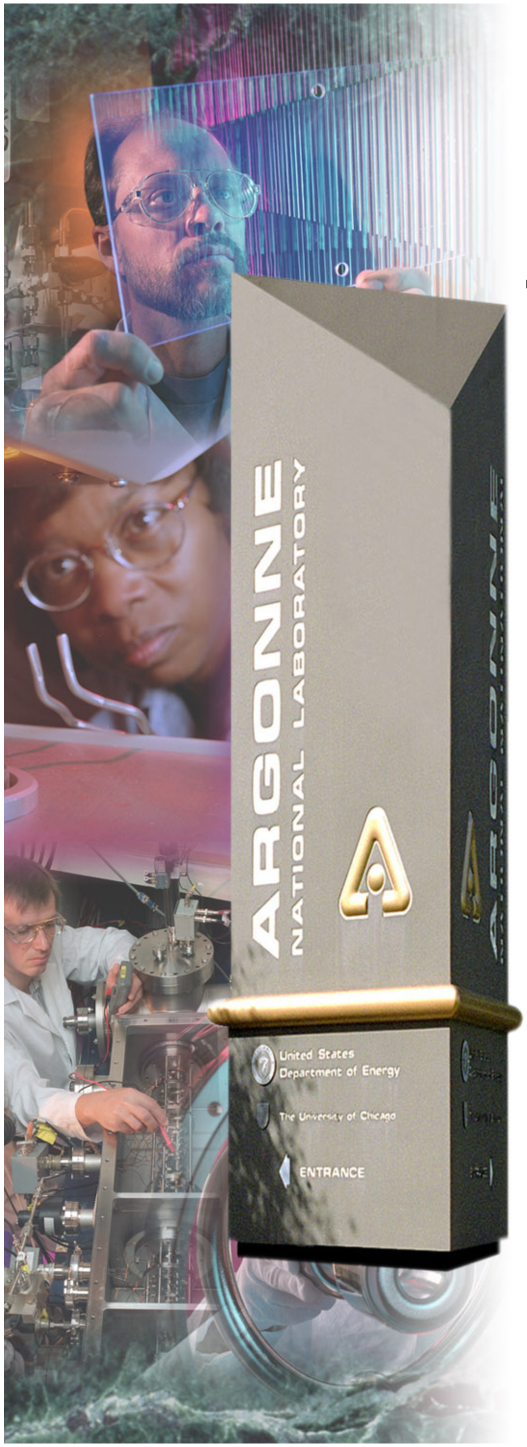

Design and Demonstration of the UREX+ Process Using Spent Nuclear Fuel

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The UREX+ Process

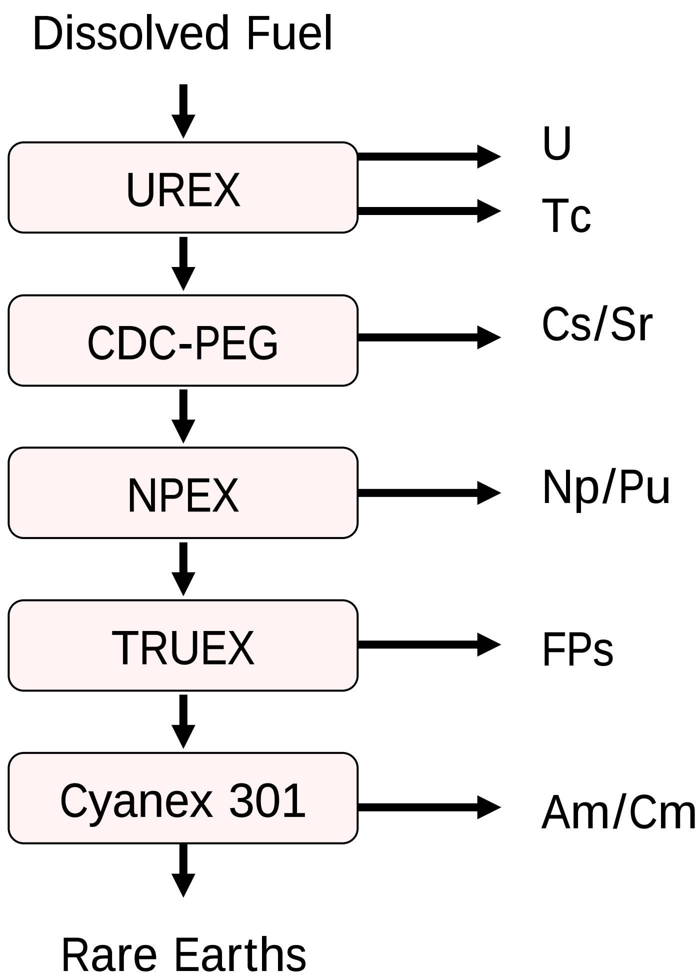
- **Process being developed as part of the Advanced Fuel Cycle Initiative (AFCI)**
- **Objective of ANL UREX+ demonstration:**
  - To demonstrate that all desired spent-fuel constituents can be separated by aqueous processing and meet required specifications for recycle or disposal
- **UREX+ work centered 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*
  - *U_3O_8 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 HNO_3
- **Flowsheets**
  - Developed using AMUSE
  - Equipment
    - *Three ANL-design 2-cm centrifugal contactors*
    - *Most operations in hot-cell contactor*



UREX+ Processing Goals

- **Uranium**
  - Recovery ≥90%.
  - Purity to allow disposal as low-level waste
    - *Fission products to meet 10CFR61.55 limits and TRU <100nCi/g*
    - *If U destined for recycle, purity requirements are more severe and would be governed by ASTM C 788-98*
- **Technetium**
  - Recovery ≥95%
  - Purity only important for use as transmutation target
- **Cesium/Strontium**
  - Recovery ≥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 ≥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 ≥ 99.5% to allow 100-fold reduction of heat load to the repository
  - Purity to meet fast-spectrum reactor lanthanide specification of <20mg/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

