

# Research Progress Of Natural Gas Hydrate Development By Thermal Injection

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**Abstract**—In the process of hydrate decomposition by thermal injection, the variation of parameter of heating and hydrate sediment can have a great influence on the outcome of decomposition. One of the most remarkable questions is the energy efficiency ratio during the process of decomposition. Thus, a deep insight on how these parameters influence the energy efficiency ratio is very significant. This paper discussed the mechanism of nature gas hydrate decomposition by thermal injection. Summarized experiments and the corresponding conclusions of nature gas hydrate decomposition by heat injection in recent years at home and abroad, discussed and compared the numerical model of nature gas hydrate decomposition by thermal injection. At the end of the paper give some suggestions on the nature gas hydrate exploitation by heat injection in the future.

**Keywords**—gas hydrate; mechanism; thermal recovery; experiment; numerical simulation; suggestion

## I. INTRODUCTION

Natural Gas Hydrate is composed of relatively small-sized guest molecules such as CH<sub>4</sub> molecules or CO<sub>2</sub> molecules and water under ideal pressure and temperature conditions (usually temperatures below 300K and pressures above 0.6Mpa [1]) The ice-like crystalline compound having a cage structure formed under contact is also called methane hydrate because the gas is mostly methane (80% to 90%). Natural gas hydrates are widely distributed in nature in the permafrost of the continent, the slopes of the islands, the uplifts of activities and passive continental margins, the polar continental shelf, and the deep water environment of the oceans and some inland lakes. It is estimated that the carbon content of natural gas hydrates in the world is equal to twice the carbon content of fossil energy sources such as petroleum and coal. Compared with conventional energy sources, natural gas hydrates have shallow burial and large scale (the global reserves are sufficient for human use for 1000 years). High energy density (the standard volume of hydrate can decompose up to 164 units of methane gas under standard conditions), burning without pollution (mainly methane gas, burning almost

no environmental pollutants), etc. Alternative energy sources for oil and gas.

In recent years, the research of hydrates has been paid more and more attention by scholars all over the world. Coupled with the continuous development of science and technology, the methods of hydrate mining are more and more diversified. The commonly used mining methods include depressurization method, heating method, replacement method and inhibitor method. And the combined use of these methods. The thermal excitation mining method directly heats the natural gas hydrate layer to make the temperature of the natural gas hydrate layer exceed its equilibrium temperature, thereby promoting the decomposition of natural gas hydrate into water and natural gas exploitation methods. This method has undergone the development process of directly injecting hot fluid heating, fire flooding, downhole electromagnetic heating and microwave heating into the natural gas hydrate layer. The thermal excitation mining method can realize the cyclic heat injection and the action mode is faster. The continuous improvement of heating methods has promoted the development of thermal excitation mining methods. However, this method has not solved the problem of low heat utilization efficiency so far, and only local heating can be performed, so the method needs to be further improved. At present, the form of heating is more and more diversified. At present, the main heating methods proposed by scholars are injection of hot water or salt water of higher temperature, utilization of lower geothermal heat, electromagnetic heating, microwave heating, steam stimulation and the like.

## II. MECHANISM OF GAS HYDRATE HEATING MINING

Because hydrates are widely distributed in rock fractures, pores, and rock masses and seafloor deposits in a crystalline solid state, they are not permeable and fluid. And the hydrate is a metastable state formed under high pressure and low temperature conditions. Once the temperature or pressure changes, the hydrate may be decomposed. Therefore, the general idea of mining hydrate is to use the prior art to promote the decomposition of the hydrate in situ, and then The natural gas is extracted by the traditional natural gas mining technology. The heating method is to promote the decomposition of the hydrate by increasing the temperature of the hydrate deposit layer.

