—Supplementary Information—

Search for Active and Inactive Ion Insertion Sites in Organic Crystalline Materials

Harshan Reddy Gopidi,[1,2,3] Alae Eddine Lakraychi,[1,2,3] Abhishek A. Panchal,[1,2,3] Yiming Chen,[1,4] Venkata Surya Chaitanya Kolluru,[1,4,8] Jiaqi Wang,[1,5] Ying Chen,[1,6] Jue Liu,[7] Kamila Wiaderek,[1,5] Maria K.Y. Chan,[1,4] Yan Yao,[1,2,3,*] and Pieremanuele Canepa[1,2,3,#]

[1]Energy Storage Research Alliance, Argonne National Laboratory, 9700 South Cass Avenue, Lemont, IL 60439, USA

[2]Texas Center for Superconductivity at the University of Houston, Houston, TX 77204, USA

[3]Department of Electrical and Computer Engineering, University of Houston, Houston, TX 77204, USA

[4]Center for Nanoscale Materials, Argonne National Laboratory, Lemont, IL 60439, USA

[5]X-ray Science Division, Argonne National Laboratory, Lemont, IL 60439, USA

[6]Physical and Computational Sciences Directorate, Pacific Northwest National Laboratory, Richland, WA 99354, USA

[7]Neutron Sciences Directorate, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

[8]Department of Materials Science and Engineering, The Ohio State University, Columbus, OH 43210, USA

