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Supporting Information for

Layered Electrides as Fluoride Intercalation Anodes

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Table S1. Performance metrics of other fluorine intercalation electrodes calculated in this study or derived from the Materials Project database.[1] The fractional volume change is given relative to the unfluoridated material.

Compound	Mass g/mol F	Gravimetric capacity (mAh/g)	V vs. PbF_2	Delta vol.	E above hull (eV/atom)
Ca2NF	94.16	284.63	-2.31	-0.13	0.00
CaF2	40.08	668.73	-2.32	-0.01	0.00
LiF	6.94	3861.88	-2.31	-0.15	0.00
Sr2NF	189.25	141.62	-2.21	-0.13	0.00
Ba2LiNF2	147.80	181.33	-2.05	-0.09	0.07
Ba3NF3	142.00	188.75	-2.02	-0.01	0.06

Compound	Mass g/mol F	Gravimetric capacity (mAh/g)	V vs. PbF_2	Delta vol.	E above hull (eV/atom)
Y2CF2	94.91	282.38	-2.01	0.09	0.00
Ba2NaOF3	104.55	256.35	-1.90	-0.24	0.06
Sr2TiO3F2	135.55	197.72	-1.89	0.23	0.06
MgF2	24.31	1102.71	-1.72	0.50	0.00
SrSiF	115.71	231.63	-1.55	0.14	0.13
BaSiF	165.42	162.02	-1.51	0.17	0.14
LaSiF	166.99	160.50	-1.47	0.15	0.06
CaSiF	68.16	393.19	-1.37	0.16	0.15
Zr2CF2	97.22	275.67	-1.07	0.37	0.00
Ti2CF2	53.87	497.50	-0.95	0.50	0.00
LaH2F	140.93	190.18	-0.91	0.00	0.17
Sr2FeO3F	279.09	96.03	-0.78	0.09	0.00
Ti2NF2	54.87	488.45	-0.71	0.57	0.03
K6Fe2O5F2	213.14	125.75	-0.67	0.07	0.12
Na4SnO3F2	129.34	207.23	-0.56	0.00	0.18
NaSnNF2	77.85	344.27	-0.45	0.31	0.05
NaSnPF2	86.34	310.43	-0.04	0.29	0.16
PbF2	103.60	258.70	0.00	0.67	0.00
ZnF2	32.70	819.74	0.13	1.36	0.00
K2Sn2O3F3	121.21	221.12	0.14	0.06	0.14
SnF2	59.09	453.61	0.17	0.58	0.00
BiF3	69.66	384.75	0.32	0.59	0.00
KSbO2F2	96.43	277.94	0.37	0.28	0.00
Sr2CrO4F2	145.62	184.05	0.55	0.36	0.14
La2CoO4F	400.75	66.88	0.55	0.04	0.10
La2CoO4F2	200.38	133.76	0.63	0.17	0.05
Sn2OF5	97.14	275.91	0.67	0.21	0.00
KSnF5	107.40	249.54	0.82	0.28	0.00
KAsOF4	85.01	315.28	0.83	-0.01	0.06
TiSn2O4F	349.29	76.73	0.88	0.08	0.08
MgFeSb4O8F2	173.80	77.11	1.06	0.01	0.11
CaSn2F8	195.74	136.92	1.08	0.11	0.09
KAsO2F	146.02	183.55	1.16	0.10	0.09
FeSb2O4F	363.41	73.75	1.18	-0.01	0.08
KVOF4	163.03	164.39	1.32	0.18	0.00
CuF2	31.77	843.53	1.33	1.99	0.00
Ca2MnO4F2	99.55	269.23	1.40	0.43	0.10
SrCuO2	183.17	146.32	1.42	0.11	0.17

FeAsO4F4	48.69	550.43	1.47	0.60	0.11
SrSbF7	152.19	176.11	1.57	0.07	0.00
Mg2MnN2	131.56	203.72	1.59	0.09	0.26
NaCuOF1.25	82.03	326.73	1.63	0.15	0.21

S2: We tested the effect of van der Waals forces using the dispersion correction of Tkatchenko and Scheffler (IVDW = 20 in VASP).[2] The change in structural parameters are shown in Table S2 for Ca_2N and Y_2C. Because the correction did not systematically improve agreement with experiment, we did not apply it to the calculations discussed in the main text.

