

Article

Formation Mechanism and Petroleum Geological Significance of (Ferro) Dolomite Veins from Fractured Reservoirs in Granite Buried Hills: Insights from Qiongdongnan Basin, South China Sea

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Abstract: This study employs logging, petrology, and geochemistry to investigate the characteristics, origin, and hydrocarbon significance of fractures and (ferro) dolomite veins in a buried hill in the Qiongdongnan (QDN) Basin, South China Sea. We show that the study area is mainly characterized by three stages of fracturing with medium-high dipping angles. The orientation of the fractures is mainly NNW–SSE, consistent with the fault system strike formed by the Mesozoic–Cenozoic tectonic activity in the basin. (Ferro) dolomite veins in the fractures can be classified into three stages, all of which can be even observed in individual fractures. The first stage is the powdery crystal dolomite veins grown mainly on the fracture surface, which have the highest strontium isotope values, as well as high contents of the Mg element and extremely low contents of the Fe and Mn elements. The first-stage veins were formed in a relatively open oxidized environment, and the vein-forming fluids exhibit characteristics of mixing formation water and atmospheric freshwater within the fractures. The second stage, involving fine-crystal dolomite veins, was formed in a buried diagenetic environment where groundwater mixed with deep hydrothermal fluids, and contained the highest carbon isotope values, more Fe and Mn elements, and less Mg element than the first stage. The third stage of medium-crystal ankerite veins was formed in the latest stage, with the lowest strontium and oxygen isotope values. This was mainly a result of deep hydrothermal formation in which the rock-forming material formed from the interaction between the hydrothermal fluid and the iron-rich and aluminosilicate minerals in the surrounding granite of the fractures. We conclude that the multi-phase tectonic movements form a massive scale reticulated fracture inside the granite buried hill, which effectively improves the physical condition of the gas reservoirs. The gas reservoirs remain of high quality, despite the filling of the three stages of (ferro) dolomite veins.

Keywords: Qiongdongnan Basin; south China sea; granite buried hills; fractured gas reservoirs; dolomite veins; reservoir quality



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1. Introduction

The global consumption of oil and gas has increased steadily in the past decades, and unconventional petroleum resources are an important exploration alternative for conventional oil and gas [1,2]. Among them, buried-hill fractured oil and gas reservoirs are an unconventional petroleum resource that have received global interest [3,4].

Buried hills are a kind of ancient geomorphology, where oil and gas can accumulate, that belongs to the basement reservoir and combined trap [3,4]. About 40 percent of the buried-hill reservoir lithology is granite [3–8]. Offshore oil and gas discoveries show a very good exploration prospect in the buried-hill area [9]. Granite buried-hill reservoirs are more heterogeneous than conventional clastic rock reservoirs [10]. In 2018, a reservoir with a thickness of over 100 m was drilled in a Mesozoic-era granite buried hill in the Qiongdongnan (QDN) Basin, South China Sea (SCS), and a high-yield and high-quality natural gas flow was obtained from the test [11–13].

The QDN Basin shows mainly two types of buried-hill reservoirs, i.e., weathering shells and granite fracture zones. The weathered-shell reservoir accounts for 11.3% and the fracture-zone reservoir accounts for 88.7% of the total thickness [14]. Fractures are essential for the generation of highly productive gas reservoirs [15]. Early generated fractures can be used as important channels for geological fluid transport, and geological fluids usually have complex interactions with surrounding rocks during transport along the fractures, forming some fillings (vein bodies) [16,17]. Among them, dolomite is one of the most common vein minerals in carbonate diagenesis and is sensitive to the medium environment [18]. Vein structure and geochemical properties can reflect the nature of rock-forming fluids, the closure and opening of the depositional environment, etc. [19]. The process of multi-phase rock-forming fluid action can be also recorded [20–22].

(Ferro) dolomite veins are widely developed in the studied gas-bearing buried-hill reservoir [23]. Previous studies of granite buried hills in the area were mostly focused on rock types and geochemical characteristics, and few studies have focused on the origin and characteristics of different stages of dolomite fillings of fractures and their influence on the quality of the reservoir [24,25].

In this paper, we examine the petrology, X-Ray fluorescence (XRF), fluid inclusions, and micro-zone in situ elements, as well as the C, O, and Sr isotopic compositions of the (ferro) dolomite veins in the granite buried-hill reservoir in the XL area of the QDN Basin. We unravel the formation process and evolutionary characteristics of these fractures and (ferro) dolomite veins and explore the influence of (ferro) dolomite vein fillings on the quality of the fractured reservoir.

2. Geological Setting

The Cenozoic QDN Basin is located in the northwest margin of SCS (Figure 1) [26–30]. Located in the north is the Hainan Uplift, in the south is the Xisha Uplift, and in the east is the Pearl River Mouth Basin. The western part of the QDN basin is bounded by faults with the Yinggehai basin, with an area of about 80,000 km² [27,28]. The QDN Basin is generally distributed in NE–SW direction and has the structural characteristics of north–south zoning in the plane. From north to south, it can be divided into five first-level structural units, namely, the northern depression area, central uplift area, central depression area, southern uplift area, and southern depression area (Figure 1) [29,30]. The Eocene to Quaternary strata are developed in the QDN Basin, which are the Lingtou Formation, Yacheng Formation, Lingshui Formation, Sanya Formation, Meishan Formation, Huangliu Formation, Yinggehai Formation and Ledong Formation, listed from old to new (Figure 1) [26,29].

The area has undergone a complex Mesozoic-era tectonic evolutionary history with intense magmatic activity. Due to tectonic uplift, granite was exposed on a large area and subjected to weathering and denudation [24,25]. Since the beginning of the Cenozoic era, extension, faulting, and deposition have formed a faulted basin with granite as its base [4,25]. The research area has full 3D seismic coverage and is in the central depressional area of the QDN Basin, with a water depth of over 1500 m [31]. The granite buried hills in the XL area have been drilled with seven wells. Granite in this area, which is mainly monzogranite and granodiorite, was formed in the Early Triassic period (240.1–252.1 Ma) [23,32].