*email: yyao4@central.uh.edu

#email: pcanepa@uh.edu

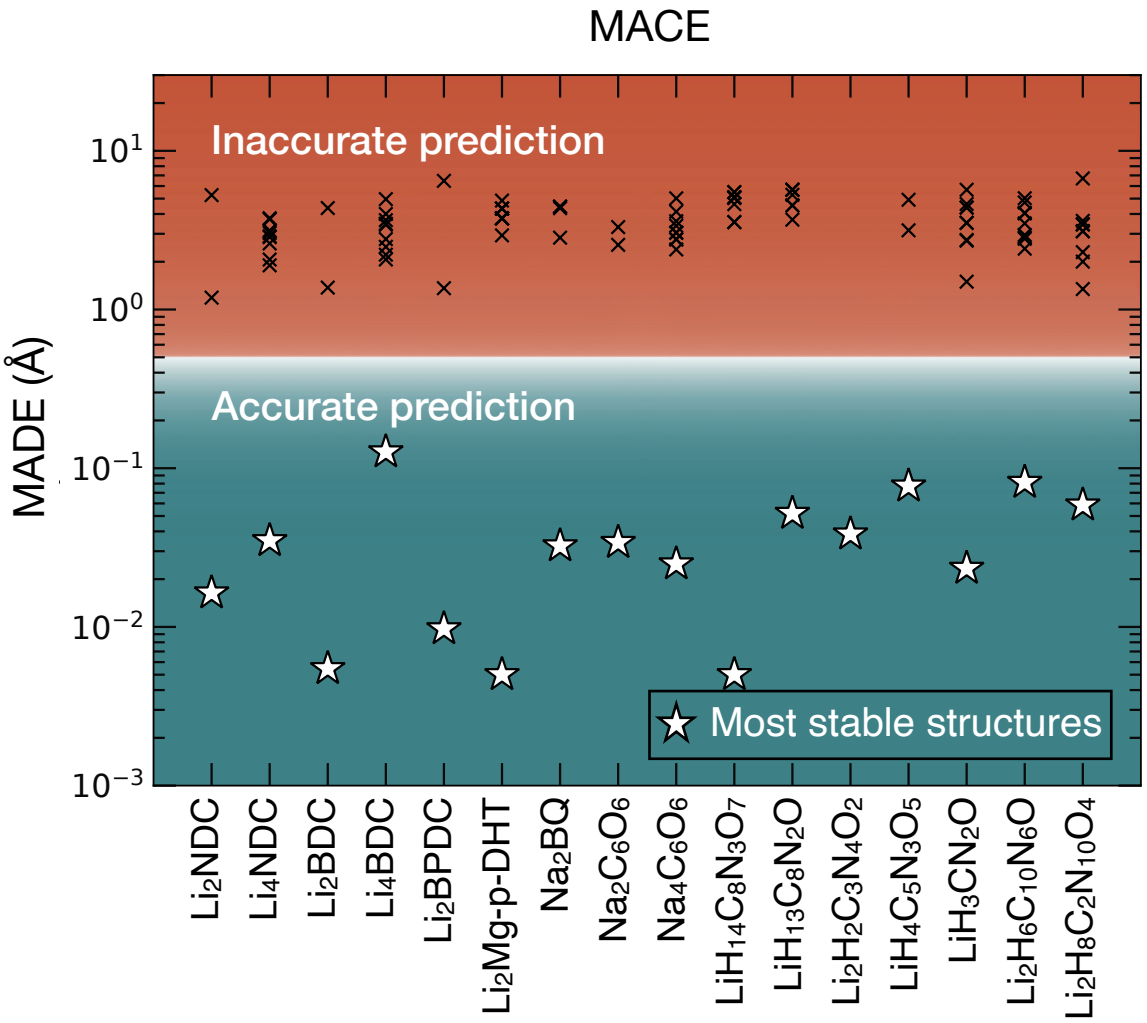

Figure S1. MADE error plot for ion sites determined using ERIN+SIIE. The active and inactive ionic positions have been optimized with MACE. Stars indicate the most stable (lowest energy) ion arrangements in each organic structure, and crosses indicate the thermodynamically unfavorable (high energy) structures.

Table S1. MADE in Å of the ELIISE and SIIE workflow, with ion positions optimized using MACE and DFT for various systems considered. Li_2NDC and Li_4NDC (di and tetra lithium 2,6-naphthalene dicarboxylate),[1,2] Li_2BDC and Li_4BDC (di and tetra lithium 1,4-benzene dicarboxylate),[1,3] Li_2-BPDC (dilithium biphenyl dicarboxylate),[4] Li_2Mg-*p*-DHT (magnesium(2,5-dilithium-oxy)-terephthalate),[5] Na_2BQ (disodium hydroquinone),[6] $Na_2C_6O_6$ (disodium rhodizonate),[7] and $Na_4C_6O_6$ (tetra sodium rhodizonate),[8] $LiH_{14}C_8N_3O_7$ (bis(diacetamide)(nitrato)lithium(I)),[9] $LiH_{13}C_8N_2O$,[10] $Li_2H_2C_3N_4O_2$,[11] $LiH_4C_5N_3O_5$ (Poly[(μ_2-nitrato-κ^2O:O')(μ_2-pyrimidin-ium-2-carboxylato-κ^2 O:O') lithium(I)]),[12] LiH_3CN_2O (Lithium carbamide),[13] $Li_2H_6C_{10}N_6O$ (Lithium tricyanomethanide hemiacetonate),[14] $Li_2H_8C_2N_{10}O_4$ (dilithium bis(1-amino-tetrazol-5-one) dihydrate).[15]

Material	**Database code**	**MADE–MACE**	**MADE–DFT**
Li_2NDC	CCDC 722281	0.016	0.017
Li_4NDC	CCDC 2450777	0.037	0.026
Li_2BDC	CCDC 145822	0.005	0.004
Li_4BDC	CCDC 2450774	0.127	0.048
Li_2-BPDC	CCDC 757041	0.011	0.012
Li_2Mg-*p*-DHT	CCDC 2247807	0.001	0.047
Na_2-BQ	CCDC 192926	0.039	0.024
$Na_2C_6O_6$	CCDC 287536	0.028	0.026
$Na_4C_6O_6$	–	0.028	0.024
$LiH_{14}C_8N_3O_7$	CCDC 1142135	0.008	0.010
$LiH_{13}C_8N_2O$	CCDC 200642	0.047	0.042
$Li_2H_2C_3N_4O_2$	CCDC 2060917	0.040	0.027
$LiH_4C_5N_3O_5$	CCDC 828497	0.076	0.071
LiH_3CN_2O	ICSD 427375	0.023	0.025
$Li_2H_6C_{10}N_6O$	ICSD 428781	0.079	0.026
$Li_2H_8C_2N_{10}O_4$	ICSD 760113	0.059	0.030
Average MADE		0.039	0.029

Table S2. Structure of the organic framework of $Na_4C_6O_6$ (*C2/m*), along with probable Na positions predicted by ELIISE.

