Supplementary Material (ESI) for CrystEngComm
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The Influence of the Guest Ion on Synthesis and Sorption Properties of an Open Framework Lanthanide Tetrakisphosphonate

*Monika Plabst, Ralf Köhn , Thomas Bein**

Department of Chemistry and Biochemistry, University of Munich, Butenandtstr.11, 81377 Munich, Germany

Supporting information 1: Additional SEM micrographs

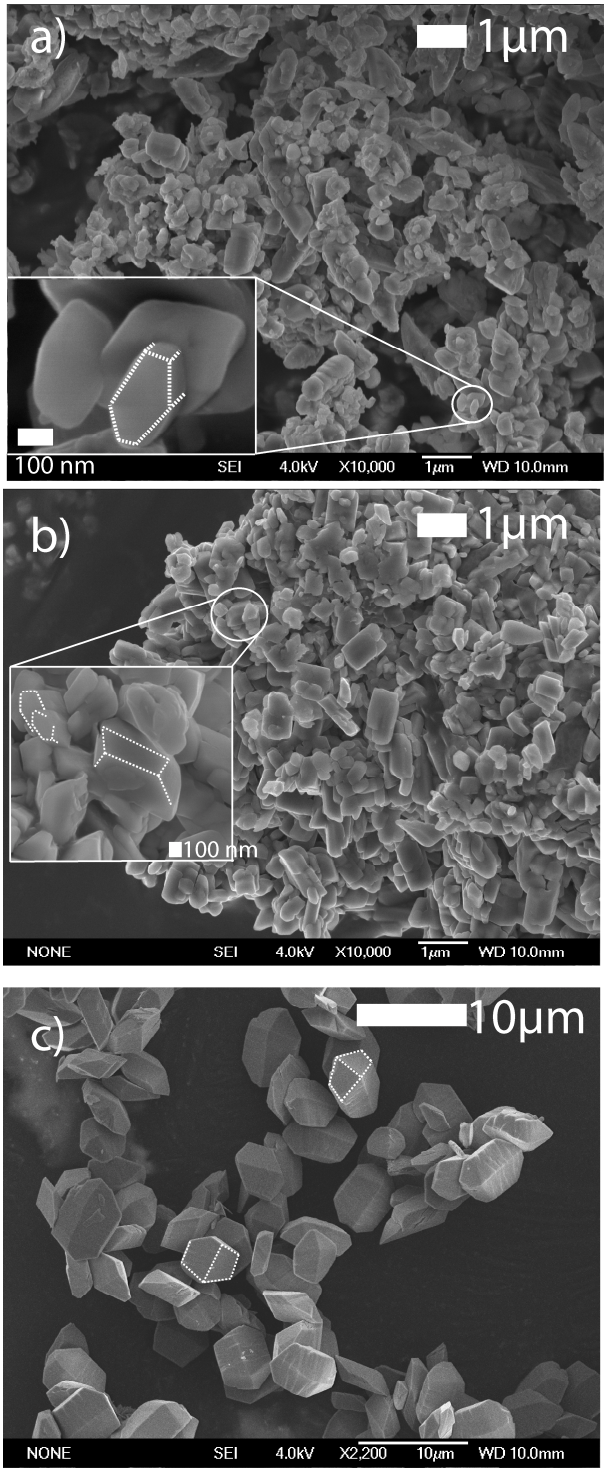

Figure S1: SEM micrographs of a) **KLa(H$_4$L),** b) **NH$_4$La(H$_4$L)** and c) **RbLa(H$_4$L)**

Supporting information 2: Illustration of the mechanism of the channel contraction of NaLa(H_4L):

