Identification of shallow gas origin in the Ulleung Basin, East/Japan Sea

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1. Introduction

The study area is the eastern part of the Ulleung Basin, East/Japan Sea. From the seismic survey on 2003, the structures such as pork mark, acoustic blanking and chimney associated with the gas were found. Additionally, the National Oceanographic Research Institute of Korea (NORI) conducted regional seafloor mapping of the East Sea on 1996 and the Korea Ocean Research and Development Institute (KORDI) performed multi-channel deep seismic survey and sampled piston core (core 98EEZ-3) in the study area. Based on the results of 1996 and 1998 surveys, Lee and Chough (2003) suggested that the shallow gas is originated by thermogenic source. However, the results of 2003 survey indicate that shallow methane gas comes from the bacterial source rather than thermogenic. Using core sediments taken from the eastern part of the Ulleung Basin, therefore, we have focused on the identification of shallow gas origin and comparison our results with Lee and Chough (2003). For these purpose, we have analyzed pore water composition, headspace gas compositions and their isotope ratios, and Rock-Eval pyrolysis of organic matter in sediments.

2. Method

Four piston cores were taken from the eastern part of the Ulleung Basin, East Sea at water depths ranging from 1,197 to 2,179 m using R/V Tamhae 2 owned by the Korea Institute of Geoscience and Mineral Resources (KIGAM) on May 2003.

For the Rock-Eval analysis, core samples were routinely taken at every 10 cm interval and the spot where the sedimentary facies is changed, and powdered in an agate vessel after drying in a freezing dryer for 24 h. The powdered sample was used

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by VINCI^Rock-Eval 6. Pore water was extracted from core samples by centrifuging for 30 min. Chloride (Cl^-) concentration was determined by Mohr titration with silver nitrate using potassium chromate/potassium dichromate as an indicator, and sulfate (SO_4^{2-}) was analyzed by ion chromatography in Seoul National University (SNU) (Gieskes et al. 1991). Headspace gas was sampled and analyzed as proposed by ODP using an HP 5890II gas chromatograph (GC) in KIGAM (Pimmel and Claypool 2001). Stable carbon isotope ratios ($\delta^{13}C$) of the headspace gas were analyzed using an isotope ratio-monitoring gas chromatograph/mass spectrometer (GC/MS) in Hokkaido University, Japan.

3. Discussion

Most organic matters belong to Type III (land-derived organic matter), which is in a good agreement with the result of core 98EEZ-3 by Lee et al (1999). However, Kim et al (2004) suggested that organic matters have originated from marine algae rather than land vascular plant using the C/N ratios and $\delta^{13}C_{org}$. Like as shown in our results, the contradiction between the Rock-Eval source characterization and C/N ratios and $\delta^{13}C_{org}$ was reported in other study areas (Meyers et al. 1996; Çagatay et al. 2001). According to their studies, such a discrepancy might be due to the heavy oxidizing of the organic matter during sinking down to the seafloor or post-deposition in the sediments.

In the case of Types II and III, organic matters reach catagenesis at T_{max} 435 ℃ (Nali et al. 2000). The analyzed T_{max} values of all the cores are lower than 435 ℃, which are the same to those of core 98EEZ-3. These results indicate that organic matter is thermally immature, i.e. it is under diagenesis in the thermal evolution. As the organic matters did not reach the categenesis, shallow methane gas could be originated by bacterial source rather than thermogenic if these organic matters are produced the shallow methane gas.

In all the cores, the sulfate concentration showed a linear depletion^with a depth. This is one of the typical characteristics when the AMO occurs (Borowski et al. 1996). Usually, this reaction that links the pore water sulfate and methane pools occurs within a local horizon (SMI), which co-consumes methane and sulfate. Therefore, in the cores, the AMO reaction has occurred. The methane concentration of the core also provides the other evidences for the AMO. Methane concentration of cores 03GHP-01, 03GHP-02 and 03GHP-04 was rapidly enriched below the SMI whereas it should be very low in core 03GHP-03 because these cores did not reach the SMI and methanogenic zone.

The $\delta^{13}C_{CH4}$ values lied within the bacterial methane region. Especially, $\delta^{13}C_{CH4}$ values in cores 03GHP-01 and 03GHP-02 were enriched in the low methane concentration than the high concentration, which is one of the typical characteristics of the AMO. These results indicate that methane has been originated from bacterial source rather than thermogenic, and the AMO occurs in cores 03GHP-01 and 03GHP-02. However, these results are very opposite to that of core 98EEZ-3. Lee and Chough (2003) assumed the origin of shallow gas to be thermogenic because $\delta^{13}C_{CH4}$ had a range of -58 to -44 ‰.

However, this conclusion has some problems on the combination of the other geochemical results such as thermal maturity of organic matter, limited sampling interval,^$C_1/(C_2+C_3)$ and SMI. Considering the SMI depth of core 03GHP-02 near the core 98EEZ-3, the analyzed interval of core 98EEZ-3 is clearly located in the methanogenic zone. And CH_4 is continually removed by the AMO, a molecular fraction occurs which relatively enriched the residual hydrocarbon gas in the higher homogeneous zone (Whiticar 1999). As a result, the $C_1/(C_2+C_3)$ ratio can decrease to values of less than 10, and thus CH_4 oxidation can lead to difficulties in the interpretation of natural gas sources. In addition, analogous to methanogenesis, the bacterial uptake of CH_4 is associated with a kinetic isotope effects (KIE) that enriches the residual CH_4 in the heavier isotope. This fractionation is most pronounced for the CH_4 carbon isotopes. Therefore, the $\delta^{13}C_{CH4}$ values for core 98EEZ-3 could be affected by the AMO.

4. References

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