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Sediment permeability change on natural gas hydrate dissociation induced by depressurization

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\textbf{A B S T R A C T}

The permeability of a natural gas hydrate reservoir is a critical parameter associated with gas hydrate production. Upon producing gas from a hydrate reservoir via depressurization, the permeability of sediments changes in two ways with hydrate dissociation, increasing with more pore space released from hydrate and decreasing due to pore compression by stronger effective stress related to depressurization. In order to study the evolution of sediment permeability during the production process with the depressurization method, an improved pore network model (PNM) method is developed to establish the permeability change model. In this model, permeability change induced by hydrate dissociation is investigated under hydrate occurrence morphology of pore filling and grain coating. The results obtained show that hydrate occurrence in sediment pore is with significant influence on permeability change. Within a reasonable degree of pore compression in field trial, the effect of pore space release on the reservoir permeability is greater than that of pore compression. The permeability of hydrate containing sediments keeps increasing in the course of gas production, no matter with what hydrate occurrence in sediment pore.

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\section{1. Introduction}

Natural gas hydrate is a group of clathrate compounds formed when small gas molecules are encapsulated in the cages constructed with water molecules via hydrogen bond (Sloan ED and Koh CA, 2007). Studies on hydrates have been ever increasing for the last few decades because of its potential as a promising energy resource. As the result, the main energy-consuming countries do much effort on the exploration and technology development for the exploitation of natural gas hydrates (Konno Y et al., 2015; Li JF et al., 2018; Matsushima J, 2006; Uchida T and Tsuji T, 2004). Since 2002, field trials of gas hydrate production have been conducted in Mallik (Dallimore SR and Collett TS, 2005), the northern slope of Alaska (Hunter RB et al., 2011), the Eastern Nankai Trough of Japan (Yamamoto K et al., 2014), and the South China Sea (Li JF et al., 2018; Wu NY et al., 2018; Ye JL et al., 2018; Zhang RW et al., 2018; Xu CL et al., 2018; Shi YH et al., 2019; Fang YX et al., 2019; Liang JQ et al., 2020). According to the production results, the depressurization method enables better production performance than other methods as thermal stimulation and carbon dioxide replacement (Yamamoto K et al., 2014). The physical properties of hydrate deposits, including its permeability, porosity, and saturation, are critical parameters to be considered for establishing appropriate production strategies (Waite WF et al., 2009), among which the permeability is even more crucial as it determines the fluid flowing behavior during production (Daigle H and Johnson A, 2016; Konno Y et al., 2015).

The permeability of hydrate-bearing sediments changes with hydrate formation and dissociation. When hydrate grows in porous sediments, the pore space is occupied, and hence the permeability decreases. On the other hand, hydrate permeability increases with hydrate dissociation. In situ measurement of the permeability during gas hydrate production is hardly accessible, so experimental methods are developed under various conditions with natural or artificial sediments, aiming at specifying the relationship between
permeability and hydrate occurrence (Kleinberg RL et al., 2003; Konno Y et al., 2015; Kumar A et al., 2010; Li CH et al., 2014; Liang H et al., 2010; Minagawa H et al., 2005; Seol Y and Kneafsey TJ, 2011). Theoretical efforts are also taken, several empirical models proposed to understand this relationship (Dai S and Seol Y, 2014; Daigle H, 2016; Delli ML and Grozic JL, 2013; Kleinberg RL et al., 2003; Masuda Y, 1997). Since the permeability of sediments decreases with hydrates formation, these empirical models are termed as “permeability reduction models” (Katagiri J et al., 2017). Because the experimental investigation of the permeability change in hydrate containing sediment is very challenging, numerical simulation can be an effective way.

