

Application of Biosorption for REE Recovery from Coal Byproducts



Yongqin Jiao, Lawrence Livermore National Laboratory

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Project Description and Objectives



Collaboration among LLNL, Duke U. and U. of Arizona

Biosorption



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Dan Park
Ziye (Jesse) Dong

Leaching



Helen Hsu-Kim
Andrew Middleton

TEA



Hongyue Jin
Majid Doolabi



Project Description and Objectives



Project Highlights from FY20

- New postdoc Ziye (Jesse) Dong started at LLNL Oct 2019
- Hongyue Jin received a *Rising Star Award* from the American Center for Life Cycle Assessment
- Two graduate students, Andrew Middleton and Majid Alipanah, are supported at Duke U. and U. of Arizona, respectively.
- Three publications in peer-reviewed journals:
 - International Journal of Coal Geology, Volume 227, 103532, 2020 (Middleton et al, 2020)
 - Separation and Purification Technology, Volume 241, 15: 116726, 2020 (Park et al, 2020)
 - ACS Substantiable Chemistry & Engineering, *Submitted* (Alipanah et al, 2020)

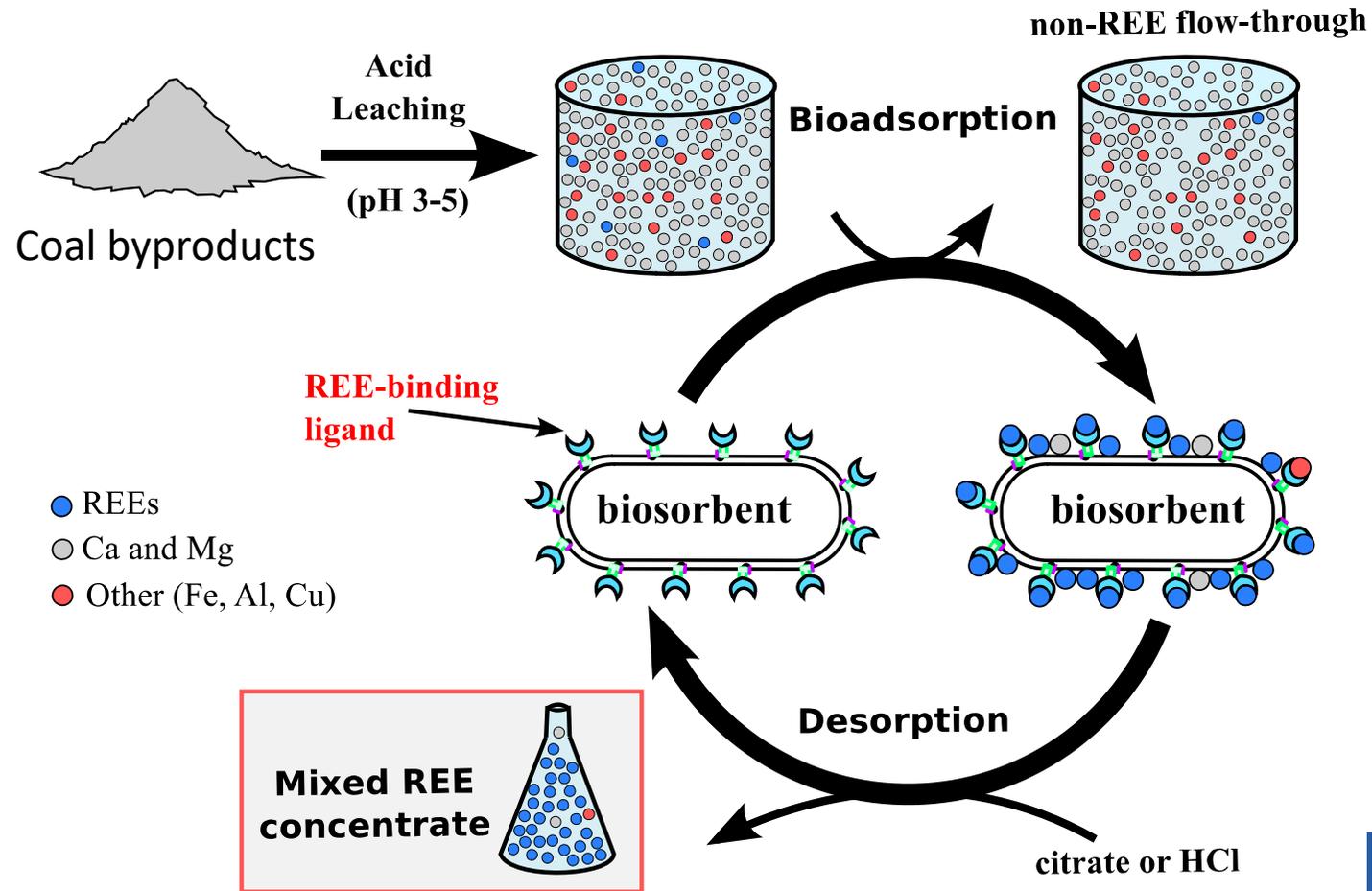


Ziye (Jesse) Dong
LLNL



Project Description and Objectives

A REE-selective biosorption approach to enrich and concentrate the REEs from leachate solutions of coal byproducts

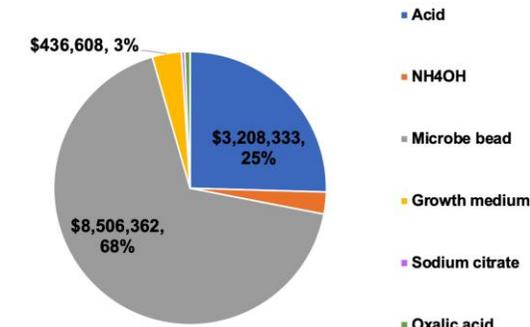
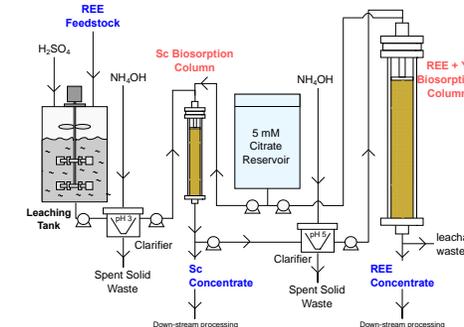
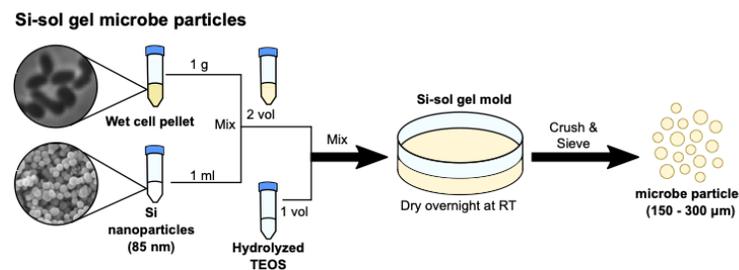
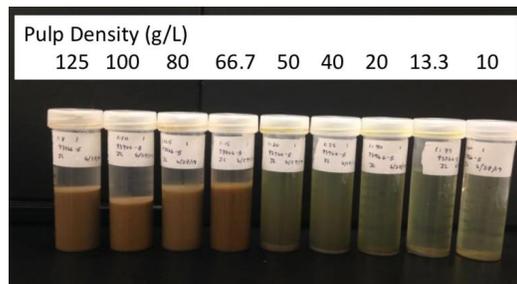


