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Automation of the radiochemical procedures for the sequential separation of radionuclides

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OUTLINE

I. Design and fabrication of an *A*utomated *S*equential *R*adionuclide *S*eparator

- ✓ *ASRS* hardware and software

II. Characterization of the *ASRS*

- ✓ Pump calibration
- ✓ Flow rate characterization
- ✓ Dead volume and memory effect

III. Optimization of an automated sequential radiochemical procedures

- ✓ Tandem column arrangement: TRU resin and Sr-spec resin
- ✓ Flow rate effects on method efficiency: separation of Fe and Sr

IV. Application of the *ASRS*

- ✓ Determination of ^{55}Fe and ^{90}Sr by *ASRS* and liquid scintillation counter

WHY Rapid analysis & Automation?

- ✓ Many samples in an emergency situation
- ✓ Repeated process in radiochemical separation
- ✓ Use of toxic chemicals in RNs separation
- ✓ Labor-intensive radiochemical process
- ✓ Human error under long-period repeated works
- ✓ Slow and time-consuming under gravity flow
- ✓ Deterioration of reproducibility in a vacuum manifold
- ✓ **To increase sample throughput and reproducibility**
- ✓ **To reduce sample preparation costs**
- ✓ **To improve worker safety**
- ✓ **To provide accurate and timely analytical results**

Conventional method & Rapid method?
On the radiochemical separations

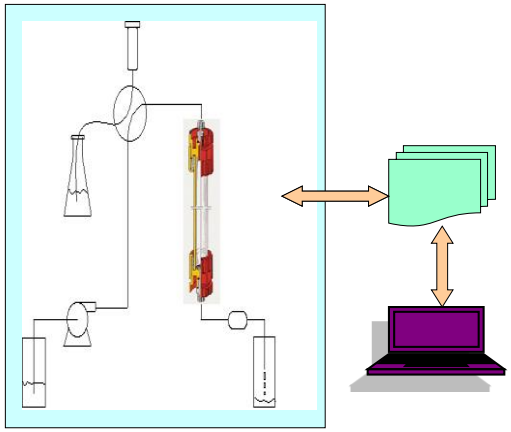
Classical method

Gravity-feed ion exchange

Gravity flow rate

Rapid method

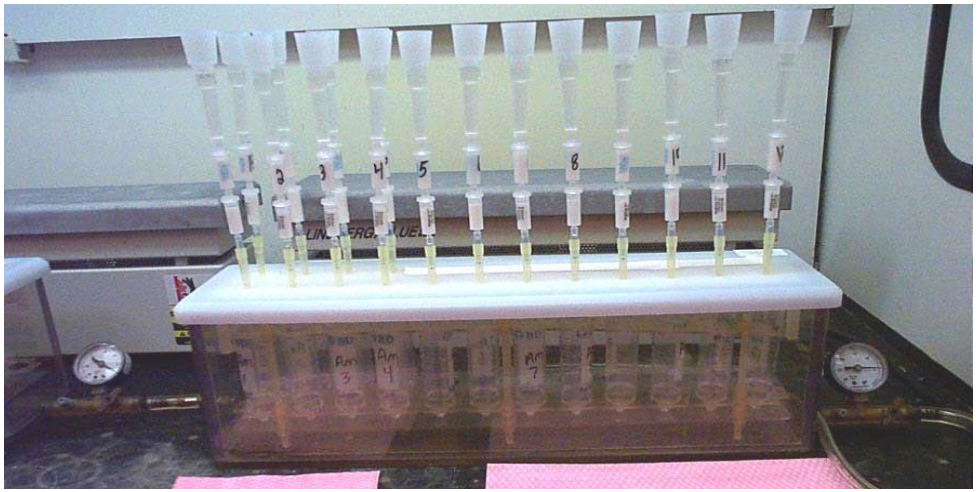
Pump or Vacuum feed ion exchange



Control of flow rate

Enhance the flow rate

Separation systems for enhancing the flow rate



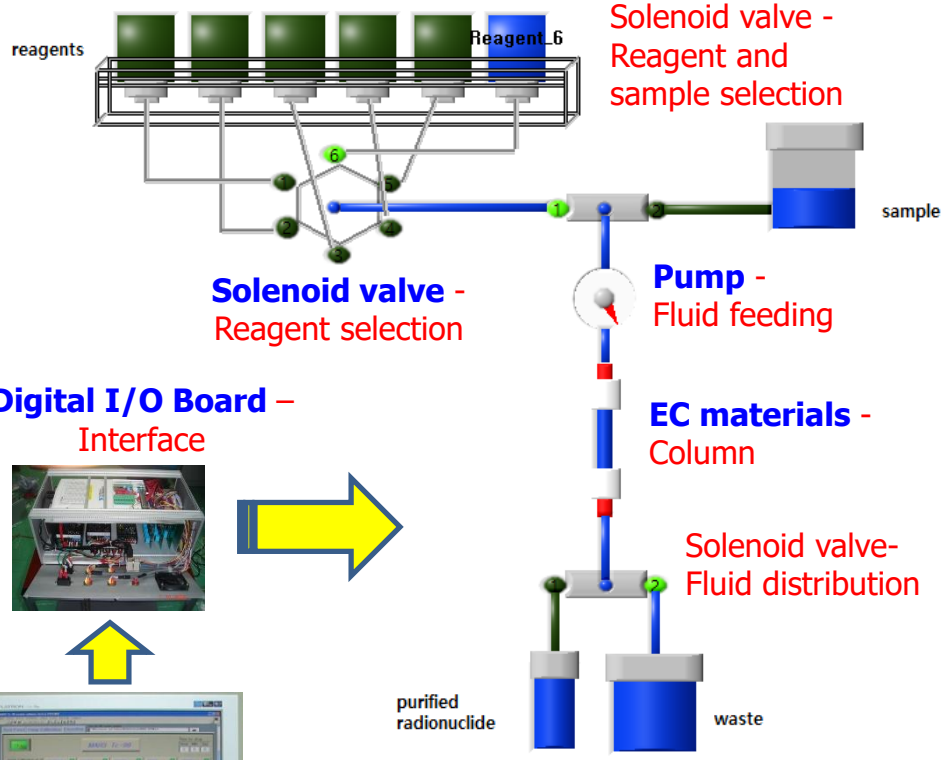
Eichrom: Vacuum Box System

- ✓ Enhanced flow rate, but manual process
- ✓ Efficient for batch processing
- ✓ Sample through-put is limited by the labor force and individual skill sets
- ✓ Exposed to the open sources of radioactivity

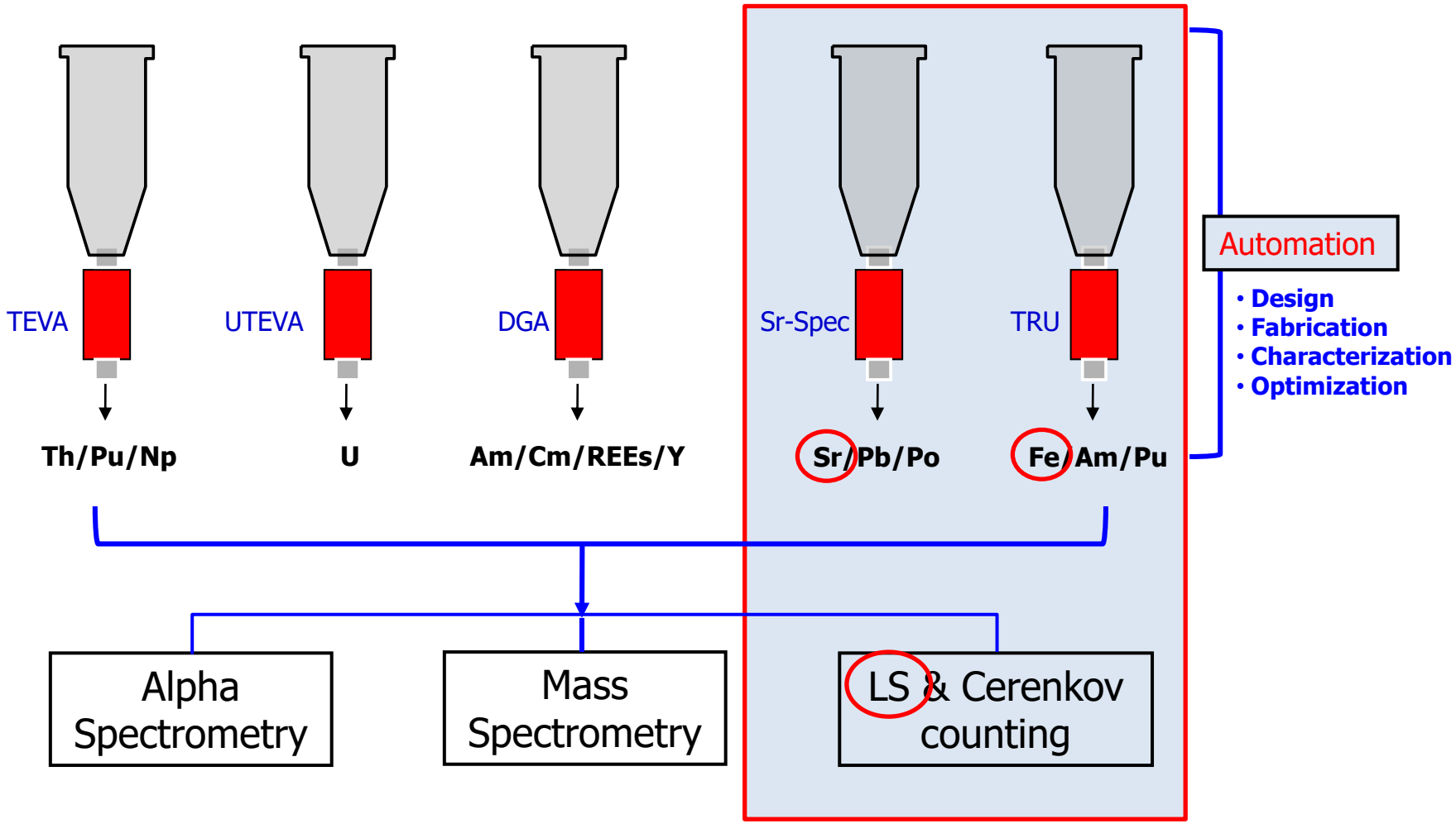
What do you need for translating manual methods into automated methods ?

