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The Effect of Phase Disengagement on Criticality Safety Calculations for Aqueous Separations

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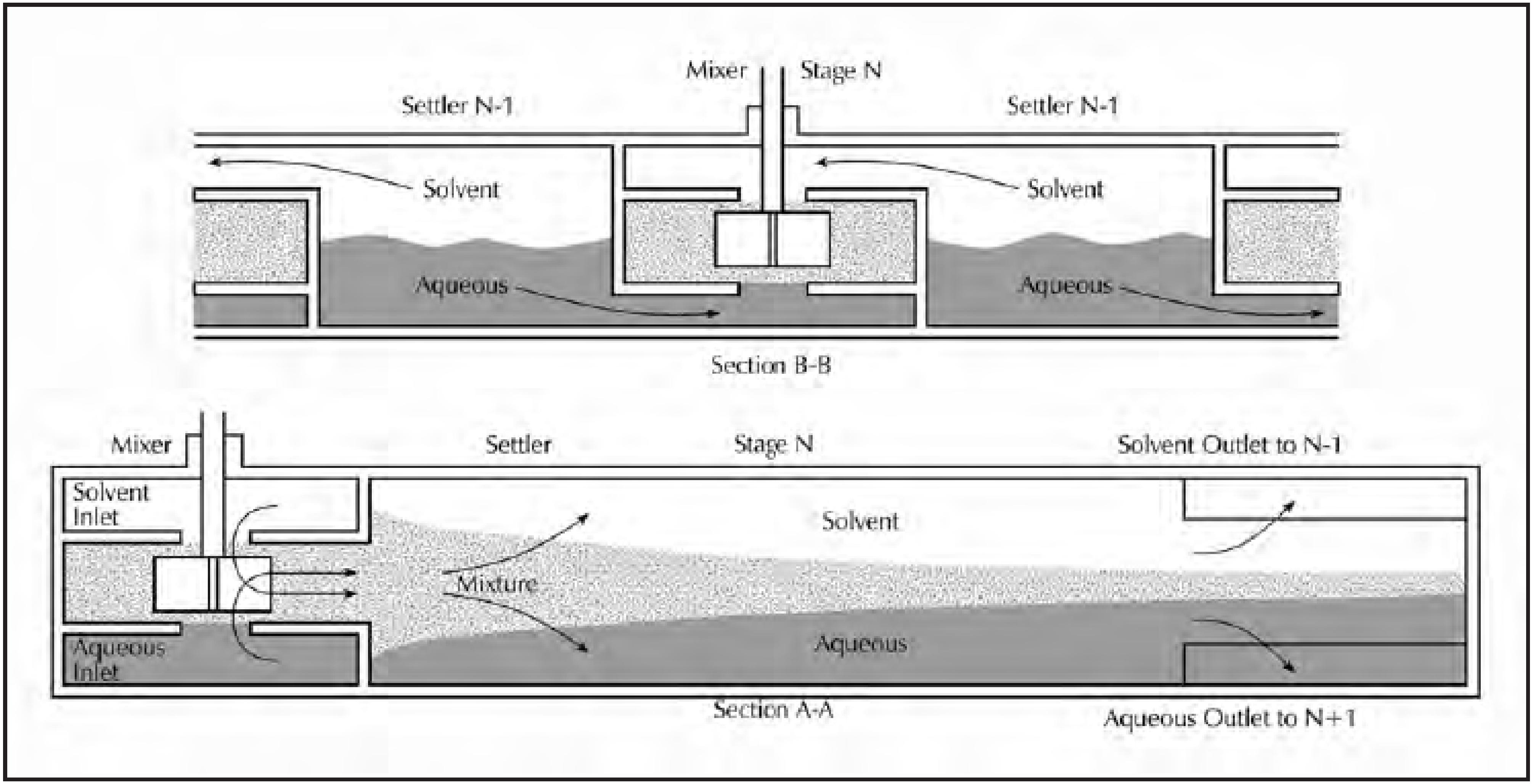
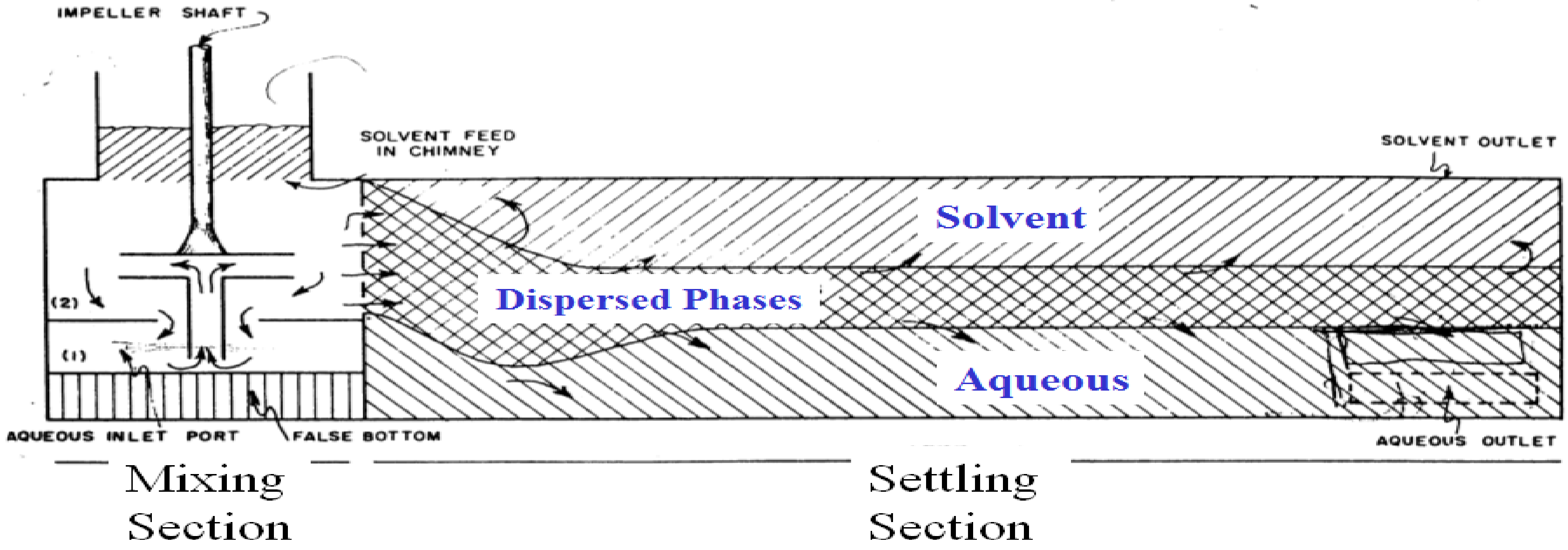
Introduction – What is a Mixer-Settler?



Introduction – What is a Mixer-Settler?

Introduction

- 2-Phase solvent extraction relies on the two phases rapidly disengaging
- Immiscible solutions of different densities
- Previous criticality safety analyses have assumed instant disengagement
- Is this conservative?



Process Description

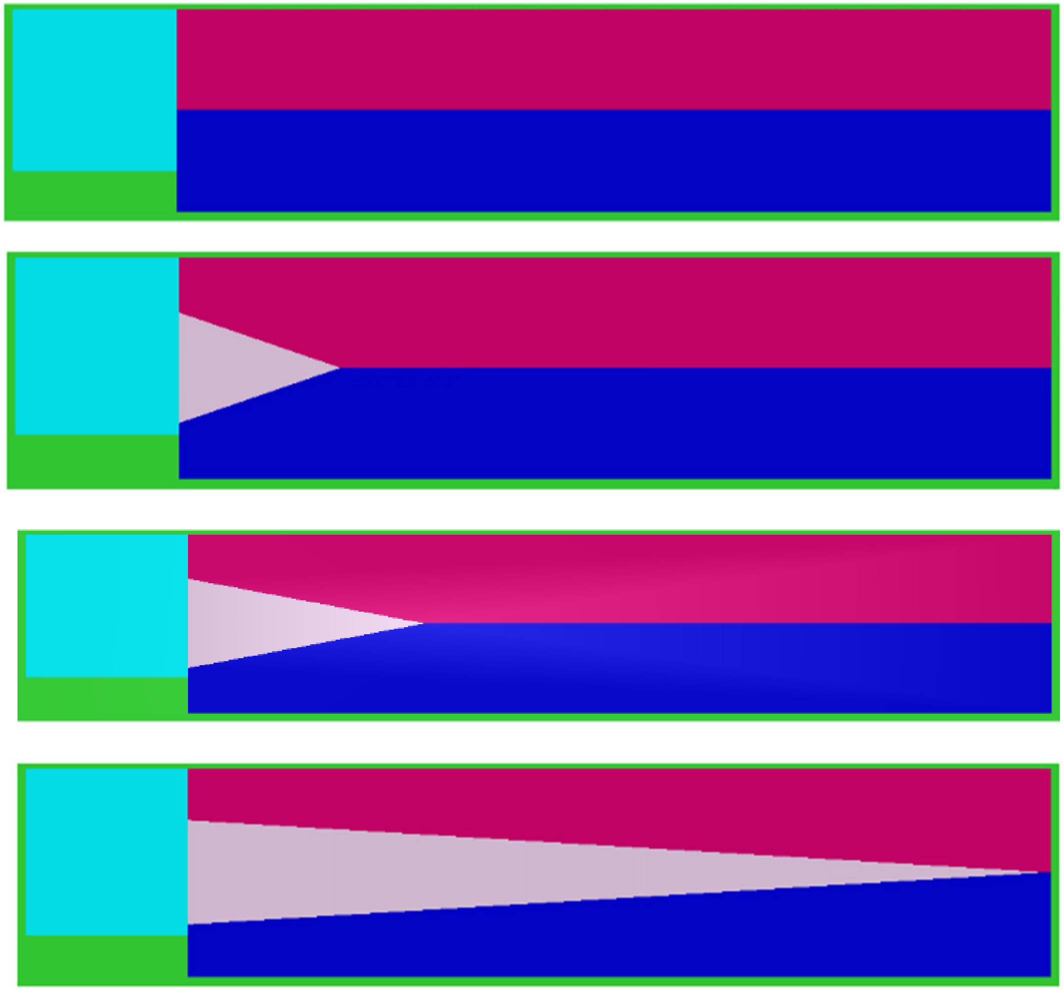
- **Examined in regard to H-Canyon's HM-Process (derivative of Purex)**
- **Two cycles of solvent extraction used for uranium separation and purification**
- **Feed: uranium, fission products, transuranics dissolved in nitric acid**
- **Aqueous Phase**
  - Higher density, nitric acid solution
- **Organic Phase**
  - Lower density, 7.5 vol.% tributyl phosphate in normal-paraffin
- **Disengagement studied by Savannah River National Laboratory**
  - Fresh, clean solvent – 29 seconds to separate
  - Degraded, contaminated solvent – 43 seconds to separate
  - Inclusion of reducing agent – additional 7 to 16 seconds
- **Time available to disengage limited by mixer-settler geometry and flowrates**

