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Electronic Supplementary Information

**Note added after first publication:** This file replaces the version published on 18th of September 2017, in which the figures were not accurately reproduced. The content of the file has not otherwise changed.

## Double Salt Ionic Liquids Based on 1-Ethyl-3-Methylimidazolium Acetate and Hydroxyl-Functionalized Ammonium Acetates: Strong Effects of Weak Interactions

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## **Supplementary Figures**

**Figure S1.** <sup>1</sup>H NMR spectra of  $[N(CH_3)_4]_x[C_2mim]_{1-x}[OAc]$  at 25 °C using CDCl<sub>3</sub> as external lock (x corresponds to the molar ratio of  $[N(CH_3)_4]^+/[OAc]^-$ , in which zero corresponds to  $[C_2mim][OAc]$ ).



**Figure S2.** <sup>1</sup>H NMR spectra of  $[Ch]_x[C_2mim]_{1-x}[OAc]$  at 25 °C using CDCl<sub>3</sub> as external lock (x corresponds to the molar ratio of  $[Ch]^+/[OAc]^-$ , in which zero corresponds to  $[C_2mim][OAc]$ ).



**Figure S3.** <sup>1</sup>H NMR spectra of  $[NH_3CH_2CH_3]_x[C_2mim]_{1-x}[OAc]$  at 25 °C using CDCl<sub>3</sub> as external lock (x corresponds to the molar ratio of  $[NH_3CH_2CH_3]^+/[OAc]^-$ , in which zero corresponds to  $[C_2mim][OAc]$ ).



**Figure S4.** <sup>1</sup>H NMR spectra of  $[NH_3OH]_x[C_2mim]_{1-x}[OAc]$  at 25 °C using CDCl<sub>3</sub> as external lock (x corresponds to the molar ratio of  $[NH_3OH]^+/[OAc]^-$ , in which zero corresponds to  $[C_2mim][OAc]$ ).



**Figure S5.** <sup>1</sup>H NMR spectra of  $[NH_3(CH_2)_2OH]_x[C_2mim]_{1-x}[OAc]$  at 25 °C using CDCl<sub>3</sub> as external lock (x corresponds to the molar ratio of  $[NH_3(CH_2)_2OH]^+/[OAc]^-$ ), in which zero corresponds to  $[C_2mim][OAc]$ ).



**Figure S6.** <sup>13</sup>C NMR spectra of  $[N(CH_3)_4]_x[C_2mim]_{1-x}[OAc]$  at 25 °C using CDCl<sub>3</sub> as external lock (x corresponds to the molar ratio of  $[N(CH_3)_4]^+/[OAc]^-$ , in which zero corresponds to  $[C_2mim][OAc]$ ).



**Figure S7.** <sup>13</sup>C NMR spectra of  $[Ch]_x[C_2mim]_{1-x}[OAc]$  at 25 °C using CDCl<sub>3</sub> as external lock (x corresponds to the molar ratio of  $[Ch]^+/[OAc]^-$ , in which zero corresponds to  $[C_2mim][OAc]$ ).



**Figure S8.** <sup>13</sup>C NMR spectra of  $[NH_3CH_2CH_3]_x[C_2mim]_{1-x}[OAc]$  at 25 °C using CDCl<sub>3</sub> as external lock (x corresponds to the molar ratio of  $[NH_3CH_2CH_3]^+/[OAc]^-$ , in which zero corresponds to  $[C_2mim][OAc]$ ).



**Figure S9.** <sup>13</sup>C NMR spectra of  $[NH_3OH]_x[C_2mim]_{1-x}[OAc]$  at 25 °C using CDCl<sub>3</sub> as external lock (x corresponds to the molar ratio of  $[NH_3OH]^+/[OAc]^-$ , in which zero corresponds to  $[C_2mim][OAc]$ ).



**Figure S10.** <sup>13</sup>C NMR spectra of  $[NH_3(CH_2)_2OH]_x[C_2mim]_{1-x}[OAc]$  at 25°C using CDCl<sub>3</sub> as external lock (x corresponds to the molar ratio of  $[NH_3(CH_2)_2OH]^+/[OAc]^-$ ), in which zero corresponds to  $[C_2mim][OAc]$ ).



**Figure S11.** <sup>1</sup>H NMR chemical shifts of the  $[C_2mim][OAc]$  ring protons of a) [NH<sub>3</sub>CH<sub>2</sub>CH<sub>3</sub>]<sub>x</sub>[C<sub>2</sub>mim]<sub>1-x</sub>[OAc], b) [NH<sub>3</sub>OH]<sub>x</sub>[C<sub>2</sub>mim]<sub>1-x</sub>[OAc], and c) [NH<sub>3</sub>(CH<sub>2</sub>)<sub>2</sub>OH]<sub>x</sub>[C<sub>2</sub>mim]<sub>1-x</sub>[OAc] (zero on the axis corresponds to [C<sub>2</sub>mim][OAc]).



**Figure S12.** <sup>13</sup>C NMR chemical shifts of the  $[C_2mim][OAc]$  ring protons of a)  $[NH_3CH_2CH_3]_x[C_2mim]_{1-x}[OAc]$ , b)  $[NH_3OH]_x[C_2mim]_{1-x}[OAc]$ , and c)  $[NH_3(CH_2)_2OH]_x[C_2mim]_{1-x}[OAc]$  (zero on the axis corresponds to  $[C_2mim][OAc]$ ).



**Figure S13.** PXRD pattern of  $[N(CH_3)_4][OAc]$  *vs.* simulated pattern from crystal structure. Unidentified peaks from the experimental pattern are labeled with positions.



Figure S14. IR spectrum of [N(CH<sub>3</sub>)<sub>4</sub>][OAc].



**Figure S15:** Optical micrographs at 50x magnification of [N(CH<sub>3</sub>)<sub>4</sub>][OAc] under ordinary transmitted light (*left*) and crossed polarizers (*right*).