

# Automated Radiochemistry Efforts at LLNL

Presented to:

TAN 2011, Sochi, Russia

September 8, 2011

**Roger A. Henderson, Kenton J. Moody, Dawn A. Shaughnessy,  
Julie M. Gostic, Evgeny Tereshatov and Warren Oldham\***

Chemistry, Materials, Earth, and Life Sciences Directorate  
Lawrence Livermore National Laboratory

\* Los Alamos National Laboratory

This work was performed under the auspices of the U.S. Department of Energy by  
Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.  
Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, CA 94551

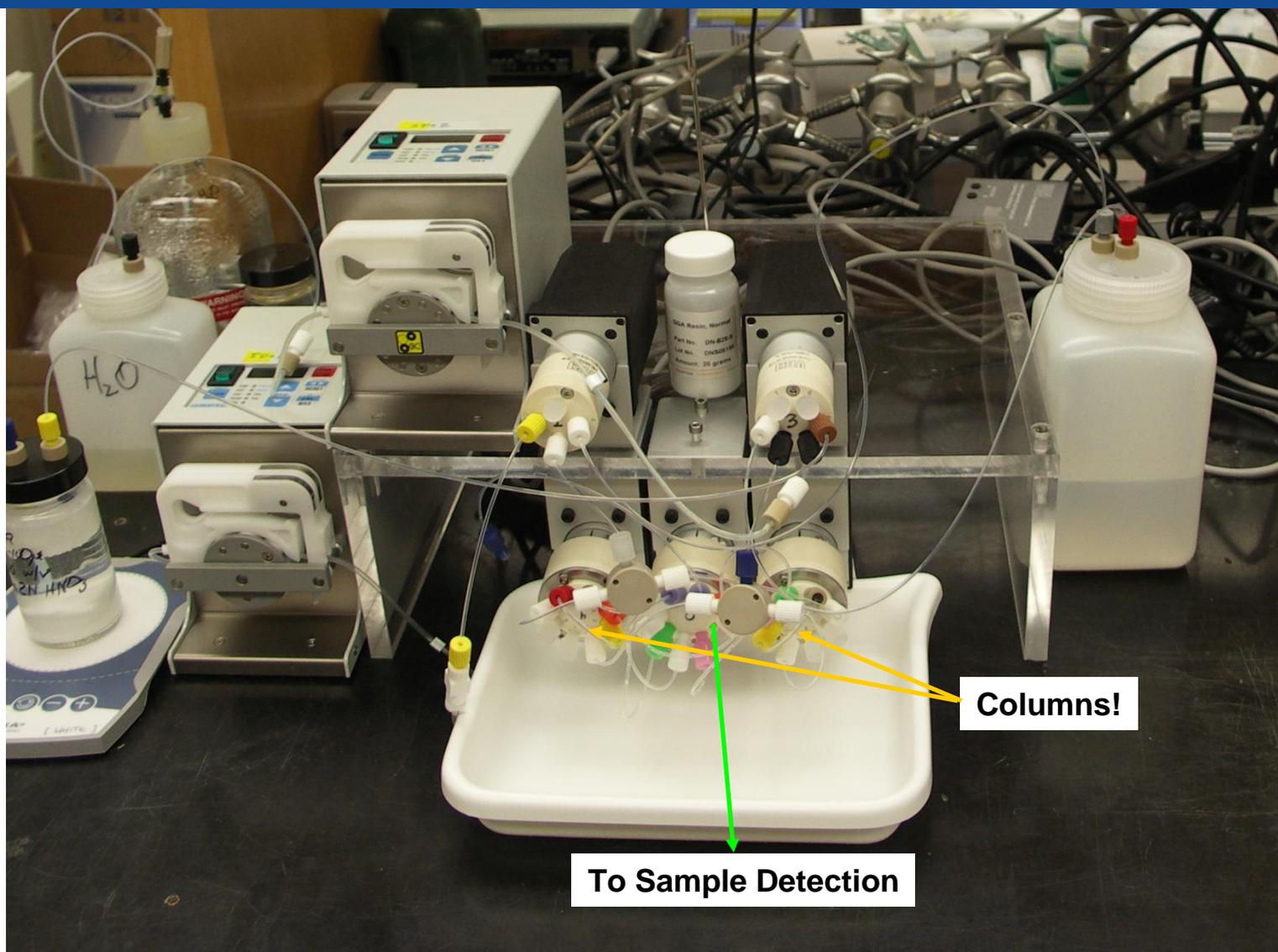
LLNL-PRES-406833

# Automated Radiochemistry Efforts at LLNL

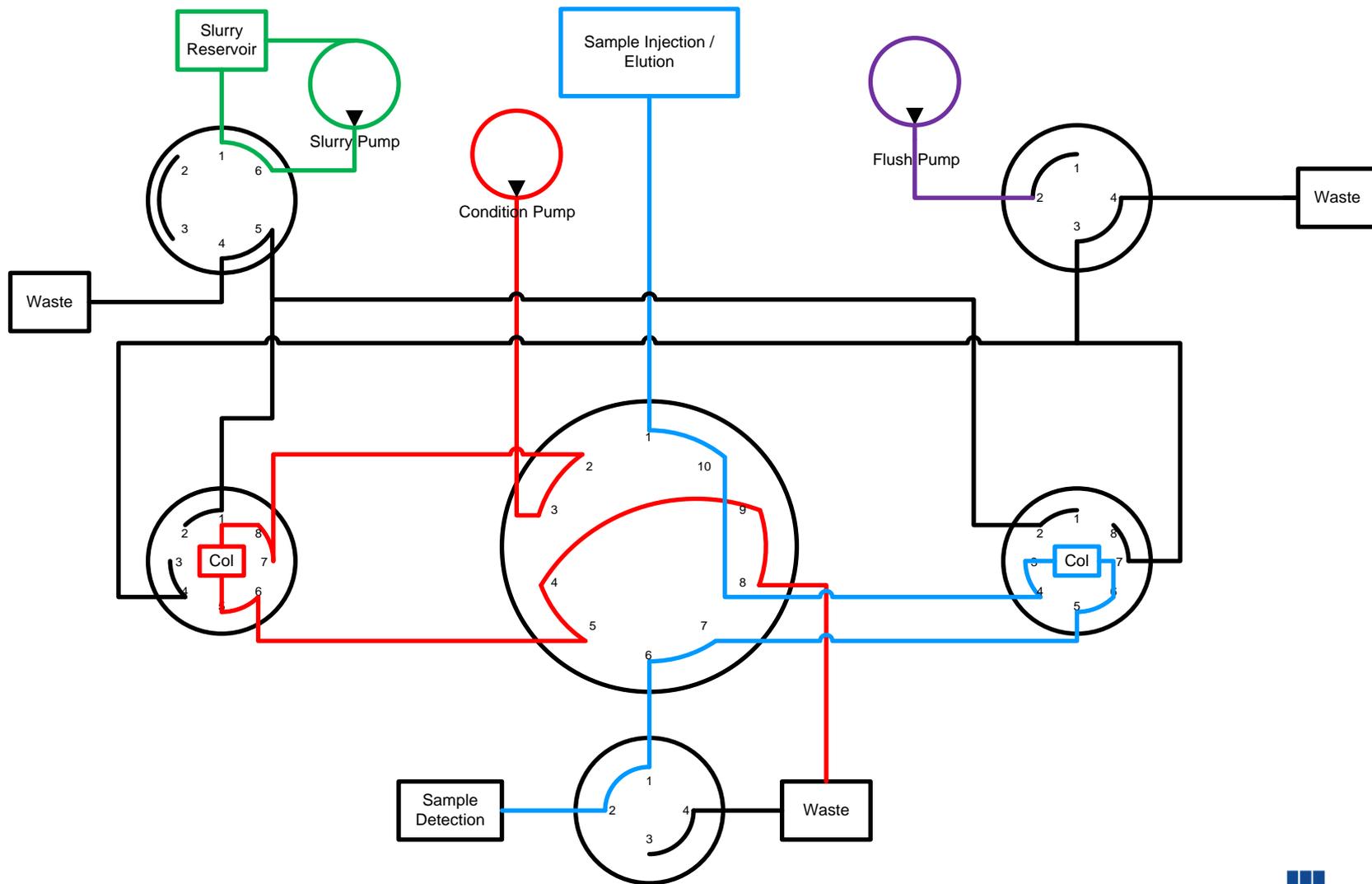
- Chemistry Desires for Automation
  - Speed
  - Reproducibility
  - Lower Dose to Operators
- Begin with a Heavy Element application
  - Single Column chemistry
    - Automated column regeneration
    - Long operating periods (months!)
- Conceptual Design – SHELA – Super Heavy Element Liquid Automation



