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MONITORING METHANE HYDRATE FORMATION AND REFORMATION IN A PARTIALLY SATURATED SAND PACK USING LOW-FIELD MRI

Yuying Zhang, Yuechao Zhao*, Yongchen Song*
Key Laboratory of Ocean Energy Utilization and Energy Conservation of Ministry of Education, Dalian University of Technology, Dalian 116024, China
Corresponding Author: zhaoychao@163.com

ABSTRACT

The hydrate formation and reformation reduce mining efficiency and safety during natural gas hydrate exploration and natural gas pipeline transportation. In this paper, the methane hydrate formation and reformation in partially water saturated consolidated porous media were studied by low field MRI. The results show that the change of water content was ununiform both in time and space. The water content reduction rate displayed as fast-slow-fast-slow periodic rhythm; Hydrate formed prior in the bigger pores due to the bigger contact area between water and gas and lower pore water saturation; The formed hydrate made the pore structure inside of porous media more complicated; The rate of hydrate reformation was faster than that of hydrate formation because the residual hydrate microcrystalline structure after decomposition provided the place of dendrites nucleated and grew. This research can give some implications for delivering natural gas safely and studying natural gas hydrate reservoirs.

Keywords: methane hydrate, formation, reformation, low-field MRI

NONMENCLATURE

Abbreviations	
MRI	Magnetic Resonance Imaging
T_2	Transverse relaxation time
MH	Methane hydrate

1. INTRODUCTION

At present, the world is facing enormous energy challenges, less per capita resources, and traditional energy is more destructive to the environment. As a new type of energy, MH has the advantages of high utilization efficiency and less pollution to the environment, so it is considered a potential energy source in the future [1, 2]. At the same time, the formation of MH may lead to unsafe flow of natural gas in pipelines. Therefore, it is of great significance to study the characteristics of MH formation and reformation. At the same time, there are many studies shown that MRI technique is a powerful analytical tool for noninvasive multidimensional quantitative analysis of flow and transport in porous media, such as the hydrate [3,4]. It provides a lot of technical support on fluid characteristics and hydrate research in recent years [5].

There are some researchers monitored MH formation/ reformation process using MRI [6-11]. Song at al. [6] investigated the effects of methane flow rate and the initial water saturation on MH formation/ reformation. Zhao et al. [7] studied natural gas production using the depressurization method in porous media. They observed that hydrate reformation always occurs in the interior of porous media near the production well, which means that the hydrate formation related to heat transfer. Wang at al. [8] studied the effects of residual water, methane flow rate and residual MHs on its reformation in a porous medium. The methane flow affected the capillary force distribution then further influenced the pore water distribution in porous media. Taewoong et al. [9] found that hydrate reformed during the process of hydrate dissociation, and the temperature and salinity of water had an impact on the regeneration. The higher the salinity is, the easier it is to inhibit the reformation of hydrate. But some researches show that low field MRI technology has the advantages of more convenient and faster than high field MRI technology [10,11]. It can directly quantify the fluid content, which makes the observation of MH change process more intuitive.

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In this study, MH formation and reformation in partially water-saturated unconsolidated porous media were studied using low-field MRI. Water content change in time/space and the priority position of MH formation in the porous media were monitored by applying the pure phase encoding MRI technique. This research can provide some experimental data for delivering natural gas safely and studying natural gas MH reservoirs.

2. EXPERIMENTAL MATERIAL AND METHODS

2.1 Experimental Apparatus

Fig. 1 shows the experimental system with a vertical magnetic field strength of 0.9 tesla. Low-field MRI system (Oxford Instruments, Witney, UK) was performed at 5 MPa/0.5 ℃. The self-designed MRI vessel was made of polyimide to hold the sample, and was placed vertically in the magnetic field. A gas cylinder was full of CH_4, which was used to supply methane gas to the sample vessel using a gas injecting ISCO pump (260D, Teledyne ISCO Inc., Lincoln, NE, USA). The sample vessel was vacuumed using a vacuum pump (E2M1.5, Edwards vacuum, Bolton, England) and was saturated with water using another ISCO pump. The temperature of the sample was controlled by a circulator (Julabo, Seelbach, Germany) , employing the Fluorinert FC-40 (US 3M) as the confining fluid. The images of fluids containing ^1H proton were measured using a NMR spectrometer, which would be displayed in the data acquisition system (the Green Imaging Technologies' advanced software).

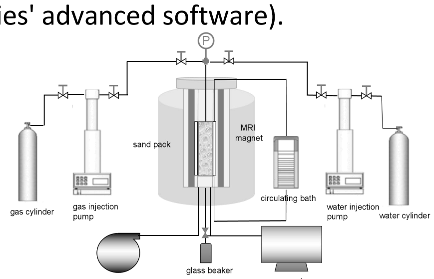

Figure 1. Experimental system

2.2 Materials

The porous media was simulated using a ununiform sand pack, with length 50mm, diameter 25 mm and a porosity of 45.8%. Initial water saturation was 51.63%. The methane gas (purity 99.99%) was supplied by Dalian Date Gas Co., LTD. The deionized water was used to saturate porous media.

