

Design and Demonstration of the UREX+ Process Using Spent Nuclear Fuel

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A U.S. Department of Energy
Office of Science Laboratory
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The UREX+ Process

- **Process being developed as part of the Advanced Fuel Cycle Initiative (AFCI)**
- **Objective of ANL UREX+ demonstration:**
 - To demonstrate that all desired spent-fuel constituents can be separated by aqueous processing and meet required specifications for recycle or disposal
- **UREX+ work centered on**
 - Process design
 - Modeling using Argonne Model for Universal Solvent Extraction (AMUSE)
 - Demonstration



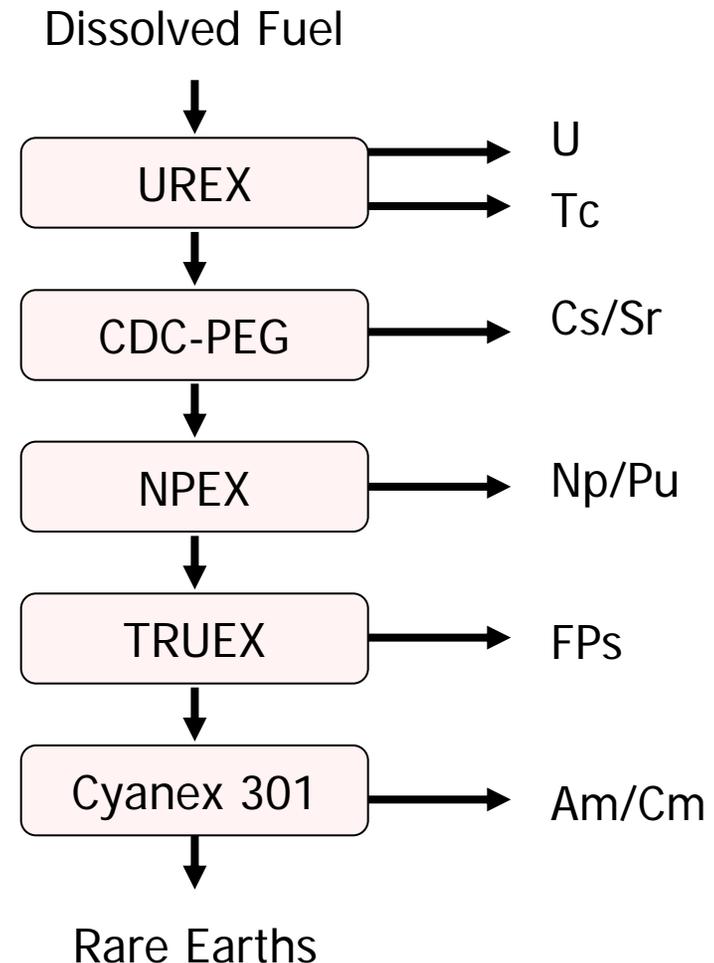
UREX+ Spent Fuel Demonstration

- **Spent fuel dissolution using nitric acid**
- **A series of five solvent-extraction process segments that separate spent fuel into seven product and waste streams**
 - *I for safe disposal*
 - *U₃O₈ for recycle or disposal as LLW*
 - *Np/Pu for mixed oxide fuel for thermal reactors*
 - *Tc for safe disposal*
 - *Am/Cm for fast-reactor fuel*
 - *Cs/Sr for decay storage*
 - *Mixed fission products for repository disposal*



Aqueous Based Separations

- **Process run twice**
 - Simulant based on ORIGEN2 calculations
 - Spent fuel dissolved in HNO₃
- **Flowsheets**
 - Developed using AMUSE
 - Equipment
 - *Three ANL-design 2-cm centrifugal contactors*
 - *Most operations in hot-cell contactor*



UREX+ Processing Goals

- **Uranium**
 - Recovery $\geq 90\%$.
 - Purity to allow disposal as low-level waste
 - *Fission products to meet 10CFR61.55 limits and TRU $< 100\text{nCi/g}$*
 - *If U destined for recycle, purity requirements are more severe and would be governed by ASTM C 788-98*
- **Technetium**
 - Recovery $\geq 95\%$
 - Purity only important for use as transmutation target
- **Cesium/Strontium**
 - Recovery $\geq 97\%$
 - *Contribution to the heat load in the repository equal to that of all other fission products*
 - Purity to allow ultimate disposal as low-level waste after decay storage