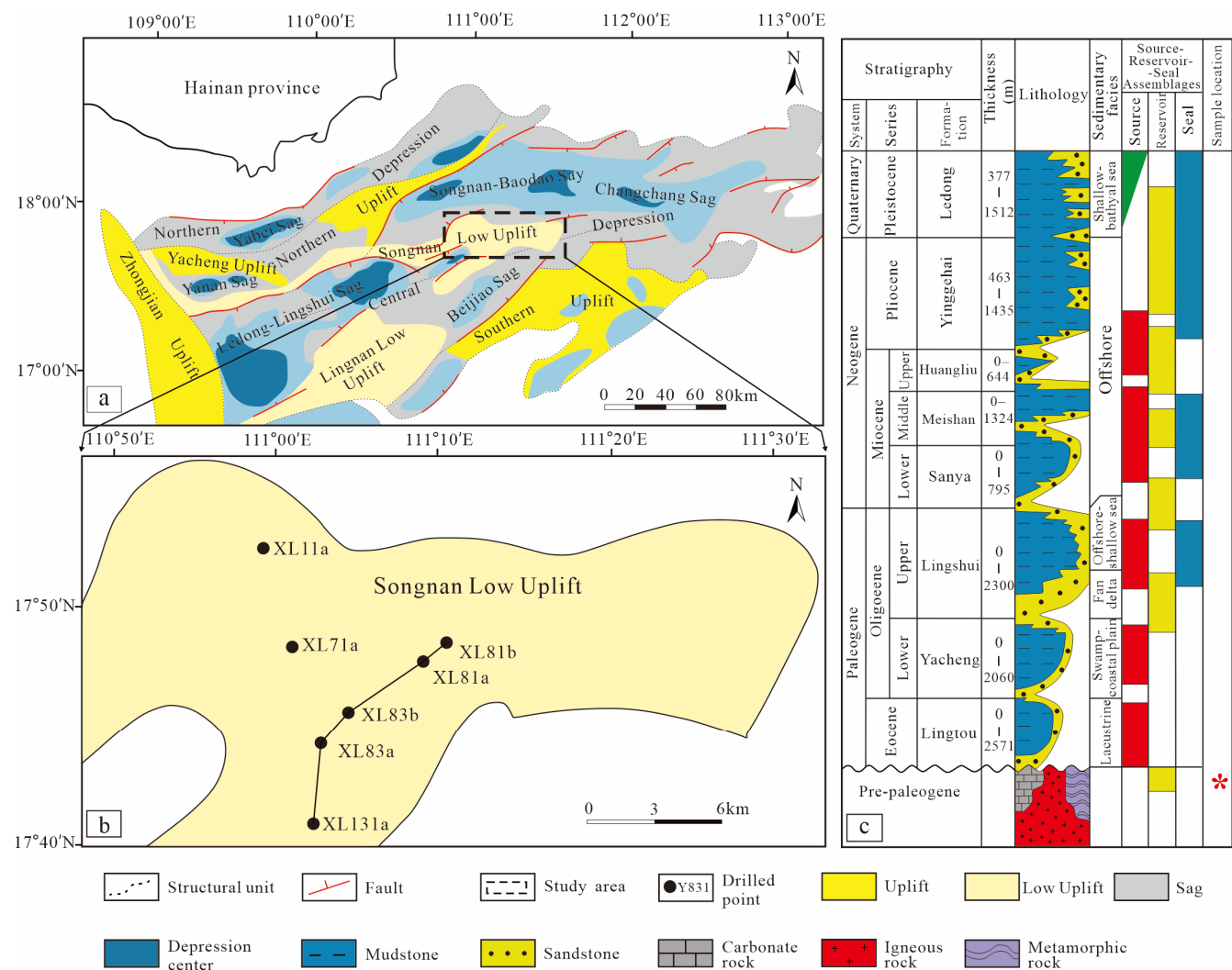


Figure 1. Tectonic framework and comprehensive stratigraphic column of Qiongdongnan Basin (modified from [4]). (a) Tectonic map; (b) drill location of intrusive buried hills in XL area; and (c) comprehensive stratigraphic column.

3. Data and Methods

All core samples were obtained from the pre-Paleogene strata. Based on the core samples and the development of veins, this study focuses on 64 samples from 7 wells (XL11a, XL71a, XL81a, XL81b, XL83a, XL83b, and XL131a). Using image logging, sidewall coring, and thin sections, we analyzed the filling characteristics of veins in the Mesozoic-era buried hill (Table S1).

Using the Bruker M4 Tornado and the Advanced Mineral Identification and Characterization System (AMICS) (Berlin, Germany), elemental analysis of rock samples was conducted. This system then converted X-Ray fluorescence spectra to mineral identifications, producing detailed mineralogical maps. The purpose of these maps was to study the chemical compositions of (ferro) dolomite veins. Additionally, emphasis was placed on analyzing representative samples that contained thick veins in favorable locations.

Micro-XRF, thin-section, fluorescence, cathodoluminescence, and fluid inclusion thermometry were conducted at the CNOOC Zhanjiang Experimental Centre. Micro-XRF analysis of polished core samples was conducted for 8 h with a resolution of 40 μm. The voltage and current were 50 kV and 600 μA, respectively. Ca, Mg, Fe, Mn, Al, Si, Ba, Sr, Rb, Pb, Na, Th, and U were determined. Based on this, thin sections were prepared, and optical, fluorescence, and cathodoluminescence identifications were conducted using a

ZEISS Axio Imager A2 microscope (Germany) and a CL8200MK5 cathodoluminescence instrument (UK). Homogenization temperature testing was carried out on 456 fluid inclusions of typical samples of dolomite veins using a ZEISS microscope equipped with a LINKAM THMS600 heating and cooling stage, with a test accuracy of 0.1 °C. From a petrographic perspective, inclusions with obvious decrepitation, stretching, and leaking were excluded in the process of temperature measurement to eliminate the possibility of re-equilibration to the greatest extent.

In situ analysis of chemical compositions, as well as Sr isotopes, in thin sections was performed at the Wuhan SampleSolution Analytical Technology Laboratory. Chemical analysis includes a Geo Las Pro laser system and an Agilent 7700× inductively coupled plasma mass spectrometer (ICP-MS) (Germany) with a spot size diameter of 50 µm. Quantitative calculations were performed using the international standards NIST SRM610 and NIST SRM612, and the generated data were processed by the ICPMSData Cal software ([33]). Based on the analysis of major trace elements, locations with relatively high Sr content in the samples were selected for in situ measurement of strontium isotope ratios using the laser ablation system and Neptune Plus multi-collector ICP-MS (MC-ICP-MS) (Germany). Due to the low Sr content in the dolomite veins, large ablation spots with a diameter of 90 µm were selected for testing. The laboratory internal standard MAD was used as the test standard, and the average value of Sr isotope measurement for the standard sample was 0.711866 ± 0.00005 .

The stable C and O isotope compositions of (ferro) dolomite veins were analyzed at CNOOC Tianjin Experimental Center. The three types of (ferro) dolomite veins were obtained using a microscope equipped with a micro-drilling system (America) with a diameter of 100 µm. The powder samples were reacted with pure phosphoric acid under vacuum conditions. Chemical separation was carried out as per [34], with acid separation conducted at 25 °C for 4 h. The isotopic ratios of the generated CO₂ were analyzed using a MAT251EM gas isotope mass spectrometer (Germany). The C and O isotope ratios were reported relative to Pee Dee Belemnite (PDB). The measurement accuracy was within $\delta^{13}\text{C}_{\text{PDB}} < 0.01\text{‰}$ and $\delta^{18}\text{O}_{\text{PDB}} < 0.02\text{‰}$. Oxygen isotopes were measured using two standards, PDB and SMOW (Standard Mean Ocean Water), with the conversion formula $\delta\text{O}_{\text{PDB}} = 0.97002\delta\text{O}_{\text{SMOW}} - 29.98$ [35]. At a hand-specimen scale, 27 samples of (ferro) dolomite were drilled from the granite fractures using a small drill bit and ground into powder. Carbon and oxygen isotope testing was conducted at the CNOOC Tianjin Experimental Center.

4. Results

4.1. Fracture Characteristics and Vein Composition

In the XL area of the QDN Basin, the gas reservoirs within the Mesozoic-era granite buried hills have well-developed fractures (Figure 2), which include structural fractures, dissolution fractures, and a minor number of diagenetic fractures. Based on the image logging from wells XL11a, XL81a, XL83a, XL83b, and XL131a, a total of 1092 fracture orientations and dip angles were statistically analyzed from top to bottom. The dominant fractures were high-angle structural fractures. While moderate-angle fractures mostly had an NNW–SSE orientation, high-angle fractures predominantly exhibited an NE–SW orientation (Figure 3). A total of 54% of the fractures are unfilled fractures and effective fractures, and they vertically form weathering-fracture zones and structural-fracture zones from top to bottom. The density of the fractures decreases with increasing burial depths. Dolomite and ankerite cements fill the fractures, forming (ferro) dolomite veins. A total number of 925 fully filled, 370 partially filled, and 709 unfilled fractures were observed on the granite core in the XL area, accounting for 46.16%, 18.46%, and 35.38% of the total count, respectively. The fracture widths range from 10 to 8000 µm, with an average of 560 µm (Figures 4 and 5). The filling materials consist of minor amounts of calcite and pyrite (Figures 5 and 6).

