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## In-situ Micro-structural Studies of Gas Hydrate Formation in Sedimentary Matrices

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The formation process of gas hydrates in sedimentary matrices is of crucial importance for the physical and transport properties of the resulting aggregates. This process has never been observed in-situ with sub-micron resolution. Here, we report on synchrotron-based micro-tomographic studies by which the nucleation and growth processes of gas hydrate were observed in different sedimentary matrices (natural quartz, glass beds with different surface properties, with and without admixtures of kaolinite and montmorillonite) at varying water saturation. The nucleation sites can be easily identified and the growth pattern is clearly established. In under-saturated sediments the nucleation starts at the water-gas interface and proceeds from there to form predominantly isometric single crystals of  $10-20\mu m$  size. Using a newly developed synchrotron-based method we have determined the crystallite size distributions (CSD) of the gas hydrate in the sedimentary matrix confirming in a quantitative and statistically relevant manner the impressions from the tomographic reconstructions. It is noteworthy that the CSDs from synthetic hydrates are distinctly smaller than those of natural gas hydrates [1], which suggest that coarsening processes take place in the sedimentary matrix after the initial hydrate formation. Understanding the processes of formation and coarsening may eventually permit the determination of the age of gas hydrates in sedimentary matrices [2], which are largely unknown at present. Furthermore, the full micro-structural picture and its evolution will enable quantitative digital rock physics modeling to reveal poroelastic properties and in this way to support the exploration and exploitation of gas hydrate resources in the future.

[1] Klapp S.A., Hemes S., Klein H., Bohrmann G., McDonald I., Kuhs W.F. Grain size measurements of natural gas hydrates. Marine Geology 2010; 274(1-4):85-94.

[2] Klapp S.A., Klein H, Kuhs W.F. First determination of gas hydrate crystallite size distribution using highenergy synchrotron radiation. Geophys.Res.Letters, 2007 ; 34 : L13608, DOI:10.1029/2006GL029134