UREX+ Processing Goals

- **Plutonium/Neptunium**
 - Recovery $\geq 99\%$ to allow 100-fold reduction of heat load to repository
 - Purity to meet mixed-oxide (MOX) fuel specifications in ASTM C833-01
- **Americium/Curium**
 - Recovery $\geq 99.5\%$ to allow 100-fold reduction of heat load to the repository
 - Purity to meet fast-spectrum reactor lanthanide specification of $<20\text{mg/g}$ Uranium plus TRU
- **Raffinates from the UREX+ process**
 - TRUEX, all soluble fission products but Cs, Sr, Tc, I, and REs
 - Cyanex 301, RE elements
 - Purity requirements are basis for recoveries in other streams



UREX+ Process Equipment

2-cm Centrifugal Contactor before Hot-Cell Placement

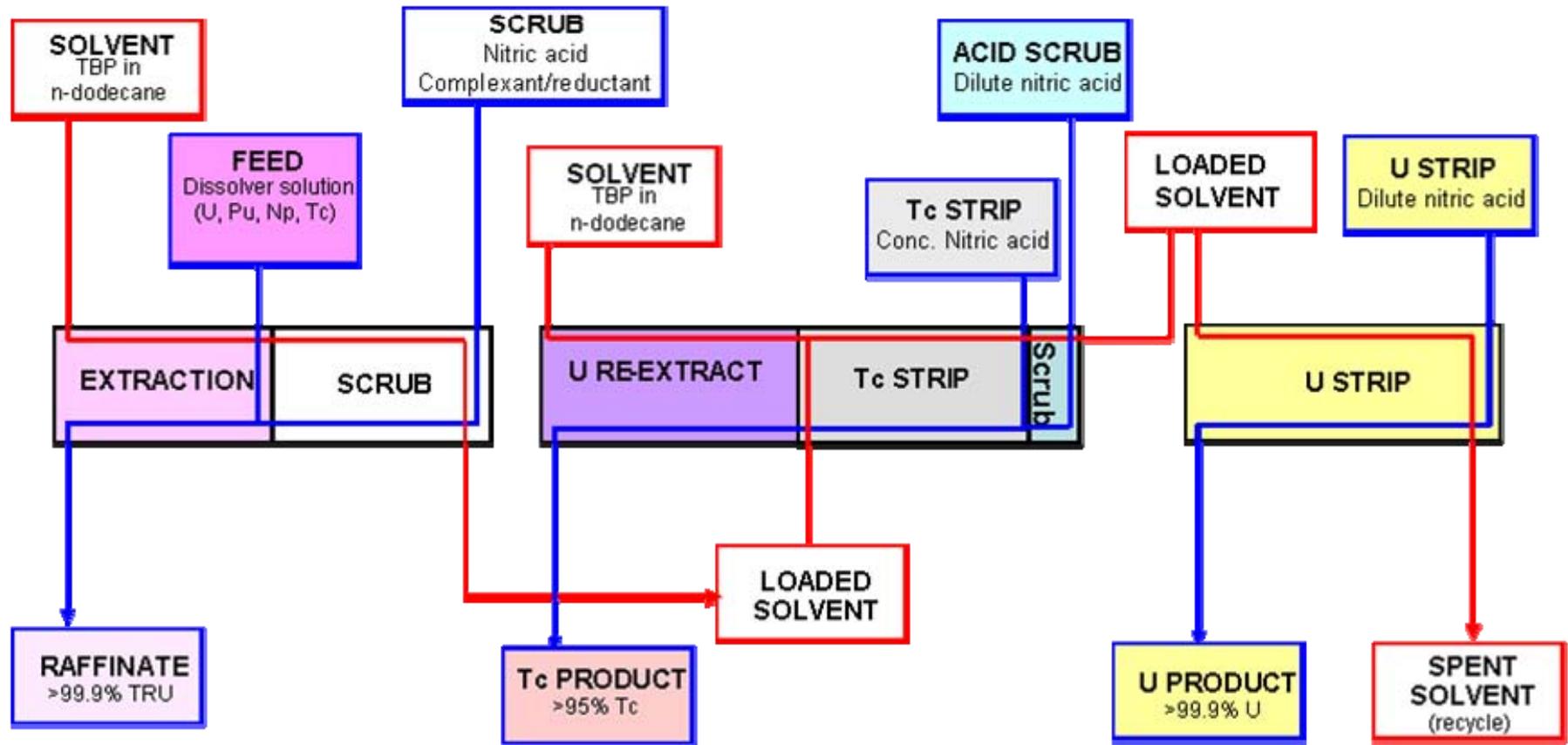


Operational Limitations

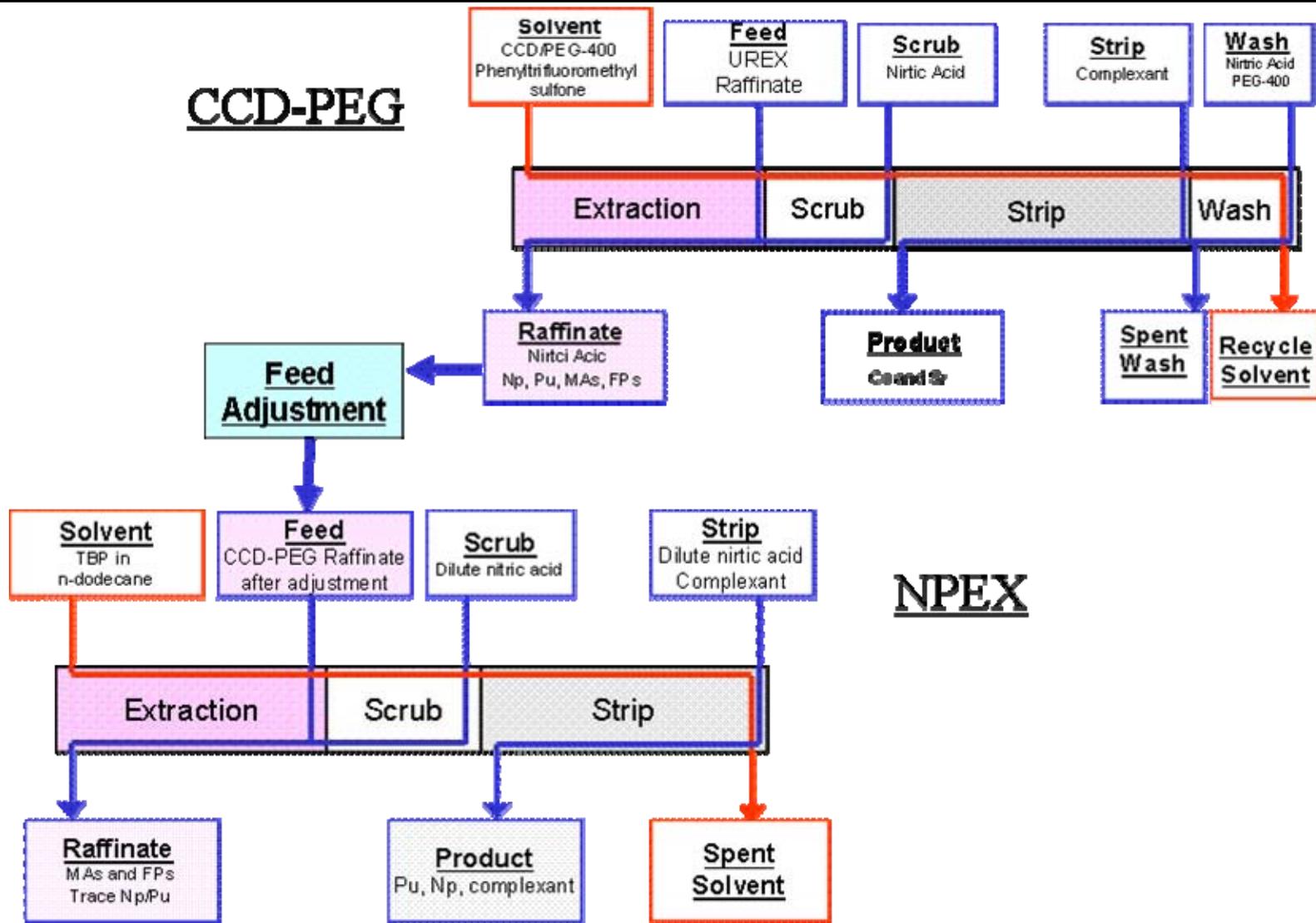
- **Equipment**
 - Three multistage contactors
 - *Shielded cell – all operations but U- and Tc-stripping*
 - *Glovebox – Tc-strip*
 - *Vacuum-frame hood – U-strip*
 - Sequential use of the hot-cell contactor for all process segments
 - *Extensive decontamination and relocation of feed and effluent lines was required between process segments*
 - *CCD-PEG solvent is denser than water*
 - Reversal of organic and aqueous flow directions
- **Reagents**
 - Cyanex 301 is commercially available but requires extensive purification for actinide /lanthanide separations
 - CCD-PEG solvent supplied by INEEL