The depressurization method has been proved to be a promising method producing gas from hydrate and has been successfully applied in the production trial conducted in the South China Sea (Li JF et al., 2018). During this trial, the reservoir pressure reduction was achieved by extracting the formation fluid from pore space, so this strategy is also termed as the fluid extraction method. This method can be described in brief, with formation fluid extracting the pressure and temperature condition of sediments deviates from the thermodynamic stability zone of hydrate, inducing hydrate dissociation into the water and hydrocarbon gas. At the same time, the reduction in pore pressure may raise the compression stress exerted on the porous sediments. Normally consolidated sandstone with large elastic modulus can withstand high compression stress. For example, the porosity of Bentheim sandstone only decreases from 21.92% to 21.76% when the confining pressure rises from 0 MPa to 20 MPa (Saenger EH et al., 2016). However, most of the natural gas hydrate systems located within 500 m below the seafloor and the sediments are unconsolidated (Liu F et al., 2013). The hydrate reservoirs in the South China Sea are mainly silty sediments with the low elastic modulus (Zhang H et al., 2016). As a result, the sediments will be compacted by the overlying layer and underlain layer with higher confining pressure. Therefore, the pore space is squeezed, and hence the permeability of sediment decreases. The risk of landslide and subsidence may also increase in the production process.

According to the triaxial compression experiments, the axial strain in the hydrate-bearing sediments of the Shenhu area can achieve 2% ~4% within the variation range of confining pressure from 0.5 MPa to 5 MPa (Zhang HW et al., 2017). There are many studies on the mechanical properties of hydrate reservoir rocks (Sun XJ et al., 2012; Hyodo M et al., 2013; Uchida S et al., 2012), while the effect of pore compression on permeability (stress sensitivity) in hydrate-bearing sediments is rarely considered.

The Pore Network Model (PNM) is a well-established method for simulating fluid transportation in porous media (Blunt MJ, 2001), which have also been widely employed to simulate the permeability variation of hydrate-bearing sediments (Dai S et al., 2014; Jang J and Santamarina JC, 2011; Mahabadi N and Jang J, 2014; Mahabadi N et al., 2016). In traditional numerical simulation experiments about the permeability of hydrate-bearing sediments using PNM, the flowing path is considered blocked once hydrate grows in it and vice versa (Dai S and Seol Y, 2014; Jang J and Santamarina JC, 2011; Mahabadi N and Jang J, 2014; Mahabadi N et al., 2016). While in practical conditions, hydrate dissociates gradually in the porous sediment, instead of abruptly disappearing. Besides, the relationship between permeability and hydrate saturation is different between hydrate dissociation from the pore surface and in the pore center (Dai S et al., 2014; Daigle H, 2016; Delli ML et al., 2013; Kleinberg RL et al., 2003; Masuda Y, 1997). In order to accurately simulate the permeability variation during hydrate dissociation in the pore space, the gradual process and the dissociation habit should both be taken into consideration.

In this paper, the authors take the hydrate dissociation as a gradual process that can start either from the surface of sediment grains (grain coating) or in the pore center (pore filling) and built the permeability change model with PNM method. Then the hydraulic conductance equation is established to depict the fluids transportation between pores in the hydrate dissociation process. Three hydrate dimensional parameters ($H_{inner}$, $H_{pore}$, and $H_{throat}$) are defined to describe the gradual hydrate dissociation process. Permeability variation, caused by hydrate dissociation anywhere in the pore network, can be obtained from changing of these parameters. Synthetic effect on permeability from hydrate dissociation and the pore compression induced by increasing of effective stress is simulated. The simulation results based on PNM method can provide the physical properties variation of the hydrate deposits, which can be used to evaluate the production performance.

2. Method

The pore network model in this research is developed based on peer-reviewed and open-source pore network modeling framework (OpenPNM), which is written in the Python environment (Gostick J et al., 2016). The OpenPNM package is originally designed for conducting simulations of multiphase transportation in porous media, such as electrode of fuel cells (Gostick JT et al., 2006, 2007, 2008, 2009), and is recently adopted to simulate the sandstones (Sharqawy MH, 2016). OpenPNM maintains good computational performance and compatibility, and is widely applied in many research areas (Aghighi M et al., 2016; Chevalier S et al., 2016; Tranter TG et al., 2016).

2.1. Geometrical characterization of sediment sample

The pore network model is composed of nodes representing the pore center and the conduit connecting adjacent nodes (Fig. 1a). According to Darcy’s law, a sparse matrix between the pore pressure and hydraulic conductivity based on material balance can be obtained and can be solved by the boundary conditions of the node pressure. With the calculated pressure value of each node, the flow rate of each conduit is gained to calculate the permeability of the sample.
Fig. 1. Schematic diagram of the pore network model. a—the nodes and the conduit (black dashed line) connecting adjacent nodes; b—simplified model of the conduit. Pore space in white, and grain particle in grey.

