

**Electronic Supplementary Information**

**Phase Equilibrium and Physical Properties of Biobased  
Ionic Liquid Mixtures**

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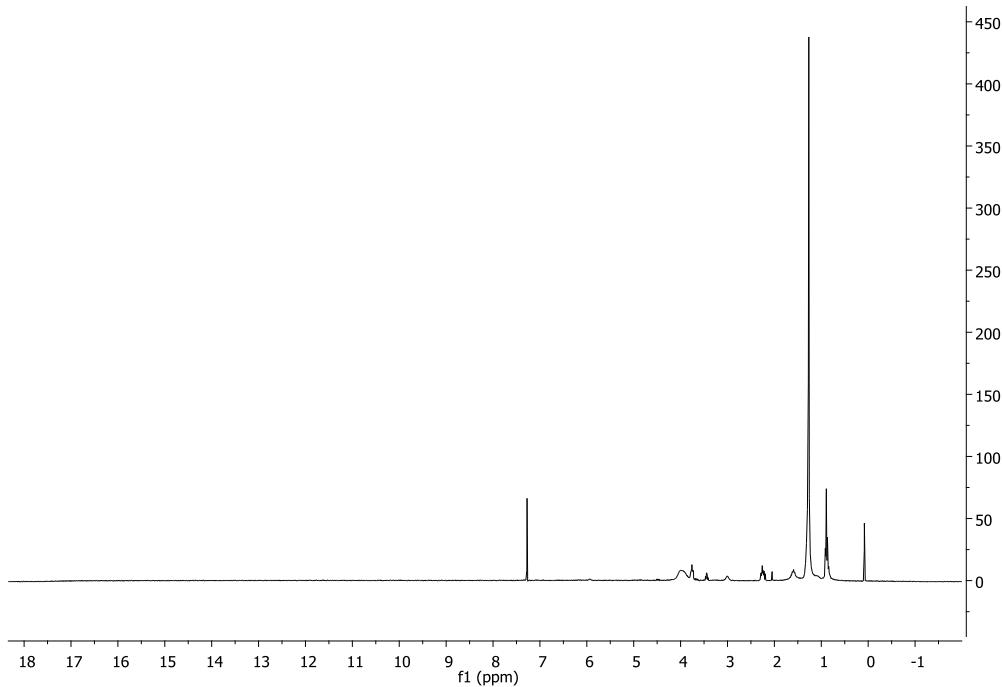
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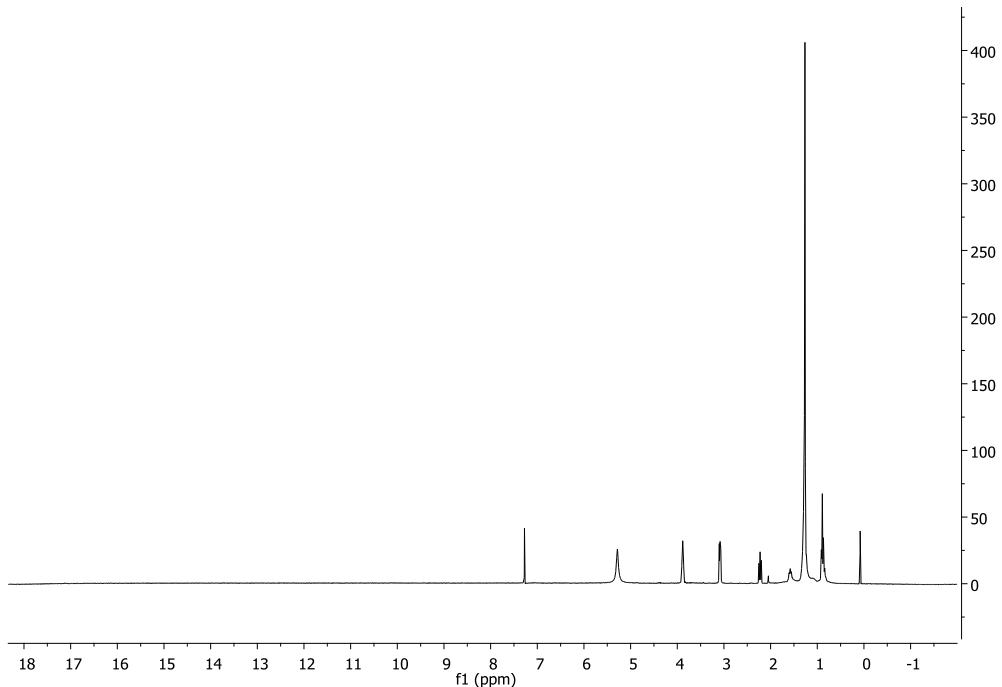
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## 1. Proton Nuclear Magnetic Resonance ( $^1\text{H}$ NMR)

