

# Influence of gas hydrate formation on methane seeps at the bottom of water reservoirs

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## Abstract

It is shown by numerical modeling that the height of gas flares above underwater methane seeps depends strongly on water parameters. A simulation model of the dynamics of a rising bubble was used. Along with gas exchange through the bubble wall, the model takes into account gas hydrate formation, whose rate is determined by turbulent heat exchange with water. Calculations were performed for depths of 250 to 1500 m with an initial bubble diameter of 0.2 to 1.5 cm. It is shown that at the water temperature of the Arctic seas, rising methane bubbles transform into gas hydrate ice within the first 1–2 m of their path. At the same time, when the water temperature is higher than the hydrate formation temperature near the sea bottom or close to it, the bubbles travel a distance of tens or hundreds of meters before complete dissolution or complete hydration.

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## Introduction

An indicator of offshore natural gas fields is the release of methane bubbles in the form of characteristic gas flares through the bottom, which are well detected by sonars (Goncharov and Klement'eva, 1996). These effects are well-studied in the Black Sea and the Sea of Okhotsk (Bobrovnikov et al., 2012; Obzhirov et al., 2012), in lake Baikal (Granin et al., 2010, 2012) and in other water bodies. According to observations and numerical simulations, the height of such flares reaches hundreds of meters if the depth of the bottom does not exceed the critical depth of hydrate formation. However, at great depths, methane in the bubbles forms gas hydrate (gas hydrate ice) with water at temperatures typical of bottom water (Egorov et al., 2012; Makagon, 2010). Calculations and laboratory experiments (Dontsov et al., 2007; Zaporozhets and Shostak, 2014) have shown that, under certain conditions, gas hydrate formation in bubbles is very fast: bubbles transform into gas hydrate particles in a few seconds. At a bubble rise velocity of up to 0.3 m/s (Degterev and Mordashev, 2008), they rise only 1 to 2 m above the bottom. This implies that at certain water temperature and pressure, a gas flare does not form near the bottom. In this

case, clouds of solid particles of methane gas hydrate form above methane seeps, which are much more difficult to detect by acoustic methods. The conditions under which this occurs are the subject of the present paper.

The dependence of the critical depth of hydrate formation  $z_{cr}$  on water temperature is well known (Fig. 1). The problem is to model the dynamics of gas exchange and hydrate formation in a bubble at depths  $z > z_{cr}$ . Due to different hydrological conditions, the values of  $z_{cr}$  differ significantly for different water bodies. In the Arctic seas, hydrate formation occurs at a depth of not less than 250–300 m, whereas, e.g., in the Black Sea, where the deep-water temperature is about 9 °C, hydration of rising methane bubbles occurs only at depths over 500–600 m. Note that the dissolution of rising methane bubbles in water due to gas exchange is slower than hydrate formation (Degterev and Mordashov, 2008), so that outside the hydrate formation zone, the height of gas flares above the bottom can reach hundreds of meters. Since  $z_{cr}$  greatly depends on water temperature, the occurrence of methane seeps at great depths is largely determined by hydrological conditions.

In recent years, several mathematical models for the dynamics of a bubble rising in water have been proposed that take into account gas exchange through the bubble wall and gas hydrate formation in the bubble (Nurislamov et al., 2015; Zaporozhets and Shostak, 2014). Typically, these models have

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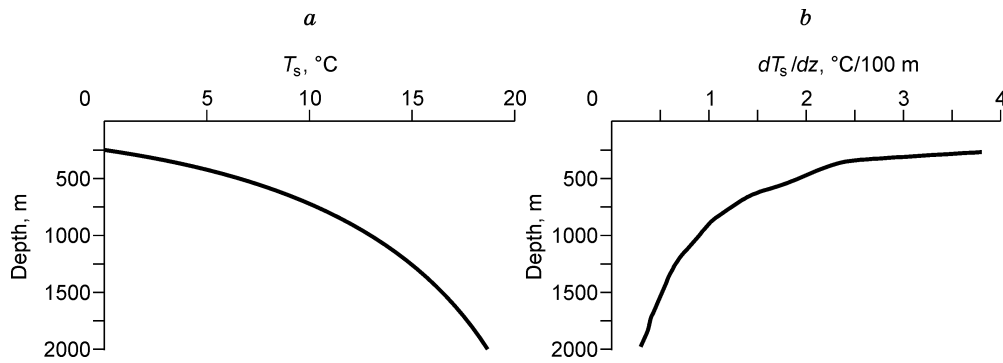