# Automated Radiochemistry Efforts at LLNL



# Automated Radiochemistry Efforts at LLNL



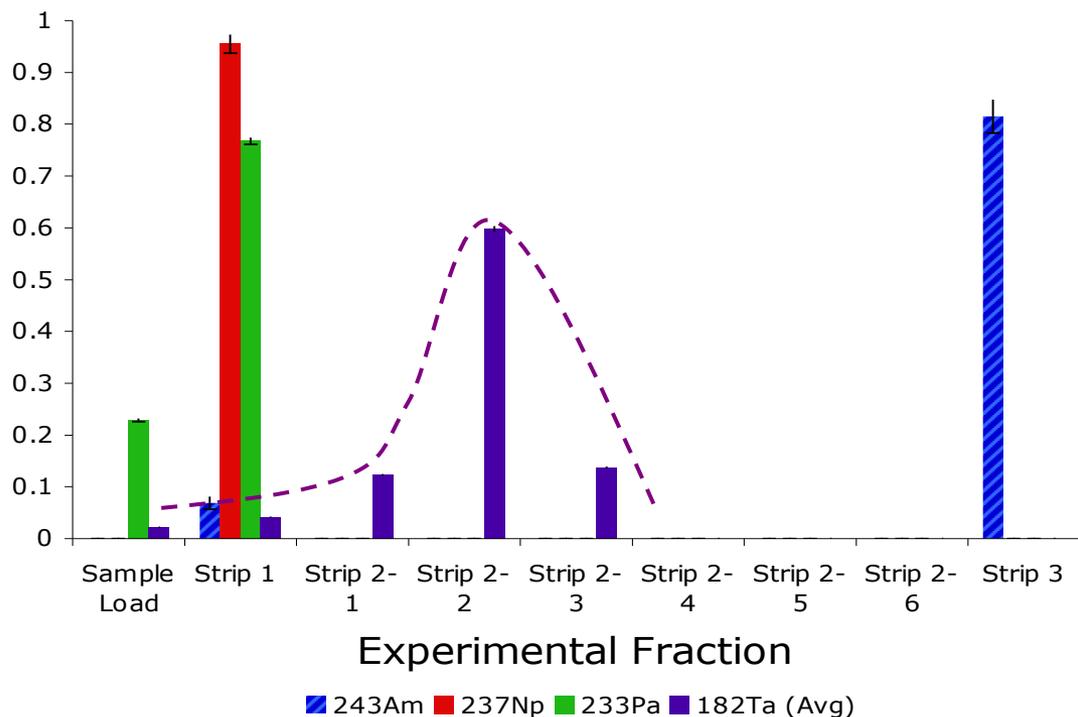
# Automated Radiochemistry Efforts at LLNL

- System Components
  - Cheminert valves, PEEK material, micro-electric actuation
  - ISCO peristaltic pumps
    - Tubing durability – time until it must be replaced?
  - Tubing – FEP – ID = 0.762 mm
  - Columns – FEP tubing, 10 cm length
    - Volumes
      - 0.762 mm tubing – 45.6  $\mu$ l
      - 0.508 mm tubing – 20.3  $\mu$ l
      - 0.254 mm tubing – 5.1  $\mu$ l
    - Frits (for column support)
      - PEEK, 0.5  $\mu$  or 2  $\mu$  Frit-In-A-Ferrule™
- LabView Control