Patented technology

Project Update

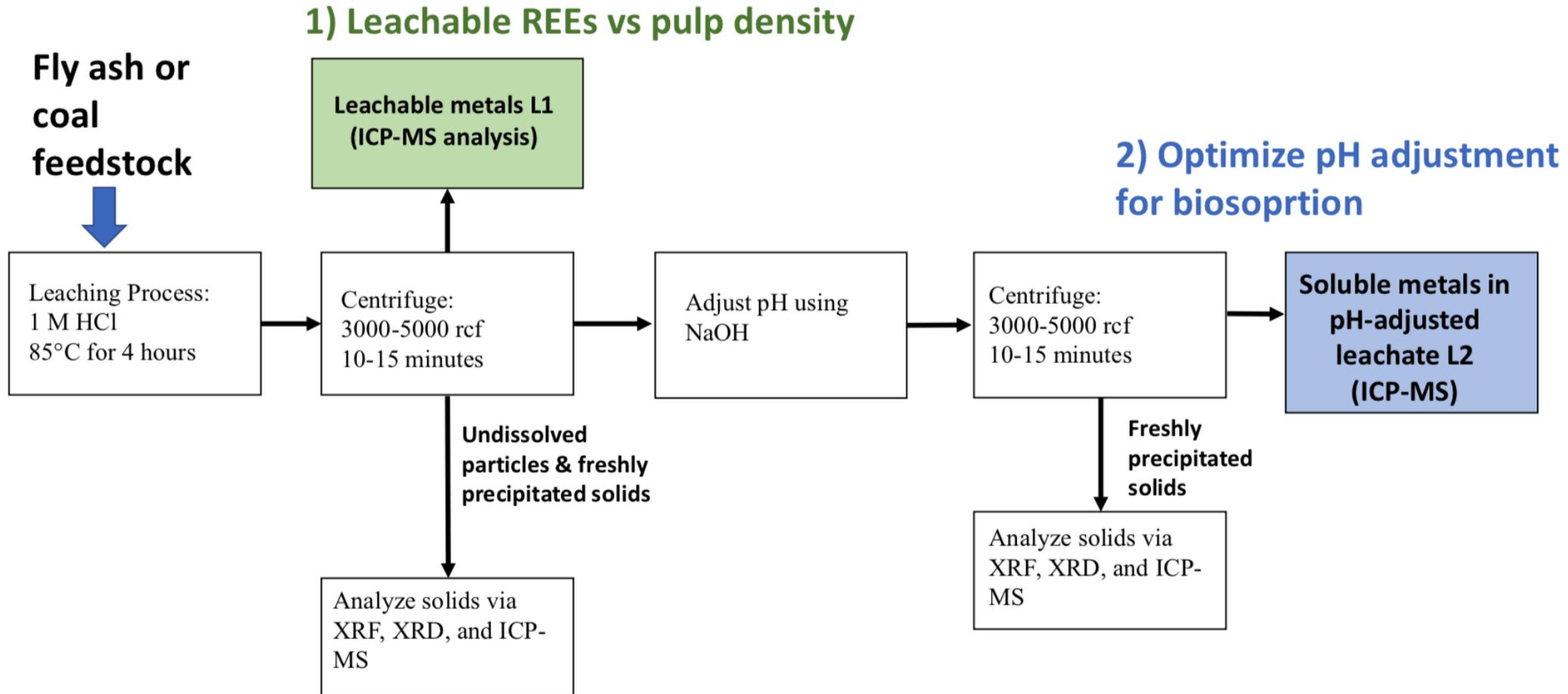
Goal: develop a cost-effective and environmentally sustainable biosorption technology for REE recovery from coal byproducts

- Completed leaching method development and biosorption tests among feedstock (lignite, PRB fly ash, Navajo coal refuse) with decision point made about feedback selection
- Developed Si Sol-gel cell encapsulation that is scalable and less expensive with higher mechanical strength than PEGDA
- Developed a two-stage Sc and REE recovery process
- Completed TEA for Si Sol-gel cell immobilization for rare earth recovery



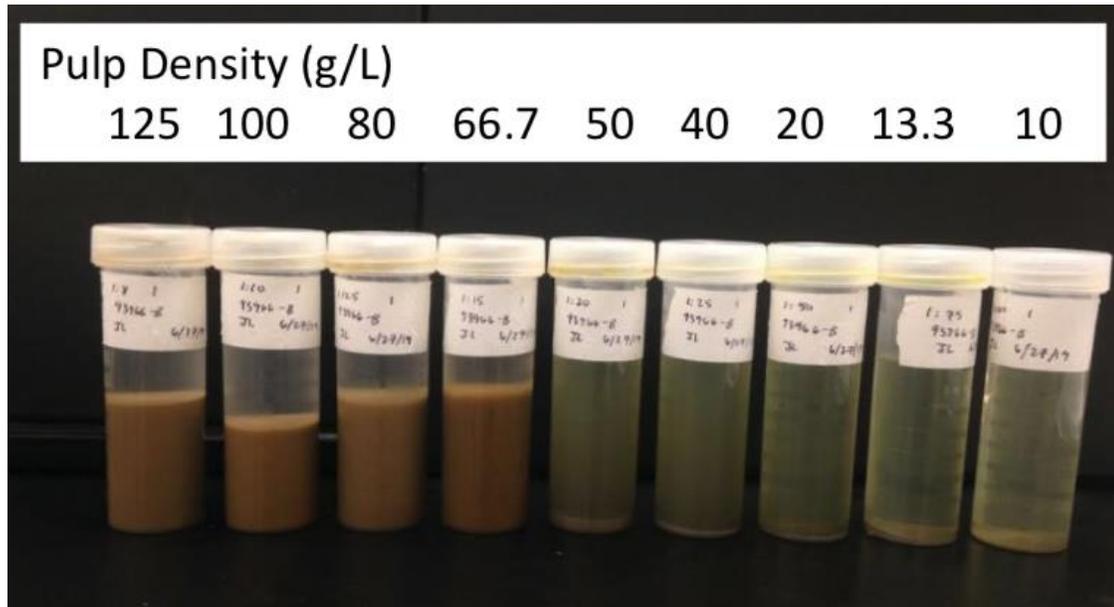
Project Update

Acid leaching of PRB fly ash followed by pH adjustment to prepare for input solution for biosorption

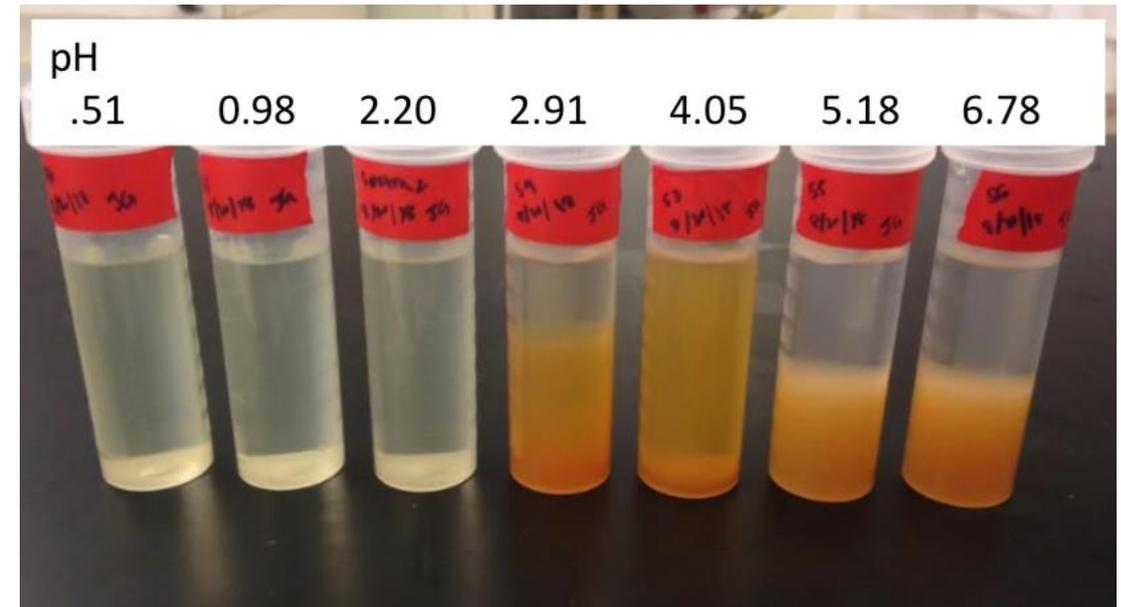


Project Update

Acid leaching of PRB followed by pH adjustment to prepare for input solution for biosorption



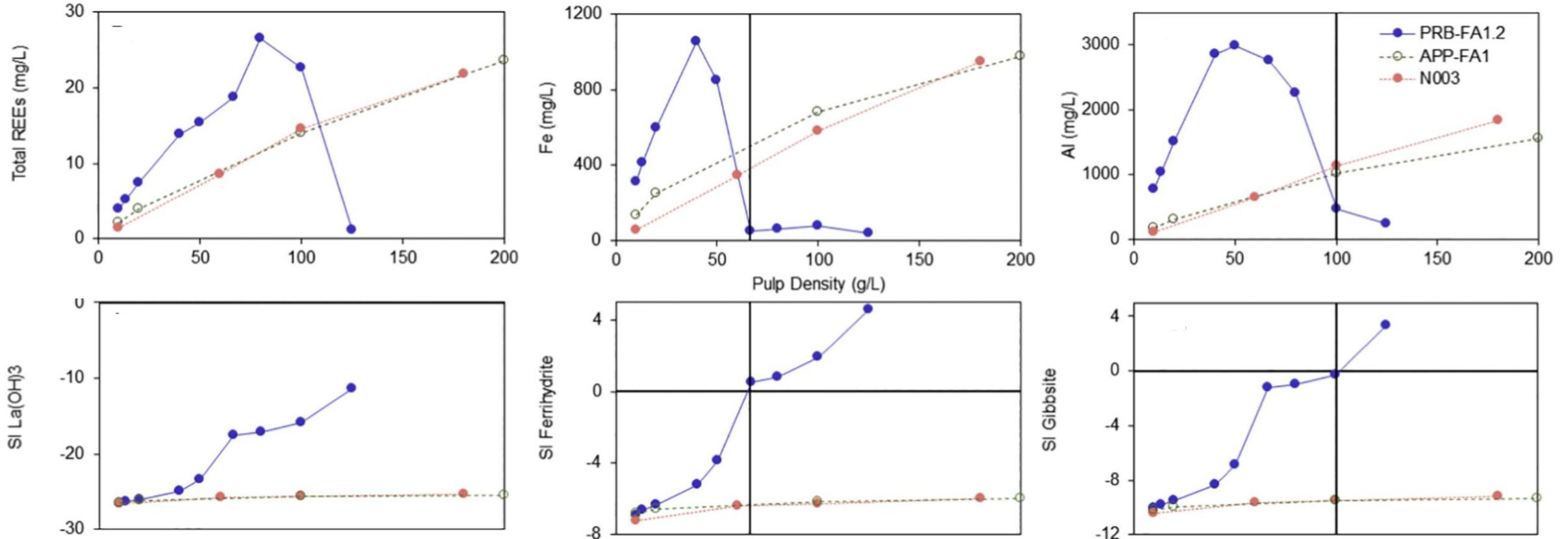
1. Acid leaching of PRB fly ash at various pulp densities



