

# **On the unusual temperature dependence of kaolinite intercalation capacity for N-methylformamide**

## **ELECTRONIC SUPPLEMENTARY MATERIAL**

### **Figures S1-S7**

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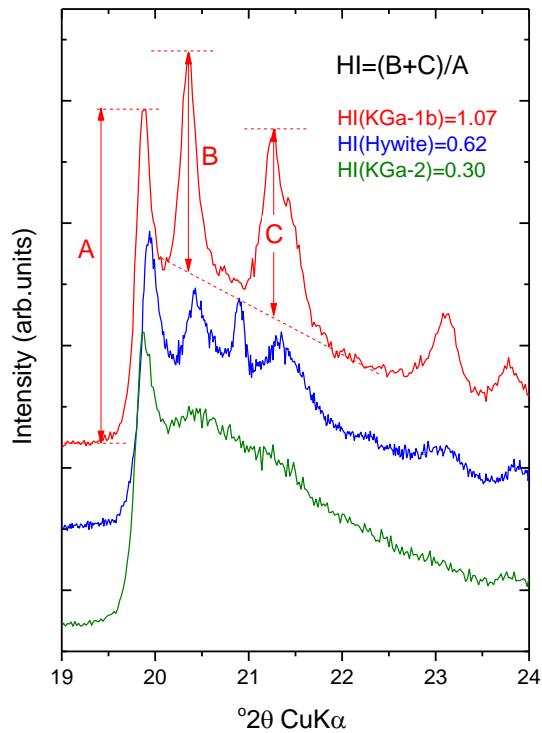


Fig. S1. Detail of the XRD patterns of KGa-1b, Hywite, and KGa-2 indicating differences in stacking order, expressed by the Hinckley index (HI).

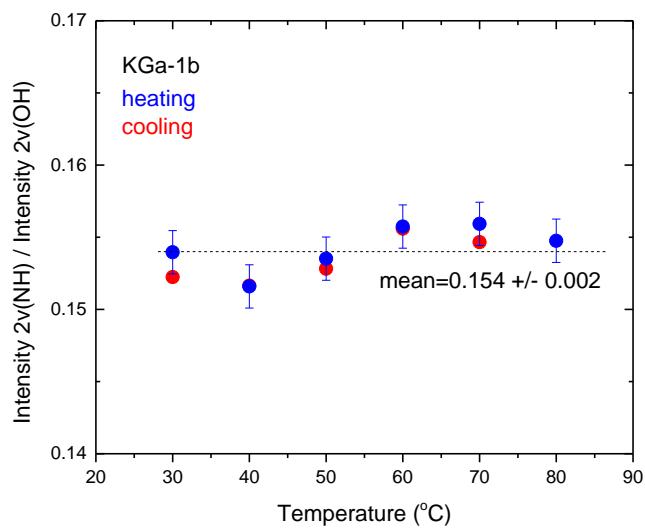


Fig. S2. Relative 2<sup>nd</sup> derivative amplitude of the 2vNH and 2vOH stretching overtones of NMF-intercalated kaolinite KGa-1b as a function of temperature. The corresponding spectra are shown in Fig. 2

