Supporting Information

Fluoride-ion shuttle battery with high volumetric energy density

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Table S1. Thermodynamically expected fluorination–defluorination potentials of several materials and calculated cell voltages and one-third of theoretical energy densities of several combinations of cathode and anode.

$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	Mat	erial	Potentia V vs. Pb I	al PbF ₂	Theoretical voltage V	One third of volumetric energy density Wh/L
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Cathode	Anode	Cathode	Anode		
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	Ag	CaF_2	1.26	-2.85	4.11	1238
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	Ag	BaF_2	1.26	-2.80	4.06	885
Ag Car 1.26 -2.41 3.67 900 Ag Car Ca -2.37 3.63 916 Ag AF 1.26 -1.72 2.99 867 Cu CaF 0.65 -2.85 3.49 1973 Cu BaF 0.65 -2.35 2.99 1988 Cu Lar 0.65 -2.35 2.99 1988 Cu CaF 0.65 -2.37 3.01 1445 Cu CaF 0.66 -2.37 3.01 1445 Cu CaF 0.66 -2.37 3.01 1445 Cu CaF 0.64 -2.35 2.99 2697 Co BaF 0.64 -2.37 3.01 2089 Co CaF 0.64 -2.37 3.01 2089 Co LaF 0.64 -2.17 3.01 2089 Co CaF 0.26 -2.35 2.61<	Ag	MgF ₂	1.26	-2.35	3.61	1294
Ag Alf 1.20 -1.21 2.00 910 Ag Alf 1.20 -1.72 2.99 867 Cu BaF2 0.65 -2.85 3.49 1973 Cu Maf5 0.65 -2.80 3.44 1432 Cu Maf5 0.65 -2.37 3.01 1445 Cu Caf7 0.65 -2.37 3.01 1445 Cu Alf3 0.65 -1.72 2.37 1319 Co Caf2 0.64 -2.85 3.49 2715 Co Baf2 0.64 -2.30 3.44 2014 Co Maf2 0.64 -2.30 3.44 2014 Co Laf3 0.64 -2.31 3.05 2069 Co Laf3 0.64 -2.41 3.05 2069 Co Alf3 0.26 -2.85 3.11 1611 Sb Baf2 0.26 -2.35	Ag	Lar ₃	1.20	-2.41	3.67	900
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	Ag	AlF ₃	1.20	-1.72	2.99	867
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	Cu	CaF_2	0.65	-2.85	3.49	1973
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	Cu	BaF ₂	0.65	-2.80	3.44	1432
Cu $CeF_3 = 0.65 - 2.37 = 3.01 = 1443$ Cu $AF_3 = 0.65 - 2.37 = 3.01 = 1445$ Cu $AF_3 = 0.65 - 1.72 = 2.37 = 1319$ Co $CaF_2 = 0.64 - 2.85 = 3.49 = 2715$ Co $MaF_2 = 0.64 - 2.35 = 2.99 = 2697$ Co $LaF_3 = 0.64 - 2.41 = 3.05 = 2063$ Co $AF_3 = 0.64 - 2.41 = 3.05 = 2063$ Co $AF_3 = 0.64 - 2.17 = 3.01 = 2089$ Co $AF_3 = 0.64 - 1.72 = 2.37 = 1868$ Sb $CaF_2 = 0.26 - 