Eichrom: Vacuum Box System
Enhanced flow rate, but manual process

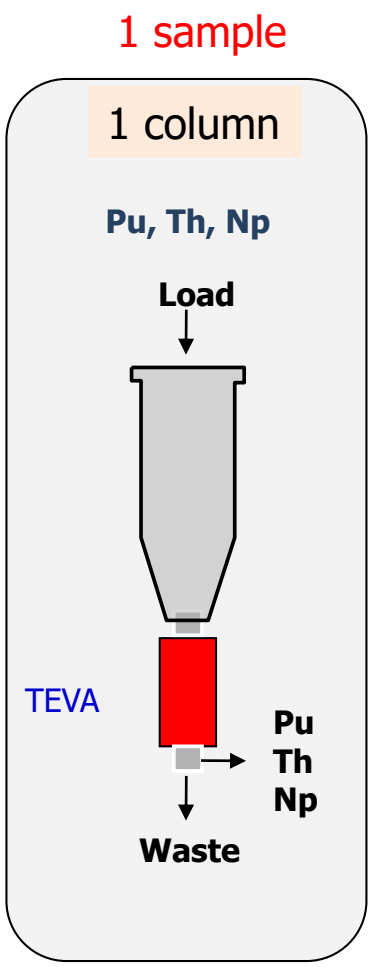
Automation



Simultaneous Determination of Hard-to-Measure Radionuclides:
Extraction Chromatography, Mass Spectrometry & Radiometry



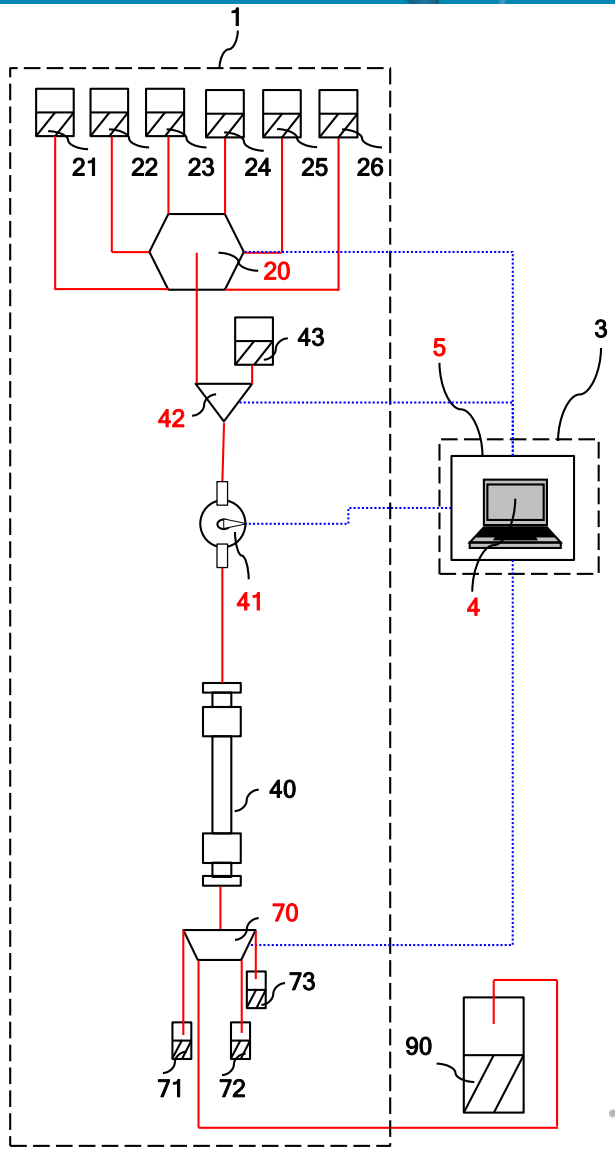
Design of an automated radionuclide separation system:
Sequential separation by single column arrangement: 1 sample



Automation

- ✓ Solid **red lines**: fluid line tubing
- ✓ Dotted **blue lines**: signal communication lines

- ✓ 6 way valve: 1ea (20)
- ✓ 2 way valve: 1ea (42)
- ✓ 4 way valve: 1ea (70)
- ✓ 1-channel pump: 1ea (41)
- ✓ Digital I/O board: 1ea (5)
- ✓ PC-control software: 1ea (4)

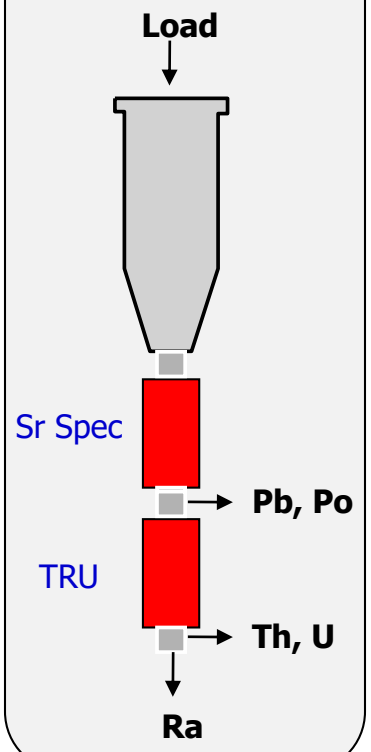


Design of an automated radionuclide separation system:
Sequential separation by tandem column arrangement: 1 sample

1 sample

2 columns

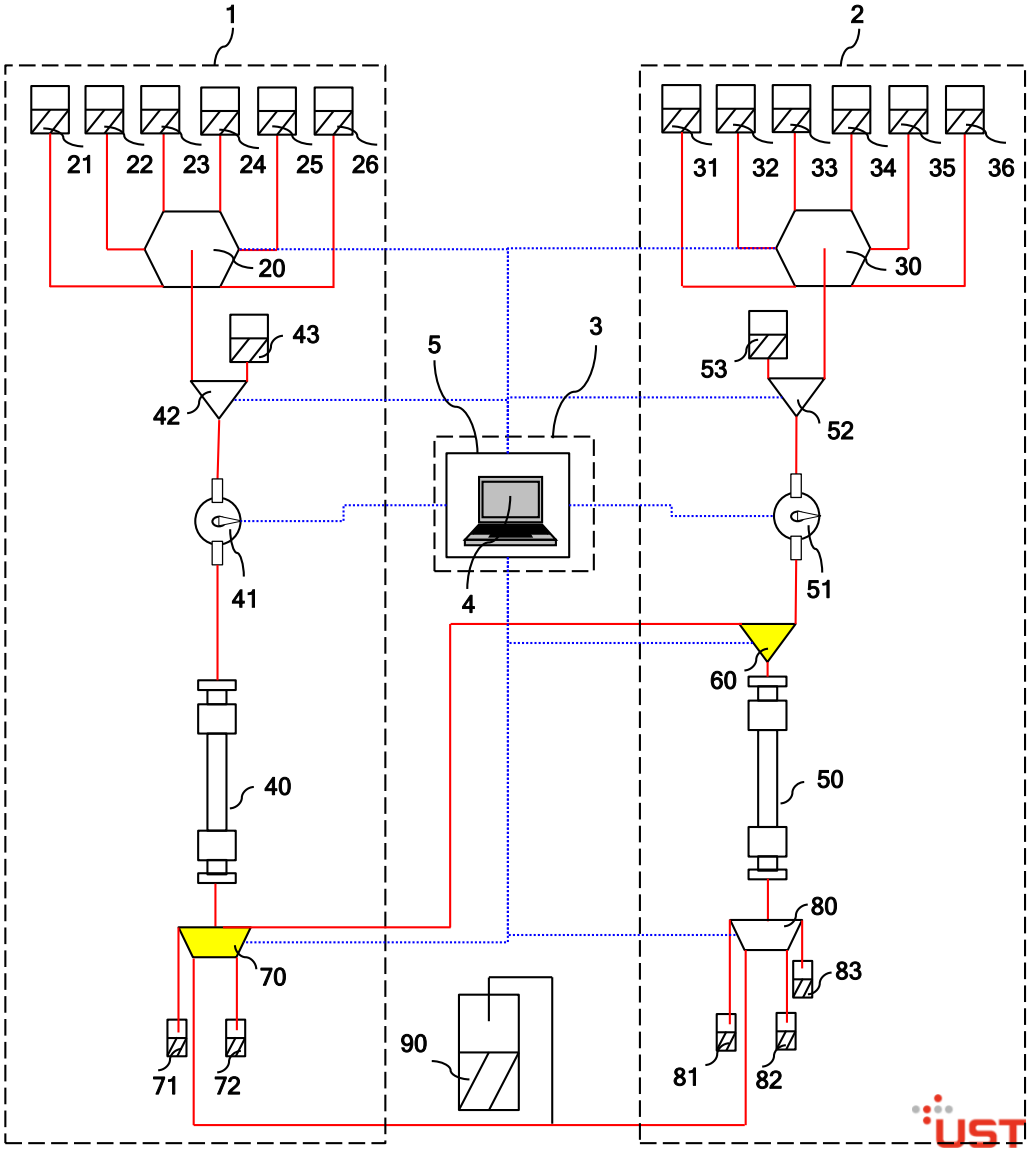
Pb, Po, U, Th, Ra



Automation

- ✓ Solid red lines: fluid line tubing
- ✓ Dotted blue lines: signal communication lines

- ✓ 6 way valve: 2ea (20,30)
- ✓ 2 way valve: 2ea (42,52,60)
- ✓ 4 way valve: 2ea (70,80)
- ✓ 1-channel pump: 2ea (41,51)
- ✓ Digital I/O board: 1ea (5)
- ✓ PC-control software: 1ea (4)



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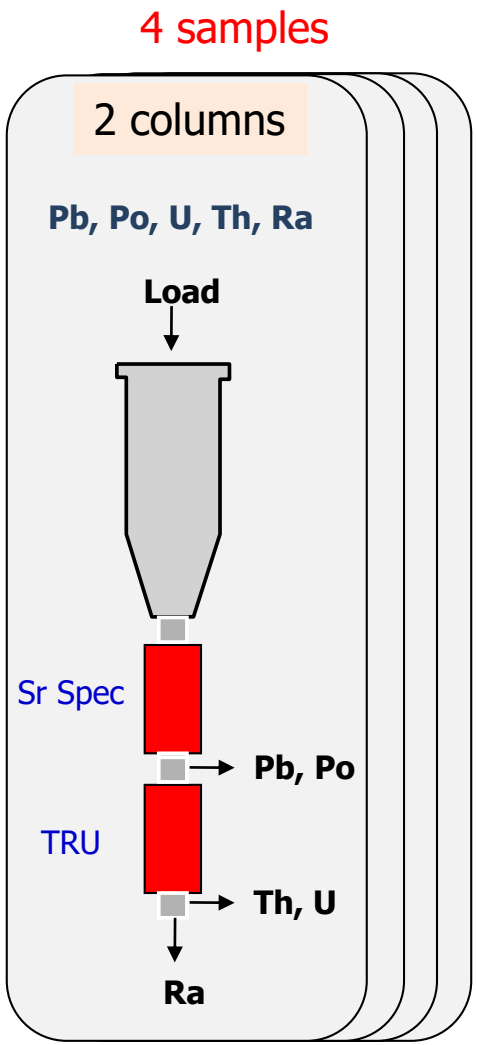
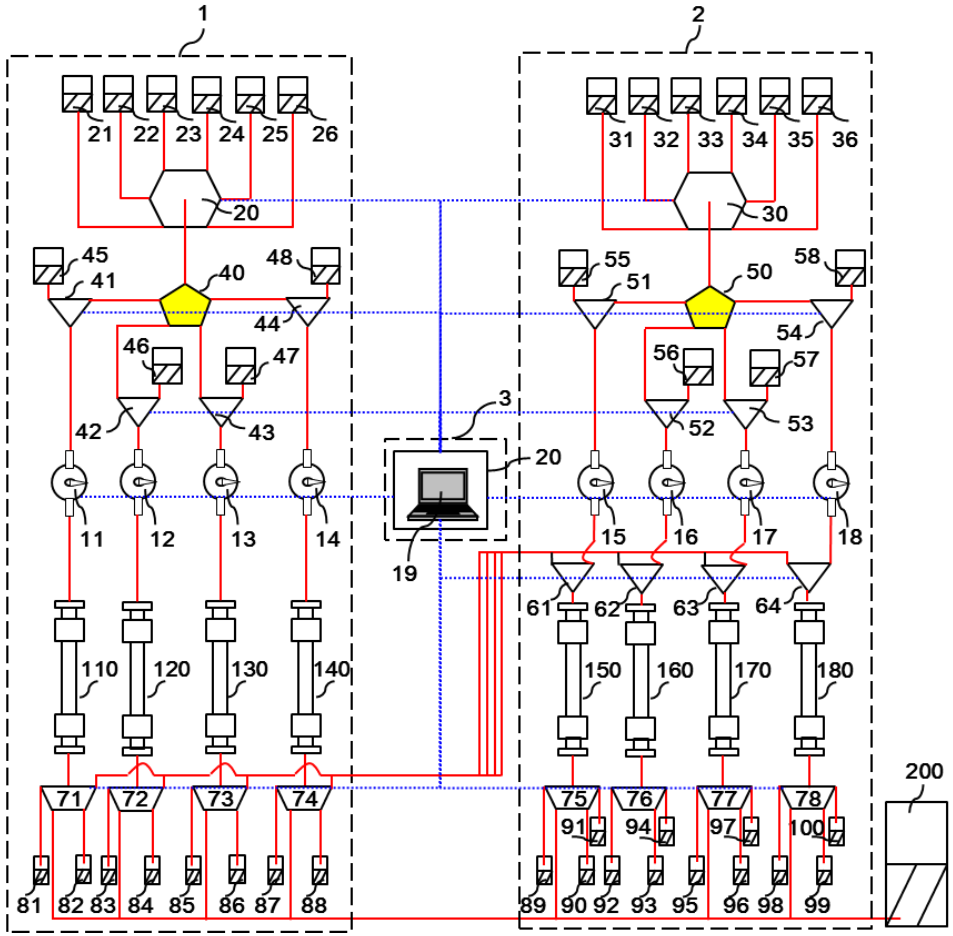
Design of an automated radionuclide separation system:
Sequential separation by tandem column arrangement: 4 samples



