

Supporting Information

Text S1. Chemicals

Betaine hydrochloride (99%) was obtained from Acros Organics. Lithium bis(trifluoromethane)sulfonimide lithium (99.95% purity) was obtained from Sigma Aldrich (Sigma-Aldrich, St. Louis, MO, USA) and Iolitec (IoLiTec-Ionic Liquids Technologies GmbH, Heilbronn, Germany). Concentrated hydrochloric acid (37 wt. %, 99.999% trace metals basis) and concentrated nitric acid (70% wt. %, 99.999% trace metals basis) were obtained from Sigma Aldrich. Ascorbic acid (AA) and sodium nitrate were obtained from Sigma Aldrich or Fisher Scientific. All chemicals were obtained at the highest purity and used without further purification. Deionized water (≥ 18 m Ω -cm) was produced from a Milli-Q water purification system (Millipore, Billerica, MA, USA).

Text S2. IL Synthesis

Aqueous solutions of betaine chloride (HbetCl) and lithium bis(trifluoromethylsulfonyl)imide (LiTf₂N) were prepared to achieve an equimolar ratio of Hbet:Tf₂N and combined at room temperature while stirring. After one hour, the aqueous phase was separated from the IL phase. No further removal of chloride impurities was performed. [Hbet][Tf₂N] is hygroscopic and absorbs 13% water by mass. IL samples were stored as water saturated samples and sealed at room temperature.

Table S1. Leaching efficiency, *L* (%), of REEs, Th, and U.

Element	93927	92801	93932	93951	93964	94012
Sc	48.4 ± 0.5	56.2 ± 6.5	-	-	73.9 ± 6.1	78.4 ± 9.5
Y	74.2 ± 0.2	85.0 ± 0.5	100.7 ± 9.3	94.7 ± 9.0	113.8 ± 8.3	123.3 ± 9.8
La	36.3 ± 0.5	81.6 ± 2.1	92.7 ± 7.1	76.9 ± 9.3	102.7 ± 7.4	77.1 ± 5.5
Ce	53.1 ± 5.2	81.4 ± 2.0	88.4 ± 8.9	70.2 ± 10.9	107.3 ± 6.8	74.3 ± 5.1
Pr	0.0 ± 0.0	103.8 ± 8.7	18.4 ± 0.7	42.0 ± 18.4	107.0 ± 42.0	104.3 ± 31.1
Nd	88.7 ± 0.9	106.5 ± 1.1	100.4 ± 5.4	102.9 ± 11.1	134.7 ± 10.8	110.7 ± 12.2
Sm	113.3 ± 3.6	88.2 ± 5.8	74.2 ± 8.7	70.1 ± 13.7	149.7 ± 21.3	111.2 ± 4.6
Eu	46.4 ± 0.6	65.5 ± 6.7	53.5 ± 4.9	61.1 ± 6.7	105.1 ± 1.5	85.3 ± 6.8
Gd	187.0 ± 2.5	107.1 ± 6.3	145.3 ± 3.2	73.7 ± 5.2	106.7 ± 4.3	103.8 ± 13.2
Tb	0.0 ± 0.0	63.9 ± 10.7	23.0 ± 6.6	15.0 ± 2.0	26.5 ± 19.9	33.6 ± 14.7
Dy	51.6 ± 2.4	83.4 ± 2.6	86.3 ± 7.8	80.0 ± 8.5	106.4 ± 9.5	75.8 ± 8.3
Ho	0.0 ± 0.0	64.9 ± 0.0	27.3 ± 0.5	73.8 ± 28.2	115.5 ± 8.1	124.0 ± 41.9
Er	49.0 ± 7.9	62.3 ± 0.0	49.0 ± 2.0	67.0 ± 7.9	116.3 ± 6.0	127.7 ± 31.8
Yb	87.9 ± 1.0	87.4 ± 0.3	95.1 ± 8.9	74.3 ± 6.0	116.8 ± 9.1	89.8 ± 7.5
Lu	55.1 ± 12.0	-	31.2 ± 2.0	38.4 ± 4.3	124.9 ± 0.7	117.4 ± 19.4
ΣCritical REE	76.0 ± 0.8	91.5 ± 1.1	91.1 ± 6.3	92.8 ± 9.4	120.5 ± 9.3	111.1 ± 11.9
ΣREE	59.6 ± 2.2	85.6 ± 2.3	81.8 ± 6.4	76.7 ± 10.6	110.7 ± 9.3	90.8 ± 9.1
U	0.0 ± 0.0	0.0 ± 0.0	-	-	0.0 ± 0.0	-
Th	208.7 ± 12.5	72.7 ± 2.3	-	-	161.0 ± 24.1	-