2. pH adjustment after acid leaching

Project Update

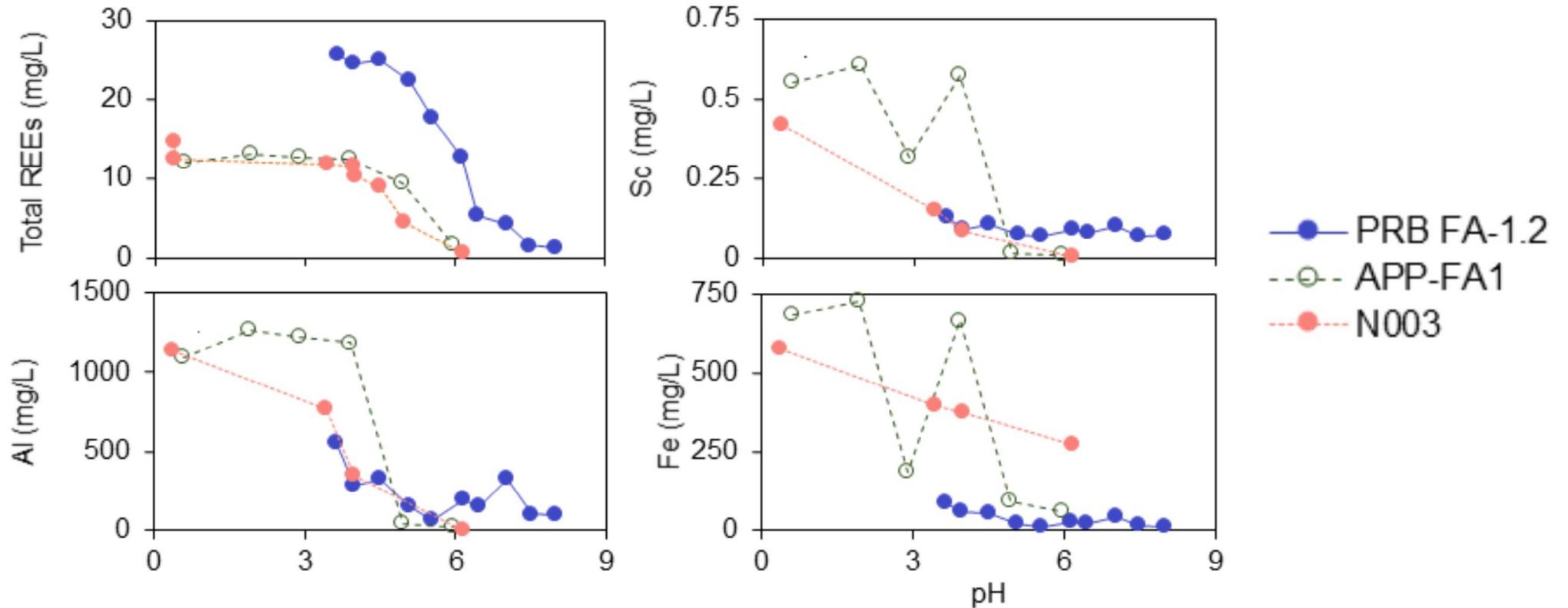
Pulp density is a key factor for selective solubilization of REEs against Al, Fe, and Si in PRB fly ash



Soluble REEs are influenced by Fe- and Al-hydroxide precipitation, not REE-hydroxide minerals.

Project Update

pH adjustment above 5 decreases REE solubility



Aqueous metal concentrations after adjusting the pH of leachates generated from three feedstock after leaching at a pulp density of 100 g/L.

Project Update

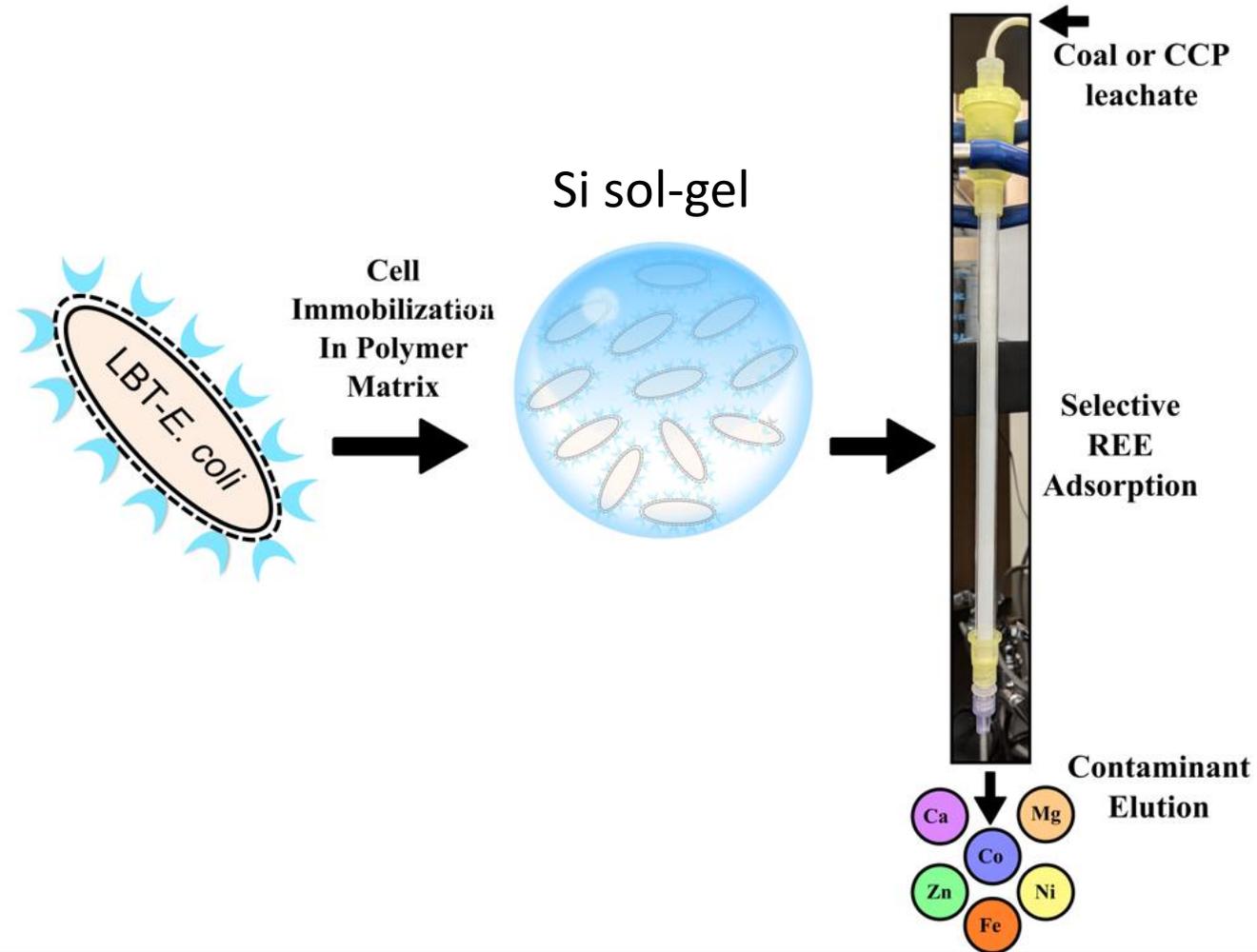
Stoplight chart on feedstock decisions based on compatibility with biosorption

