Recovery of High Purity Rare Earth Elements from Coal Ash

2019 Annual Project Review Meeting for Crosscutting, Rare Earth Elements, Gasification, and Transformative Power Generation

NATIONAL ENERGY TECHNOLOGY LABORATORY

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RARE EARTH SALTS

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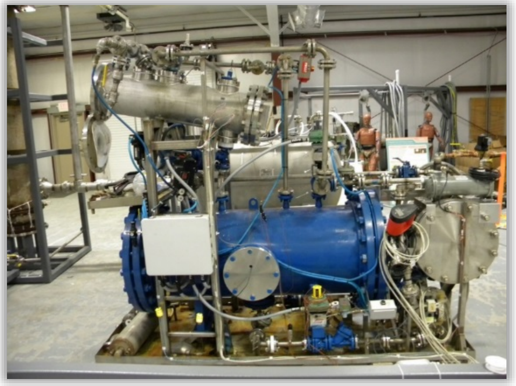
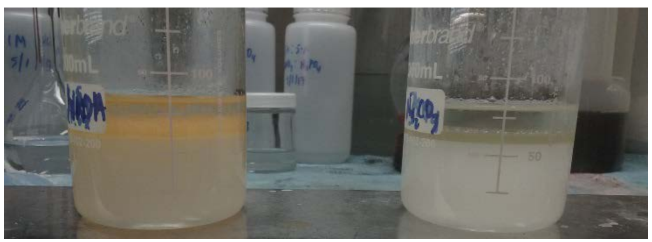
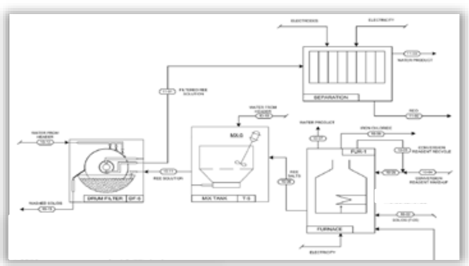
Outline



