

Electronic Supplementary Material (ESI) for RSC Advances.

Table S1 Main parameters of macroscopic experiment for hydrate formation in sediments

Median granularity	Initial porosity	Hydrate saturation
150 μm	42.11%	35.61%
87 μm	41.91%	34.32%
55 μm	38.13%	36.25%
38 μm	38.66%	35.01%
24 μm	38.04%	33.22%
13 μm	37.84%	32.43%

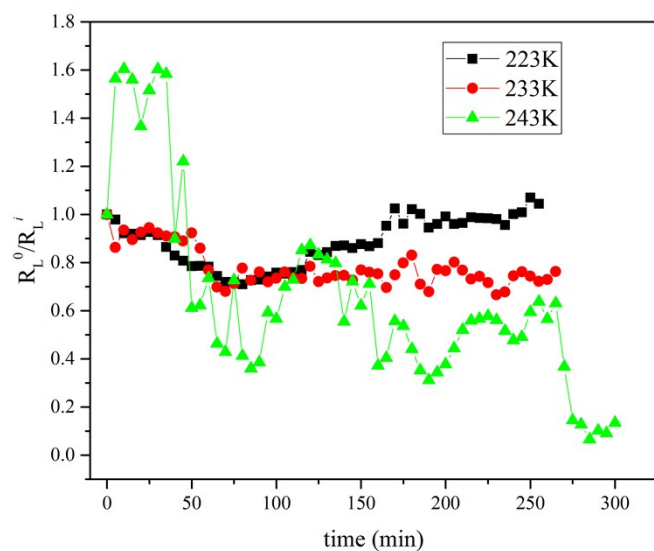


Fig.S1 Time-resolved integrated Raman intensity for the C-H stretching mode of CH_4 in large cages (2902 cm^{-1}) in bulk hydrate.

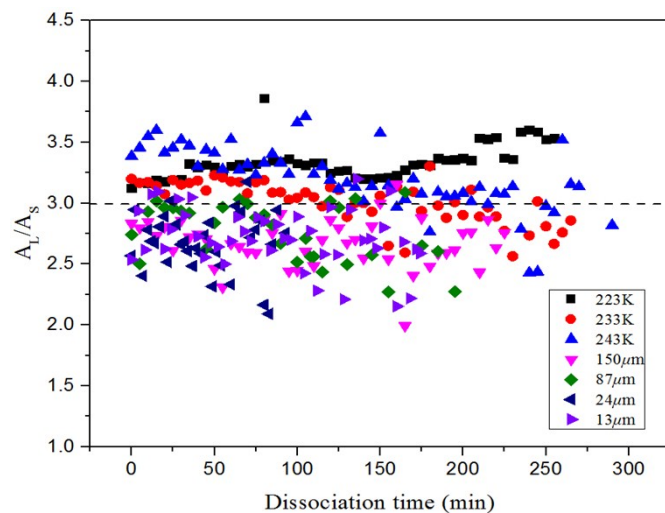


Fig. S2 Integrated intensity ratio of large to small cages for methane hydrate (A_L/A_S) during dissociation.

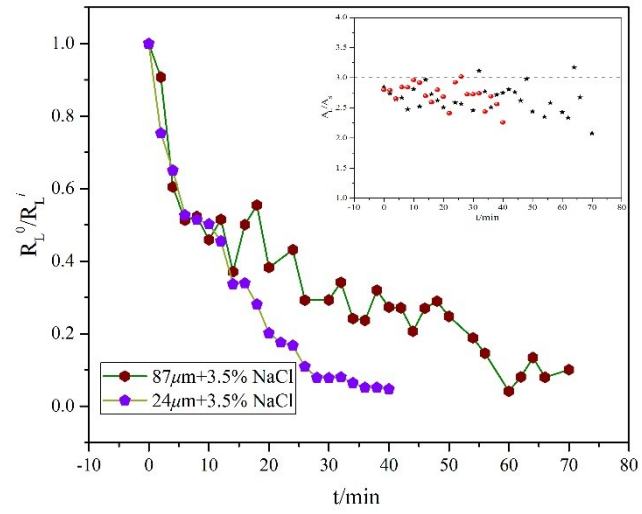


Fig. S3 Changes in R_L^0/R_L^i and AL/AS with time during hydrate decomposition in natural sand with 3.5% NaCl.