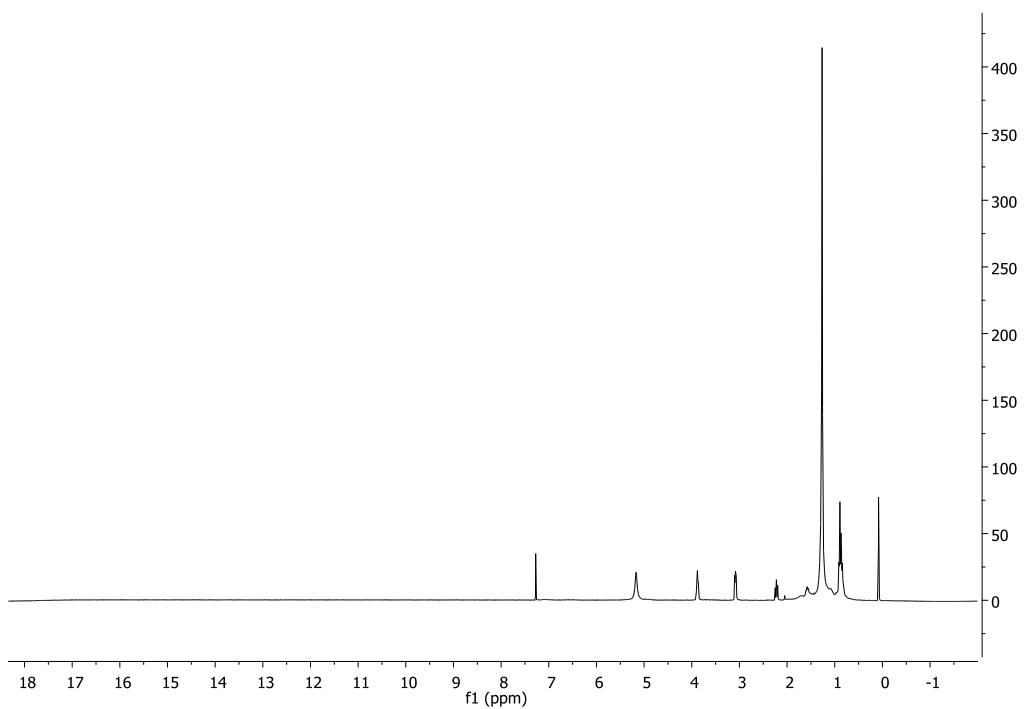
The spectrums for all PILs obtained by  $^1\text{H}$  NMR are presented below:



