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Elimination of Acid Base Criticality Scenarios in the HM-Process

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Savannah River Nuclear Solutions, LLC
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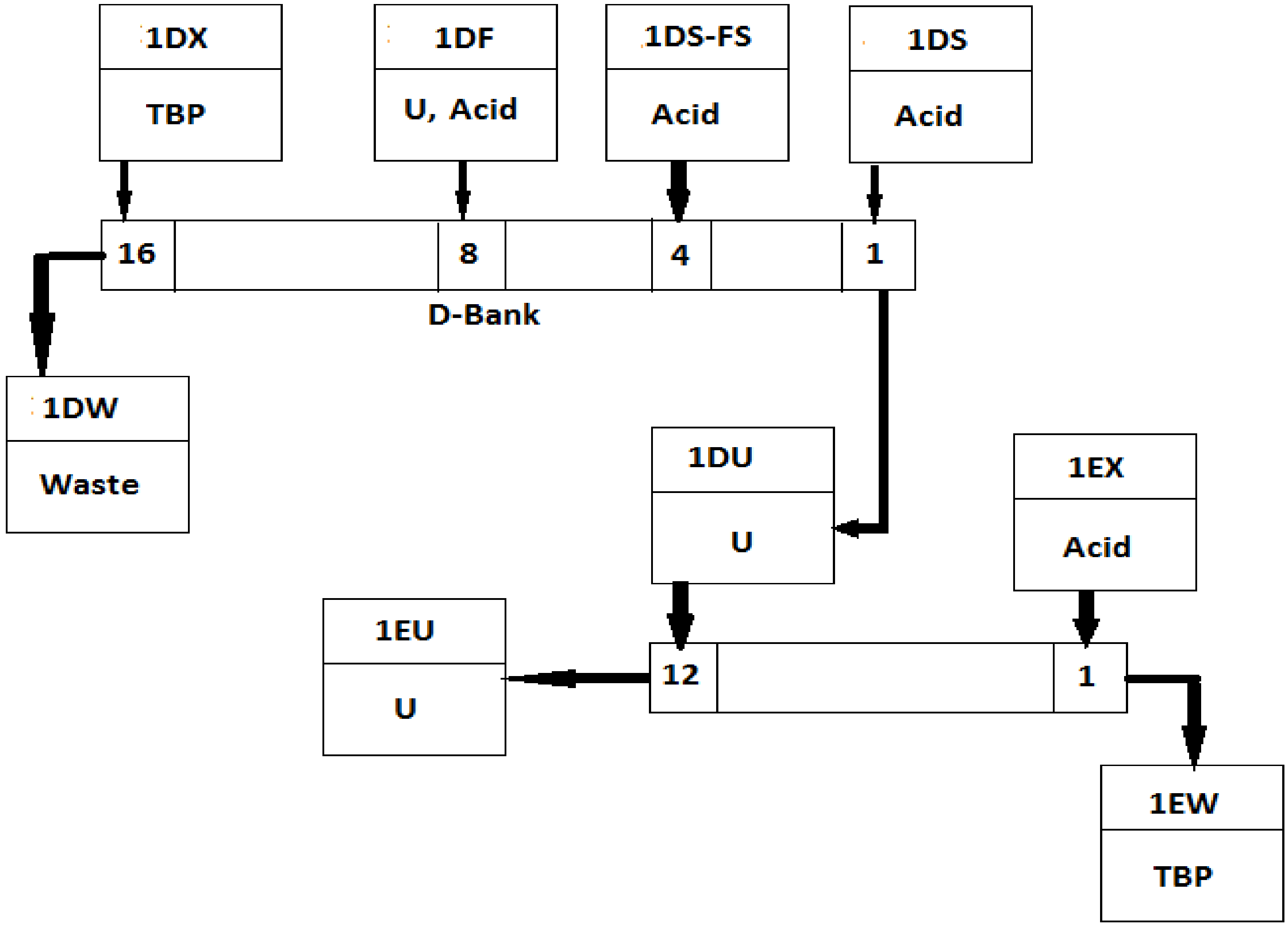
Introduction

- The HM-Process
  - Aqueous 2 cycle uranium extraction and purification using low volume percent TBP to extract uranium from nitric acid solution
  - Mixer-settlers are not geometrically safe
- Safety analysis postulates 36 criticality scenarios based on reflux leading to accumulation of fissile material
  - Due to upset in a process stream
  - Conservative assumptions made many years go with less sophisticated models
  - Assumption that reflux continuously builds up fissile material in 3 stages
    - *Distribution goes to 0 or infinity – not physically achievable*

HM-Process

- 1st Uranium Cycle
  - Process that separates the uranium from the fuel matrix, fission products, and most of the transuranic actinide content
- 2nd Uranium Cycle
  - One 16-stage bank (D) and one 12-stage bank (E)
  - Purifies uranium and removes any remaining transuranic content