Automation

- ✓ Solid **red lines**: fluid line tubing
- ✓ Dotted **blue lines**: signal communication lines

- ✓ 6 way valve: 2ea (20,30)
- ✓ 4 way connector: 2ea (40, 50)
- ✓ 2 way valve: 8ea (41~44,51~54)
- ✓ 4 way valve: 8ea (71~78)
- ✓ 1-channel pump: 8ea (11~18)
- ✓ Digital I/O board: 1ea (20)
- ✓ PC-control software: 1ea (19)



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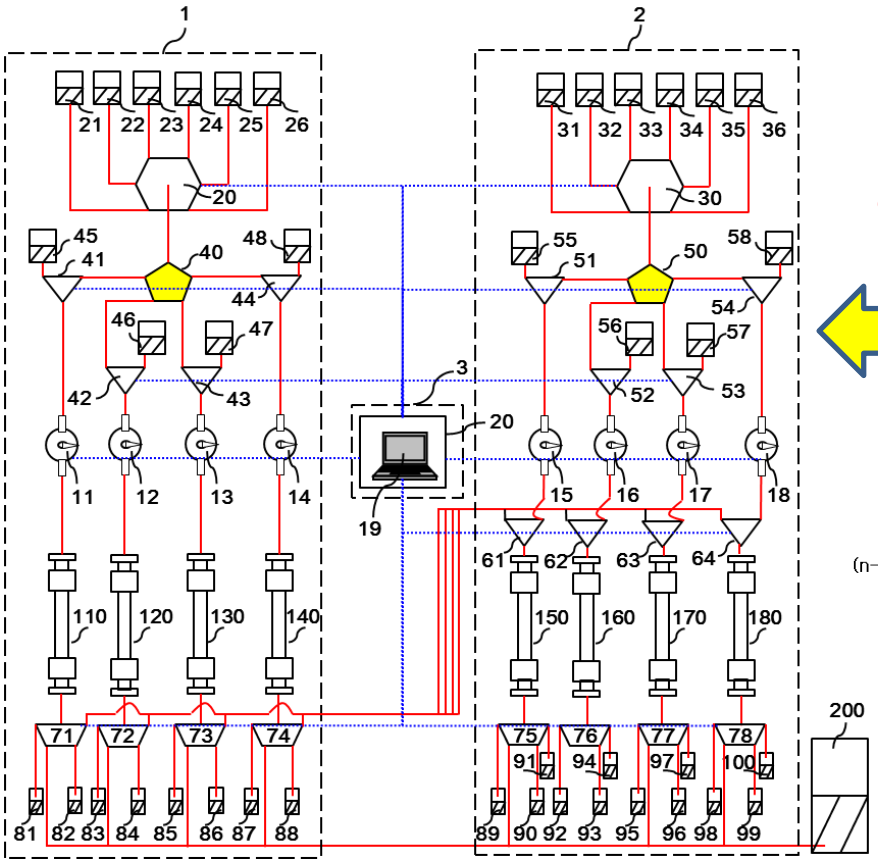
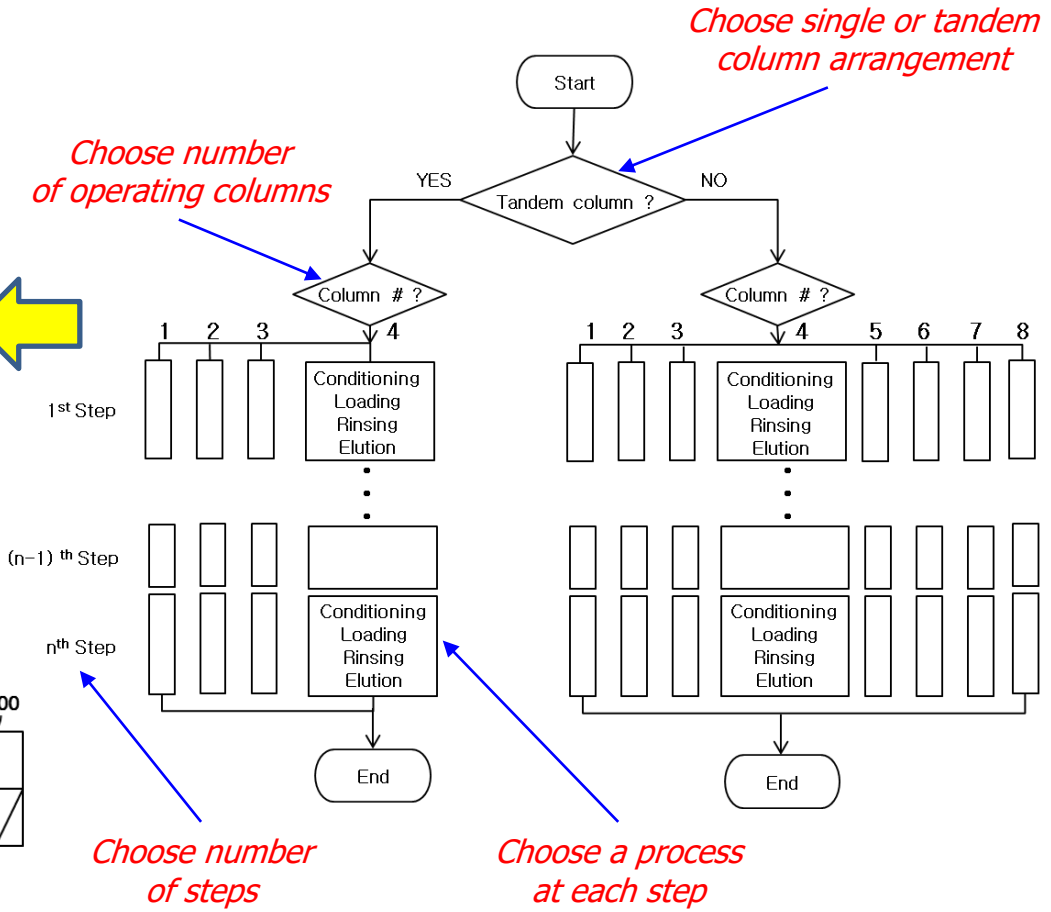
Design of an automated radionuclide separation system:
Algorithm used to the source code for executing the automated protocol

Signal and fluid connection of the *ASRS*



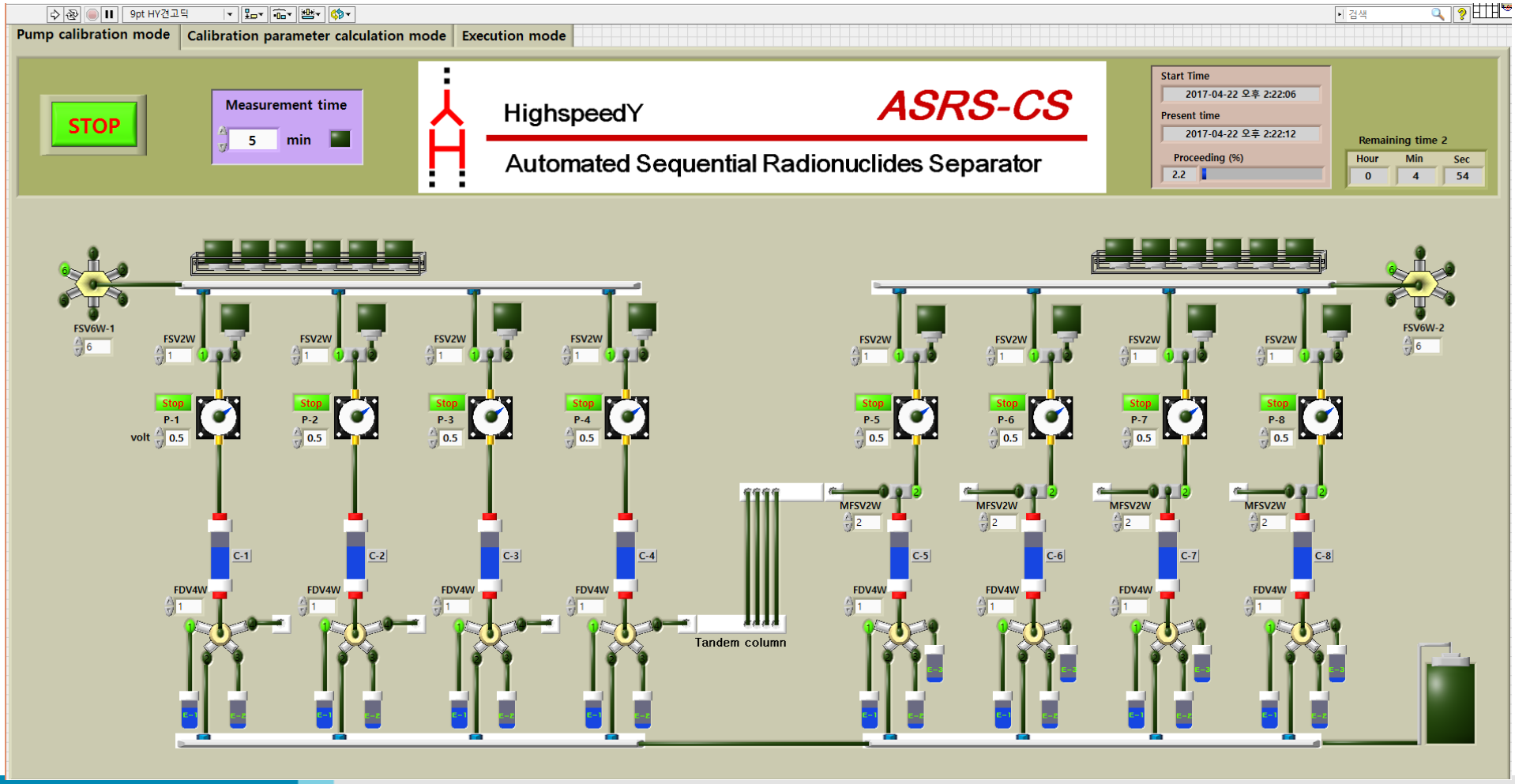
Algorithm to control the *ASRS*



Fabrication of an automated radionuclide separation system:
Control software of ASRS

