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Original research paper

Scale and distribution of marine carbonate burial dissolution pores☆

Anjiang Shen [a,b,*], Min She [a,b], Anping Hu [a,b], Liyin Pan [a,b], Junming Lu [a]

[a] PetroChina Hangzhou Institute of Geology, Hangzhou, Zhejiang 310023, China
[b] CNPC Key Laboratory of Carbonate Reservoirs, Hangzhou, Zhejiang 310023, China

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Abstract

It is gradually accepted that porosity can be created in burial settings via dissolution by organic acid; TSR derived or hydrothermal fluids. The role of deep-buried carbonate reservoirs is becoming more and more important since the degree and difficulty in petroleum exploration of shallow strata are increasing. A profound understanding of the development scale and prediction of the deep-buried carbonate reservoirs is economically crucial. In addition to the formation mechanism, scale and distribution of burial dissolution pores in burial settings are focused on in recent studies. This paper is based on case studies of deep-buried (>4500 m) carbonate reservoirs from the Tarim Basin and Sichuan Basin. Case studies mentioned includes dissolution simulation experiments proposes that an open system is of crucial importance in the development of large-scale burial dissolution pores, the distribution pattern of which is controlled by lithology, pre-existing porosity, and pore throat structures. These findings provided the basis for evaluation and prediction of deep-buried carbonate reservoirs.

Keywords: Carbonate; Large scale reservoir; Burial dissolution pore; Simulation experiment; Open system; Pore throat structure

1. Introduction

In previous studies, scholars often focused on the formation mechanism of burial dissolution pores. Surdam et al. [1,2] presented that secondary dissolution pores in sandstone were created via dissolution of feldspar and clipped by the organic acid produced from hydrocarbon generation. Meanwhile, Cai et al. [3] studied the origin and distribution of organic acid and the effects of pore increase in carbonate reservoirs in the Tarim Basin. Another finding is that Fan et al. [4] proved that organic acid increased pores in carbonate reservoirs in burial settings through simulation experiments. Additionally Xiao et al. [5] discussed the distribution characteristics of H_2S in the Eastern Sichuan and its effect on the phenomenon in which pore increase in oolitic beach reservoirs of the Feixianguan Formation. Both Bildstein et al. [6] and Cross et al. [7] analyzed the reaction mechanism and dynamic characteristics of TSR and its influence on diagenesis. Relatively, Cai et al. [8] investigated the origin of TSR in sedimentary basins and its influence on reservoir development. Zhu et al. [9] and Zhang et al. [10] stated that TSR was an important type of formation of deep high-quality carbonate reservoirs in the Sichuan Basin, and it had a catalytic effect on crude oil pyrolysis. Luo et al. [11] discussed the reaction mechanism of TSR and its influence on crude oil pyrolysis and reservoir reformation. Zhao et al. [12] believed that acid fluid such as organic acid and H_2S was crucial to the expansion of secondary pores and the increase of new dissolution pores. Graham et al. [13] worked on the hydrothermal dolostone reservoirs controlled by faults. Jin et al. [14] studied the

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* Corresponding author. PetroChina Hangzhou Institute of Geology, Hangzhou, Zhejiang 310023, China.
E-mail address: shenaj_hz@petrochina.com.cn (A. Shen).

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influence of hydrothermal activity on reservoir reformation in the Tarim Basin, and presented that a lot of micro dissolution vugs produced improved the reservoir properties significantly. It is progressively being accepted that burial dissolution pores can be created in burial settings via dissolution by organic acid, TSR derived, or hydrothermal fluids.