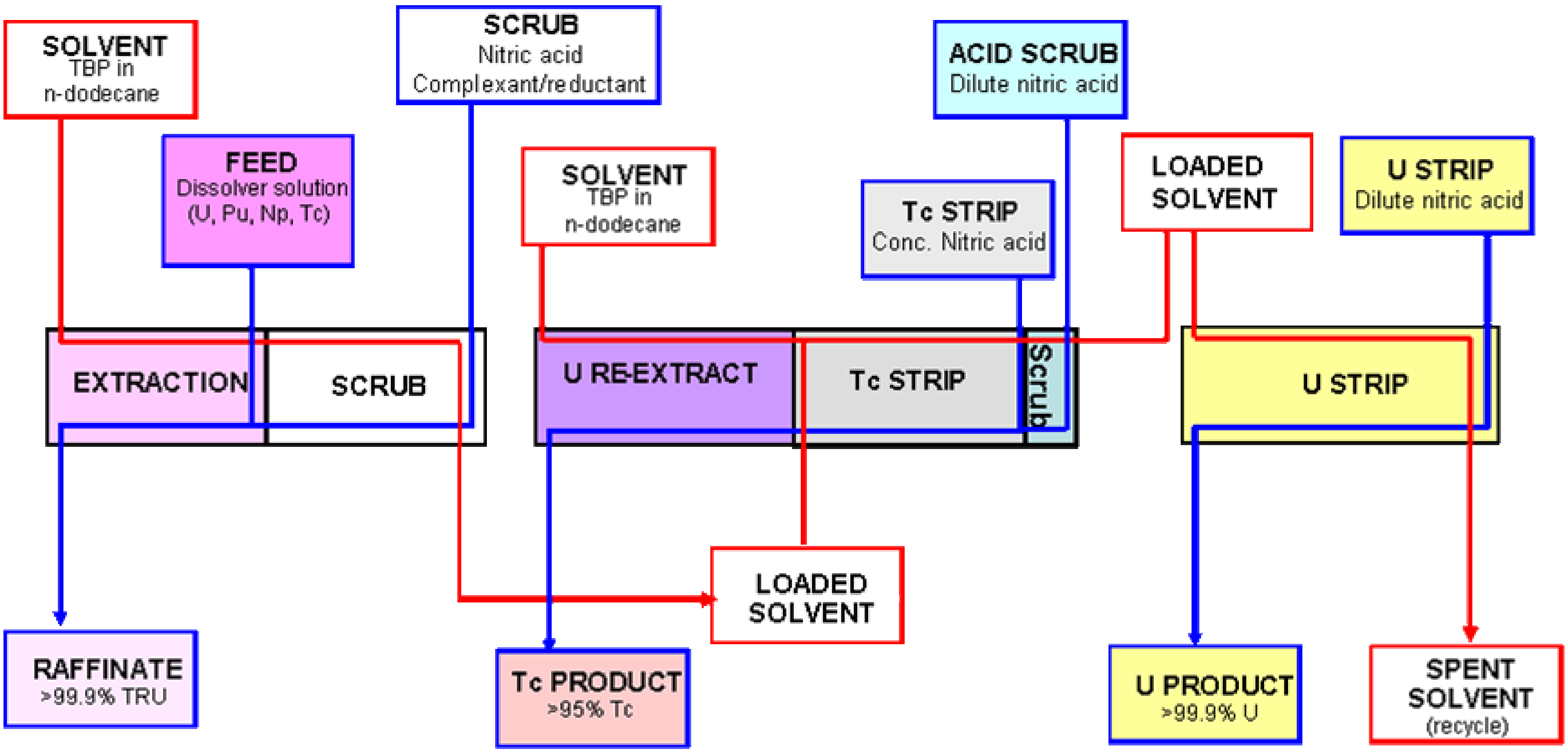
UREX+ Process Equipment

2-cm Centrifugal Contactor before Hot-Cell Placement

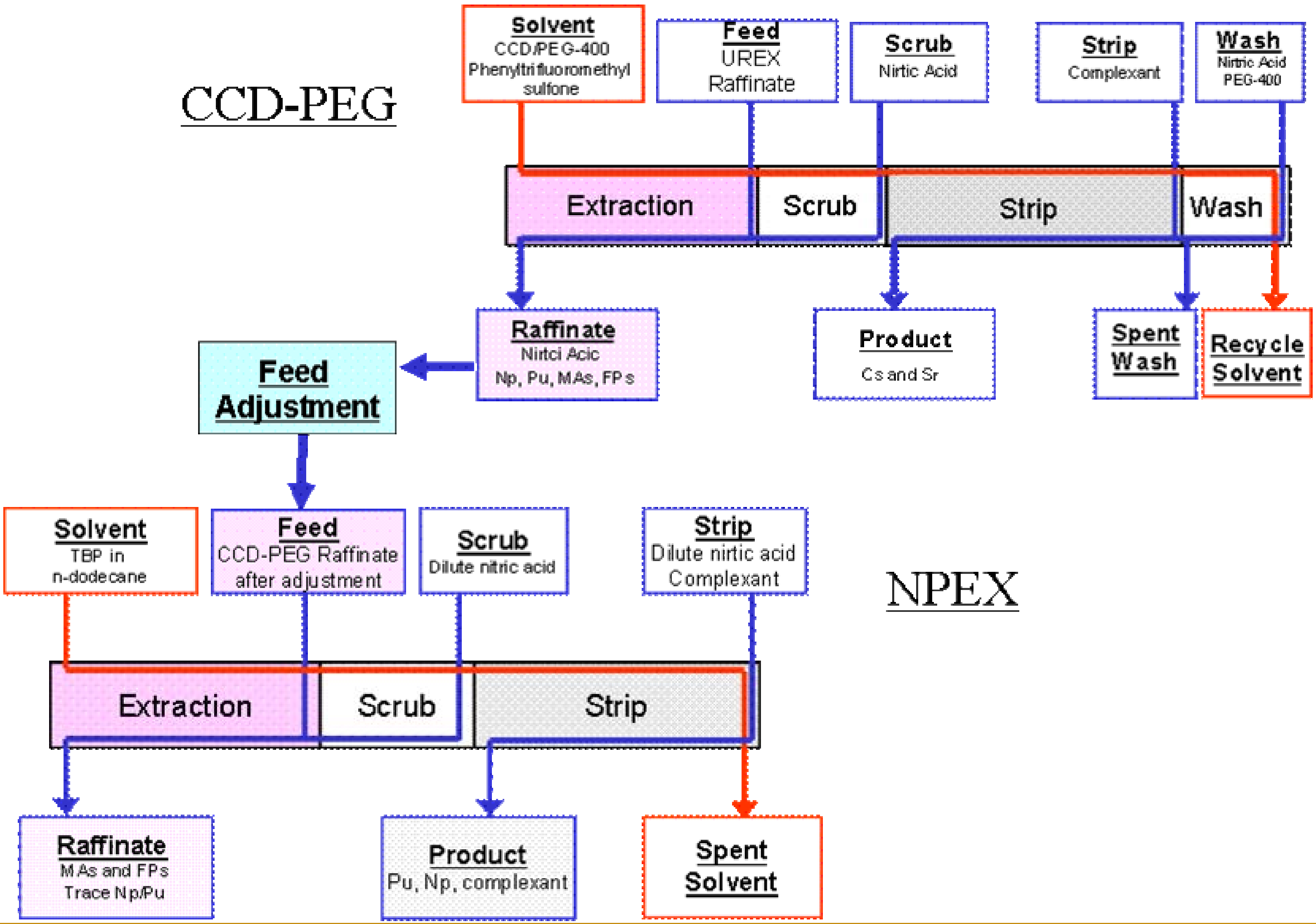
Operational Limitations

