

## Electronic Supporting Information

### Strong negative thermal expansion and relaxor ferroelectricity driven by supramolecular patterns

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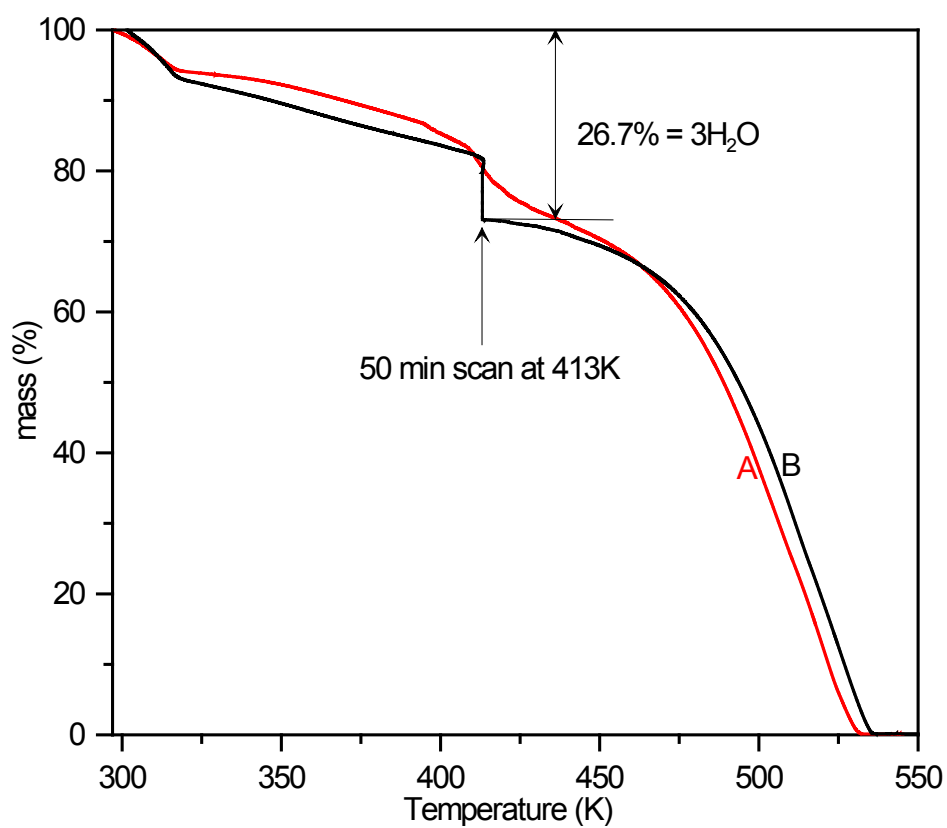
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#### Elemental analysis

The stoichiometry of the crystals obtained from an aqueous solution was checked by elemental analysis, which gave the following results: N – 13.98 (calc. 13.83)%, C – 35.87(35.56)%, H – 9.99(9.39)% and Cl – 17.39(17.51)%. The values in brackets calculated for crystal formula  $[C_6H_{13}N_2]^+Cl^- \cdot 3H_2O$  are very close to those obtained from the elemental analysis.

#### Thermogravimetric study

The stoichiometry of the crystal has also been confirmed by thermogravimetric measurements presented in Figure S1. The measurements were performed using TGA Q50 (TA Instruments) analyzer. The sample was surrounded by dry  $N_2$  and therefore the dehydration process was seen even at room temperature. Two runs were performed for two different samples. The temperature was changed with a rate of 3 K/min. Additionally in the run B the isothermal scan at 413 K was made. These measurements indicate that the loss of the crystal mass, corresponding to three water molecules per formula unit, takes place, as it is marked in Figure S1.



**Figure S1.** TGA runs for dabcoHCl·3H<sub>2</sub>O measured: (A) at a rate of temperature changes of 3 K/min, and (B) at the same temperature rate, but with isothermal 50 min scan at 413 K.