In 2005, Makogan et al. [4] plotted the pressure drop required for the decomposition of hydrates in some known hydrate-deposited layers into a function image related to the temperature of the deposited layer, by which the hydrate can be seen at a certain temperature and it can be stably existed under pressure. The higher the temperature, the greater the pressure drop required for the stable hydrate. This indicates that if the pressure is higher or the pressure is lower, the hydrate may be stable in the crystalline solid state. Existence, this inevitably leads to the existence of a critical line of temperature and pressure, which divides the hydrate into two regions—the hydrate-stable region and the hydrate-unstable region. It is a region where the hydrate is stably present, and the region on the upper right side of the critical line is a region where the hydrate is unstable. This is also the reason for the widespread stable presence of hydrates and terrestrial permafrost and marine sediments. The heating method is to change the temperature conditions of the hydrate storage layer beyond the critical decomposition temperature of the hydrate at this pressure to promote hydration. The phase equilibrium is destroyed and decomposed into water and natural gas.

Contrary to the hydrate formation reaction, the hydrate decomposition reaction is an endothermic reaction. During the decomposition process, it is necessary to continuously absorb a large amount of heat into the surrounding environment to destroy the crystal cage of the hydrate, release the natural gas molecules enclosed therein, and heat-exploit natural gas hydrate is the condition that keeps the pressure of the hydrate deposit layer constant, and the hydrate is continuously decomposed by continuously injecting the heat required for the decomposition of the hydrate into the sediment layer. The heat is mainly transferred through the sediment layer during the injection process. The way forward, the hydrate is decomposed when the temperature reaches the critical temperature required for the decomposition of the hydrate, and the interface is continuously moving forward, so that the hydrate is thermally decomposed in the sediment layer, while maintaining the constant pressure of the sediment layer. The process can be seen as a moving boundary ablation problem. Considering the hydrated deposit as a semi-infinite cylinder, the heat-decomposing hydrate can divide the region into two parts—hydrate decomposition and hydration.

### III. EXPERIMENTAL RESEARCH ON HEAT INJECTION OF NATURAL GAS HYDRATE

In recent years, the importance of natural gas hydrates as a future energy source has been recognized by more and more scholars. The research on natural gas hydrates has been deepened in various countries, and the continuous development of science and technology has gradually improved the hydrate experimental system and the degree of visualization of equipment. And the accuracy of testing is getting higher and higher, and the methods for detecting

hydrate formation and decomposition are becoming more and more diverse. In recent years, research on hydrated pyrolysis mining experiments has focused on heat injection processes (including hot water temperature, speed, hot brine salinity, etc.) and hydrate-related parameters (hydrate initial saturation, permeability, etc.). The effect of hydrate decomposition gas production and energy efficiency of heat injection mining. Through experimental research, it laid the foundation for future economic and effective mining of hydrates.

In 2007, Hao Yongzheng et al [6] conducted an experimental study on natural gas hydrates, and injected 100 ° C, 130 ° C, 160 ° C, 190 into the prepared hydrates at a constant rate while maintaining other conditions. The hot water of ° C records the gas production and water production curves. Through analysis and comparison, the optimal hot water temperature under the experimental conditions is obtained, and the gas production law and water production law obtained by the change of the hydrate decomposition process will be. The hydrate decomposition gas production process is divided into three stages, namely, the free gas production stage, the hydrate decomposition gas production stage, and the pressure reduction mining gas production stage. The experiment illustrates the feasibility of heat injection of natural gas hydrate by calculating the heat transfer efficiency (heat exchange efficiency is about 1.0). At the same time, by optimizing the parameters of temperature and speed of hot water injection, the benefit of hot water injection is improved. The feasibility and economics of hydrate injection heating provide experimental basis.

