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# Experimental set-up design for gas production from the Black Sea gas hydrate reservoirs



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## ABSTRACT

Gas hydrate deposits which are found in deep ocean sediments and in permafrost regions are supposed to be a fossil fuel reserve for the future. The Black Sea is also considered rich in terms of gas hydrates. It abundantly contains gas hydrates as methane (CH<sub>4</sub> ~ 80–99.9%) source. In this study, by using the literature seismic and other data of the Black Sea such as salinity, porosity of the sediments, common gas type, temperature distribution and pressure gradient, the optimum gas production method for the Black Sea gas hydrates was selected as mainly depressurization method. It was proposed that CO<sub>2</sub>/N<sub>2</sub> injection as a production method from the potential Black Sea gas hydrates might not be favorable. Experimental set-up (high pressure cell, gas flow meter, water-gas separator, mass balance, pressure transducers and thermocouples) for gas production from the Black gas hydrates by using depressurization method was designed according to the results of HydrateResSim numerical simulator. It was shown that cylindrical high pressure cell (METU Cell) with 30 cm inner length and 30 cm inner diameter with a volume 21.64 L in this study might reflect flow controlled conditions as in the real gas hydrate reservoirs. Moreover, 100 mesh portable separator in METU cell might be very useful to mimic Class 1 hydrate reservoirs and horizontal wells in gas hydrate reservoirs experimentally.

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## 1. Introduction

With the decline of the amount of gas in conventional gas reservoirs, unconventional gas reservoirs such as gas hydrates and shale gas reservoirs have become very popular recently (Kok and Merrey, 2014). Gas hydrates are ice like crystalline structures formed by water and gas molecules at high pressure and low temperature values. They are defined as nonstoichiometric compounds, which means the ratio of the atoms present in the composition is not a simple integer (Carroll, 2009). Hydrocarbon molecules such as methane (CH<sub>4</sub>), ethane (C<sub>2</sub>H<sub>6</sub>), propane (C<sub>3</sub>H<sub>8</sub>) and i-butane (i-C<sub>4</sub>H<sub>10</sub>) form their own hydrate (simple hydrate) at high pressure and low temperature conditions when there is enough water in the system. Similarly, carbon dioxide (CO<sub>2</sub>), hydrogen sulfide (H<sub>2</sub>S), nitrogen (N<sub>2</sub>), oxygen (O<sub>2</sub>) and other gases form hydrate at their hydrate equilibrium conditions (Sloan and Koh, 2007).

According to the gas in place calculations of Johnson (2011) in

hydrate bearing sands in the world, there is a huge range of gas hydrate resource between 133 and 8891 tcm. It can be concluded that even the most conservative estimates of the total quantity of gas in gas hydrate are much larger than the conventional gas resources (404 tcm) and shale gas (204–456 tcm) (Chong et al., 2015). The magnitude of this resource can make hydrate reservoirs a substantial future energy resource. Currently, there are mainly four gas production methods from gas hydrate reservoirs: depressurization, thermal stimulation, chemical injection, and CO<sub>2</sub> injection. Depressurization is thought to be the most economically viable production method for gas hydrates because there is no extra heat introduced into the system. This method is applied by decreasing reservoir pressure within hydrate stability zone, causing hydrate to decompose and release gas and water that will migrate towards the wellbore. Although there is no additional heat input cost of depressurization method, its disadvantages are low gas production rates, high amounts of water production, the risk of hydrate reformation due to fast depressurization, and the risk of reservoir subsidence (Konno et al., 2010; Chong et al., 2015; Xu and Li, 2015). By increasing the temperature of hydrate deposits, reservoir conditions are shifted the outside of hydrate equilibrium conditions. Below hydrate equilibrium line, hydrate starts to dissociate after

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the increase of temperature. There are several ways to increase temperature such as steam injection, hot water injection, electric heating or microwave heating (Liang et al., 2008; Xu and Li, 2015; Chong et al., 2015). Thermal stimulation is very effective to dissociate hydrate. However, the injection of heat into hydrate deposits is very expensive. Moreover, injected heat is not only absorbed by hydrate deposits, it is also absorbed by non-hydrate deposits such as sands, boundaries, etc. Therefore, cost estimations and energy efficiency ratio calculations are important for thermal stimulation studies. The aim of chemical injection into gas hydrate reservoirs is to shift hydrate equilibrium line upward and then reservoir conditions is below hydrate equilibrium line. At these conditions, hydrate starts to dissociate. Compared to other production methods such as depressurization and thermal injection, this method is not preferred much by scientists because it is very expensive and environmentally harmful (Demirbas, 2010; Moridis et al., 2013; Chong et al., 2015). CO<sub>2</sub> injection is quite different among the production methods of gas hydrate reservoirs. This method was firstly suggested by Ohgaki et al. (1996). Basically, the difference of thermodynamic stability between CH<sub>4</sub> and CO<sub>2</sub> causes CH<sub>4</sub> molecules' leaving the cages of its hydrates and empty cages are filled by CO<sub>2</sub>. This replacement is called CO<sub>2</sub>–CH<sub>4</sub> swapping (Chong et al., 2015; Xu and Li, 2015). This method is advantageous both in terms of CH<sub>4</sub> production from hydrates and CO<sub>2</sub> sequestration for environmental purposes. CO<sub>2</sub>–CH<sub>4</sub> swapping also keeps the sediments geomechanically stable (Park et al., 2006; Zhao et al., 2012; Ors, 2012; Abbasov, 2014; Hyodo et al., 2014).

