

## Electronic Supplementary Information for

# Insights into the Climate-Driven Evolution of Gas Hydrate-Bearing Permafrost Sediments: Implications for Prediction of Environmental Impacts and Security of Energy in Cold Regions

Mehrdad Vasheghani Farahani<sup>1</sup>, Aliakbar Hassanpouryouzband<sup>1,2</sup>, Jinhai Yang<sup>1,\*</sup>, Bahman Tohidi<sup>1</sup>

<sup>1</sup> Hydrates, Flow Assurance & Phase Equilibria Research Group, Institute of GeoEnergy Engineering, Heriot-Watt University, Edinburgh, EH14 4AS, United Kingdom

<sup>2</sup> School of Geosciences, University of Edinburgh, Grant Institute, West Main Road, Edinburgh, EH9 3JW, United Kingdom

Detailed experimental conditions and measured elastic wave velocities and ETC values at different methane hydrate saturations are available at Mendeley Data, <http://dx.doi.org/10.17632/xj4bhkw9sz.1>.

## Contents of this file

**Text S1.** Providing the detailed procedure followed to obtain the experimental measurement errors of the elastic wave velocities and ETC measurements.

**Table S1.** Values of density ( $\rho_h$ ) and hydration number ( $\gamma$ ) of methane hydrates together with methane specific volume ( $\bar{V}_g$ ) used to calculate the saturation of each phase for the measured elastic waves velocity data points.

**Table S2.** Values of density ( $\rho_h$ ) and hydration number ( $\gamma$ ) of methane hydrates together with methane specific volume ( $\bar{V}_g$ ) used to calculate the saturation of each phase for the measured ETC data points.

### Text S1.

The experimental errors of the elastic wave velocities and ETC measurements could be determined by the following equation:

$$E = \sqrt{E_{Sys.}^2 + E_{Rand.}^2} \quad (S1)$$

where  $E$  is the relative experimental measurement error, and  $E_{Sys.}$  and  $E_{Rand.}$  stand for the systematic and random errors.

The elastic wave velocities were measured indirectly by measuring the travel time ( $t$ ) of the elastic waves and the propagation distance ( $L$ ). Therefore, the measurement error could be obtained via the following equation:

$$E_i = \sqrt{\left(\frac{\Delta L}{L}\right)^2 + \left(\frac{\Delta t_i}{t_i}\right)^2}, i:P/S \quad (S2)$$

where  $\Delta L$  and  $\Delta t$  are the absolute experimental measurement errors of the length and travel time. Thus, the experimental measurement errors of the elastic wave velocities measurements are calculated using the following equation:

$$E_i = \sqrt{\left(\frac{\Delta L_{Sys.}}{L}\right)^2 + \left(\frac{\Delta t_{i, Sys.}}{t_i}\right)^2 + \left(\frac{\Delta L_{Rand.}}{L}\right)^2 + \left(\frac{\Delta t_{i, Rand.}}{t_i}\right)^2}, i:P/S \quad (S3)$$

The propagation distance was measured using a LVDT (ABSOLUTE Digimatic IP42 Bore Gauge Digital Indicator (ID-C) - 543 Series, Mitutoyo UK Ltd) with a nominal systematic error ( $\Delta L_{Sys.}$ ) of 0.003 mm. The random error of the length measurement ( $\Delta L_{Rand.}$ ) could be neglected because the reading did not change during the measurement. The travel time was measured using digital oscilloscope (DSO-2102M, Link Instruments Ltd). The random measurement error of the travel time ( $\Delta t_{i, Rand.}$ ) is mainly caused by the uncertainty of identification of the first arrival of the acoustic signal. According to our experimental examination, the random error is estimated about 1  $\mu$ s for P-wave (the first arrival can be easily picked up) and up to 5% for S-wave (the first arrival of S-wave is often difficult to determine, particularly for low hydrate saturation at unfrozen conditions). The measurement accuracy of the DSO is  $\pm 0.01\%$  of the time base i.e. 10 ns, which can be neglected compared to the random error. Having the instantaneous propagation distance and travel time, the experimental measurement error for both P- and S-wave velocities were calculated and reported together with the measured experimental value.