Modeling of Phase Disengagement

- **Simplified Simulation, Four Degrees of Disengagement**
  - – Retain combined interspersed solution composition from mixing section
  - – Triangular wedge penetrating the settling section
  - – Base length of penetration on disengaging time from chemical experiments: $L = \frac{V_{org}+V_{aq}}{A} t$
  - – Total fissile mass in system preserved
  - – Mid-density interspersed phase is simulated
  - – Simplified model
    - *More complex models could be made*
    - *Computational fluid dynamics, drop-rise simulation, etc.*
    - *Additional rigor shown not to be necessary due to low impact*
  - – Baffles neglected

Modeling of Phase Disengagement

- **Phases:**
  - –Instant
  - –Clean solvent
    - *29 seconds*
  - –Degraded solvent
    - *43 seconds*
  - –Incomplete
    - *Adjusted time such that phases have not separated by end of settling section*
    - *52 to 207 seconds depending on stage size and flow rate*



Selection of Process Conditions

- **Two separations cycles with 30+ postulated scenarios to choose from**
- **Selected 1st Uranium Cycle**
  - Availability of SPEHIS-ACM model
  - Partitioning cycle that includes Np and Pu
  - Three banks to examine
- **Selected upsets in A-Bank and B-Bank acid concentration**
  - Operator controlled process with active engineered interlocks
  - Analyzed significantly beyond credible abnormal range
  - Acid upsets previously analyzed to have most significant changes in multiplication
- **Isotopics assumed:**
  - U: 73 wt.% U-235 and 27 wt.% U-238. U-234 and U-236 content are neglected
  - Np: 99.9 wt.% Np-237, 0.1 wt.% Np-236
  - Pu: 93.6 wt.% Pu-239, 6 wt.% Pu-240, and 0.4 wt.% Pu-241

SEPHIS-ACM Inputs

Parameter	Description
Process feed stream	6.1 L/min, 5.00 M acid 4.5 g U/L 0.0365 g Pu/L 0.000125 g Np/L
A-Bank acid stream	1.4 L/min, nominally 3.70 M, **modeled from 0.20 to 8.00 M**
A-Bank solvent stream	10.7 L/min
B-Bank acid stream	6.3 L/min, nominally 1.55 M, **modeled from 0.20 to 4.50 M**
B-Bank solvent stream	18.7 L/min
C-Bank acid stream	7.6 L/min, 0.02 M

SEPHIS-ACM Tracked Species

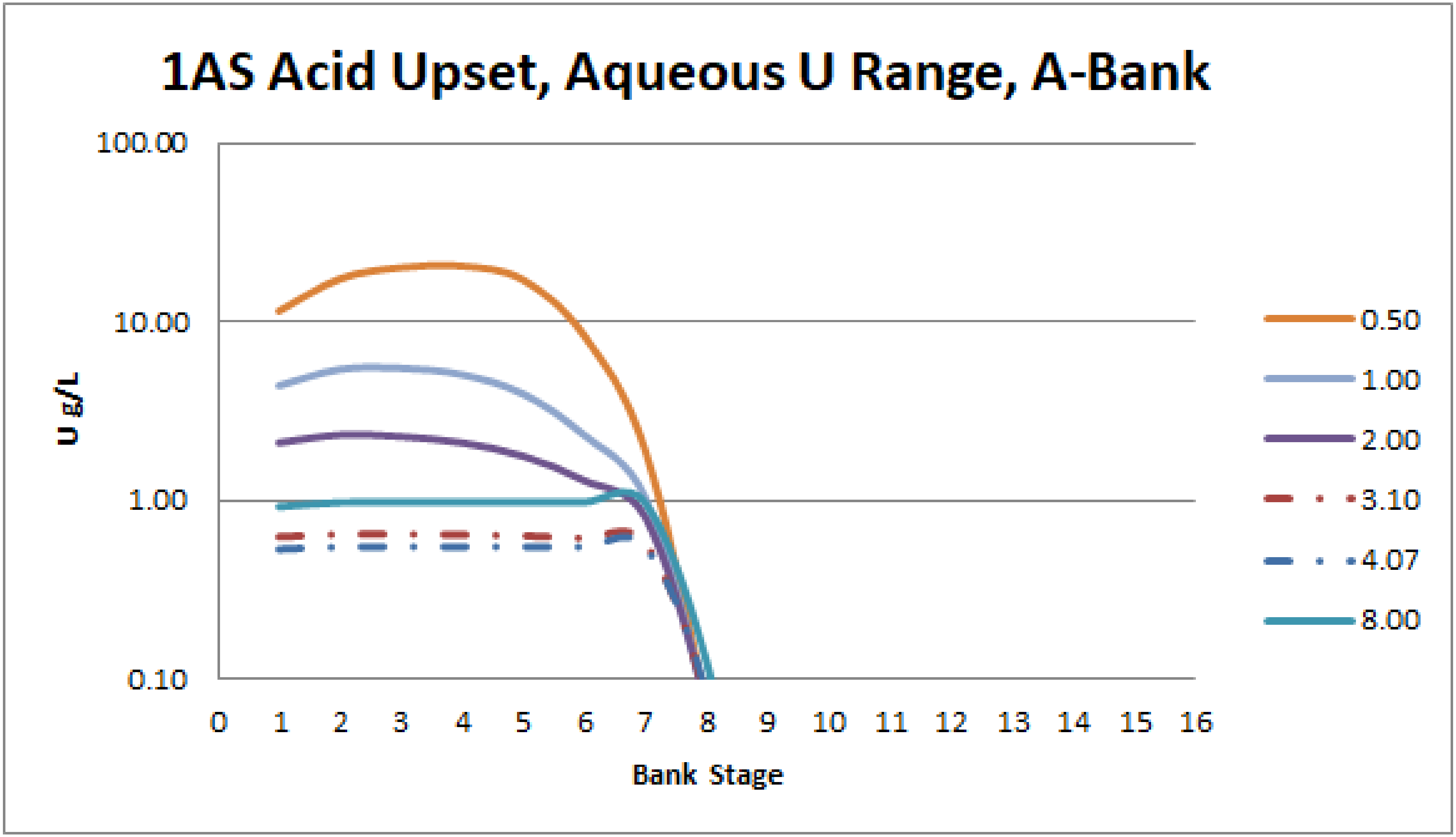
Species	Purpose
Aqueous Stream	
Al^{+3}	Al from salting solution and dissolved fuel
CO_3^{-2}	Carbonate from solvent wash
Fe^{+2}, Fe^{+3}	Fe from ferrous sulfamate reducing agent
H_2O	Solvent
H_3NO	Hydroxylamine reducing agent
H^+	Free acid
$NH_2SO_3^-$	Sulfamate ion from reducing agent
NO_2^-	Nitrite, reaction product
NO_3^-	Free nitrate
OH^-	Free base
SO_4^{-2}	decomposition product from ferrous sulfamate reducing agent
U^{+4}	U from reducing agent
UO_2^{+2}	Extractable U
Np^{+4}	Extractable Np
NpO_2^{+2}, NpO_2^{+1}	Less extractable Np species
Pu^{+4}	Extractable Pu
Pu^{+3}	Less extractable Pu species

Organic Stream	
$Al(NO_3)_3{*}2(C_{12}H_{27}O_4P)$	Extracted Al
$C_{12}H_{26}$	Organic diluent
$Fe(NO_3)_3{*}3(C_{12}H_{27}O_4P)$	Extracted Fe species
$HNO_3{*}2(C_{12}H_{27}O_4P)$	Dominant extracted nitric acid species
$HNO_3{*}(C_{12}H_{27}O_4P)$	
$Np(NO_3)_4{*}2(C_{12}H_{27}O_4P)$	Dominant extracted Np species
$NpO_2(NO_3)_1{*}3(C_{12}H_{27}O_4P)$	
$NpO_2(NO_3)_2{*}2(C_{12}H_{27}O_4P)$	
$Pu(NO_3)_3{*}3(C_{12}H_{27}O_4P)$	Dominant extracted Pu species
$Pu(NO_3)_4{*}2(C_{12}H_{27}O_4P)$	
$C_{12}H_{27}O_4P$	Tri-n-butyl phosphate extractant
$UO_2(NO_3)_2{*}2(C_{12}H_{27}O_4P)$	Dominant extracted uranium species