2.85 = 3.11 = 1611$ Sb $MaF_2 = 0.26 - 2.85 = 3.11 = 1611$ Sb $MaF_2 = 0.26 - 2.35 = 2.61 = 1488 = 55$ Sb $LaF_3 = 0.26 - 2.41 = 2.67 = 1399$ Sb $LaF_3 = 0.26 - 2.41 = 2.67 = 1399$ Sb $AF_3 = 0.26 - 1.72 = 1.99 = 1171$ Bi $CaF_2 = 0.30 - 2.85 = 3.15 = 1492$ Bi $BaF_2 = 0.30 - 2.85 = 3.15 = 1492$ Bi $BaF_2 = 0.30 - 2.80 = 3.10 = 1223$ Bi $MaF_3 = 0.36 - 2.41 = 2.67 = 13355$ Bi $LaF_3 = 0.30 - 2.41 = 2.71 = 13355$ Bi $LaF_3 = 0.30 - 2.37 = 2.63 = 1369$ Bi $LaF_3 = 0.30 - 2.37 = 2.67 = 1340$ Bi $LaF_3 = 0.30 - 1.72 = 2.02 = 1114$ Ni $CaF_2 = 0.07 - 2.85 = 2.92 = 1675$ Ni $MaF_2 = 0.07 - 2.85 = 2.92 = 1675$ Ni $MaF_3 = 0.07 - 2.35 = 2.42 = 1636$ Ni $MaF_3 = 0.07 - 2.37 = 2.44 = 2.47 = 1182$ Ni $MaF_3 = 0.07 - 2.37 = 2.44 = 1196$ Ni $MaF_3 = 0.07 - 2.37 = 2.44 = 1196$ Ni $MaF_3 = 0.07 - 2.37 = 2.44 = 1196$ Ni $MaF_3 = 0.07 - 2.37 = 2.44 = 1196$ Ni $MaF_3 = 0.07 - 2.35 = 2.85 = 1190$ Pb $CaF_3 = 0.00 - 2.37 = 2.37 = 2.37 = 958$ Pb $AF_3 = 0.00 - 2.41 = 2.41 = 2.41 = 952$ Pb $AF_3 = 0.00 - 2.41 = 2.41 = 2.41 = 952$ Pb $AF_3 = 0.00 - 2.41 = 2.41 = 2.41 = 952$ Pb $AF_3 = 0.00 - 2.41 = 2.41 = 2.25 = 1502$ Fe $MaF_3 = 0.00 - 2.41 = 2.41 = 2.25 = 1502$ Fe $LaF_3 = -0.16 - 2.41 = 2.25 = 1502$ Fe $LaF_3 = -0.16 - 2.41 = 2.25 = 1502$ Fe $LaF_3 = -0.16 - 2.41 = 2.25 = 1502$ Fe $LaF_3 = -0.16 - 2.41 = 2.25 = 1502$ Fe $LaF_3 = -0.16 - 2.41 = 2.25 = 1502$ Fe $LaF_3 = -0.16 - 2.41 = 2.25 = 1502$ Fe $LaF_3 = -0.16 - 2.41 = 2.25 = 1502$ Fe $LaF_3 = -0.16 - 2.41 = 2.25 = 1502$ Fe $LaF_3 = -0.16 - 2.41 = 2.25 = 1502$ Fe $LaF_3 = -0.16 - 2.41 = 2.25 = 1502$	Cu	MgF ₂	0.65	-2.35	2.99	1988
Cu AE 2.37 130 Cu AE 3.65 -1.72 2.37 1319 Co CaF ₂ 0.64 -2.85 3.49 2715 Co BaF ₂ 0.64 -2.85 2.99 2697 Co LaF ₃ 0.64 -2.37 3.01 2089 Co AEF ₃ 0.64 -2.37 3.01 2089 Co AF ₅ 0.64 -1.72 2.37 1868 Sb CaF ₂ 0.26 -2.85 3.11 1611 Sb BaF ₂ 0.26 -2.85 3.10 1300 Sb LaF ₃ 0.26 -2.41 2.67 1399 Sb CaF ₂ 0.26 -2.37 2.63 1406 Sb LaF ₃ 0.26 -1.72 1.99 1171 Bi CaF ₂ 0.30 -2.37 2.67 1340 Bi <td>Cu</td> <td>CeF.</td> <td>0.05</td> <td>-2.41</td> <td>3.05</td> <td>1445</td>	Cu	CeF.	0.05	-2.41	3.05	1445
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	Cu	AlF ₃	0.65	-1.72	2.37	1319
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Co	CaF_2	0.64	-2.85	3.49	2715