In 2010, Yang et al [7] used a three-dimensional medium-sized reaction vessel to carry out hot water circulation injection to promote the decomposition of gas production by hydrate. The experimental results show that the overall temperature change trend is accompanied by the increase of hot water injection. As the gas production decreases, the temperature distribution and fluctuations in the vessel are related to the porosity, permeability, etc. of the hydrate sample, in addition to the location of the production or injection well. They introduce the energy efficiency ratio and the heat utilization rate. An important parameter (Equation 1 and Equation 2) studies some factors affecting energy efficiency, including hydrate saturation, initial temperature, hot water temperature, hot water volume, and well pressure. It is found that energy efficiency is the same when other conditions are the same. The saturation of the hydrate increases as the initial temperature of the hydrate increases, and decreases as the heat injection temperature and the well pressure increase. It is also pointed out that the method of circulating hot water is beneficial to the exploitation of hydrates with high hydrate saturation and low permeability, because the gas and water generated by hydrate decomposition are difficult to penetrate by other methods. This experiment comprehensively evaluates the effects of heat injection process and

hydrate reservoir parameters on temperature distribution, gas production law and energy efficiency, and explains the causes of relevant experimental phenomena, which is beneficial to formulate corresponding parameters according to different parameters of hydrate reservoirs.

In 2013, S. Li et al. [8] designed a new one-dimensional experimental system for the exploitation of natural gas hydrates. The injection parameters affecting the gas production rate and energy efficiency were analyzed. They believed that the higher the temperature of the injection, the faster the injection speed, the faster the rate of gas generation. The parameters of the sensitivity to energy efficiency are, in order, the temperature of the hot brine, the rate of hot brine injection, and the injection time. In 2014, Li Shuxia et al [9] conducted a pilot study on the sensitive factors of natural gas hydrate reservoir hot water mining. They used different factors and thermal parameters to test the hydrate saturation by using five-factor four-level orthogonal design. The effects of five parameters, initial temperature, hot water injection temperature, hot water injection time and hot water injection rate on energy efficiency, and the following conclusions: the factors affecting the energy efficiency of hydrate storage hot water mining from large to small In turn, hot water injection temperature, hydrate saturation, initial temperature, hot water injection time and hot water injection speed. At the same time, the optimal combination of hydrate injection mining is obtained. In this experiment, different geological factors and heat injection parameters are obtained for the sensitivity of the gas-fired natural gas hydrate and the optimal mining combination. The conclusions obtained are easy to highlight the main factors in the process of heat injection mining, and the most important factors are ignored. The superior combination economically and efficiently extracts natural gas hydrates.

In 1987, Kamath et al. conducted experiments on the evaluation of gas decomposition by hydrated brine. The experimental study found that gas hydrated by gas injection has higher gas production rate and energy efficiency than steam injection. Jeonghwan L [2010] 10] Experiments were carried out on the effect of hot brine injection on hydrate decomposition behavior and gas production. The experiment mainly studied the effects of different concentrations of brine and permeability on gas production. They found that too high salinity would reduce gas production. Because the excessive salinity hinders the flow of the liquid and reduces the permeability, the greater the permeability of the porous medium, the larger the gas production, and the smaller the gas production rate. In 2015, Li Shuxia [11] conducted the hot water salinity The impact of hydrate mining is to first generate natural gas hydrate reservoirs with the same initial conditions in a self-made one-dimensional gas hydrate mining simulation experimental device, and then inject hot water with a salinity of 2%, 10%, and 20% for mining experiments. . The results show that when the

salinity of the hot water is large (after reaching 10%), the hydrate decomposes rapidly and is produced simultaneously with the free gas, and the peak gas production rate is large; while the salinity of the hot water is small (2%) When the free gas is first produced, then the hydrate decomposes the gas to produce. The higher the salinity of the hot water, the faster the decomposition rate of the hydrate, the greater the rate of movement of the thermal front, and the higher the energy efficiency of the large decomposition stage of the hydrate. Therefore, the increase of hot water salinity can accelerate the exploitation rate of natural gas hydrate and increase the economic feasibility of heat injection.