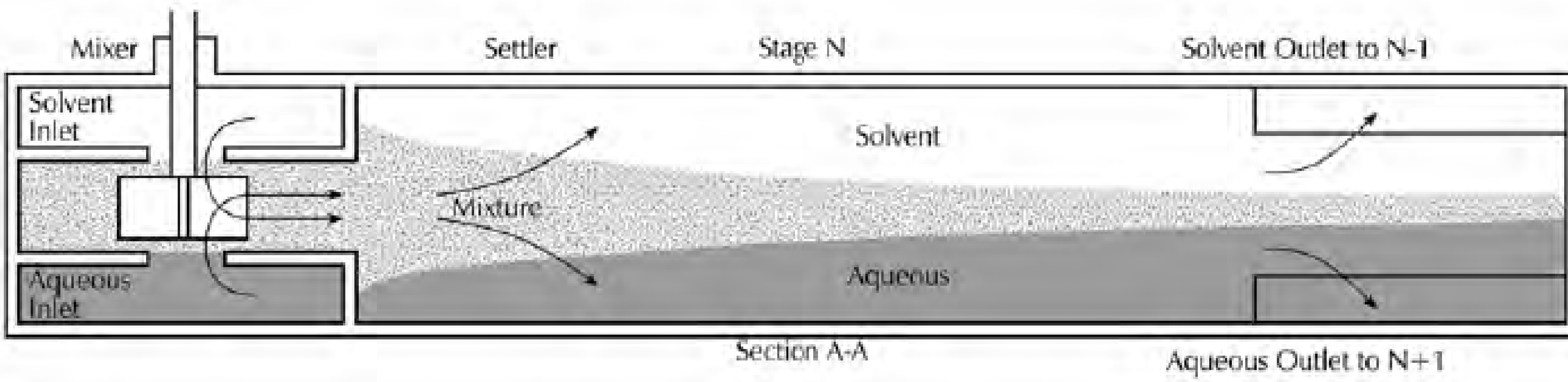
HM-Process



1DX
TBP
1DF
U, Acid
1DS-FS
Acid
1DS
Acid
16
8
4
1
D-Bank
1DW
Waste
1DU
U
1EX
Acid
1EU
U
12
1
1EW
TBP

What is a Mixer-Settler?

Parameter (all dimensions in inches)	16-Stage Bank	12-Stage Bank
Settling section height x width x length	10x8x48	12x12x108
Mixing section height x width x length	8x8x9	10x12x13.5
False bottom height	2	2
Bottom, front, and back plate thickness	0.5	0.5
Top and side plate thickness	0.25	0.25



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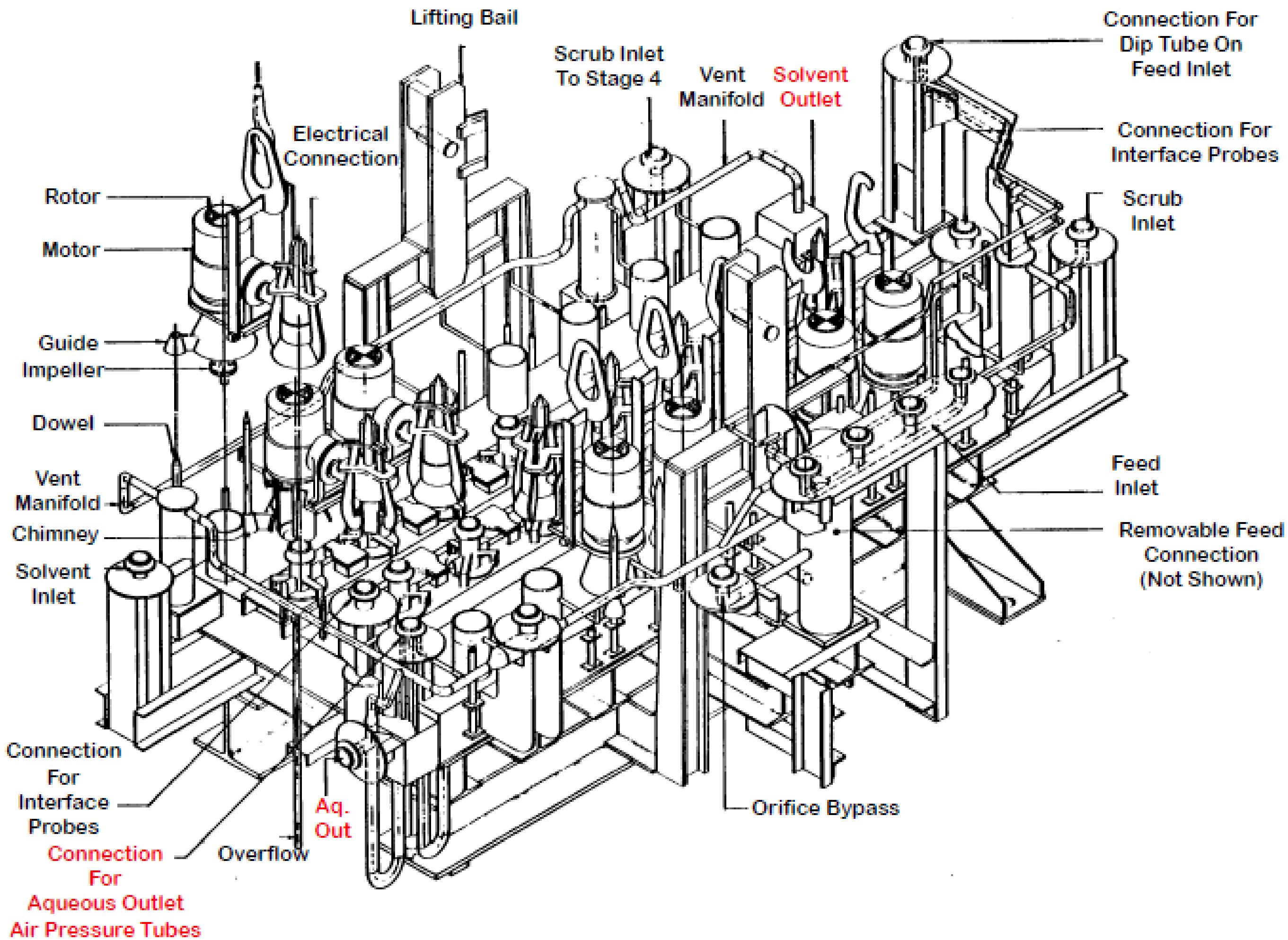