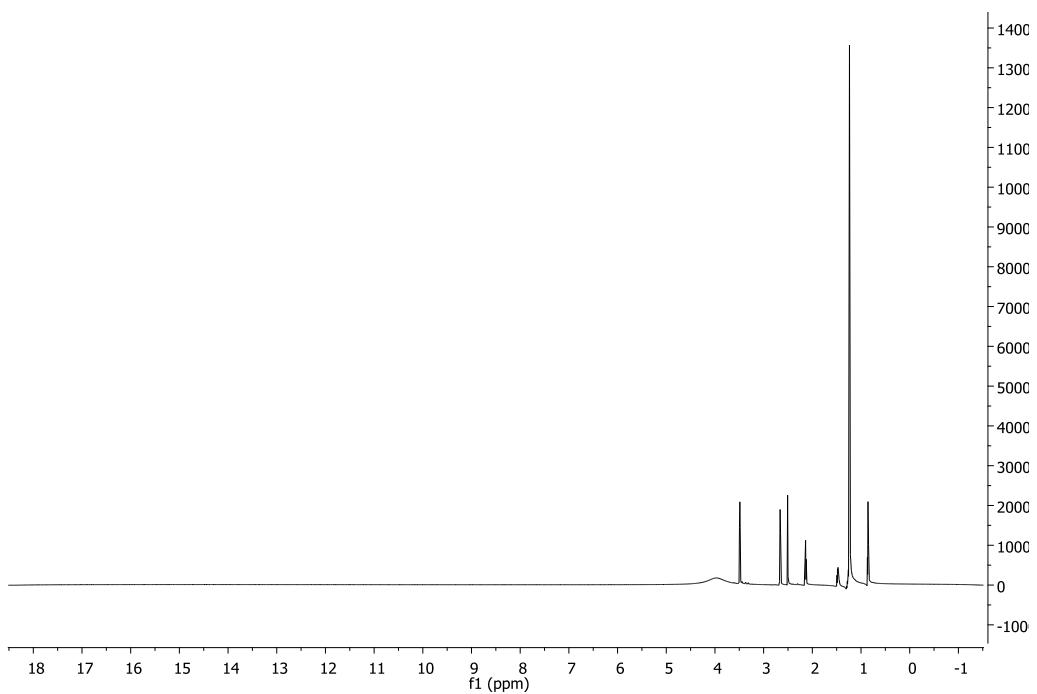
**Figure 1.**  $^1\text{H}$  NMR spectrum for 2-hydroxy ethylammonium tetradecanoate [HEA][ $\text{C}_{14}\text{OO}$ ].



**Figure 2.**  $^1\text{H}$  NMR spectrum for bis(2-hydroxy ethyl)ammonium tetradecanoate [ $\text{H}_2\text{EA}$ ][ $\text{C}_{14}\text{OO}$ ].



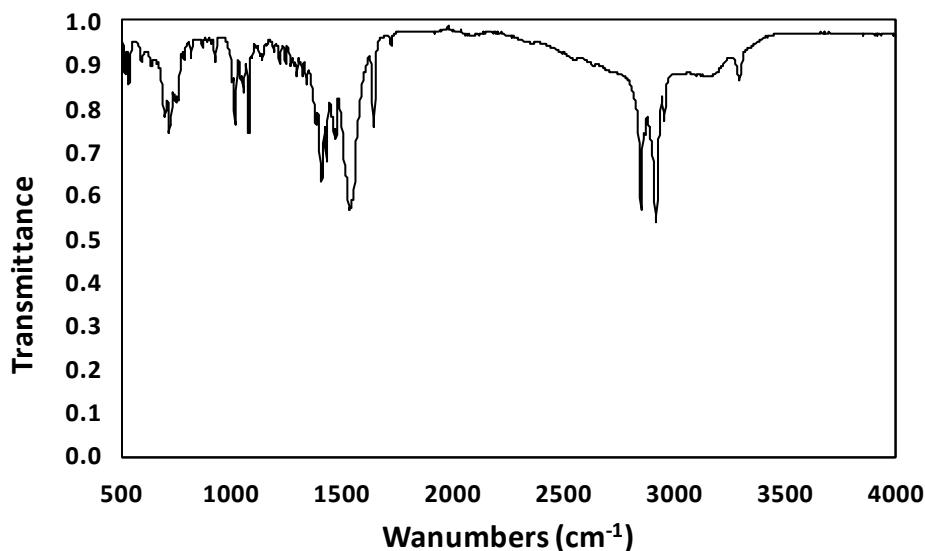
**Figure 3.** <sup>1</sup>H NMR spectrum for bis(2-hydroxy ethyl)ammonium hexadecanoate [H<sub>2</sub>EA][C<sub>16</sub>OO].