Note: a “-” indicated that the original mass concentration of the element was unknown for the CFA.

Table S2. Mass distribution of REEs, Th, and U in the alkaline solution after pretreatment (PT%).

Element	92801	93932	93951	93964	94012
Sc	0.0 ± 0.0	-	-	0.3 ± 0.4	0.3 ± 0.4
Y	0.2 ± 0.0	0.3 ± 0.1	0.3 ± 0.0	0.7 ± 0.4	0.8 ± 0.4
La	0.8 ± 0.1	1.9 ± 0.6	1.3 ± 0.2	2.4 ± 0.6	1.7 ± 0.7
Ce	1.6 ± 0.6	1.5 ± 2.1	0.0 ± 0.0	1.7 ± 0.7	1.8 ± 1.0

Pr	0.0 ± 0.0	0.0 ± 0.0	5.3 ± 0.8	19.2 ± 27.1	9.1 ± 12.9
Nd	6.3 ± 2.8	10.6 ± 1.1	9.7 ± 0.6	17.0 ± 0.2	17.0 ± 2.5
Sm	6.5 ± 3.2	1.8 ± 0.7	3.3 ± 3.5	24.0 ± 11.4	16.6 ± 3.7
Eu	5.0 ± 4.4	1.6 ± 0.1	4.5 ± 1.7	3.3 ± 3.6	4.3 ± 0.1
Gd	0.0 ± 0.0	17.3 ± 7.3	7.0 ± 1.5	16.4 ± 2.7	12.6 ± 3.0
Tb	11.8 ± 1.4	15.5 ± 6.7	8.9 ± 9.3	12.4 ± 17.5	0.0 ± 0.0
Dy	0.0 ± 0.0	0.5 ± 0.7	0.8 ± 0.9	1.7 ± 2.0	0.7 ± 1.0
Ho	3.4 ± 0.9	8.1 ± 0.8	14.2 ± 17.3	37.8 ± 8.2	34.3 ± 43.3
Er	0.0 ± 0.0	2.8 ± 3.9	0.0 ± 0.0	26.9 ± 1.8	33.7 ± 28.2
Yb	1.6 ± 0.7	6.2 ± 0.3	4.1 ± 1.4	3.7 ± 0.8	5.4 ± 0.7
Lu	-	8.6 ± 0.2	9.0 ± 0.6	32.0 ± 4.2	30.6 ± 25.8
U	0.0 ± 0.0	-	-	0.0 ± 0.0	-
Th	5.2 ± 2.9	-	-	12.5 ± 14.7	-

Note: a “-” indicated that the original mass concentration of the element was unknown for the CFA.

Table S3. Distribution coefficient, *D*, of REEs, Th, and U.