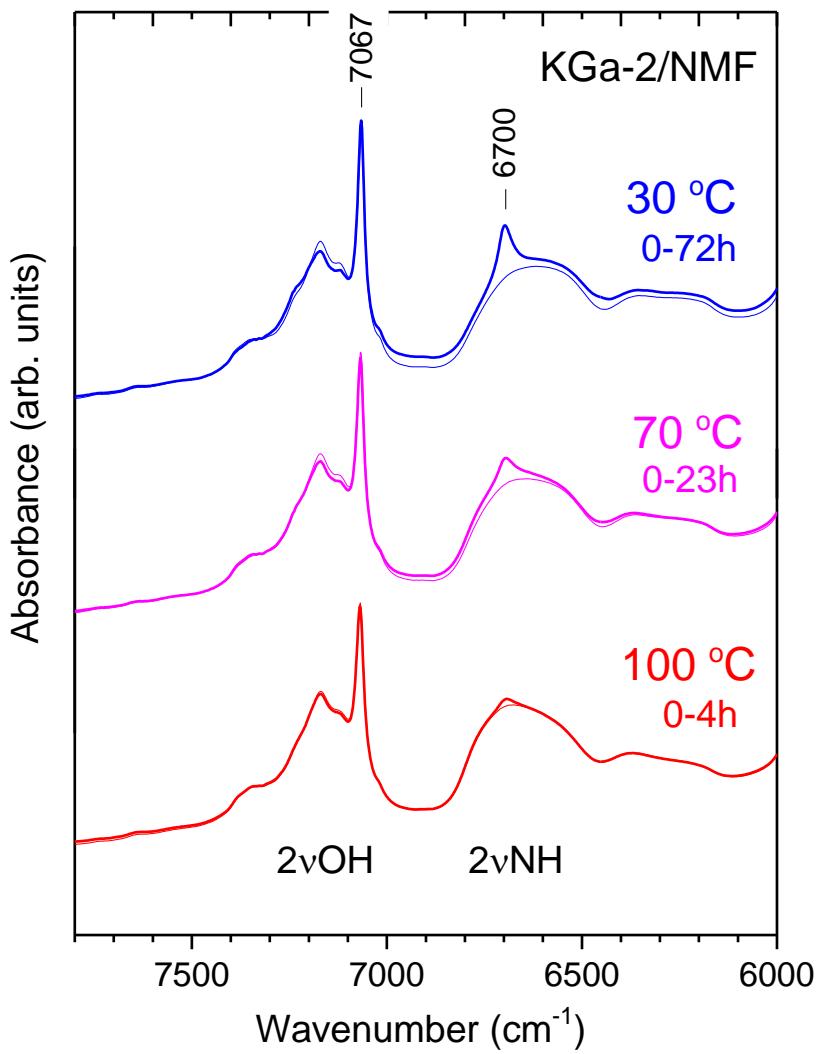


Fig. S3. First (thin lines) and last (thick lines) NIR absorbance spectra of the KGa-2/NMF real-time kinetic series at 30°C (blue), 70°C (magenta), and 100°C (red). Each pair of spectra is offset for clarity. The spectra are from the runs presented in Fig. 4.