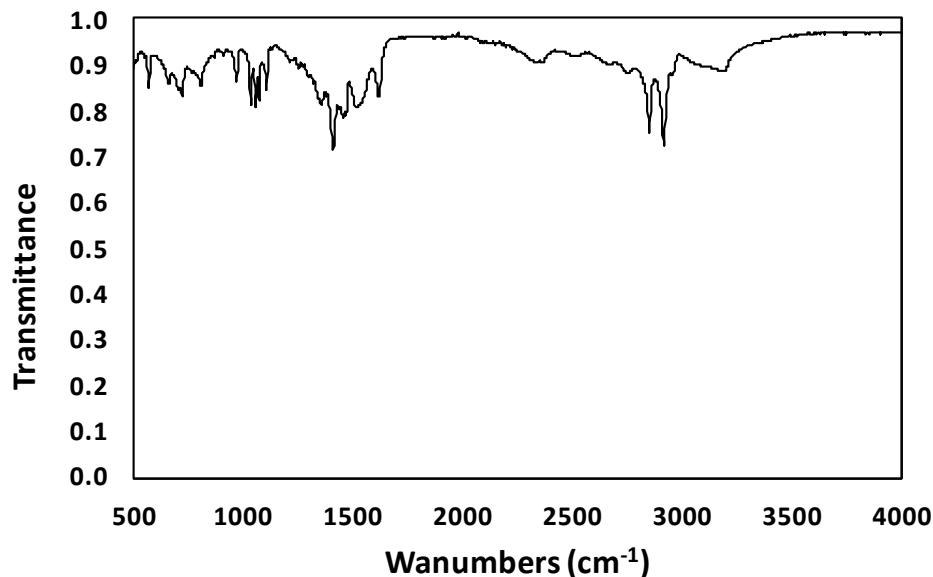
**Figure 4.** <sup>1</sup>H NMR spectrum for bis(2-hydroxy ethyl)ammonium octadecanoate [H<sub>2</sub>EA][C<sub>18</sub>OO].

## 2. Attenuated Total Reflectance-Fourier Transform Infrared spectroscopy (ATR-FTIR)

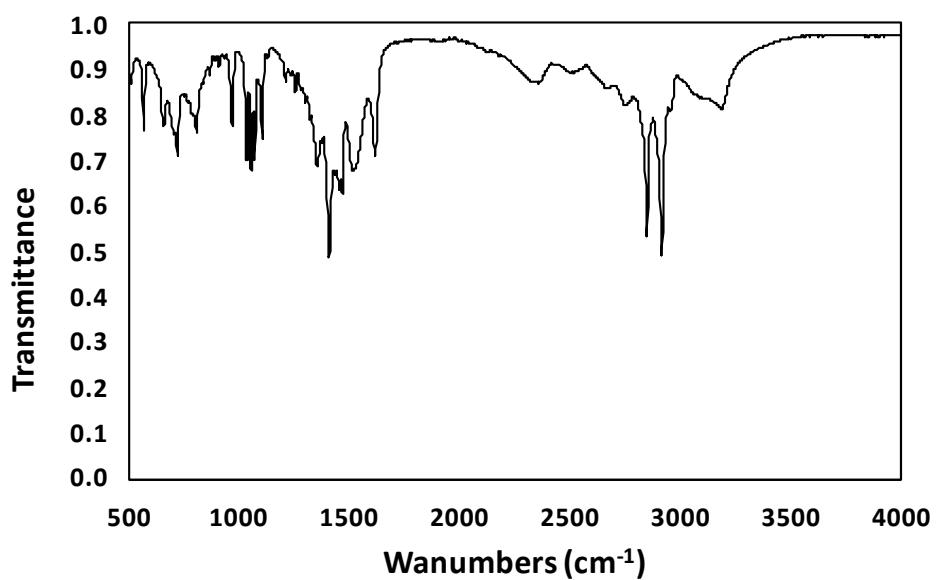
The spectrums for all PILs obtained by ATR-FTIR are presented below:



