of all oil and gas reservoirs are hosted in carbonate rocks, with a significant proportion involving dolomitization processes. Understanding the dolomitization model is thus essential for predicting potential reservoirs related to dolostone formations. Current exploration efforts indicate a strong correlation between Paleozoic reservoirs and dolostones in the offshore Bohai Bay Basin [69,70]. The physical properties of dolostone reservoirs are superior to those of limestone, making them the primary targets in exploration activities. Most of the high-quality reservoirs occur in layered areas and predominantly develop in crystalline dolostone strata. In the Lower Paleozoic marine of the North China craton, early diagenetic evaporative pumping and reflux dolomitization were prevalent phenomena, which is the fundamental setting for the development of dolostone reservoirs (Figure 6a). Burial dolomitization is responsible for forming more regular dolomitic crystalline, which reduces occupied volume spatially, thereby increasing rock porosity (Figure 6b). It occurs on top of early-stage dolostones, converting pre-existing layered dolostone into a porous medium with enhanced permeability. By contrast, dolomite rocks are also an important carbonate reservoir in Ordos Basin, the west part of North

China Block, similar to those observed in the offshore Bohai Bay Basin. Their primary mechanisms of dolomitization all involve seepage reflux dolomitization and later burial dolomitization [71–73]. As the burial temperature and pressure increase, early diagenetic micritic dolostones can undergo recrystallization to form fine-crystalline dolostones [74–76]. This recrystallization process exhibits a volume reduction effect, which can give rise to an increase in intercrystalline porosity [75]. The transformation from micritic to fine-crystalline dolomite is driven by thermodynamic and kinetic factors that favor the growth of larger, more stable crystals at the expense of smaller, less stable ones [75,76]. This process alters the mineralogical and textural characteristics of the rock and significantly impacts its porosity and permeability, which are crucial for hydrocarbon reservoir quality [76]. The development of intercrystalline pores during recrystallization can enhance the storage capacity and fluid flow properties of the dolostone, making it an important consideration in subsurface geology and resource exploration [77].