# Automated Radiochemistry Efforts at LLNL

## MS-4 Fractional Radionuclide Activity Distribution



### Sample Loading:

0.4 M HNO<sub>3</sub>/ 0.02 M HF, 1 mL

### Strip 1 (Pa):

0.4 M HNO<sub>3</sub>/0.1 M HF, 5 mL

### Strip 2 (Ta):

8M HNO<sub>3</sub>/0.1 M HF, 6 x 5 mL

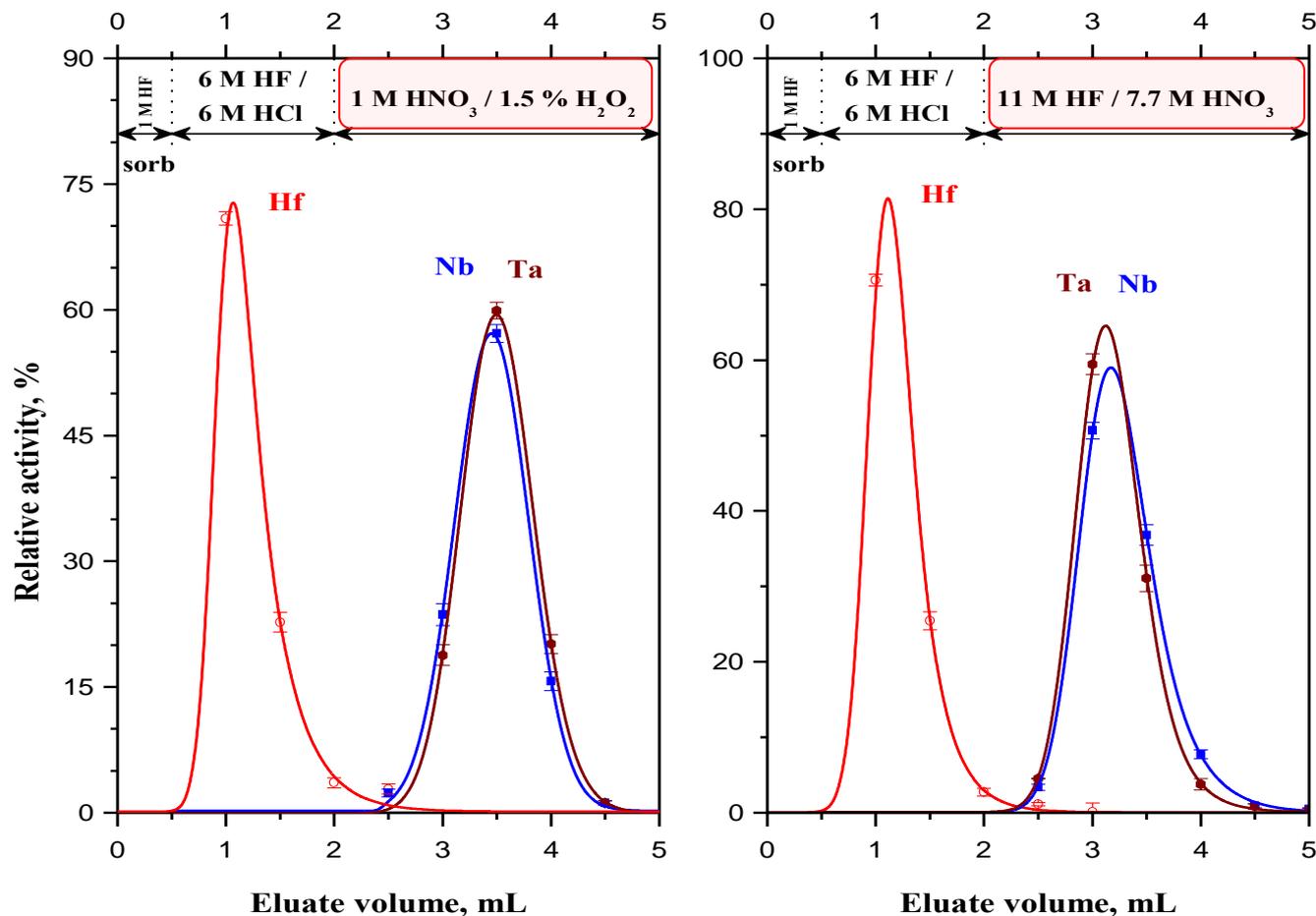
### Strip 3:

