Supporting information available

The exchange model

A comparison between two-site exchange models, using two different quadrupole coupling constants of 51 and 30 kHz, was conducted in order see the influence of internal motions of the guest molecules on the resulting ^{2}H MAS NMR spectra. As mentioned in the main text, a quadrupole coupling constant of 51 kHz is a typical value for a CD_3 group in an organic molecule, which can be scaled furthermore to 30 kHz when an additional anisotropic motion occurs inside the examined molecule, as seen in the case of methionine-d_3 and ala-ala-$d_3 - OH$. Simulated ^{2}H MAS spectra for $k_{bf} = 10^3$ and $3.2 \cdot 10^5\ s^{-1}$, and varying P_b values, for $\nu_r = 6kHz$, $\nu_Q = 51$ (a,c), and 30 kHz (b,d) are presented in fig. 1S.

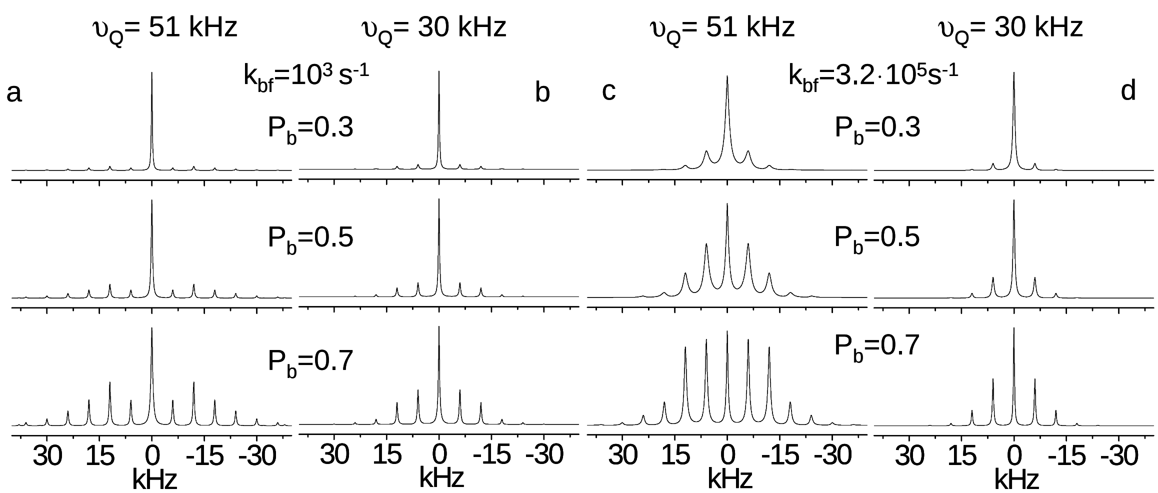

Figure 1S

Two parameters were used in the comparison between calculated and experimental spectra, i.e. the FWHH of the centerband of the ^{2}H MAS spectra, and the ratio between the intensities of the first sideband (I_1) and centerband (I_o). Contours presenting these ratios as a function of the $\{\log(k_{bf}), P_b\}$ parameters for $\nu_Q = 51\ kHz$ (a) and $\nu_Q = 30\ kHz$ (b), respectively, with $\nu_r = 6\ kHz$, as well as our experimental data points are shown in fig. 2S.