Through the increase of the degree and difficulty in petroleum exploration of medium to shallow strata, the exploration and development of deep strata are becoming ever more vital. However, in comparison with medium to shallow strata, petroleum exploration in deep strata would be much more expensive. Therefore, higher requirements are put forward for the development scale and prediction accuracy of deep-buried carbonate reservoirs. In addition to the formation mechanism, the scale and distribution of burial dissolution pores in marine carbonate reservoirs are the focus in recent studies. In this paper, based on numerous experimental studies, including case studies and quantitative dissolution simulation experiments of deep-buried (>4500 m) carbonate reservoirs from both the Tarim and Sichuan basins, simulation experiments for the influence of mineral components on dissolution intensity, simulation experiments for the influence of reservoir properties on dissolution intensity and simulation experiments for the influence of lithology and pore throat structure on dissolution effect, it is presented that open system is of crucial importance in the development of large-scale burial dissolution pores, the distribution of burial dissolution pores is controlled by pre-existing pore developed zones, and the distribution pattern of burial dissolution pores is controlled by reservoir lithology and pore combinations. These findings provide the basis for evaluation and prediction of deep-buried carbonate reservoirs.

2. Open system's crucial importance in the development of large-scale burial dissolution pores

In order to identify the importance of the open system to the development of large-scale burial dissolution pores, a quantitative dissolution simulation experiment was designed. This experiment was performed on the HTHP dissolution kinetics physical simulation apparatus in the CNPC Key Laboratory of Carbonate Reservoir. This apparatus is a set of independently designed nonstandard equipment manufactured in France. It could simulate chemical reactions between rock and fluid from room temperature up to 400 °C, and ordinary pressure up to 100 MPa. It is close to real diagenetic environment, it could simulate internal dissolution of many kinds of rocks in a variety of fluids in open−close system, in dynamic−static (at different flow rates) state, and different temperature − pressure conditions, detect the fluid components and contents quantitatively after reaction, and monitor real-time permeability. Therefore, it is very helpful in studying the mechanism of pore development in deep-buried reservoirs. This apparatus have been granted a Utility Model Patent (Patent No.201120344178.X) and a Patent for Invention (Patent No. 201110271800.3).

In the simulation experiment, two kinds of reservoir samples were used, i.e. porous reservoir sample and fractured−vuggy reservoir sample. The samples were taken from the Longwangmiao Formation, the Changxing Formation, and the Feixianguan Formation in the Sichuan Basin. They had a porosity of 9.85%, 11.78%, and 10.15%, permeability of 2.17×10^{-3} μm^2, 1.84×10^{-3} μm^2, and 6.19×10^{-3} μm^2, respectively. Pores are believed to be a result of burial dissolution and expansion on the basis of pre-existing pores in a supergene environment (Fig. 1) [15,16]. The reaction liquid measures to be 2 ml/L acetic acid solution in an open−flowing system at the flow rate of 1 ml/min. The dissolution is internal, and simulation experiments at 11 temperature/pressure points were performed. See Fig. 2 for experiment results.

The experiment results showed that comparing with fractured−porous dolostone reservoir samples, the porous dolostone reservoir samples reaches chemical balance easier. As the acetic acid flow at the rate of 1 ml/min, when all these three samples reach the common point of chemical thermodynamic balance, the corresponding pressure is 40 MPa, the temperature is 135 °C, and the depth is about 4000 m (threshold depth of deep burial). Additionally, the concentration of $Ca^{2+}+Mg^{2+}$ is 12−18 mmol/L. When the temperature and pressure increase to 60 MPa and 189 °C, the concentration of $Ca^{2+}+Mg^{2+}$ changes slightly. It may be understood that the solution saturation of $Ca^{2+}+Mg^{2+}$ in dolostone at the deep-buried reservoir is 12−18 mmol/L approximately. It indicates that dolostone could be dissolved greatly in an acid environment at high temperature and pressure as well as the dissolution is controlled by pore throat structure. Therefore, in a deep-buried open system, with the continuous flow of acid