- **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

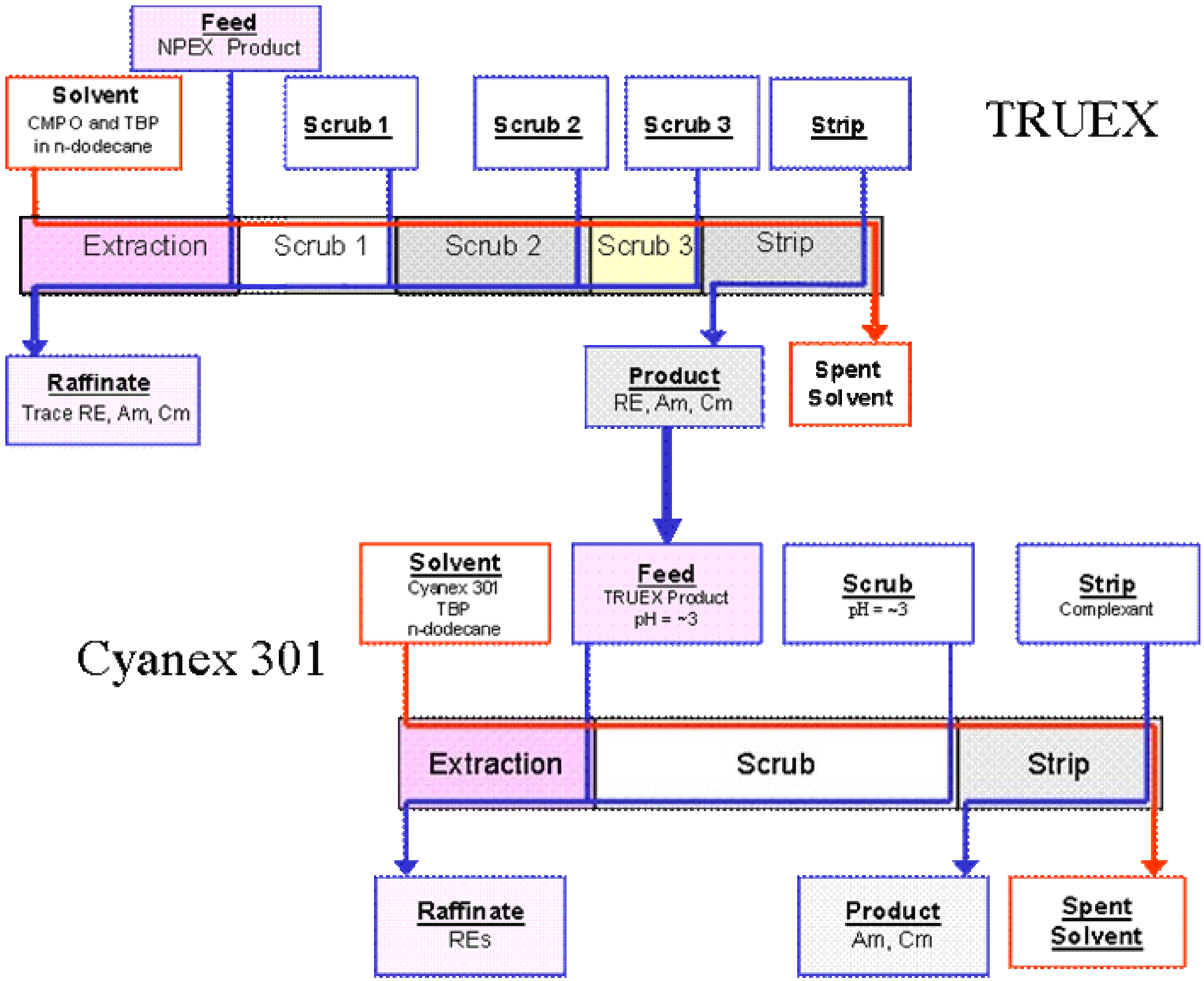
Solvent Extraction Processes - UREX



UREX+ Solvent Extraction Processes



UREX+ Solvent Extraction Processes