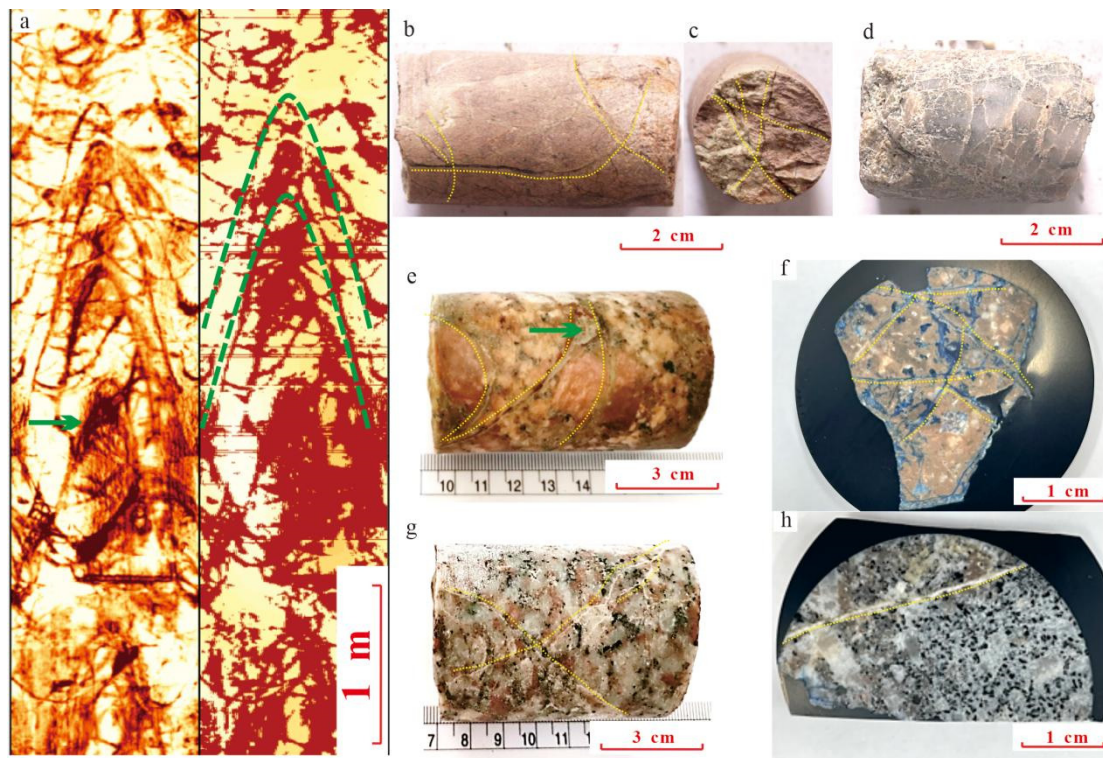


Figure 2. Fracture characteristics of buried-hill cores in XL area. (a) XL83a, 2840.0–2845.0 m, acoustic image logging, tectonic fractures (showing a complete sine or cosine shape) and dissolution fractures (the green arrow); (b) XL81a, 3006.0 m, fractures are in ‘X’ shape filled by dolomite; (c) cross-section of core cut in (b), with sample diameter of 3.8 cm; (d) XL81a, 3066.0 m, fractures filled with dolomite; (e) XL83a, 2908.8 m, fractures are mostly filled by dolomite, and partial dissolution widens the fracture surface (the green arrow); (f) cross-section of core cut in (e), with sample diameter of 3.8 cm; (g) XL11a, 3736.0 m, shear fracture is ‘X’ type and filled by ankerite; and (h) cross-section of core cut in (g), with sample diameter of 3.8 cm. The dotted lines in the figure represent fractures.

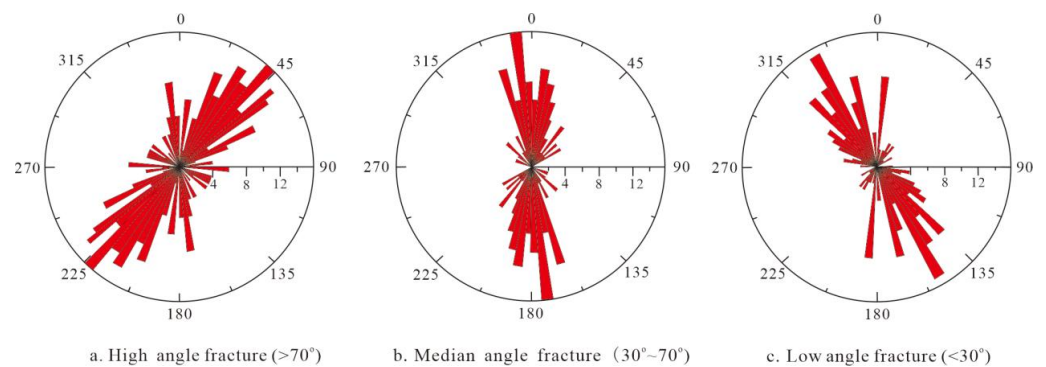


Figure 3. Rose diagrams of different occurrences of fractures from buried hills in XL area.

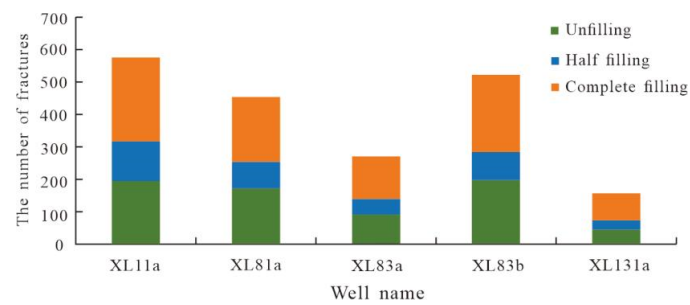


Figure 4. Histogram of core fracture filling degree from buried hills in XL area.

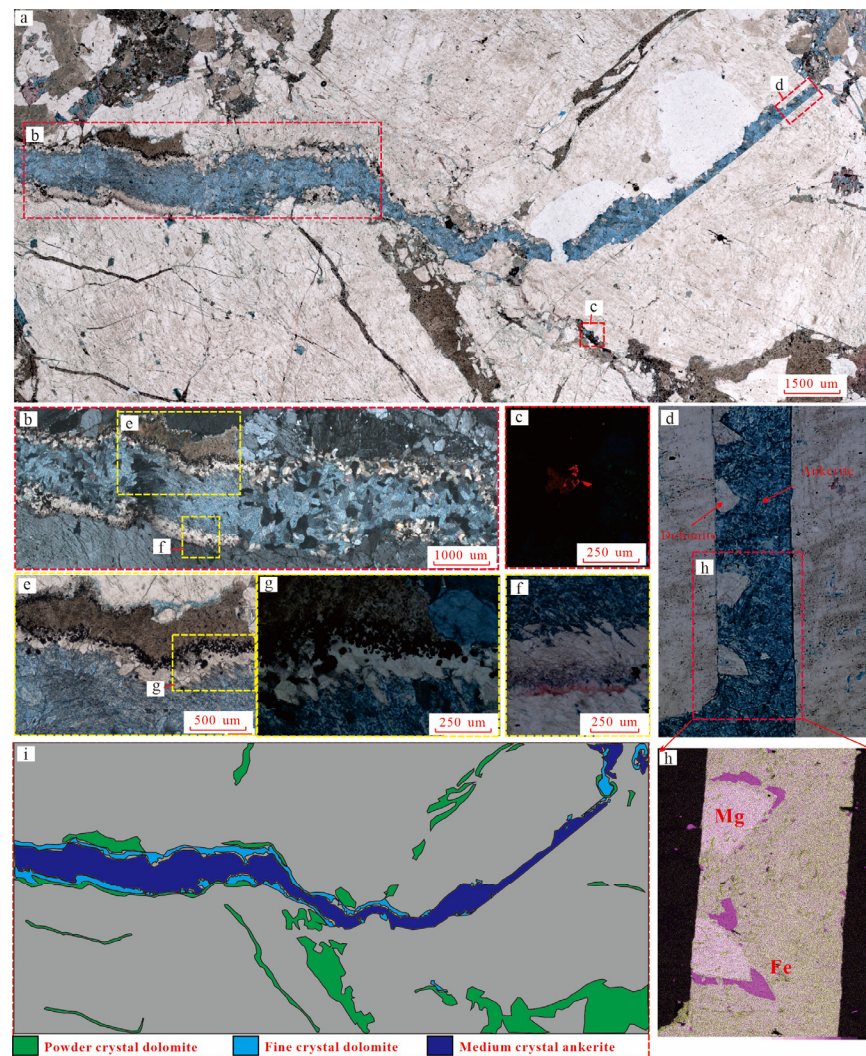


Figure 5. Microscopic characteristics and filling stage division of (ferro) dolomite veins in core samples from buried hills in XL area. (a) XL11a, 3897.0 m, dolomite and ankerite veins in granite, PLM; (b) vein has an undulose extinction, XPLM; (c) ankerite (bright red), dolomite (dark red), CL; (e–g) adhering to edge of fracture, powdery crystal dolomite grows at first stage; fine-crystal dolomite grows at second stage and crystals are highly euhedral and mostly diamond-shaped; a large number of medium-crystal ankerite grows at third stage; (d,h) adhering to edge of fracture, diamond dolomite occupies fracture space firstly, and ankerite fills remaining fracture space in later stages, indicating that iron contents in fluid increase in late diagenesis (brightness is proportional to content); and (i) three stages of (ferro) dolomite vein filling.