ETC was directly measured using a thermal conductivity measurement system (TPSYS02, Hukseflux, The Netherlands). The expected accuracy ( $E_{Sys.}$ ) and repeatability ( $E_{Rand.}$ ) given by the manufacturer are 3%+0.02 W/m.K and 2%, respectively.

**Table S1.** Values of density ( $\rho_h$ ) and hydration number ( $\mathcal{V}$ ) of methane hydrates together with methane specific volume ( $\mathcal{V}_g$ ) used to calculate the saturation of each phase for the measured elastic waves velocity data points.

No.	$P_{\text{Pore}}$ (MPa)	$T$ (°C)	Density	Hydration Number	Specific Volume
			g/cc	(-)	cc/mol
$S_h = 0.00$					
1	3.6	3.1	-	-	584.56
2	2.8	0.2	-	-	756.41
3	2.6	-0.5	-	-	816.19
4	2.3	-3.1	-	-	919.05
$S_h = 0.09$					
1	3.4	3.0	0.912	5.796	621.70
2	2.7	0.5	0.911	5.767	787.53
3	2.4	-0.5	0.911	5.757	888.79
4	2.2	-3.0	0.911	5.761	963.84
$S_h = 0.20$					
1	3.4	3.0	0.912	5.796	621.70
2	2.7	0.5	0.911	5.767	787.53
3	2.4	-0.4	0.911	5.757	889.19
4	2.3	-3.0	0.911	5.761	919.46
$S_h = 0.31$					
1	3.4	3.0	0.912	5.796	621.70
2	2.7	0.5	0.911	5.767	787.53
3	2.4	-0.6	0.911	5.758	888.39
4	2.3	-3.0	0.911	5.761	919.46
$S_h = 0.44$					
1	3.4	3.0	0.912	5.796	621.70
2	2.7	0.6	0.911	5.768	787.89
3	2.4	-0.5	0.911	5.757	888.79
4	2.3	-3.0	0.911	5.761	919.46
$S_h = 0.59$					
1	3.4	3.0	0.912	5.796	621.70
2	2.7	0.5	0.911	5.767	787.53
3	2.4	-0.5	0.911	5.757	888.79
4	2.2	-3.1	0.911	5.761	963.40

**Table S2.** Values of density ( $\rho_h$ ) and hydration number ( $\mathcal{V}$ ) of methane hydrates together with methane specific volume ( $\mathcal{V}_g$ ) used to calculate the saturation of each phase for the measured ETC data points.

No.	$P_{\text{Pore}}$ (MPa)	$T$ (°C)	Density	Hydration Number	Specific Volume
			g/cc	(-)	cc/mol
$S_h = 0.00$					
1	3.5	3.1	-	-	602.74
2	3.4	0.6	-	-	614.56
3	3.4	-0.6	-	-	610.97
4	3.4	-0.7	-	-	610.68
5	3.3	-1.0	-	-	629.91
6	3.3	-1.6	-	-	628.07
7	3.9	-2.0	-	-	521.94
8	3.9	-2.3	-	-	521.13
9	3.9	-3.3	-	-	518.43
10	3.7	-9.7	-	-	531.29
$S_h = 0.20$					
1	3.4	2.6	0.91	5.79	620.52
2	2.7	0.5	0.91	5.77	787.53
3	2.5	-0.5	0.91	5.76	851.03
4	2.3	-2.8	0.91	5.76	920.29
5	1.8	-9.7	0.91	5.77	1156.02
$S_h = 0.29$					
1	3.5	2.8	0.91	5.79	601.87
2	2.7	0.5	0.91	5.77	787.53
3	2.5	-0.6	0.91	5.76	850.64
4	2.3	-3.0	0.91	5.76	919.46
5	1.8	-9.7	0.91	5.77	1156.02
$S_h = 0.39$					
1	3.7	2.9	0.91	5.80	566.80
2	2.8	0.5	0.91	5.77	757.46
3	2.5	-0.4	0.91	5.76	851.42
4	2.3	-2.8	0.91	5.76	920.29
5	2.0	-9.7	0.91	5.77	1034.33
$S_h = 0.51$					
1	3.5	2.8	0.91	5.79	601.87
2	2.8	0.6	0.91	5.77	757.81
3	2.6	-0.4	0.91	5.76	816.55
4	2.3	-2.9	0.91	5.76	919.88
5	2.0	-9.7	0.91	5.77	1034.33