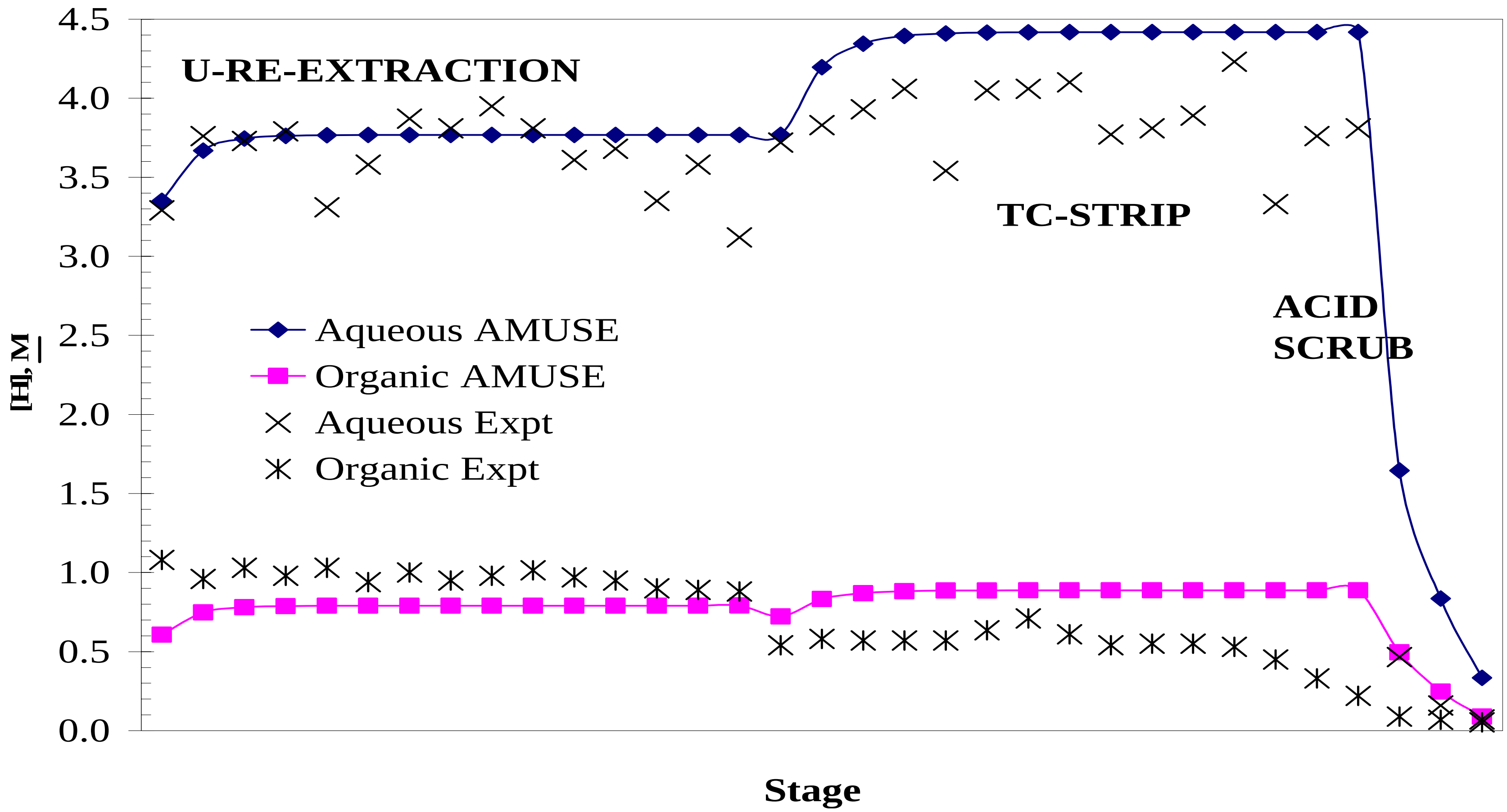


Improved Predictions of Hydrogen Partitioning at high Solvent Loadings

- **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

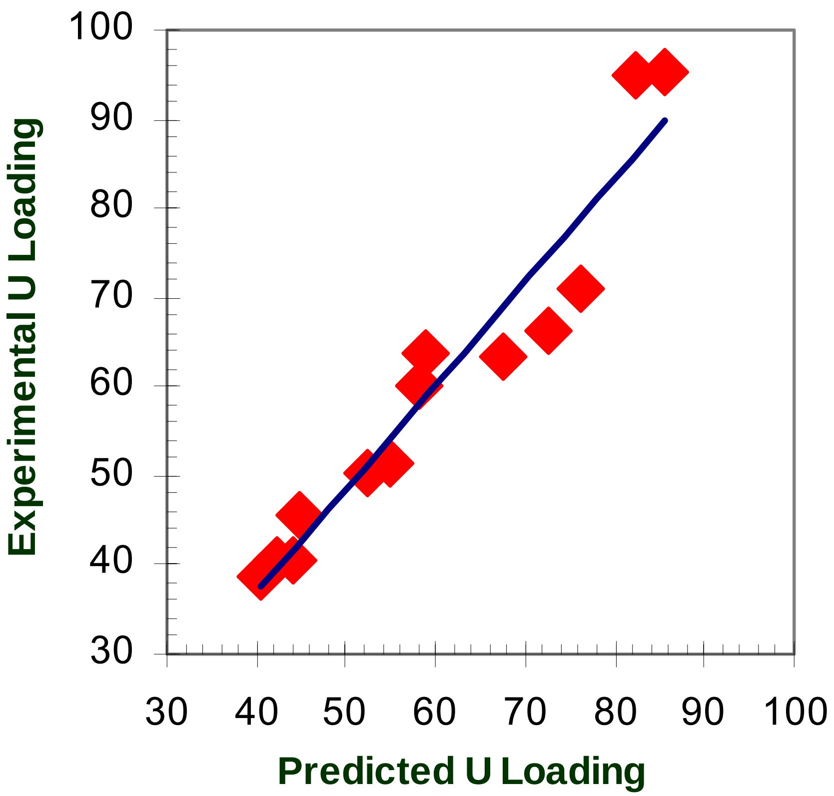