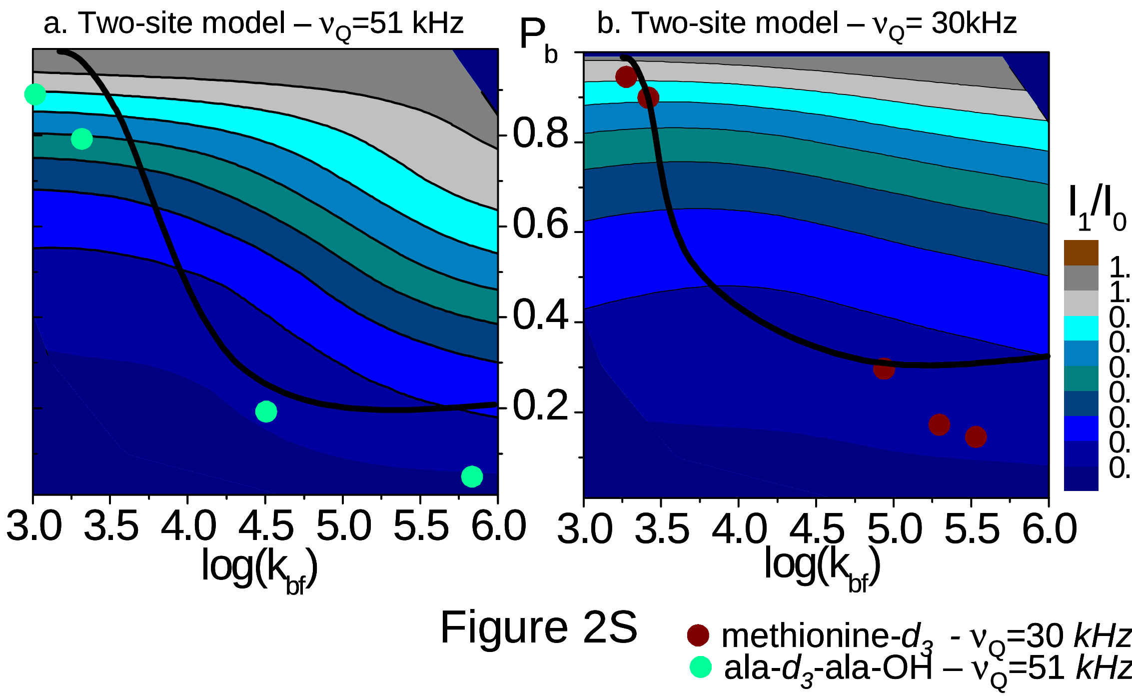

Figure 2S

Guest molecules inside SBA-15

As mentioned in the main text, the ^{2}H MAS NMR spectra of the guest molecules can be decomposed into two spectral components a narrow sideband pattern and a broad one. Since the narrow components describe guest molecules that are bound to the surface wall, the broad components correspond to guest molecules that participate in an exchange process and therefore they change their shape during the drying process. These dynamic sideband spectra are analyzed using the two-site exchange model, and are compared to calculated ^{2}H MAS spectra to retrieve the average dynamic parameters $P_b(p)$ and $k_{bf}(p)$. Fig. 3S. shows the experimental ^{2}H MAS spectra of methionine-d_3 in a sample of SBA-15 at different p values in (a). Each spectrum is amplified to show its different components, and the multiplication factors are chosen with respect to the $p = 11$ spectrum. The amplification causes a truncation at the top of part of the spectra. In (b) and (c) the narrow- and broad-line components are shown after deconvolution of the experimental ^{2}H MAS spectra.