2.3 MRI measuring methods

A Carr−Purcell−Meiboom−Gill (CPMG) method and an inversion recovery method were utilized to determine the bulk T_2 relaxation time of the sample [12,13], respectively.

The bulk T_2 distribution acquired with two signal averages evaluated the pore structure and the fluids distribution change. T_2 Max was 300 ms, while the recycle time was 2250 ms.

2.4 *Experimental steps*

Put the sample vessel into the central of the magnetic field and connected the piping; Tested the leakage of the whole system at 6 MPa for 12 h; Vacuumed and saturated the porous media, and then tested the parameters in the fully-saturated state when the temperature was stable at 0.5 ℃ ; Drained some water out of the sample until it reached a certain initial water saturation; Injected gas to 5 MPa and used the ISCO pump to maintain the pressure, then started the sequence collection during the process of MH formation; At the end of MH formation test, reduced the pressure to 2 MPa and started the MH decomposition test; Injected gas to 5 MPa again, collected the sequence during the process of MH reformation; Reduced the pressure to 0.1 MPa and finished the test. Cleared the piping and prepared for the next one test.

3. RESULTS AND DISCUSSION

In a series of tests, the effect of different initial water saturation on MH formation and reformation process were studied. The experimental results showed that when the initial water saturation increased to 51.63%, the rate of MH formation and reformation both started to display as a fast-slow-fast-slow rhythm until the end of test. The specific causes need to be found and verified by experimental data, so the process of MH formation and reformation in the partially water saturated sand-pack with initial water saturation of 51.63% was selected as the research object. The experimental results are as follows:

3.1 Overall change in residual water saturation

Figure 2 shows the variation of the T_2 distribution in the process of MH formation and reformation. The transverse relaxation time represents the pore size, while the integral area under the T_2 distribution represents the water content [14]. According to the T_2 transverse relaxation time, the pores could be divided into two types: bigger pores (T_2 > 10 ms) and smaller pores (1ms < T_2 < 10 ms).

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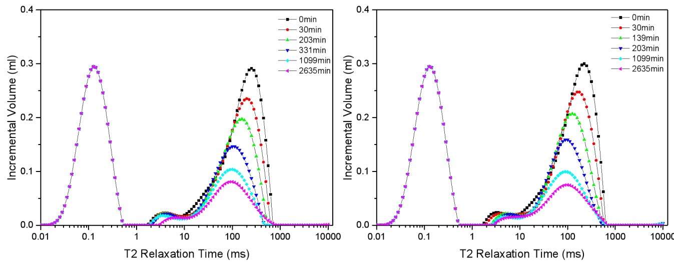


(a) (b)

Figure 2. The variation of the T_2 distribution in the process of MH formation (Figure 2(a)) and reformation (Figure 2(b)).

Figure 2(a) shows the variation of the T_2 distribution in the process of MH formation. T_2 distribution shifted from the black points to the pink points, while transverse relaxation time of water in the bigger pores shifted from high to low and that of water in the smaller pores shifted from low to high. And the signal amplitude decreased significantly, which meant that the formed MH changed the pore structure. Some smaller pores were completely occupied by the formed MH, while bigger pores were partially occupied.

Besides, the residual water saturation could be calculated as the ratio of water content at different times to that in the fully water saturated state. Within initial 30 min of MH formation, the water saturation decreased significantly from 51.63% to 42.26%. After 203 min of MH formation, the residual water saturation decreased to 35.14%. After 331 min of MH formation, the residual water saturation decreased to 26.90%. After 2635 min of MH formation, the residual water saturation decreased to 15.70%. After that, it barely changed until the end of test. The results showed that the MH formation process was ununiform in time and space [2], while the same results could be seen in the process of MH reformation (Figure 2(b)).

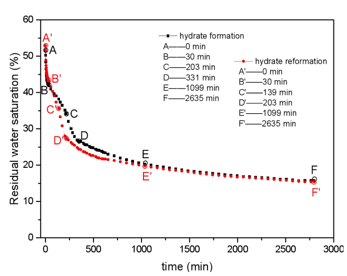


Figure 3. The residual water saturation during the whole MH formation/reformation process.

Figure 3 shows the residual water saturation during the whole MH formation/reformation process. The reduction rate of water saturation displayed as fast-slow-fast-slow rhythm, so the process could be divided into four sections.

Section A-B: 0-30min, 9.36% residual water was consumed; Section A'-B': 0-30min, 9.55% residual water was consumed.

Section B-C: 30-203 min, 7.12% residual water was consumed; Section B'-C': 30-139 min, 7.79% residual water was consumed.