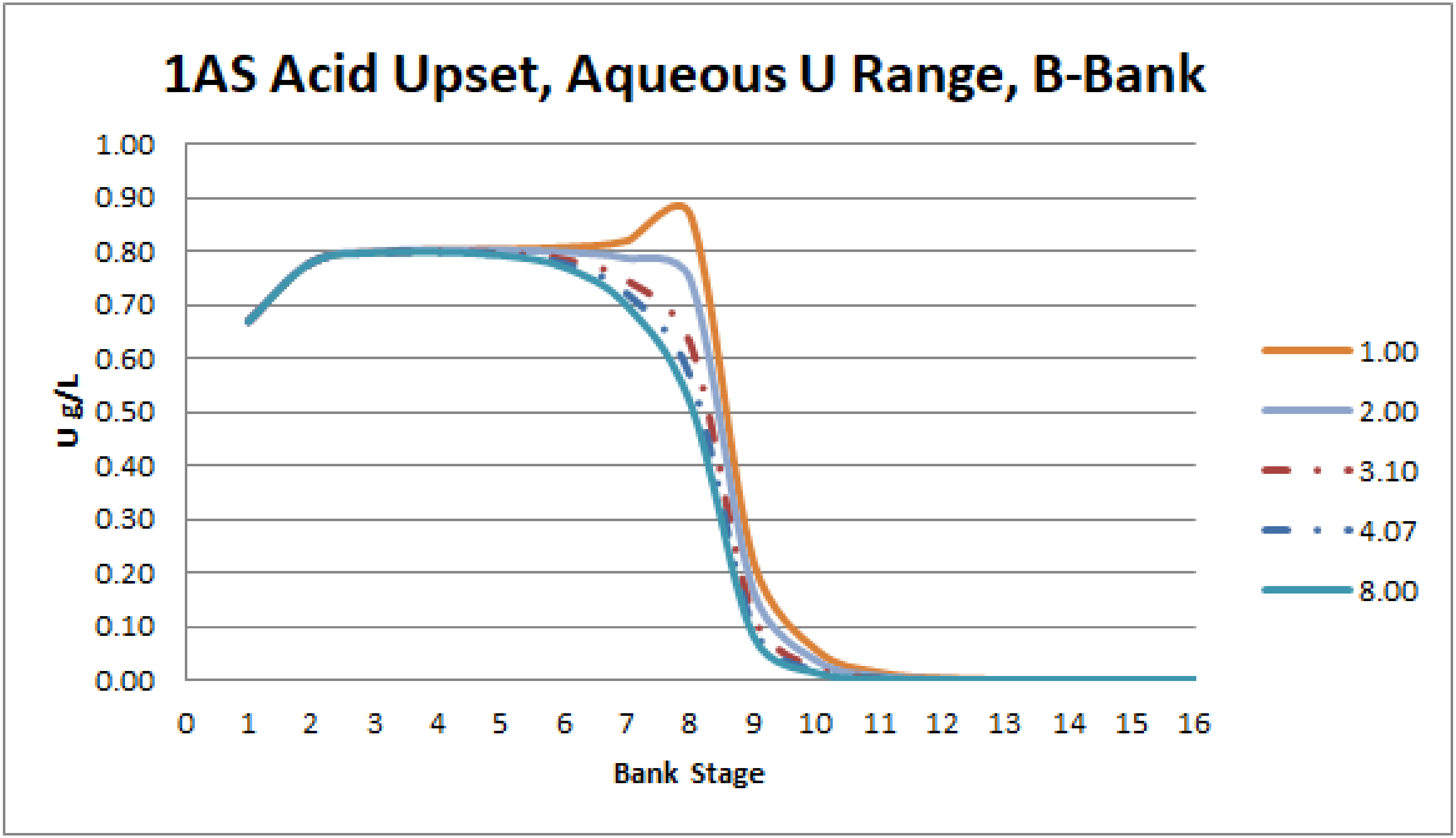
A-Bank Acid Upset

- Chemical behavior shown is representative, sufficient to show behavior of system
- Data series are the differing molarity of acid
- Dashed lines represent credible abnormal band

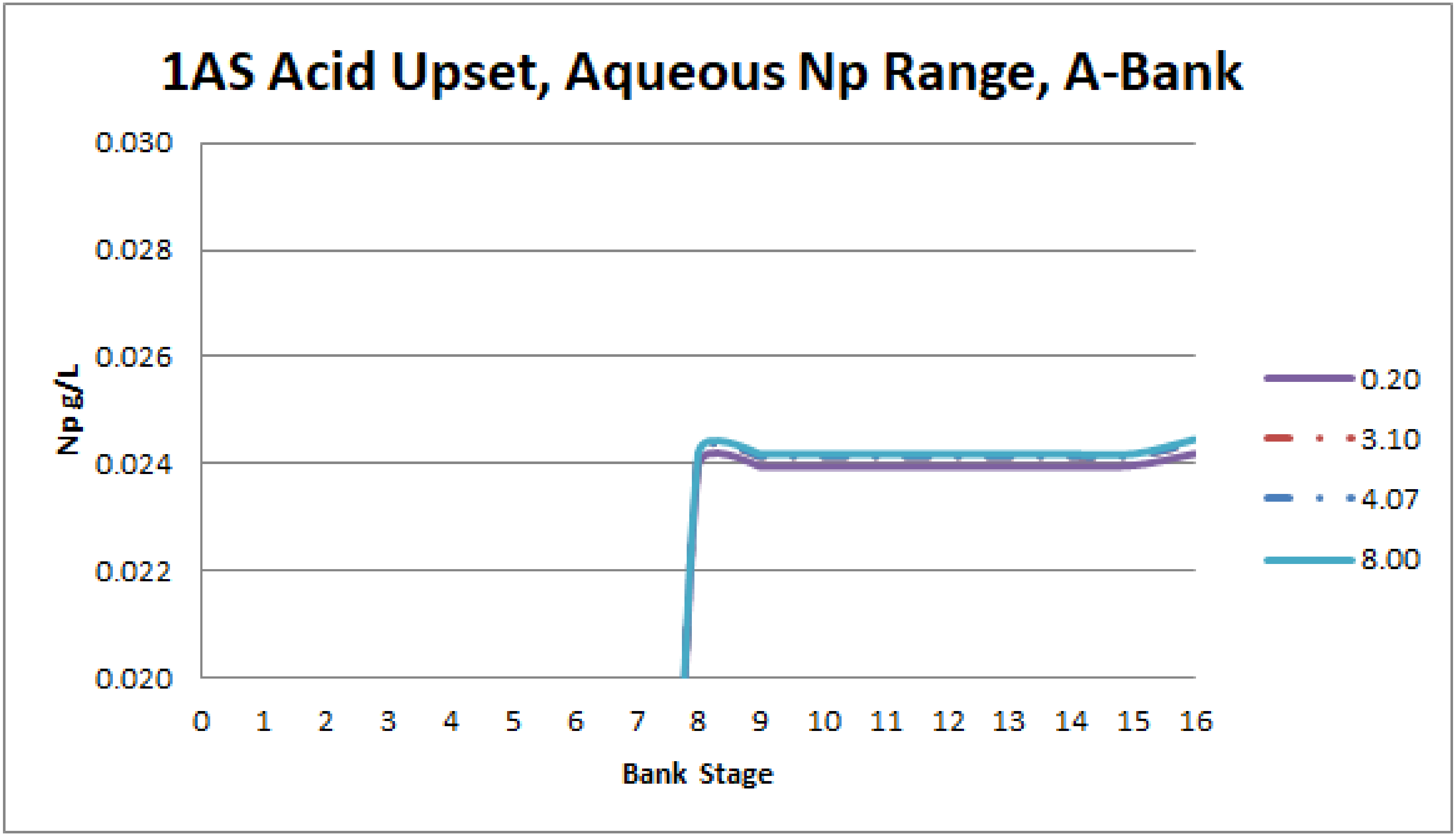
Aqueous Uranium Behavior During A Acid Upset, A-Bank