Element	93927	92801	93932	93951	93964	94012
Sc	†	270.3 ± 73.2	†	†	†	†
Y	3.2 ± 0.0	1.1 ± 0.1	1.4 ± 0.0	1.2 ± 0.0	1.3 ± 0.1	1.5 ± 0.2
La	†	1.3 ± 0.1	1.6 ± 0.1	1.3 ± 0.0	1.4 ± 0.1	1.7 ± 0.3
Ce	5.9 ± 1.9	1.6 ± 0.1	1.8 ± 0.0	1.5 ± 0.1	1.6 ± 0.1	2.0 ± 0.3
Pr	0.0 ± 0.0	2.3 ± 0.6	†	6.2 ± 0.0	†	†
Nd	4.4 ± 0.3	1.9 ± 0.1	2.1 ± 0.1	1.7 ± 0.0	1.8 ± 0.1	2.2 ± 0.3
Sm	2.2 ± 0.0	2.1 ± 0.1	1.9 ± 0.0	1.9 ± 0.1	1.6 ± 0.1	2.0 ± 0.3
Eu	2.2 ± 0.2	1.6 ± 0.0	1.6 ± 0.0	1.6 ± 0.2	1.3 ± 0.2	1.6 ± 0.3
Gd	0.8 ± 0.0	1.3 ± 0.0	2.0 ± 0.3	1.6 ± 0.1	1.6 ± 0.0	1.9 ± 0.2
Tb	0.0 ± 0.0	182.3 ± 174.6	†	†	†	†
Dy	†	1.5 ± 0.0	1.8 ± 0.0	1.6 ± 0.0	1.7 ± 0.2	2.0 ± 0.3
Ho	0.0 ± 0.0	1.0 ± 0.0	2.3 ± 0.7	1.7 ± 0.0	1.9 ± 0.2	2.2 ± 0.2
Er	3.9 ± 1.6	0.9 ± 0.2	1.7 ± 0.2	1.4 ± 0.0	1.4 ± 0.3	1.3 ± 0.2
Yb	3.3 ± 0.1	1.2 ± 0.1	1.4 ± 0.0	1.3 ± 0.1	1.4 ± 0.1	1.6 ± 0.2
Lu	5.3 ± 0.0	1.2 ± 0.2	1.8 ± 0.1	1.4 ± 0.2	1.8 ± 0.3	2.1 ± 0.4
U	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Th	2.5 ± 0.4	6.4 ± 1.2	2.1 ± 0.2	4.8 ± 1.2	1.7 ± 0.0	1.8 ± 0.3

Note: a † indicated that $D \gg 1000$ because no elements were detected in the AQ phase.

Table S4. Recovery efficiency, *R* (%), of REEs, Th, and U.

Element	93927	92801	93932	93951	93964	94012
Sc	48.4 ± 0.5	56.0 ± 6.6	-	-	73.6 ± 5.8	78.2 ± 9.9
Y	56.5 ± 0.3	45.0 ± 1.7	58.2 ± 5.3	51.5 ± 5.9	64.0 ± 6.9	73.5 ± 10.3
La	36.3 ± 0.5	46.2 ± 0.1	55.6 ± 5.3	42.2 ± 5.5	57.9 ± 6.1	47.4 ± 6.6
Ce	44.9 ± 2.1	49.0 ± 0.8	56.3 ± 4.9	42.0 ± 7.1	64.9 ± 5.8	48.0 ± 6.3
Pr	0.0 ± 0.0	71.4 ± 0.5	18.4 ± 0.7	33.3 ± 12.7	87.9 ± 14.9	95.2 ± 18.3
Nd	72.3 ± 0.2	65.7 ± 1.2	60.6 ± 4.2	58.8 ± 7.8	75.6 ± 8.5	64.0 ± 9.9
Sm	77.6 ± 2.1	55.4 ± 1.0	47.8 ± 5.1	44.1 ± 6.0	78.3 ± 8.2	62.9 ± 9.2
Eu	31.8 ± 1.2	37.0 ± 1.5	32.3 ± 3.1	34.6 ± 5.1	58.3 ± 4.2	50.3 ± 7.9
Gd	80.6 ± 1.9	59.6 ± 2.9	84.7 ± 12.0	40.7 ± 3.1	56.2 ± 4.9	59.3 ± 8.9
Tb	0.0 ± 0.0	51.6 ± 9.7	7.4 ± 0.1	6.0 ± 7.3	14.1 ± 2.4	33.6 ± 14.7
Dy	51.6 ± 2.4	50.4 ± 2.2	55.6 ± 4.6	49.0 ± 6.3	66.2 ± 8.0	50.2 ± 7.5
Ho	0.0 ± 0.0	30.1 ± 0.4	13.2 ± 1.0	37.3 ± 6.9	51.0 ± 2.2	61.3 ± 0.8
Er	38.2 ± 2.9	29.2 ± 3.0	28.8 ± 2.3	38.9 ± 4.5	51.9 ± 7.6	52.9 ± 5.8