Fig. 1. Hydrate formation temperature as a function of depth (a), and the hydrate formation temperature gradient as a function of depth (b).

been developed for specific tasks not related to the detection of methane seeps at the bottom of water bodies, so that they are difficult to use in the case considered. In particular, they do not adequately describe the dynamics of physical processes in rising bubbles with consideration of the influence of environmental parameters. For this purpose, it is better to use a simulation model that describes not only the changes in pressure and gas content due to gas exchange through the bubble wall and the change in depth, but also the processes related to the formation and melting of gas hydrate. Of course, this model uses some parametrization schemes proposed in the already existing models.

## Model

The dependences of the parameters of rising bubbles on water temperature and pressure taking into account the formation of methane gas hydrate were studied using a numerical model which has previously been applied to the description of gas bubble dynamics in the upper layer of the sea (Degterev and Kolobaev, 1993). Even the usual problem of the rise of a gas bubble at shallow depths cannot be solved analytically. The need to consider the viscosity of water and the gas flow through the bubble wall leads to a system of equations which is solved only numerically. The proposed model is based on the calculation of parameters characterizing the state of the bubble at successive times. In view of the rate of hydrate formation, the corresponding time step was taken to be 0.05–0.5 s.

The state of a bubble with a single-component gas is described by specifying the current values of the radius  $r$ , the rising velocity  $v$ , and the coordinate  $z$ . In this case, it is necessary to take into account the presence of methane hydrate, in addition to methane, in the bubble. Usual condensation of methane does not occur in this case since the water temperature in natural water bodies is above the critical temperature of methane. The bubble rises under the action of buoyancy and the viscous drag of water. The balance between them is achieved within the first millimeter of bubble rise. As a result, the bubble of constant radius rises at a constant velocity. For spherical bubbles of radius  $r$ , it is described by

well-known Stokes theory, which, neglecting the weight of the bubble, gives the formula

$$v = 2gr^2/9\nu, \quad (1)$$

where  $\nu$  is the kinematic viscosity coefficient of water and  $g$  is the free-fall acceleration. For not very small bubbles, this formula uses a multiplicative correction for the nonsphericity of large bubbles type (Thorpe, 1984):

$$v = M_1(2gr^2/9\nu) M_2, \quad (2)$$

where  $M_1 = 1/((G^2 + 2G)^{1/2} - G)$  and  $G = 10.82\nu^2/gr^3$ . In addition, unlike problems related to the upper layer of the sea (Degterev and Mordashov, 2008), here it is necessary to take into account the density of the compressed gas in the bubble, which at depths of 1000–2000 m reaches values comparable with the density of water  $\rho$ . The weight of the bubble also increases due to the formation of gas hydrate in it. In this connection, formula (1) was supplemented with a factor of the form

$$M_2 = 1 - (m_g + m_h)/V\rho, \quad (3)$$

where  $m_g$  is the mass of the gas in the bubble,  $m_h$  is the mass of gas hydrate in the bubble, and  $V$  is the volume of the bubble. This correction describes the decrease in the bubble rising velocity due to gas hydrate formation in the bubble because, in this case, the total mass  $m_g + m_h$  increases due to the water molecules entering the hydrate. In particular, in the case of complete inclusion of methane in gas hydrate, the velocity of the bubble transformed into a gas hydrate particle is reduced by an order of magnitude.

Thus, knowing the current values of the bubble radius  $r$ ,  $m_g$ , and  $m_h$ , we can calculate  $v$ . The bubble radius is related to the hydrostatic pressure and hence to the coordinate  $z$  through the gas equation of state. In the absence of gas hydrate, the ideal gas equation of state has the form

$$m_g = V(P_{\text{atm}} + \rho gz + 2\sigma/r)/RT, \quad (4)$$

where  $m_g$  is the mass of the gas in the bubble,  $P_{\text{atm}}$  is the atmospheric pressure,  $\sigma$  is the surface tension of water,  $R$  is the universal gas constant, and  $T$  is the gas temperature. When taking into account the gas hydrate, which also occupies part

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