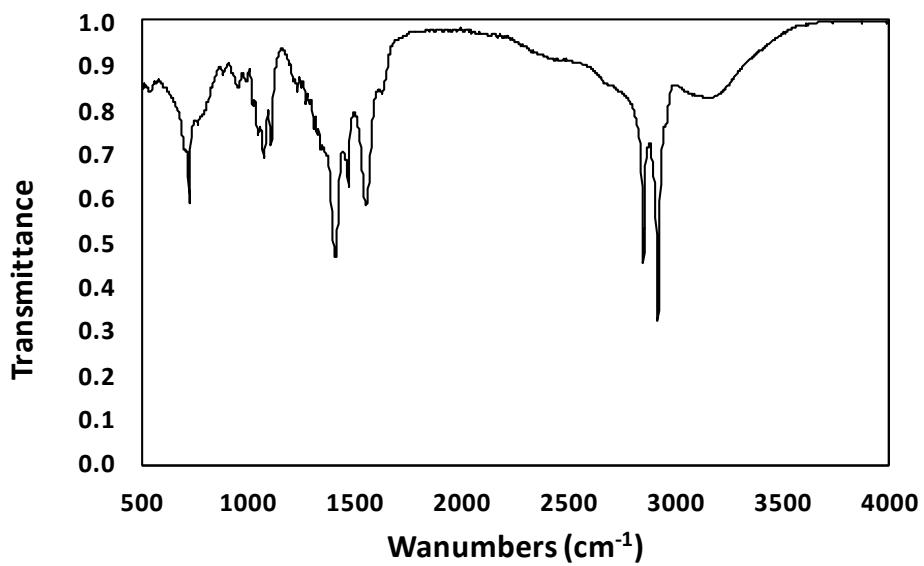
**Figure 5.** ATR-FTIR spectrum for 2-hydroxy ethylammonium tetradecanoate [HEA][C<sub>14</sub>OO].



**Figure 6.** ATR-FTIR spectrum for bis(2-hydroxy ethyl)ammonium tetradecanoate [H<sub>2</sub>EA][C<sub>14</sub>OO].

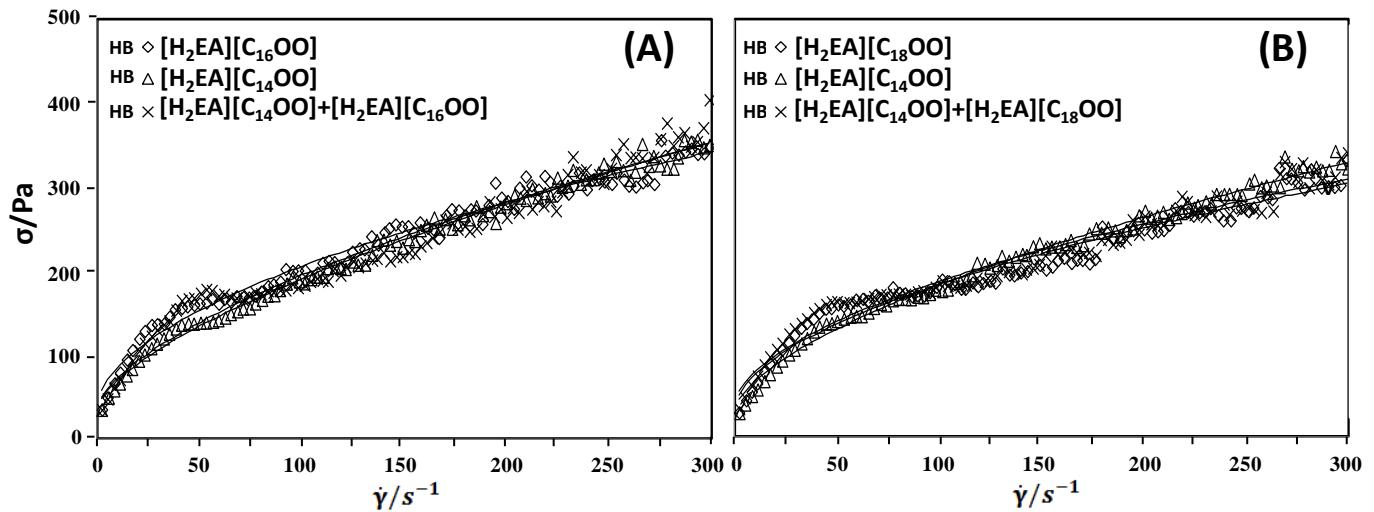


**Figure 7.** ATR-FTIR spectrum for bis(2-hydroxy ethyl)ammonium hexadecanoate  $[\text{H}_2\text{EA}][\text{C}_{16}\text{OO}]$ .

