

Electronic Supporting Information

for

**On the impact of metal ions proportion on physical properties of heterometallic metal-organic frameworks**

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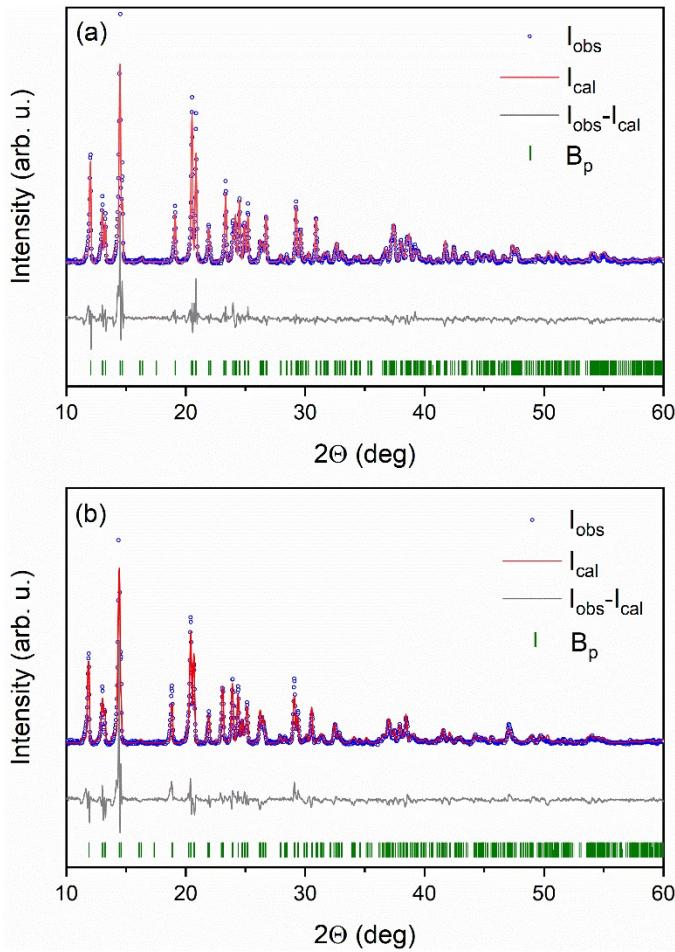
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**Table S1.** The Na and K content determined by the ICP-OES analysis

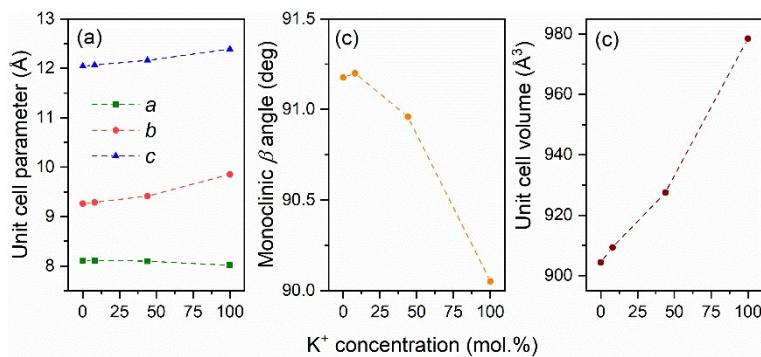
| Sample   | Na (mol%) | K (mol%) |
|--|-----------|----------|
| [CH <sub>3</sub> CH <sub>2</sub> NH <sub>3</sub> ][K <sub>0.04</sub> Na <sub>0.46</sub> Cr <sub>0.5</sub> (HCOO) <sub>3</sub> ] ( <b>2</b> ) | 91.5      | 8.2      |
| [CH <sub>3</sub> CH <sub>2</sub> NH <sub>3</sub> ][K <sub>0.22</sub> Na <sub>0.28</sub> Cr <sub>0.5</sub> (HCOO) <sub>3</sub> ] ( <b>3</b> ) | 56.1      | 43.4     |