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Co	BaF_2	0.64	-2.80	3.44	2014
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Co	MgF_2	0.64	-2.35	2.99	2697
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Co	LaF ₃	0.64	-2.41	3.05	2063
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Co	AlF ₂	0.64	-2.37	2.37	2089
S0 Car_2 0.26 -2.85 3.11 1011 Sb MgF_2 0.26 -2.80 3.066 1300 Sb MgF_2 0.26 -2.35 2.61 1488 Sb LaF_3 0.26 -2.37 2.63 1406 Sb AlF_3 0.26 -1.72 1.99 1171 Bi BaF_2 0.30 -2.85 3.15 1492 Bi BaF_2 0.30 -2.85 2.65 1369 Bi LaF_3 0.30 -2.37 2.67 1340 Bi CaF_2 0.07 -2.85 2.92 1675 Bi AlF_3 0.30 -1.72 2.02 1114 Ni CaF_2 0.07 -2.85 2.92 1675 Ni BaF_2 0.07 -2.35 2.42 1636 Ni MgF_2 0.07 -2.37		,	0.04	2.05		
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	SD	Car ₂ BaF	0.26	-2.85	3.11	1011
Sb LaF_3 0.26 -2.41 2.67 1399 Sb CeF_3 0.26 -2.37 2.63 1406 Sb AIF_3 0.26 -1.72 1.99 1171 Bi CaF_2 0.30 -2.85 3.15 1492 Bi BaF_2 0.30 -2.80 3.10 1223 Bi MgF_2 0.30 -2.41 2.71 1335 Bi LaF_3 0.30 -2.37 2.67 1340 Bi AIF_3 0.30 -2.37 2.67 1340 Bi AIF_3 0.30 -1.72 2.02 1114 Ni CaF_2 0.07 -2.85 2.92 1675 Ni BaF_2 0.07 -2.35 2.42 1636 Ni LaF_3 0.07 -2.37 2.44 1196 Ni LaF_3 0.07 -1.72 1.79 1009 Pb BaF_2 0.00 -2.35	Sh	MgF ₂	0.26	-2.80	2 61	1488
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Sb	LaF ₃	0.26	-2.41	2.67	1399
Sb AIF ₃ 0.26 -1.72 1.99 1171 Bi CaF ₂ 0.30 -2.85 3.15 1492 Bi BaF ₂ 0.30 -2.80 3.10 1223 Bi MgF ₂ 0.30 -2.35 2.65 1369 Bi LaF ₃ 0.30 -2.37 2.67 1340 Bi AIF ₃ 0.30 -1.72 2.02 1114 Ni CaF ₂ 0.07 -2.85 2.92 1675 Ni BaF ₂ 0.07 -2.35 2.42 1636 Ni MgF ₂ 0.07 -2.35 2.42 1636 Ni LaF ₃ 0.07 -2.37 2.44 1196 Ni CeF ₃ 0.07 -1.72 1.79 1009 Pb CaF ₂ 0.00 -2.85 2.85 1190 Pb BaF ₂ 0.00 -2.37 2.35 1103 Pb LaF ₃ 0.	Sb	CeF ₃	0.26	-2.37	2.63	1406
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Sb	AlF ₃	0.26	-1.72	1.99	1171
Bi BaF_2 0.30 -2.80 3.10 1223 Bi MgF_2 0.30 -2.35 2.65 1369 Bi LaF_3 0.30 -2.41 2.71 1335 Bi CeF_3 0.30 -2.37 2.67 1340 Bi AlF_3 0.30 -1.72 2.02 1114 Ni CaF_2 0.07 -2.85 2.92 1675 Ni BaF_2 0.07 -2.85 2.92 1675 Ni BaF_2 0.07 -2.85 2.42 1636 Ni LaF_3 0.07 -2.37 2.44 1196 Ni CaF_3 0.07 -1.72 1.79 1009 Pb CaF_2 0.00 -2.85 2.85 1190 Pb BaF_2 0.00 -2.35 2.35 1103 Pb DaF_2 0.00 -2.37 2.37 958 Pb LaF_3	Bi	CaF_2	0.30	-2.85	3.15	1492