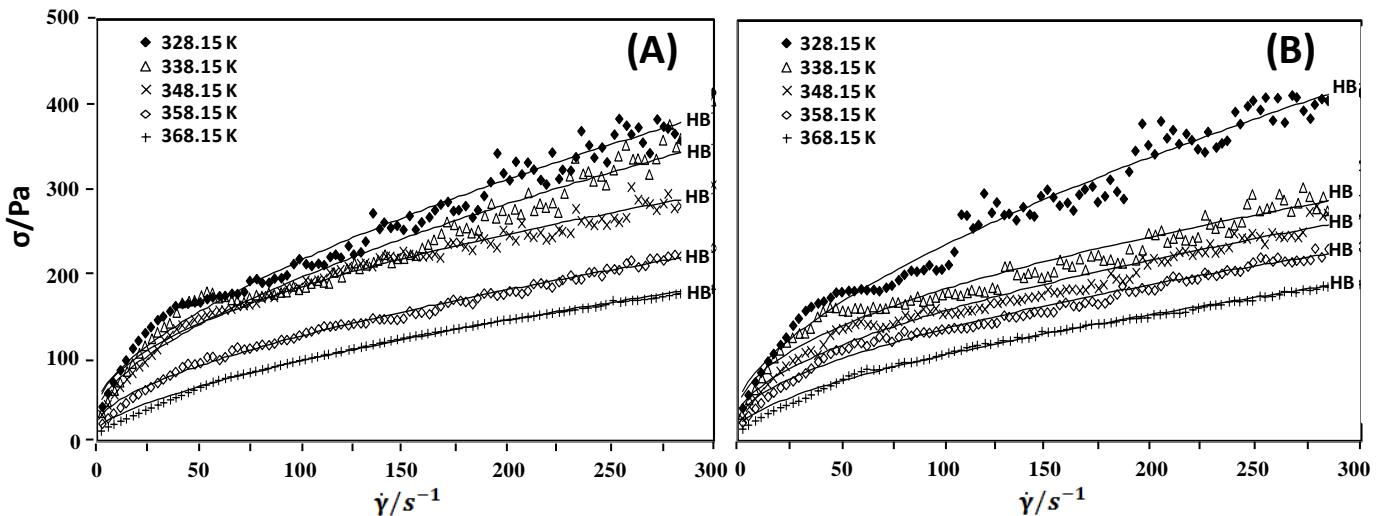


**Figure 8.** ATR-FTIR spectrum for bis(2-hydroxy ethyl)ammonium octadecanoate  $[\text{H}_2\text{EA}][\text{C}_{18}\text{OO}]$ .

