Savannah River Remediation
AMENTUM | BECHTEL | JACOBS | BWXT

December 9, 2020

TANK GROUT BULK CHEMISTRY EXPERIMENTS

Greg Flach

Safety SRR Integrity SRR Ownership SRR Teamwork SRR Continuous Improvement

SRR-CWDA-2020-00085 Rev. 1

Savannah River Remediation
AMENTUM | BECHTEL | JACOBS | BWXT

Uncertainties Motivating Grout Bulk Chemistry Experimentation

- Inability to achieve modeled grout Eh ranges in SRNL real waste solubility experiments; uncertainty in Eh ranges to test in any future solubility experiments
- Is the range of Eh values under realistic conditions tighter than assumed in PA analyses, such that different solubilities for key radionuclides could be predicted?
- Is Eh > +0.45v a realistic possibility, such that Pu solubility is orders of magnitude higher than assumed in PA modeling?
- Can Eh < -0.29v be achieved with slag-bearing cementitious materials, such that Tc is controlled by solubility under reducing conditions?
- How far above +0.1v can Eh rise under realistic conditions, such that Np solubility increases rapidly?

Summary of Grout Samples and Test Methods for SREL Experiments

Formulations

- Tank Fill Grout (TFG) used in Tanks 5, 6, 12, 16, 18, & 19 (Mix LP#8-16)
- TFG no BFS (slag replaced with equal weight of additional fly ash)
- Common Controlled Low-Strength Material (CLSM) = fly ash + small amount of cement

Forms

- Size-reduced (~2 mm) paste + sand
- Crushed paste + sand
- Monolithic grout cylinders

Solution contact

- Batch
- Column

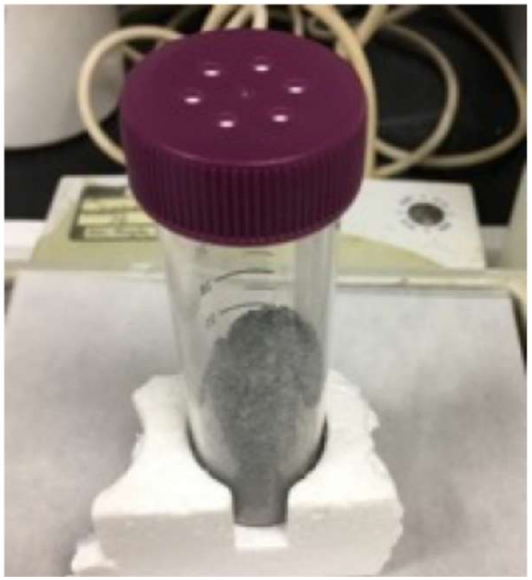
Exposure environments

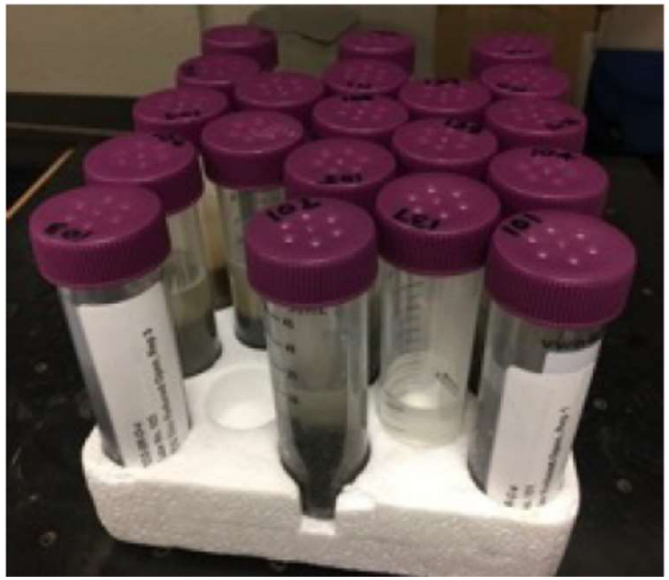
- Open atmosphere / Oxic
- H2 / Reducing
- N2 / Anoxic