Therefore, the critical input parameters in the PNM model include node position, node connectivity, and hydraulic conductivity of conduits.

The arrangement of nodes in the three-dimensional pore network model is generally divided into two categories: (1) Structured node network, which is the most commonly adopted network generated by a specific algorithm that follows a given topological pattern. (2) The unstructured network, that is generated by extracting from the 3D CT reconstruction data (Blunt MJ et al., 2013; Dong H and Blunt MJ, 2009; Gostick JT, 2017; Raeini AQ et al., 2017). The applicability of the unstructured network is constrained to characterize specific samples from which the network is generated. In this study, the structured pore network with better generality is used to eliminate the influence of the topological features on simulation results.

The connectivity between nodes is represented by the coordination number. The coordination number indicates the number of conduits connected to the nodes, and a larger coordination number results in more conduit (throat) and higher permeability. For cubic grids, the coordination number can be 6 (face connection), 8 (corner connection), 12 (edge connection), 14 (face and corner connection), 18 (face and edge connections), 20 (edge and corner joints), and 26 (face, edge, and corner joints). The node position and coordination number are generally referred as the topology information of the pore network model.

The shape and size of pores and throats in the pore network model are termed as the geometric information of the pore network model. The geometric information can be characterized by conduit, which can be divided into three sections: The left half pore (①), throat (②), and the right half pore(③) (Fig. 1a). The shape of the pores and throats are spherical and cylindrical, in other words, the ball-and-tick model.

In the structured pore network, the number of pores can be as large as 125000, and the sizes of each pore and throat are randomly generated with a random function. The algorithm can provide pore and throat sizes obeying the same statistical distribution, while the geometry values obtained in each random arrangement can be different. The geometric dimensions of the network follow the normal distribution, determined by the average value and standard deviation of the input. The number of pores should be large enough so that the random algorithm will not affect the result. In this study, it is found that in case the number of pores exceeds 100000, the deviation of permeability calculated from randomly generated network is less than 0.1%, which is acceptable.

Instead of generating from the random algorithm, the throat diameter is determined by the diameter of the smaller pore connected to the throat multiplied by the throat-pore ratio defined as:

$$d_{\text{throat}} = d_{i/p} \times d_{\text{min}} \quad (1)$$

Where $d_{\text{throat}}$ is throat diameter, $d_{\text{min}}$ is the pore diameter of the smaller pore connected to the throat.

Depending on the shape of the network, the values of the throat-pore ratio are different. For example, the throat-pore ratio of a cubic arrangement of identical spheres is 0.565, a face-centered arrangement is 0.374, while a body-centered arrangement is 0.688 (Kruyer S, 1958). For the stacking of glass beads with different sizes, the measured throat-pore ratio range is 0.242–0.698 (Al-Raoush RI and Willson CS, 2005). The throat-pore ratio also can be used to adjust model porosity and permeability for better fitting the network with the real samples.

As shown in Fig. 1, the conduit in the pore network model is simplified from hemisphere-cylinder-hemisphere to cylindrical-cylindrical-cylindrical to calculate the permeability of the conduit as follows:

$$\bar{S} = \frac{\pi D_i^4}{128L_\mu} \quad (2)$$

Where $D_i$ and $L_\mu$ is the diameter and length of the cylinder in the $i_{th}$ segment, and $\mu$ is the viscosity of the fluid.

According to the Hagen-Poiseuille equation, the permeability of the entire conduit can be given as:

$$\frac{1}{\bar{S}} = \frac{1}{\bar{S}_{cyl1}} + \frac{1}{\bar{S}_{cyl2}} + \frac{1}{\bar{S}_{cyl3}} \quad (3)$$