Results

- **Nitric acid data compared with old AMUSE code predicitions**



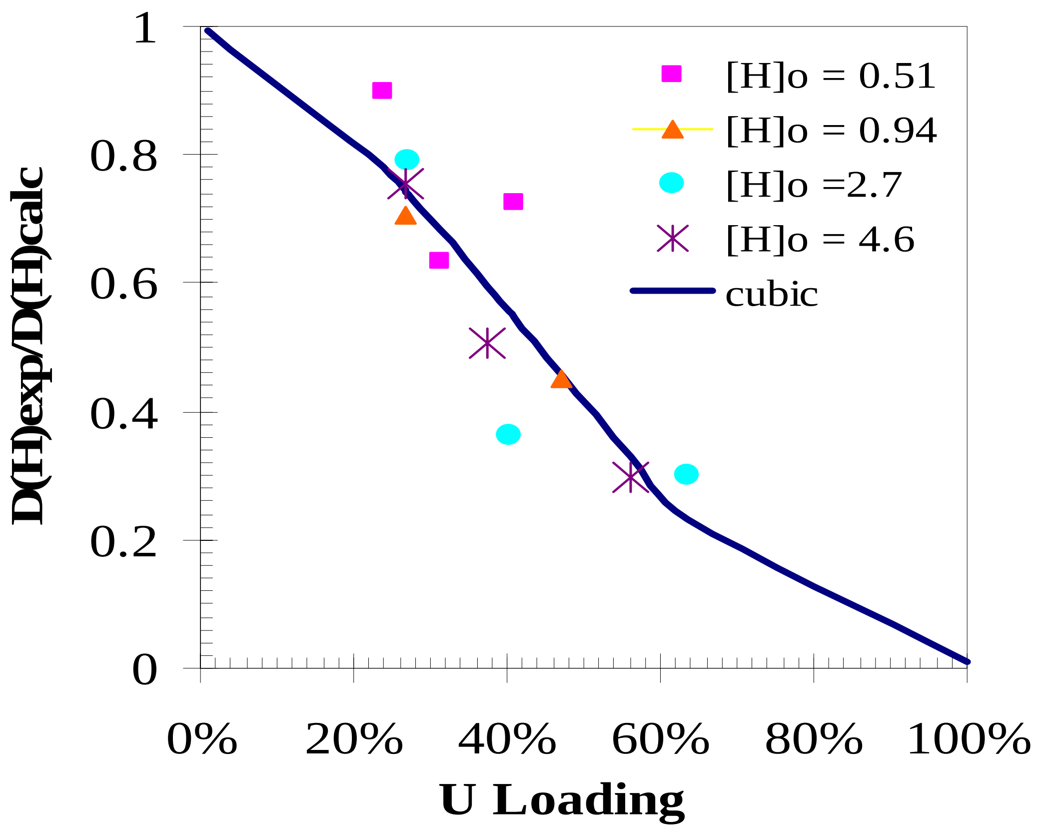
Experimental vs. Predicted U loading

- **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 HNO_3*
  - Organic = 30% TBP in n-dodecane
  - O/A = 4/1 -2/1
- **U distribution ratios and loading were well predicted by AMUSE**
- **HNO_3 partitioning at high loadings were not.**



