

U-Th radioactive disequilibrium analyses and ^{14}C dating of methane-derived carbonates, eastern margin of Japan Sea

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Gas hydrates and gas plumes have been observed off Joetsu, eastern margin of Japan Sea. Carbonate concretions occur in and on gas-hydrate bearing sediments near the plume sites, and also in gas-hydrate free sediments far away from gas plumes. Carbonates are a useful chronometer which records the timing of dissociation or formation of gas hydrate. CH_4 oxidation and SO_4^{2-} reduction and an increase in $[\text{HCO}_3^-]$ are accelerated by high CH_4 flux, which also accumulate CH_4 hydrate in surface sediments. On the other hand, massive dissociation of CH_4 hydrate also supplies CH_4 to SMI horizons in shallow sediments, to form carbonates. A number of carbonate concretions were recovered by piston cores, dredges and ROV dives from the plume sites on the mounds and around pockmarks of gas hydrate fields at Umitaka Spur (UT) and Joetsu knoll (JK) off Joetsu. Pockmarks are depressions on the seafloor and mounds are mud domes on the seafloor. Carbonate concretions are grouped into 5 types. Types A, B and C were observed on the sea floor by ROV Hyper Dolphin, and Types D and E were recovered by piston cores or MBARI push corers. Type A is a carbonate mound, 50 cm to 250 cm in diameter. They are dominated by aragonite (75 -100% aragonite and 0-25% calcite). Type B is a thin carbonate plate, 5 cm to 15 cm in thickness. They are composed of calcite (90-100% calcite and 0-10% aragonite). Type C is a nodular carbonate, 5 cm to 30 cm in diameter. Type D is an indurated and Type E is a soft to semi-indurated micro nodule, 1 mm to 1 cm in diameter. Type C, D and E are composed of either calcite, aragonite, or the mixture of calcite and aragonite. Carbon isotopic compositions of carbonates are between -5 and -32 permil PDB, suggesting a contribution from methane-derived, isotopically-light carbon. Oxygen isotopic compositions of aragonite dominant concretions are from +3.5 to +5.5 permil. Assuming that the oxygen isotope value of sea water was +0.3 permil, paleo temperatures are calculated to be unusually low between -1.2 and -8.2 degree. Considering that the bottom water temperatures was around 0.3 degree throughout the glacial-interglacial periods, above calculations are likely to suggest that the oxygen isotope value of aragonite precipitating fluids were +3.3 to +3.4 permil. The heavy oxygen water have been possibly derived from the dissociation of gas hydrates, therefore aragonite dominant concretions indicate the past gas hydrate dissociation events. To the contrary, the oxygen isotopic compositions of calcite dominant concretions are +1.5 to +4.8 permil, suggesting that these concretions were precipitated either under higher temperature or O-18 depleted waters. This indicates that calcite dominant concretions are thought to have been formed in light-oxygen waters, probably the residual waters of gas hydrate formation.

U-Th radioactive disequilibrium analyses indicate that carbonates from UT were precipitated between 12 and 62 ka, centered around 20 ka (Watanabe and Nakai, 2006). Ages of carbonate concretions from UT were also measured by ^{14}C dating. These ages were older than U-Th dating, indicating the effect of dead carbon. On the other hand, carbonate concretions from JK were precipitated between 11 and 390 ka, in which the age of Type A was very old. Type A is observed in only JK. Considering that the carbonate precipitation was triggered and facilitated by strong methane seeps, observed U-Th ages of carbonates seem to indicate that methane venting was strongly enhanced during the Last Glacial Maximum at UT spur, whereas the carbonates from JK knoll seem to be older than UT-spur. Implying that large gas hydrate dissociation occurred older than LGM.