Supporting Information

Tetramethylurea Dimer/Lithium Salt-Based Deep Eutectics as a Novel Class of Eutectic Electrolytes

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Scheme 1. Synthesis of tetramethylurea (TMU) dimer.

Relative humidity = 42 %



Figure S1. Photographs of DMU dimer obtained after the recrystallization and after leaving to stand for 5 h under a humid condition.



Figure S2. DSC (top) and TGA (bottom) traces of TMU dimer.



Figure S3. (a) ¹H NMR and (b) ¹³C NMR spectra of DMU dimer in CDCl₃.

Table S1. Solubility test of DMU dimer^{a)}

	Hexane	Toluene	Diethylether	CHCl ₃	CH_2Cl_2	Acetone	THF
Dimer	-	+	+	+	+	+	+
	AcOEt	МеОН	EtOH	dioxane	DMSO	DMF	H ₂ O
Dimer	+	+	+-	+	+	+	+

 a^{a} +: soluble at r.t. +- : soluble at 60°C -: insoluble at r.t. conc. = 0.3-1.0 mg/mL.



	DMU dimer:LiTFSI							
	10 :1	5:1	3:1	1:1				
100 ∘C								
	Liquid	Liquid	Liquid+Solid	Liquid+Solid				
r.t.								
	Liquid	Liquid	Solid	Solid				

Figure S4. Synthetic pathway (top) and photographs (bottom) of TMU dimer/LiTFSI obtained at different feed ratios (dimer/LiTFSI = 10:1-1:1) after heating at 100 °C for 2 h and after leaving to stand at room temperature (25 °C or less) for 12 h.





Figure S5. Synthetic pathway (top) and photographs (bottom) of TMU dimer/LiFSI obtained at different feed ratios (dimer/LiFSI = 10:1-1:1) after heating at 100 °C for 2 h and after leaving to stand at room temperature (25 °C or less) for 12 h.



Figure S6. (a) Structures of TMU (tetramethylurea) and TMU dimer and (b) TGA traces of DMU (red dotted line) and DMU dimer (blue dotted line) and their mixture with LiTFSI (solid line) prepared at DMU (or dimer)/LiTFSI of 5:1 at 100 °C for 2 h.



Figure S7. Typical coordination diagrams of Li⁺/TMU dimer: one Li⁺ was interacted with (a) one amido (C=O) and (b) two amide (C=O) units, respectively.