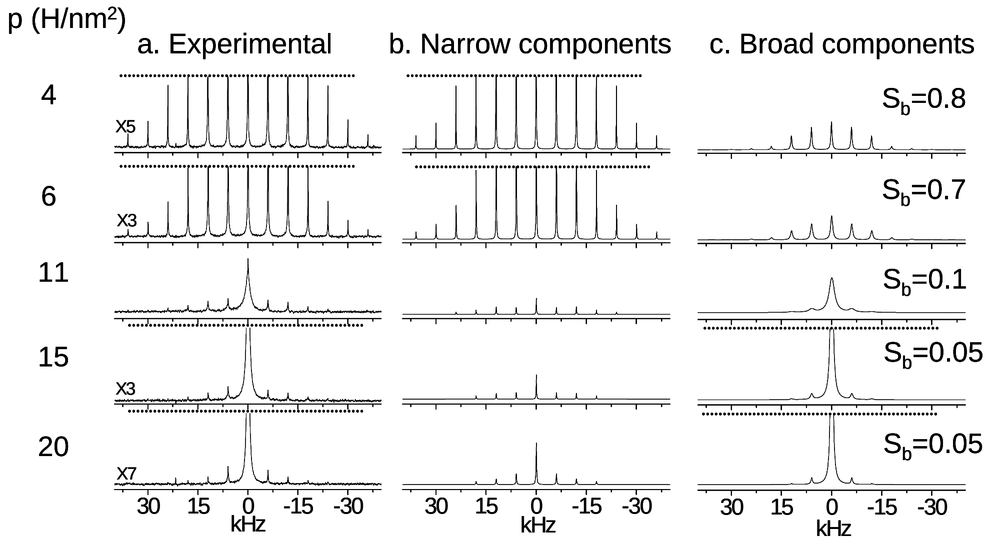

Figure 3S

The dynamic components of the temperature-dependent ^{2}H spectra of a sample of methionine-d_3 inside SBA-15 with $p = 11$ were analyzed using the two-site exchange model with $\nu_Q = 30\ kHz$. The temperature dependence of $S_b(T)$(squares), $P_b(T)$(circles), and $\log(k_{bf}(T))$ are shown in fig. 4S. (a) The lines are only for eye guidance and to demonstrate the possible similarity of the T dependences of S_b and P_b. The straight line in (b) corresponds to an activation energy of 7 $kcal/mol$, assuming Arrhenius temperature dependence.

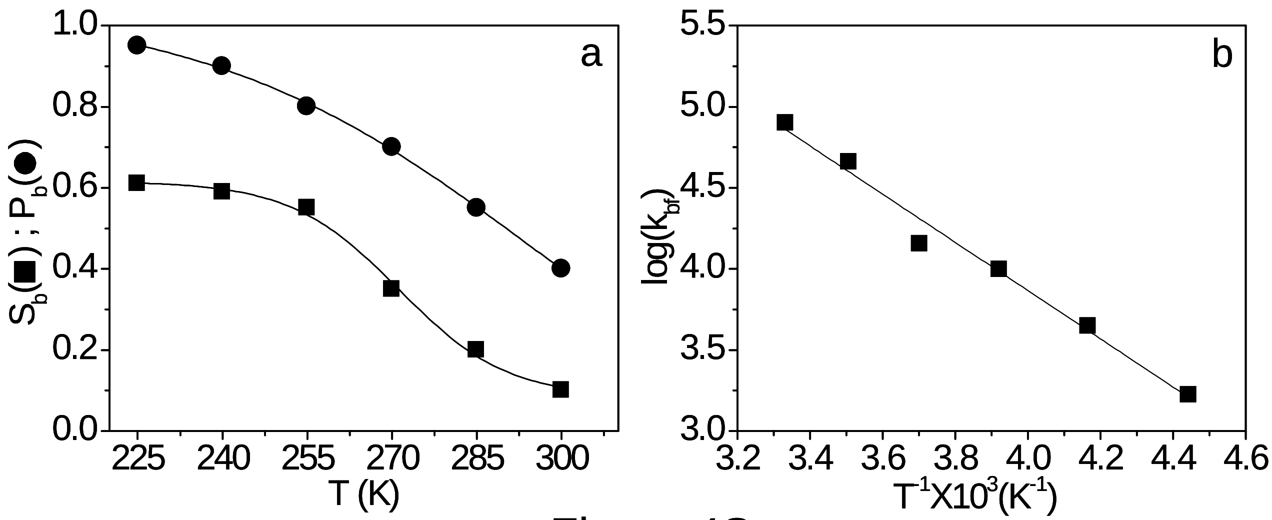

Figure 4S

Fig. 5S presents a schematic illustration of the possible binding of surface-bound methionine-d_3 in (a), and ala-d_3-ala$-OH$ in (b), with respect to the surface silanol. These orientations can explain the different quadrupole coupling constants observed in the experiments.