Feedstock type	Location	REE content (ppm)	Leaching efficiency	Leachate purity	Sorption efficiency	Product purity
Lignite	North Dakota, ND	551	high	high	high	high
Fly ash	Power River Basin coal	296-399	high	medium	medium	medium
Fly ash	Appalachian Basin coal	655-703	medium	medium	low	low
Coal refuse	Navajo Indian Reservation AZ	175-210	high	medium	low	low

high
 medium
 low

Project Update

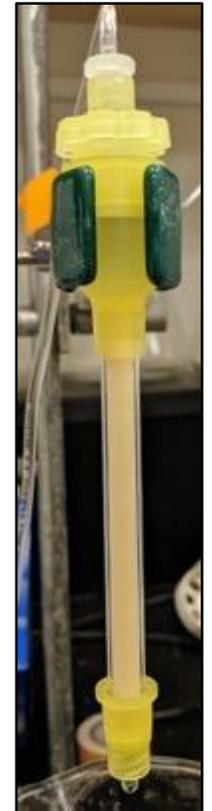
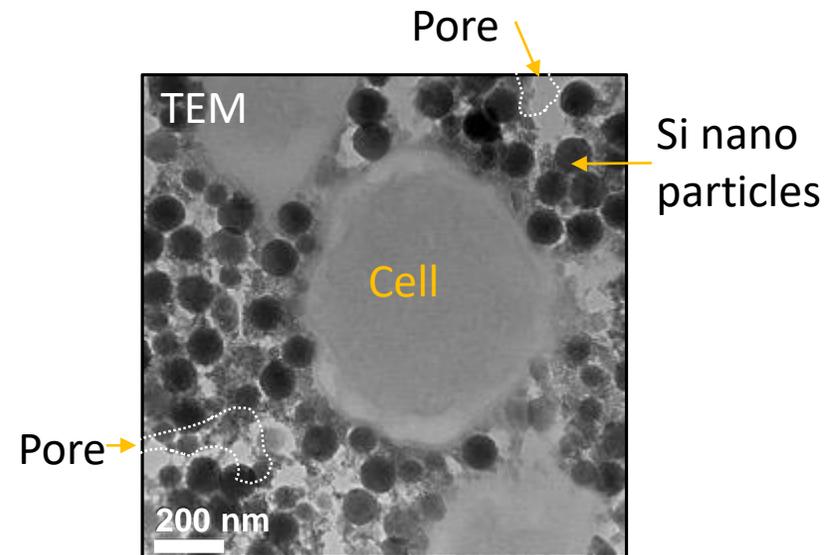
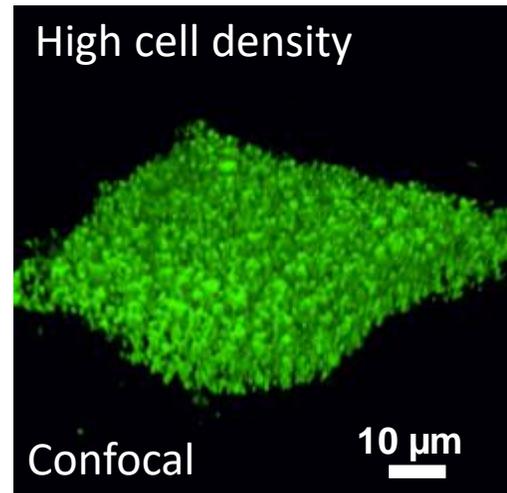
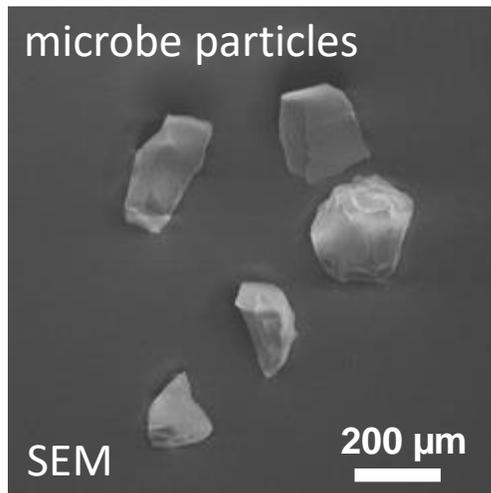
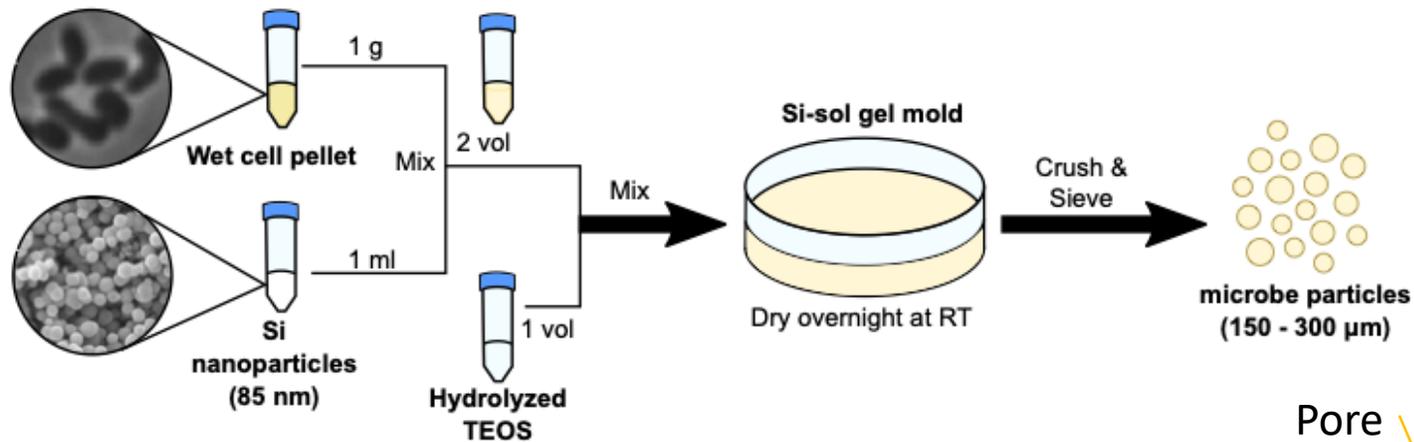
Develop a flow-through biosorption process



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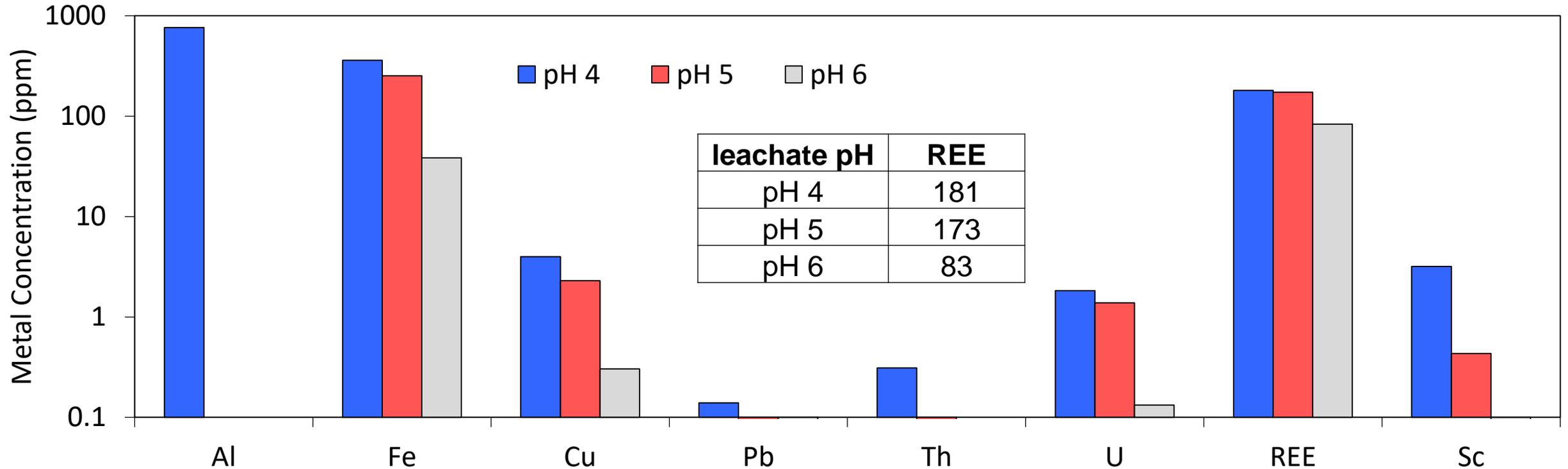
Developed scalable Si Sol-gel sorbent by microbe encapsulation