Yb	67.6 ± 0.1	46.1 ± 2.9	52.1 ± 4.5	39.6 ± 4.8	65.6 ± 7.4	51.9 ± 7.8
Lu	50.1 ± 4.9	-	14.6 ± 1.7	17.0 ± 3.1	59.8 ± 6.1	58.5 ± 7.7
ΣCritical REE	60.8 ± 0.1	52.5 ± 1.7	53.9 ± 4.3	52.1 ± 6.7	67.6 ± 7.5	64.8 ± 9.6
ΣREE	48.5 ± 0.9	51.2 ± 0.8	50.4 ± 4.2	44.6 ± 6.9	66.3 ± 6.8	57.7 ± 8.2
U	0.0 ± 0.0	0.0 ± 0.0	-	-	0.0 ± 0.0	-
Th	149.0 ± 15.0	58.4 ± 6.0	-	-	93.8 ± 6.7	-

Note: a “-” indicated that the original mass concentration of the element was unknown for the CFA.

Table S5. Leaching efficiency, *L* (%), of major oxides and trace elements.

Elements	93927	92801	93932	93951	93964	94012
Mg	104.6 ± 6.1	83.3 ± 6.5	117.9 ± 20.9	72.1 ± 15.1	123.0 ± 5.2	40.2 ± 1.7
Al	69.5 ± 0.6	38.3 ± 0.1	52.2 ± 6.2	31.0 ± 4.8	54.1 ± 7.6	19.6 ± 2.1
Si	6.4 ± 0.8	37.3 ± 0.4	27.2 ± 0.9	28.2 ± 3.2	24.2 ± 2.5	18.6 ± 1.0
Ca	87.1 ± 3.6	95.6 ± 8.4	114.7 ± 14.5	85.1 ± 11.4	118.9 ± 7.2	44.3 ± 2.3
Ti	8.8 ± 0.1	4.1 ± 0.5	3.0 ± 0.2	4.5 ± 2.5	3.5 ± 0.6	3.4 ± 0.2
Fe	74.3 ± 2.1	33.9 ± 1.9	55.5 ± 7.3	26.3 ± 6.1	30.9 ± 1.9	23.6 ± 1.9
V	71.4 ± 1.8	71.2 ± 2.8	43.3 ± 4.0	23.3 ± 8.8	15.4 ± 3.8	13.8 ± 4.3
Cr	137.9 ± 1.9	23.2 ± 2.3	26.1 ± 1.1	10.4 ± 1.3	8.7 ± 1.4	6.6 ± 1.0
Mn	144.4 ± 4.4	85.4 ± 15.8	95.9 ± 15.2	48.7 ± 9.3	65.9 ± 3.0	32.2 ± 0.4
Ni	37.3 ± 5.3	43.4 ± 5.2	157.5 ± 98.4	55.3 ± 10.9	85.1 ± 27.4	33.6 ± 2.2
Cu	0.0 ± 0.0	170.2 ± 54.7	147.4 ± 16.8	99.9 ± 22.0	159.5 ± 5.5	79.0 ± 9.5
Zn	0.0 ± 0.0	80.9 ± 1.4	117.0 ± 2.8	64.3 ± 12.3	56.1 ± 2.0	44.9 ± 1.0
As	0.0 ± 0.0	78.6 ± 2.6	65.0 ± 44.2	46.1 ± 47.6	65.4 ± 5.5	56.7 ± 12.3
Se	-	10.1 ± 7.4	-	-	-	-
Cd	0.0 ± 0.0	0.0 ± 0.0	30.7 ± 43.4	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Pb	40.7 ± 2.1	74.3 ± 15.3	35.5 ± 19.7	12.8 ± 1.6	16.4 ± 16.4	0.0 ± 0.0
Σbulk	62.8 ± 2.4	38.8 ± 0.5	43.4 ± 3.3	30.0 ± 4.4	36.3 ± 3.6	20.3 ± 0.5
Σtrace	48.3 ± 1.5	71.8 ± 8.6	81.3 ± 11.2	41.4 ± 12.8	52.4 ± 6.5	32.1 ± 4.1

