

## Supporting Information

### Solid-State $^1\text{H}$ and $^{13}\text{C}$ NMR Studies of New Ionic Plastic-Crystals with Branched Structures: $[\text{NEt}_x\text{Me}_{(3-x)}(i\text{-Pr})][\text{BEt}_{(4-y)}\text{Me}_y]$ ( $x = 1\text{--}3$ , $y = 0, 1$ )

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Table S1 Melting ( $T_{\text{mp}}$ ) and phase transition temperatures of some alkylammonium salts with the  $\text{BEt}_3\text{Me}$  and  $\text{BEt}_4$  anions. Here, symbols of  $T_{\text{R}}$  and  $T_{\text{P}}$  are transition temperatures from solid to rotator and plastic phases, respectively.  $M_w$  is molecular weight of the cations.

	$M_w$	BEt <sub>3</sub> Me			BEt <sub>4</sub>		
		$T_{\text{R}}$	$T_{\text{P}}$	$T_{\text{mp}}$	$T_{\text{R}}$	$T_{\text{P}}$	$T_{\text{mp}}$
NMe <sub>4</sub> *	74	-	252.6	dec.	-	333	dec.
NEtMe <sub>3</sub> *	88	-	239.8	dec.	-	278	dec.
NEt <sub>2</sub> Me <sub>2</sub> *	102	-	-	dec.	-	242	dec.
<b>NMe<sub>3</sub>(i-Pr)</b>	<b>102</b>	<b>-</b>	<b>227</b>	<b>dec.</b>	<b>-</b>	<b>271</b>	<b>dec.</b>
NEt <sub>3</sub> Me*	116	-	-	dec.	-	238	dec.
NEtMe <sub>2</sub> Pr**	116	267	325	355	253	-	345
<b>NEtMe<sub>2</sub>(i-Pr)</b>	<b>116</b>	<b>-</b>	<b>197</b>	<b>dec.</b>	<b>-</b>	<b>242</b>	<b>dec.</b>
NEt <sub>4</sub> *	130	-	233.4	dec.	-	295	dec.
NEt <sub>2</sub> MePr**	130	240	322	370	235	-	325
NEtMe <sub>2</sub> Bu**	130	202	-	326	210	-	320
<b>NEt<sub>2</sub>Me(i-Pr)</b>	<b>130</b>	<b>-</b>	<b>-</b>	<b>dec.</b>	<b>-</b>	<b>235</b>	<b>dec.</b>
NEt <sub>3</sub> Pr*	144	-	243.3	391.2	341	-	363
NEt <sub>2</sub> MeBu**	144	222	-	310	-	-	303
<b>NEt<sub>3</sub>(i-Pr)</b>	<b>144</b>	<b>-</b>	<b>184</b>	<b>dec.</b>	<b>-</b>	<b>245</b>	<b>dec.</b>
NEt <sub>2</sub> Pr <sub>2</sub> *	158	269.1	-	358.7	341	-	361
NEtPr <sub>3</sub> *	172	328.4	-	359.3	358	-	366
NPr <sub>4</sub> *	186	-	-	377.9	-	-	388
NBu <sub>4</sub> *	242	-	-	423.7	-	-	418

Data of \* and \*\* are employed in reference [6, 37, 38, 40, 41].