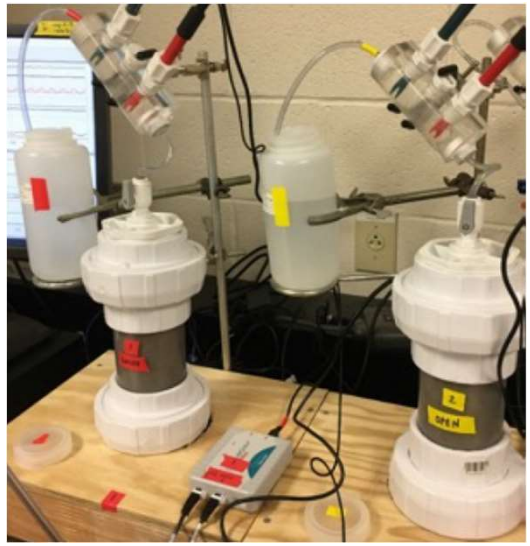
Measurements

- pH, Eh, major cations
- XRF, XRD

Batch and Column Tests (SREL)

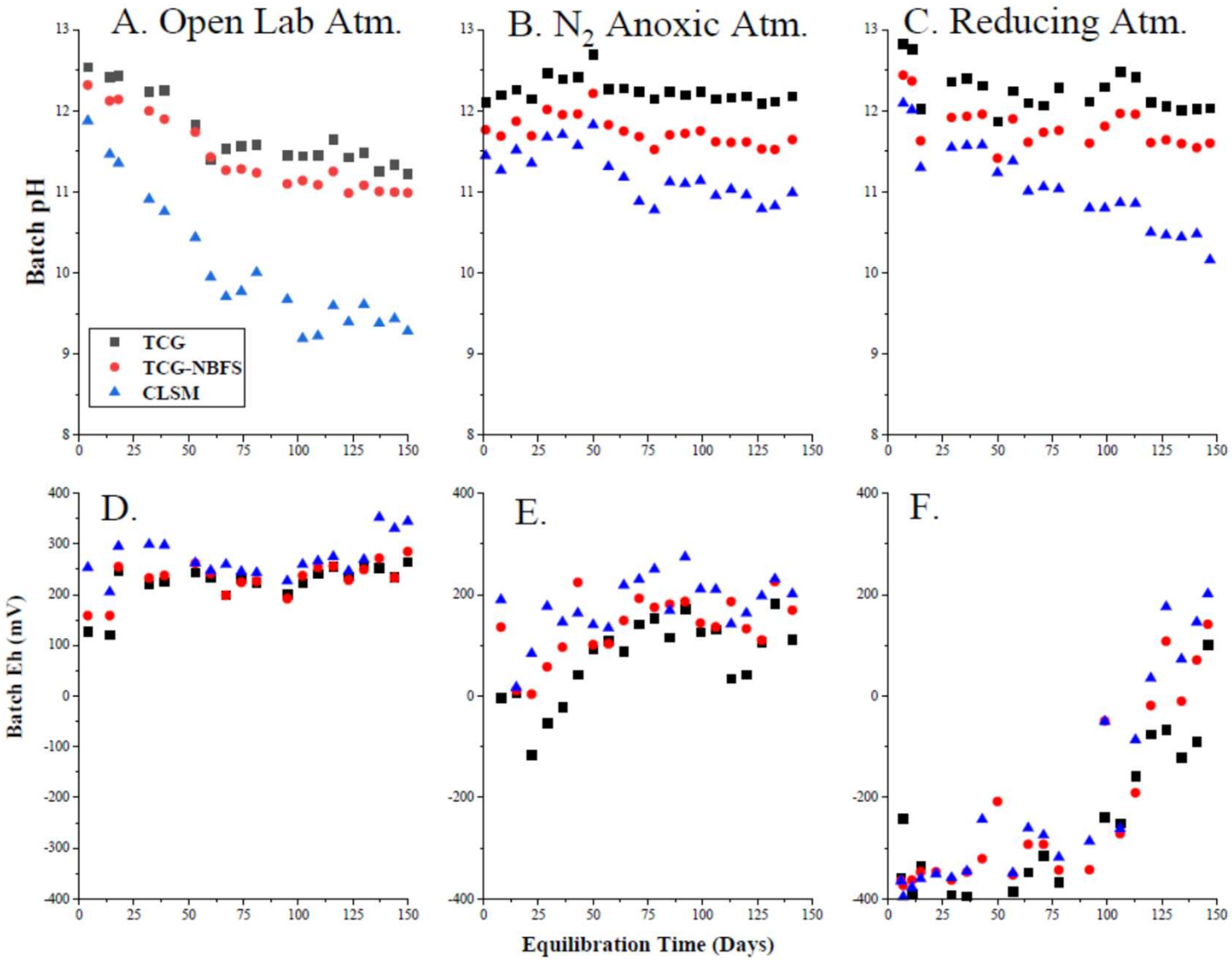




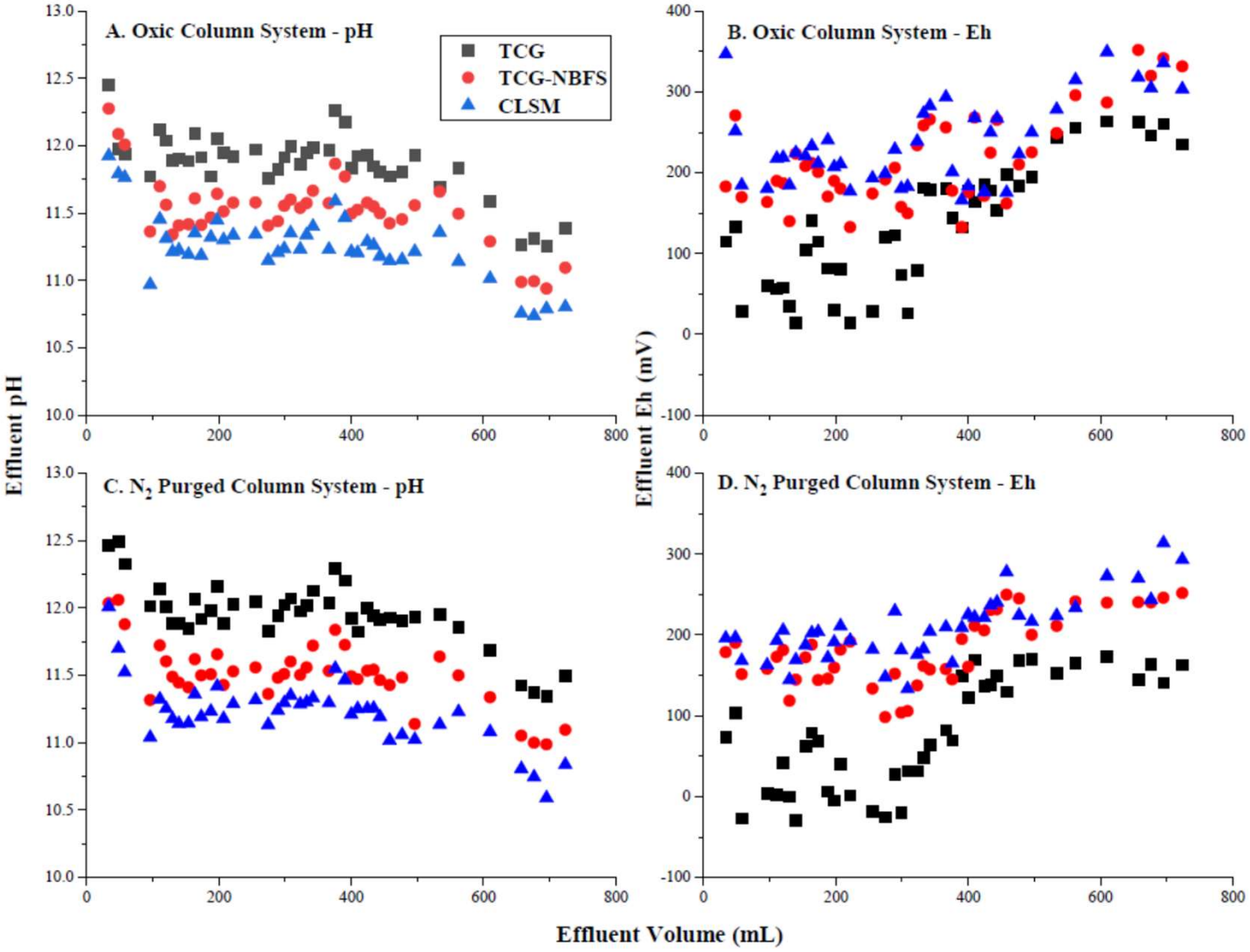
SRR-CWDA-2020-00085 Rev. 1

Batch Study (SREL R-21-0001 Fig. 9)



SRR-CWDA-2020-00085 Rev. 1

Column Study (SREL R-21-0001 Fig. 12)



SRR-CWDA-2020-00085 Rev. 1

XRD Results (SREL R-21-0001)

Table 16. Results for the Rietveld Refinement of the batch-treated TCG formulations.

TCG Sample	Phase (wt%)											
	Amorphous	*Mullite*	*Quartz*	*Hematite*	*Magnetite*	*Calcite*	*Ettringite*	*Strätlingite*	*Hydrotalcite*	*Kuzelite*	*Portlandite*	*Hemicarbo-aluminate*