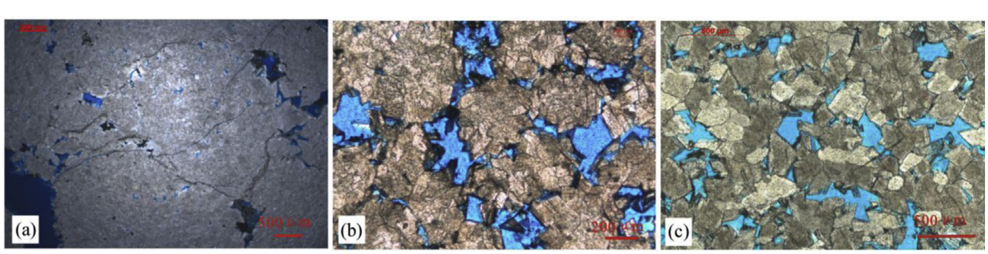

Fig. 1. The characteristics of samples for quantitative dissolution simulation experiment. (a) Dolarenite, fractured-vuggy dolostone reservoir, Cambrian Longwangmiao Formation, Moxi area in the Sichuan Basin, Well MX13, 4614.75 m, casting thin section, single polar; (b) Oolitic dolostone, oolite composed of aplite dolostone, porous dolostone reservoir, Triassic Feixianguan Formation, Luojiazhai area in the Sichuan Basin, Well LJ1, 3509.71 m, casting thin section, single polar; (c) Fine crystal dolostone, porous dolostone reservoirs, Permian Changxing Formation, Longgang area in the Sichuan Basin, Well LG001, 6070.55 m, casting thin section, single polar.

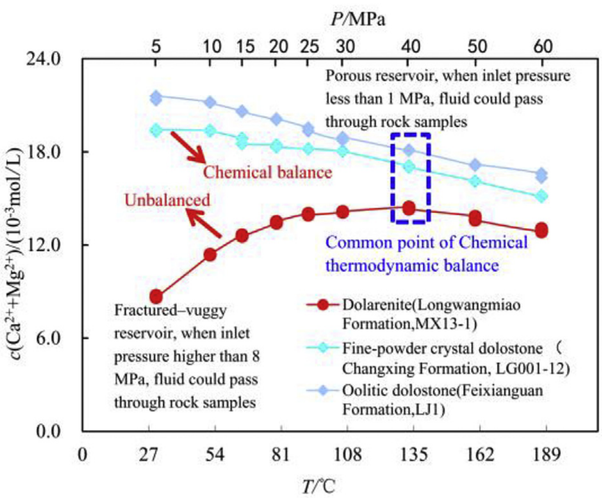

Fig. 2. Quantitative dissolution simulation experiment.

fluid, acid fluid with saturated Ca^{2+}+Mg^{2+} may be transported continuously, acid fluid with unsaturated Ca^{2+}+Mg^{2+} may be supplemented in time, and dolostone may be dissolved continuously. When time is sufficient, large-scale dissolution pores may be created. The property changes of fine crystal dolostone of Changxing Formation from 40 MPa to 135 °C to 60 MPa and 189 °C show that, large-scale pores are produced in burial environment via dissolution by organic acid, TSR, and hydrothermal fluid (Table 1), and the properties besides pore throat structure of pre-existing reservoirs are improved significantly.

At 60 MPa and 189 °C, together with static acetic acid, the fluid concentration after 30 min changed a little, indicating that dolostone did not dissolve or precipitate, it may also imply that dissolution and precipitation reached dynamic balance without an increase or decrease in the amount of pores. It shows that a close system is important for preservation of pre-existing pores.

In an open system in a burial setting, new pores may be created from burial dissolution, but it mainly occurs at high fluid potential positions. At low fluid potential positions, fluid with supersaturated Ca^{2+}+Mg^{2+} will precipitate and clog the pre-existing pores. The Sinian Dengying Formation in the Gaoshiti−Moxi area, Sichuan Basin is a good example of precipitation in an open system. In the Dengying Formation, two phases of interstratal karstification were developed. At the end of the sedimentary stage of the Deng-2 Member, Episode I of Tongwan movement made the Deng-2 Member uplift and experience weathering denudation, thus, forming interstratal karst reservoirs on top of the Deng-2 Member. At the end of the sedimentary stage of the Deng-4 Member, due to the uplift of Episode II of the Tongwan movement, the Deng-4 Member was subject to leaching and denudation to different degrees, resulting in different formation thickness. The Deng-3 Member in local areas (e.g. Weiyuan and Ziyang) was denudated partly or completely. The Deng-2 Member was immediately covered by the Lower Cambrian strata with unconformable contact forming interstratal karst reservoirs on top of the Dengying Formation [17,18]. The reservoir spaces are vugs in centimeters to tens of centimeters, which were packed with cement at different stages, forming the so-called snowflake-shaped or botryoidal structure [19,20]. The cementing order from surrounding rock to the center of vug is, surrounding rock → dark lace dolostone → light lace dolostone → fine − coarse crystal dolostone → saddle dolostone (Fig. 3a, b, c), with some vugs remaining. On the Nanjiang Yangba profile and the Ebian Xianfeng profile, fracture-vug rate of the Deng-2 Member algal dolostone is over 30%. After multi stages of cement packing, the fracture-vug rate remains at 5%−10%. Such large-scale cement in burial environment must result from large-scale dissolution at high fluid potential positions, which provides supersaturated diagenetic fluid or origin for dolostone precipitation at low fluid potential positions.