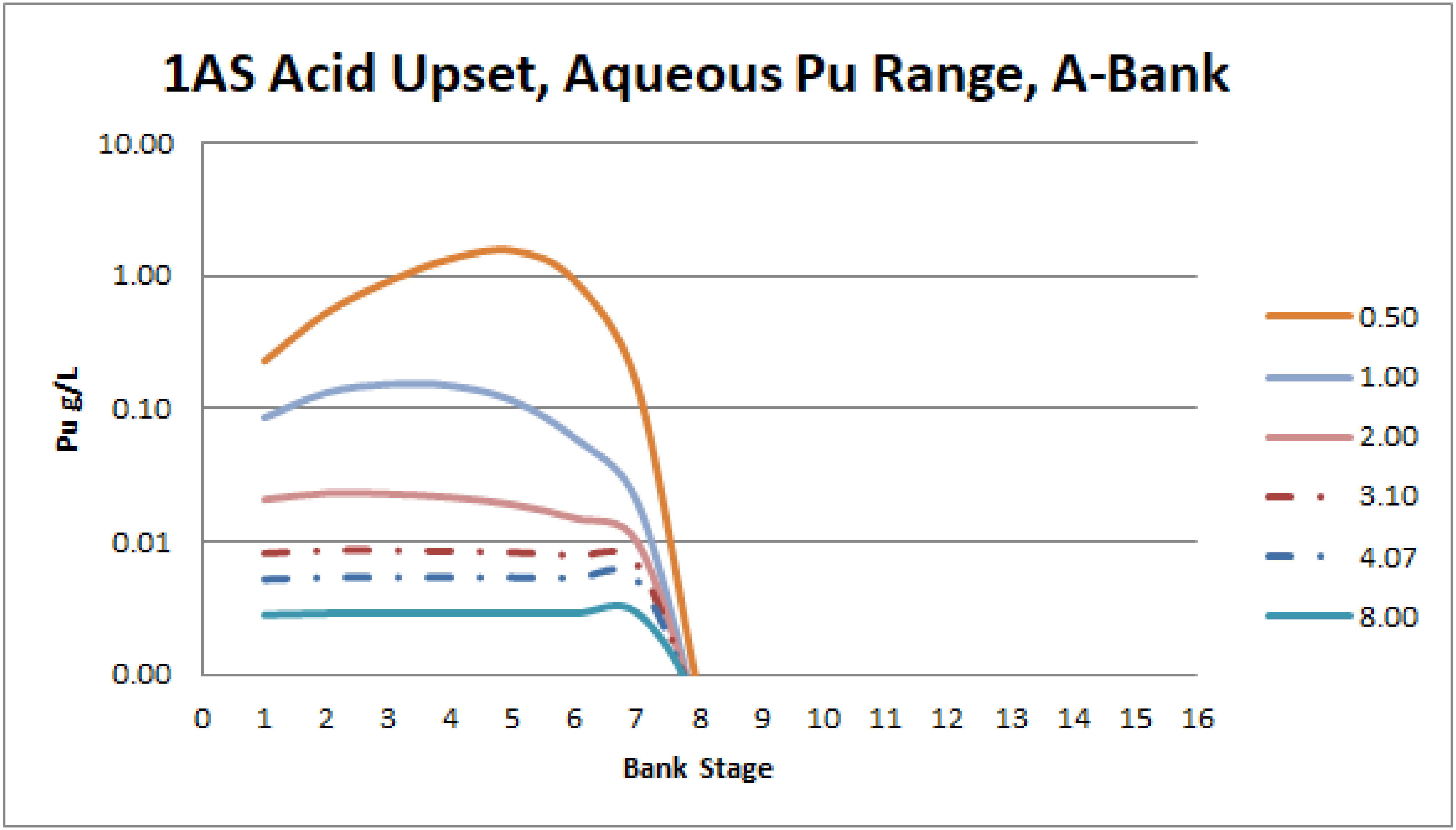
Aqueous Uranium Behavior During A Acid Upset, B-Bank



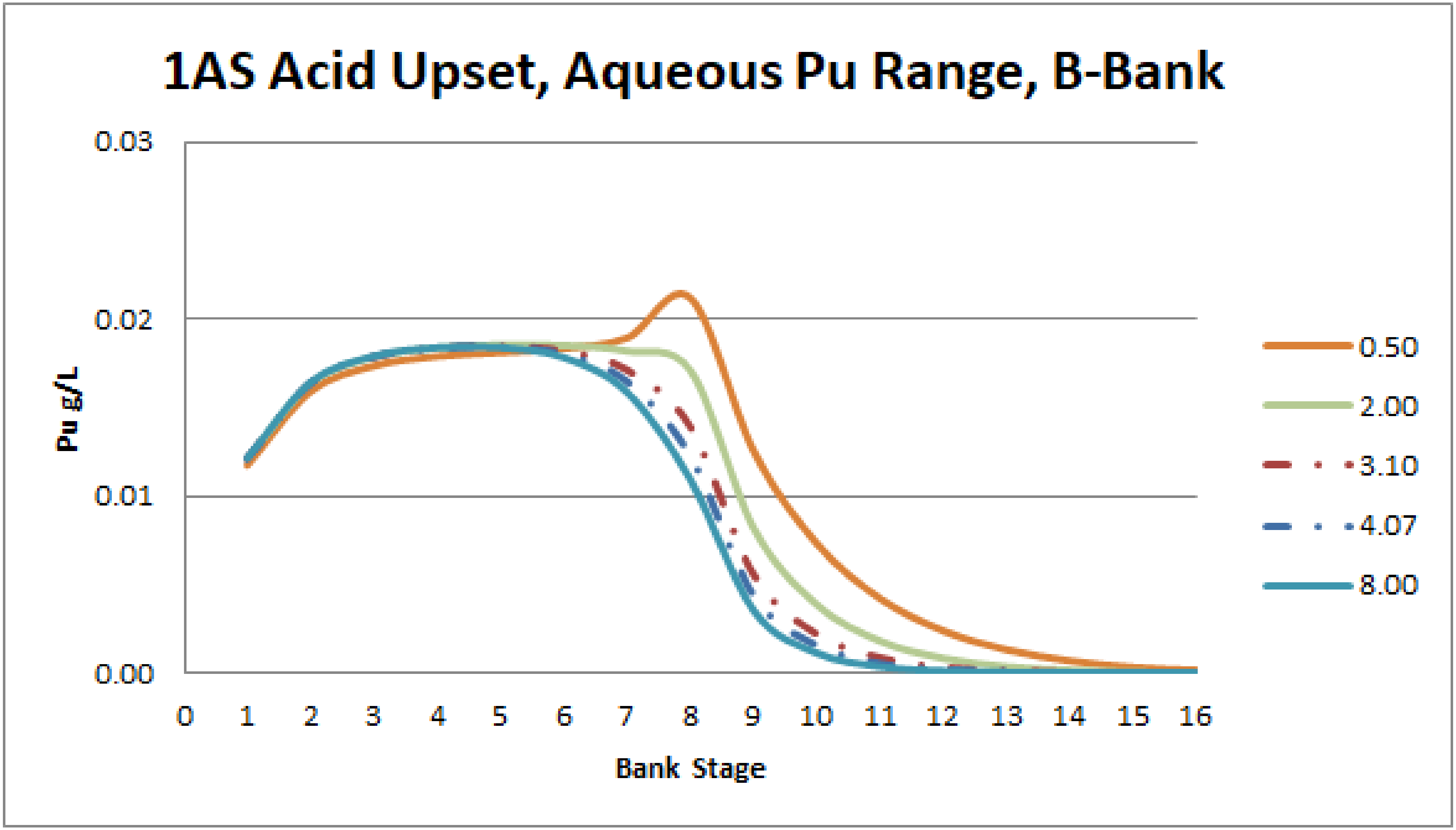
Aqueous Neptunium Behavior During A Acid Upset, A-Bank