In 2005, Komai, T et al. [12] observed the formation and decomposition of hydrates by microscope and Raman spectroscopy. Through analysis of Raman spectroscopy, they found that hydrates were first formed at the interface between water and sedimentary layers. It is the interface between gas and liquid, and then gradually develops toward the center of the pore. On the contrary, the decomposition of hydrate starts at the interface between the hydrate and the sediment layer, and then develops toward the center of the pore, which indicates the decomposition of hydrate and the mass transfer phenomenon of heat and mass. There is a big connection between them.

In 2013, Garrett C. Fitzgerald et al. [13] designed hydrate formation and decomposition in containers with water saturation of 21%, 41%, and 60%, respectively, in order to obtain the characteristics of hydrate formation and decomposition under different water saturations. In the experiment, the hydrate saturations were 20%, 35%, and 50%, respectively. It was observed that when the pressure was increased, the container hydrate containing the lower initial water saturation was generated at a faster rate, including the initial water. The peak of hydrate formation with the lowest saturation is the largest, and when the hydrate saturation is low, the heating rate is higher, and the efficiency peak and accumulation efficiency are lower. This experiment is of great significance for the future adoption of different heat injection processes for different hydrated sediment layers with different saturations.

In 2014, Prathyusha Mekala et al. [14] compared the formation and decomposition kinetics of methane hydrate in seawater and silica sand by experimental methods. Under the same conditions, they were synthesized using pure water and 3.03wt% seawater brine respectively. Hydrate, by measuring the amount of methane gas consumed to react to produce hydrates, they found that 72% of water was converted to hydrates when pure water was used, while only 11.6% of seawater was converted to hydrates under the same conditions. They obtained salt as a thermodynamic inhibitor, making the amount of seawater salt converted to hydrate nearly one-tenth that of pure water, while also reducing the rate of synthesis of hydrates. In the hydrate formation process, the temperature at different points in the

container is measured, and the intermittent temperature peak is found. This is because hydrates are formed in these places, and hydrate formation is an exothermic reaction. A similar phenomenon is in the literature [15–18] also got it. After the hydrate production process, hot water is injected to promote the decomposition of the hydrate. When the hydrate is decomposed, it is found that the decomposition rate of the hydrate synthesized in pure water is significantly higher than that of the sea salt.

In 2014, Garrett C. Fitzgerald [19] designed four sets of experiments to investigate the effects of hydrate saturation and heat injection rate on gas production efficiency and thermal frontal movement. The layer has high energy efficiency when injected at high temperature or low temperature. The effect of heat injection rate on energy efficiency is less than that of hydrate saturation. However, the faster the heat injection rate, the higher the energy efficiency, and the study of the movement of the thermal front is found. Increasing the hydrate saturation of the sediment will slow the movement of the hot front (similar to the literature 20). This is because hydrate decomposition is an endothermic reaction, and the greater the hydrate saturation, the larger the volume of hydrate per unit volume. The larger the heat, the more heat is used to break down the hydrate, and they believe that more research is needed to further understand the energy efficiency and thermal frontal movement laws.

In 2016, Shuxia Li [21] analyzed the energy efficiency of the hydrated hydrate by experiments. Like the literature [8], they first defined the energy efficiency, and then analyzed the geological parameters (the hydrate saturation) experimentally. , permeability, initial temperature of the sedimentation layer, and the effect of heat injection parameters (hot brine temperature, hot brine injection rate, concentration, and hot brine injection) on energy efficiency, compared with previous scholars, they pointed out more refinement The energy efficiency and energy efficiency of different hydrate parameters vary with the parameter.

#### IV. PROGRESS IN NUMERICAL SIMULATION OF THERMAL INJECTION MINING

In 1994, Islam et al. [24] established a two-phase thermal model. The hydrate pyrolysis decomposition is divided into undecomposed zone and decomposition zone, and the undecomposed zone, decomposition zone and moving boundary equation are established respectively. It consists of a continuity equation, a Darcy equation and an energy conservation equation. The model considers the effects of heat conduction and gas-liquid two-phase seepage. The model can simulate the production dynamics of hydrates in vertical and horizontal wells and analyze the relationship between instantaneous and cumulative gas production over time. The model can also discuss the relationship between instantaneous and cumulative gas production in the case of continuous heating,

intermittent heating, preheating, and low temperature heating of the wellbore.