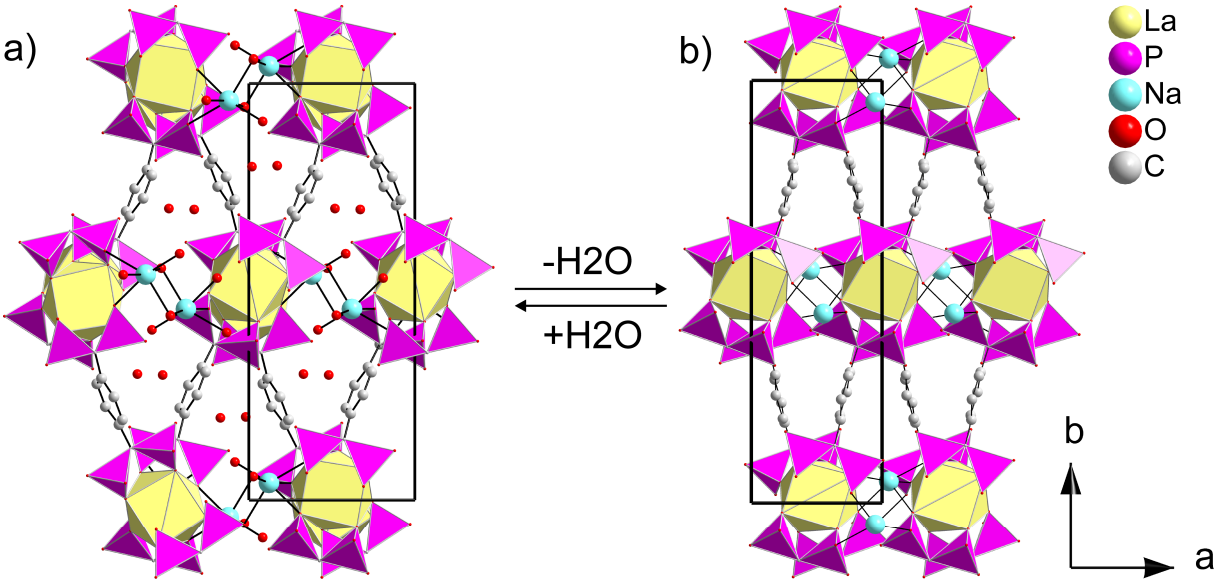

Figure S2: Contraction and expansion of the channels in **NaLa(H_4L)** during the dehydration/hydration process: [001] projection of the crystal structure of (a) **NaLa(H_4L)**, and (b) **NaLa(H_4L)$_{dehyd}$** . Hydrogen atoms have been omitted for clarity.

Supporting information 3: Additional X-Ray diffractogramms

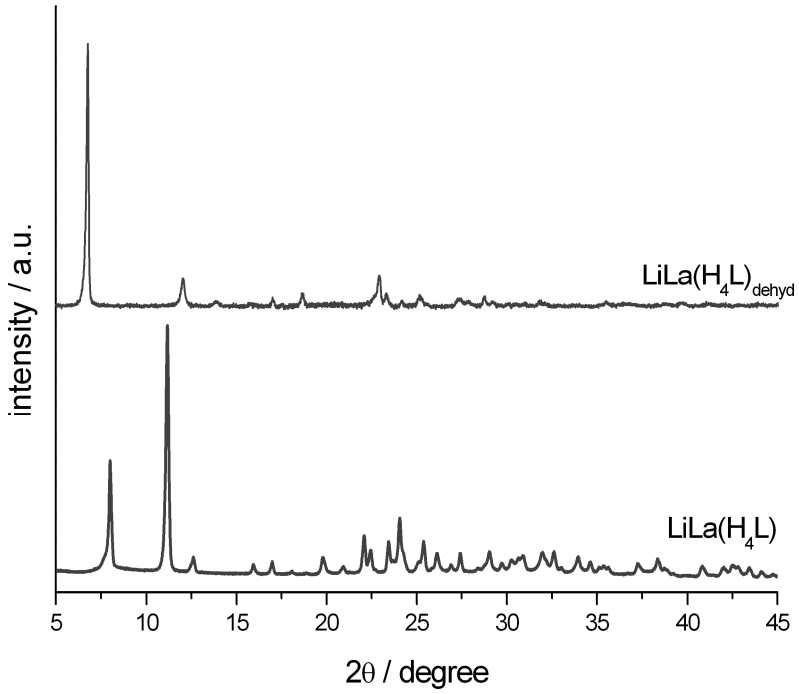

Figure S3-1: Powder pattern of **LiLa(H$_4$L)** · 4H$_2$O and the dehydrated form **LiLa(H$_4$L)$_{dehyd.}$** The reversible switch of the pattern upon dehydration indicates a contraction of the structure similar as demonstrated for **NaLa(H$_4$L)**.