Table S2: Theoretical cell volume and *c*-axis length for relevant electrode materials, with or without van der Waals interactions, compared to experimental values.

Compound	Volume (PBE)	Volume (IVDW=20)	Volume (Exp.)	*c*-axis (PBE)	*c*-axis (IVDW=20)	*c*-axis (Exp.)
Ca_2N[3]	217.0 Å^3	199.9 Å^3	216.6 Å^3 (2 K)	19.24 Å	18.81 Å	19.06 Å
Ca_2NF	189.5 Å^3	172.2 Å^3	N/A	17.43 Å	16.66 Å	N/A
Y_2C[4]	208.5 Å^3	191.0 Å^3	203.5 Å^3 (298 K)	18.41 Å	17.67 Å	17.96 Å
Y_2CF_2[5]	75.50 Å^3	72.26 Å^3	73.25 Å^3 (300 K)	6.453 Å	6.394 Å	6.297 Å

S3: We list below the most favorable decomposition pathways identified by the Materials Project, and their decomposition energies. Positive values correspond to stable materials which are on the convex hull.

$$2Ca_2N \rightarrow Ca_3N_2 + Ca,\ E_{decomp} = +99\ meV/atom$$

$$2Ca_2NF \rightarrow Ca_3N_2 + CaF_2,\ E_{decomp} = +72\ meV/atom$$

$$5Y_2C \rightarrow Y_4C_5 + 6Y,\ E_{decomp} = +91\ meV/atom$$

$$9Y_2CF_2 \rightarrow Y_4C_5 + 4Y_2C + 6YF_3,\ E_{decomp} = +84\ meV/atom$$

S4: In this section, we provide the data and fitting plots obtained from AIMD simulations, which we used to obtain the diffusivity and the activation energy for diffusion in Y_2CF_2.

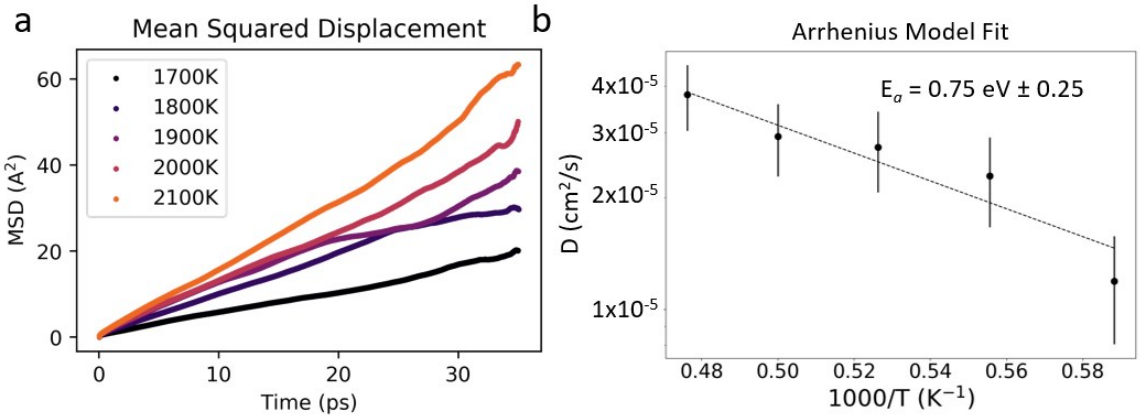

Figure S1: a. Mean squared displacement over time for MD simulations of stoichiometric Y_2CF_2 at different temperatures. b. The Arrhenius relationship of diffusivity vs. temperature using the data derived from *a*.

Additional References

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