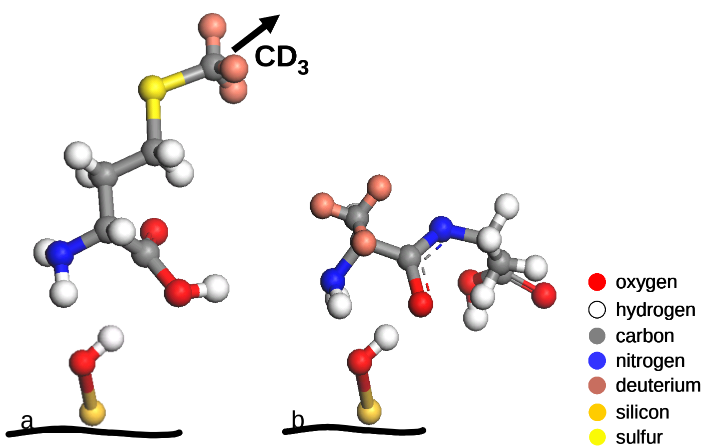

Figure 5S

Fig. 6s presents the total integrated intensity of ^{1}H spectra of a sample of SBA-15 loaded with ala-d_3-ala$-OH$ with p = 15 at different temperatures, with the relevant spectra on the right. The relative intensity of the different line components is also shown. The total integrated intensity of these spectra is reduced when lowering the temperature mainly as a result of a reduction in the $x > 3$ clusters, which is accompanied by a small increase in the $x \leq 3$ clusters. Thus, the larger clusters and bulk water start to solidify and the relative amount of smaller clusters is maintained or even increases. However, the spectrum at the lowest temperature still reflects the significant contribution of clusters with $x > 3$.

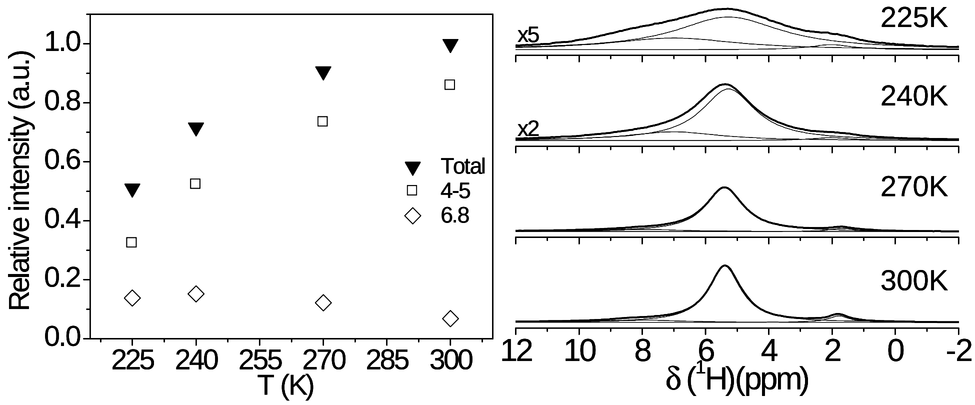

Figure 6S

The dynamic components of the temperature-dependent ^{2}H spectra of SBA-15 samples loaded with ala-d_3-ala-OH at $p = 15$ were analyzed using the two-site exchange model with $\nu_Q = 51kHz$. The temperature dependence of $S_b(T)$(squares), $P_b(T)$(circles) and $\log(k_{bf}(T))$ are shown in fig. 7S.

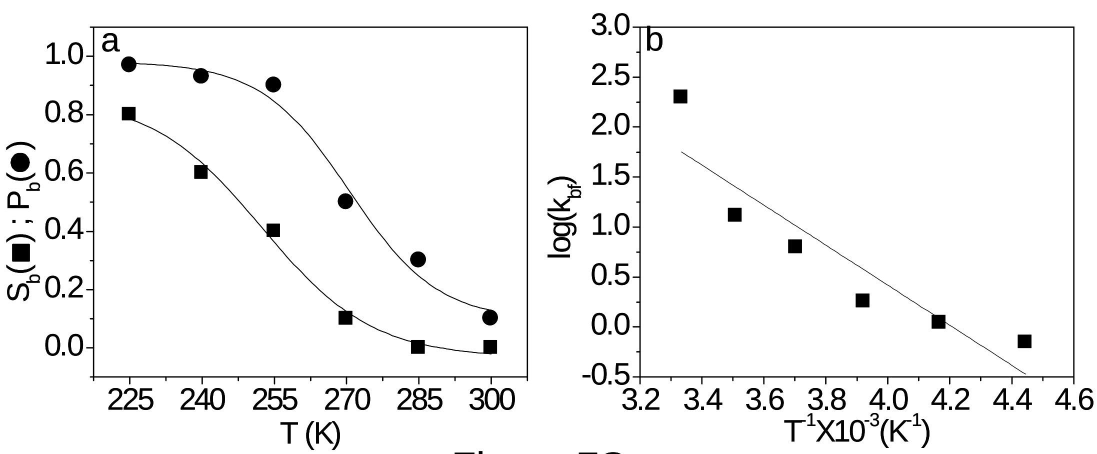

Figure 7S