Process Description

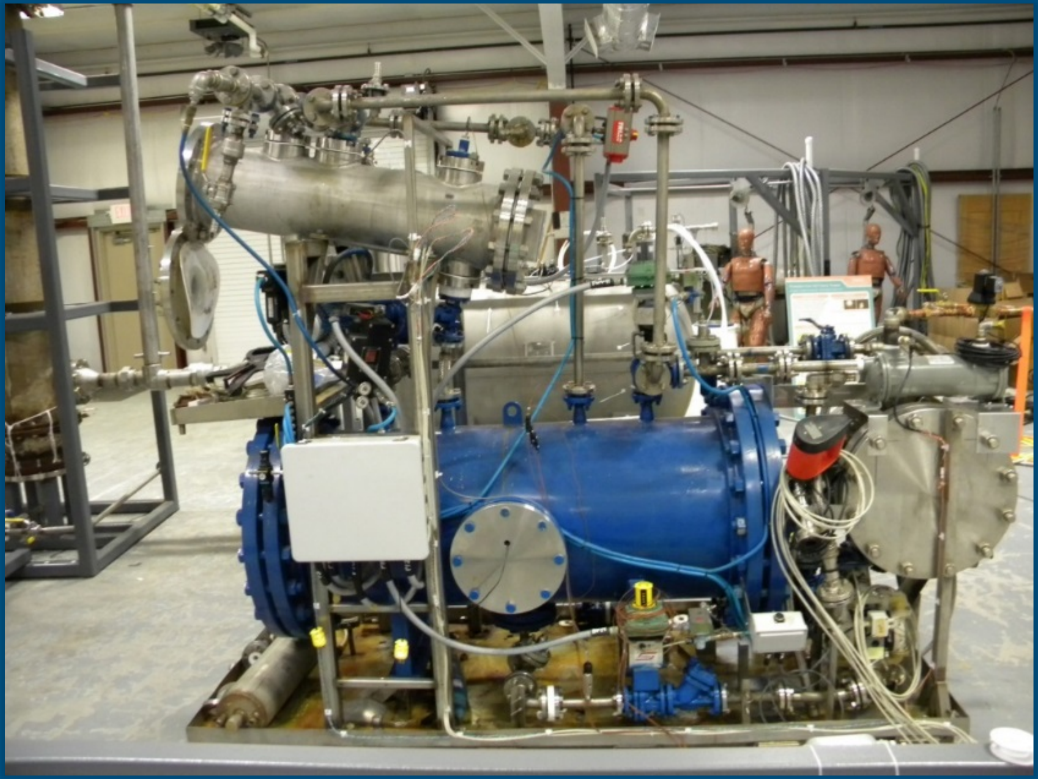


Key Results

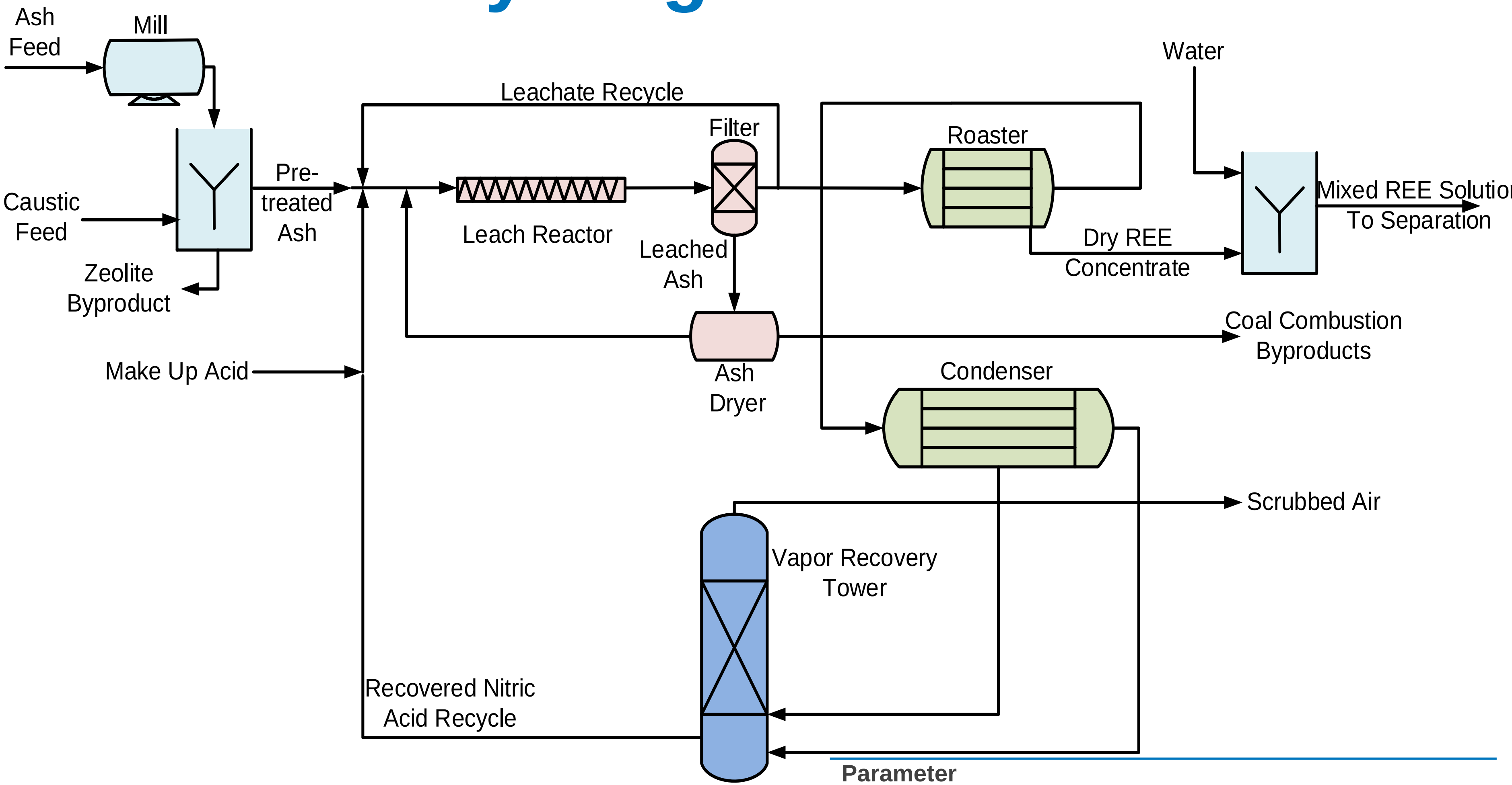


Next Steps

Process Description

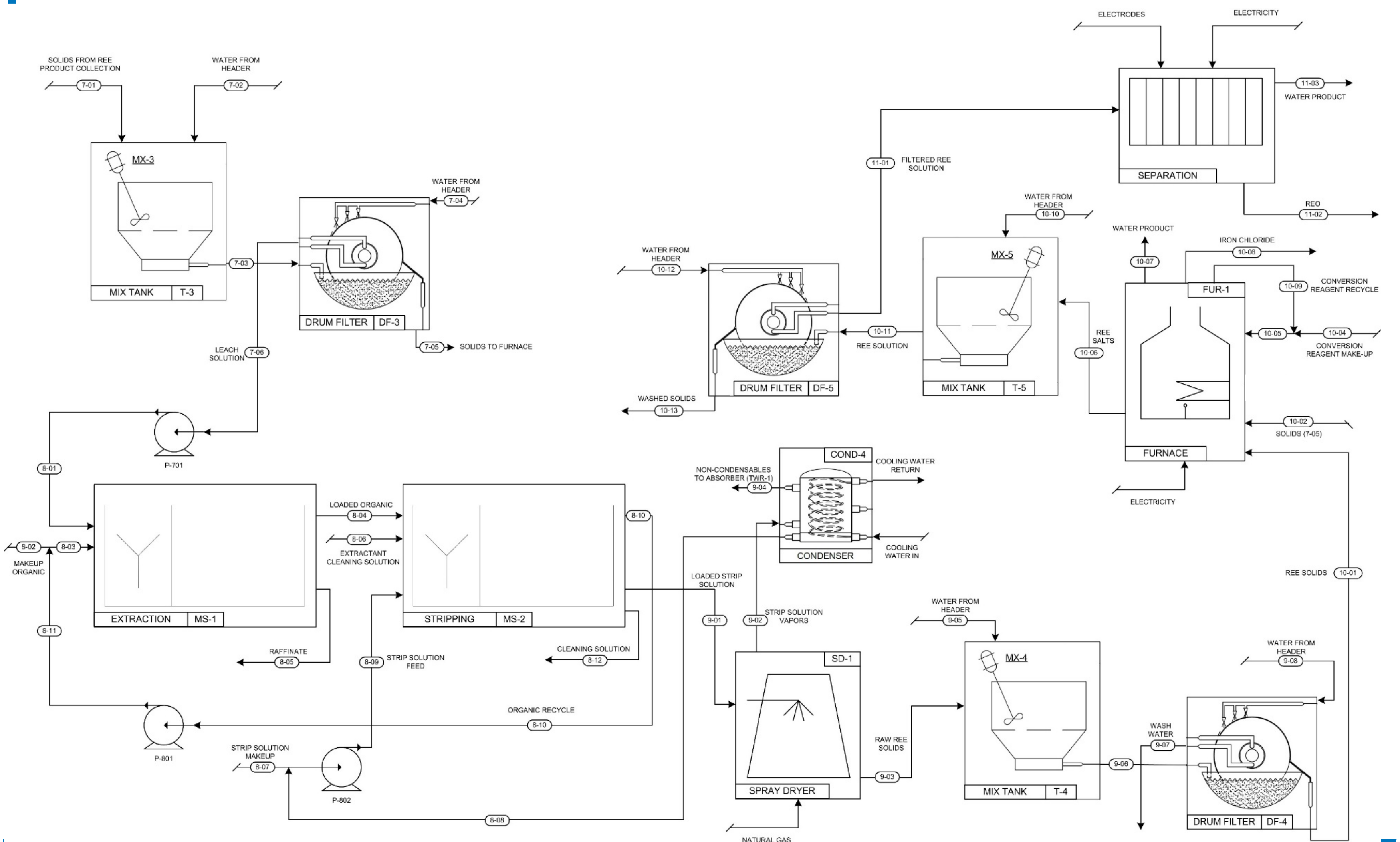


Acid consumption is reduced by thermal recycling



Parameter	
Average REE Yield (% Leached)	85%
Final REE Product Concentration	4.0%
Process Acid Recovery	97.88%

Upgrading and purification will result in greater than 99% purity separated rare earth oxide products