2.2. Permeability change model

In Fig. 2a, three hydrate dimensional parameters, ranging from 0 to 1, are defined to represent hydrate forming in different positions of the pore space. For the hydrates formed in the pore center (pore filling), $H_{\text{center}}$ is defined as the ratio of hydrate radius ($h_{\text{center}}$) with the pore radius ($r_{\text{pore}}$) and its value changes from 0 to 1 meaning hydrate start to form in the pore center until filling the whole pore space. In the dissociation process depicted in Fig. 2b, $H_{\text{center}}$ change from 1 to 0. On the
other hand, when hydrates formed at the pore surface (grain coating), \( H_{\text{pore}} \) represents the ratio of hydrate thickness (\( h_{\text{pore}} \)) with the pore radius. \( H_{\text{throat}} \) is the ratio of hydrate thickness (\( h_{\text{throat}} \)) with the radius of the cross-section of the cylindrical throat (\( r_{\text{throat}} \)). During the hydrate formation process, \( H_{\text{pore}} \) and \( H_{\text{throat}} \) changes from 0 to 1 at the pore and throat surface. While when hydrate dissociates, as indicated in Fig. 2c, \( H_{\text{pore}} \) and \( H_{\text{throat}} \) decreases from 1 to 0.

Based on the defined geometrical information of the pore network, the hydraulic conductivity of the conduit can be calculated. As shown in Fig. 3, the conduit models are simplified to gain the analytical expression of the sediment conductivity. When hydrate grows at the grain surface (grain coating), the conduit can be divided into three zones, all of which are simplified to be cylindrical. While in the case of pore filling, five segments are included in the conduit, in which three are cylindrical and the rest two of them are annular zones of half-pores. The hydraulic conductivity of the cylindrical annulus can be given by:

\[
 \kappa_{\text{hydrate}} = \frac{\pi}{128L\mu} \left[ D^2 - d^2 - \frac{(D^2 - d^2)^2}{\log \left( \frac{D}{d} \right)} \right]
\]

Where \( D \) and \( d \) is the outer and inner diameter of the annular section, respectively. The permeability of the pore network can be obtained by Eq. 3.

3. Results and discussion

3.1. Pore network

In this simulation, the geometry of the pore network model is built from the core sample of hydrate-bearing sediments in the Shenhu area of the South China Sea (Chen F et al., 2011; Li CH et al., 2014). The sample is siltstone with grain sizes ranging from 0.008 mm to 0.063 mm and porosity of 0.26. The intrinsic permeability of the sample without hydrate is 2.8×10⁻¹² m². As shown in Fig. 4, the total number of pores in the cubic pore network is 125000 with the coordination number of 6. The throat-pore ratio can be modified to better fit with the measured porosity and permeability of the core sample and the best fit occurs at 0.64. Since the gas-water ratio from hydrate dissociation is fixed, the two-phase flow in the pore network is treated as a single-phase flow. The injection phase is water, and the simulation temperature is 293 K.

Fig. 2. Demonstration of the hydrate dissociation under morphology of pore filling and grain coating. a—the definitions of \( H_{\text{pore}}, H_{\text{throat}}, H_{\text{pore}} \); b—hydrate dissociation underhydrate morphology of pore filling; c—hydrate dissociation under hydrate morphology of grain coating. Pore space in white, grain particle in grey, hydrate cluster in blue and hydrate dissociation direction represented by blue arrow.

Fig. 3. The pore-throat-pore conduit and the simplified conduit under different hydrate occurrence morphology. (Hydrate: blue; Pore space: white; Sediment grain: grey). a—the pore-throat-pore conduit with grain coating hydrates (Zone 1 and 3: half pore; Zone 2: throat); b—the pore-throat-pore conduit with pore filling hydrates (Zone 1 and 5: annular zones of half-pores; Zone 2 and 4: cylindrical zones of half-pores; Zone 3: throat); c—the simplified conduit of a; d—the simplified conduit of b.

Fig. 4. The pore network built from the hydrate-bearing sediments of the Shenhu area in South China sea, in which balls with different colors correspond to pores with different sizes and for better visualization, throats are not shown. The dimension of the pore network is 50×50×50 (non-dimensional).
3.2. Dynamical behavior of the reservoir during hydrate production

In the first offshore natural gas hydrate production trail in the South China Sea, formation fluid extraction method was adopted. As depicted in Fig. 5, formation pressure drop can be achieved by extracting fluid from the hydrate-bearing sediments to induce hydrate dissociation. To simulate the permeability change during hydrate production adopting formation fluid extraction method, pore space variation caused by the opposite effects from pore compression and hydrate dissociation are both taken into consideration. Moreover, the dynamical behavior of hydrate dissociation from hydrate morphology of pore filling and grain coating is also considered.