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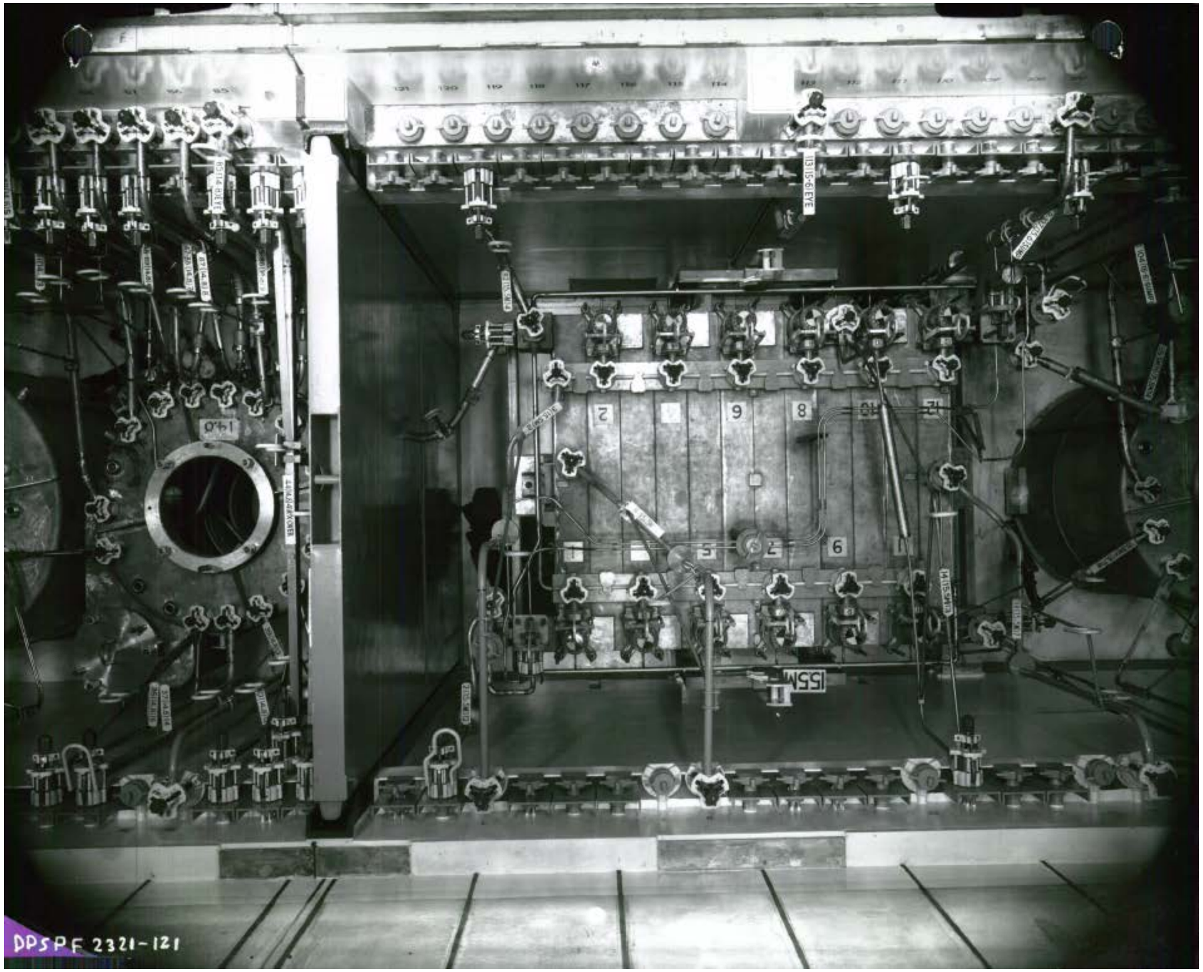


What is a Mixer-Settler?

What is a Mixer-Settler?

Postulated Events

- Conclusion of a two year process to begin removing incredible events
- D-Bank acid (1DS stream) becomes dilute in nitric acid
  - Drops below minimum acid limit
  - Uranium not extracted into organic phase and accumulates in aqueous phase of D-Bank
- D-Bank acid (1DS stream) becomes concentrated in nitric acid
  - Exceeds maximum acid limit
  - Acid carry over to E-Bank increases, acidity in E-Bank increases reducing the amount of purified uranium leaving the bank

Normal, Credible Abnormal, & K-safe

- Normal = Operational Setpoint
  - 1DS is set to 0.95 M nitric acid at 3.31 L/min flow
- Credible Abnormal = Operational Band Limits
  - 0.8 M to 1.5 M
  - Flowrate remains the same
  - Once exceeded process must be immediately corrected by operations or shut down
- Safe multiplication factor
  - K-safe = 0.9564

Validation

- SEPHIS and SEPHIS-P
  - 1970s era FORTRAN code maintained by SRS
  - 1998 Formally validated for HM-Process chemistry modeling
  - 2015 ported to PYTHON programming language and given basic GUI, identical methodology – no changes to results
- SCALE 6.1
  - Internal, bias determined for HEU aqueous processing systems

Need for Experimental Basis

- SEPHIS historically used for flowsheet development – not safety analysis
- No simulation is perfect – SEPHIS-P has some bias to it
  - Overprediction for HM-process tends to be toward aqueous uranium concentration
- Debated various ways to show SEPHIS-P was bounding the chemistry conditions
- Concluded needed experimental basis
- 1DS acid upset were modeled in process mock-up equipment by SRNL
- Confirmed SEPHIS-P was adequately modeling the process chemistry with a bias toward aqueous uranium

SEPHIS Calculations

- Based on the 73 wt.% uranium enrichment flowsheet
- 1DS Acid Molarity:
  - Normal: 0.95 M
  - Credible Abnormal: 0.8 M (low), 1.5 M (high)
  - Analyzed Range: 0.05 M to 3.5 M
- 1DS Flow: 3.31 L/min
- 1DF Properties:
  - 8.67 L/min
  - 5.5 M acid
  - 5.25 g U/L (4.52 g U-235/L)
- 1DX: 26.02 L/min solvent at 7.5 vol% TBP