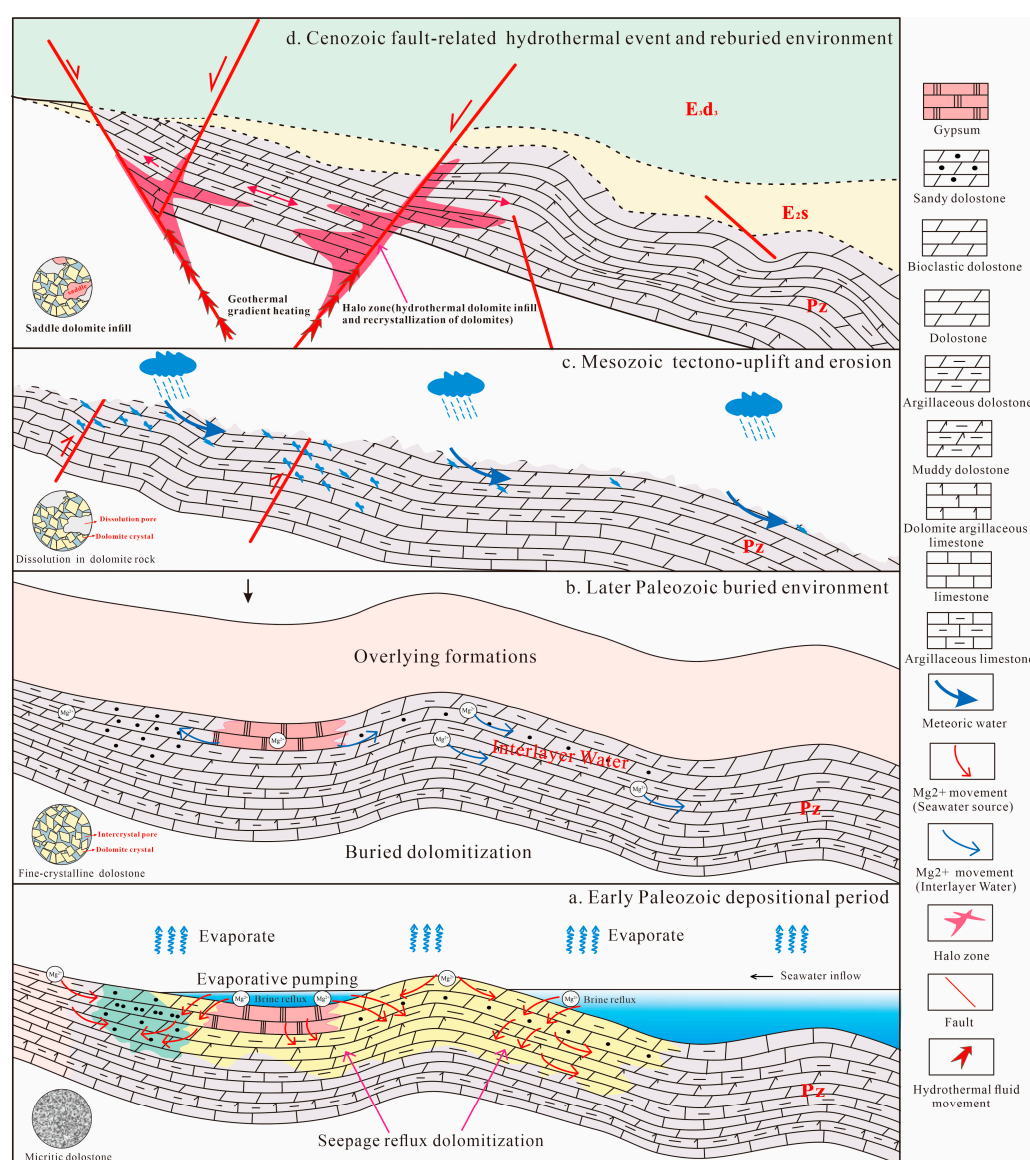


Figure 6. Multiple dolomitization process for Lower Paleozoic dolostones. (a) The evaporative pumping dolomitization and seepage reflux dolomitization model; (b) the buried dolomitization model; (c) mesozoic tectono-uplift and erosion; (d) the hydrothermal dolomitization model.

Furthermore, research has demonstrated that fractures are more prevalent in fine-crystalline dolostone than in micritic dolostone [78]. Our core observation results corroborate this finding, indicating greater fracture development in the layers of fine-crystalline dolostone. These fractures initially acted as conduits for diagenetic fluids. During the Mesozoic exposed interval, the interaction between meteoric and subsurface diagenetic solutions with carbonate rocks along fractures resulted in the development of extensive fracture-related dissolution pores and caves, significantly modifying the geological characteristics of the region (Figure 6c). Meanwhile, these second pores or caves would capture some meteoric fluid that partly influenced the recrystallization of dolomites during the later Cenozoic burial process, which could explain why they partly had meteoric fluid characteristics. Additionally, the Cenozoic hydrothermal event might destroy the prior reservoir as a “halo zone” surrounding these fault-related saddle dolomites where the host rock underwent recrystallization (Figure 6d) [79]. But such occurrences of saddle dolomite, filling these pores or caves, are rare and do not significantly impact the overall reservoir quality, due to the volume of saddle dolomite being minimal [80]. Thus, some of the faults and fractures remain open. In addition, the recrystallization can enhance the reservoir properties of the matrix [81]. So the fractures and related pores or caves yet serve as critical pathways and storage sites for subsequent oil and gas injections, contributing to the potential productivity of the reservoirs within these formations.

6. Conclusions

- (1) The dolomitizing fluids of the Lower Paleozoic dolostone reservoirs include syndepositional to early post-depositional seawater and hydrothermal fluids. The diagenetic fluids responsible for the formation of micritic dolostone originated primarily from contemporaneous seawater. Fine-crystalline dolostone also formed from penecontemporaneous dolostone but is influenced by later burial processes. The geochemical characteristics of saddle dolomite indicate it originated from hydrothermal fluids linked to magmatic activity and occurs sporadically within pre-existing dissolution pores or caves.
- (2) The carbonate reservoirs in the west of offshore Bohai Bay Basin primarily underwent syndepositional to early post-depositional evaporative pumping, reflux dolomitization, burial dolomitization, and hydrothermal dolomitization. The syndepositional reflux dolomitization is the initial mechanism for the formation of extensive layered dolostones. The superimposed effects of later burial dolomitization led to the development of fine-crystalline dolostone which represents the predominant reservoir type in the carbonate basement of offshore Bohai Bay Basin.

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