To sum things up, it is easy to understand that close system preserved pre-existing pores. In a close system, when dissolution and precipitation reach chemical balance, no pores are created or destroyed. Therefore, a close system is important to preserve pre-existing pores. In an open system, pores are enriched or depleted, large-scale dissolution pores are created by dissolution at high fluid potential positions, and pores at low fluid potential positions are destroyed by precipitation.

3. The influence of pre-existing pore developed zones in the distribution of burial dissolution pores

As mentioned above, burial environment is a place to preserve and adjust pre-existing pores. Pore adjustment

Table 1
Change of reservoir physical property and pore throat characteristics during quantitative dissolution simulation experiment.

Item	Analysis item	40 MPa, 135 °C	60 MPa, 189 °C
General statistics	Porosity/%	16.82	19.54
	Pore throat volume/μm^3	4.97e+10	5.67e+10
	Connecting volume/μm^3	1.84e+10, as 35.60% of total volume	4.05e+07, as 71.40% of total volume
Pores	Quantity	53857	25889
	Volume/μm^3	3.92e+10	4.32e+10
	Radius/μm	Average: 30.17; min: 3.107; max: 211.5	Average: 36.12; min: 3.324; max: 961.8
Throats	Quantity	39617	17322
	Volume/μm^3	1.06e+10	1.36e+10
	Radius/μm	Average: 18.67; min: 2.77; max: 152.7	Average: 26.02; min: 3.061; max: 746.9

Fig. 3. Botryoidal dolostone of the Sinian Dengying Formation in the Moxi-Gaoshiti area, Sichuan Basin. (a) Algal dolostone, showing surrounding rock → dark lace dolostone → light lace dolostone → fine-coarse crystal dolostone → saddle dolostone, remaining vugs (>2 cm), Deng-2 Member, Ebian Xianfeng outcrop profile; (b) Algal dolostone, showing surrounding rock → dark lace dolostone →vlight lace dolostone → fine-coarse crystal dolostone, remaining vugs, Deng-2 Member, Ebin Xianfeng outcrop profile; (c) Algal dolostone, Showing surrounding rock → light lace dolostone→fine-coarse crystal dolostone, Deng-2 Member, Nanjiang Yangba outcrop profile.

includes the enrichment of pores due to developed burial dissolution pores and the depletion of pores due to precipitation. The investigation on pore enrichment mechanism is critical in studying the distribution rule of large-scale high quality deep reservoirs. For this purpose, the simulation experiment used in order to study the influence of mineral components on dissolution degree and the simulation experiment used to study the influence of reservoir properties on dissolution degree were formulated. Through comparing and analyzing the results of these two simulation experiments, it is understood that pre-existing pore developed zones control the distribution of burial dissolution pores, which provides a basis for distribution prediction of large-scale high-quality deep reservoirs.

In the simulation experiment performed to study the influence of mineral components on dissolution degree, two groups of samples were used, i.e. fine-powder crystal dolostone and limestone (micritic limestone, marl, and bioclasticmicritic limestone). For these tight rocks, surface dissolution experiment was performed, with the reaction fluid of 2 ml/l acetic acid solution and an open flowing system with the flow rate of 3 ml/min. The simulation experiment was performed at 13 temperature and pressure points, and the experiment time on each temperature and pressure point was 30 min. The experiment results are shown in Fig. 4. The experiments reveal that the dissolution rate of limestone in an acetic acid solution is higher than that of dolostone; with the increase of burial depth, the dissolution rate of both limestone and fine−powder crystal dolostone increases and converges.