SEPHIS Calculations

- Establish a SEPHIS model of the 2nd Uranium cycle
- Hold all process parameters other than 1DS acid molarity at normal value
  - No two concurrent, independent upsets
- Generate steady state (equilibrium conditions) at 1DS acid molarity values:
  - 0.05, 0.10, 0.30, 0.50, 0.65, <u>0.80</u>, 0.95, 1.25, <u>1.50</u>, 1.75, 2.00, 2.25, 2.75, 3.00, and 3.50 M

SEPHIS Calculations

- Assumptions
  - No plutonium, transuranics, or fission products modeled
  - Ferrous sulfamate reductant added to D-Bank stage 4
  - Utilized the setpoints and ranges from the 73 wt.% enriched uranium flowsheet
  - Settling sections are controlled to 50% by volume organic, and 50% aqueous

KENO Calculations

- Acquire stage-wise D and E Bank compositions
  - Aqueous and organic components
  - Mixing and settling sections
- Transform into SCALE compatible compositions
  - Organic: n-paraffin, TBP, uranyl nitrate, acid
  - Aqueous: acid, uranyl nitrate hexahydrate, water
  - Reduce to H, C, O, P, N, U-235, and U-238
  - Separate in settling, combined in mixing section → 3 compositions per stage

Model of Mixer Settler – Chemical Species

- 3 unique compositions per stage
- Each stream in each stage composed of H, C, O, P, N, U-235, U-238
  - 73 wt.% uranium enrichment
- Chemical species listed below:

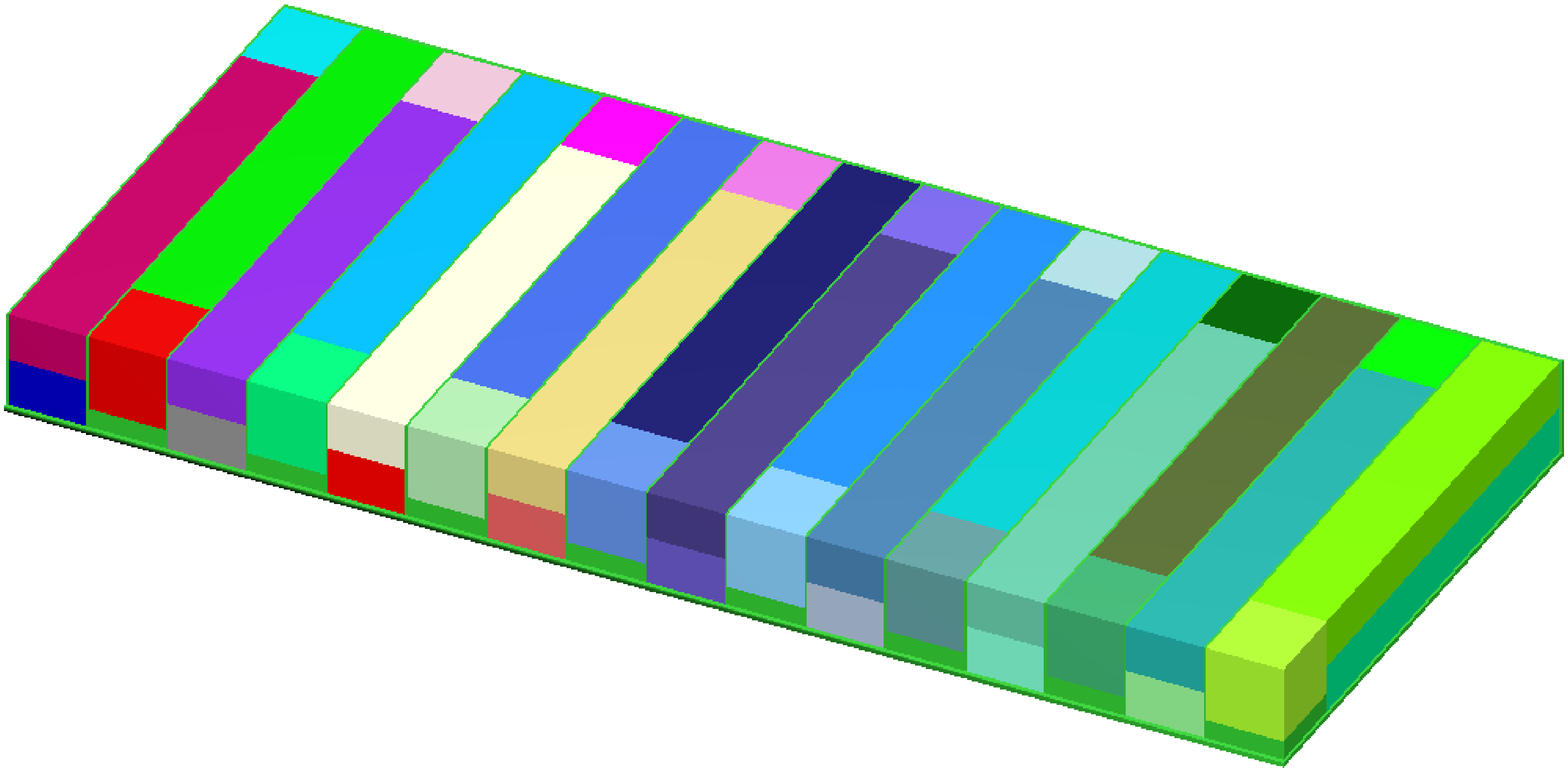
Organic

Name	Chemical Formula	Theoretical Density (g/cm^3)
Tributyl Phosphate (TBP)	$C_{12}H_{27}O_4P$	0.9727
Nitric Acid	HNO_3	1.55
n-paraffin (assumed):	--	--
20% dodecane	$C_{12}H_{26}$	0.749
40% tridecane	$C_{13}H_{28}$	0.756
40% tetradecane	$C_{14}H_{30}$	0.763
Uranyl nitrate	$UO_2(NO_3)_2$	2.203