Si-sol gel microbe particles



Project Update

REE and Sc solubility of lignite leachate upon pH adjustment



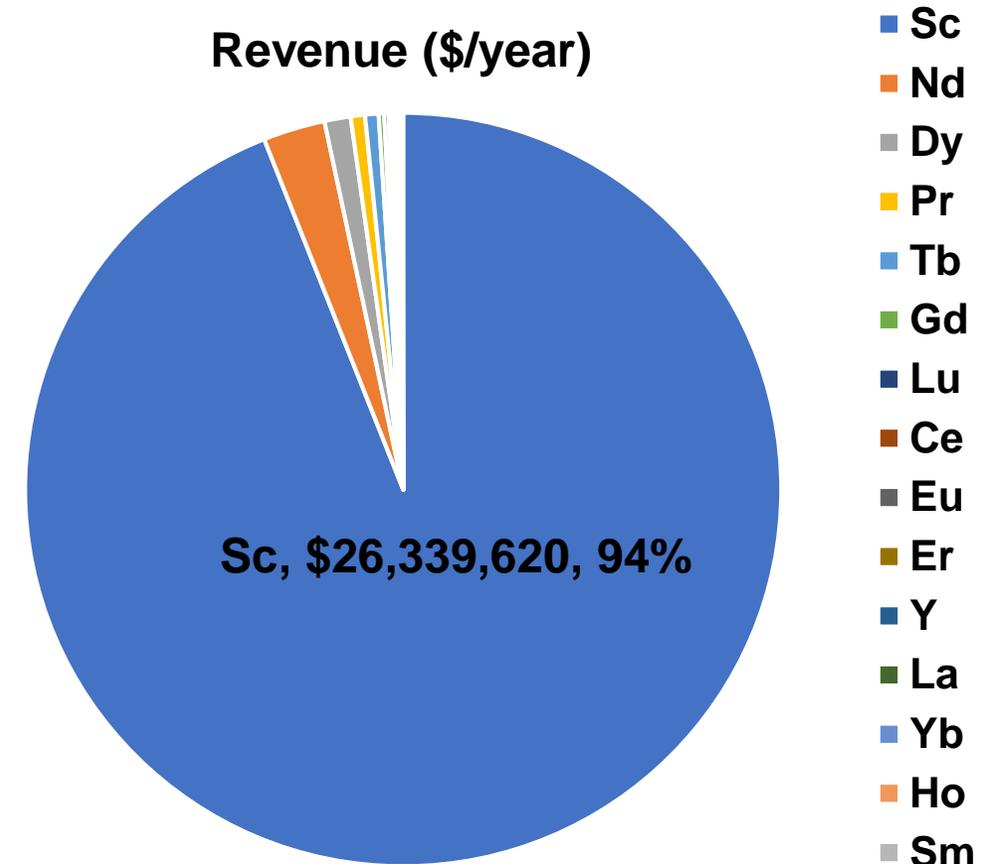
- Lanthanides are soluble up to pH 5
- Sc is soluble only up to pH 4

Project Update

Sc is an important revenue source in coal byproducts

Plant operation assumptions (200 ktonnes/year)

Debt/equity (Capital)	60/40
Term of debt financing	10 years
Interest for debt financing	8% per annum
Plant Life	20 years
Depreciation periods	7, 15, 39 years
Income tax rate	27%
Start up time	6 months
Operating time	8000 hours/year

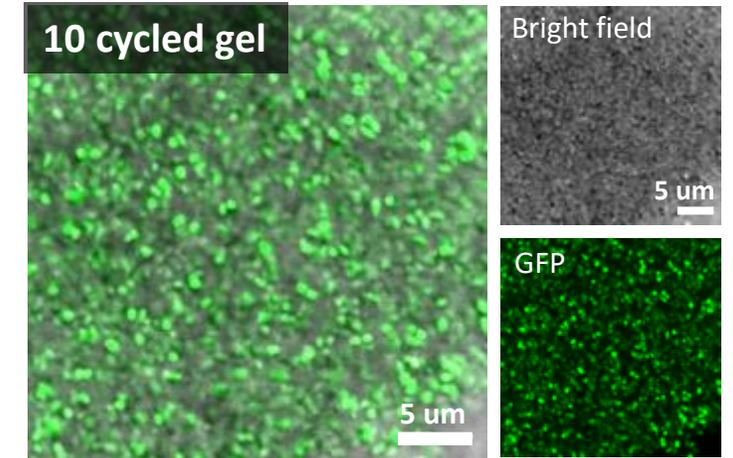
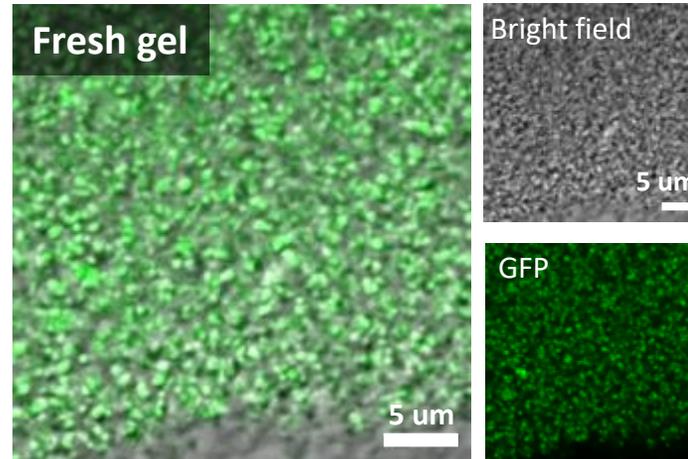
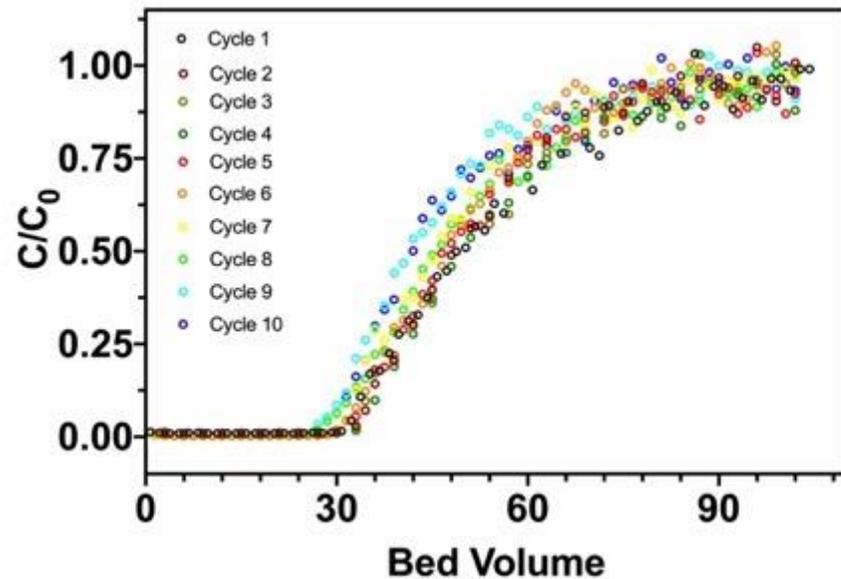


Based on 35.4 ppm Sc in lignite (Laudal, 2017); Sc oxide price: \$4,600/kg (USGS, 2019)

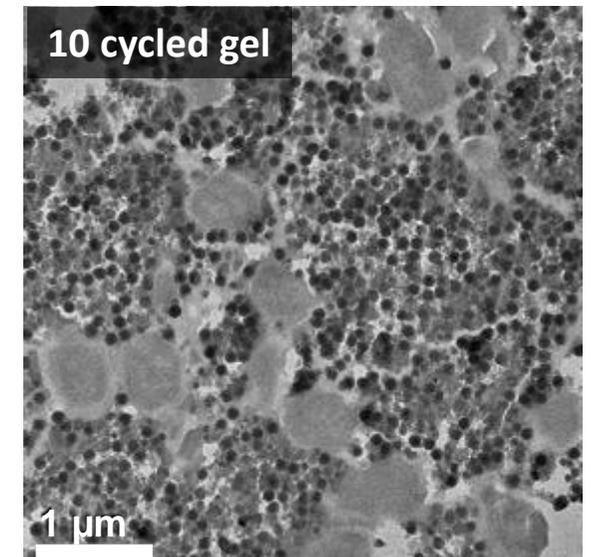
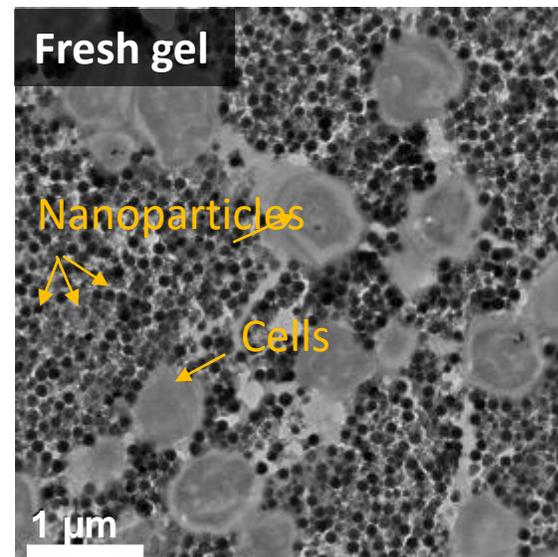
Project Update