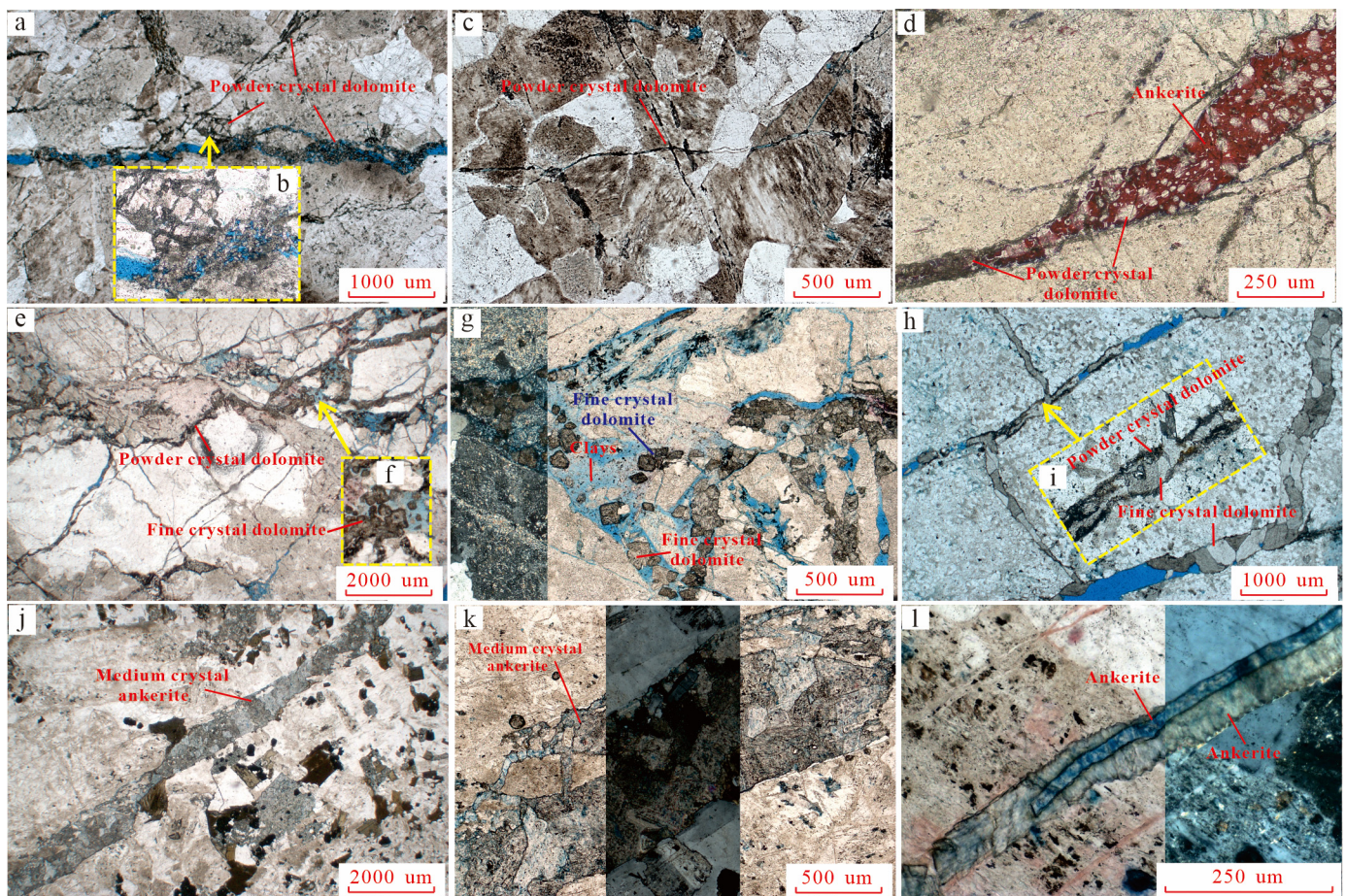


Figure 6. Microscopic characteristics of different stages of (ferro) dolomite veins of core samples from buried hills in XL area. (a) XL83a, 2830.0 m, powdery crystal dolomite fills fractures. Smaller opening fractures are almost completely filled, and larger opening fractures are mostly half-filled, with little remaining reservoir space; (b) partial enlargement of (a); (c) XL83a, 2902.0 m, powdery crystal dolomite filling ‘X’ type shear fracture (tectonic); (d) XL81a, 3012.0 m, powdery crystal dolomite preferentially fills fractures; (e) XL81a, 3033.0 m, powder-crystal and fine-crystal dolomite fills fractures. Among them, powdery crystal dolomite mainly fills small opening fractures, and fine-crystal dolomite fills large opening fractures (tectonic-dissolution fracture); (f) partial enlargement of (e); fine-crystal dolomite is diamond-shaped; (g) XL81a, 3002.0 m, fractures are filled with large number of clay minerals and fine-crystal dolomite. Most of the surface of fine-crystal dolomite has a (blue) coating composed of iron oxide and hydroxide; sericitization of feldspar components is common (left XPLM in diagram), and some of them are dissolved to form clay minerals, indicating that rock is affected by atmospheric water leaching; (h) XL83a, 2908.8 m, powdery crystal dolomite preferentially grows along fracture surface, and fine-crystal dolomite further fills remaining fracture space in later stage; (i) partial enlargement of (h); (j) XL11a, 3736.0 m, medium-crystal ankerite fills fractures; (k) locally enlarged image of (j), and medium-crystal ankerite has an undulose extinction (XPLM in the middle of the (k)); and (l) XL11a, 3744.0 m, fine-crystal dolomite preferentially grows along fracture surface, and medium-crystal ankerite further fills remaining fracture space in later stage.

4.2. Lithological Characteristics of Dolomite Veins and Filling Stages

The late-stage development of at least three stages of (ferro) dolomite veins was observed in the buried-hill fractures of the XL area (Figures 5 and 6). These authigenic mineral veins significantly reduce both porosity and permeability, as well as the reservoir’s homogeneity and connectivity (Figures 5 and 6). There are significant differences in the mineral lattice characteristics of (ferro) dolomite veins at different positions within the

fractures (Figures 5 and 6). In the first stage, dolomite directly contacting the surrounding rocks at the edge of the vein exhibits a rhombic shape. It primarily grows along the fracture surface, with predominantly muddy and powdery crystal forms. The surface appears yellowish and turbid (rich in impurities). In the 2nd stage, crystals in the vein center, slightly away from the fracture surface, generally increase in size, with a predominance of fine crystals. They often appear rhombic in shape with a high degree of idiomorphism. One to two sets of cleavage can be observed. The surfaces are relatively clean, and the crystals primarily grow vertically to the fracture surface (Figure 5b,d–g). Cathodoluminescence shows a weak red color (Figure 5c). This phase formed later than the powdery dolomite (continuing growth on the powdery dolomite as a substrate). In the third stage, medium-sized crystals of ankerite fill the remaining space within the fractures. They exhibit a deep blue-green color when stained with AFeS dye and show undulatory extinction (Figure 5b). The crystal size of ankerite is coarser than that of fine-crystal dolomite, and it is enriched in iron elements (Figure 5e–h). Cathodoluminescence shows a bright red color (Figure 5c). In the later stage, dissolution processes are generally absent. This phase formed later than the fine-grained dolomite (continuing growth on the fine-crystal dolomite as a substrate).

Under a microscope, both (ferro) dolomite veins and associated quartz fractures contain predominantly gas–liquid inclusions. These inclusions appear in ellipsoidal, triangular, or irregular shapes, ranging in size from 2.8 to 9.8 μm . They are commonly observed in a banded arrangement (Figure 7). The homogenization temperatures and salinity of the fluid inclusions within quartz fractures (Figure 8) indicate that the first-stage powdery crystal dolomite and second-stage fine crystalline dolomite are associated with saline fluid inclusions. The salinity ranges primarily from 1.1% to 6.5%, while the temperature ranges uniformly from 91.2 to 144.5 $^{\circ}\text{C}$. The third-stage medium-crystalline ankerite is associated with saline fluid inclusions. The salinity ranges primarily from 6.2% to 12.3%, while the temperature ranges uniformly from 173.6 to 191.8 $^{\circ}\text{C}$.