Note: a “-” indicated that the original mass concentration of the element was unknown for the CFA.

Table S6. Mass distribution of major oxides and trace elements in the alkaline solution after pre-treatment (PT%).

Elements	92801	93932	93951	93964	94012
Mg	0.7 ± 0.3	0.6 ± 0.1	0.5 ± 0.1	0.4 ± 0.1	0.2 ± 0.1
Al	0.8 ± 0.0	1.2 ± 0.1	0.9 ± 0.3	1.0 ± 0.0	0.6 ± 0.1
Si	30.9 ± 0.1	24.8 ± 0.4	25.2 ± 4.4	22.5 ± 2.5	16.3 ± 1.0
Ca	10.3 ± 3.0	5.2 ± 1.2	13.8 ± 1.0	5.7 ± 1.5	5.0 ± 1.5
Ti	0.3 ± 0.1	0.4 ± 0.0	0.7 ± 0.5	0.4 ± 0.0	0.2 ± 0.0
Fe	1.0 ± 0.0	0.8 ± 0.3	0.9 ± 0.3	0.3 ± 0.1	0.6 ± 0.0
V	67.9 ± 2.7	43.1 ± 4.0	23.3 ± 8.8	15.4 ± 3.8	13.8 ± 4.3
Cr	9.8 ± 3.0	3.8 ± 0.2	0.8 ± 1.1	0.0 ± 0.0	0.0 ± 0.0
Mn	14.8 ± 11.1	1.0 ± 1.3	2.4 ± 2.5	0.0 ± 0.0	0.0 ± 0.0
Ni	1.7 ± 2.2	0.0 ± 0.0	0.2 ± 0.2	0.3 ± 0.5	0.9 ± 1.3
Cu	111.3 ± 49.3	44.4 ± 43.6	56.2 ± 9.7	13.7 ± 10.5	49.5 ± 9.3
Zn	41.7 ± 1.6	53.2 ± 2.3	30.4 ± 5.8	19.5 ± 1.8	24.5 ± 1.3
As	78.6 ± 2.6	65.0 ± 44.2	46.1 ± 47.6	65.4 ± 5.5	56.7 ± 12.3
Se	10.1 ± 7.4	-	-	-	-
Cd	0.0 ± 0.0	30.7 ± 43.4	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Pb	26.0 ± 13.3	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0

Note: a “-” indicated that the original mass concentration of the element was unknown for the CFA.

Table S7. Distribution coefficient, D , of major oxides and trace elements.