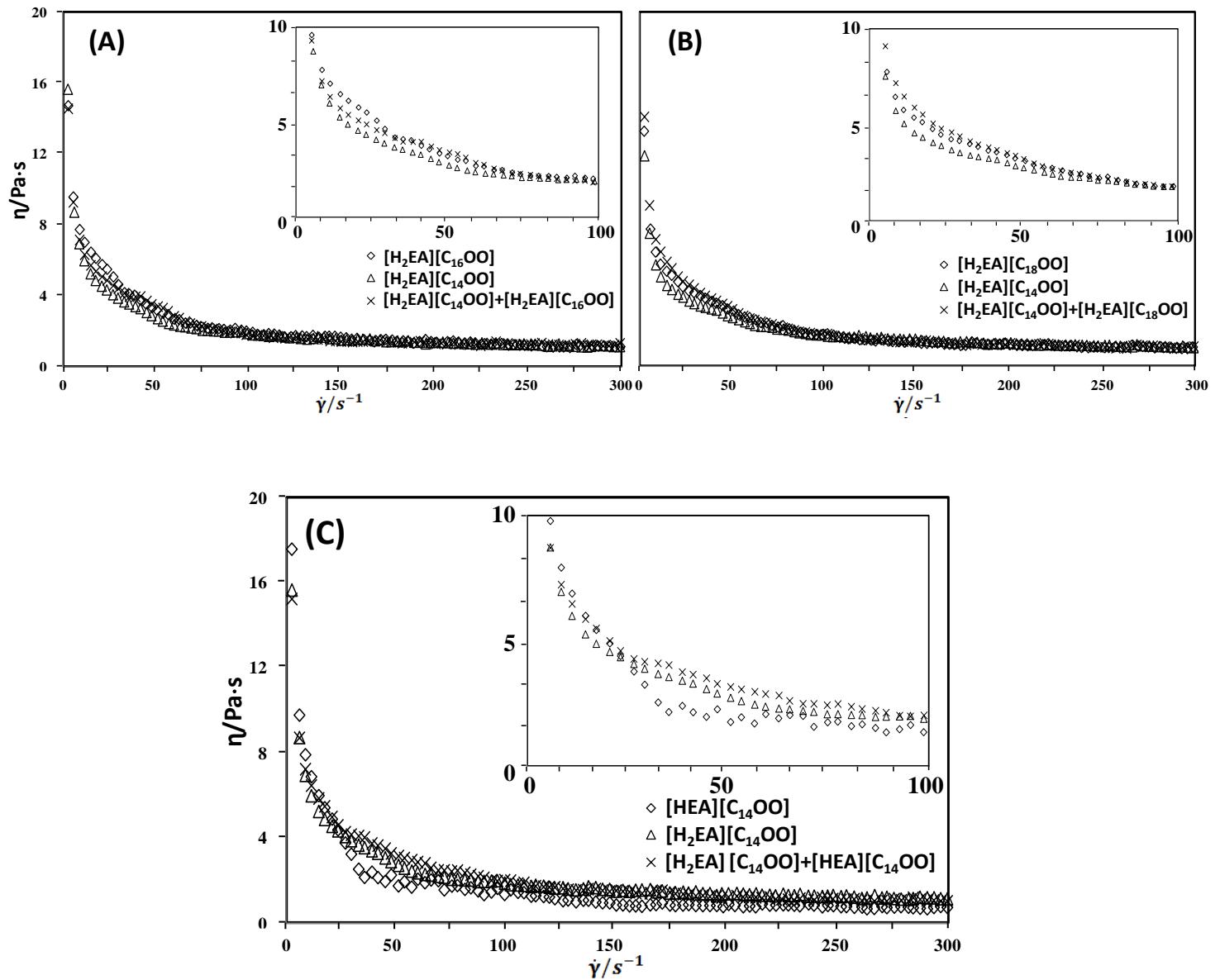
### 3. Rheological profiles



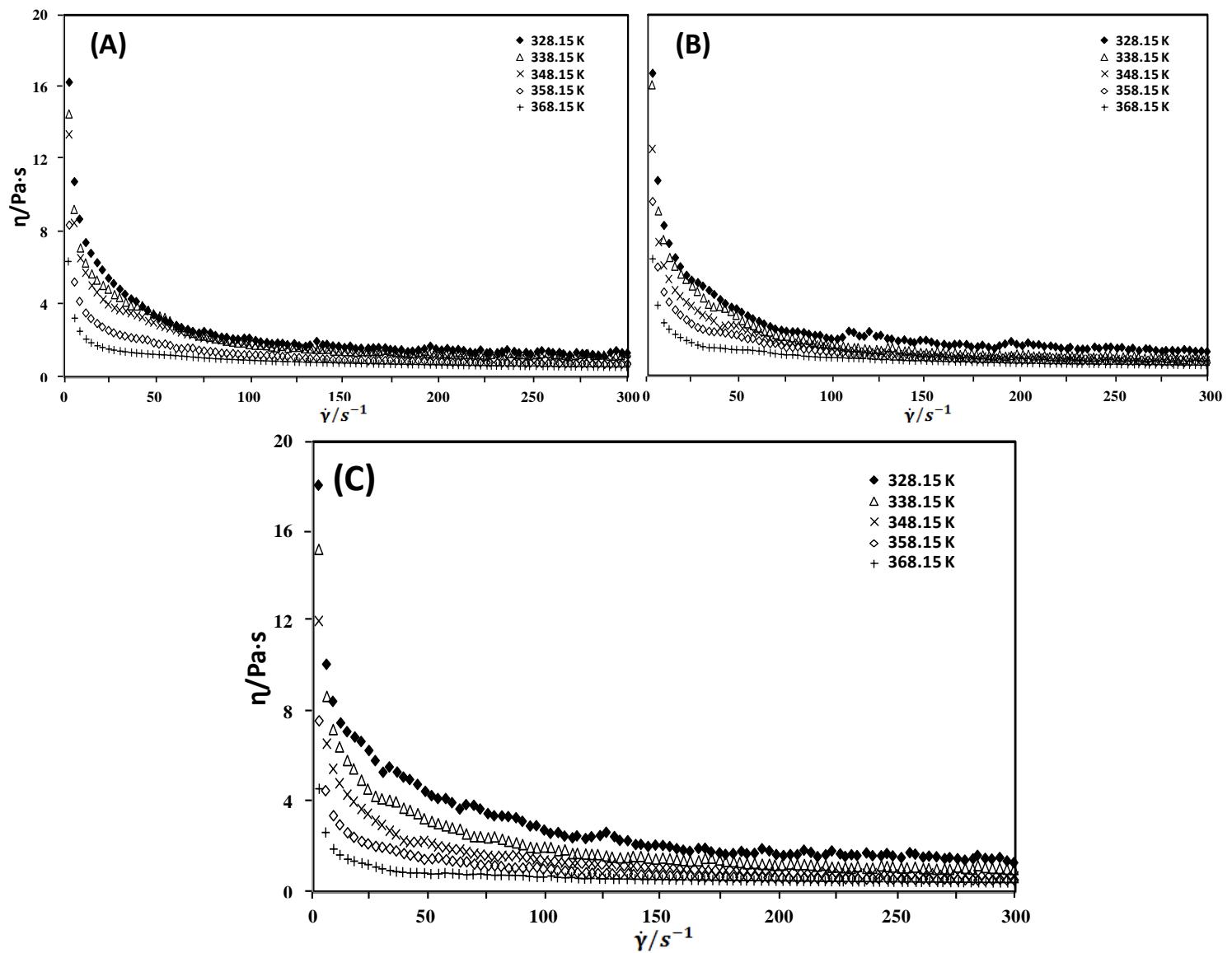
**Figure 9.** Shear stress ( $\sigma$ ) versus shear rate ( $\dot{\gamma}$ ) curves of pure protic ionic liquids (PILs) and their binary mixtures at  $x_1 = 0.5$ : (A)  $[H_2EA][C_{14}OO]$  +  $[HEA][C_{16}OO]$  at 338.15 K, (B)  $[H_2EA][C_{14}OO]$  +  $[HEA][C_{18}OO]$  at 338.15 K. Solid lines are the fitted curves for Herschel-Bulkley model, represented by **HB** label.



**Figure 10.** Shear stress ( $\sigma$ ) versus shear rate ( $\dot{\gamma}$ ) curves of protic ionic liquids (PILs) binary mixtures ( $x_1 = 0.5$ ) at different temperatures 328.15–368.15 K: (A)  $[H_2EA][C_{14}OO]$  +  $[HEA][C_{16}OO]$ , (B)  $[H_2EA][C_{14}OO]$  +  $[HEA][C_{18}OO]$ . Solid lines are the fitted curves for Herschel-Bulkley model, represented by **HB** label.