Biosorbent is stable for reuse

Sc adsorption at pH 3



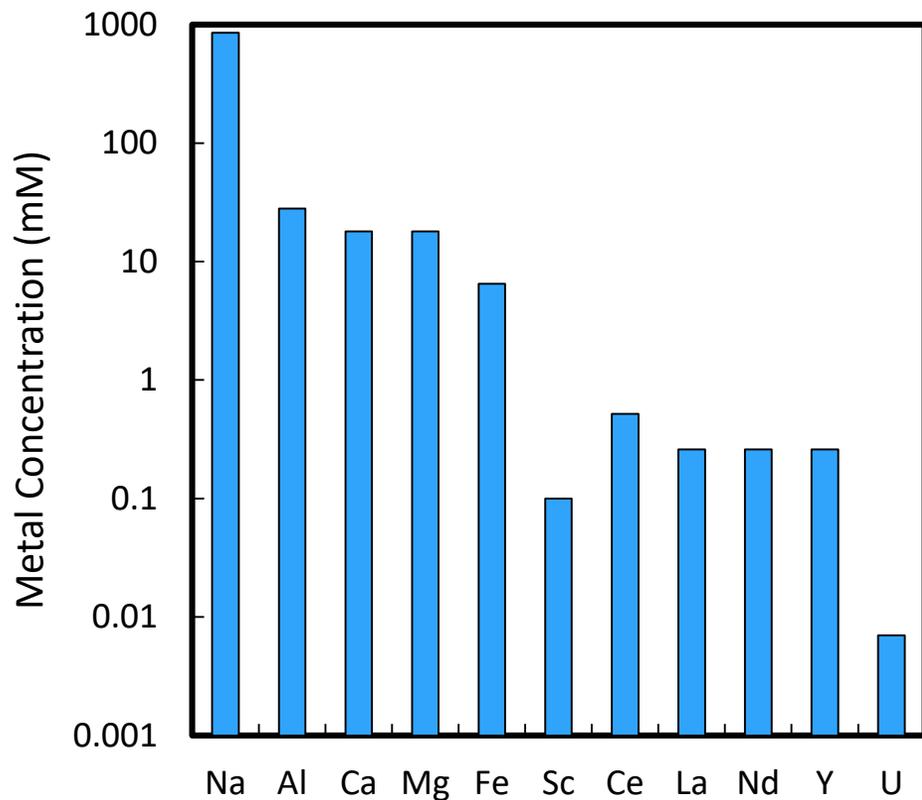
90.0-102 % capacity retained after 10 cycles