REF (Non-Leached)	78.40	6.06	5.44	1.22	0.14	1.14	0.91	Trace *	1.42	0.78	0.42	4.07
OPEN	77.85	6.55	5.64	1.30	0.22	5.37	-	Trace	2.67	0.41	-	-
CLOSED	78.92	5.48	5.28	1.29	0.15	3.38	-	Trace	3.07	1.75	0.08	-
N_2	79.66	6.64	5.96	1.44	0.24	1.30	-	Trace	2.78	1.77	0.20	-

Table 15. Results for the Rietveld Refinement of the batch-treated TCG-NBFS formulations.

TCG-NBFS Sample	Phase (wt%)									
	Amorphous	*Mullite*	*Quartz*	*Hematite*	*Magnetite*	*Calcite*	*Strätlingite*	*Ettringite*	*Hemicarbo-aluminate*	*Monocarbo-aluminate*
REF (Non-Leached)	66.97	11.69	8.64	1.79	0.99	2.63	1.85	1.41	2.61	1.43
OPEN	68.66	11.87	9.07	1.81	1.01	6.08	1.51	-	-	-
CLOSED	72.58	11.32	9.12	1.73	1.07	2.79	-	-	-	1.39
N_2	71.17	11.77	8.55	1.86	0.79	2.27	1.75	-	-	1.84

Preliminary Thoughts

- **PA validations: Alkali metals leave quickly, predicted mineralogy generally consistent, pH prediction good, solubilities based on Eh lower than model prediction for oxidized conditions**
- **Opportunities: Add hemicarboaluminate to mineral set, consider partial hydration, update C-S-H solid solution model, better predict Eh, reconsider Eh range for solubility assignments**
- **Eh challenges:**
  - Eh probes biased with respect to some redox couples
  - Disequilibrium in experiments due to short timescale
  - Modeling assumes equilibrium but inherent redox disequilibrium even at long times
  - Need to integrate experimental and modeling insights