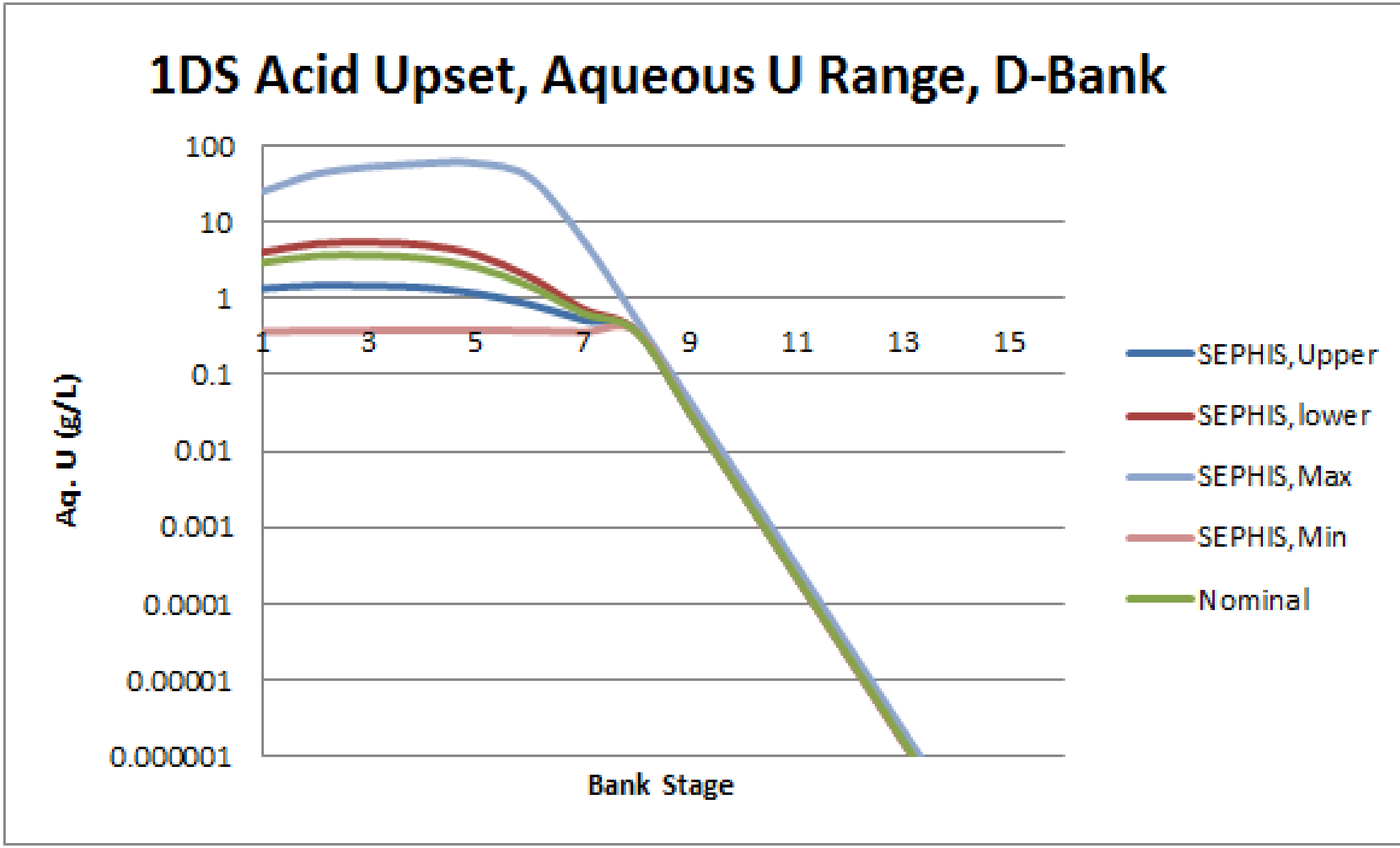
Aqueous

Name	Chemical Formula	Theoretical Density (g/cm^3)
Water	H_2O	0.9982
Nitric Acid	HNO_3	1.55
Uranyl Nitrate (aqueous)	$UO_2(NO_3)_2 * 6(H_2O)$	2.4183