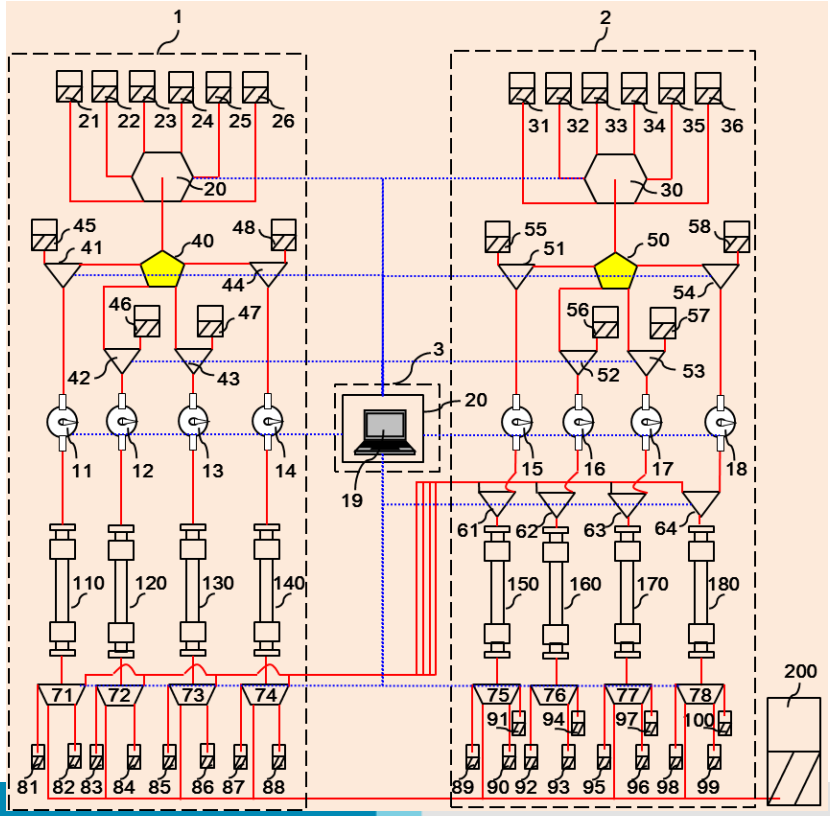
System control software: VIRTUAL INSTRUMENT

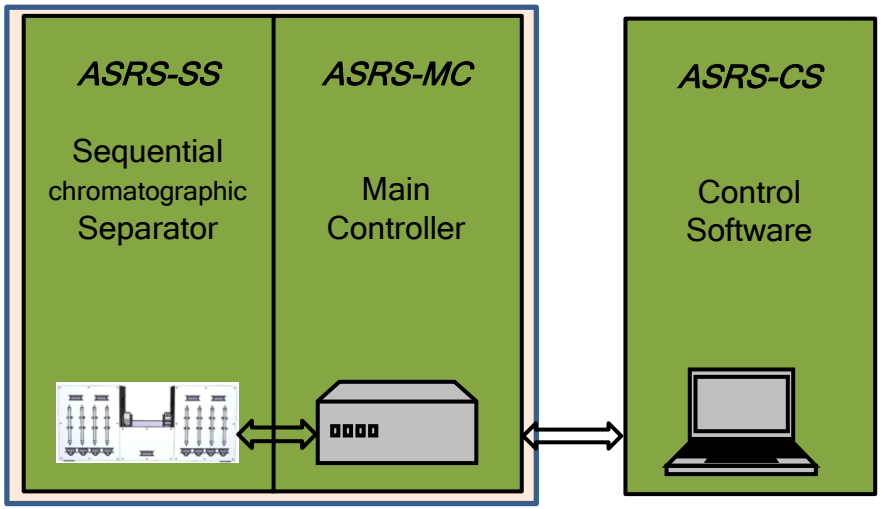
Graphical User Interface: easy to learn and use the software



Fabrication of an automated radionuclide separation system:
Conceptual design of ASRS

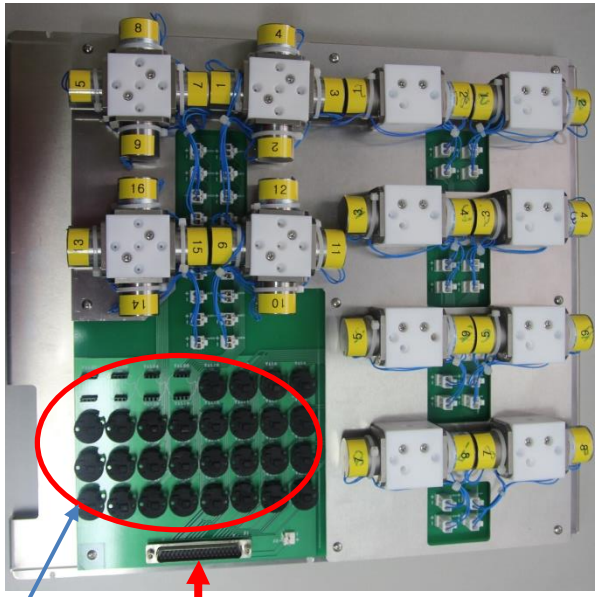
- ✓ **Automated column-based extraction chromatography**
- ✓ **2 RNs: 8 samples in parallel with a single column arrangement**
- ✓ **5 RNs: 4 samples in parallel with a tandem column arrangement**
- ✓ **PC control: system software (LabVIEW) for instrument control**



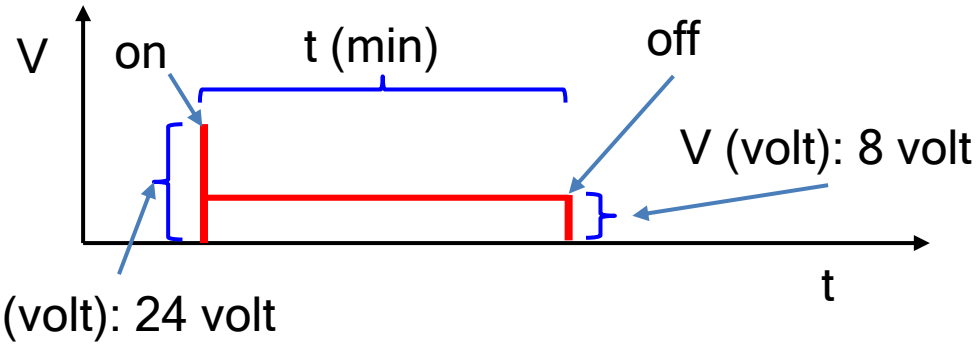


Automated Sequential Radionuclide Separator:
Fabrication of ASRS

Design of a solenoid valve driver circuit board to actuate solenoid valves



- ✓ **Solenoid valve ON: connect 24V DC**
- ✓ **Solenoid valve OFF: disconnect 24V DC**



To prevent overheating valves: 24V → 8V

Switching

24V DC

Digital I/O Board

On

Off

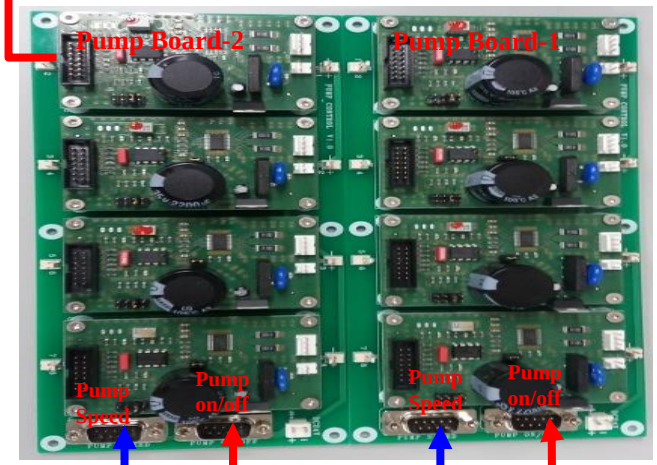
PC-Control Software

Power Supply

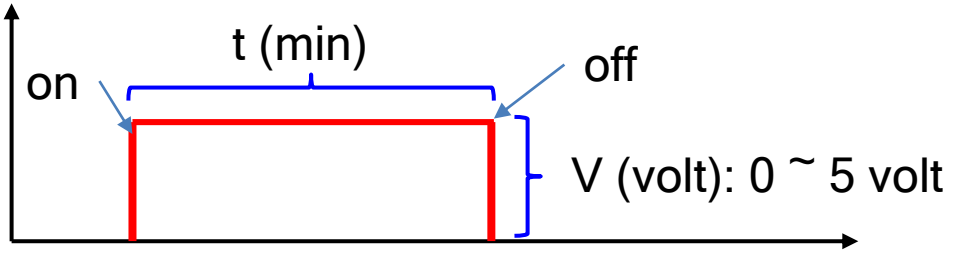
24 V DC

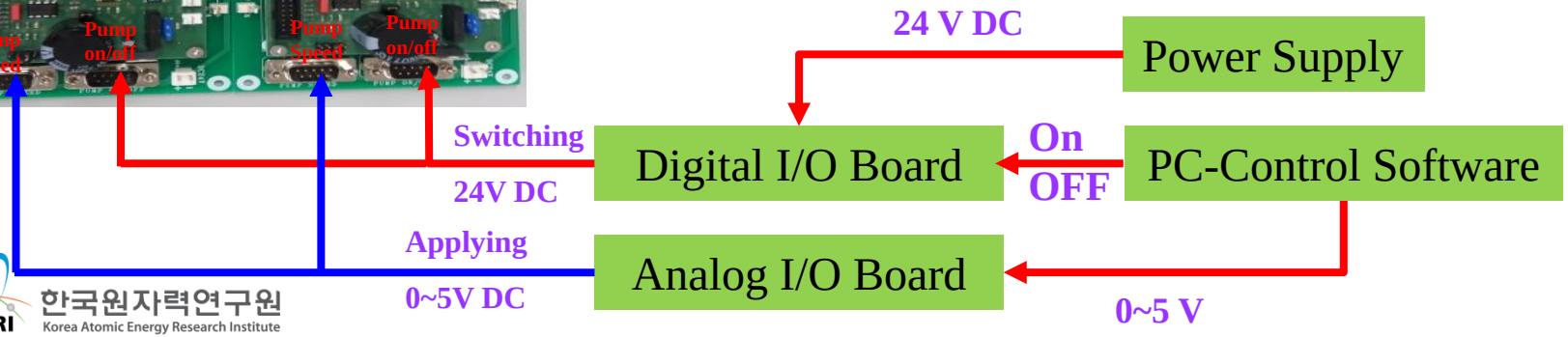
Automated Sequential Radionuclide Separator:
Fabrication of ASRS

Design of a peristaltic pump driver circuit board to control peristaltic pumps



- ✓ **Pump calibration function:**
  - * Flow rate (f, mL/min) = a (mL/min/volt)*voltage (V, volt) + b (mL/min)
- ✓ **Input parameters: Flow rate (f, mL/min), Volume (mL)**
- ✓ **Pump working time (t): calculated by input parameters**
  - * t (min) = Volume/Flow rate
- ✓ **Pump speed control (volt): Calculated by pump calibration function**
  - * V (volt) = (f – b)/a





KAERI development history of Automated Radionuclide Separator

MARS Sr-90, 2007~2009

MARS Tc-99, 2008~2010

- ✓ The peristaltic pump has a 4-channels head
- ✓ Flow rate differences between the channels

ASRS-two pumps, 2012~2014

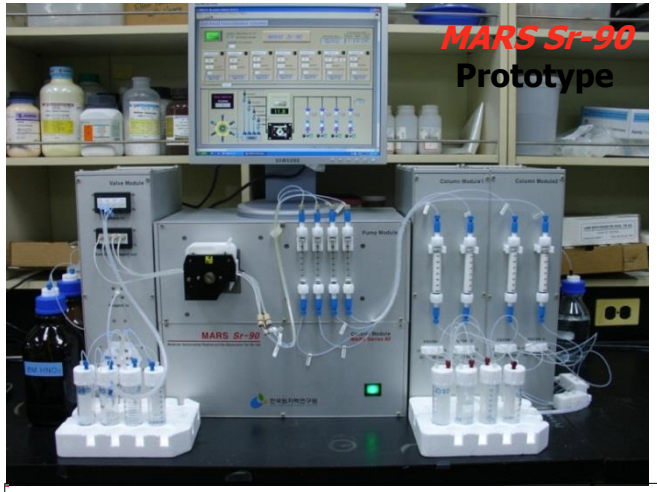
- ✓ Minimized flow difference between the pumps
- ✓ Slow deterioration of the pump tubing