Aqueous Plutonium Behavior During A Acid Upset, A-Bank

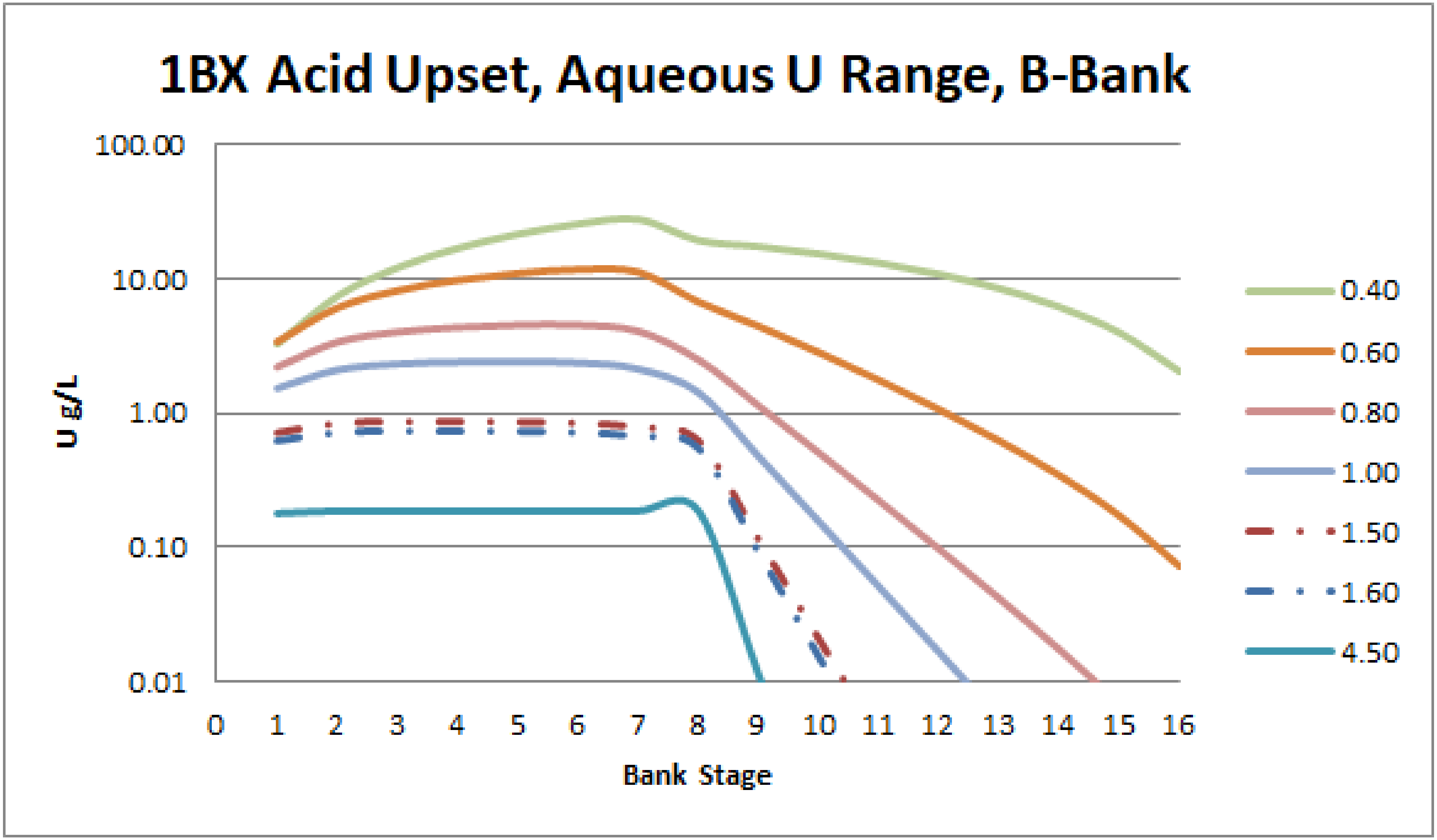


Aqueous Plutonium Behavior During A Acid Upset, B-Bank



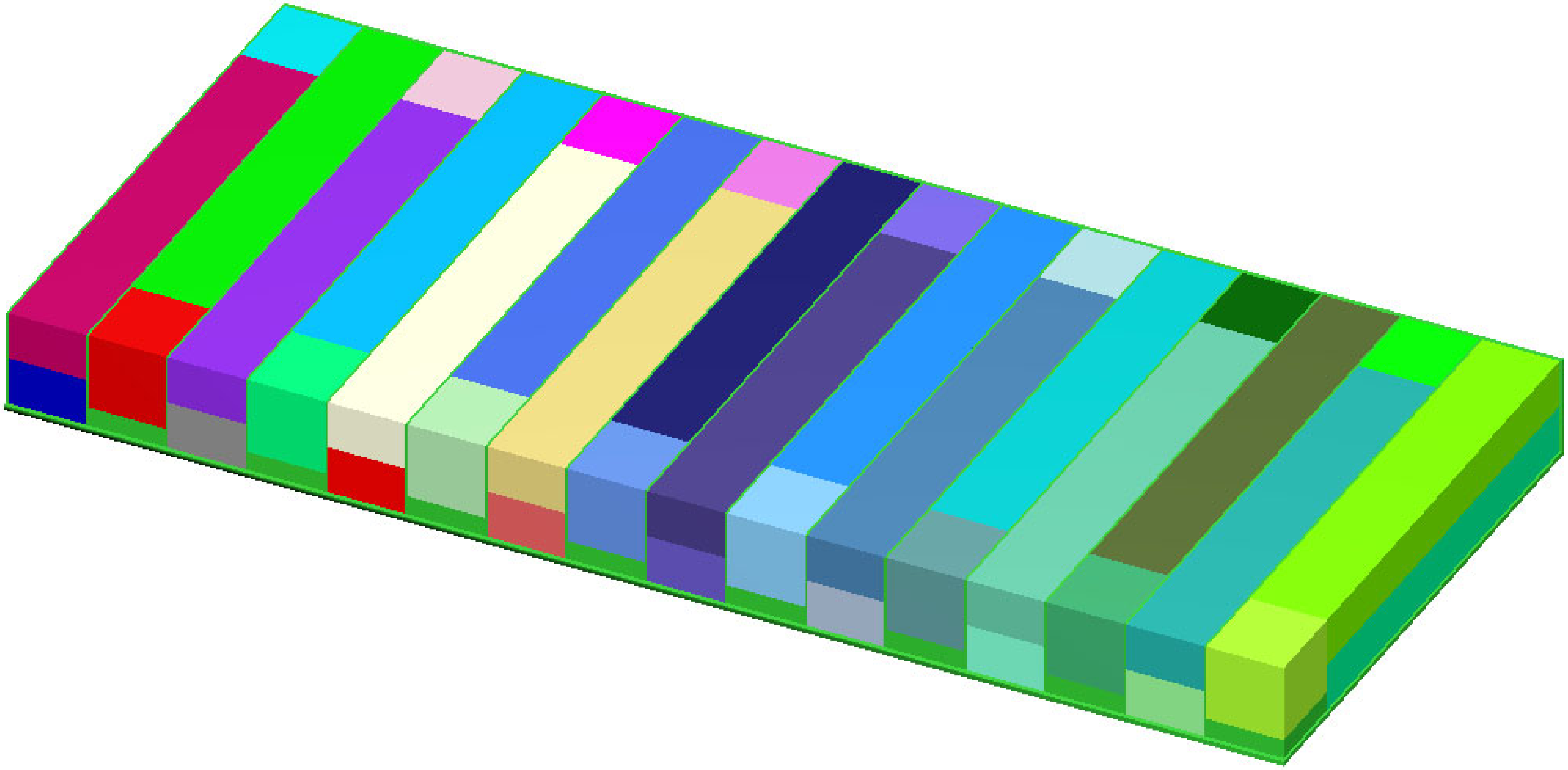
B-Bank Acid Upset

Aqueous Uranium Behavior During B Acid Upset, B-Bank



SCALE-Keno VI Simulation

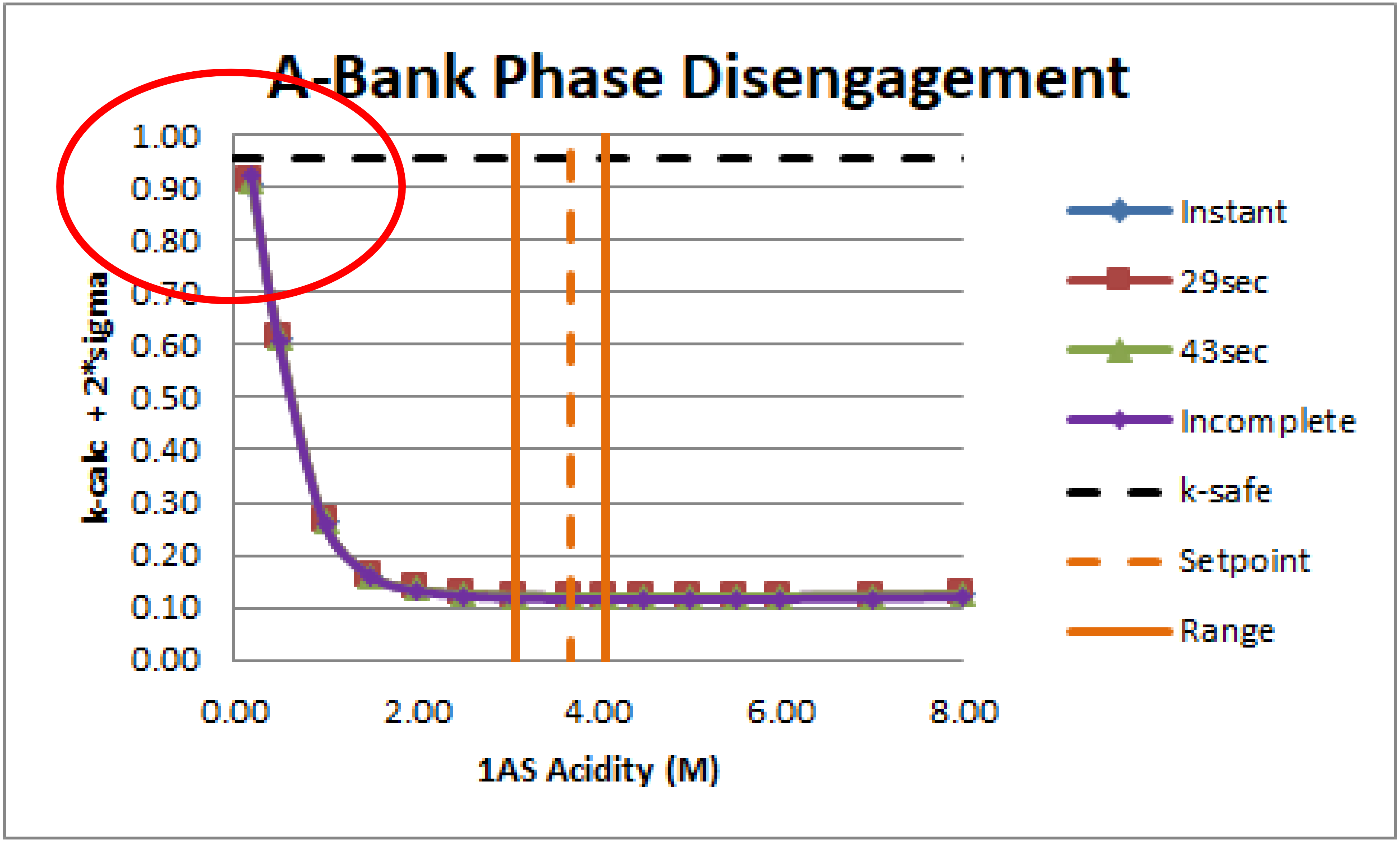
- **Neutron multiplication simulation with ENDF-VII 238-group cross sections**
- **All chemical species tracked in SEPHIS-ACM are modeled in SCALE**
  - – Broken down to isotopics and three compositions per stage
  - – Stainless steel plates explicitly modeled
  - – Baffles, flow weirs, impeller, and flow inlets neglected
  - – Bank surrounded by an air pocket and then given a reflective boundary condition beyond



