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## Multiple crystal pathways and crystal polymorphs in protic ionic liquids

Hiroshi Abe <sup>a,\*</sup>, Takahiro Takekiyo <sup>b</sup>, Yukihiro Yoshimura <sup>b</sup>, Akio Shimizu <sup>c</sup>, Shinichiro Ozawa <sup>a</sup>

<sup>a</sup> *Department of Materials Science and Engineering, National Defense Academy, Japan*

<sup>b</sup> *Department of Applied Chemistry, National Defense Academy, Japan*

<sup>c</sup> *Graduate School of Environmental Engineering for Symbiosis, Soka University, Hachioji, Tokyo 192-8577, Japan*

### ABSTRACT

Multiple crystal pathways and crystal polymorphs were observed in protic ionic liquids (pILs), which are liquid or solid at room temperature. The pILs studied were methylammonium nitrate (MAN), dimethylammonium nitrate (DAN), ethylammonium nitrate (EAN), and propylammonium nitrate (PAN). MAN and DAN solidified as plastic crystals at room temperature, whereas EAN and PAN are liquids at room temperature. Solid MAN and DAN indicated crystal polymorphs. In spite of its simple molecular structure, complicated phase behaviors such as multiple crystal pathways and crystal polymorphs were observed in liquid EAN at low temperature [Abe *et al.*, *J. Mol. Liq.* **241** (2017) 301]. Liquid PAN also possesses low-temperature anomalies in analogy with liquid EAN. Proton fluctuations, which disturb the crystal nucleation of liquid pILs at room temperature, cause multiple crystal pathways via a non-equilibrium state.

Keywords: Protic ionic liquids; Multiple pathways of crystallization; Crystal polymorphs; Plastic crystal; Proton fluctuations; Hydrogen-bonding network

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