ASRS-eight pumps, 2014~2015

Commercialized models, 2016~

An automated separation system developed by KAERI: Modular Automated Radionuclide Separator for Sr-90

- ✓ **Modular type system: control module, pump module, valve module, column module**
- ✓ **Four samples in parallel**
- ✓ **User friendly system software: Virtual Instrument**
- ✓ **Input parameters: volume, flow rate, reagent number in conditioning, rinsing, loading and elution steps**



J Radioanal Nucl Chem (2015) 304:293–300
DOI 10.1007/s10967-014-3594-z

Rapid determination of radiostrontium in milk using automated radionuclides separator and liquid scintillation counter

Kun Ho Chung · Hyuncheol Kim · Jong Myoung Lim · Young-Yong Ji · Geun-Sik Choi · Mun Ja Kang

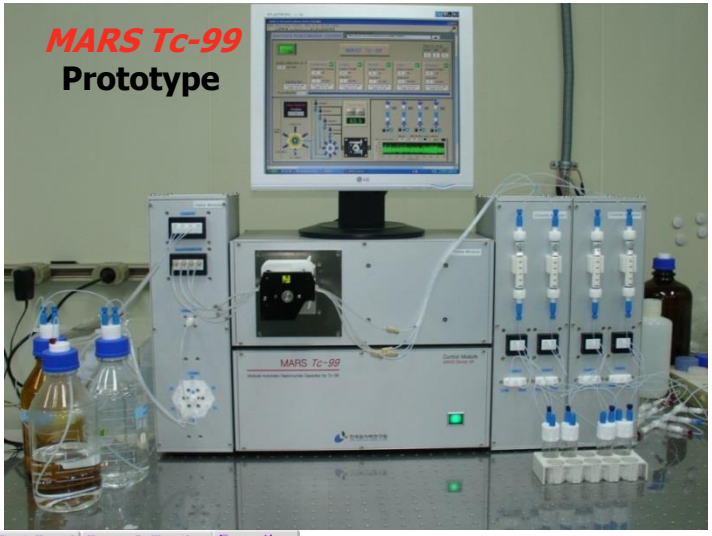
Received: 1 September 2014 / Published online: 19 September 2014

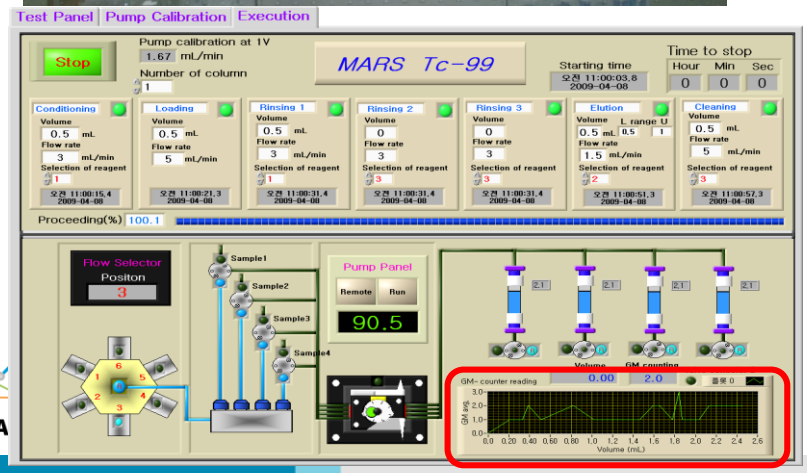
Abstract A modular automated radionuclide separator for $^{89/90}$Sr (MARS Sr-90) has been developed for the rapid and reproducible isolation of radiostrontium. The automated radiochemical method for separation and purification of $^{89/90}$Sr in milk was developed and applied to the literature [1–8]. Both ^{89}Sr and ^{90}Sr, produced by the fission reaction of nuclear fuel in a nuclear reactor, are beta emitters that require radiochemical separation for radiometric analysis. The conventional methods for the determination of radiostrontium in milk are using time consuming sample

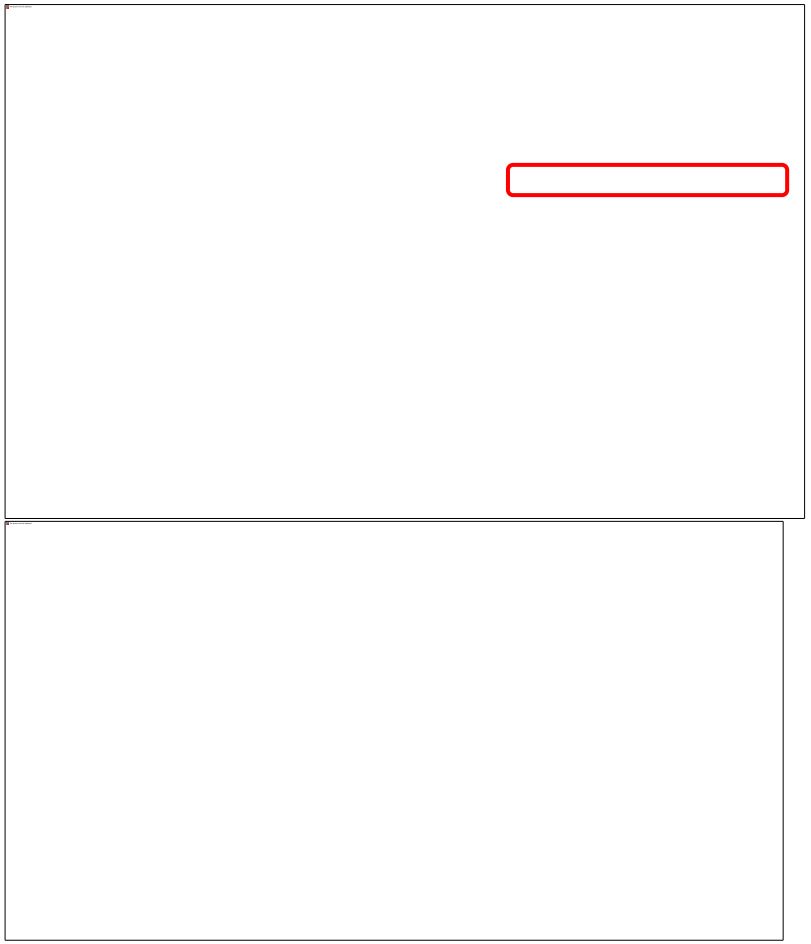
Chung *et al.*, J. Radioanal. Nucl. Chem., 2015, 304,293-300

An automated separation system developed by KAERI: Modular Automated Radionuclide Separator for Tc-99

- ✓ **Modular type system: control module, pump module, valve module, column module**
- ✓ **User friendly system software: Virtual Instrument**
- ✓ **GM counter: monitor and select of $^{99m}Tc/^{99}Tc$ from the sample**
- ✓ **GM counter signal: Used to switch the 3W-FDV**







Chung *et al.*, Appl. Radat. Isot., 2013, 81, 57-61

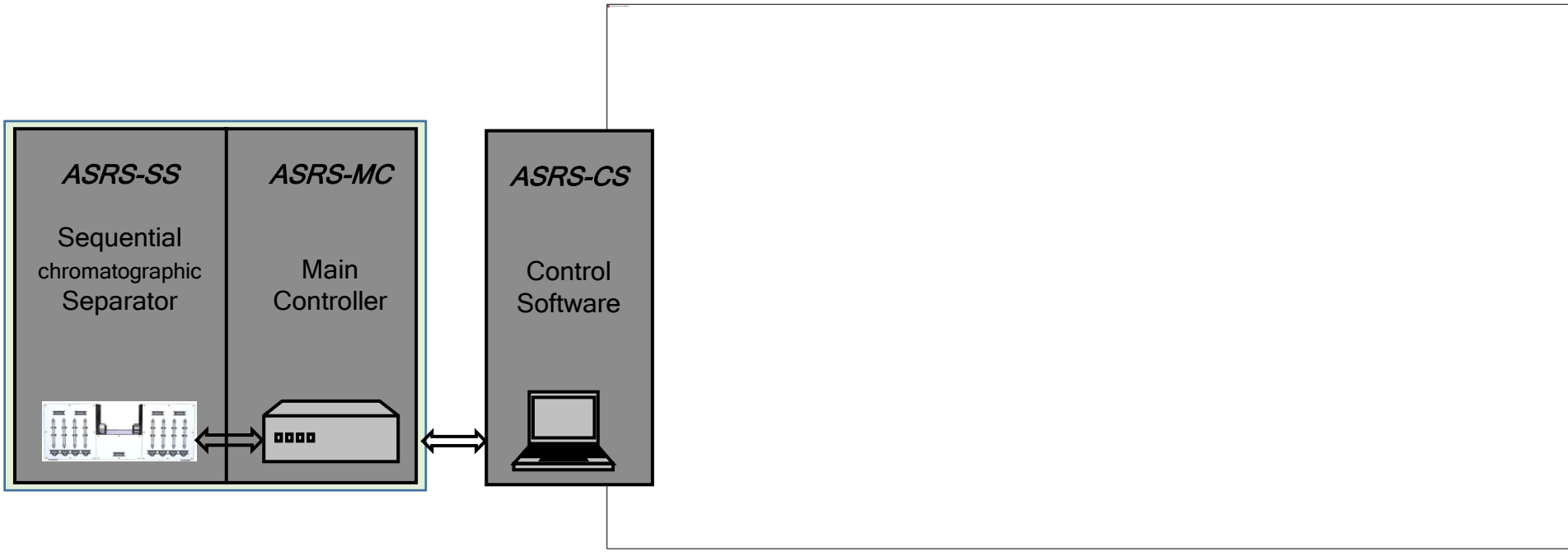
Automated Sequential Radionuclide Separator:
Developed ASRS

System Design Specification

Automated Sequential Radionuclide Separator: *Developed ASRS*

System Configuration

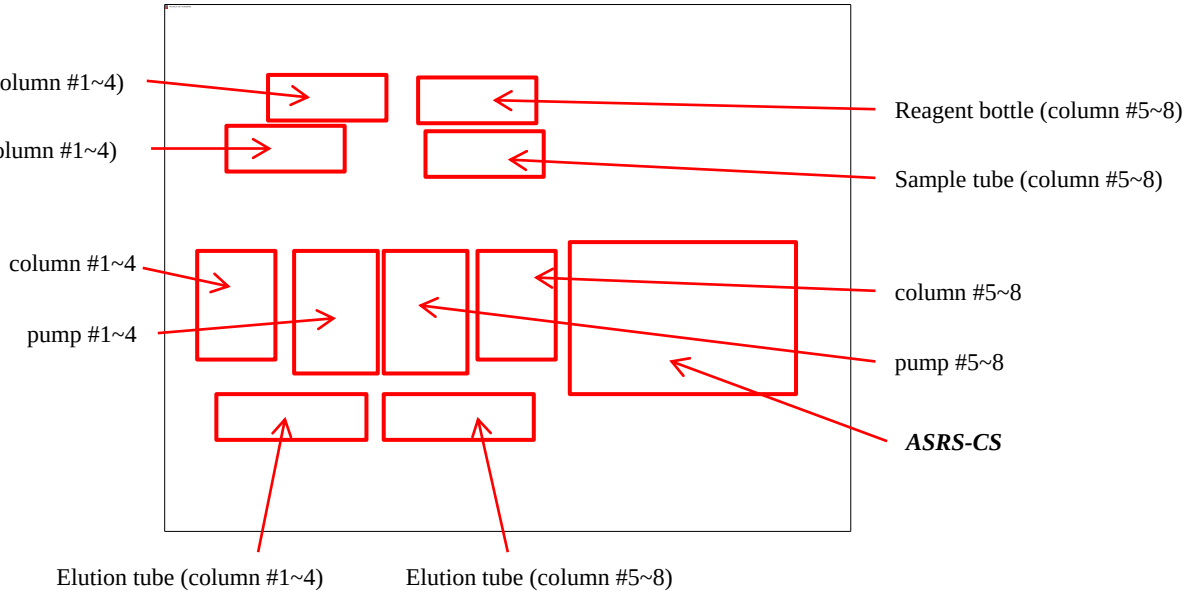
- The ASRS consists of three parts: *ASRS-CS, ASRS-MC, ASRS-SS*.
- The *ASRS-CS* means “Automated Sequential Radionuclide Separator-Control Software.
- The *ASRS-MC* means “Automated Sequential Radionuclide Separator-Main Controller, which is built in ASRS-SS.
- The *ASRS-SS* means “Automated Sequential Radionuclide Separator-Sequential chromatographic Separator.