**Figure 11.** Apparent viscosity data of protic ionic liquids (PILs) and their binary mixtures at  $x_1 = 0.5$ : (A)  $[\text{H}_2\text{EA}][\text{C}_{14}\text{OO}] + [\text{HEA}][\text{C}_{16}\text{OO}]$  at 338.15 K, (B)  $[\text{H}_2\text{EA}][\text{C}_{14}\text{OO}] + [\text{HEA}][\text{C}_{18}\text{OO}]$  at 343.15 K, (C)  $[\text{H}_2\text{EA}][\text{C}_{14}\text{OO}] + [\text{HEA}][\text{C}_{14}\text{OO}]$  at 338.15 K.



**Figure 12.** Apparent viscosity data of protic ionic liquids (PILs) binary mixtures ( $x_1 = 0.5$ ) at different temperatures 328.15-368.15 K: (A)  $[\text{H}_2\text{EA}][\text{C}_{14}\text{OO}] + [\text{HEA}][\text{C}_{16}\text{OO}]$  (B)  $[\text{H}_2\text{EA}][\text{C}_{14}\text{OO}] + [\text{HEA}][\text{C}_{18}\text{OO}]$  (C)  $[\text{H}_2\text{EA}][\text{C}_{14}\text{OO}] + [\text{HEA}][\text{C}_{14}\text{OO}]$ .