Bi MgF2 0.30 -2.33 2.05 1309 Bi LaF3 0.30 -2.41 2.71 1335 Bi CeF3 0.30 -2.37 2.67 1340 Bi AF3 0.30 -1.72 2.02 1114 Ni CaF2 0.07 -2.85 2.92 1675 Ni BaF2 0.07 -2.35 2.42 1636 Ni MgF2 0.07 -2.35 2.42 1636 Ni LaF3 0.07 -2.37 2.44 1196 Ni CeF3 0.07 -1.72 1.79 1009 Pb CaF2 0.00 -2.85 2.85 1190 Pb BaF2 0.00 -2.35 2.35 1103 Pb MgF2 0.00 -2.35 2.35 1103 Pb LaF3 0.00 -2.37 2.37 958	Bi	BaF ₂	0.30	-2.80	3.10	1223
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	B1 P;	MgF ₂	0.30	-2.35	2.05	1309
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Bi	CeF.	0.30	-2.41	2.71	1335
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	Bi	AlF ₃	0.30	-1.72	2.02	1114
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Ni	CaF ₂	0.07	-2.85	2.92	1675
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Ni	BaF_2	0.07	-2.80	2.86	1206
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Ni	MgF_2	0.07	-2.35	2.42	1636
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	Ni	LaF_3	0.07	-2.41	2.47	1182
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	N1 Ni	CeF ₃ AIF ₂	0.07	-2.37	2.44	1196
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$,	0.00	2.02	2.00	1100
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	Pb	CaF ₂	0.00	-2.85	2.85	024
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	PD Ph	MgF.	0.00	-2.80	2.80	1103
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	Pb	LaF.	0.00	-2.41	2.41	952
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	Pb	CeF ₃	0.00	-2.37	2.37	958
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	Pb	AlF ₃	0.00	-1.72	1.72	780
Fe BaF_2 -0.16 -2.80 2.64 1521 Fe MgF_2 -0.16 -2.35 2.19 1928 Fe LaF_3 -0.16 -2.41 2.25 1502 Fe CeF_3 -0.16 -2.37 2.21 1514 Fe AlF_3 -0.16 -1.72 1.56 1218	Fe	CaF_2	-0.16	-2.85	2.69	2050
re Mgr_2 -0.16 -2.35 2.19 1928 Fe LaF_3 -0.16 -2.41 2.25 1502 Fe CeF_3 -0.16 -2.37 2.21 1514 Fe AlF_3 -0.16 -1.72 1.56 1218	Fe	BaF ₂	-0.16	-2.80	2.64	1521
re Lar ₃ -0.10 -2.41 2.25 1502 Fe CeF ₃ -0.16 -2.37 2.21 1514 Fe AlF ₃ -0.16 -1.72 1.56 1218	Fe	MgF ₂	-0.16	-2.35	2.19	1928
Fe AlF ₃ -0.16 -1.72 1.56 1218	re	Lar ₃	-0.10	-2.41	2.25	1502
	Fe	AlF ₃	-0.16	-1.72	1.56	1218