0.1 M Ammonium Bioxalate, 5 mL



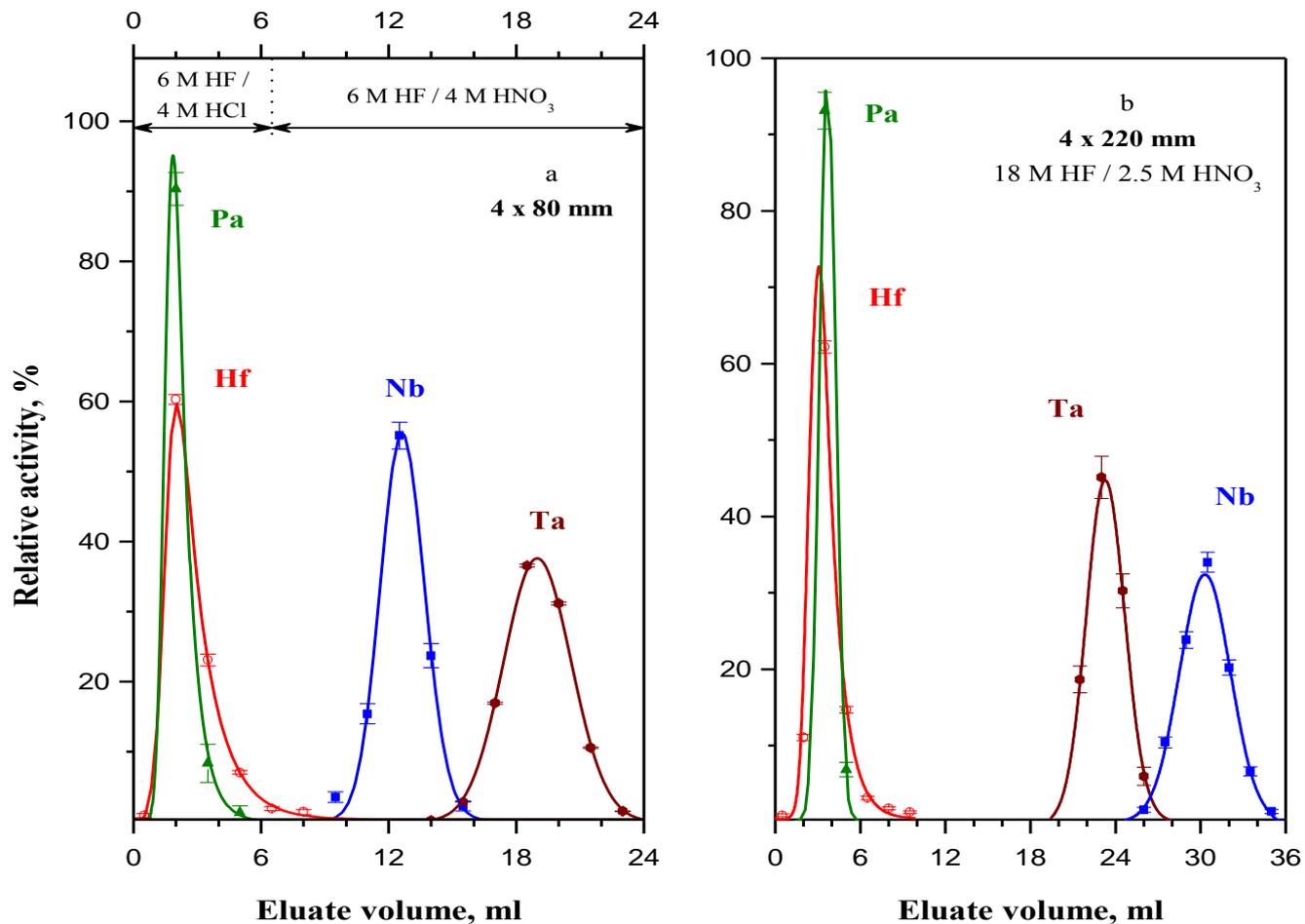
# Automated Radiochemistry Efforts at LLNL