At the initial state, the three-dimensional parameters in the pore network have the value of $H_{inner} = 0.9$ and $H_{pore} = H_{throat} = 0.5$, enabling the hydrate saturation matches the core sample of the sediment from the Shenhua area of the South China Sea.

In the following, the pore compression effect is considered in this model. The pore compression effect can be expressed by compression rate $R$, defined as the reduction rate of the length of the cubic network. Here it assumes all the three directions is uniformly compressed. Additionally, it is assumed that the mineral grains are incompressible and the volume reduction originates from the pore space condensation. Then the relationship between the porosity ($por_i$) of the $i$ calculation step, the compression rate ($R_i$) and the initial porosity ($por_0$) of the pore network can be given by:

$$por_i = \frac{por_0 - [1 - (1 - R_i)^3]}{(1 - R_i)^3}$$

The pore network is rebuilt at every calculation step since the porosity has been changed. It is found that when the compression rate is not larger than 5%, the porosity of the sediment sample is deemed to be proportional to the compression rate.

In this simulation, the hydrate size in pore space is assumed linearly change with the compression rate, that is, the compression rate is proportional to the hydrate dimensional parameters that describe its shape. Therefore, the compression rate at the $i$ calculation step is as following:

$$R_i = R_{end} \times \frac{H_i}{H_{max}}$$

Where $R_i$ is the compression rate of the $i$ step, $R_{end}$ is the final compression rate defined as the compression rate when the hydrate is completely dissociated and in this simulation $R_{end}$ has different values of 0%, 1%, 2%, 3%, 4% and 5%, $H_g$ is either $H_{inner}$ or $H_{pore}$ that depends on the hydrate morphology, $H_{max}$ is the maximum value of the hydrate dimensionless parameters of $H_{inner}$ and $H_{pore}$. As shown in Fig. 6b, the sample length of the pore network linearly changes with the hydrate dimensional parameter in both hydrate morphology.

As mentioned earlier, the pore space will be modified by pore compression. After considering the combination effects of pore compression and hydrate dissociation, the hydrate saturation at the $i$ calculation step is given by:

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**Fig. 5.** Schematic diagram of the formation fluid extraction method.

**Fig. 6.** Hydrate saturation (a) and sample length (b) variation with hydrate dimensional parameters in the process of producing gas from hydrate via depressurization method.
\[ S_{\text{hydrate}, i} = \frac{V_{\text{hydrate}, i}}{V_{\text{por}, i}} \]  

Where \( V_{\text{hydrate}, i} \) is the total volume of hydrate at step \( i \), \( V_{\text{por}, i} \) is the total volume of pore space at step \( i \). It should be noticed that the hydrate dissociation rate should be larger than the pore compression rate to avoid the hydrate occupying the whole pore space. The calculation results show that even at the maximum final compression rate of 5%, the pore volume released at each step of hydrate dissociation in the pore space is greater than the pore volume reduced by compression.

3.3. Permeability change with hydrate dissociation and pore compression

The dissociation of hydrate from the pore center is firstly simulated. Fig. 7a presents the relationship between the sediment porosity and hydrate dimensional parameter with and without considering hydrate dissociation in the pore space. The case without hydrate dissociation effect shown as the dotted line in Fig. 7a represents the pore compression effect. In this case, the porosity linearly decreases with the hydrate dimensional parameter reduction. The solid lines depict the combination results from pore compression and hydrate dissociation. In the case of the final compression rate is 0, i.e., the pore compression effect is absent. The porosity increase with hydrate size and the increasing rate gradually declines, indicating that hydrate dissociates very fast at the early stage and slows down later. This tendency is coincident with Fig. 6a. In other cases, the porosity firstly increases until to a certain value and then decreases afterward.