In the simulation experiment to study the influence of reservoir properties on dissolution degree, two groups of samples, i.e. dolarenite and calcarenite, were used. These samples were taken from the Longwangmiao Formation and the Changxing Formation, both in the Sichuan Basin, their porosity was 19.76% and 4.44%, and permeability was 3.70×10^{-3} μm^2 and 1.71×10^{-3} μm^2, respectively. The physical properties of dolarenite are obviously better than those of calcarenite. The experiment was performed with a reaction fluid of 1 ml/L, which is an acetic acid solution, and an open flowing system, at the flow rate of 1 ml/min. The internal dissolution simulation experiment was performed at 11 temperature and pressure points. The temperature and pressure points were taken based on the recovered burial

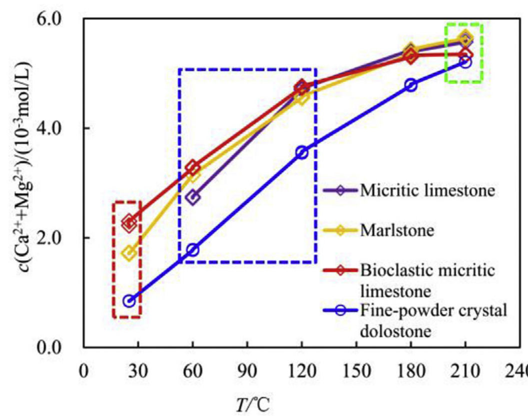

Fig. 4. Dissolution simulation experiment to study the influence of mineral components on dissolution degree (limestone ion concentration: Ca^{2+}, dolostone ion concentration: Ca^{2+}+Mg^{2+}).

history of real samples, and the experiment time at each temperature and pressure point was 30 min. The experiment results are shown in Fig. 5.

The simulation experiment to study the influence of mineral components on dissolution degree shows (Fig. 5) that in terms of the dissolution capacity, calcite is much easier to dissolve in

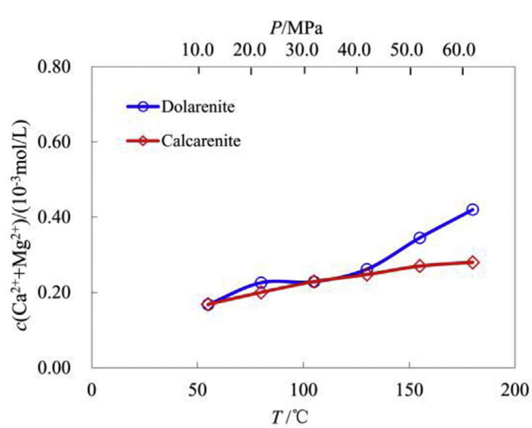

Fig. 5. Dissolution simulation experiment to study the influence of reservoir physical property on dissolution degree (limestone ion concentration: Ca^{2+}, dolostone ion concentration: Ca^{2+}+Mg^{2+}).

acid than dolostone. However, the simulation experiment to study the influence of reservoir properties on dissolution degree made the opposite conclusion, which is attributed to the property difference between dolarenite and calcarenite. The properties of dolarenite are much higher than those of calcarenite. Consequently, the specific surface area of dolarenite contacting acid fluid is more than that of calcarenite. Even if the dissolution rate of calcite is higher than that of dolostone, the concentration of Ca^{2+}+Mg^{2+} is still higher than the concentration of Ca^{2+} in the solution. This indicates that the rock porosity and permeability in burial environment have more control on the dissolution rate than the mineral components. In fact, it is easy to understand the influence of pre-existing pores on burial dissolution. For high porosity and high permeability rock, burial diagenetic media may easily enter the rock. The specific surface area of dissolution is great, and the dissolved products are easy to be removed. Even for insoluble dolostone, large-scale dissolution pores may be created within adequate time. On the contrary, for low porosity and low permeability rock, burial diagenetic media is difficult to enter, the specific surface area of dissolution is small, and the dissolved products are difficult to be removed. Even for soluble calcite, it is difficult to form large-scale dissolution pores within enough time. Therefore, original porosity and permeability of rock are necessary conditions for burial dissolution.