In 2008, Liu et al. [25] used a one-dimensional mathematical model to simulate the depressurization and pyrolysis decomposition of a hydrated deposit in a one-dimensional radial seepage system, ie, an axisymmetric reactor. He believes that when the reactor is large enough, depressurization is considered a good method of mining because of the hydration production of a single well while maintaining a constant boundary temperature for the boundary of the hydrate deposit. The impact is limited because of the deterministic effect of convective heat transfer that allows little heat to pass into the well. For a multi-well system, it has a hot well and a gas producing well, making the injection method an effective way to mine large hydrate reservoirs. And for a poorly permeable sediment layer, the heat injection provides sufficient additional heat for the hydrate decomposition, so it is a good mining method.

In 2012, Xiao-Sen Li et al. [27] conducted a numerical simulation study on the potential of Qilianshan hydrate for single gas well steam stimulation. They obtained the distribution of temperature and pressure in the sedimentary space. Drawing the graphs under different parameters, it is found that the proper heat injection parameters and geological parameters can obtain the ideal energy efficiency during the steam stimulation process. However, the gas production rate is low throughout the mining process, and the sensitivity is sensitive to the parameters. It is found that the characteristics of gas production are related to the permeability, porosity, velocity of steam injection, temperature, and saturation of bound water. The relative permeability index is related to gas production during steam stimulation. Limited impact. It can also be found from this numerical simulation that gas hydrates have higher gas production efficiency than pure methane hydrates. Compared with the previous numerical simulations, the simulation directly obtained the evaluation of the gas production process from different geological conditions.

In 2014, Wang Lina [28] established a three-dimensional three-phase five-component hydrate injection thermal mining mathematical model, which includes the mass conservation equations of each component, the energy conservation equation and the decomposition kinetic equation of natural gas hydrate. The energy conservation equation considers heat conduction, heat convection, hydrate decomposition heat, heat carried by the injected production fluid, and heat exchange between the hydrate layer and the underlying layer. The mass conservation equation considers not only the Darcy flow but also the gas diffusion. Multiphase seepage in hydrate reservoirs takes into account the effects of temperature, composition, phase equilibrium and other factors.

In 2014, Liu Di [29] analyzed the heat transfer of natural gas hydrate decomposition process in porous media by numerical simulation method, studied the heat conduction of porous media, separated the parameters for controlling heat transfer, and classified the heat conduction in porous media of hydrate. Heat transfer between the porous media layer of the hydrate and the upper and lower cap layers, convective heat transfer in the hydrated porous medium, and sensible heat of the porous media layer of the hydrate to the decomposition process of methane hydrate in the porous medium. The system analysis was carried out. Through the gasification of natural gas hydrate gas production and pressure, temperature and hydrate saturation distribution at different decomposition moments, the gas pressure hydrate depressurization, heat injection and combined mining heat transfer control in porous media were revealed. mechanism.

In 2015, ZHAO [30] and so on, in order to understand the effect of heat transfer on the thermal exploitation of methane hydrate, a two-dimensional axisymmetric model was established, which consists of three components (gas, water, hydrate) and three phases (gaseous state, liquid, solid), including the mass conservation equation, energy conservation equation and reaction kinetic equation of hydrate pyrolysis decomposition, the model considers the effects of heat conduction, thermal convection and gas-liquid two-phase percolation, and the results show that in the hydrate injection. In the process of thermal decomposition, increasing the specific heat capacity of the porous medium containing hydrate will reduce the gas production rate; the initial saturation of water and the convective heat transfer of water and methane condensate gas hydrate decomposition gas production rate and gas production have little effect, increase the porous medium. The thermal conductivity will inhibit the decomposition of hydrates at the initial stage, but will promote the decomposition of hydrates over time.