The permeability of hydrate-bearing sediments changes with the hydrate saturation under synthetic effects from pore procession and hydrate dissociation is simulated and the results are shown in Fig. 7b. Similar to Fig. 7a, the dotted lines indicate the separate effect from pore compression. The permeability decreases with the hydrate saturation and especially when the saturation is as low as 0.1, the permeability quickly decreases. The blue solid line demonstrates the relationship between the permeability of sediments sample and hydrate saturation without pore compression. In this case, the permeability increases with hydrate saturation with accelerated rate. Then taking consideration of both effects from pore compress with different final compression rates and hydrate dissociation, the permeability increases resulted from the space released by hydrate dissociation is greater than pore compression. While when hydrate saturation is weighing too much lower, the pore compression is superior to hydrate dissociation effect and the permeability becomes smaller accordingly. With increasing of the final compression rate, the hydrate saturation corresponding to the inflection point increases.

In the following, the simulation of grain-coated hydrate dissociation is conducted and the results are shown in Fig. 8. As shown in Fig. 8a, the trend of porosity variation with the hydrate size without considering hydrate dissociation is the same with the pore filling type which is linear. While different from the pore filling type, the porosity linearly increases with hydrate size reduction in the whole range when both pore compression and hydrate dissociation are considered. This result demonstrates that hydrate dissociation effect plays a dominant role in the grain coating hydrates. Since hydrate dissociation rate is constant as depicted in Fig. 6a, the porosity also linearly change with hydrate size, and its increasing rate decrease with a higher pore compression rate.

Next, the relationship between the permeability of the sediment sample and the hydrate saturation is given in Fig. 8b. When neglecting hydrate dissociation effect that shown as the dotted lines, pore compression induces permeability reduction with a gradually decreasing rate. While hydrate dissociation will release pore space and hence permeability increases. Adding up the effects from the pore compression and hydrate dissociation, the permeability still increases with hydrate dissociation that shown as the solid line. The dominant role of
hydrate dissociation over pore compression is again verified. Under different hydrate morphology, the relationship between the permeability of the sample and the hydrate saturation in the process of hydrate dissociation is quite different. The permeability of pore-filling type increases at the early stage and then decreases with saturation. The greater of the compression degree, the more severe of the decrease in permeability. However, the permeability in the sediments with grain coating hydrates keeps increasing with saturation. This is because the increasing rate of permeability caused by the dissociation of pore-filled hydrates is gradually reduced, if the hydrate saturation decreases to a certain value, the pore space released by hydrate dissociation is smaller than the pore compression. On the other hand, dissociation of grain-coated hydrate with constant rate plays the dominant role in the whole production process and the sediment permeability keeps increasing. The difference of relationships between permeability and hydrate saturation under these two hydrate morphology results from the different hydrate volume variation with hydrate dissociation. When the pore-filled hydrates dissociate, the hydrate volume change is proportional to the square of the hydrate dimensional parameter. Therefore, released pore space contributing to permeability increment is gradually reduced with hydrate dissociation until the saturation decreases to a certain value that the hydrate dissociation is comparative with pore compression effect. While in case of hydrate at the grain surface dissociates, the hydrate volume change slightly increases with the increase of pore diameter, so the effect of hydrate decomposition on permeability is gradually increased and keeps playing a dominant role during the whole hydrate dissociation process.

Also, it should be pointed out that in a mechanical experiment conducted with the core sample from the northern part of the South China Sea (Zhang HW et al., 2017), the porosity of the sample withstands a reduction of 0.05 under 16 MPa triaxial compression. In this simulation, the maximum value of the final compression rate is 5%, corresponding to the porosity reduction of 0.13. Besides, in a practical production trial, the pressure reduction is normally less than 5 MPa, much less than the pressure in the experiment. Therefore, the degree of pore compression in practical field tests should be less than that in this simulation. That is to say, under these simulation conditions, the effect induced by pore compression is less than the hydrate composition, no matter what types of hydrate occurrence morphology. During hydrate dissociation induced by depressurization, the permeability of the reservoir keeps increasing.

4. Conclusion

In this study, the pore network model method is employed to establish a reservoir permeability change model by pore compression and pore space release due to hydrate dissociation in the course of producing gas from hydrate via depressurization. It is also revealed that the permeability changes differently when hydrate occurrence morphology is different. For the pore filling hydrates, the permeability of sediment decreases after goes up first, while for grain coating hydrates, the permeability keeps increasing throughout the production process. However, within a certain range (<5%) of pore compression rate, the hydrate dissociation effect is more significant than the pore compression effect, so reservoir sediment permeability keeps increasing in the hydrate production process.

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