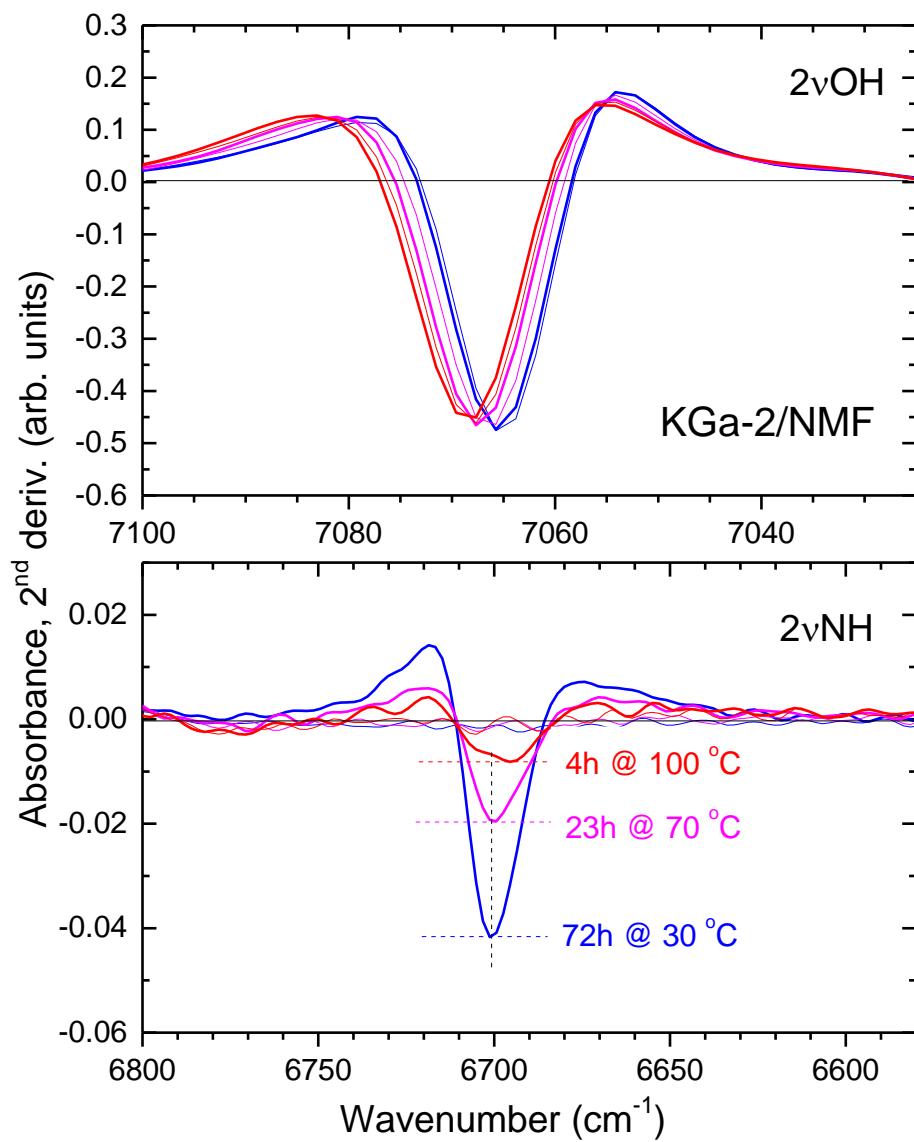


Fig. S4. 2<sup>nd</sup> derivatives of the spectra in Fig. S3, following vector normalization over the 2vOH range (7100-7025  $\text{cm}^{-1}$ , upper panel, c.f. Fig. 3) to desensitize the intensity of the 2vNH mode (6700  $\text{cm}^{-1}$ , lower panel) from irrelevant temperature effects. Lines and colors are as in Fig. S3.