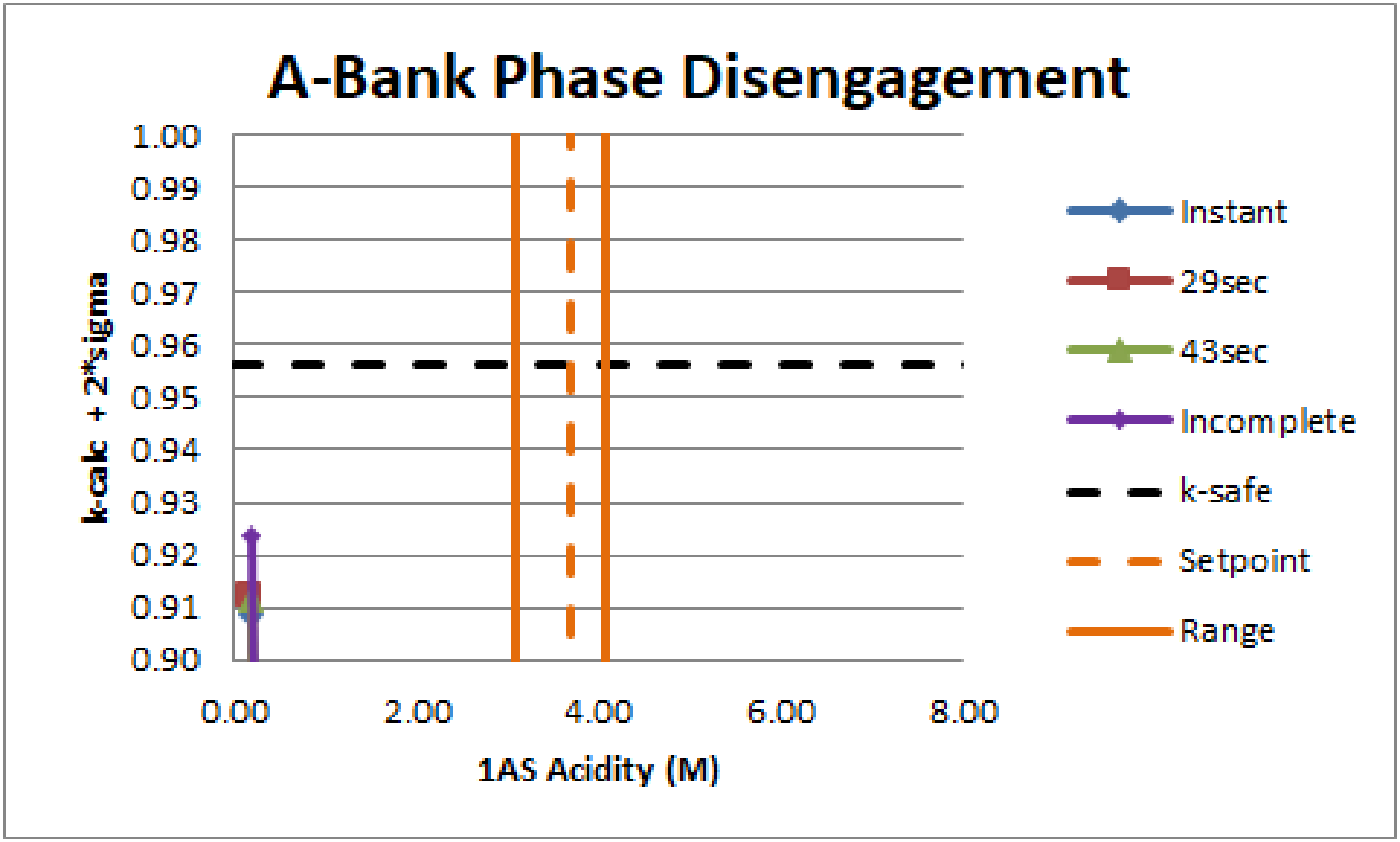
Determination of k-safe

An in-house validation for the SCALE 6.1 KENO-VI code for HEU solution systems was performed. A conservative k_{SAFE} assumed for this work was 0.9564.

Bank Multiplication Factor During A Acid Upset, A-Bank



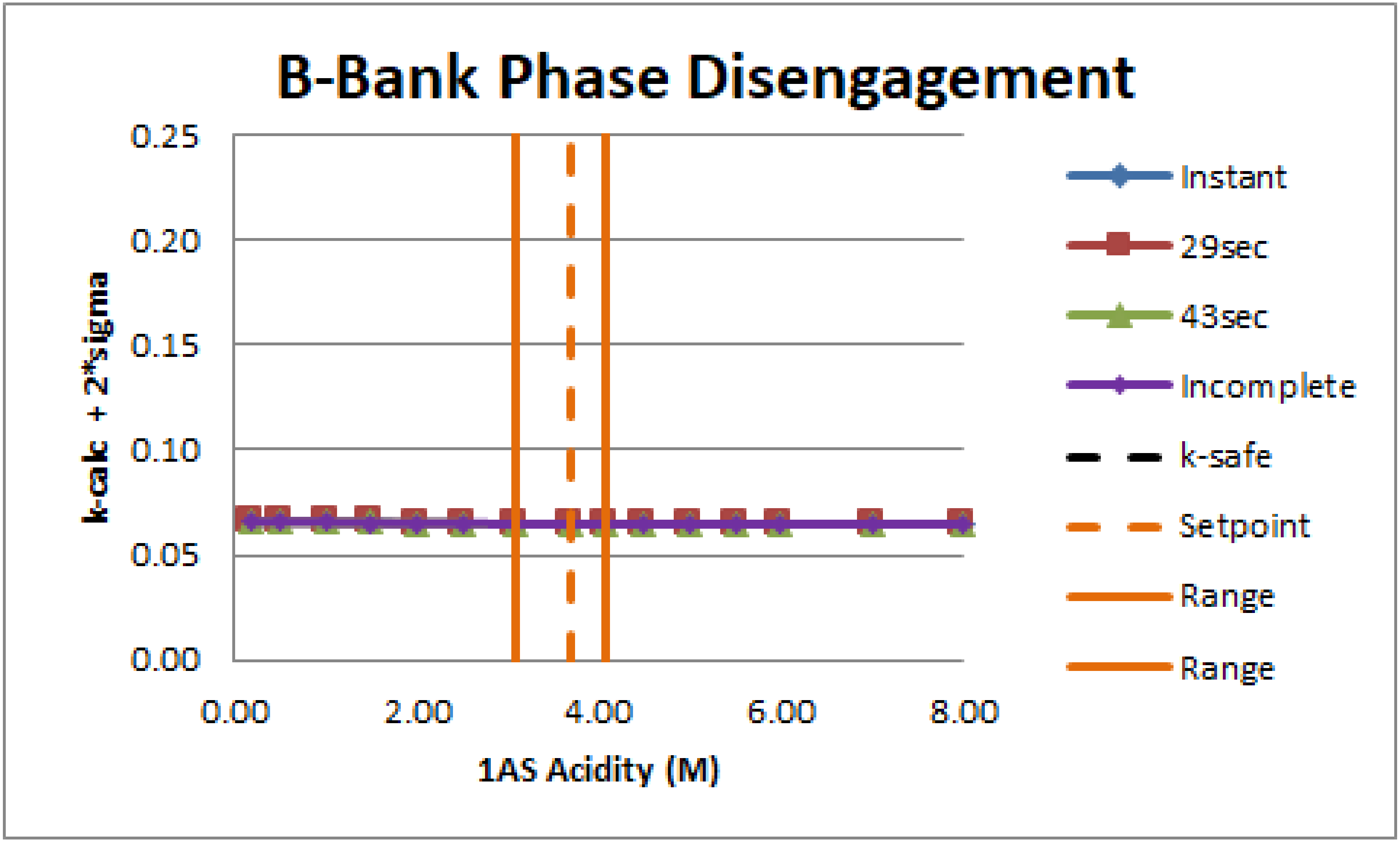
Bank Multiplication Factor During A Acid Upset, A-Bank