IPF MAP: Normal Direction

Figure S1. a) Illustration of the pre-treatment of the LaF₃ substrate for EBSD analysis. The raw LaF₃ surface was etched by Ar for obtaining a very smooth surface. b) SEM image of the pre-treated LaF₃ surface. The etched surface and EBSD analysis area are indicated by green and yellow, respectively. c) Crystal orientation mapping analysed by EBSD. Because there is no grain boundary of the LaF₃ substrate, the crystal orientation map was given by only one colour in a relatively wide area of 500 μ m². This indicates that the LaF₃ substrate used in this study is a single crystal.

1		• ⁻¹⁻¹⁻³	• ⁰⁻²⁻² • ¹⁻³⁻¹
d ₃	$\begin{array}{c} d_1 \\ 1\overline{1}1 \\ d_2 \end{array}$,000 ● ¹⁻¹¹ _2-2:
022	[™] 113		• ⁰²² 113
Material	Cu	d-sp	acing
Zone axis Crystal system	[2 1 1] Cubic	Experimental / nm	Simulated / nm
Space group	$Fm\overline{3}m$	(d ₁) 0.201	(d ₁₋₁₁) 0.2087
	a = 0.361 nm	(d ₂) 0.110	(d ₁₁₃) 0.1090
		(d ₃) 0.130	(d ₀₂₂) 0.1278

Figure S2. Experimental (upper left) and simulated (upper right) ED patterns collected from cross section of Cu half-cells with various charge states. Miller indexes are also shown in the simulated ED pattern. Italic numbers shown in the experimental ED pattern indicate the analysed points or areas as shown in Figure 3 in the text. The assumed crystal structure for simulation is shown in the lower right. Three sets of d-spacing values calculated from experimental ED pattern and determined by simulation are shown in lower left. All sets of d-spacing values were almost identical, indicating that the assumed crystal structure is true.



Material	CuF_2	d-spacing			
Zone axis	[2 2 1]	Experimental / nm	Simulated / nm		
Space group	$\frac{1}{P} \frac{2}{2_1} c$	(d ₁) 0.156	(d ₀₋₁₂) 0.1550		
	a = 0.459 nm h = 0.454 nm	(d ₂) 0.150	(d ₁₀₂) 0.1496		
	c = 0.332 nm	(d ₃) 0.323	(d ₁₁₀) 0.3217		
	$\beta = 96.67^{\circ}$				

3	$\begin{array}{c} 111\\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1$	•3-11 •2-20 •1-3-1 •	.222 .111 .000 131 131 131 131 220 220 220 31-1
Material	Cu	d-spa	acing
Zone axis	[1 1 2] Cubic	Experimental / nm	Simulated / nm
Space group	$Fm\overline{3}m$	(d ₁) 0.208	(d ₁₁₁) 0.2087
	a = 0.361 nm	(d ₂) 0.109	(d ₋₁₃₁) 0.1090
		(d ₃) 0.125	(d ₋₂₂₀) 0.1278

4 111 d ₁	$d_2, 2\overline{11}$ $d_3, 1\overline{22}$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$
Material	LaF ₃	d-spa	acing
Zone axis	$\begin{bmatrix} 0 \ \overline{1} \ 1 \end{bmatrix}$	Experimental / nm	Simulated / nm
Space group	$P\overline{3}c1$	(d ₁) 0.320	(d ₁₁₁) 0.3228
	a = 0.7185 nm	(d ₂) 0.322	(d ₂₋₁₋₁) 0.3228
	a = 0 (2b1 nm)		

Figure S3. Experimental (upper left) and simulated (upper right) ED patterns collected from the cross section of LaF₃ half-cells with various charge states. Miller indexes are also shown in the simulated ED pattern. Italic numbers shown in experimental ED pattern indicate the analysed points or areas as shown in Figure 5 in the text. The assumed crystal structure for simulation is shown in the lower right. Three sets of d-spacing values calculated from the experimental ED pattern and determined by simulation are shown in lower left. All sets of d-spacing values were almost identical, indicating that the assumed crystal structure is true.