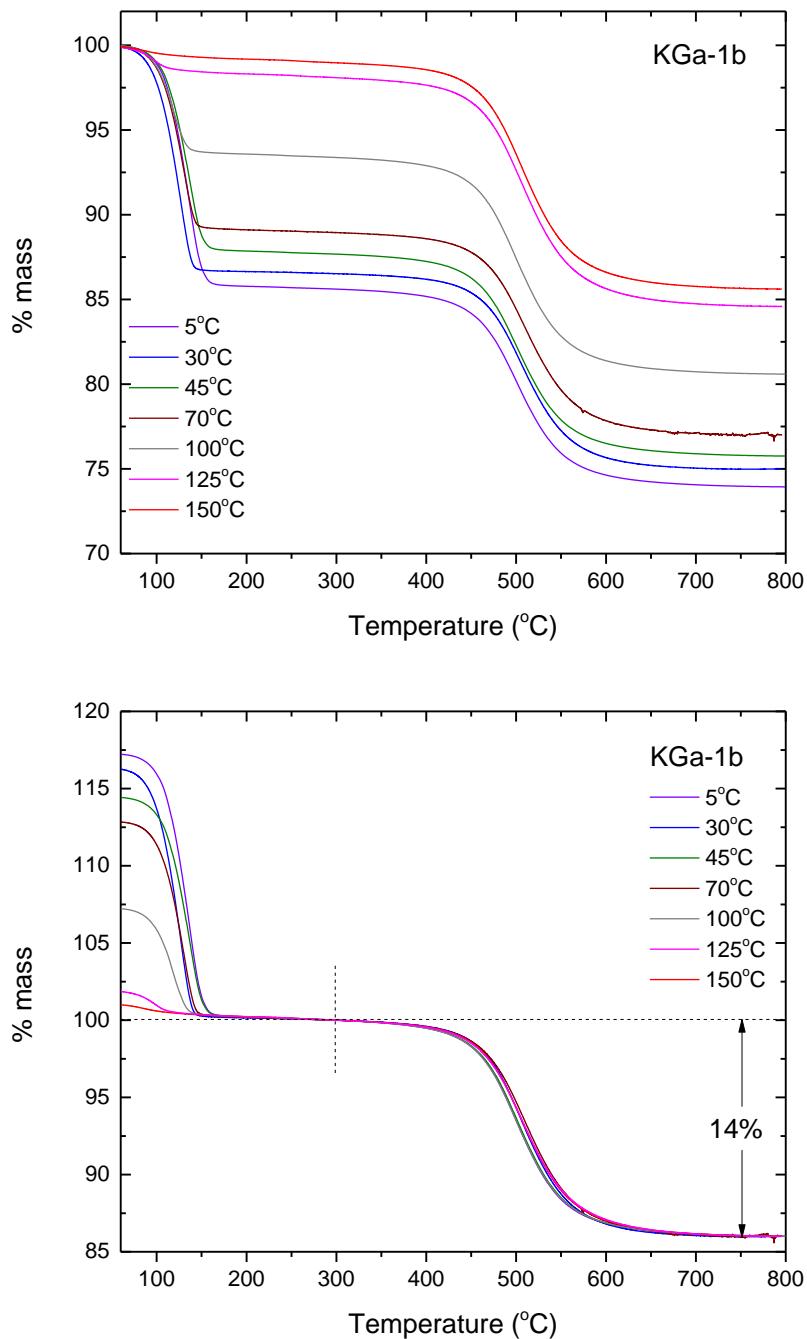


Fig. S5 Mass loss TGA traces of KGa-1b, pre-intercalated with NMF at various temperatures and rinsed with dioxane to remove excess liquid. Both raw (upper) and normalized (lower) data are depicted. The traces are normalized by dividing the raw data with the value at 300°C and scaling the dehydroxylation loss in the 300–800°C to match that of pure kaolinite (~14%). The mass loss below 300°C is assigned to the amount of previously intercalated NMF per 100 g of pure kaolinite. Full uptake of two NMF molecules per unit cell,  $\text{Al}_4\text{Si}_4\text{O}_{10}(\text{OH})_8$ , corresponds to 22.9 g NMF per 100 g kaolinite.

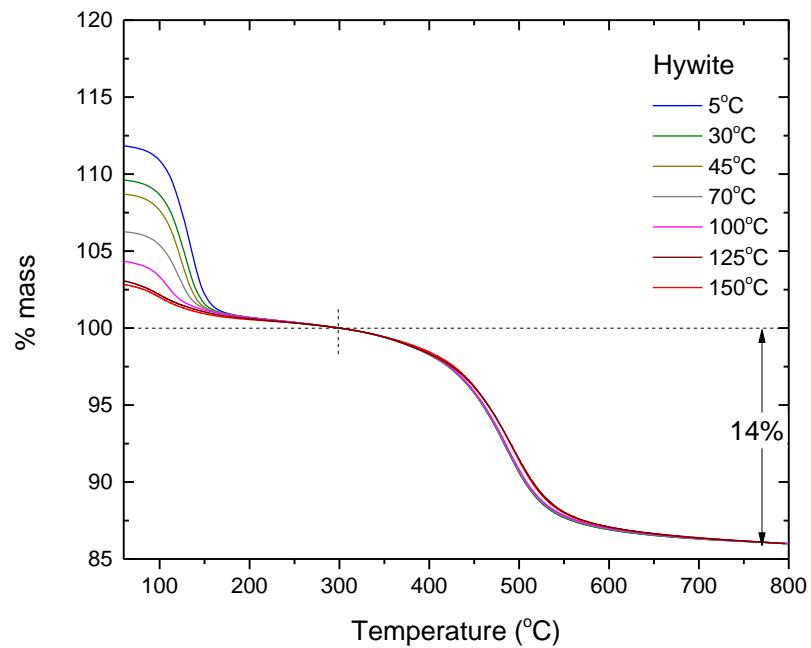
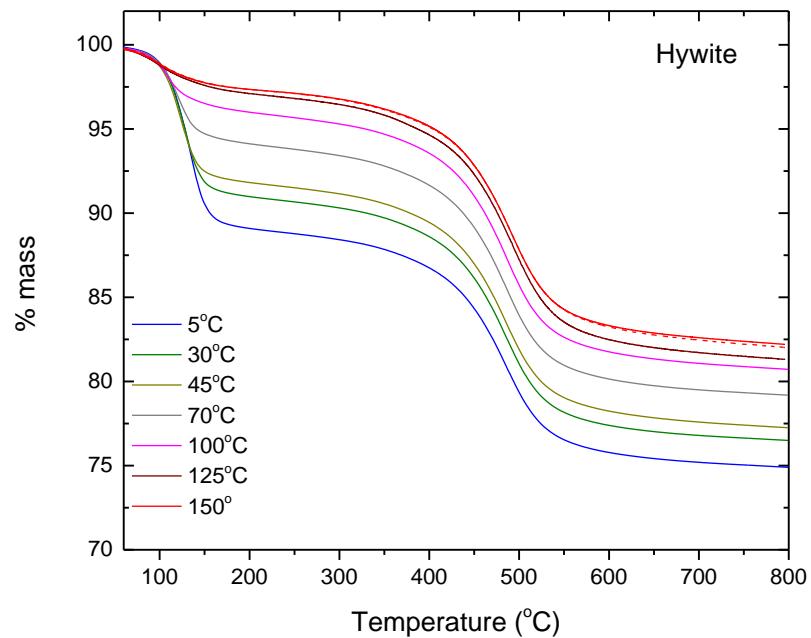