Element	93927	92801	93932	93951	93964	94012
Mg	0.3 ± 0.0	0.2 ± 0.0	0.2 ± 0.0	0.2 ± 0.0	0.2 ± 0.1	0.2 ± 0.0
Al	2.9 ± 0.1	1.0 ± 0.2	1.3 ± 0.0	1.2 ± 0.2	1.1 ± 0.4	1.6 ± 0.2
Si	0.0 ± 0.0	0.2 ± 0.1	0.2 ± 0.0	0.1 ± 0.1	0.2 ± 0.0	0.1 ± 0.0
Ca	0.4 ± 0.0	0.3 ± 0.1	0.3 ± 0.0	0.3 ± 0.0	0.3 ± 0.1	0.3 ± 0.1
Ti	6.1 ± 0.5	2.8 ± 0.1	8.7 ± 3.0	4.8 ± 0.3	10.5 ± 2.3	3.8 ± 0.4
Fe	0.5 ± 0.0	0.3 ± 0.1	0.3 ± 0.0	0.3 ± 0.0	0.2 ± 0.1	0.3 ± 0.1
V	7.6 ± 0.4	2.1 ± 0.0	×	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Cr	12.8 ± 2.8	8.3 ± 4.1	17.8 ± 2.9	26.5 ± 3.3	13.1 ± 0.8	×
Mn	0.4 ± 0.0	0.2 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.2 ± 0.1	0.3 ± 0.0
Ni	0.2 ± 0.2	0.2 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.5 ± 0.5	0.3 ± 0.0
Cu	0.0 ± 0.0	0.3 ± 0.0	0.2 ± 0.0	0.2 ± 0.0	0.2 ± 0.2	0.2 ± 0.0
Zn	0.0 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.2 ± 0.1	0.3 ± 0.0
As	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Se	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	×
Cd	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Pb	0.1 ± 0.1	0.2 ± 0.1	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0

Note: a × indicated that the element was not detected in the AQ phase but had minor amounts in the IL phase.

Table S8. Recovery efficiency, R (%), of major oxides and trace elements.

Element	93927	92801	93932	93951	93964	94012
Mg	23.7 ± 1.8	13.2 ± 3.7	20.8 ± 3.8	13.4 ± 1.6	19.0 ± 7.4	6.5 ± 0.6
Al	51.6 ± 0.8	18.8 ± 1.9	29.2 ± 3.2	16.6 ± 1.4	27.7 ± 8.4	11.6 ± 1.9
Si	0.0 ± 0.0	0.8 ± 0.3	0.4 ± 0.1	0.3 ± 0.0	0.2 ± 0.0	0.3 ± 0.0
Ca	24.1 ± 1.1	19.7 ± 4.0	24.2 ± 3.3	17.8 ± 1.5	23.4 ± 7.4	10.1 ± 1.0
Ti	7.5 ± 0.0	2.8 ± 0.4	2.4 ± 0.3	3.2 ± 1.7	2.9 ± 0.6	2.5 ± 0.0
Fe	23.2 ± 2.1	7.0 ± 1.5	12.2 ± 1.4	6.1 ± 1.4	6.0 ± 2.5	5.3 ± 1.4
V	63.1 ± 2.0	2.3 ± 0.1	0.2 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Cr	127.7 ± 0.3	11.8 ± 1.3	21.1 ± 1.5	9.3 ± 0.2	8.1 ± 1.3	6.6 ± 1.0
Mn	43.7 ± 1.8	14.1 ± 3.0	20.9 ± 3.5	10.5 ± 0.7	12.4 ± 4.5	6.6 ± 0.8
Ni	5.3 ± 7.5	7.7 ± 1.6	40.7 ± 27.7	12.6 ± 1.3	26.9 ± 26.5	7.5 ± 0.4
Cu	0.0 ± 0.0	12.5 ± 2.9	18.7 ± 9.9	8.3 ± 2.1	27.6 ± 18.4	4.2 ± 0.5
Zn	0.0 ± 0.0	9.5 ± 0.7	15.1 ± 0.2	8.6 ± 1.4	6.6 ± 1.9	5.1 ± 0.2
As	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Se	-	0.0 ± 0.0	-	-	-	-
Cd	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Pb	2.0 ± 2.8	8.3 ± 5.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Σ_{bulk}	22.1 ± 0.9	8.2 ± 0.1	11.9 ± 1.3	6.8 ± 0.7	9.1 ± 2.9	4.9 ± 0.8
Σ_{trace}	28.3 ± 1.0	5.9 ± 0.9	13.0 ± 4.4	5.1 ± 0.0	9.4 ± 5.7	3.2 ± 0.1

Note: a “-” indicated that the original mass concentration of the element was unknown for the CFA.