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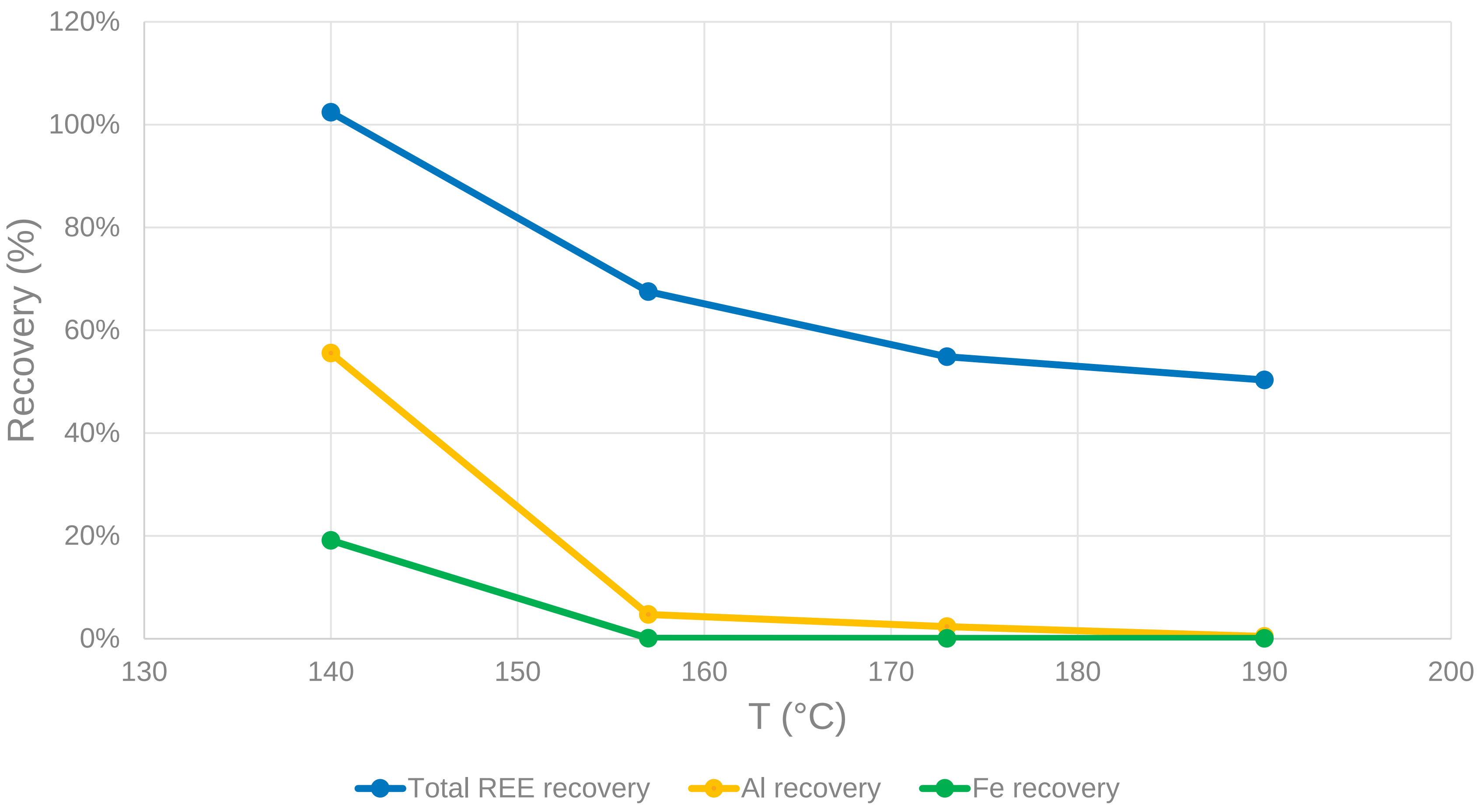
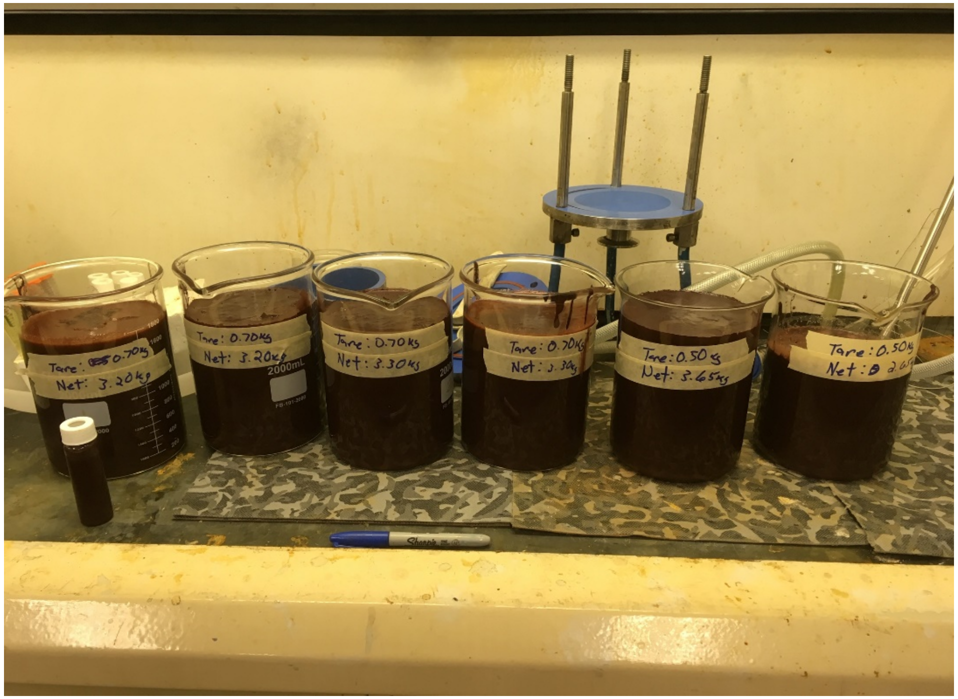
Key Results