Methodology – SCALE Models

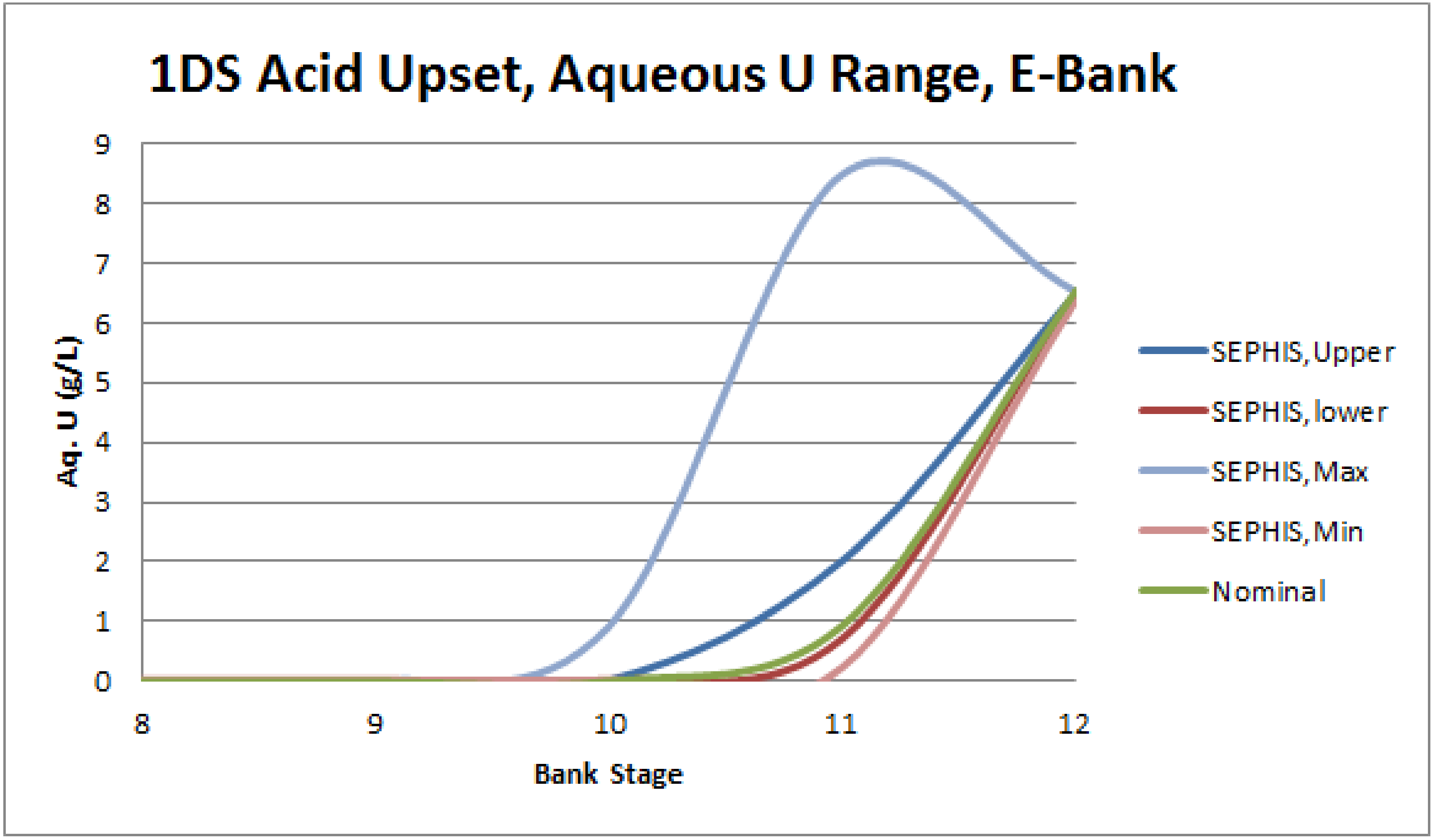


Results



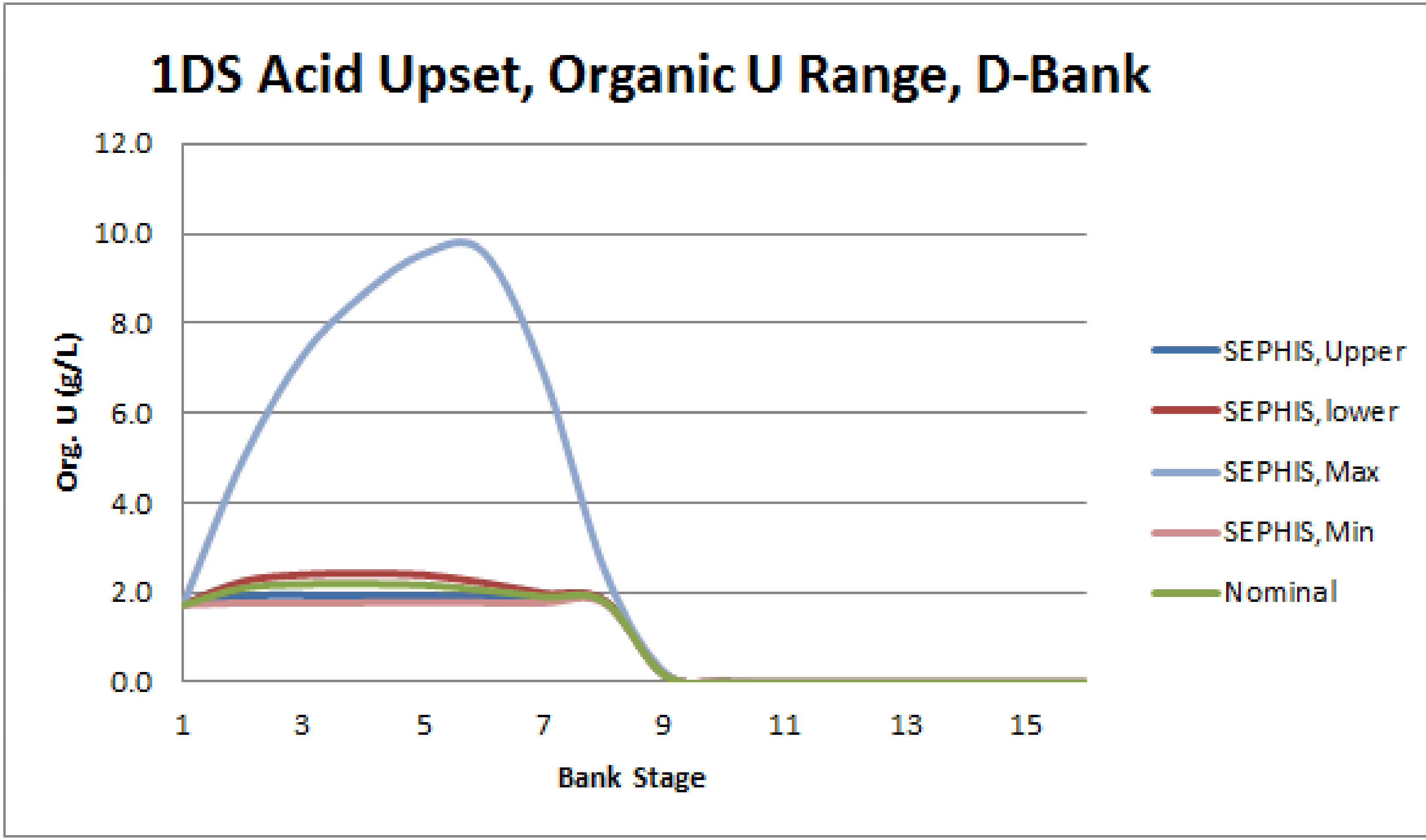
1DS Acid Upset, Aqueous U Range, D-Bank
100
10
1
0.1
0.01
0.001
0.0001
0.00001
0.000001
1
3
5
7
9
11
13
15
Aq. U (g/L)
Bank Stage
SEPHIS, Upper
SEPHIS, lower
SEPHIS, Max
SEPHIS, Min
Nominal

