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Supporting Information for:

Molecular dynamics study of guest-host hydrogen bonding in alcohol clathrate hydrates

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Rotational relaxation of guest molecules

The relaxation times of the rotational motion of the guest molecules are computed by fitting $M_2(t)$ to a sum of exponential functions corresponding to different intramolecular geometry changes and rotation of the molecules as follows,

$$M_2(t) = \sum_{k=1}^{n} A_k \exp\left(-\frac{t}{\tau_k}\right).$$

Fit parameters for the molecular rotations of the guest molecules in Figure 6 at different temperatures in the hydrates are given in Table S1. To fit the decay of the $M_2(t)$ related with molecular rotations, one to four exponential functions were used. The multiple relaxation times in the $M_2(t)$ decay reflect the complicated motions and environment in the cage due to the guest-host hydrogen bonding, especially for the $5^{12}6^4$ cages in the sII hydrate.

Table S1: Relaxation times in the $M_2(t)$ decay

Guest (*Temp.*)	$A1$	t_1 / ps	$A2$	t_2 / ps	$A3$	t_3 / ps	$A4$	t_4 / ps
3-methyl-1-butanol (113 K)	0.048	0.327	0.018	244	0.934	112567	-	-
3-methyl-1-butanol (200 K)	0.120	0.923	0.080	103	0.213	1216	0.585	35527
3-methyl-1-butanol (270 K)	0.209	0.600	0.185	25.1	0.360	289	0.245	4023
2-methyl-2-butanol (113 K)	0.102	0.985	0.894	256066	-	-	-	-
2-methyl-2-butanol (200 K)	0.243	0.667	0.754	69315	-	-	-	-
2-methyl-2-butanol (270 K)	0.396	0.680	0.258	1238	0.342	23403	-	-