Group IV, V separation using Dowex 1x4, -400 mesh, 4x16 mm



# Automated Radiochemistry Efforts at LLNL

## Separation of Hf, Pa, Nb, and Ta using Dowex 1x4, -400 mesh

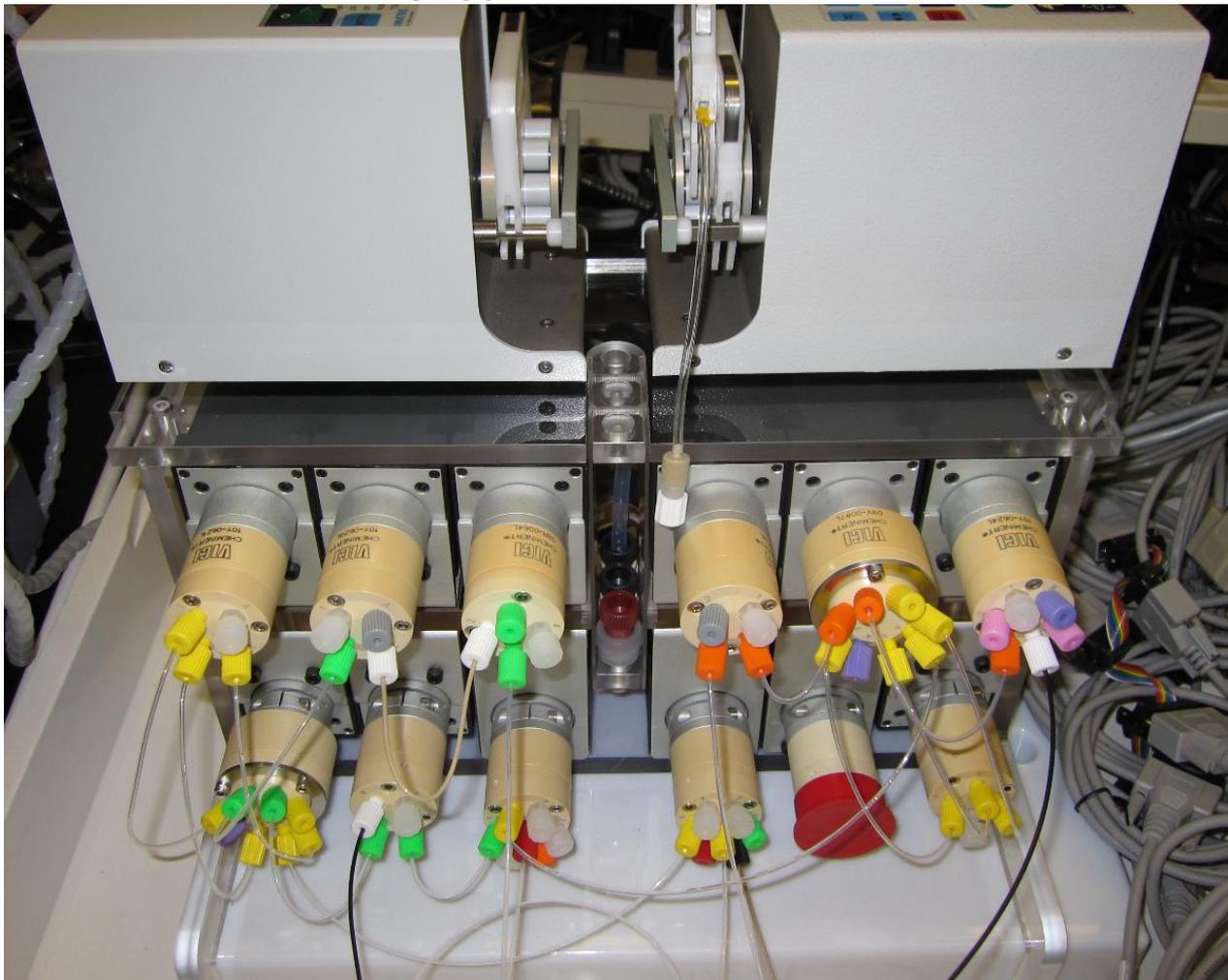


Tereshatov E.E. et al. (2010) J Radioanal Nucl Chem 286, pp. 9-16



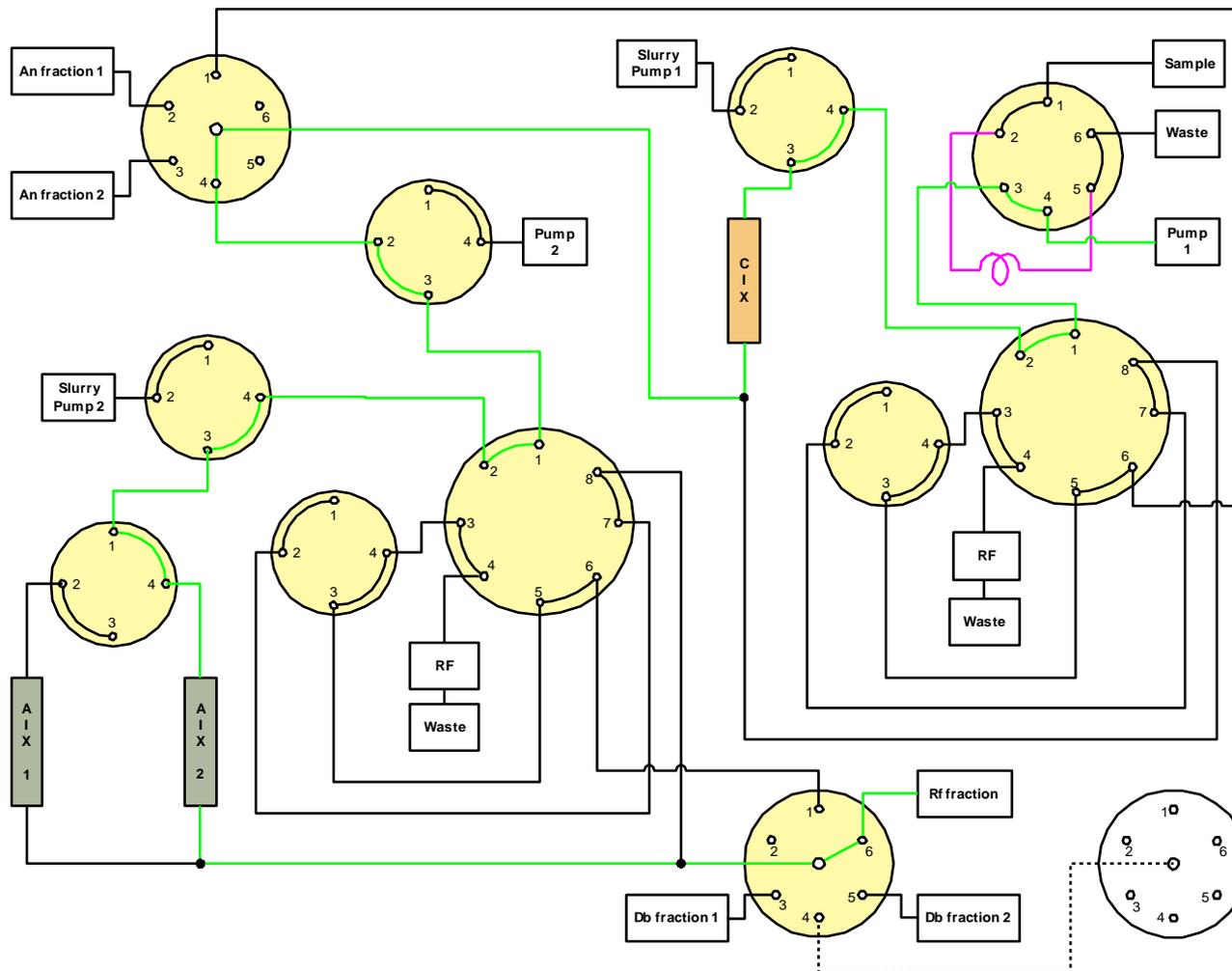
# Automated Radiochemistry Efforts at LLNL

The newest automated chemistry apparatus – hardware view



# Automated Radiochemistry Efforts at LLNL

The newest automated chemistry apparatus – ‘wiring’ diagram



# Automated Radiochemistry Efforts at LLNL

- System Components
  - Cheminert valves, PEEK material, micro-electric actuation
  - ISCO peristaltic pumps – resin slurry only
    - Tubing durability – time until it must be replaced?
  - Tubing – FEP – ID = 0.762 mm
  - Columns – Variable Size
  - Main Pump – Dionex Dual Quaternary Gradient Pump
    - Four eluants into each of two final outputs
    - Chromeleon™ Express controlled
- LabView Control
  - Communications issue with Chromeleon
    - Use ActiveX controls from LabView to tell Chromeleon how to run the pump



# Automated Radiochemistry Efforts at LLNL

- Sample production
  - Sample introduction system
    - Current design at engineering for SolidWorks digital prototyping
    - Three station system - GLITTAR
      - Gas jet collection of a frit
      - Sample removal from the frit
      - Rinse / replace the frit



# Automated Radiochemistry Efforts at LLNL

## Final steps...

- Source Preparation
  - Traditionally a time consuming step of evaporating a solution onto a suitable substrate
  - LANL Functionalized Surface technology
    - CMP (carbamoylmethylphosphonate) ligand functionalized glass or silicon substrates (Initially for Pu<sup>+4</sup> work)
    - Other ligands possible for different projects
      - Alpha spectra are comparable to the best electroplated sources
        - FWHM = 22 keV for <sup>242</sup>Pu, <sup>239/240</sup>Pu
      - Investigating polymer substrates to improve detection efficiency and be able to detect both fission fragments
      - Fast – absorption from solution is rapid
- Detection
  - Array of PIPs detectors for alpha / SF detection

