**Supplementary Table 1.** Lattice parameters from the Rietveld refinements and hydrate/ice phase fractions from the Bragg patters obtained from the total scattering instrument (NOMAD) and from high resolution neutron powder diffraction (POWGEN) in previous work.

| Feed gas composition | Lattice parameter (Å) | Phase fraction ice |
|----------------------|-----------------------|--------------------|
|                      | NOMAD (this work)     | POWGEN (Everett et al.) | NOMAD (this work) |
| 100% CH$_4$          | 11.830(2)             | 11.83210(8)         | 0.10(1)           |
| 50% CH$_4$ 50% CO$_2$| 11.8244(3)            | 11.82487(8)         | 0.21(1)           |
| 100% CO$_2$          | 11.8192(7)            | 11.82216(9)         | 0.13(1)           |

**Supplementary Table 2.** Hydrate cage occupancies high resolution neutron powder diffraction (POWGEN) in previous work.$^1$

| Feed gas composition | Large cage occupancy | Small cage occupancy | Total composition % filled % |
|----------------------|----------------------|----------------------|-----------------------------|
|                      | CH$_4$ | CO$_2$ | CH$_4$ | CO$_2$ | CH$_4$ | CO$_2$ | Total cages |
| 100% CH$_4$          | 0.73(3) | -      | 0.93(5) | -      | 79(4)  | -      | 79(4)      |
| 50% CH$_4$           | 0.08(3) | 0.77(3) | 0.54(4) | 0.21(2) | 20(3)  | 63(1)  | 83(5)      |
| 50% CO$_2$           | -      | 1.00(6) | -      | 1.00(4) | -      | 100(6) | 100(6)     |

$^1$ References and sources of data should be provided.
Supplementary Figure 1. Comparison of mixed CH₄-CO₂ hydrate PDF data with the ice impurity and with ice subtracted shows that the impurity has little effect on the PDF.

Supplementary Table 3. Summary of physical constraints applied in the RMC simulations.

| Closest approach pair distances (Å) | O-O | O-D | O-C | O-H | D-D | D-C | D-H | C-C | C-H | H-H |
|-----------------------------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
|                                   | 2.2 | 0.85| 1.1 | 2.5 | 1.2 | 2.5 | 2.0 | 5.0 | 1.0 | 1.5 |
| Maximum distance per move (Å)     |     |     |     |     |     |     |     |     |     |     |
|                                   | O   | D   | C   | H   |
|                                   | 0.05| 0.05| 0.05| 0.05|
| Distance window for O-D hydrogen bonded pairs (Å) | Minimum distance | Maximum distance |
|                                           | 1.6 | 2.25 |

Supplementary References

1 Everett, S. M. et al. Insights into the structure of mixed CO₂/CH₄ in gas hydrates. *American Mineralogist* **100**, 1203-1028 (2015).
