International Seminar on Interaction of Neutrons with Nuclei: Fundamental Interactions & Neutrons, Nuclear Structure, Ultracold Neutrons, Related Topics (ISINN-30), 14-18 April 2024, Sharm El-Sheikh, Egypt.

# Innovative neutron activation approach for analysis of large liquid samples based on short-lived radionuclides


**Mohamed Soliman, Ph.D.**

Head of NAA laboratory

Member of $k_0$-NAA International Scientific Committee ($k_0$-ISC)

Egypt Second Research Reactor (ETRR-2),
Egyptian Atomic Energy Authority (EAEA).
E-mail: soliman.ma@gmail.com

Egyptian Atomic Energy Authority

# Introduction

Neutron Activation Analysis (NAA) is a mature multi-elemental analytical technique know for its excellent accuracy and precision, and good limits of detection.

But……

It is not effective for analysis of liquid and water samples because……..

i. Radiolysis of water molecules causes pressure build-up problem during irradiation – Safety issue

ii. Natural water samples have relatively low elemental content – analysis of small sample is not effective.

Several solutions have been proposed:

i. Chemical pre-concentration method

ii. Analysis of large volume – combined with radiochemical process

# Introduction (cont'd)

## Limitations of the existed NAA methods for analysis of liquid samples

1. Chemical pre-concentration method

- Costly and time consuming,
- Losses of elements may be occurred, and/or
- Cross-contamination problems.

2. Analysis of large volume

- Almost all reactors aren't equipped with large volume irradiation facility.
- Not suitable for analysis of elements via short lived isotopes
- Radiolysis of water molecules - pressure build-up problems

# To overcome these issues…

In 2011, we started to design the new set-up: Flowing Sample Neutron Activation Analysis (FSNAA).

(My Ph.D. Thesis experimental works "Enhancements and Health Related Studies of Neutron Activation Analysis Technique", 2012, Ain Shams University, Egypt)

It involves the continuous flowing (pumping) of the sample between an irradiation cell (Cf-252) and measurement station (HPGe).

It can be operated in 2 modes: cyclic & one-way

Expected advantages:

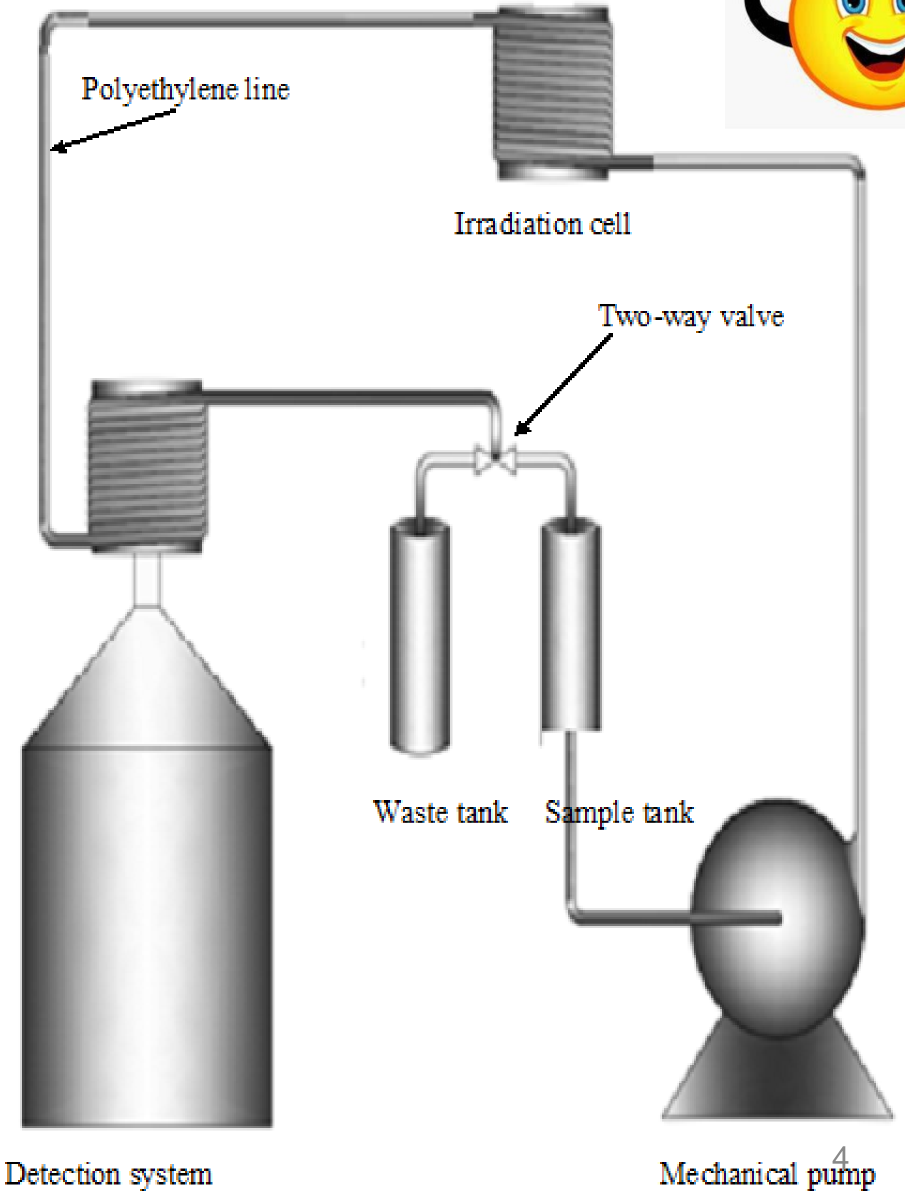
No pretreatment/post irradiation processes