Results



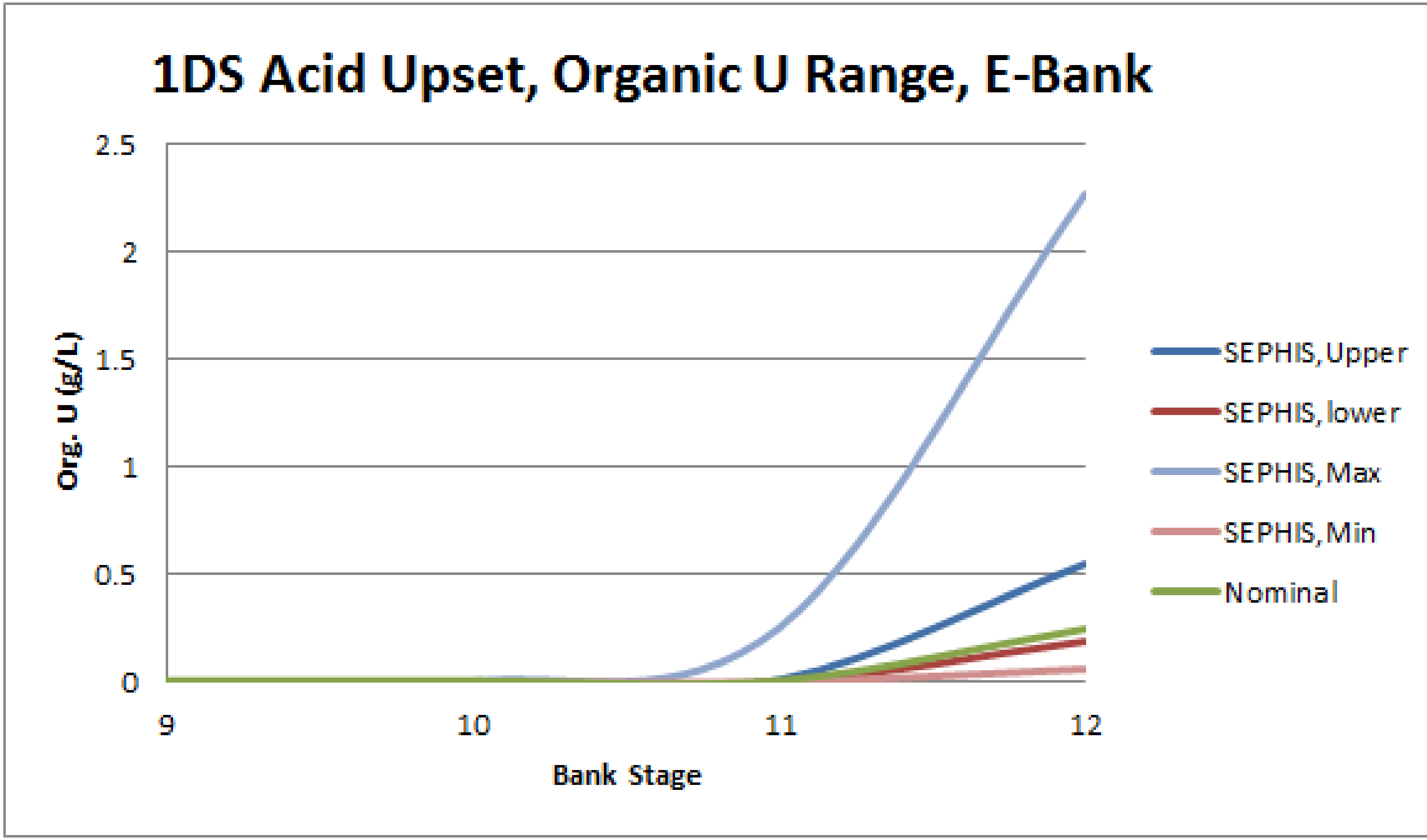
1DS Acid Upset, Aqueous U Range, E-Bank
9
8
7
6
5
4
3
2
1
0
Aq. U (g/L)
8
9
10
11
12
Bank Stage
SEPHIS, Upper
SEPHIS, lower
SEPHIS, Max
SEPHIS, Min
Nominal