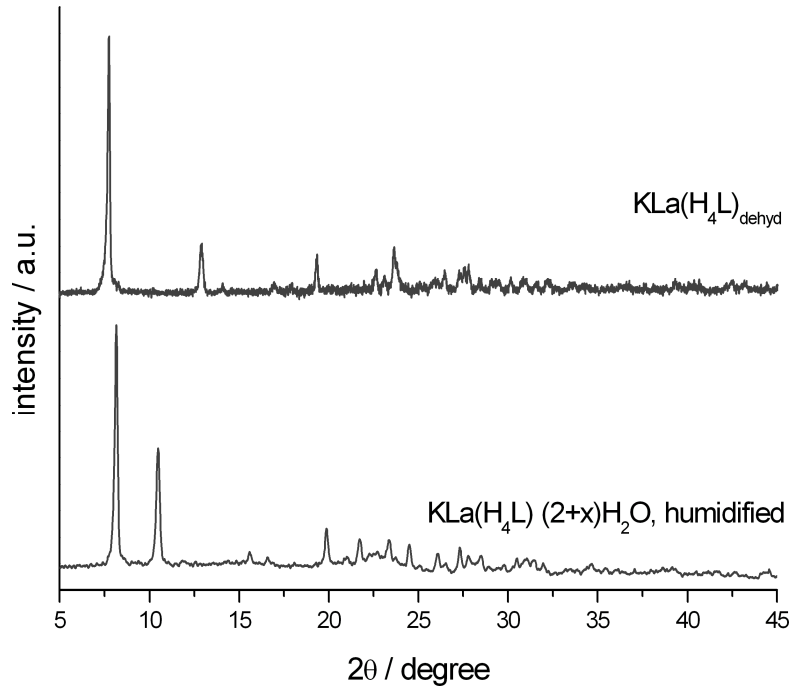

Figure S3-2: Powder pattern of a humidified sample of **KLa(H$_4$L)** · (2+x) H$_2$O and the dehydrated form **KLa(H$_4$L)$_{dehyd.}$** The reversible switch of the pattern upon dehydration indicates a contraction of the structure similar as demonstrated for **NaLa(H$_4$L)**.

Supporting information 4: Additional water sorption isotherms

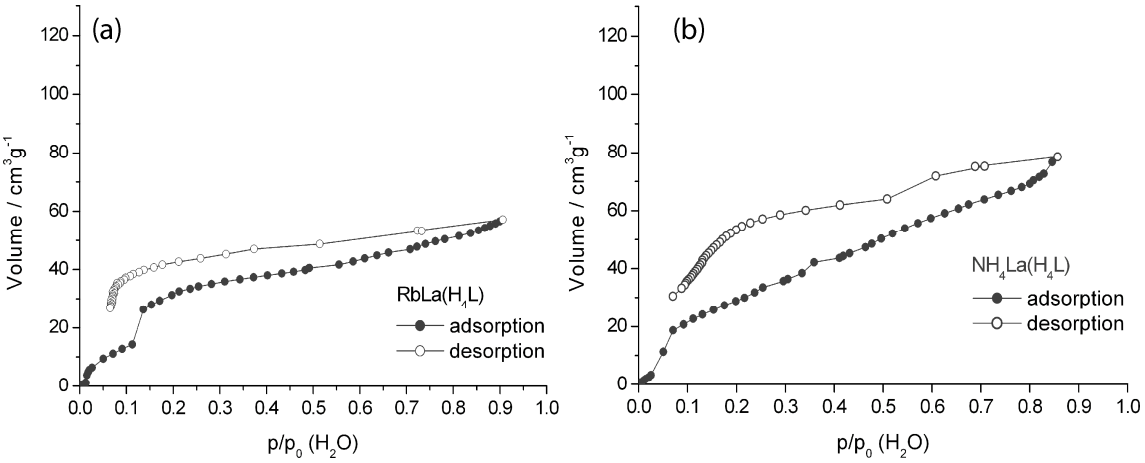

Figure S4: Water sorption isotherms of a) **RbLa(H$_4$L)** and b) **(NH$_4$)La(H$_4$L)**

Supporting information 5: Additional Thermogravimetric analyses

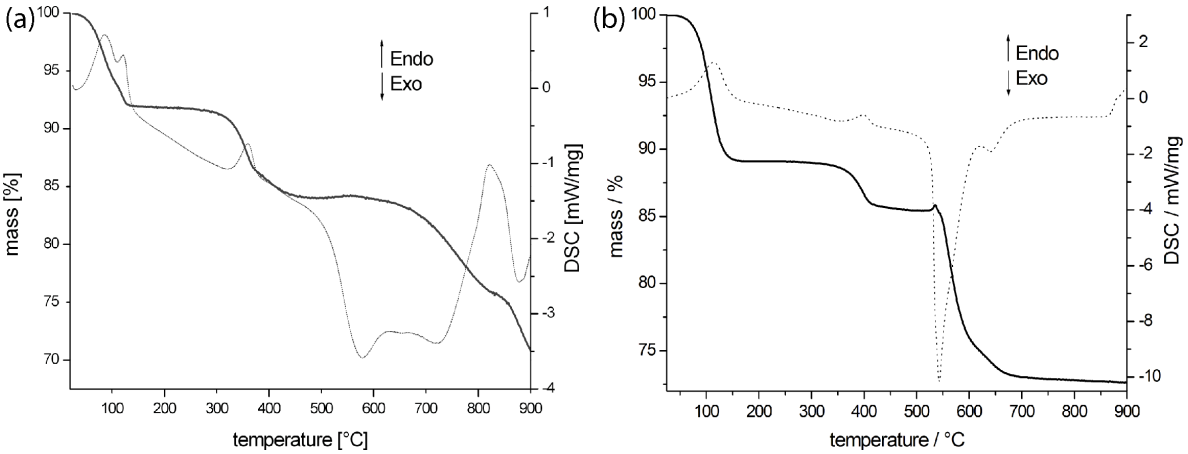

Figure S5: Thermogravimetric Analysis and Differential Calorimetry Measurement of a) **(NH$_4$)La(H$_4$L)** and b) **NaLa(H$_4$L)**. In **(NH$_4$)La(H$_4$L)** water removal occurs in two energetically distinct endothermic steps.