**Table S2.** The results of Rietveld refinement for samples with x = 0.04 and 0.22 compared to the literature data for [CH<sub>3</sub>CH<sub>2</sub>NH<sub>3</sub>][Na<sub>0.5</sub>Cr<sub>0.5</sub>(HCOO)<sub>3</sub>] (x = 0) and [CH<sub>3</sub>CH<sub>2</sub>NH<sub>3</sub>][K<sub>0.5</sub>Cr<sub>0.5</sub>(HCOO)<sub>3</sub>] (x = 1)

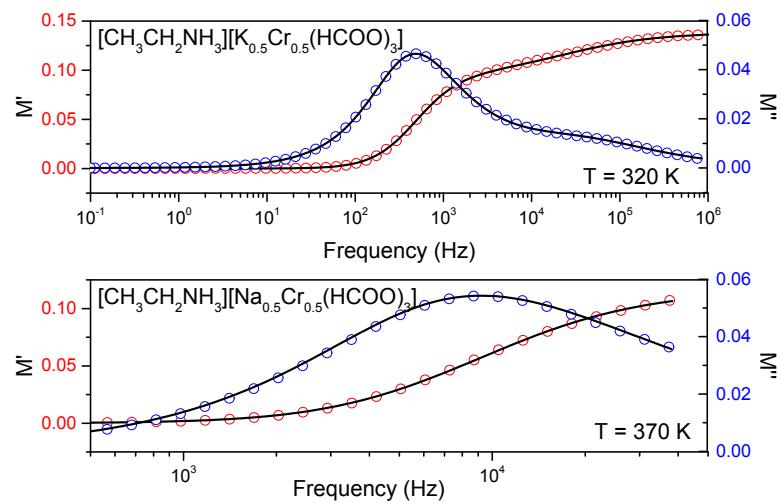
| Sample   | x    | a (Å)     | b (Å)     | c (Å)      | β (Å)       | V (Å <sup>3</sup> ) |
|--|------|-----------|-----------|------------|-------------|---------------------|
| [CH <sub>3</sub> CH <sub>2</sub> NH <sub>3</sub> ][Na <sub>0.5</sub> Cr <sub>0.5</sub> (HCOO) <sub>3</sub> ] ( <b>1</b> )                    | 0    | 8.1047(1) | 9.2639(1) | 12.0459(1) | 91.1766(10) | 904.24(2)           |
| [CH <sub>3</sub> CH <sub>2</sub> NH <sub>3</sub> ][K <sub>0.04</sub> Na <sub>0.46</sub> Cr <sub>0.5</sub> (HCOO) <sub>3</sub> ] ( <b>2</b> ) | 0.04 | 8.1125    | 9.2897    | 12.0664    | 91.20       | 909.36              |
| [CH <sub>3</sub> CH <sub>2</sub> NH <sub>3</sub> ][K <sub>0.22</sub> Na <sub>0.28</sub> Cr <sub>0.5</sub> (HCOO) <sub>3</sub> ] ( <b>3</b> ) | 0.22 | 8.0985    | 9.4155    | 12.1641    | 90.96       | 927.53              |
| [CH <sub>3</sub> CH <sub>2</sub> NH <sub>3</sub> ][K <sub>0.5</sub> Cr <sub>0.5</sub> (HCOO) <sub>3</sub> ] ( <b>4</b> )                     | 1    | 8.0147(2) | 9.8547(3) | 12.3886(3) | 90.052(3)   | 978.48(5)           |



**Figure S1.** Experimental ( $I_{\text{obs}}$ ), calculated ( $I_{\text{cal}}$ ) and difference ( $I_{\text{obs}} - I_{\text{cal}}$ ) XRD patterns together with Bragg positions ( $B_p$ ) for the  $x = 0.04$  (2) (a) and  $x = 0.22$ (3) (b) samples; resulting parameters of fitting:  $R_p = 3.38$ ,  $wR_p = 5.09$ ,  $Gof = 11.99$  (for  $x = 0.4$ ) and  $R_p = 3.48$ ,  $wR_p = 5.88$ ,  $Gof = 14.62$  (for  $x = 0.22$ )



**Figure S2.** Evolution of unit cell parameters as a function of  $K^+$  concentration



**Figure S3.** Frequency dependence of the complex dielectric permittivity