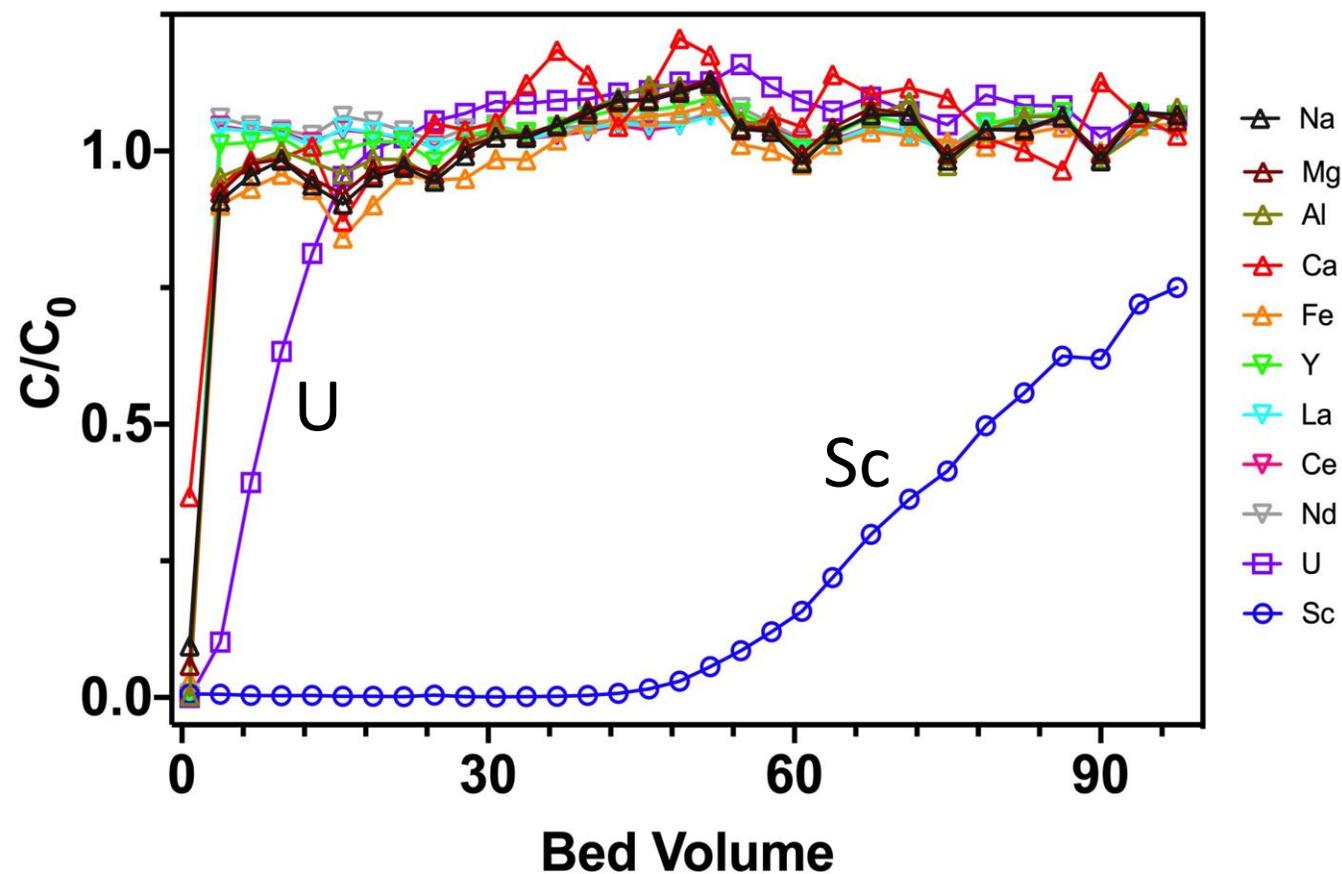
Project Update

Sc is preferentially extracted from a synthetic lignite leachate at pH 3

Synthetic lignite leachate, pH 3



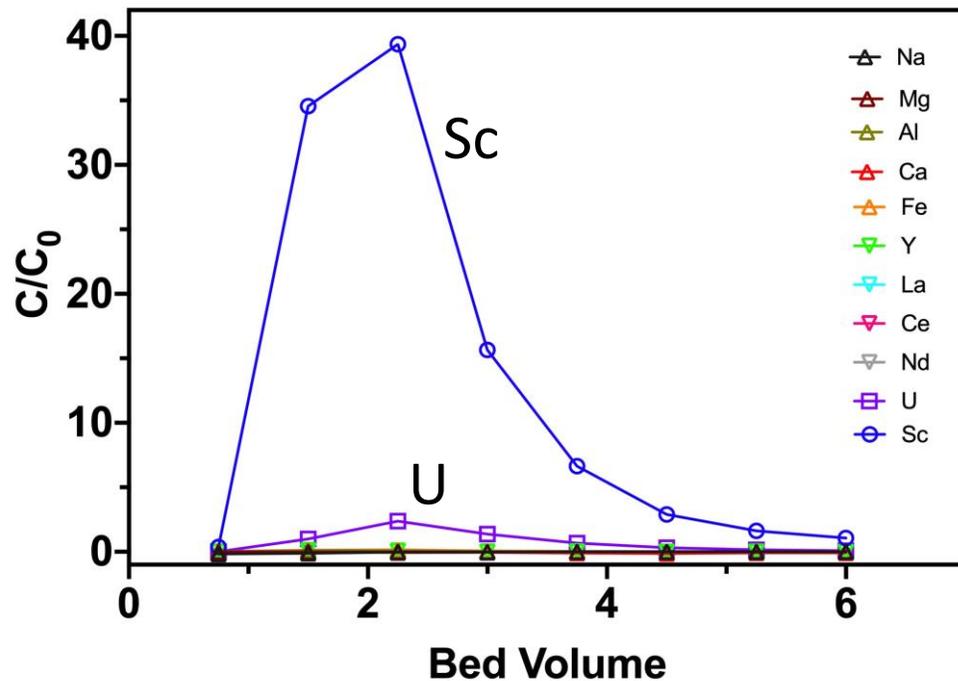
Breakthrough curve



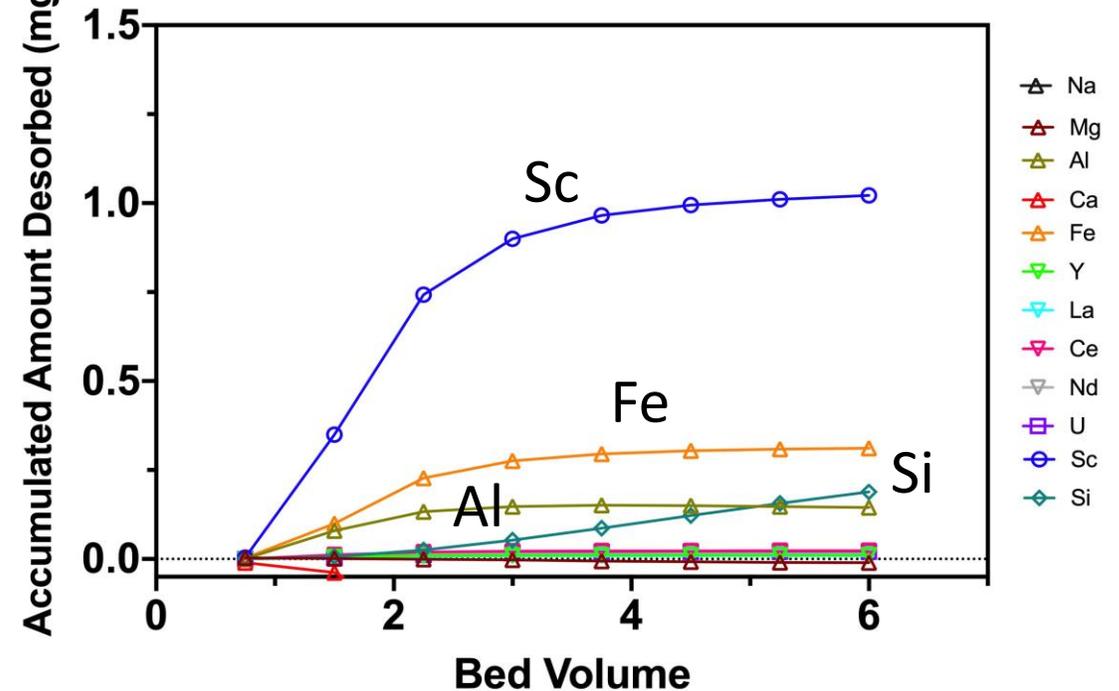
Project Update

Sc is preferentially extracted from a synthetic lignite leachate solution

Desorption using 50 mM citrate pH 6

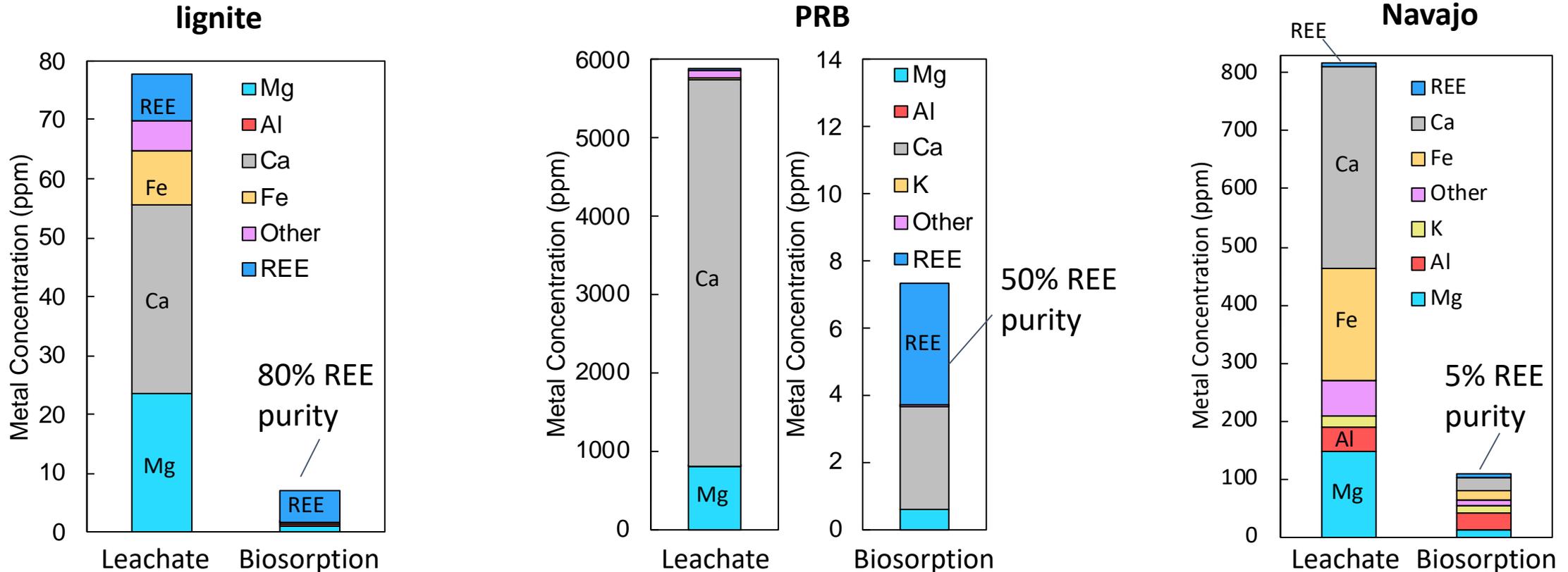


Sc concentrated up to 40-fold



Project Update

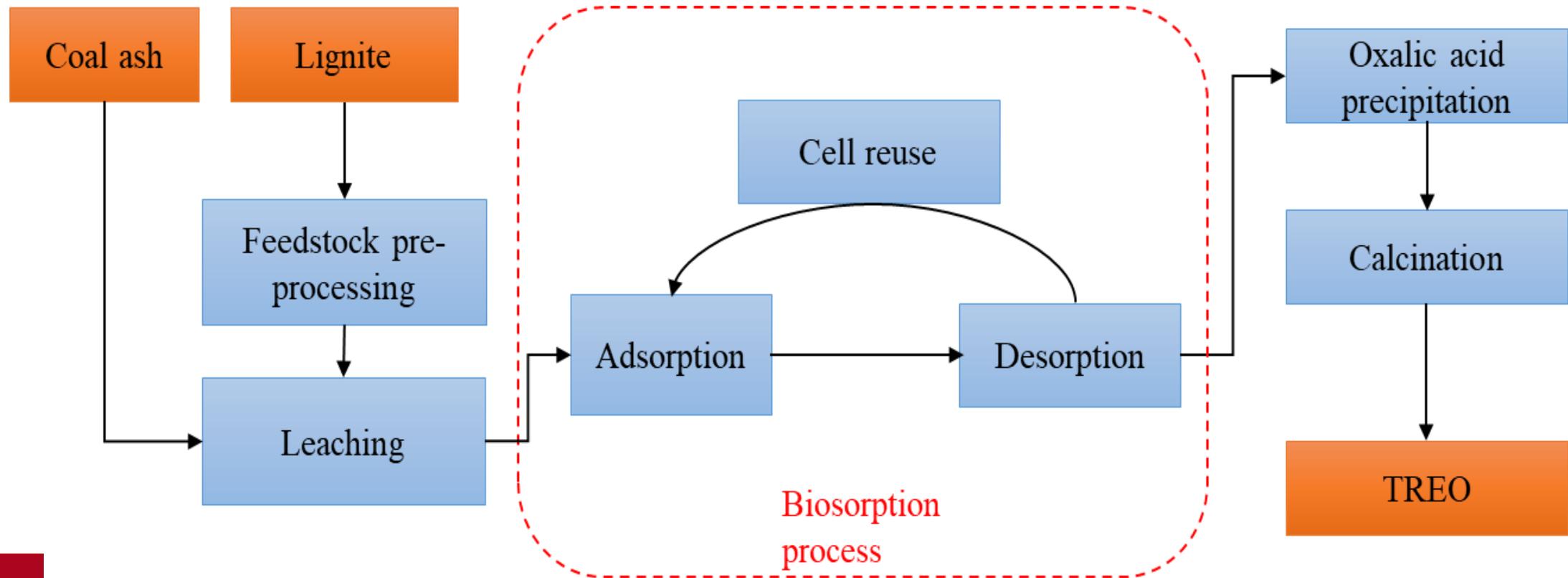
A single round of biosorption produces high purity mixed REEs from lignite and PRB fly ash, but not Navajo coal refuse



High Al precludes high purity REE recovery in Navajo leachate.

Project Update

Overall process flow diagram used in TEA



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Project Update

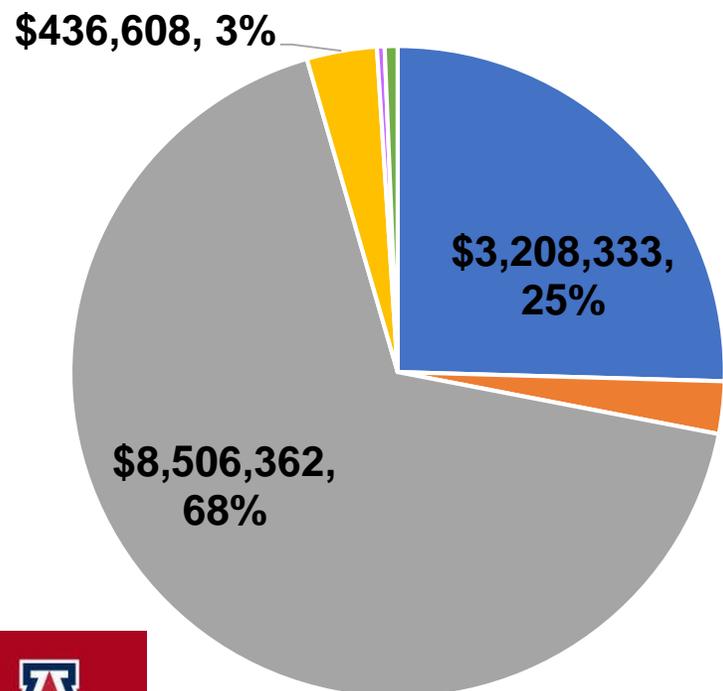
TEA summary and comparison with competing technologies

Data source	Feedstock	REE content (ppm)	REO output quality	REO basket price (\$/kg)	Total cost (\$/kg REO)	Profit rate
Our study	PRB fly ash	337	95% TREO	16	1,518	-99%
Our study	ND lignite	551	95% TREO	338	292	18%
Das et al. (2018)	Coal ash	608-934	Individual REO	577-1,150	680-2,545	-73%~30%
Zhang & Honaker (2018)	Coal coarse refuse KT	7	94% TREO	N/A	34	N/A
Carlson (2018)	Louisville fly ash	480	TREO	33	2,669	-99%
Peterson et al. (2017)	Ohio fly ash	532-558	Individual REO	179	235	-24%