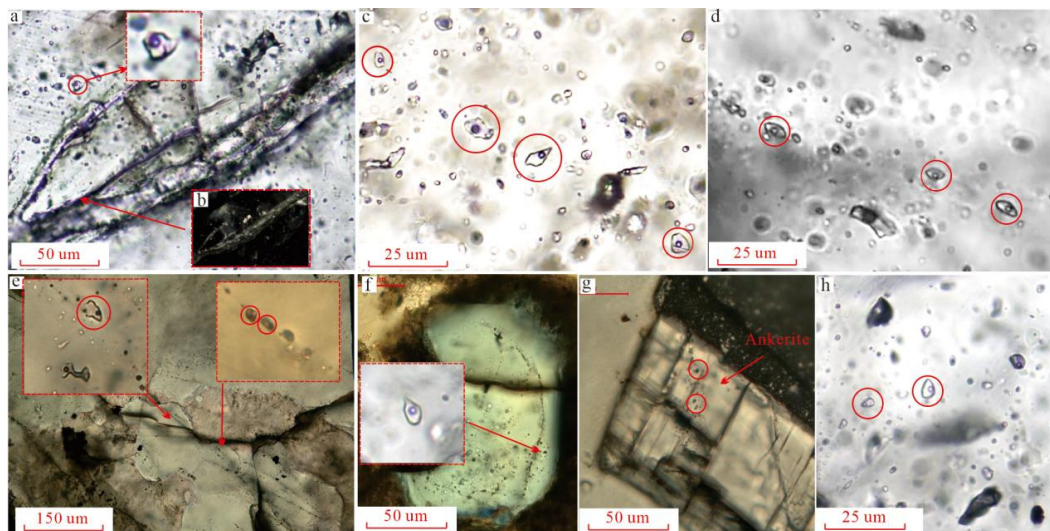


Figure 7. Microscopic characteristics of buried-hill (ferro) dolomite veins and associated quartz fissure brine inclusions in the XL area. (a,b) XL83a, 2860.0m, PPL, cataclastic granite. A large number of saline inclusions are developed in quartz fissures associated with dolomite veins, distributed in zones; (b) fluorescence (UV) photo; (c) XL71a, 3842m, PPL, A large number of saline inclusions are developed in quartz fissure; (d) XL71a, 3818 m, PPL, A large number of saline inclusions are developed in quartz fissures; (e) XL83b, 2890.0 m, PPL, a large number of saline inclusions are developed along microfissures of quartz particles; (f) XL131a, 2642 m, PPL, brine inclusions in enlarged edges of quartz; (g) XL83b, 2915.0 m, PPL, brine inclusions in microfissures of ankerite vein cements; and (h) XL71a, 3310 m, PPL, saline inclusions are developed along microfissures of quartz particles. The red circles indicate fluid inclusions.

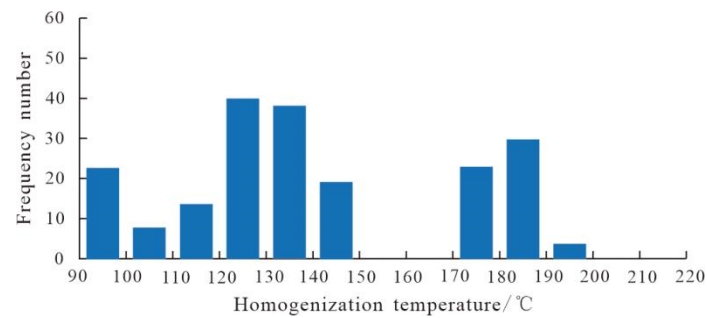


Figure 8. Histogram of homogeneous temperature distribution of brine inclusions in buried-hill (ferro) dolomite veins and associated quartz fissures in XL area.

4.3. Geochemical Characteristics of (Ferro) Dolomite Veins

In the XL area, the first-stage powdery crystal dolomite veins exhibit high Magnesium, low iron, and extremely low manganese concentrations (Figures 9 and 10). In the second stage, the fine-crystalline dolomite veins exhibit an increased Fe and Mn content, accompanied by a decrease in Mg content. In the third stage, the medium crystalline ankerite veins shows significantly higher content of Fe, Mn, Al, and Si compared to the previous stages, while the Mg content continues to decrease.

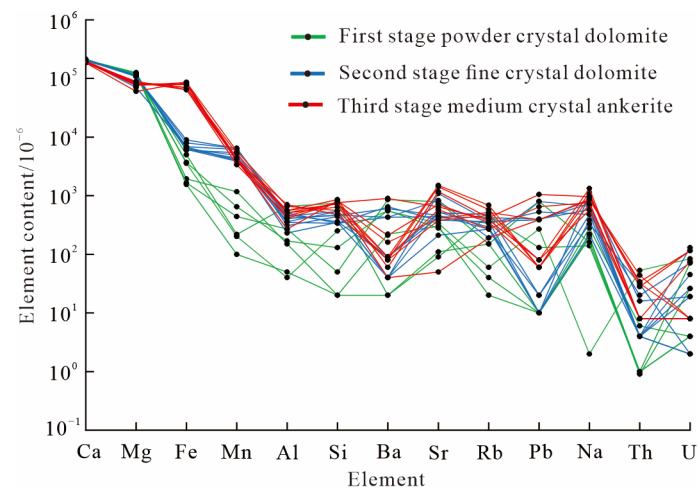


Figure 9. Distribution diagram of major and trace elements of different stages of (ferro) dolomite veins from buried hills in XL area.

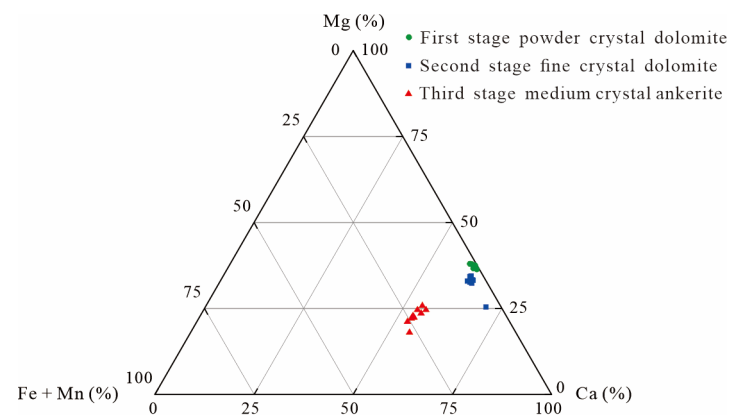


Figure 10. Triangle diagram of relative content of Fe + Mn, Mg, and Ca elements in different stages of (ferro) dolomite veins from buried hills in XL area.

Rare earth element (REE) distribution in (ferro) dolomite veins exhibit significant differences after normalization based on chondrite normalized REE values (Figure 11) [36]. The first-stage powdery crystal dolomite veins show no significant differentiation in the content of LREE and HREE, with a strong negative Ce anomaly and a weak negative Eu anomaly. The second-stage fine-crystalline dolomite veins exhibit a right-leaning distribution pattern of REE, characterized by significant enrichment of LREE, moderate positive Eu anomaly, and positive Ce anomaly. The third-stage medium-crystalline ankerite veins exhibit decreased REE content, accompanied by strong positive anomalies of Eu and Ce.