Fig. S6 Same as Fig. S5 for Hywite

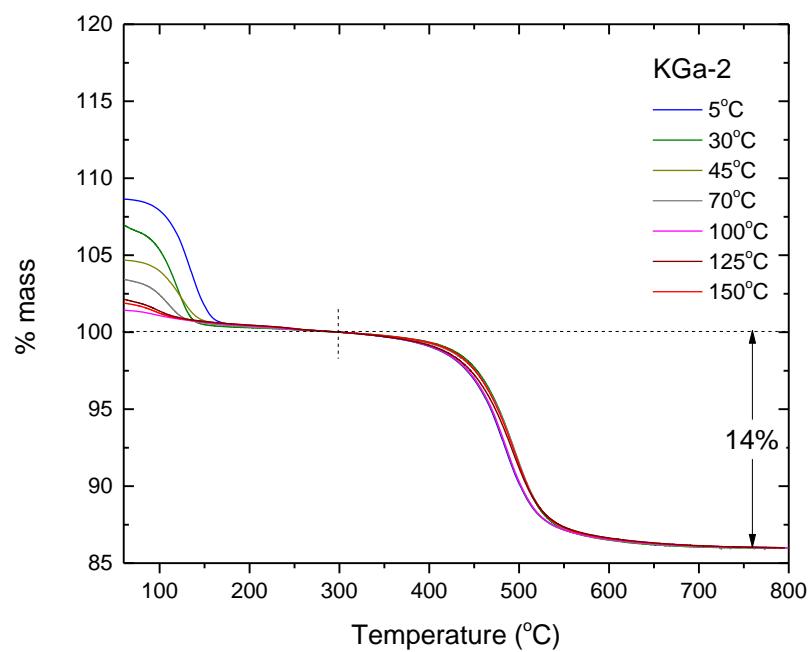
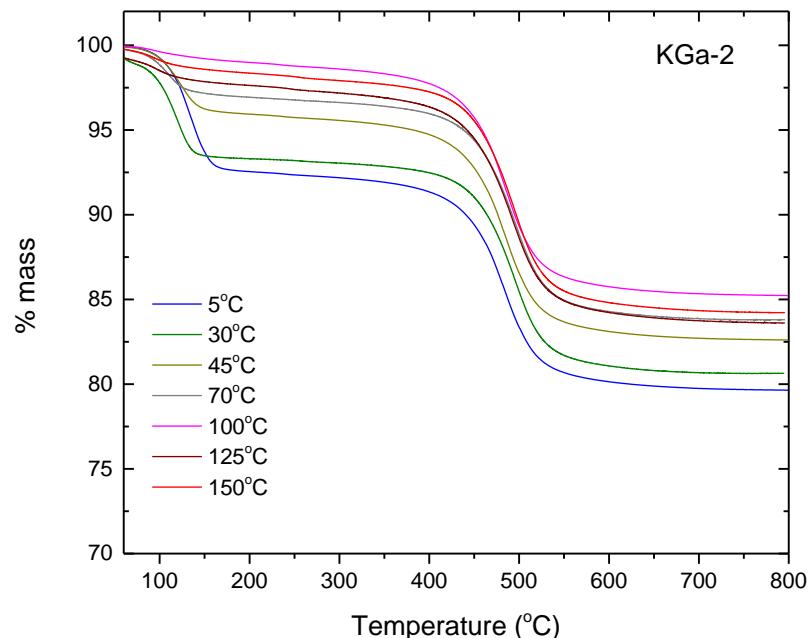


Fig. S7 Same as Fig. S5 for KGa-2