A supergene environment is important for pore development in reservoirs including sedimentary primary pores, early supergene dissolution pores, and late supergene dissolution pores. The sedimentary primary pores are related to the sediment characteristics controlled by sedimentary environment, the early supergene dissolution pores are related to synsedimentary exposed surface, and the late supergene dissolution pores are related to the interstratal karst surface and buried hill unconformity surface [21,22]. After the strata containing these pores were buried, they became the areas with the most active deep burial diagenetic media (organic acid, TSR, and hydrothermal fluid), where pores were adjusted. New pores were created as a result of dissolution at high fluid potential positions, and pores were plugged by precipitation at low fluid potential positions. This gives a good explanation for the case that the pores created by dissolution of the organic acid, TSR, and hydrothermal fluid are controlled by the original pore distribution prior to burial and have inheritance. This finding provides a basis for predicting the distribution of deep-buried large-scale reservoirs.

4. Lithology and pore combination in controlling the distribution pattern of burial dissolution pores

As discussed above, large-scale dissolution pores may be created by the dissolution of the organic acid, TSR, and hydrothermal fluid in deep burial strata and the distribution of pores is controlled by the original pore distribution prior to burial. In the following parts, the control of both the lithology and pore combination on the distribution pattern of burial dissolution pores will be discussed.

In order to identify the controlling factors in the distribution pattern of burial dissolution pores, the simulation experiment was performed to study the influence of lithology and pore combination on the dissolution effect. Four groups of samples (i.e. porous dolostone reservoir, fractured−vuggy dolostone reservoir, porous limestone reservoir, and fractured limestone reservoir) were used. The dolostone samples were taken from the Feixianguan Formation and the Longwangmiao Formation in the Sichuan Basin, they had a porosity of 10.15% and 10.80%, a permeability of 6.18×10^{-3} μm^2 and 32.41×10^{-3} μm^2, respectively. The limestone samples were taken from the Yijianfang Formation and the Yingshan Formation in the Tarim Basin, the porosity was 7.08% and 2.79%, and their permeability was 0.65×10^{-3} μm^2 and 3.34×10^{-3} μm^2, respectively. The experiment was performed using the reaction fluid of 2 ml/L acetic acid solution and an open flowing system with a flow rate of 2 ml/min. The internal dissolution simulation experiment was performed at 5 temperature and pressure points, ranging from 40 MPa to 135 °C to 60 MPa and 189 °C. The temperature and pressure points were taken based on the recovered burial history of real samples, and the experiment time at each temperature and pressure point was 60 min. The experiment results are shown in Fig. 6 and Fig. 7.

The simulation experiment results (Fig. 6) show that porous dolostone reservoirs' burial diagenetic media entered pore system dispersively, which increased the specific surface area of dissolution. The matrix porosity also increased. After the dolostone dissolution, porosity increased by 2%−3%, and permeability increased by $(4.75−7.48) \times 10^{-3}$ μm^2. It's still a porous pore combination. For fractured-porous dolostone reservoirs, due to the existence of fractures burial diagenetic media transported along fractures mostly (fractures are high-rate passages for fluid), and seldom entered pore systems dispersively. The dissolution fractures and vugs expanded along the fractures. The fracture-vuggy porosity increased, but the matrix porosity did not increase. The permeability may increase by three orders of magnitude. The pore combination changed from fractured-porous type to fractured-vuggy type.