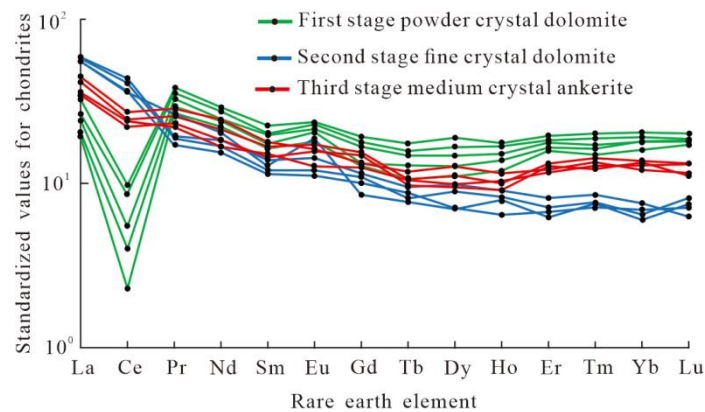


Figure 11. Standard curve of chondrite-normalized REE spectra of (ferro) dolomite veins from buried hills in XL area.

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the first-stage and second-stage powdery crystal dolomite veins ranged from 0.7088 to 0.71149 (average of 0.71033), and from 0.70769 to 0.70785 (average 0.70776), respectively (Figure 12). The third-stage medium-crystalline ankerite veins showed $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 0.70446–0.70526 (average of 0.70486).

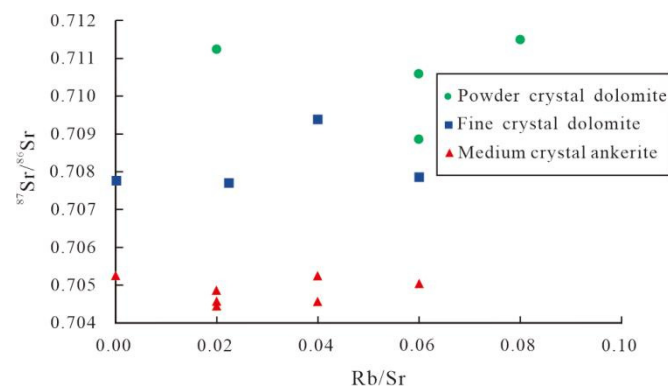


Figure 12. Strontium isotope ratios of (ferro) dolomite veins from buried hills in XL area.

The first-stage powdery crystal dolomite veins have $\delta^{13}\text{C}_{\text{PDB}}$ values ranging from -3.61‰ to -6.56‰ (average of -4.47‰) (Figure 13). The $\delta^{18}\text{O}_{\text{PDB}}$ values range from -7.26‰ to -24.43‰ , with an average value of -13.60‰ . The second-stage fine-crystalline dolomite veins have $\delta^{13}\text{C}_{\text{PDB}}$ values ranging from -0.94‰ to -7.53‰ (average of -2.72‰). The $\delta^{18}\text{O}_{\text{PDB}}$ values range from -7.212‰ to -15.84‰ , with an average value of -10.69‰ . The third-stage medium-crystalline ankerite veins have $\delta^{13}\text{C}_{\text{PDB}}$ values ranging from -2.42‰ to -8.34‰ (average of -5.26‰). The $\delta^{18}\text{O}_{\text{PDB}}$ values range from -20.05‰ to -25.82‰ (average of -22.75‰).

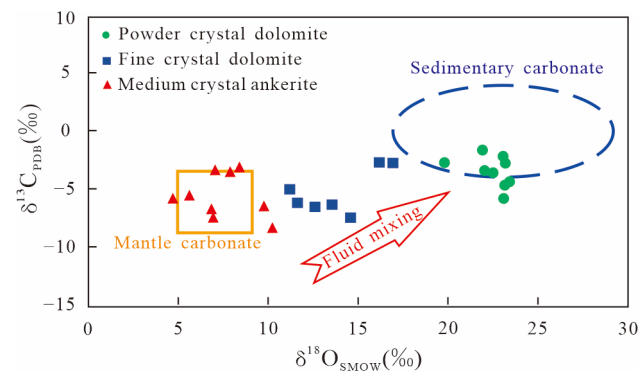


Figure 13. The $\delta^{13}\text{C}_{\text{PDB}}$ and $\delta^{18}\text{O}_{\text{SMOW}}$ isotopic characteristics of (ferro) dolomite veins from the buried hills in the XL area.

5. Discussions

5.1. Tectonic Activity and Fracture Formation

According to zircon $^{206}\text{Pb}/^{238}\text{U}$ dating, the granite intruded 240.1–252.1 Ma ago [23,32]. The buried hill with the low uplift has experienced several stages of tectonic events, such as orogeny, strike-slip movement, and extensional tilting, and three groups of faults formed a fracture network system [37].

The fault system with an NNW–SSE strike and a SWW dip had a strong impact on the fracture development of the buried hills [14]. For example, the core observation and image logging of well XL83a show that the fracture strikes were mainly NNW–SSE, consistent with the strike of regional fault system.

The subduction of the Pacific plate beneath the Eurasian plate uplifted the buried hills and resulted in weathering and erosion. The buried hills were also subjected to sinistral strike-slip stress, forming a fracture system along the NNE–SSW direction [38].

With continued extension in the Eocene era, the rifting scale expanded, and a rifted basin formed and began to accept deposition [39]. A series of normal faults with a NE–SW direction formed in a tensile environment [38], and mainly high-angle shear fractures with NE–SW orientations accompanied the faults. After the Early Miocene period, the fault activity in western SCS weakened, and the low uplift entered the depression stage, with few fractures [40].

These multi-stage tectonic activities fostered the development of buried-hill fractures and formed a fracture network dominated by a NNW–SSE strike, supplemented by an NNE–SSW strike and a NE–SW strike (Figures 2 and 3).

5.2. Formation Mechanism of (Ferro) Dolomite Veins

The formation of (ferro) dolomite veins in the different stages of buried-hill fractures in the XL area originated from fluids with different chemical characteristics.

The earliest precipitated dolomite mainly occurs close to the fracture surface, with weak fluorescence and no fluorescence band (Figures 5 and 6), and is usually formed by the opening and closing of fractures [41,42]. Dolomite was formed in multiple stages, possibly ascribed to the seasonality of organic matter or episodic charging of hydrocarbon. Episodic hydrocarbon charging can form banded fluorescent carbonate authigenic minerals [43]. However, there are no fluorescent oil–gas inclusions in the first-stage powdery dolomite veins in the research area, and the REE pattern is completely different than the right-leaning distribution curve of the REE pattern in the carbonate veins caused by diagenetic hydrocarbon-generating fluids [44], so the formation of the first-stage veins may have no linkage with hydrocarbon charging.

The first-stage powdery crystal dolomite is enriched in Mg but depleted in Fe and Mn (Figures 9 and 10). Freshwater-derived carbonate minerals in fractures lack Fe and Mn elements [42], so we speculated that the first stage of powdery dolomite was influenced by fresh water in the precipitation process.

REEs are good tracers to explore the genesis of rocks. In the first stage of powdery dolomite veins after chondrite standardization treatment, there is no obvious difference between the contents of LREE and HREE, and there is a strong negative Ce anomaly and a weak negative Eu anomaly (Figure 11), indicating that the first-stage veins are precipitated in a strong oxidation environment [45,46].

In the first stage, the $\delta^{13}\text{C}_{\text{PDB}}$ values of dolomite veins are $-3.61 \sim -6.56 \text{ ‰}$, the $\delta^{18}\text{O}_{\text{PDB}}$ values are $-7.26 \sim -19.14 \text{ ‰}$, and the $\delta^{18}\text{O}$ values are obviously negative (Figure 13). The $\delta^{13}\text{C}_{\text{PDB}}$ of carbonate minerals formed under the influence of atmospheric water is mostly $-7.0 \text{ ‰} \sim -3.5 \text{ ‰}$ [47–49]. There may be three causes for this negative $\delta^{18}\text{O}$ value: (1) atmospheric water infiltration and its corresponding water–rock interaction [50,51]; (2) increased pore water temperature [52]; and (3) decomposition of organic matter [53]. It is generally considered that atmospheric precipitation and/or diagenetic temperature are the most important factors in the control of $\delta^{18}\text{O}$, and the influence of other factors is relatively limited.

For formation water, the more intense the evaporation, the lighter the isotopes are that enter the steam, and the corresponding heavier isotopes are left in the water, causing the fluid oxygen isotopes to become heavier. The temperature during deposition can also affect the oxygen isotope value. The higher the temperature, the lower the oxygen isotope value in water, and the smaller the oxygen isotope value in cement. There is a corresponding functional relationship between the two [54,55]: $1000 \ln a_{\text{dolomite-water}} = 3.2 \times 10^6 \times T^{-2} - 1.5$, $a_{\text{dolomite-water}} = (1 + (\delta^{18}\text{O}_{\text{dolomite}}/1000))/(1 + (\delta^{18}\text{O}_{\text{water}}/1000))$.