Automated Sequential Radionuclide Separator: *Developed ASRS*

System Configuration

The ***ASRS-SS*** consists of five parts: Reagent bottle part (column #1~4 & column #5-8), Sample tube part (column #1~4 & column #5-8), Column part (column #1~4 & column #5-8), Pump part (pump #1~4 & pump #5~8), Elution tube part (column #1~4 & column #5-8).



Automated Sequential Radionuclide Separator:
Developed Control software of ASRS : VIRTUAL INSTRUMENT

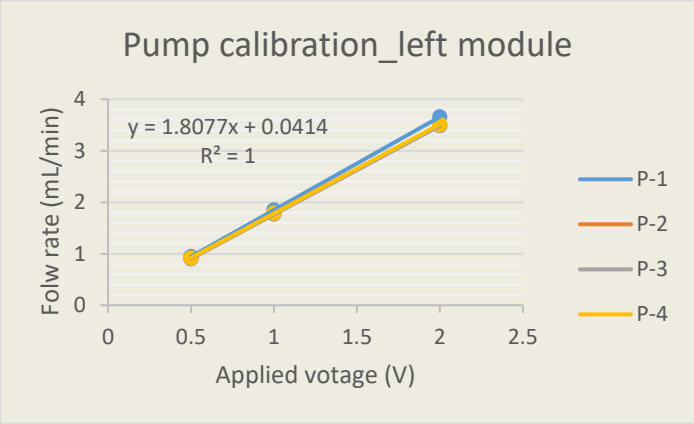
Control Software Configuration

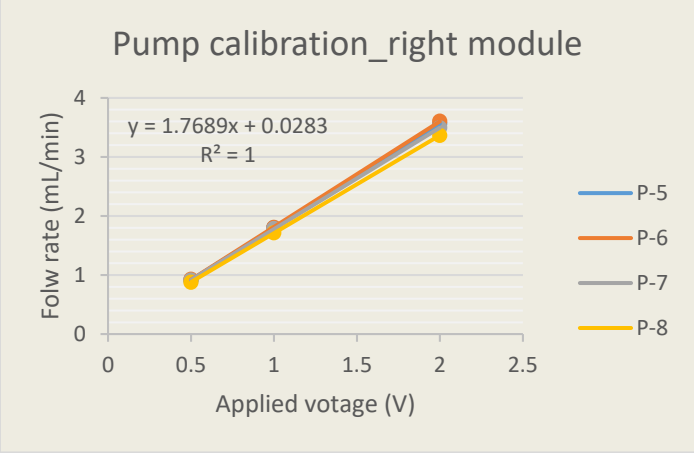
The ***ASRS-CS*** consists of three parts: Pump calibration mode, Calibration parameter calculation mode and Execution mode.

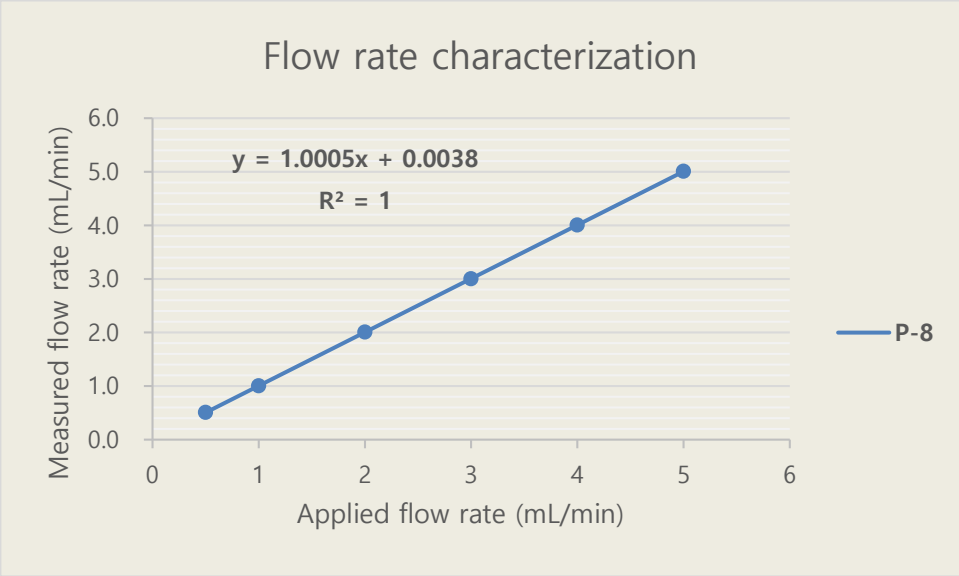
Input parameters: Flow rate, Volume, Reagent, Tandem column mode, column number, elution bottle selection

Characterization of the ASRS:
Pump Calibration of ASRS

- ✓ **Flow rate characterization of the ASRS: excellent precision, accuracy and linearity**
- ✓ **Measured by weighing the masses of deionized water**



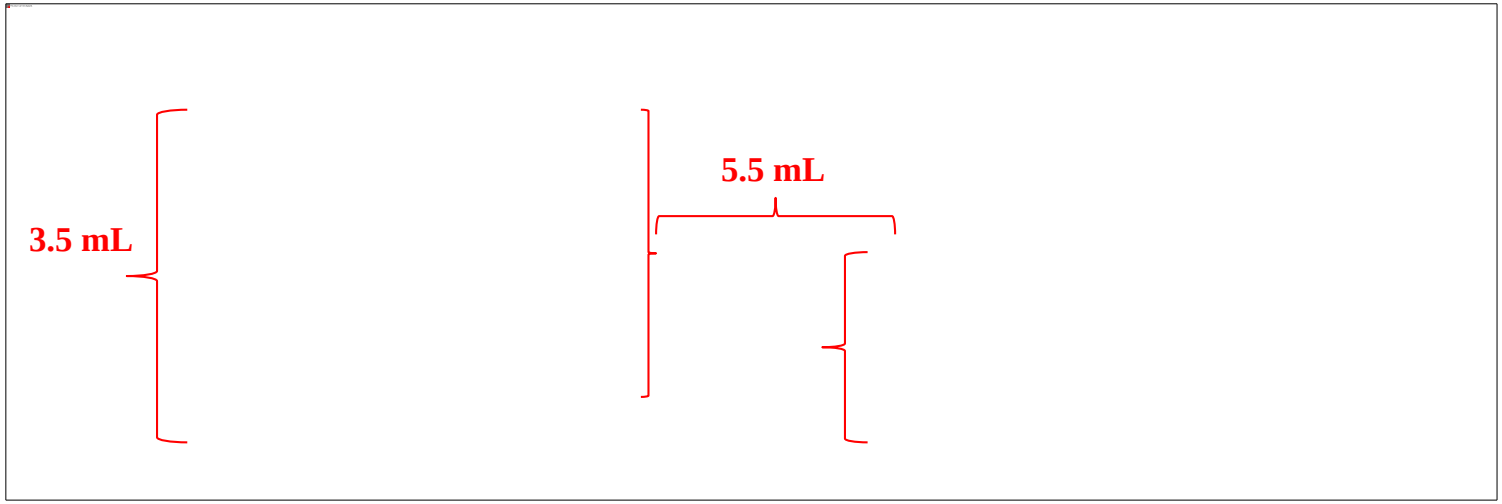


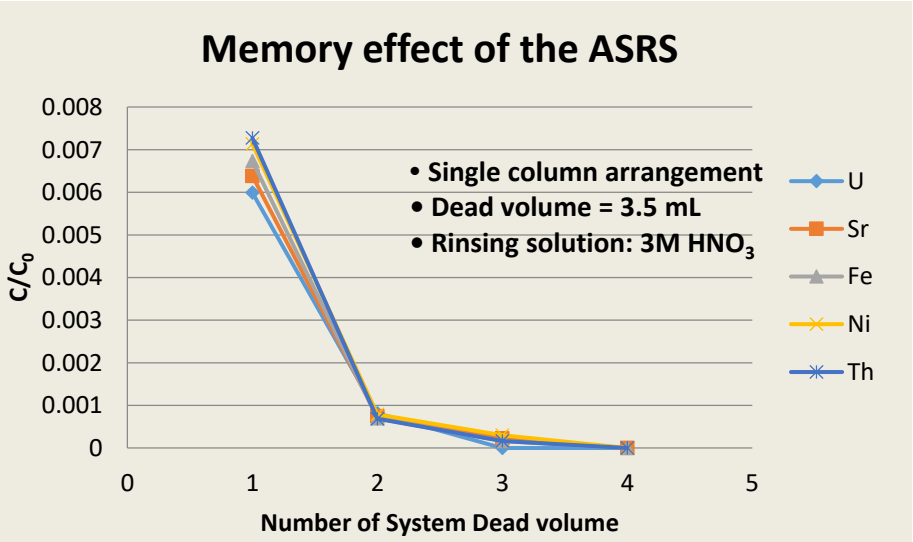


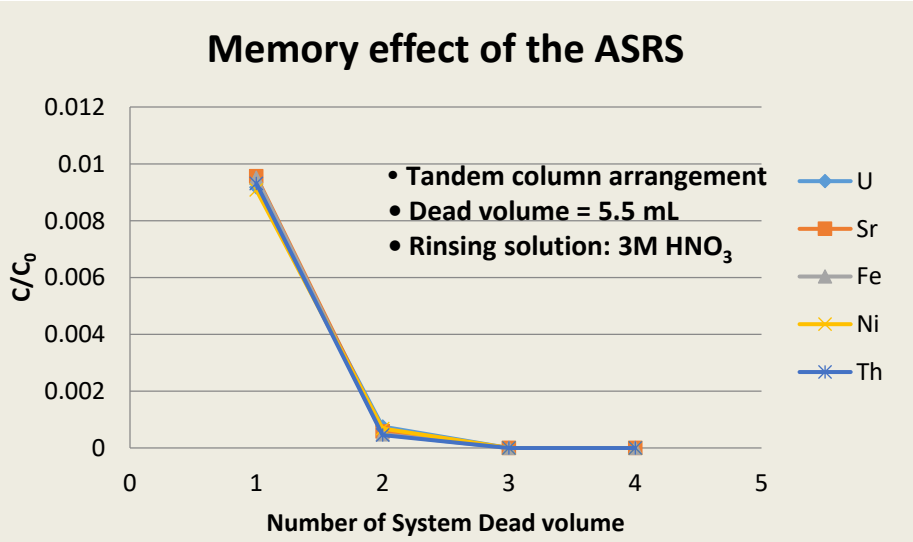
Pump-8	n=3		
Set flow rate (mL/min)	Measured flow rate (mL/min)	std	rsd (%)
0.5	0.5058	0.0010	0.2057
1	1.0035	0.0019	0.1921
2	2.0053	0.0093	0.4640
3	3.0030	0.0087	0.2885
4	4.0042	0.0097	0.2415
5	5.0087	0.0086	0.1710