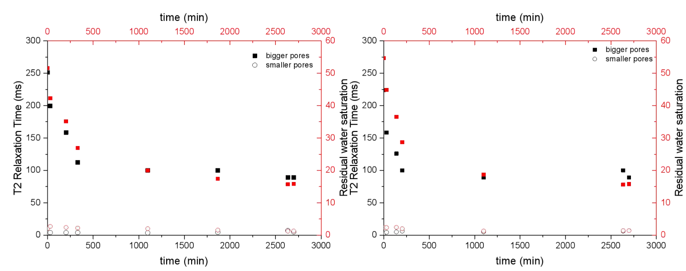
Section C-D: 203-331 min, 8.24% residual water was consumed; Section C'-D': 139-203 min, 7.57% residual water was consumed.

Section D-E: 331-1099 min, 6.93% residual water was consumed; Section D'-E': 203-1099 min, 8.56% residual water was consumed.

Section E-F: 1099-2635 min, 4.27% residual water was consumed; Section E'-F': 1099-2635 min, 3.94% residual water was consumed.

It could be seen that in the first three sections, the reduction rate of residual water saturation was faster and it took less time in the MH reformation than that in the MH formation. Except this, there was no significant difference of total time required between MH formation and reformation.

3.2 *Pore structure and fluids occupancy change*



(a) (b)

Figure 4. The variation of the T_2 peak and the residual water saturation in the pores in the MH formation (Figure 4(a)) and reformation (Figure 4(b)).

Figure 4 shows the variation of the T_2 peak and the residual water saturation in both the bigger pores and smaller pores. Water consumption and MH formation mainly occurred in the bigger pores. There were about two reasons: the first one was that the contact area of water was bigger than that in the smaller pores after draining some free water, which accelerated the MH formation rate. The other one reason was that the pore water saturation in the smaller pores was higher. The only gas resource to form MH in the case of high pore water saturation was the dissociated gas, which was not conducive to MH formation.

Figure 5 shows the fractal dimension and correlation coefficients at different time in the MH formation and reformation, which were obtained by linear regression analysis on the T_2 distribution curves. The responding fractal dimension increased with the

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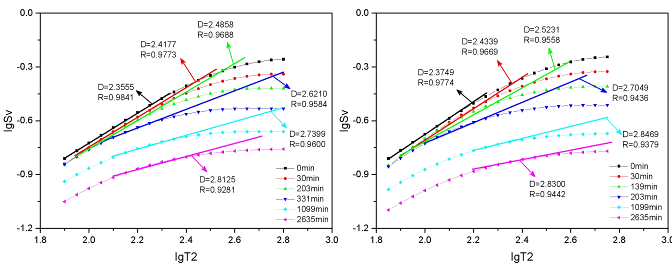
MH formed, which meant that the pore structure inside of porous media became more complicated [15]. During 0min to 2635min of MH formation, the fractal dimension increased as a logarithmic growth trend from 2.3555 to 2.8125, while the residual water saturation decreased from 51.63% to 15.70%. It meant that there was always MH formation in the process of water consumption to the end of MH formation; During 0min to 1099min of MH reformation, the fractal dimension increased as a logarithmic growth trend from 2.3749 to 2.8469, while the residual water saturation decreased from 52.94% to 19.47%. During 0min to 1099min, the fractal dimension basically unchanged, while the residual water saturation still decreased from 19.475 to 15.53%. It meant that there were water consumption and MH formation in the initial 1099min of MH formation, but there was mainly the fractal growth from 1099min to 2635min, which has less effect on the pore structure.



(a) (b)

Figure 5. The fractal dimension (D) and correlation coefficients (R) during MH formation (Figure 5(a)) and reformation (Figure 5(b)).

Besides, it could be seen that there was residual MH microcrystalline structure after MH decomposition. When the MH formation conditions were reached again, the MH dendrites first nucleated and grew in the place with residual MH microcrystalline structure, which made the rate of consuming water more rapidly than that in the initial MH formation process.

4. CONCLUSIONS

In this paper, MH formation and reformation process was studied in the partially water saturated sand pack, using low-field MRI technique. The pure phase encoding MRI technique, a bulk T_2 distribution were used to monitor the change of fluids content, Pore structure and fluids occupancy. The results are as followed: In the MH formation and reformation, the rate of water content reduction displayed as fast-slow-fast-slow rhythm, which had significant stages. And the reduction rate of residual water saturation was faster in the MH reformation than that in the MH formation, which was because that there was residual MH microcrystalline structure remained in the porous media after MH decomposition and they provided place for the MH dendrites nucleated and grew again; Besides, the T_2 distributions clearly reflected that MH preferred forming in the bigger pores to smaller pores due to the bigger contact area between water and gas, and lower of pore water saturation; The formed MH made the pore structure inside of porous media more complicated.

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