Roasting tests suggested that good separation can be realized between REE and base metal contaminants



Concentrating the leach solution leads to a molten salt

- 58kg of PCC fly ash was leached in a batch reactor



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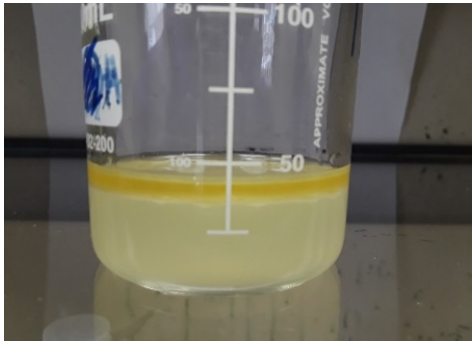
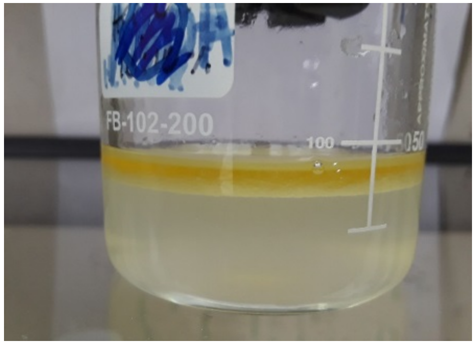
The solvent extraction formulation required a modifier to achieve good phase disengagement