Characterization of the ASRS:
System dead volume and memory effect of ASRS

✓ **Line tubing was completely washed by 4 dead volume with 3M HNO_3**

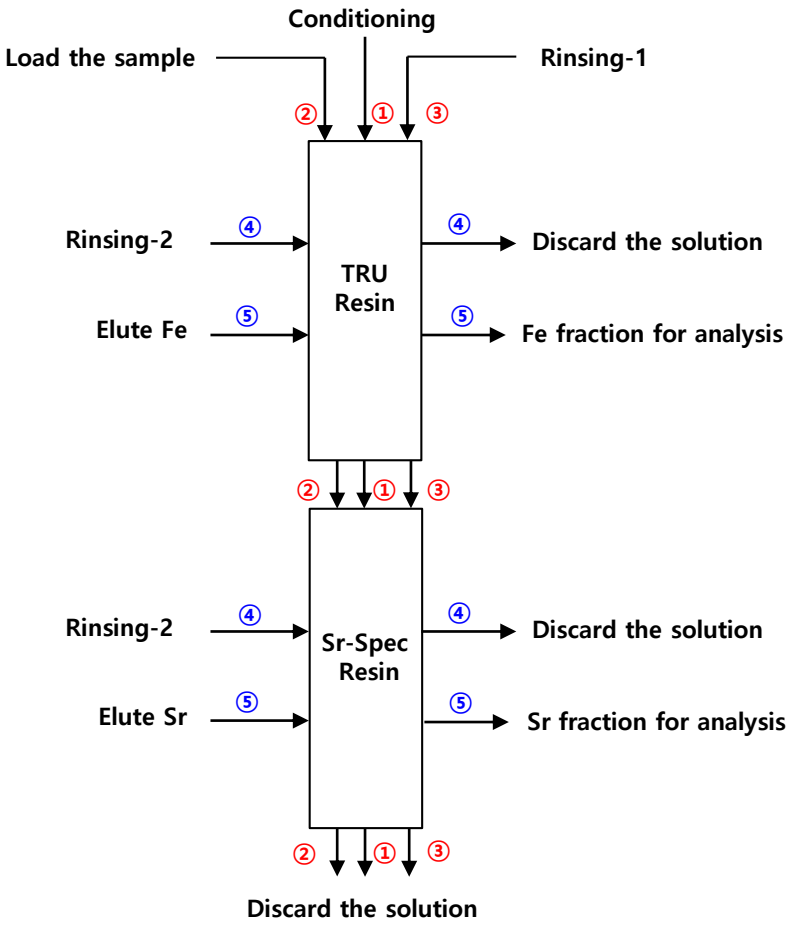




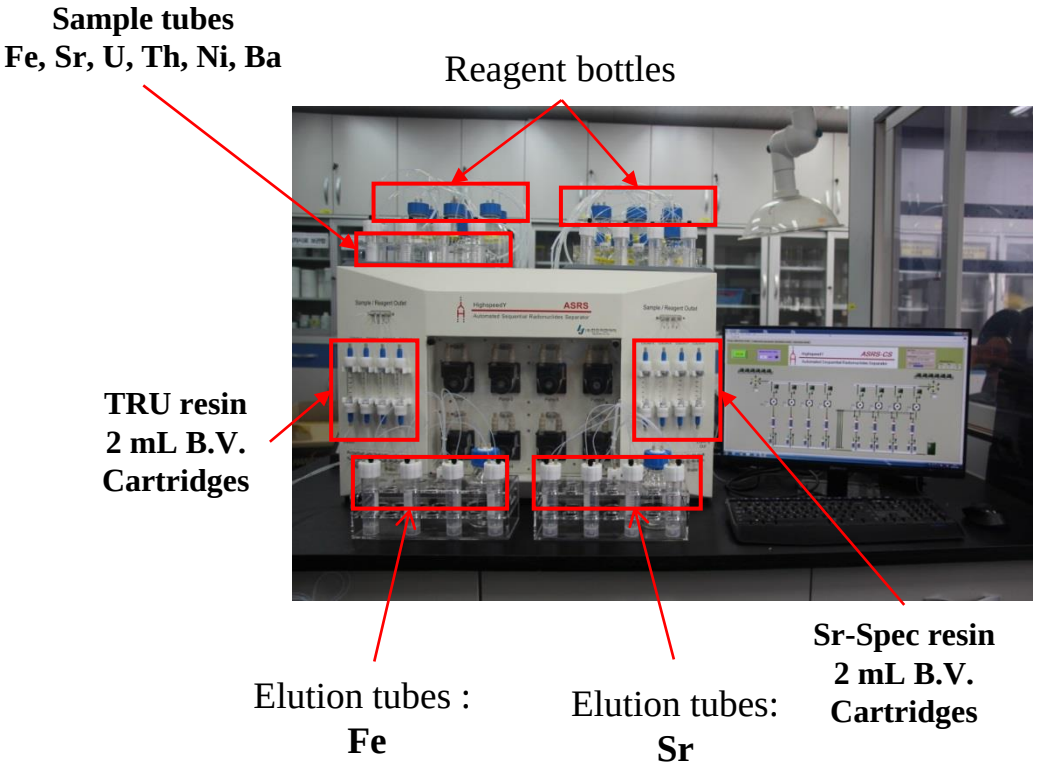


Optimization of an automated sequential radiochemical procedures:
Tandem column arrangement : TRU and Sr-spec resin

Sequential analysis of Fe & Sr in water: a tandem column arrangement



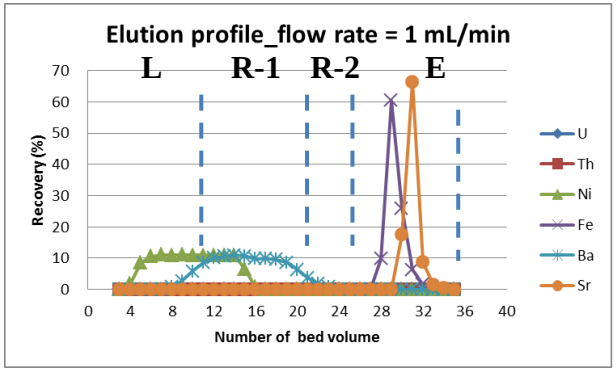
Column # 1~4

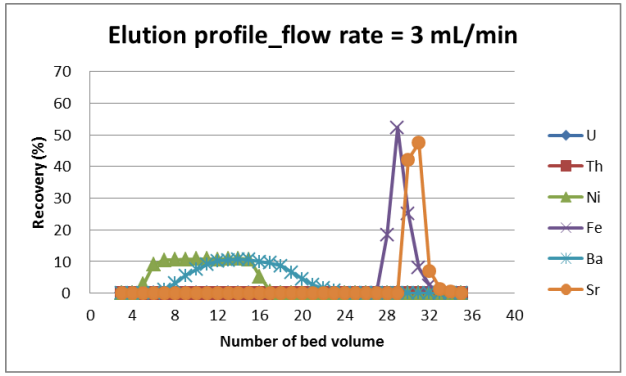


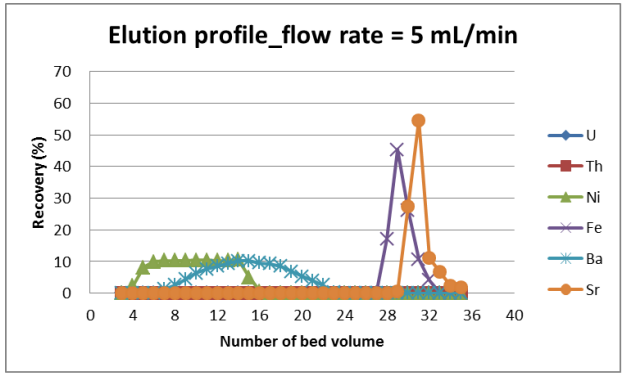
① Conditioning : 20 mL, 8M HNO_3
② Loading : 20 mL, Sample
③ Rinsing-1 : 20 mL, 8M HNO_3
④ Rinsing-2 : 6 mL, 8M HNO_3 — 6 mL, 8M HNO_3
⑤ Elution : 20 mL, 2M HNO_3 — 20 mL, D.I. water

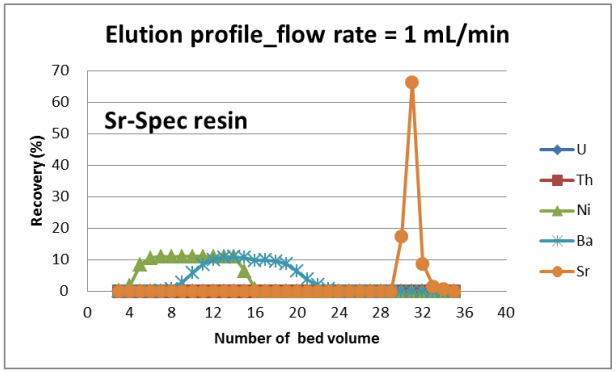
Optimization of an automated sequential radiochemical procedures:
Tandem column arrangement : TRU and Sr-spec resin

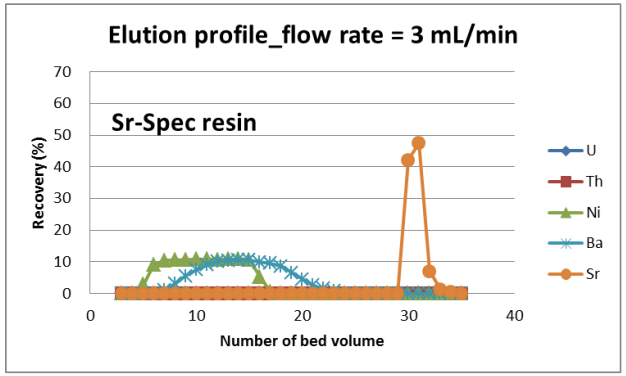
Flow rate effects on the method efficiency: separation of stable Fe & Sr, measured by ICP-OES