As well as in the world, it is considered that the Black Sea has a huge biogenic and thermogenic gas hydrate potential (Vassilev, 2006; Johnson and Max, 2015; Haeckel et al., 2015; Mery and Sinayuc, 2016). Table 1 shows potential CH<sub>4</sub> amount in gas hydrate stability zone (GHSZ) of the Black Sea. However, all deposits including gas hydrates in GHSZ is not considered as an energy source. Gas hydrates are found in arctic sand reservoirs, marine sand reservoirs, non-sand marine reservoirs, massive sea floors and marine shales. However, only gas hydrates in Arctic sand reservoirs and marine sand reservoirs are considered as an energy source (Boswell and Collett, 2006; Johnson, 2011; Johnson and Max, 2015; Mery and Sinayuc, 2016). Gas productions from non-sand marine hydrates, massive seafloor or shallow hydrates, and marine shale hydrates are very difficult even though there is a huge gas hydrate potential in these reservoirs compared to arctic and marine sand reservoirs. In these hydrate reservoirs, reservoir quality and fractional gas production recovery are quite low. Therefore, Table 2 shows the potential CH<sub>4</sub> deposited in sands as gas hydrate in the Black Sea. The Black Sea might include CH<sub>4</sub> as an energy source up to 13.6 trillion cubic meter (tcm) in its potential gas hydrates (Mery and Sinayuc, 2016).

The Black Sea is an inland sedimentary basin, located between the latitudes of 41°–46°N and longitudes of 28°–41.5° E with an area of 423,000 km<sup>2</sup>, a volume of 547,000 km<sup>3</sup> and a maximum depth of 2200 m (Murray, 1991). It has a connection to the Sea of Azov by the Kerch Strait in the north, while it is connected to the Mediterranean Sea with the Bosphorus Strait through the Sea of Marmara in the south. Near the shores of the Black Sea, the depth of

sea is shallow but after little far away from the shores, the sea level depth suddenly decreases up to 2212 m (Railsback, 2010; Stanev et al., 2014). As shown in Fig. 1, the salinity of the Black sea increases from 1.75% to 2.15% between sea surface and 200 m below sea level (mbsl). After 200 mbsl, the increment of salinity with sea depth slows and then the salinity of sea water becomes approximately 2.23% on the sea floor. As many places in the world, the Black Sea also has a huge gas hydrate potential and it is also considered as the world's most isolated sea, the largest anoxic water body on the planet and a unique energy-rich sea (Overmann and Manske, 2006). It abundantly contains gas hydrates and H<sub>2</sub>S as CH<sub>4</sub> and hydrogen source, respectively (Demirbas, 2009). CH<sub>4</sub> seepage is extremely intense on the shelf and on the slope of the Black Sea (Sozansky, 1997; Vassilev and Dimitrov, 2000; Naudts et al., 2006; Küçük et al., 2015). Naudts et al. (2006) observed gas seepages between 66 and 825 mbsl (meters below sea level) in the Dnepr paleo-delta, northwestern Black Sea as shown in Fig. 2. However, in this area below 825 mbsl, there is no gas seepage observed because gas hydrate stability zone is below 825 mbsl for this delta. If these seepages are natural and slow, most of gas released is oxidized in sea water. However, if there are sudden gas releases because of slope failures, etc., it is harmful for environment (Xing, 2013). According to Vassilev and Dimitrov (2003), the area of the Black Sea suitable for gas hydrate formation is evaluated at 288,100 km<sup>2</sup>, representing about 68.5% of the total Black Sea or almost 91% of the deep-water basin.

Numerical simulation of gas production from gas hydrates in laboratory scale and reservoir scale is very important for gas hydrate studies because there are not many real gas production data from hydrate reservoirs. There are several numerical codes for the simulation of gas production from hydrate reservoirs such as HydrateResSim, CMG Stars, MH-21 Hydres, Tough + Hydrate, the code of University of Houston, Hyres, Stomp-Hyd-Ke and Mix3-HydrateResSim (Garapati, 2013). Most of these codes are used to simulate gas production from hydrate reservoirs by using depressurization, thermal injection, chemical injection and combination of different production methods. Differently, Stomp-Hyd-Ke and Mix3HydrateResSim are used to simulate CH<sub>4</sub>–CO<sub>2</sub> swapping and CH<sub>4</sub>–CO<sub>2</sub> & N<sub>2</sub> swapping respectively (Gaddipati, 2008; Garapati, 2013). Experimental studies related to gas hydrates are quite important especially when there are not much field data available of real gas hydrate reservoirs. Therefore, experimental studies try to investigate gas production mechanism in hydrate reservoirs. Furthermore, they give necessary data for the simulation studies. Previously, the volumes of high pressure hydrate cells (reactors) were quite small for gas production experiments from hydrates. Masuda et al. (1999) conducted depressurization experiments in a 589 cm<sup>3</sup> high pressure cylindrical cell. Similarly, the depressurization experiments on Berea sandstone were conducted in a 171 cm<sup>3</sup> cylindrical high pressure cell by Yousif et al. (1991). Experimental studies of Masuda et al. (1999) were simulated by different scientists (Gamwo and Liu, 2010; Ruan et al., 2012; Zhao et al., 2012). In all these studies, with the kinetic equation of Kim et al. (1987), there is a good fit to the experimental study of Masuda et al. (1999). Moreover, Gamwo & Liu (2010) compared the

**Table 1**  
CH<sub>4</sub> potential of the Black Sea Hydrates.

Source	Initial gas (methane) in place in the Black Sea hydrates, tcm
Korsakov et al., 1989	40–50
Klauda and Sandler, 2003	850
Shi, 2003	42
Vassilev and Dimitrov, 2003	42 to 49 (10–50)
Mery and Sinayuc, 2016	13.6 (0.021–138)

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