Lattice Parameters			
a	12.88 Å		
b	8.17 Å		
c	7.68 Å		
β	120.83°		
Space Group	*C2/m*		
Atomic coordinates			
Species	**Site label**	**Wyckoff label**	**Fractional coordinates**
C	C1	*4i*	0.0336 0.0000 0.2807
C	C2	*4i*	0.1773 0.0000 0.1334
C	C3	*8j*	0.0214 0.1553 0.8224
C	C4	*8j*	0.1235 0.1566 0.0326
O	O1	*4i*	0.1207 0.0000 0.4682
O	O2	*4i*	0.2191 0.5000 0.6950
O	O3	*8j*	0.0126 0.2925 0.2809
O	O4	*8j*	0.1739 0.2941 0.1195
Na	Na1	*2a*	0.0000 0.0000 0.0000
Na	Na2	*2b*	0.0000 0.5000 0.0000
Na	Na3	*2d*	0.0000 0.5000 0.5000
Na	Na4	*4f*	0.2500 0.2500 0.5000
Na	Na5	*4h*	0.0000 0.1667 0.5000
Na	Na6	*4i*	0.1208 0.5000 0.8833
Na	Na7	*4i*	0.1583 0.5000 0.3375
Na	Na8	*4i*	0.2000 0.0000 0.7958
Na	Na9	*8j*	0.1292 0.2458 0.6208

Table S3. Structure of the organic lattice of Na_2BQ (*$P4_2/ncm$*), along with candidate Na positions predicted by ELIISE.

Lattice Parameters			
a	10.5114 Å		
b	10.5114 Å		
c	5.6593 Å		
Space Group	*$P4_2/ncm$*		
Atomic coordinates			
Species	**Site label**	**Wyckoff label**	**Fractional coordinates**
H	H1	*16j*	0.0536 0.1672 0.0986
C	C1	*8i*	0.1798 0.3202 0.0887
C	C2	*16j*	0.1344 0.2025 0.1651
O	O1	*8i*	0.1114 0.3886 0.9307
Na	Na1	*4a*	0.0000 0.0000 0.2500
Na	Na2	*4e*	0.0000 0.5000 0.1905
Na	Na3	*8h*	0.1187 0.1187 0.7500
Na	Na4	*8i*	0.1094 0.3906 0.5298

Table S4. Structure of the Li_4NDC determined from single crystal X-ray diffraction from Ref 1 and predicted from Li_2NDC using the ERIN+SIIE method. The lattice parameters and fractional coordinates are in close agreement between the experimental data and the predicted structure.

		Experimental	Predicted
Lattice Parameters			
a		9.7425 Å	9.6243
b		5.9515 Å	6.0565
c		8.3569 Å	8.3492
β		105.758°	107.268°
Space Group		*$P2_1/c$*	*$P2_1/c$*
Atomic coordinates			
Species	**Wyckoff label**	**Fractional coordinates**	
H	*4e*	0.0130 0.1000 0.3480	0.0114 0.0884 0.3515
H	*4e*	0.1960 0.6730 0.0480	0.1924 0.6541 0.0362
H	*4e*	0.2240 0.2150 0.2990	0.2304 0.2213 0.2910
Li	*4e*	0.3845 0.0886 0.5422	0.3838 0.0800 0.5418
Li	*4e*	0.4309 0.6831 0.6684	0.4295 0.6810 0.6667
C	*4e*	0.0308 0.6077 0.5302	0.0293 0.6089 0.5272
C	*4e*	0.0513 0.2482 0.3895	0.0536 0.2522 0.3937
C	*4e*	0.1602 0.6724 0.5093	0.1577 0.6766 0.5015
C	*4e*	0.1764 0.1820 0.8637	0.1779 0.1749 0.8639
C	*4e*	0.2383 0.5349 0.4204	0.2370 0.5416 0.4159
C	*4e*	0.3569 0.6174 0.3754	0.3572 0.6222 0.3718
O	*4e*	0.4135 0.5129 0.2685	0.4153 0.5186 0.2668
O	*4e*	0.4186 0.6895 0.9353	0.4193 0.6854 0.9326

