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OPINION
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The Impact of Accurate Prediction of Natural Gas Compressibility Factor on the CO_2 Replacement Method for Natural Gas Hydrate Exploitation

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INTRODUCTION

Carbon capture, utilization, and storage (CCUS) is a new development trend of carbon capture and storage (CCS), which is a technology that captures and purifies the carbon dioxide (CO_2) emitted during the production process and then puts CO_2 into a new process of production for recycling or storage. This technology is not only a key technology to reduce CO_2 emissions from fossil energy power generation and industrial processes but also a bottom-line technology to achieve carbon neutrality. CO_2 geological storage is a core component of CCUS technology, which determines the development potential and direction of CCUS technology.

The oil and gas industry has relative technical advantages in CO_2 sequestration. Injecting captured CO_2 into oil and gas reservoirs can not only enhance the recovery of oil and gas but also achieve long-term CO_2 storage. At the same time, CO_2 can be injected into depleted oil layers, shale layers, and unmineable coal beds, which are used to enhance crude oil recovery (CO_2-EOR), enhance shale gas recovery (CO_2-ESGR), and improve coalbed methane recovery (CO_2-ECBM), respectively, while storing CO_2 (Zhang et al., 2020). The deep saline aquifer in the sedimentary basin is also considered as the main storage place for CO_2 geological storage due to its huge storage potential.

The environmentally friendly technologies for gas hydrate extraction based on CCUS are constantly developing. The combined technology of natural gas hydrate (NGH) exploitation using the CO_2 replacement method and ocean storage is also considered as potential long-term CO_2 storage solution due to its promising prospects for capturing, storing, and utilizing CO_2 (Tewari et al., 2021).

The natural gas compressibility factor is one of the most important physical parameters of natural gas, which characterizes the ratio of the same amount of real gas to the ideal gas volume under certain temperature and pressure conditions. This study focuses on the combined technology of NGH extraction with CO_2 replacement and ocean storage and points out that the impact of the prediction accuracy of the gas compressibility factor on the exploitation of NGH by the replacement with CO_2.

THE COMBINED TECHNOLOGY OF NGH EXTRACTION WITH CO_2 REPLACEMENT AND OCEAN STORAGE

Fundamental

The combined technology of the CO_2 replacement method for NGH exploitation and ocean storage is a new coupling technology type which is safe and environmentally friendly. Its basic principle is to capture industrial waste CO_2 to mine NGH and achieve CO_2 sequestration under the premise of ensuring the stability of the seabed reservoir. This technology has improved the utilization of CO_2 resource greatly and maximized the storage of CO_2 by taking advantage of the huge potential of ocean storage space, which is a beneficial supplement to CCUS.

Ebinuma and Ohgaki et al. first proposed the idea of NGH exploitation using the CO_2 replacement method (Ebinuma, 1993; Ohgaki et al., 1996). After long-term research and demonstration, the idea has been proven to be highly feasible in terms of kinetics and thermodynamics. The replacement reaction formula is shown below:

$$CH_4 \cdot nH_2O + CO_2(g) \rightleftharpoons CH_4(g) + CO_2 \cdot nH_2O\,(n \geq 5.75) \quad (1)$$

In the process of NGH exploitation using the CO_2 replacement method, the generation of CO_2 hydrate is an exothermic process, and the decomposition is an endothermic process. It can be seen from equations (Tewari et al., 2021) and (Ebinuma, 1993) that the exotherm is greater than the endothermic, so this process can proceed spontaneously (Yezdimer et al., 2002). The equations are given as follows:

$$nH_2O + CO_2(g) \rightarrow CO_2 \cdot nH_2O\,(\Delta H_f = -57.98\,kJ/mol) \quad (2)$$

$$CH_4 \cdot nH_2O \rightarrow CH_4 + nH_2O\,(\Delta H_f = 54.49\,kJ/mol) \quad (3)$$

The combined technology of CO_2 replacement mining NGH and ocean storage has unique development advantages. It can ensure the stability of the geological structure during the mining process, while storing the injected CO_2 on the seabed for a long time, which expands the application scope of CCUS technology to a certain extent. At present, the exploitation technology of NGH is still in the stage of trial production at home and abroad, and there are still many key technologies that need to be broken through. With the continuous innovation and development of related technologies, this technology will definitely become the main force of NGH extraction technology in the future.