For porous limestone reservoirs (Fig. 7), burial diagenetic media entered the pore system dispersively at first, but continuous dissolution will dissolve or collapse pore framework completely because limestone is much more soluble than dolostone, thus, forming fractured−vuggy pore combination. For fractured or fractured−porous limestone reservoirs like fractured-porous dolostone reservoirs, burial diagenetic media are transported mostly along the fractures which are high-rate flowing passages for fluid, it is seldom that the pore system gets entered dispersively. The dissolution fractures and vugs expanded along the fractures. The fracture-vug porosity will increase, but matrix porosity does not increase. The permeability may increase by three orders of magnitude. The pore combination changed from fractured type and fractured-porous type to fractured−vuggy type.

To sum things up, lithology and pore combination are important factors to control the distribution pattern of burial dissolution pores. For porous dolostone reservoirs, the porosity increased after burial dissolution, but the pore combination did

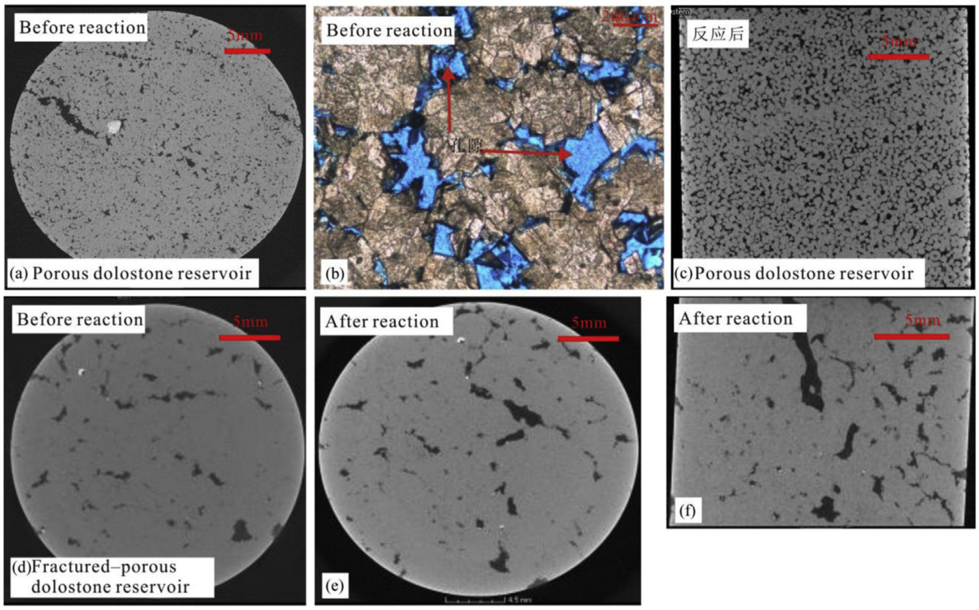

Fig. 6. Dissolution simulation experiment to study the influence of lithology and pore combination on the dissolution effect (dolostone). (a)–(c) Porous dolostone reservoir. (a) and (b) are pore combination before reaction, (c) is pore combination after the reaction. The porosity increased obviously, but pore combination type did not change, still porous dolostone reservoir. (d)–(f) Fractured-porous dolostone reservoir. (d) is pore combination before reaction, (e) and (f) are combination after the reaction. Fractures became the prevailing flowing passages, the fracture porosity increased but matrix porosity did not change. As a result, fractured-porous dolostone reservoir changed to fractured-vuggy dolostone reservoir, and the pore combination changed.