0 124 d ₁	$\begin{array}{c} 0\overline{32} \\ d_2 \\ d_3 \end{array} \overline{112} \\ d_3 \end{array}$	2-1-6 1-2 22-4 11-2 23-3 12-1 140 0 151	1 -4 -5 -4 0 -3 -2 -1 -3 1 -2 -4 3 -1 -1 2 -2 -2 4 -2 -1 5 -3 2 -1 2 4 -1 3 5
			1997 - A.
Material	LaF ₃	d-spa	acing
Material Zone axis	LaF ₃ [8 2 3]	d-spa Experimental / nm	ncing Simulated / nm
Material Zone axis Crystal system Space group	LaF ₃ [8 2 3] trigonal <i>P</i> 3c1	d-spa Experimental / nm (d ₁) 0.162	acing Simulated / nm (d ₁₋₂₋₄) 0.1636
Material Zone axis Crystal system Space group	LaF ₃ [8 $\overline{2}$ 3] trigonal $P\overline{3}c1$ a = 0.7185 nm	d-spa Experimental / nm (d ₁) 0.162 (d ₂) 0.180	acing Simulated / nm (d ₁₋₂₋₄) 0.1636 (d ₀₋₃₋₂) 0.1806



Material	La	a d-spacing				
Zone axis	[2 1 1]	Experimental / nm	Simulated / nm			
Space group	$P6_3/mmc$	(d ₁) 0.222	(d ₁₀₂) 0.2218			
	a = 0.375nm c = 0.607nm	(d ₂) 0.177	(d ₁₁₁) 0.1798			
	γ = 120 °	(d ₃) 0.289	(d ₀₁₋₁) 0.2870			





Material	La	d-spacing			
Zone axis	[1 3 3]	Experimental / nm	Simulated / nm		
Space group	ce group P6 ₃ /mmc	(d ₁) 0.318	(d ₀₁₋₁) 0.31532		
a = 0.375 c = 0.607 β = 120 °	a = 0.375nm c = 0.607nm	(d ₂) 0.119	(d ₃₋₁₂) 0.1181		
	$\beta = 120^{\circ}$	(d ₃) 0.120	(d ₃₋₂₃) 0.1209		

	• 030 300 110 • -120 2-10 • -330 • -330 • -210 • 1-20 • -1-10 • -300 • 0-30
Material LaF ₃	d-spacing

matorial	Laig	u ope	lonig
Zone axis	[0 0 1] trianal	Experimental / nm	Simulated / nm
Crystal system	trigonal		
Space group	$P\overline{3}c1$	(d ₁) 0.355	(d ₋₁₂₀) 0.3593
	a = 0.7185 nm	(d ₂) 0.351	(d ₁₁₀) 0.3593
	$\gamma = 120^{\circ}$	(d ₃) 0.351	(d ₂₋₁₀) 0.3593

10 d	$ \begin{array}{c} 002\\ d_{2}\\ d_{3}\\ \overline{151}\\ \overline{151}\\ \end{array} $	• 1-5 • 1-51 • 1-50 • 1-5-1 • 1-5-2 1-5-3 • 00-4 • 00-6	53 006 004 002 000 2 000 2 000 000 00
Material	LaF ₃	d-spa	icing
Zone axis	[5 1 0]	Experimental / nm	Simulated / nm
Space group	$P\overline{3}c1$	(d ₁) 0.371	(d ₀₀₂) 0.36755
	a = 0.7185 nm c =0 7351 nm	(d ₂) 0.133	(d ₋₁₅₁) 0.13352
	$\gamma = 120^{\circ}$	(d ₃) 0.132	(d ₋₁₅₋₁) 0.13352

 Table S2. Detailed conditions for the sputtering of thin-film electrodes.

Thin film	Thickness nm	RF power W	Sputtering rate nm s ⁻¹	Flow gas	pressure Pa	substrate temperature °C
Cu	2.3 or 60	50	0.033	Ar	1.0	RT
W	50	120	0.033	Ar	1.0	RT
Pt	50	50	0.025	Ar	1.0	RT
Pb	1000	50	0.025	Ar	1.0	RT
PbF ₂	20	100	0.25	Ar	1.0	300