Calculation of Decomposition Heat of NGH

When studying the phase equilibrium problem of the NGH system developed using the CO_2 replacement method, it is not only difficult to measure the heat of hydrate decomposition directly but also impossible to measure the heat of hydrate decomposition under all conditions directly. The main reason is that some gas hydrates have high formation pressure and the purity of the hydrates formed is difficult to determine. It is a simple and feasible method to use the Clausius–Clapeyron equation to calculate the heat of decomposition of hydrate. The decomposition heat of natural gas hydrate can be obtained by using the Clausius–Clapeyron equation based on the NGH phase equilibrium pressure, phase equilibrium temperature, gas constant, and gas compressibility factor (Sloan and Fleyfel, 1992). The specific formula is as follows:

$$\frac{d(\ln P)}{d(1/T)} = \frac{-\Delta H}{ZR} \quad (4)$$

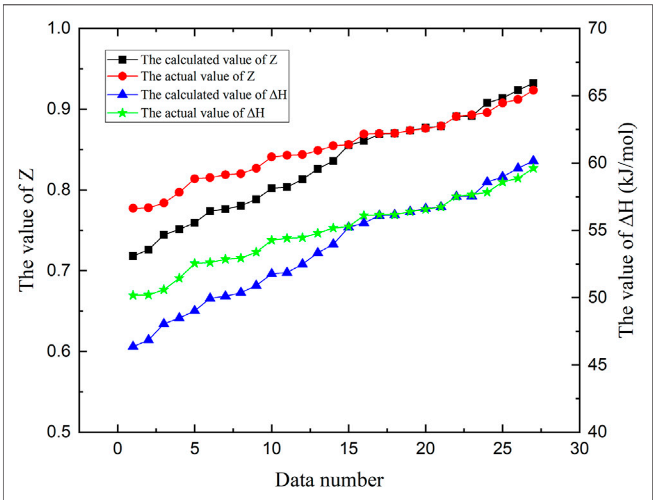

FIGURE 1 | Comparison of compressibility factor (Z) and decomposition heat (ΔH) (The data were obtained from reference (Li, 2018)).

where: P—phase equilibrium pressure, MPa;

T—phase equilibrium temperature, K;

Z—gas compressibility factor; R—gas constant, 8.314 J/(molK).

As shown in equation (Ohgaki et al., 1996), it is necessary to use the gas equation of state to calculate the gas compressibility factor so as to obtain the decomposition heat of NGH under different phase equilibrium conditions. The slope of the logarithm of the NGH phase equilibrium pressure and the reciprocal curve of the phase equilibrium temperature is 10^4–10^5 orders of magnitude. Therefore, a small deviation in the calculation result of the compressibility will cause a larger calculation error of the decomposition heat; in other words, the prediction accuracy of the natural gas compressibility factor determines the calculation accuracy of NGH decomposition heat (Chen et al., 2019). Therefore, it is necessary to compare and analyze the calculation results of the compressibility factor of different equations of state and find the equation of state that calculates the gas compressibility factor accurately, so as to improve the calculation accuracy of the Clausius–Clapeyron equation to calculate the hydrate decomposition heat.

ANALYSIS AND DISCUSSION

When using the CO_2 displacement method to exploit NGH, the concentration of CO_2 in the formation fluid will affect the calculation accuracy of the gas compressibility factor. The study displays that when the CO_2 concentration is higher than 50%, the deviation between the compressibility factor calculated using the cubic equation of state and the actual value can be as high as 5–40% (Li, 2018). By comparing the experimental test value of the compressibility factor with its calculated result, and using the Clausius–Clapeyron equation to compare the decomposition heat corresponding to the compressibility factor, the comparison result is shown in **Figure 1**, which means that the small deviation of the compressibility factor calculation result will bring a larger calculation error of decomposition heat.