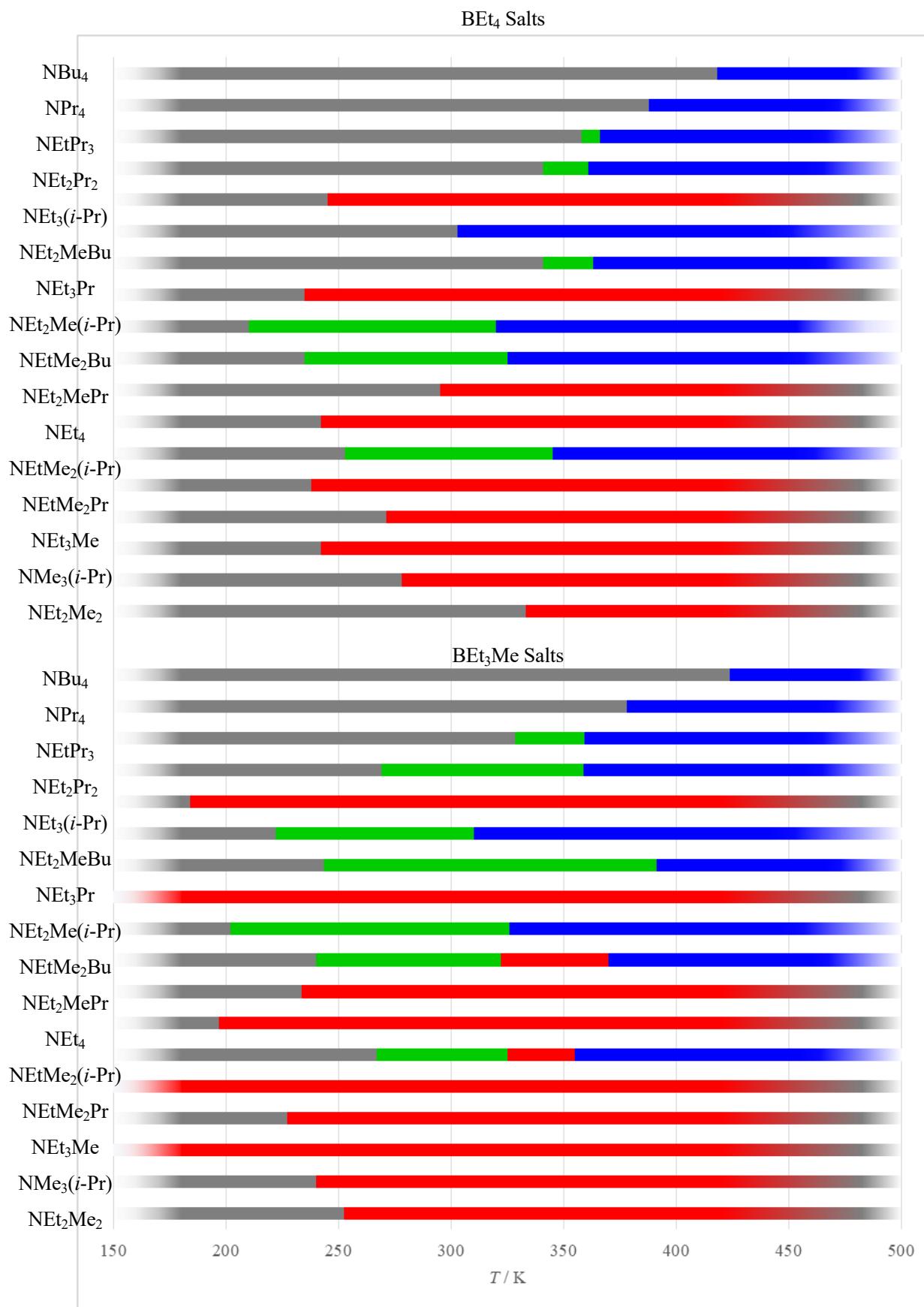


Figure S1 Phase transition temperatures. Gray, green, red, and blue lines are ordinal crystal, rotator,

plastic, and liquid phases, respectively. In this figure, the values listed in [Table S1](#) are employed.

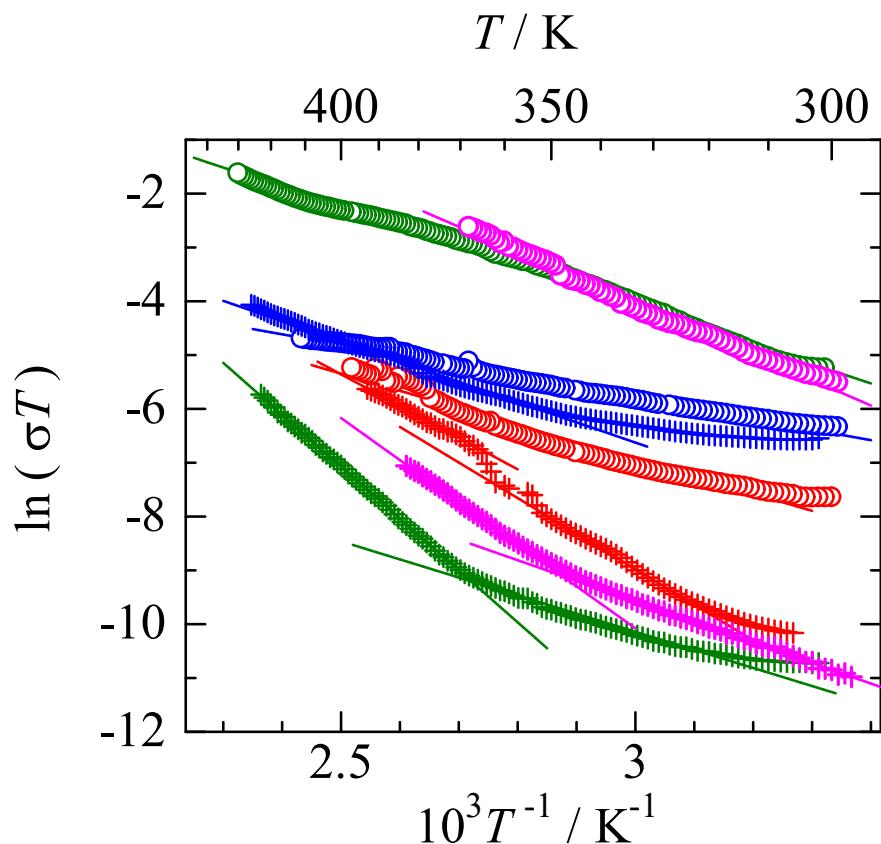


Figure S2 Arrhenius plots of electrical conductivity.

$[\text{NMe}_3(i\text{-Pr})][\text{BEt}_3\text{Me}]$  (○),  $[\text{NEtMe}_2(i\text{-Pr})][\text{BEt}_3\text{Me}]$  (○),  $[\text{NEt}_2\text{Me}(i\text{-Pr})][\text{BEt}_3\text{Me}]$  (○),  $[\text{NEt}_3(i\text{-Pr})][\text{BEt}_3\text{Me}]$  (○),  $[\text{NMe}_3(i\text{-Pr})][\text{BEt}_4]$  (+),  $[\text{NEtMe}_2(i\text{-Pr})][\text{BEt}_4]$  (+),  $[\text{NEt}_2\text{Me}(i\text{-Pr})][\text{BEt}_4]$  (+),  $[\text{NEt}_3(i\text{-Pr})][\text{BEt}_4]$  (+)