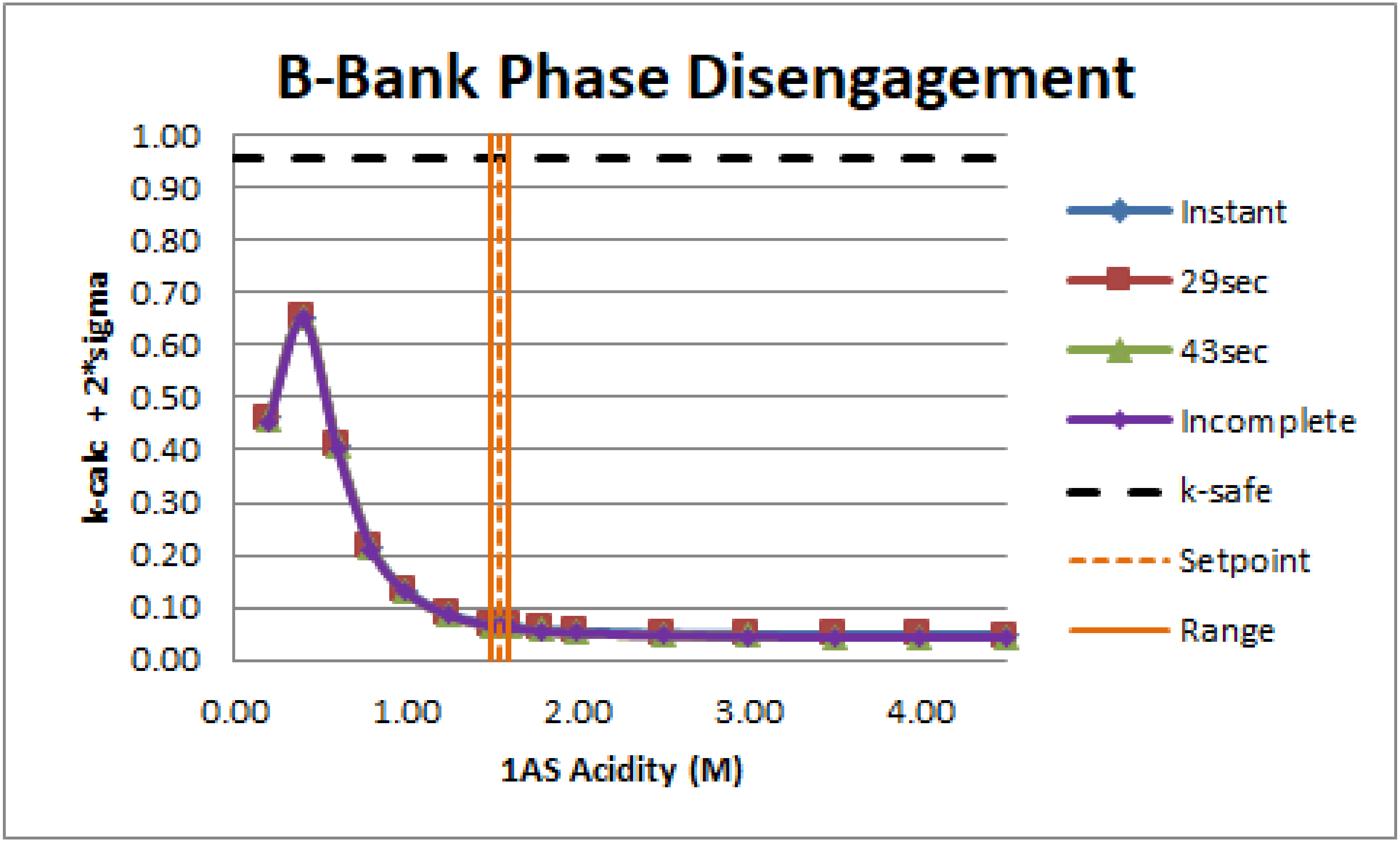
Bank Multiplication Factor During A Acid Upset, A-Bank

- **One exception was extreme low acid introduced to A-Bank**
  - Generally, solutions are highly over moderated.
  - U concentration in the aqueous phase is such that in several stages the aqueous composition is closer to optimal moderation
  - Multiplication jumps from ~0.6 to ~0.9 between 0.5M and 0.2M acid cases
  - Organic phase is still well over moderated
  - **When mixed with the aqueous phase, the interspersed phases are also nearer to optimal moderation than in other higher acid cases.**
  - The result is **unique** to this case and the condition is not credible for the A-Bank in the HM-Process.

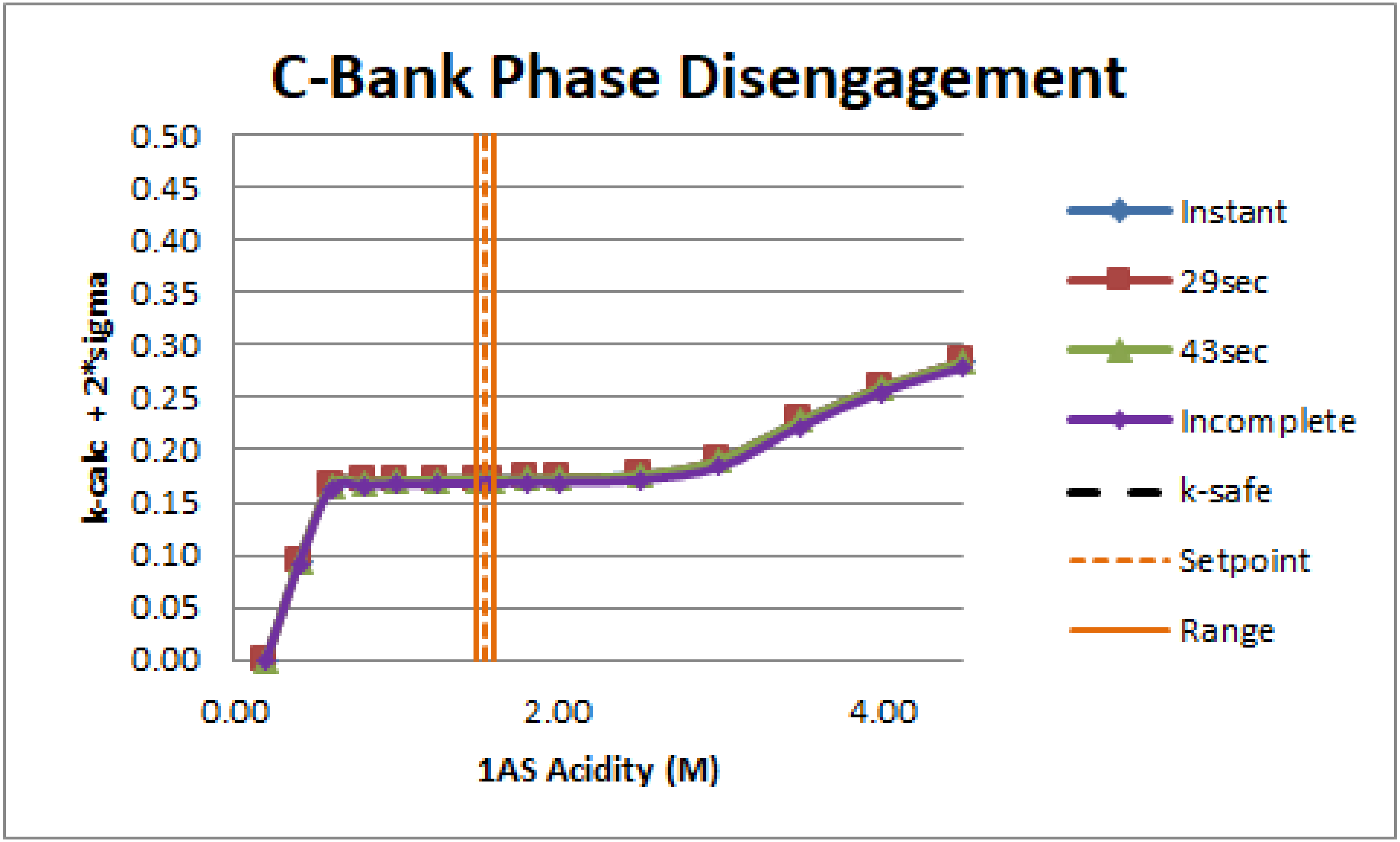
Bank Multiplication Factor During A Acid Upset, B-Bank



Bank Multiplication Factor During B Acid Upset, B-Bank



Bank Multiplication Factor During B Acid Upset, C-Bank



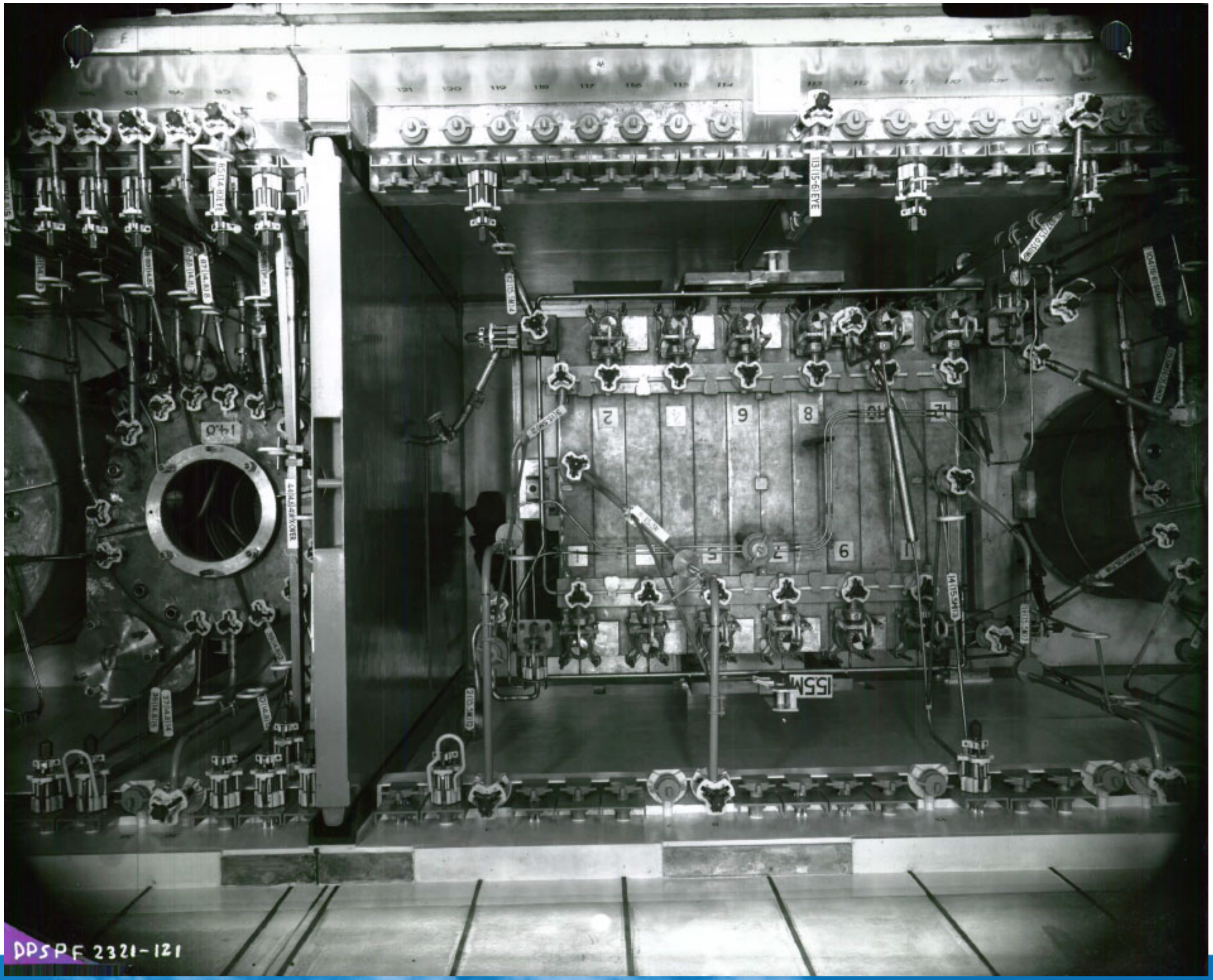
Discussion & Conclusions

- **Acid upsets simulated far beyond credible abnormal conditions**
  - Even then always met k-safe
- **For practical considerations, phase disengagement had no impact on criticality calculations**
  - Delayed disengagement slightly reduces multiplication
- **Conservative to assume instantaneous disengagement**
- **One exception was extreme low acid introduced to A-Bank**