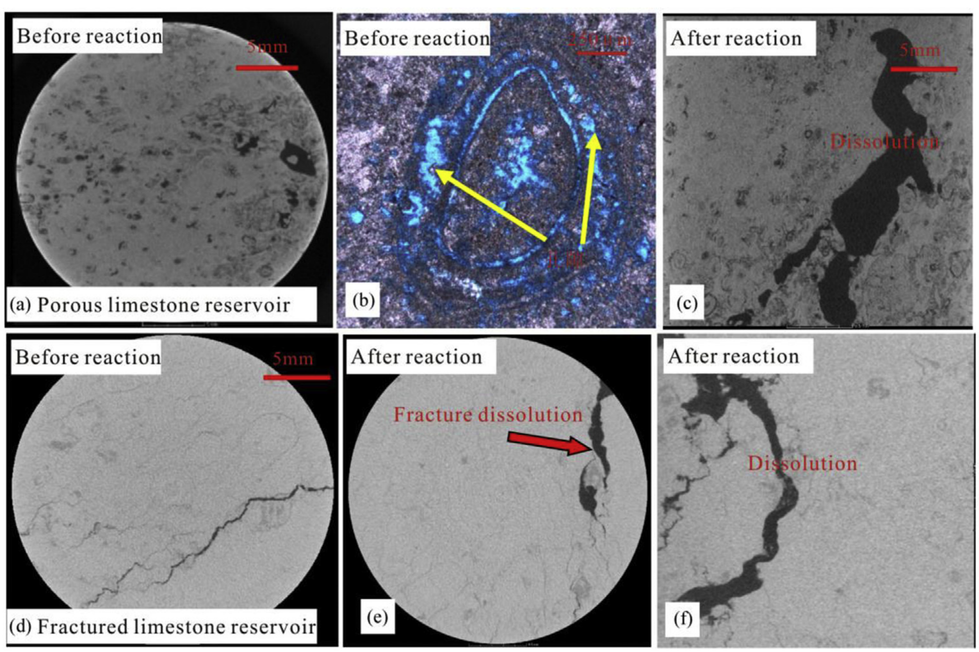

Fig. 7. Dissolution simulation experiment to study the influence of lithology and pore combination on the dissolution effect (limestone). (a)–(c) Porous limestone reservoir. (a) and (b) are pore combination before reaction, (c) is pore combination after the reaction. The fracture porosity increased but matrix porosity did not increase. As a result, porous limestone reservoir changed to fractured-vuggy limestone reservoir. (d)–(f) Fractured limestone reservoir. (d) is pore combination before reaction, (e) and (f) are pore combinations after the reaction. Fractures became the prevailing flowing passages. The fracture porosity increased but matrix porosity did not increase. As a result, fractured limestone reservoir changed to fractured-vuggy limestone reservoir.

not change it remained a porous type. For fractured-porous dolostone reservoirs, continuous burial dissolution changed the pore combination to fractured-vuggy type. For limestone reservoirs, no matter what the original pore combination was (porous, fractured-porous, or fractured), it finally changes to fractured-vuggy pore combination after continuous burial dissolution. This well explains the pore combination characteristics of ancient deep-buried marine carbonate reservoirs in China, fractured-vuggy reservoirs seen in limestone and dolostone strata, and porous reservoirs mainly seen in dolostone strata.

5. Conclusions

Through the simulation experiments for carbonate samples from the Tarim Basin, the Sichuan Basin, and the Ordos Basin, main findings are obtained in three aspects.

(1) The close system is important for the preservation of pre-existing pores and an open system is the key for development of large-scale burial dissolution pores. Large-scale dissolution pores were created by means of dissolution at high fluid potential positions, and pores at low fluid potential positions were destroyed by precipitation. In a very lengthy burial environment, absolute close systems are very rare; instead, an open system alternated with the close system, and positions will change between high fluid potential positions and low fluid potential positions due to structural inversion. Therefore, a right judgment is crucial to the prediction of reservoir distribution.

(2) Pre-existing pore developed zones control the distribution of burial dissolution pores with the estate. After the strata containing sedimentary primary pores, namely, early supergene dissolution pores and late supergene dissolution pores, were buried, they became the areas with the most active deep burial diagenetic media (organic acid, TSR, and hydrothermal fluid). New pores were created as a result of dissolution at high fluid potential positions, and pores were plugged by precipitation at low fluid potential positions. This finding provides a basis for predicting the distribution of deep-buried large-scale reservoirs.

(3) Lithology and pore combination control the distribution pattern of burial dissolution pores. For porous dolostone reservoirs, porosity increases after burial dissolution, but the pore combination did not change, it remains a porous type. For fractured-porous dolostone reservoirs, continuous burial dissolution changed the pore combination to fractured-vuggy type. For limestone reservoirs, no matter what the original pore combination was, such as porous, fractured-porous, or fractured, after continuous burial dissolution it may finally change to fractured-vuggy pore combination.

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Conflict of interest

The authors declare no conflict of interest.

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