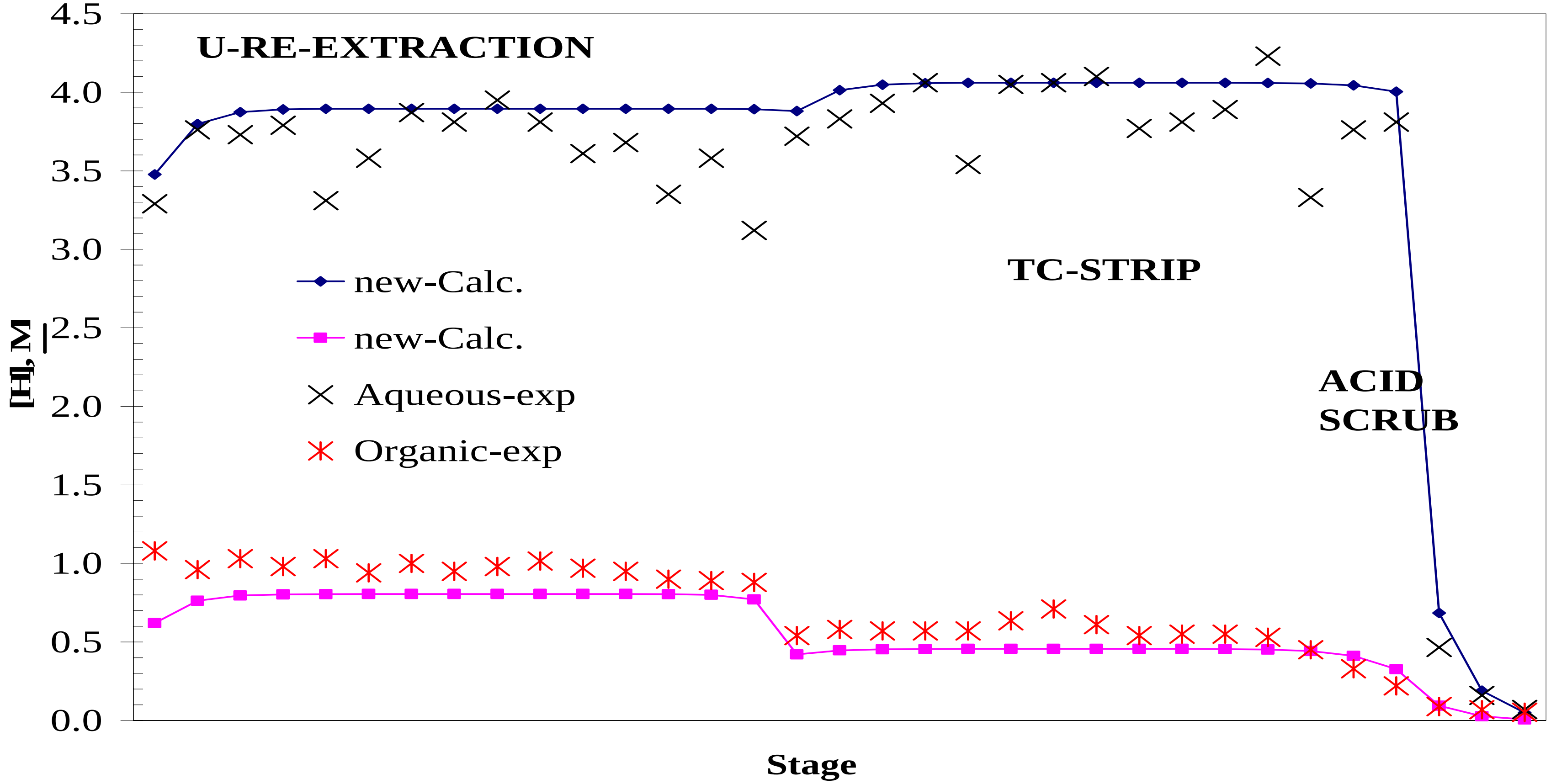
Results—U(VI)/H⁺ Studies

- **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 HNO_3 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**