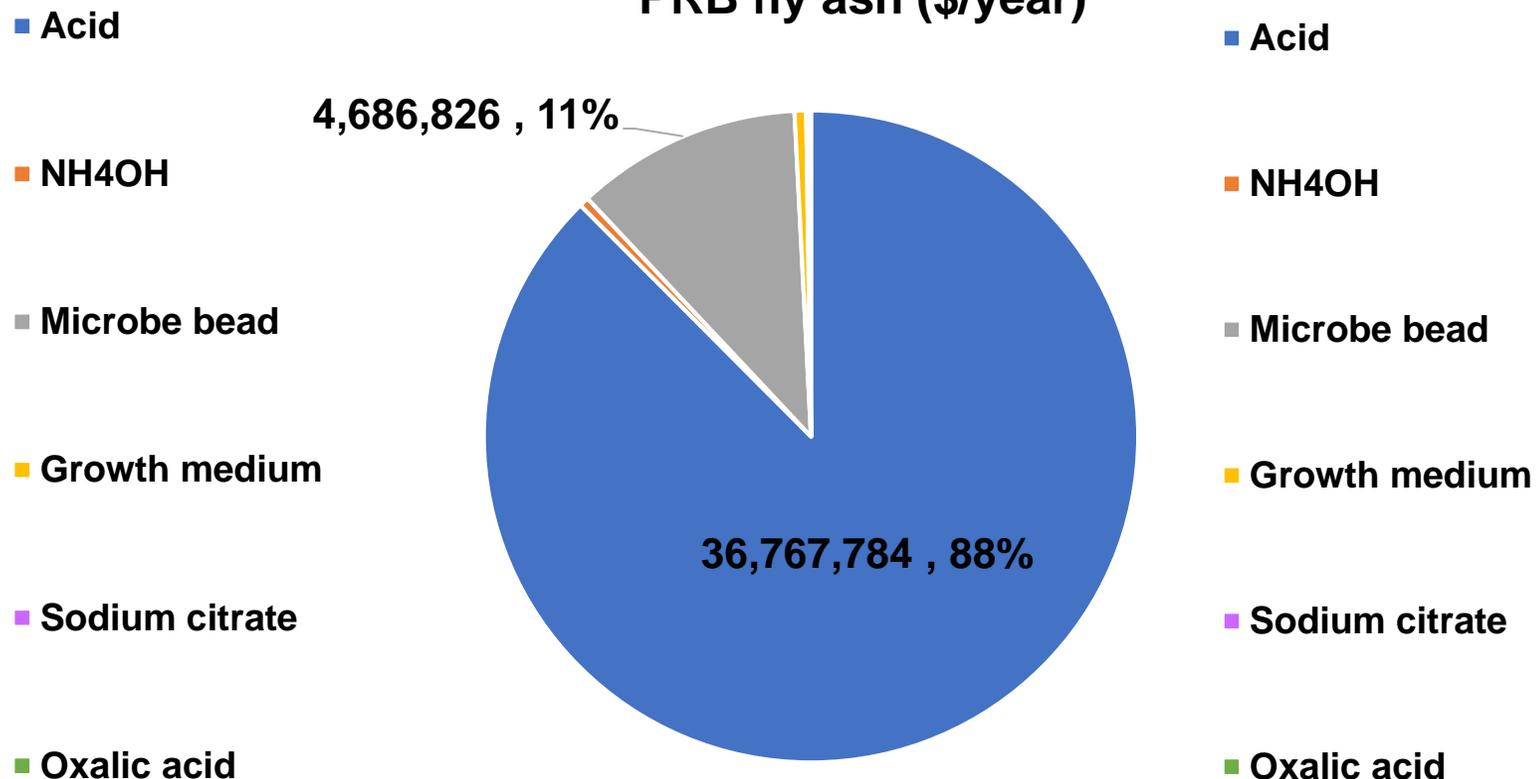
Project Update

Material cost breakdown – acid consumption is a major cost factor for PRB

ND lignite (\$/year)



PRB fly ash (\$/year)



Preparing Project for Next Steps



Transitioning and scale-up into bench-scale production

Market Benefits

- Fill a technology gap by converting coal byproducts to REE concentrate intermediate that can be further refined by existing technologies
- Provide an environmentally friendly alternative for REE recovery and refinement.

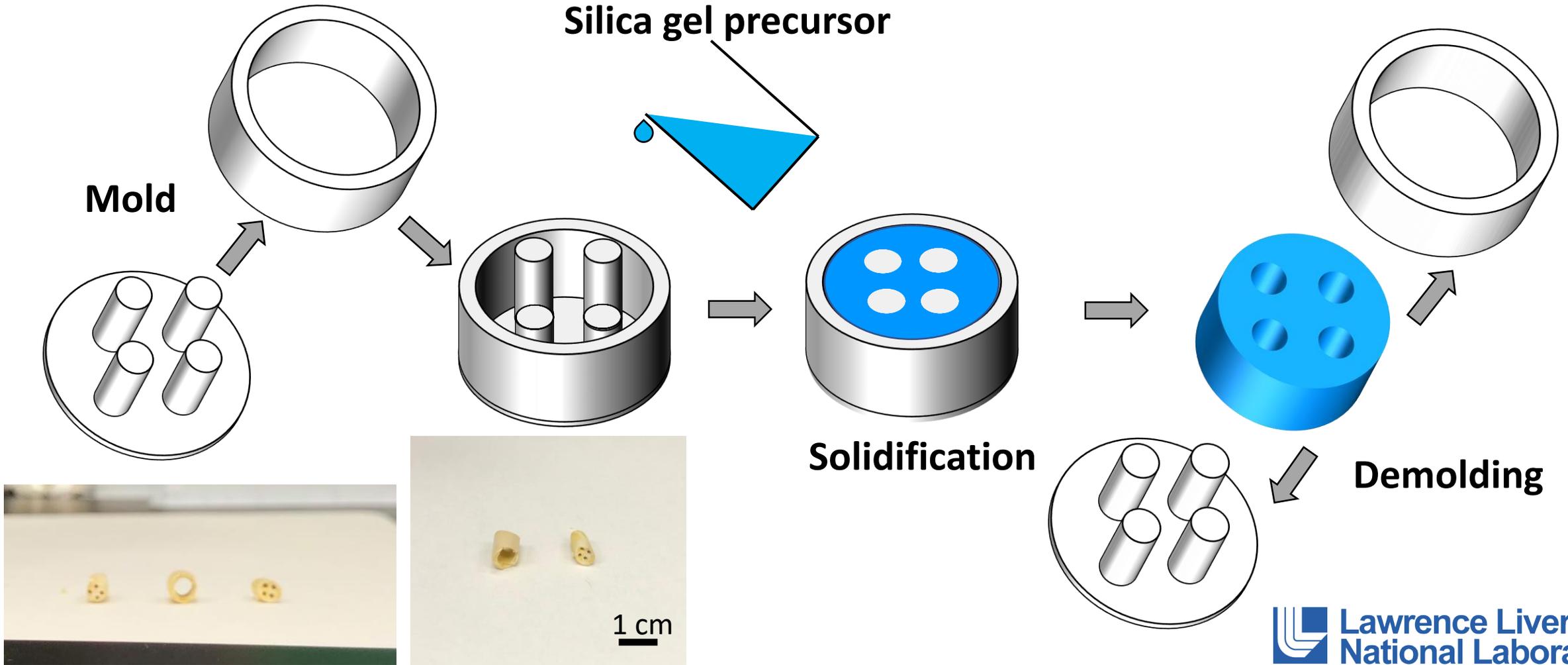
Technology-to-Market Path

- Improve on economics and transition towards scale-up
- Engage with partners to scale up biosorbent production
- Integrate packed-bed bioreactors within NETL pilot programs
- Potential industrial partners: La Paz Rare Earth Project, Solvay, Drylet



Preparing Project for Next Steps

Silica gel monolith for scale-up



Concluding Remarks

Develop REE-selective biosorbent to enrich for REEs and Sc from acid leachate of coal byproducts

- Provide an economical and environmentally friendly option for enriching and concentrating REEs from coal byproducts, with decision points made:
 - Feedstock selection – lignite and PRB fly ash
 - Cell immobilization strategy – Si Sol-gel
 - Two-stage Sc and REE+Y recovery
 - A TEA informed process design
- Next steps for tech development
 - Complete 2-stage Sc and REE+Y recovery tests with feedstocks
 - Convert solution upon desorption into solids
 - Transitioning and scaling to bench scale production

In FY21 we will achieve an extraction efficiency of >80% and total REE purity >20 wt% from pre- and post- combustion coal byproducts.

Acknowledgements



Collaboration among LLNL, Duke U. and U. of Arizona

Dan Park



Co-PI

Helen Hsu-Kim



Hongyue Jin



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