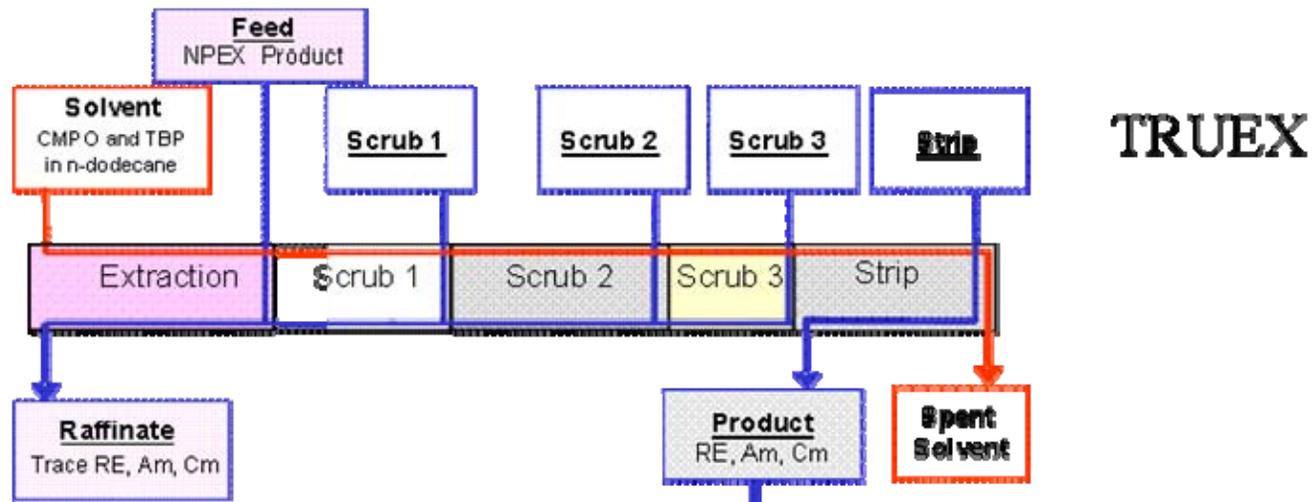
Solvent Extraction Processes - UREX



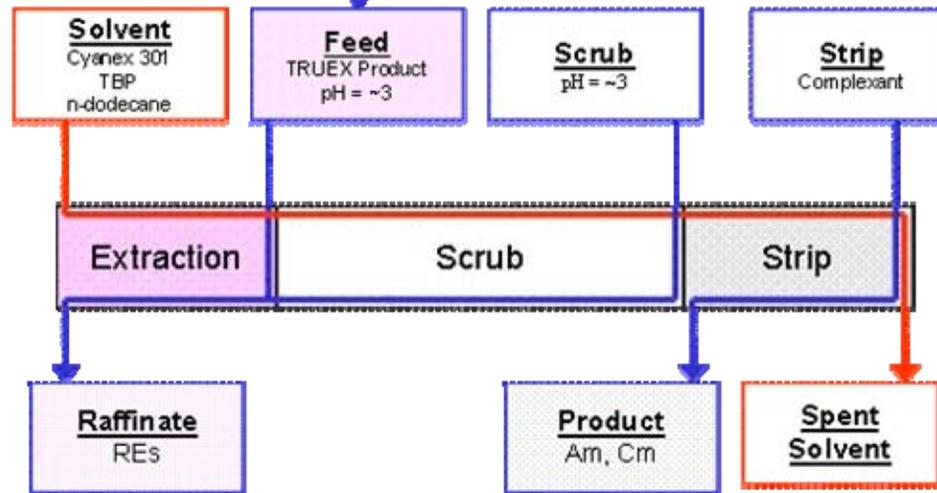
UREX+ Solvent Extraction Processes



UREX+ Solvent Extraction Processes



Cyanex 301

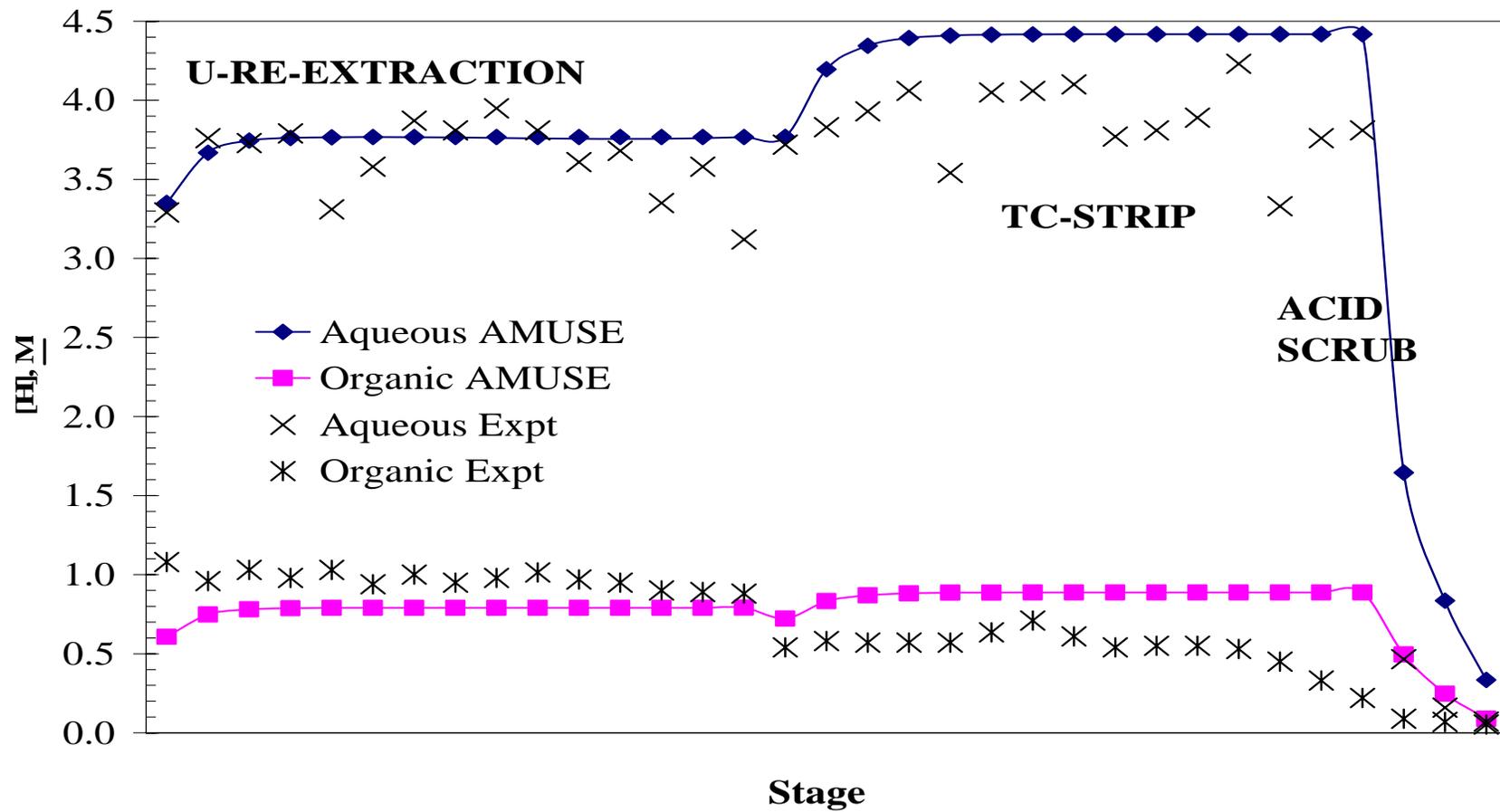


Improved Predictions of Hydrogen Partitioning at high Solvent Loadings

- **Why required?**
 - Evaluation of the nitric-acid stage profiles from the FY 03 UREX+ Demonstration showed that AMUSE did not properly fit the analytical results when solvent was highly loaded
 - AMUSE predictions did not agree with SEPHIS and PAREX predictions at high solvent loadings
 - UREX process flowsheets are designed to keep uranium loading of the solvent in the scrub section between 70-72%
- **Solution**
 - Measure hydrogen- and uranyl-ion distribution ratios under conditions of high solvent loading
 - Develop a hydrogen-ion correlation to put into the Loading Module in AMUSE