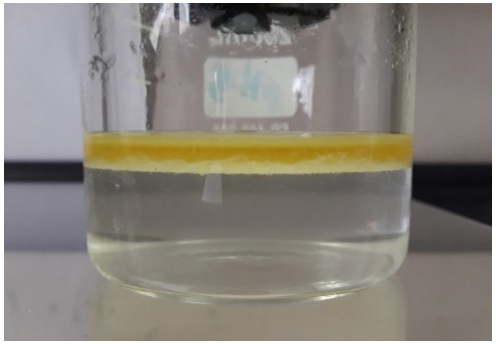


15% Cyanex 572 in aliphatic diluent



13.2% Cyanex 572 and 12% Isoecanol in aliphatic diluent

13.2% Cyanex 572 and 12% tributyl phosphate in aliphatic diluent



A surface response model was generated to understand REE extraction

- The design of experiment considered pH, mixing time, and TBP concentration

Test Number	Starting pH	Equilibrium pH	Mixing time (min)	Addition of TBP (Cyanex 572:TBP)	REE Recovery	REE Purity
1	3.28	2.51	20	13.2%:12%	69.7%	12.1%
2	2.52	2.34	20	14.1%:6%	69.2%	19.5%
3	2.52	2.48	1	14.1%:6%	24.1%	26.6%
4	3.28	2.86	1	15%:0%	37.3%	7.5%
5	4.03	3.06	1	14.1%:6%	75.2%	9.4%
6	4.03	2.73	10	13.2%:12%	78.0%	14.3%
7	3.26	2.65	10	14.1%:6%	79.6%	17.7%
8	3.26	2.65	10	14.1%:6%	79.2%	22.6%
9	2.51	2.43	10	13.2%:12%	65.6%	17.4%
10	4.02	2.74	20	14.1%:6%	84.3%	13.4%
11	3.27	2.64	10	14.1%:6%	77.8%	24.8%
12	2.49	2.37	10	15%:0%	35.7%	12.7%
13	3.26	2.74	1	13.2%:12%	65.7%	13.0%
14	4	2.79	10	15%:0%	71.2%	7.4%
15	3.26	2.56	20	15%:0%	58.9%	15.3%

Parameter	Model Predictions
Maximum REE recovery	~97% at equilibrium pH 3.06, mixing time 10.4 mins, and TBP of 0.55 (Cyanex 572:TBP = 13.9%:7.6%)
Maximum REE purity	~25% at equilibrium pH 2.46, mixing time 1 min, and TBP of 0.54 (Cyanex 572:TBP = 13.9%:7.5%)

The stripping tests left room for improvement

- Of the remaining 40-50% contaminants, over half is zinc and the balance primarily aluminum

Test Number	Starting pH	Equilibrium pH	Mixing time (min)	REE Purity
1	1.80	1.86	1	53.7%
2	1.80	1.87	10	53.7%
3	1.80	1.86	10	55.1%
4	1.31	1.37	10	52.6%
5	1.79	1.84	10	53.0%
6	1.31	1.35	1	53.2%
7	1.29	1.28	20	34.6%
8	1.29	1.21	20	37.4%
9	2.32	2.35	20	21.8%
10	2.31	2.41	10	24.7%
11	1.78	1.70	20	46.2%
12	2.31	2.40	1	20.5%
13	1.28	1.25	20	38.9%

Zinc leads to downstream processing challenges and has to be removed

- Selective precipitation achieved high purity with tradeoffs in recovery:
  - Route 1: Two step process with a 96% pure REE product (~3.5% zinc) with an 80% average REE recovery (68% Y, 90% Nd/Pr, ~86% all other REE)
  - Route 2: Two step process with a 99% pure REE product (<0.8% zinc) with an average REE recovery of ~65%.
- A new extractant formulation showed good selectivity for zinc:

Element	% Extracted
Sc	91.8%
La	3.6%
Ce	4.1%
Rest of REEs	0.98%
Zn	25.1%
Fe	29.4%

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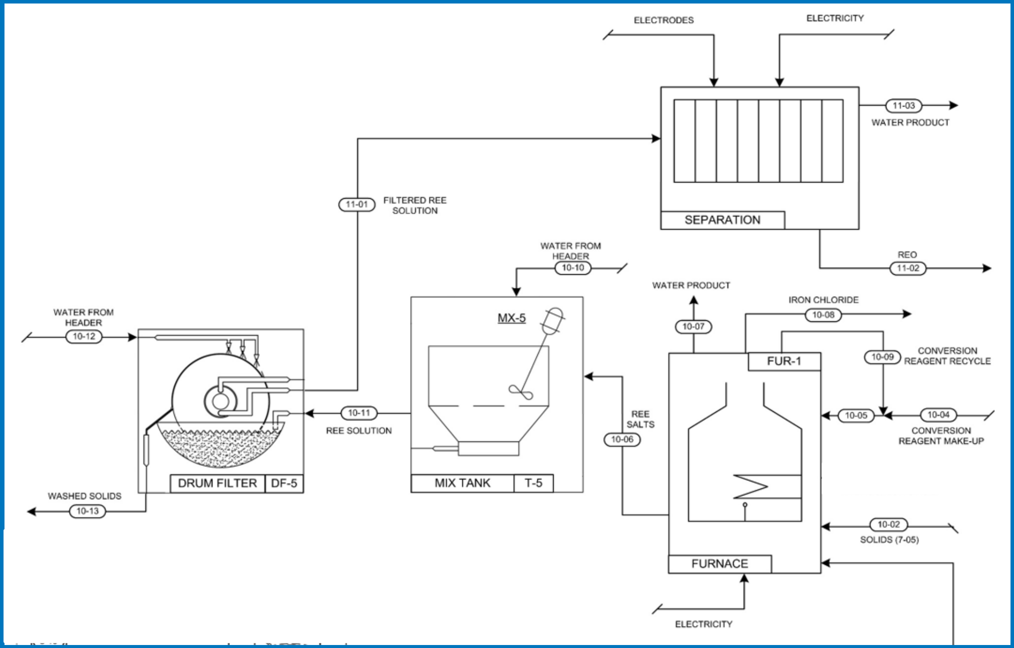
At higher leach solution concentrations, the selectivity inverted