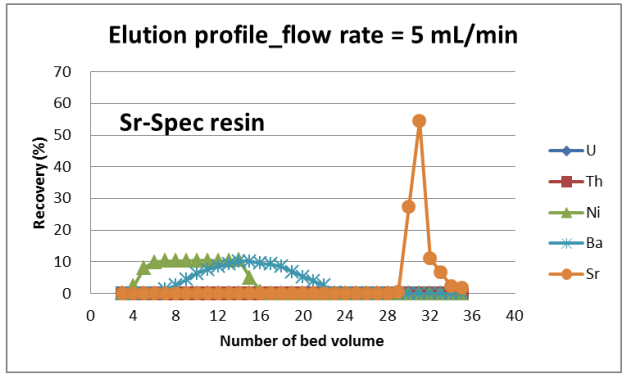


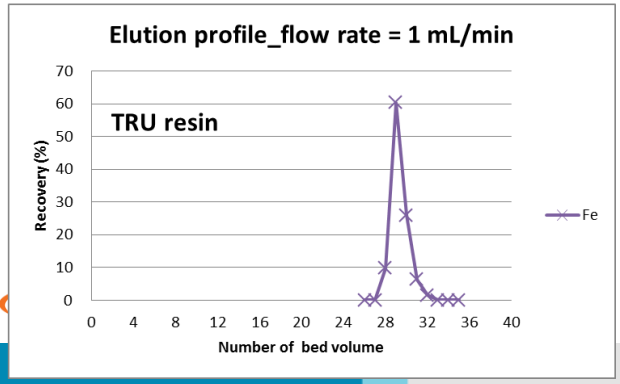


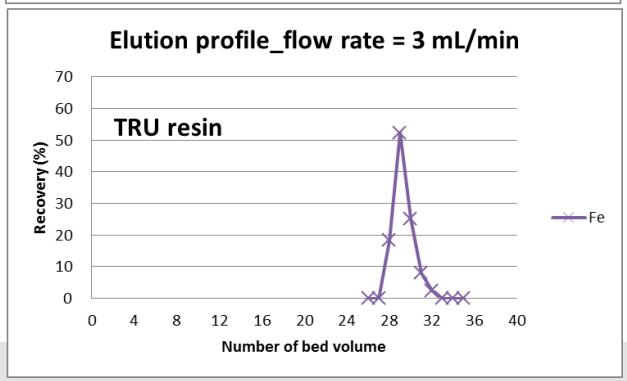


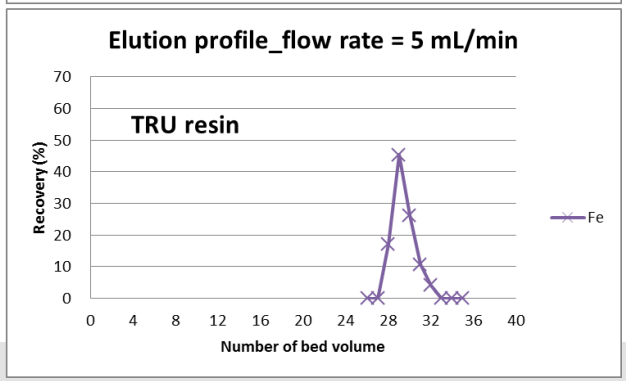






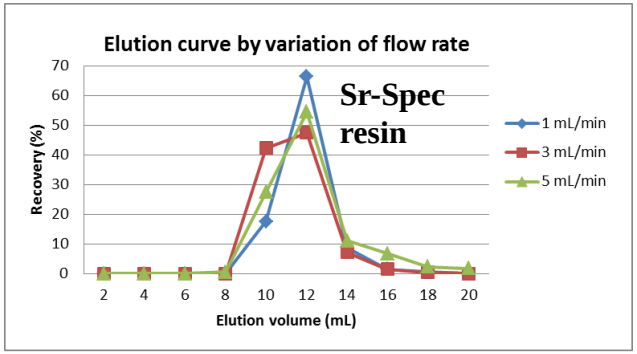


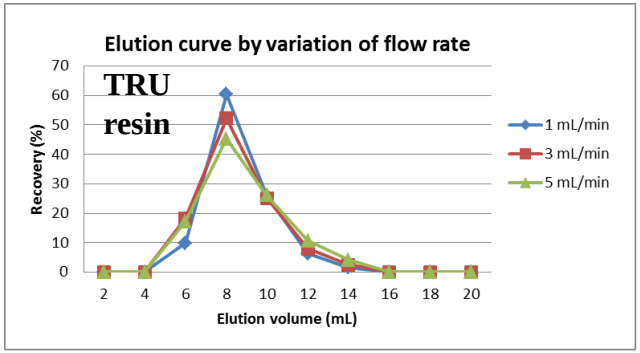


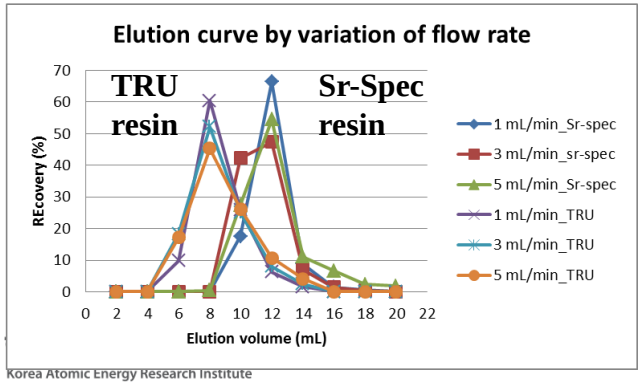


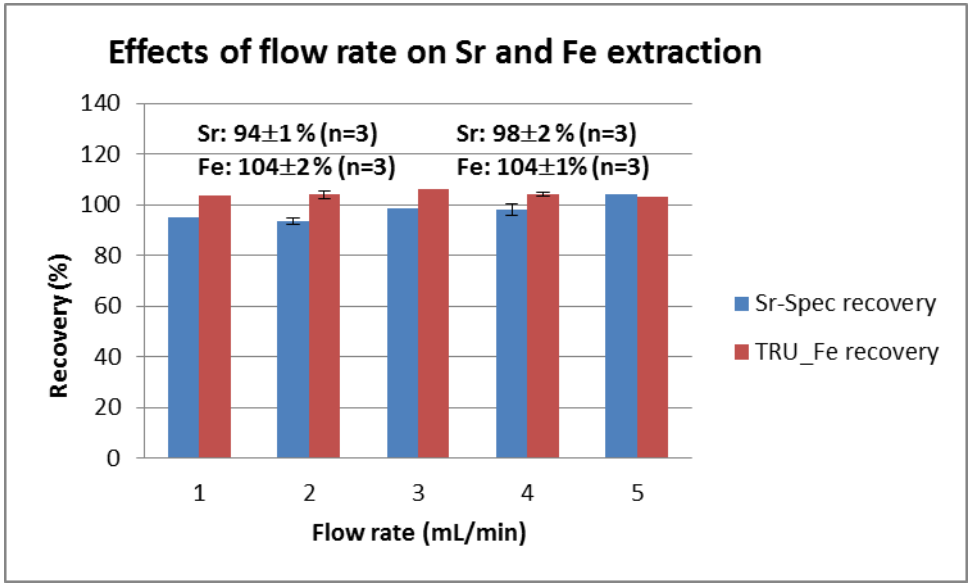
Optimization of an automated sequential radiochemical procedures:
Tandem column arrangement : TRU and Sr-spec resin

Flow rate effects on the method efficiency: separation of Fe & Sr









- ✓ Sr recoveries exceeded 94%
- ✓ Fe recoveries showed 100%
- ✓ Over a wide range flow rate (1~5 mL/min)
- ✓ Highly reproducible (r.s.d. : less than 2%)
- ✓ In-house column packing: 2 mL dry pack cartridge
- ✓ Sufficient removal of interferences

Application of the *ASRS*: Determination of ^{55}Fe and ^{90}Sr by ASRS and LSC

Sample tubes

- ✓ Fe, Sr, U, Th, Ni, Ba: ~ 1mg
- ✓ ^{55}Fe : 13.99 Bq
- ✓ ^{90}Sr : 13.60 Bq

① Conditioning	: 20 mL, 8M HNO_3	
② Loading	: 20 mL, Sample	
③ Rinsing-1	: 20 mL, 8M HNO_3	
④ Rinsing-2	: 6 mL, 8M HNO_3	6 mL, 8M HNO_3
⑤ Elution	: 20 mL, 2M HNO_3	20 mL, D.I. water

- ✓ Tandem column mode
- ✓ TRU resin: 2mL B.V.
- ✓ Sr-spec resin: 2 mL B.V.
- ✓ Flow rate: 3 mL/min
- ✓ Separation time: 29 min

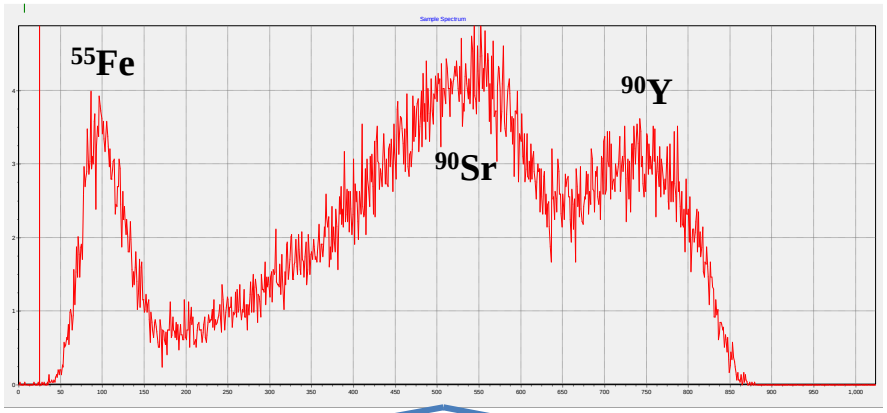
Application of the *ASRS*: Determination of ^{55}Fe and ^{90}Sr by ASRS and LSC

Performance test of *ASRS* : simultaneous measurement of ^{55}Fe & ^{90}Sr in spiked sample

LSC Samples

- ✓ ^{55}Fe :
- ✓ 1 mL 2M H_3PO_4
- ✓ 2 mL D.I. water
- ✓ 15 mL Ultima Gold AB

- ✓ ^{90}Sr :
- ✓ 10 mL 0.1M HNO_3
- ✓ 10 mL Ultima Gold AB



Sample tubes

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- ✓ TRU resin: 2mL B.V.
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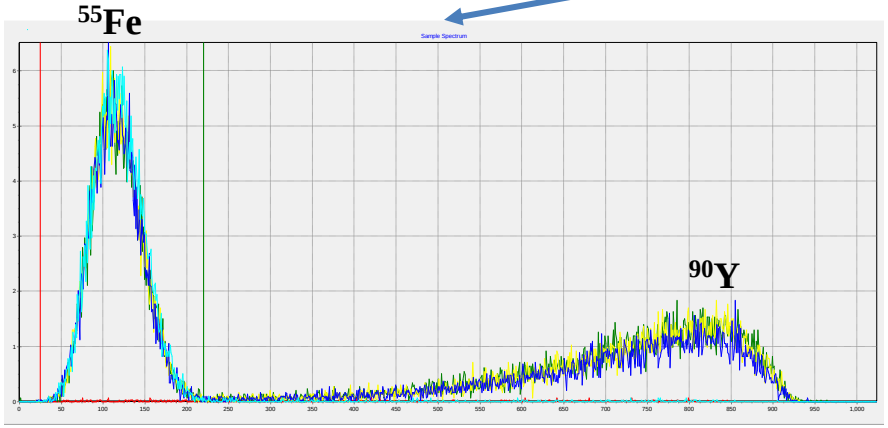

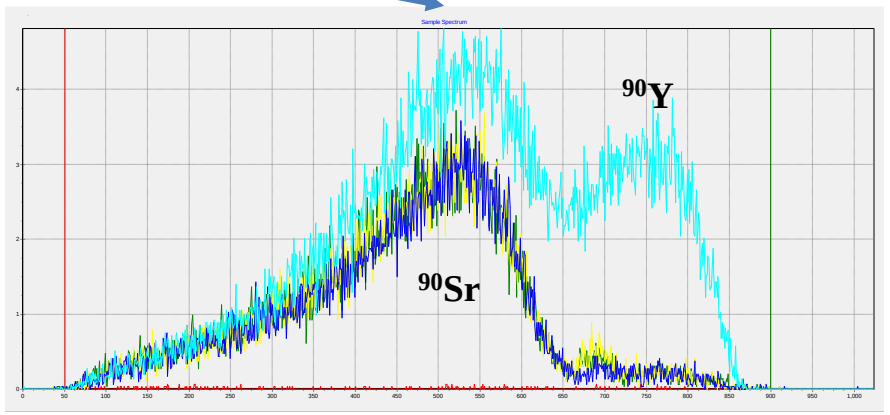

(n=3)	Chemical yield (%)	Activity	Relative vias (%)
Mean	92.4	14.11	0.8
std	6.6	1.14	
rsd(%)	7.2	8.05	

(n=3)	Chemical yield (%)	Activity	Relative vias (%)
Mean	91.3	14.00	2.9
std	1.4	0.59	
rsd(%)	1.6	4.21	

Closing Remarks: Automated Radiochemical Separation!!!

- *ASRS*: fully automated column based extraction chromatography
- Flowrate characterization: excellent accuracy, precision and linearity, minimized flow difference between the 8 pumps (RSD: < 1%)
- System memory effect: washing the tubing with 4 dead volume
- Reliable and reproducible : recovery (>94%), relative bias(<3%), RSD (< 2%, n=3)
- High-throuput: simultaneous 4 samples, up to 5 RNs, within 1 hour
- Rapid and safe : compared to conventional methods
- Less labour-intensive : fully automated radiochemical procedures
- Powerful and convenient tool: an automated chemical separation and purification of RNs at trace levels in environmental samples
- Suitable for a routine and an emergency responce: Rapid, reliable, high-throughput

Thank you

for your attention !