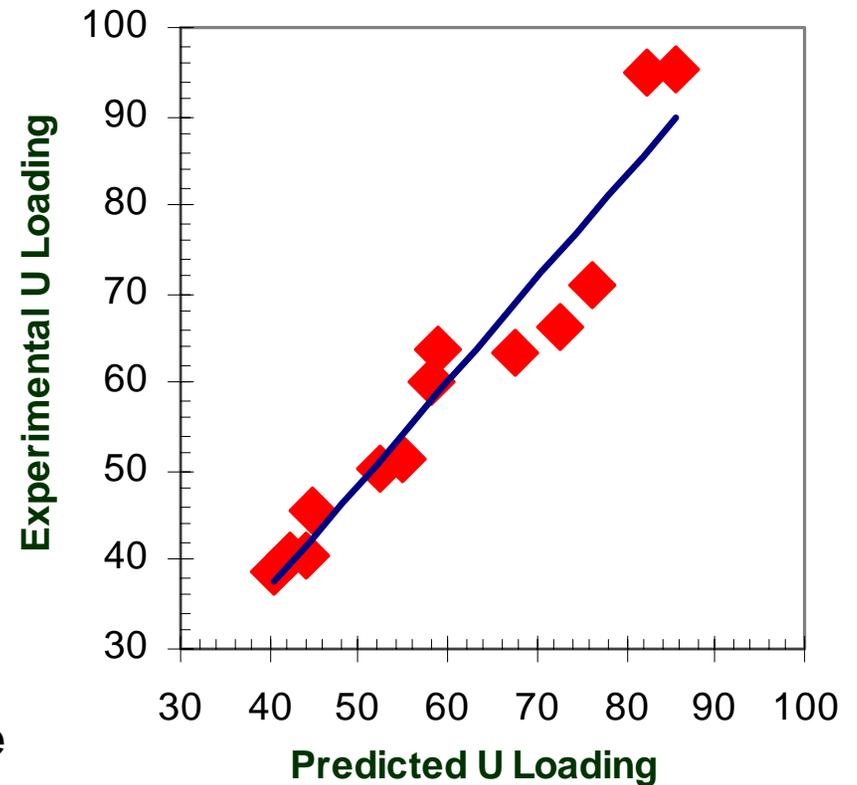
Results

- Nitric acid data compared with old AMUSE code predictions



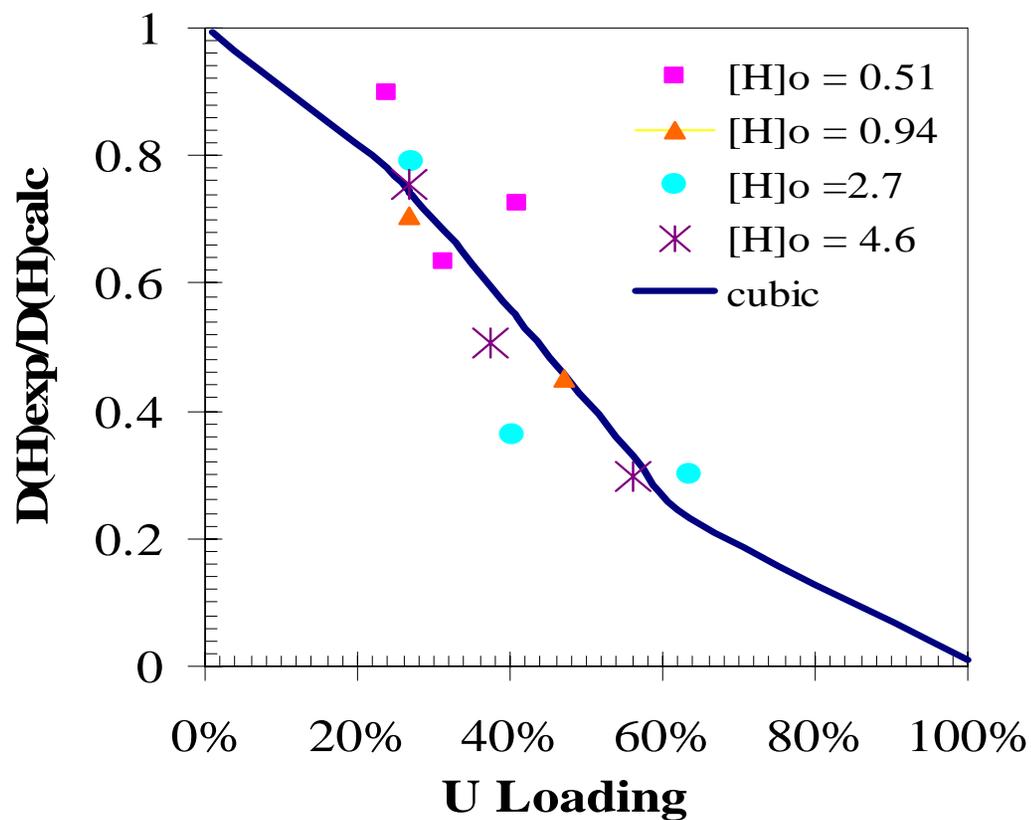
Experimental vs. Predicted U loading

- **U(VI) partitioning was measured in the laboratory at conditions that would load the solvent**
 - Initial aqueous composition =
 - 150 g/L U
 - 0.5 – 5M HNO₃
 - Organic = 30% TBP in n-dodecane
 - O/A = 4/1 -2/1
- **U distribution ratios and loading were well predicted by AMUSE**
- **HNO₃ partitioning at high loadings were not.**