In this study, through the comprehensive analysis of mineral precipitation temperatures and oxygen isotopes in the (ferro) dolomite veins, combined with the above fractionation formula, the relationship between the precipitation temperatures and oxygen isotopes of (ferro) dolomite veins in the XL area is established, and the oxygen isotope value of formation water during the different stages of mineral precipitation is given (Figure 14). During the formation of the first-stage powdery crystal dolomite, the variation in the oxygen isotopes of the fluid is small, and it basically varies between the mixed fluids of atmospheric freshwater and formation water [56,57]. Therefore, the formation process of the first-stage dolomite vein is mainly influenced by atmospheric fresh water.

The second-stage dolomite veins generally show an increased crystal size and a high degree of idiomorphism (Figures 5 and 6). The content of iron and manganese elements in the dolomite increased, while the content of Mg decreased (Figures 9 and 10). The interactions between the formation water and surrounding granite rock, e.g., near-surface weathering and leaching [58,59], and/or deep processes under elevated temperature–pressure conditions [60], can increase the content of iron and Manganese elements in the fluid. The REEs in the second stage of the dolomite veins after chondrite standardization treatment show a right-leaning distribution pattern (Figure 11). Most of the samples showed an obvious LREE enrichment, medium-intensity Eu positive anomaly and Ce positive anomaly (Figure 11), showing certain characteristics of hydrothermal dolomite [61]. The stable C and O isotope composition also shows that the formation of dolomite in the second stage is related to fluid characteristics rather than atmospheric water (Figure 13).

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in global seawater is basically the same in a certain geological history period, considering the long residence time of Sr [62]. Moreover, strontium isotopes do not produce significant fractionation in most geological processes [63], so the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in dolomite veins can represent the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of paleofluids, thus tracing the source of paleofluids. In the second stage, the Sr isotope ratio of the veins is $0.70769 \sim 0.70785$ (Figure 12), which shows a combined influence of formation water and deep hydrothermal fluid [64]. To sum up, the second-stage fine-crystal dolomite veins were produced in a buried diagenetic environment influenced by deep hydrothermal fluid. During evolution, the vein-forming fluid gradually became enriched in metal elements, such as Fe and Mn, and gradually lost REE (Figures 9–11).

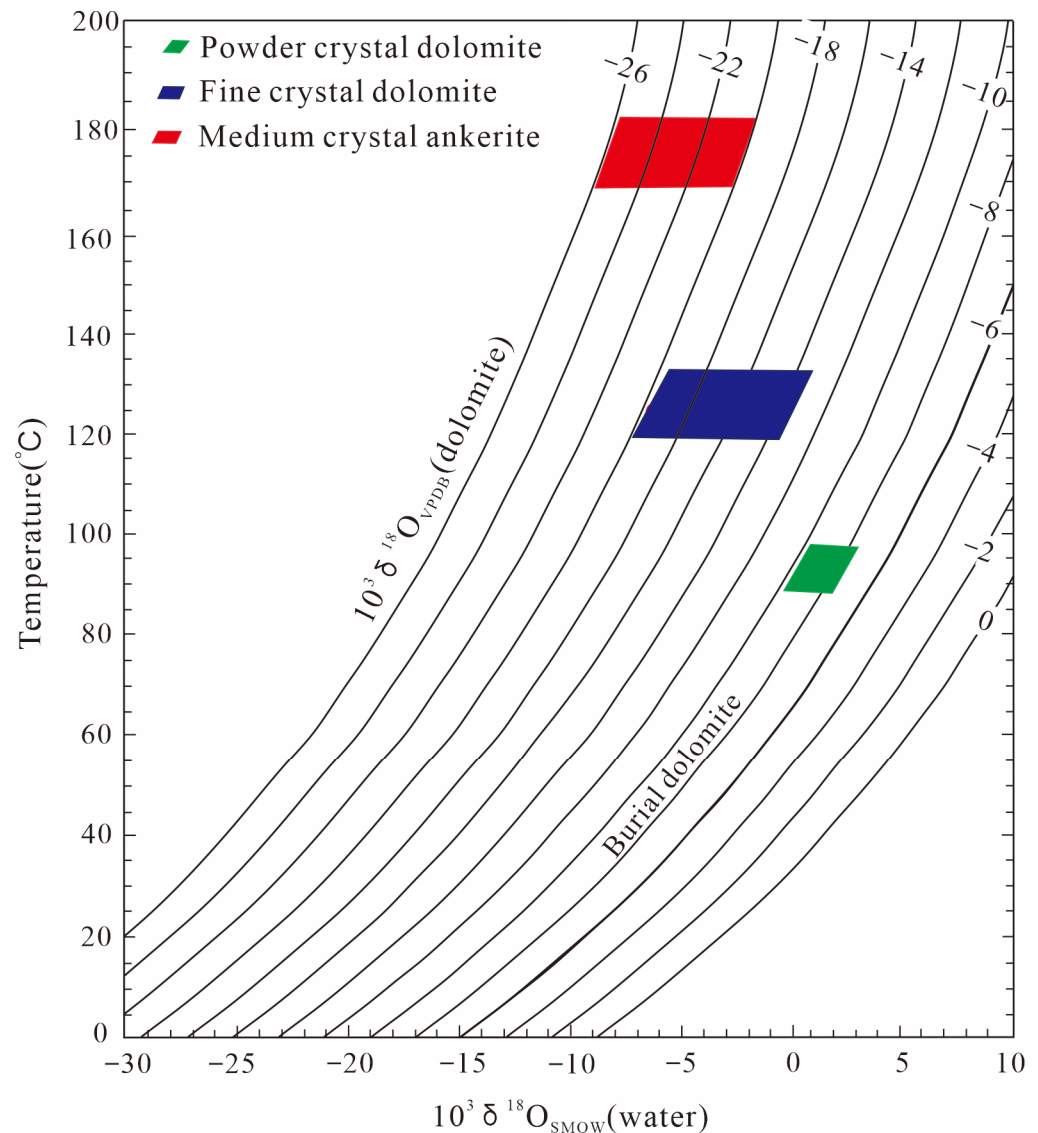


Figure 14. Temperature versus $\delta^{18}\text{O}$ diagenetic fluid for various $\delta^{18}\text{O}$ dolomite values that were reconstructed from equation $10^3 \ln a = 3.2 \times 10^6/T^{-2} - 3.3$ [54,55]. Green shaded areas mark preferred temperature ranges for powdery crystal dolomite veins, whereas blue and red shaded areas mark preferred temperature ranges for fine-crystal dolomite and medium-crystal ankerite veins, respectively.

The third-stage medium-crystal ankerite veins mainly extend along the edge of the previous second-stage veins, filling the remaining fracture space (Figure 5). The strontium isotope ratio of these veins is 0.70446–0.70526 (Figure 12), showing characteristics of deep hydrothermal fluid [65]. The stable C and O isotope compositions also show that the diagenetic fluid of ankerite in the third stage mainly comes from deep hydrothermal fluid (Figures 13 and 14). The contents of iron, manganese, aluminum, and silicon are obviously higher than those in the previous two stages (Figure 9), caused by iron-rich and aluminosilicate minerals in the granite around the fracture. The interactions between the deep hydrothermal fluid in the upwelling process and the surrounding granite minerals continuously increase the Fe, Mn, Al, and Si elements in the fluid [66]. In the third stage, the REE distribution curve of the veins is right-leaning, with an obvious LREE enrichment, strong Eu positive anomaly, and Ce positive anomaly (Figure 11), showing strong hydrothermal genetic characteristics [61]. The homogenization temperature of the second-stage fine-crystal dolomite vein, the third-stage medium-crystal ankerite veins,

and their associated quartz fracture brine inclusions (Figure 8) show that the buried-hill fractures in the XL area were invaded by at least two episodes of deep hydrothermal fluids.

