

Formation of gas hydrate and carbonate nodules around active seeps of thermogenic methane at the eastern margin of Japan Sea

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A number of pockmarks and mud-mounds develop on a tongue-shaped ridge (UT04 Ridge) in the Naoetsu-oki sedimentary basin along the stress concentrated zone in the eastern margin of Japan Sea. These characteristic features suggest strong methane flux in this region. Geologic survey and sampling of sediments and waters were conducted on the ridge during the UT04 cruise of R&T/V Umitaka-(Tokyo University of Marine Science and Technology) in July-August, 2004.

Echo-sounding survey has exhibited magnificent flares of gas plume over the area. Fifteen piston cores, 5.5 m long, were deployed either on the mounds, into the pockmarks, or on a flat basin floor far from the plumes. Sediments of each core were grey or olive silty clay. Thirteen cores among 15 are made up of inter-bedded units of structure less, bioturbated beds and laminated beds. PC06 is totally made up of bioturbated unit. PC15 contained gas hydrate and carbonate nodules (ca.5cm in diameter) in intensively disturbed sediments, and the original sedimentary structures were not observed. Glove sampler dropped near PC15 recovered many carbonate nodules. PC05 contained carbonate nodules which are a little larger (ca.7cm) than PC15 and Glove samples. One piston core (PC15) near the plume successfully recovered white massive gas hydrate. On board GC analysis has revealed that the gas hydrate is largely composed of methane (methane to ethane ratio is 7738:1), though, the carbon isotopic composition of the hydrate methane (-38.9 to -39.4 permil PDB) seems to indicate thermogenic source. XRD analysis has revealed that the carbonate nodules are composed of calcite and/or aragonite, without dolomite. Microscopic observations have clearly demonstrated aragonite needles. Carbon isotopic composition of carbonates range between -10 and -30 permil PDB, which are a bit heavier than methane of gas hydrate. This may suggest that bicarbonate ion in sea water (DIC) was also used for the formation of carbonates.

Ion concentration of the interstitial waters showed that sulfate-methane interface (SMI) at the piston coring sites range between 0.5 to 3 m. These are remarkably shallow when compared with the SMI on the Blake Ridge (5.0m to 20m, Borowski et al., 1999) and Nankai Trough (4.0m to 63m; Matsumoto and Chen, 2003). Methane flux on and around the UT04 Ridge are very strong. The shallow SMI (1.5m) was observed at PC03 and PC04, both were located close to gas plumes. The existence of gas plume and the shallow SMI means that the methane flux is strongest at this part of the Ridge. SMI at PC05, close to pockmarks, was observed to be very shallow (0.5m), but this may not indicate strong methane flux. Lower water content (53.8 weight%) than other cores (over 60 weight%) seems to suggest an erosion of surface sediment.

Interstitial water chemistry of the PC03 sediments illustrated an increase of Mg, Na, K, and Cl. This is explained as the result of gas hydrate formation in semi-closed system. Gas hydrate excludes salts from the structure, causing salinity increase of ambient waters. In most cases, the anomaly would be erased by diffusive mixing of the interstitial water system. Anomalously high concentration of cations and chloride in the PC03 sediments are likely to indicate that gas hydrate formation and consumption of waters are not balanced by supply of surrounding waters. In other words, large supply of methane caused water-deficient environments in shallow sediments. We would conclude that gas hydrate is now being formed in free-gas charged sediments of the UT04 Ridge.