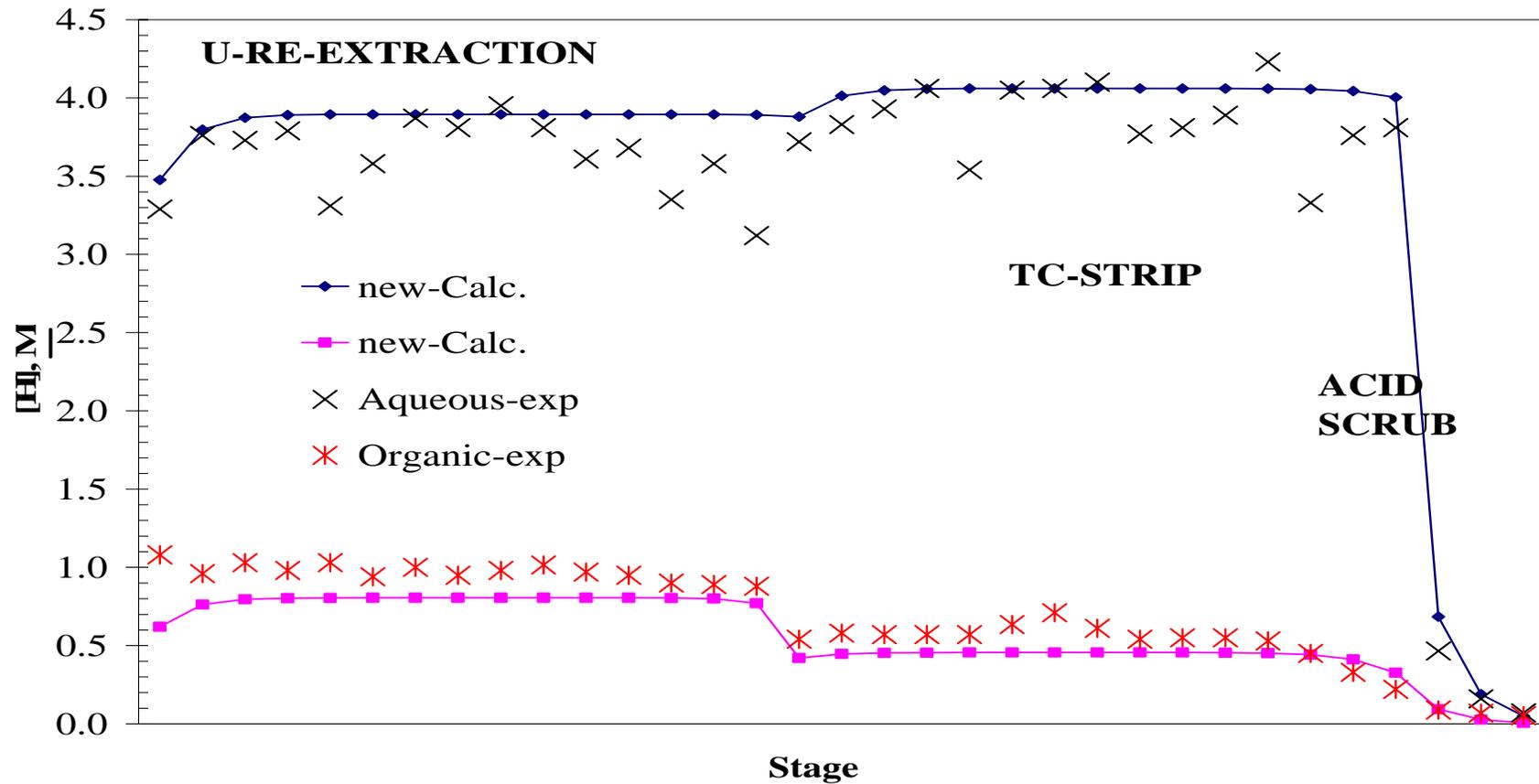
Results—U(VI)/H⁺ Studies

- Experimental vs. Calculated D_H vs. Uranium Loading



Results

- Nitric acid data compared with enhanced AMUSE code



UREX+ Results Summary

- **Most processing goals were met**
 - Those missed were due to operational glitches
- **Demonstrated the feasibility of meeting AFCI goals by aqueous solvent extraction**
 - However, a stable solvent system is still required for actinide/lanthanide separations
 - *Cyanex 301 will work at the laboratory scale, but its stability makes it an unlikely candidate for industrial processing*
- **Stage samples in Tc-strip sections showed that H⁺ partitioning was being over-predicted under conditions of high solvent loading**
 - Experimental data were collected to develop an algorithm for HNO₃ extraction for the AMUSE Loading Module



Next Steps

- **The demonstration being planned for FY-04 will have several improvements**
 - PC-based monitoring and control of pumps
 - *Load cells to measure feed and effluent flow rates*
 - *Three-way ball valves for sample collection and changing feed and effluent tanks during runs*
 - Hot cell will have
 - *Sample carousel*
 - *Sample aliquot and dilution station using automated pipettes*
 - *Automated titration station*
- **Alternate flowsheet for U, Tc, Pu/Np, and Cs/Sr recovery**
 - Co-extraction of U, Tc, Np, and Pu
 - *Separate strips for Np/Pu and U/Tc*
 - CDC/PEG extraction of Cs/Sr



Acknowledgements

- **Research funded by U.S. Department of Energy, Office of Nuclear Energy, Science and Technology**