5.3. Petroleum Geological Significance

The pattern and layout of the pre-Paleogene era buried-hill reservoirs in the XL area can be established with the help of cross-well seismic profiling (CSP) (Figure 15) and belong to a basement reservoir and combined trap. The reservoirs can be classified vertically as a weathering dissolution, weathering fracture, or structural fracture zone [63]. The reservoir space is dominated by fractures, with a few dissolution pores. Due to different degrees of weathering leaching, the dissolution pores gradually decrease with depth, with less dissolution pores in the structural fracture and weathering fracture zones. The vertical fracture development is characterized by a thin weathering dissolution and weathering fracture zone, and a thick structural fracture zone, as well as a wide horizontal distribution range [67]. Many open fractures can be discriminated in both the image logging and thin sections.

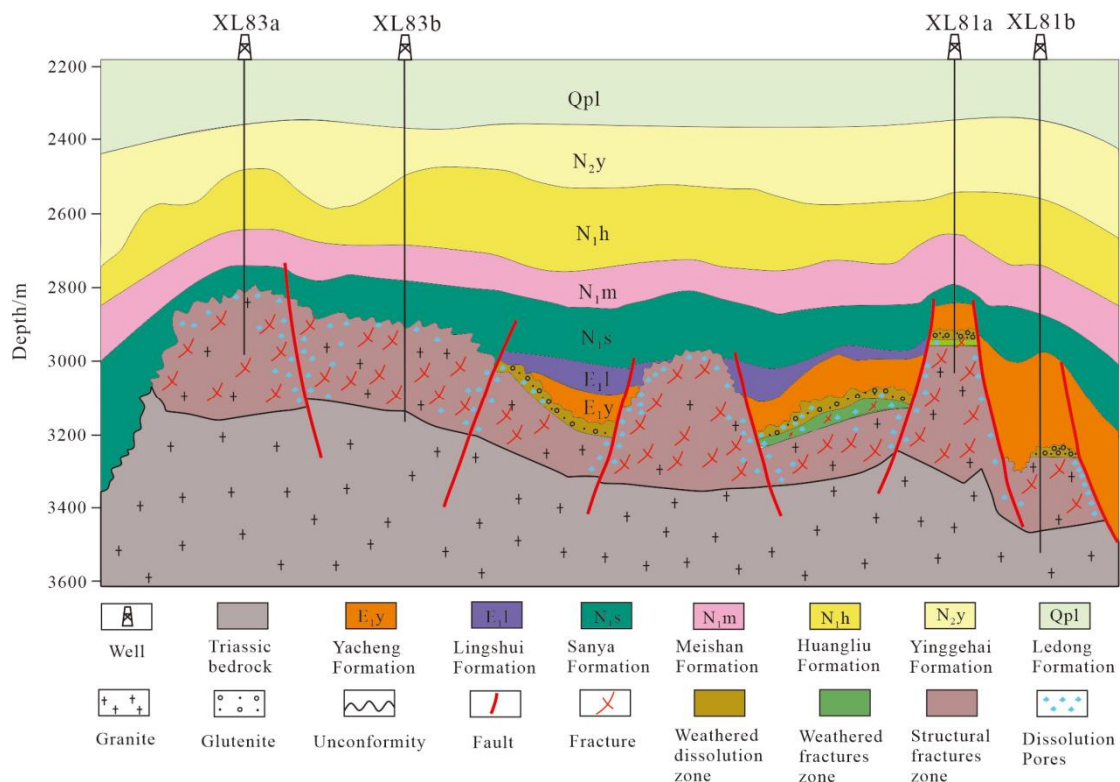


Figure 15. Schematic diagram of fractured gas reservoirs from buried hills in XL area.

Shear and tensile fractures have been extensively developed in the buried hills after multi-stage tectonic activities. The dissolution fractures related to tectonic movement also developed, from further dissolution and expansion of the original structural fracture due to the massive acidic substances derived from the hydrothermal fluid rising from the deep and large fault. The thin sections show that (ferro) dolomite and pyrite are developed in fractures, indicating deep hydrothermal fluid activity (Figure 6). The third type of diagenetic fracture rarely develops in the XL area. The comprehensive analysis shows that the granite buried-hill reservoir in the XL area is mainly controlled by three factors: tectonic activity, weathering leaching, and hydrothermal dissolution. There is a certain connection between the formation of the above three kinds of fractures. Diagenetic fractures are formed when magma cools and are solidified after magma intrusion, while structural fractures are formed under multi-stage tectonic movement, and dissolution fractures are formed when the fluid dissolves along the surface of structural fractures. The three types of fractures

show a crossing distribution in space and form a widely distributed fracture system in the buried hill, effectively improving the physical conditions of the reservoirs.

In the XL area, constructive diagenesis includes weathering, denudation, leaching, and dissolution, whereas destructive diagenesis involves filling, especially during the three-stage filling of (ferro) dolomite veins. Both core data and microscopic observations show that 45% of the fractures are fully or half-filled by (ferro) dolomite veins (Figures 4 and 5). In the first stage, powdery crystal dolomite mainly filled primary pores or fractures with small openings. In the second stage, fine-crystal dolomite mainly filled fractures with large openings, and in the third stage, the medium-crystal ankerite further filled residual fractures (Figures 5 and 6). Large quantities of faults and fractures were generated by several tectonic movements, and the relatively oxidized diagenetic environment was formed by atmospheric fresh water infiltration along the faults into the fracture formation water in the buried hill in the first stage. After the Early Miocene era, the influence of atmospheric water gradually declined with the depth of the buried hill. Combined with the intrusion of deep hydrothermal fluid and the influence of iron-rich and aluminosilicate minerals in the surrounding rock, the elements Fe, Mn, Al, and Si in the diagenetic fluid increased. The second and third stages of (ferro) dolomite precipitation further filled residual fractures. The fractured reservoirs of the buried hill in the XL area are still of high quality even though they have been affected by the three-stage filling of the (ferro) dolomite veins, and the extensively distributed fractures have effectively improved the physical condition of the reservoirs. The resource assessment shows that the fractured reservoirs of the granite buried hills in the XL area have a natural gas potential of over 100 billion m³ [11].

6. Conclusions

The buried-hill fractures in the XL area can be classified into structural, dissolution, and diagenetic types, based on their formation characteristics. They are dominated by medium-high angle structural fractures and are mainly divided into three stages. The different stages of fractures are interlaced with each other and associated with faults. The fractures strikes are mostly NNW–SSE, followed by NNE–SSW and NE–SW, all of which are consistent with the strike of the fault patterns formed by the tectonic events of the QDN Basin. The three types of fractures are cross-cutting, forming a network of fractures widely distributed inside the buried hills.

The three stages of (ferro) dolomite veins developed from the three stages in the buried-hill fracturing of the XL area. The first stage, involving powdery crystal dolomite veins, mainly grew close to the fracture surface, with the highest strontium isotope value, while being rich in Mg and having an extremely low content of Fe and Mn. The first stage of veins formed in a relatively open oxidation diagenetic environment. The vein-forming fluid shows mixed characteristics of formation water and meteoric freshwater. The second stage, involving fine-crystal dolomite veins, grew on the first type of powdery crystal dolomite, perpendicular to the fracture surface, and had a high degree of idiomorphism. Between one and two groups of cleavages were observed. The content of Fe and Mn increased, and the content of Mg decreased. The second stage of the veins formed in a buried diagenetic environment mixed with formation water and deep hydrothermal fluids. The third stage, involving medium-crystal ankerite veins, continued growing on the second-stage fine-crystal dolomite veins. They have the lowest strontium isotope value and the lightest oxygen isotope value. The contents of Fe, Mn, Al, and Si elements were significantly higher than those of the first and second stages, as a result of the deep hydrothermal effect. The diagenetic material was sourced from the interaction between the hydrothermal fluid and the iron-rich and aluminosilicate minerals in the surrounding granite of the fractures.

The QDN Basin of SCS has undergone multi-stage tectonic movements of extrusion, strike-slip transformation, extension, and tilting uplift. A large-scale fracture network system has been formed inside the Mesozoic-era buried hills in the XL area. Despite the filling of the three stages of (ferro) dolomite veins, the widely distributed networked

fractures have improved the physical conditions of the gas reservoirs, which are still of high quality.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/jmse12111970/s1>, Table S1: List of methods used, sample types, and sample numbers.

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