In 2015, Wang et al. [31] established an analytical model for heat and mass transfer in natural gas hydrate pyrolysis. The pyrolysis decomposition was divided into two processes. The first process was a gas release process, and the second was a note. The process of heat-induced decomposition of hydrates establishes two stages of heat and mass transfer equations. They find that the decomposition rate of hydrates increases with the increase of injection temperature in the low temperature section, while the change is not obvious in the high temperature section. The results of the analytical model were compared with the experimental results. It was found that the results obtained by the analysis were basically consistent with the experimental results.

## V. SUMMARY AND OUTLOOK

At present, the core problem of hydrate mining is how to improve the energy efficiency ratio and how to avoid a series of environmental problems caused by hydrate mining. This requires a deeper understanding of the mechanism of hydrate formation and

decomposition, thereby improving the economics of hydrate mining and Safety, in the current mining methods, the injection method, the pressure reduction method and the joint mining of the two methods are considered to be the most promising mining methods at present, and the technical conditions of the heat injection mining are more complicated and need to be solved. There are more problems. From the current basic experiments and numerical simulation results of hydrated pyrolysis, the following conclusions can be drawn:

(1) Natural gas hydrate is a metastable crystalline material formed under certain temperature and pressure conditions. Once the equilibrium condition destroys the hydrate, it will decompose to produce a flowable gas and water. The heating method is used to maintain the hydrate deposition. Under the condition that the layer pressure does not change much, the equilibrium condition of the hydrate is broken by increasing the temperature of the reservoir, and the hydrate is decomposed to generate gas. The gas is then recovered using conventional natural gas recovery techniques.

(2) The heat injection process (heat injection method, speed, hot water salinity, etc.) and the relevant parameters of the hydrate storage (permeability, initial water saturation, etc.) have a greater impact on the hydrate decomposition process, among which One of them is that it has a greater impact on the energy efficiency ratio of mining. It is concluded that understanding how these parameters affect the energy efficiency ratio of hydrates helps to obtain the parameters of economical and efficient mining of hydrates, and selects the most suitable injection for improving energy efficiency ratio. Thermal process.

(3) The current numerical simulations of hydrate mining are established under a series of hypothetical ideal conditions, such as not considering the secondary formation of hydrates, but the mechanism of hydrate formation and decomposition is known to be involved in the actual mining process. To the secondary formation of hydrate, how to establish a mathematical model under the premise of considering the secondary formation of hydrate is also a direction of future research.

(4) The pyrolysis decomposition of natural gas hydrate will cause the volume of sediment to expand, affecting the mechanical properties of marine sediments, and the excess pore water pressure will reduce the strength of the sediment and affect the mining factor such as permeability. If it is not handled properly, it will occur. The destruction of the gas hydrate layer in the area will even lead to catastrophic events [40]. However, the research on these practical problems is very limited. How to use the experimental and mathematical simulation methods to verify it is of great significance for the actual mining in the future.

In summary, although many hydrate decomposition problems and mining technology difficulties have been

broken, but not very systematic. Compared with pressure-reducing mining, thermal mining technology is more complicated and there are more problems to be solved. Recently, high-quality hydrates have also been discovered in the Shenhu area of the South China Sea, which may accelerate the pace of commercial exploitation of natural gas hydrates in China in the next few years, and this must further accelerate the research on hydrate accumulation mining technology. The study of thermal mining is more practical and strategic, and can be used as one of the options for actual mining in China in the future. "Gas hydrates will likely change the current geopolitical model, and countries such as the United States, Japan, and India may achieve energy self-sufficiency. This incident has a strong impact on international affairs and foreign policy... The existing world energy trade will be completely changed" (quoted from US experts Michael), so accelerating gas hydrate research is of great significance for China's energy strategy.

#### VI. PROGRESS IN NUMERICAL SIMULATION OF THERMAL INJECTION MINING

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