- With a higher PLS concentration, the new extractant still exhibited high selectivity, but extracted REE preferentially over zinc
- Stripping will be done with nitric acid then roasted and washed to remove contaminants and prepare for the conversion process

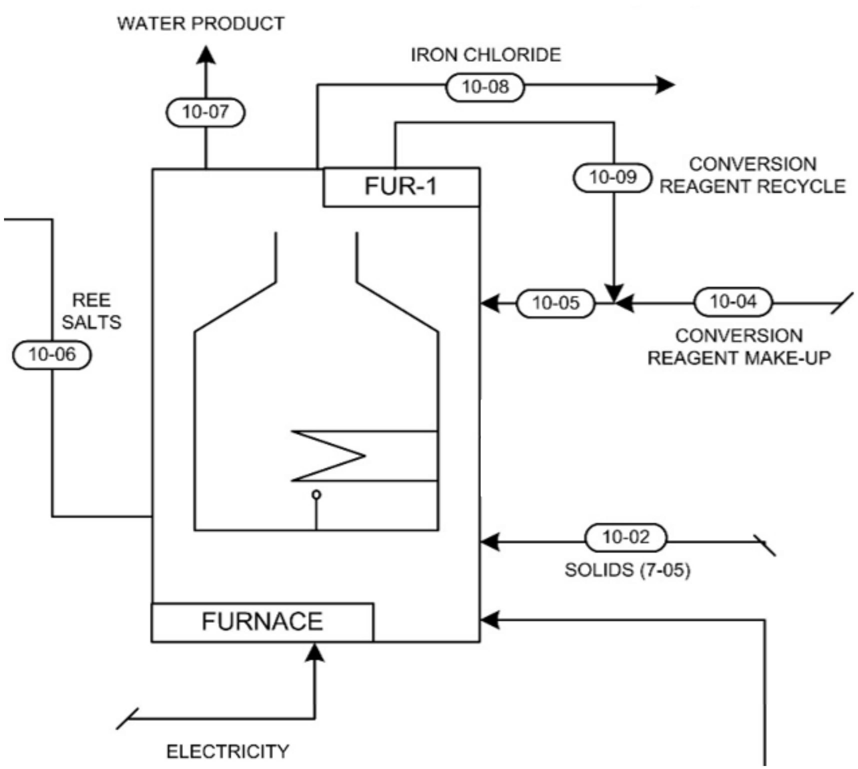
Species	% Extracted (single pass)
Sc	23.08%
Y	39.62%
La	93.02%
Ce	93.11%
Pr	92.04%
Nd	91.00%
Sm	86.52%
Eu	83.17%
Gd	72.52%
Tb	70.99%
Dy	69.39%
Ho	62.37%
Er	55.49%
Tm	40.67%
Yb	32.46%
Lu	21.32%
Total REE+Sc+Y	72.90%
Zn	6.78%
Fe	0%

Next Steps


ELECTRODES
ELECTRICITY
11-03
WATER PRODUCT
SEPARATION
11-01
FILTERED REE SOLUTION
WATER FROM HEADER
10-10
MX-5
REO
11-02
WATER PRODUCT
10-07
IRON CHLORIDE
10-08
WATER FROM HEADER
10-12
FUR-1
10-09
CONVERSION REAGENT RECYCLE
10-11
REE SOLUTION
REE SALTS
10-06
10-05
10-04
CONVERSION REAGENT MAKE-UP
DRUM FILTER
DF-5
MIX TANK
T-5
WASHED SOLIDS
10-13
10-02
SOLIDS (7-05)
FURNACE
ELECTRICITY

Chloride conversion upgrades the mixed REE, allows for recovery of Sc, and can produce a ferric chloride byproduct

- Scandium containing solids from the initial roasting step are recovered in the conversion process



Separated lanthanum and neodymium products will be generated electrochemically

- Separated REEs have been generated from mineral sources and surrogate solutions to determine process parameters for coal sources
- Targeting 99%+ purity separated REE

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