References

1. Griffiths, K. *et al.* Chemical lithiation reveals the structures of conjugated tetralithium dicarboxylates. *Chem. Commun.* **61**, 11991–11994 (2025).
2. Banerjee, D., Kim, S. J. & Parise, J. B. Lithium Based Metal–Organic Framework with Exceptional Stability. *Crystal Growth & Design* **9**, 2500–2503 (2009).
3. Kaduk, J. A. Terephthalate salts: salts of monopositive cations. *Acta Crystallogr B Struct Sci* **56**, 474–485 (2000).
4. Banerjee, D., Borkowski, L. A., Kim, S. J. & Parise, J. B. Synthesis and Structural Characterization of Lithium-Based Metal–Organic Frameworks. *Crystal Growth & Design* **9**, 4922–4926 (2009).
5. Jouhara, A. *et al.* Unveiling the Peierls effect during electrochemical delithiation/lithiation process in magnesium (2,5-dilithium-oxy)-terephthalate using combined experimental and computational studies. *Energy Storage Materials* **81**, 104456 (2025).
6. Couhorn, U. & Dronskowski, R. Alkali Metal Compounds of Hydroquinone: Synthesis and Crystal Structure. *Zeitschrift anorg allge chemie* **629**, 647–652 (2003).
7. Dinnebier, R. E., Nuss, H. & Jansen, M. Disodium rhodizonate: a powder diffraction study. *Acta Crystallogr E Struct Rep Online* **61**, m2148–m2150 (2005).
8. Yamashita, T., Momida, H. & Oguchi, T. First-Principles Investigation of a Phase Transition in $Na_x C_6 O_6$ as an Organic Cathode Material for Na-ion Batteries: Role of Intermolecule Bonding of $C_6 O_6$. *J. Phys. Soc. Jpn.* **84**, 074703 (2015).
9. Bray, D. D. & Bray, N. F. X-ray structure of bis(diacetamide)(nitrato)lithium(I). *Inorganica Chimica Acta* **111**, L39–L41 (1986).
10. Zi, G., Li, H.-W. & Xie, Z. Synthesis, Structural Characterization, and Reactivity of 'Carbons-Adjacent' *n ido* - and *a rachno* -Carborane Anions of the $C_2 B_{10}$ Systems and Their Metal Complexes. *Organometallics* **21**, 5415–5427 (2002).
11. Liang, C. *et al.* Solvent-Free Lithium/Sodium-Based Metal–Organic Frameworks with Versatile Nitrogen-Rich Ligands: Insight for the Design of Promising Superheat-Resistant Explosives. *Inorg. Chem.* **60**, 9282–9286 (2021).

12. Starosta, W. & Leciejewicz, J. Poly[(μ_2 -nitrato-κ^2 *O*: *O ′*)(μ_2 -pyrimidinium-2-carboxylato-κ^2 *O*: *O ′*)lithium(I)]. *Acta Crystallogr E Struct Rep Online* **67**, m818–m818 (2011).
13. Sawinski, H. & Dronskowski, R. Synthesis and Crystal Structure of a “High-temperature” Phase of Li[H_2 NC(O)NH], the Missing Alkali-Metal Ureate. *Zeitschrift für Naturforschung B* **69**, 651–654 (2014).
14. Reckeweg, O. & DiSalvo, F. J. Two lithium tricyanomethanide compounds: syntheses and single-crystal structure determination of LiK[$C(CN)_3$]$_2$ and Li[$C(CN)_3$]·½ (H_3 C)$_2$ CO. *Zeitschrift für Naturforschung B* **70**, 191–196 (2015).
15. Yang, J. *et al.* Alkaline and Earth Alkaline Energetic Materials Based on a Versatile and Multifunctional 1-Aminotetrazol-5-one Ligand. *Inorg. Chem.* **57**, 15105–15111 (2018).