In summary, the accurate prediction of the natural gas compressibility factor is of great significance for the combined technology of NGH exploitation using the CO_2 displacement method and ocean storage. Therefore, it is particularly critical to determine this parameter quickly and accurately.

The various methods for calculating the compressibility factor proposed by domestic and foreign scholars can be summarized into four categories, which are experimental measurement method, chart method, equation of state method, and empirical formula method, respectively. The experimental measurement method has accurate calculation results, but the experimental cost is high, the cycle is long, and it is difficult to meet the needs of a large number of engineering calculations. The chart method is simple and easy to implement and can meet the engineering needs of a large number of calculations, but the error is large and the accuracy is not high. Although the equation of state method can accurately describe the gas compressibility factor, it is difficult to use for mixtures containing a large number of components due to the complexity of the parameters involved. This difficulty has prompted scholars to seek simpler and more reliable methods to obtain more accurate predictions of gas compressibility factors. The HY method, DPR method, and DAK method in the empirical formula method are the classic methods of applying the equation of state to calculate the natural gas compressibility factor. They can all represent the Standing–Katz chart relatively accurately within a certain range. In particular, the DAK method has the highest calculation accuracy, but its error at high temperature and high pressure is also slightly larger, and there is room for improvement.

Most methods for calculating the compressibility factor are applicable to conventional natural gas reservoirs. If the CO_2 concentration in the gas reservoir is high, it will affect the critical parameters of natural gas and cause the change of the compressibility factor. Hence, the critical parameters must be corrected when calculating the natural gas compression factor to obtain more accurate data. The commonly used correction methods are W-A (Chen, 1989; Guofa et al., 2011) and GXQ (Guo et al., 2000).

The artificial intelligence technology has been applied to the evaluation of the gas compressibility factor, which has shown excellent performance in terms of accuracy (Mohamadi-Baghmolaei et al., 2015; Attia et al., 2016; Irene et al., 2016; Adel et al., 2018; Tariq et al., 2019). In future research work, we should make full use of new technologies such as artificial intelligence to establish a calculation model for accurately predicting the gas compression factor, further improving the calculation accuracy of the natural gas compression factor and providing more accurate basic data for the combined technology of NGH exploitation using the CO_2 displacement method and marine storage, thereby accelerating the promotion of CCUS technology to achieve the goal of carbon neutrality.

CONCLUSION

1) As a core component of CCUS technology, CO_2 geological storage mainly includes storage in hydrocarbon reservoirs such as depleted oil layers, shale layers, and unmineable coal bed stages and storage of deep saline aquifers. The environmentally friendly technologies for gas hydrate extraction based on CCUS are constantly developing. The combined technology of NGH extraction with CO_2 replacement and ocean storage have enhanced the recovery of oil and gas at the same time by realizing geological sequestration of CO_2 and achieved the goal of "integration of sequestration and utilization, emission reduction and benefit win-win."
2) When studying the phase equilibrium problem of the NGH system developed using the CO_2 replacement method, according to the Clausius–Clapeyron equation, it was found that a small deviation in the calculation result of the compressibility factor will cause a larger calculation error of the decomposition heat. Therefore, the accurate prediction of the natural gas compressibility factor is of great significance to the combined technology of NGH extraction with CO_2 replacement and ocean storage. In order to improve the accuracy of the calculation results, it is necessary to compare and analyze the calculation results of different equations of state to find an accurate calculation of the gas compressibility factor, thereby improving the calculation accuracy of the hydrate decomposition heat.
3) In the future research work, we should make full use of new technologies such as artificial intelligence to establish a calculation model to predict the gas compressibility factor accurately, further improving its calculation accuracy, to provide more accurate basic data for the combined technology of NGH extraction with CO_2 replacement and ocean storage, so as to accelerate the development of CCUS technology and help achieve carbon neutrality.

AUTHOR CONTRIBUTIONS

YN: mainly responsible for the construction of thesis ideas, literature research, and manuscript writing. GJ: mainly responsible for framework adjustment and thesis guidance. YL: mainly responsible for literature research.

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Conflict of Interest: Author YL was employed by the company Sinopec Zhongyuan Petroleum Engineering Design Co., Ltd.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.