Future Work

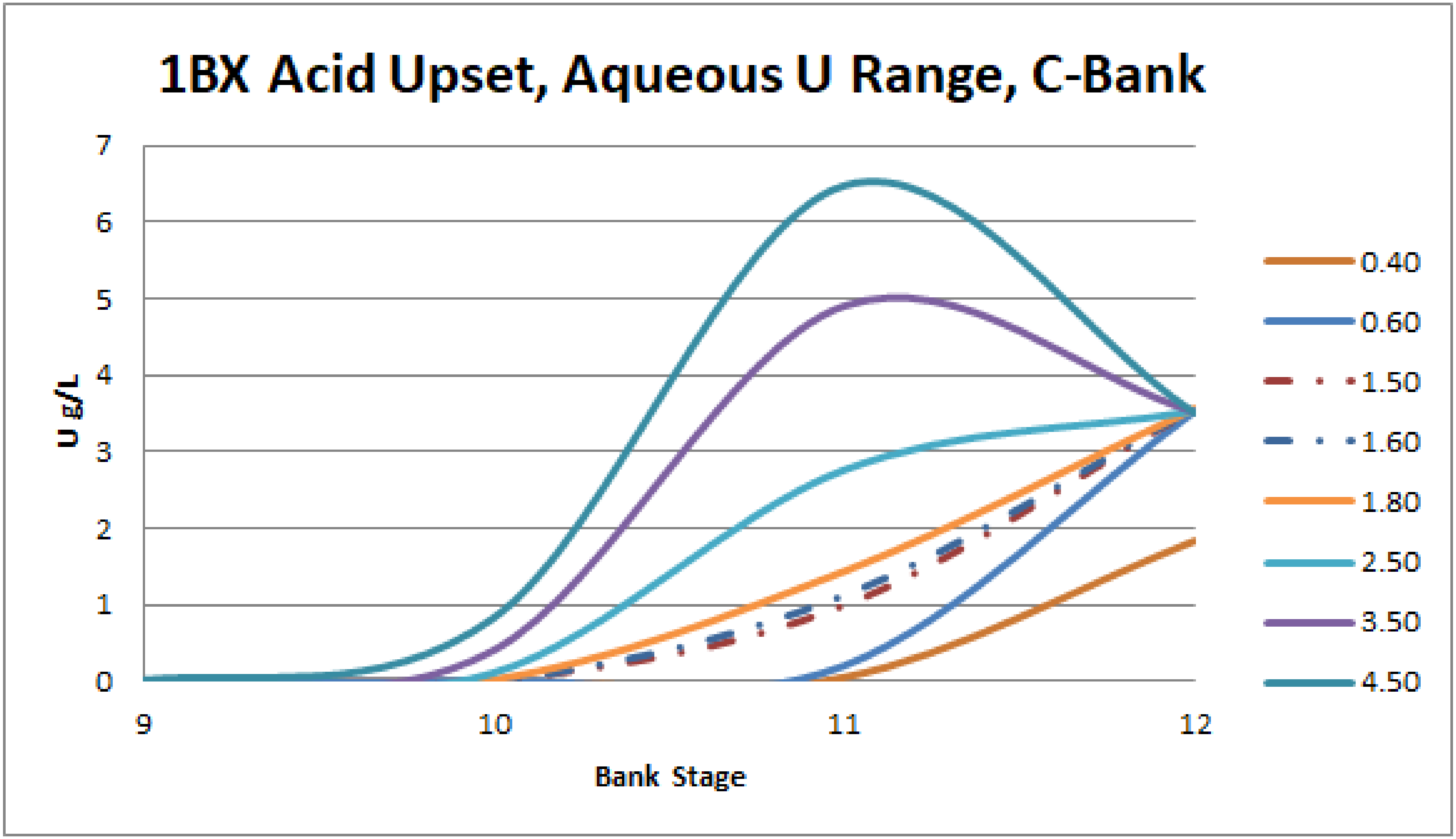
- Use SEPHIS-ACM to examine various first and second cycle upsets that are still identified as credible scenarios
- Assume instantaneous phase disengagement

Questions?

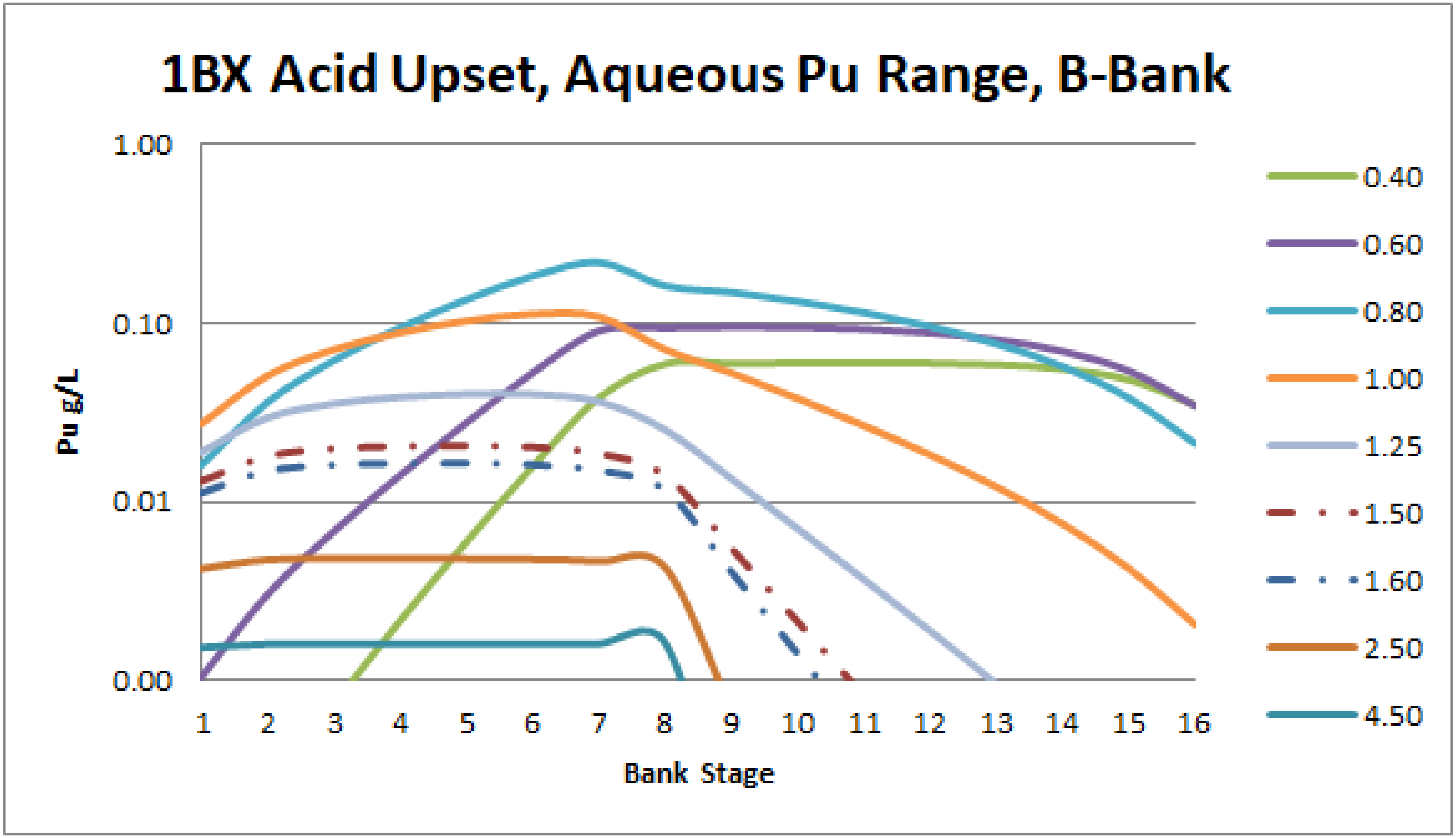


Additional Slides

Aqueous Uranium Behavior During B Acid Upset, C-Bank



Aqueous Plutonium Behavior During B Acid Upset, B-Bank



Aqueous Plutonium Behavior During B Acid Upset, C-Bank