Results



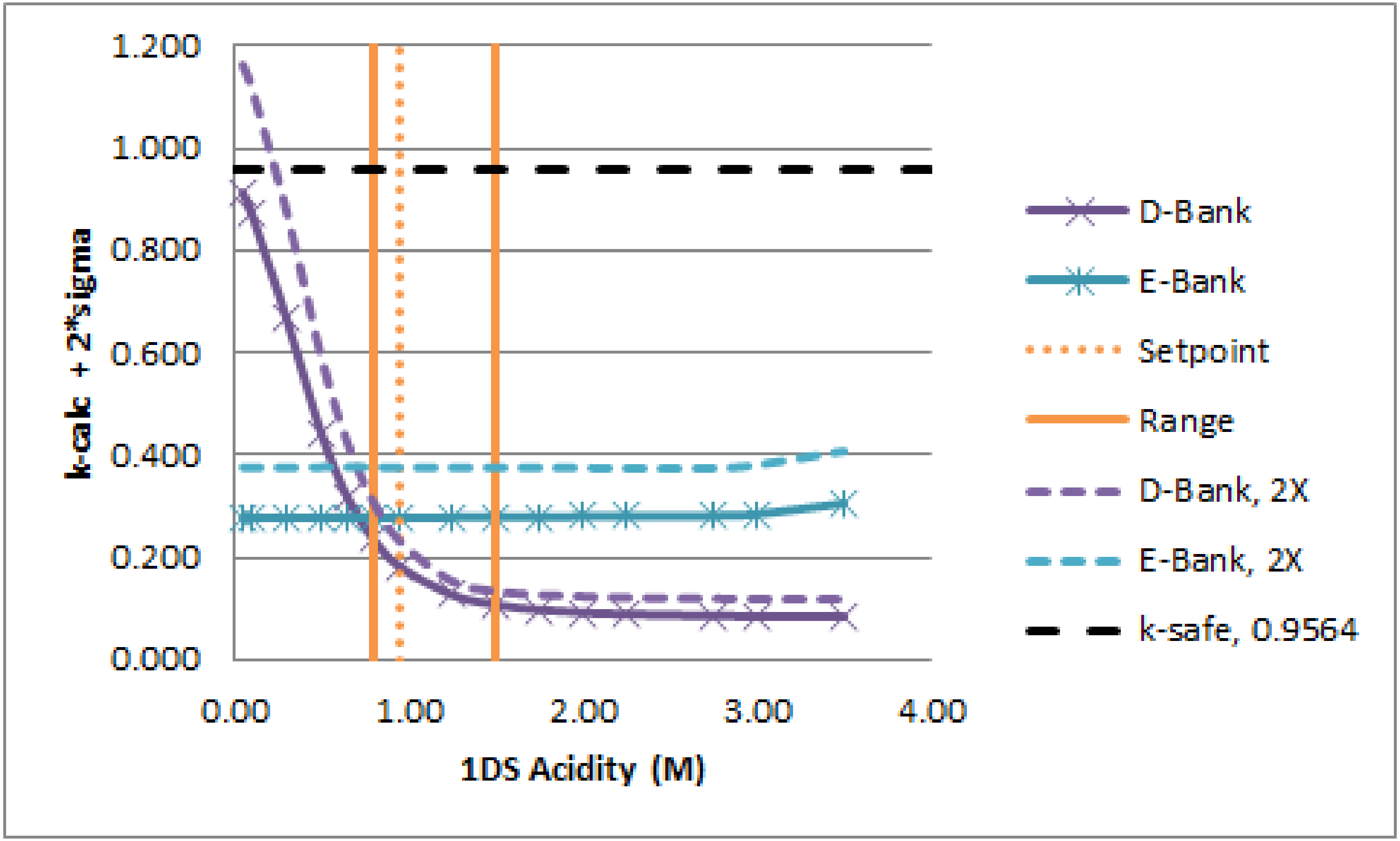
1DS Acid Upset, Organic U Range, D-Bank
12.0
10.0
8.0
6.0
4.0
2.0
0.0
Org. U (g/L)
1
3
5
7
9
11
13
15
Bank Stage
SEPHIS, Upper
SEPHIS, lower
SEPHIS, Max
SEPHIS, Min
Nominal

Results



1DS Acid Upset, Organic U Range, E-Bank
2.5
2
1.5
1
0.5
0
Org. U (g/L)
9
10
11
12
Bank Stage
SEPHIS, Upper
SEPHIS, lower
SEPHIS, Max
SEPHIS, Min
Nominal

Results



1.200
1.000
0.800
0.600
0.400
0.200
0.000
k-calc + 2*sigma
0.00
1.00
2.00
3.00
4.00
1DS Acidity (M)
D-Bank
E-Bank
Setpoint
Range
D-Bank, 2X
E-Bank, 2X
k-safe, 0.9564

Aqueous Phase vs. Multiplication

1DS Acid Conc.	Organic Volume Fraction	Aqueous Volume Fraction	Bank	k_{BE}
0.05 M	67%	33%	D	0.754
0.05 M	50%	50%	D	0.901
0.05 M	33%	67%	D	1.020
0.05 M	50%, 2x height	50%, 2x height	D	1.160
0.10 M	67%	33%	D	0.717
0.10 M	50%	50%	D	0.859
0.10 M	33%	67%	D	0.979
0.10 M	50%, 2x height	50%, 2x height	D	1.112
3.00 M	67%	33%	E	0.213
3.00 M	50%	50%	E	0.285
3.00 M	33%	67%	E	0.329
3.00 M	50%, 2x height	50%, 2x height	E	0.379
3.50 M	67%	33%	E	0.223
3.50 M	50%	50%	E	0.305
3.50 M	33%	67%	E	0.354
3.50 M	50%, 2x height	50%, 2x height	E	0.408

Approval Process

- No user qualification exists for SEPHIS-P, expert based
- Author learned SEPHIS-P under guidance of expert user
- Analysis reviewed by facility personnel familiar with process chemistry
- Analysis reviewed by SRNL process chemists along with experimental work for 1DS upset
- Concurrence issued that the analysis was appropriate and SEPHIS-P was conservative for this application
- Facility and Criticality Safety management both satisfied with concurrence → accept analysis

Conclusions & Future Work

- Eliminates 2 of 36 postulated cycle criticality scenarios
- Sets basis for future analysis with SRNL support
- Allows facility to eventually downgrade the functional classification of certain equipment
  - Saves time, money, dose

Acknowledgements

- Funded by DOE-EM under operational budget for H-Canyon
- Thanks to SRNL chemists Tara Smith and Edward Kyser
- Dedicated to Mark Crowder