It allows analysis of large sample – improving LDs

No pressure build-up problem

It allows measurement of short lived radionuclides

It keeps a constant dead time – more accurate



# FSNAA…
# What we did…….

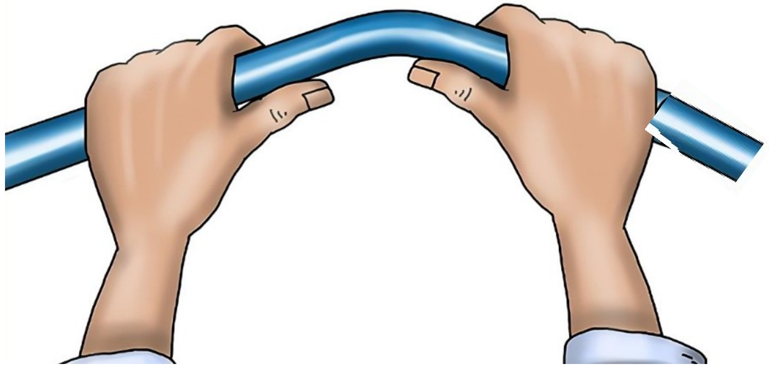
- Selection of tube material
- Testing and characterizing
  - Leakage test
  - Pump (flow rate and its stability)
  - Repeatability
  - Accuracy
  - Stability of dead time
  - Efficiency calibration
  - etc….
- Optimizing the counting and irradiation configurations using Mote Carlo simulations – Tube diameter
- Installation of the FSNAA in a reactor
  - Horizontal neutron beam neutron @ ETRR-2 – flux ~ $10^7$ $cm^{-2}s^{-1}$.
  - Slant Irradiation tube @ Kyoto University Research Reactor – flux, 4.82 x $10^{12}$ $cm^{-2}s^{-1}$
- Applications

# FSNAA…
# What we did…….

- Selection of tube material

Durability test

Irradiation of polyethylene, TYGON, and Ethylene tetrafluoroethylene tubes for different times (max. 4 hr @ flux: $5 \times 10^{13}$ $cm^{-2}s^{-1}$)followed by simple bending test.



TYGON is the best tube

- Leakage test
  - Cold run with deionized water & without neutrons
  - Hot run with deionized water (neutron irradiation) – check for $^{41}Ar$ and $^{16}N$ radioactivity

# FSNAA…
# What we did…….

- □ System repeatability

Fifteen measurements of Indium solutions ($^{116m2}In$, $t_{1/2}$=2.2sec)

- ➢ Repeatability<3%

- □ Analysis of solutions containing suspended matters

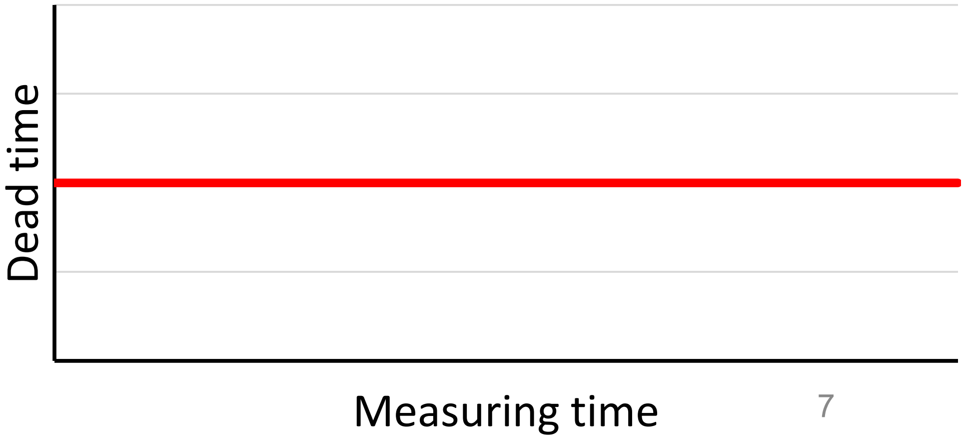
Analysis of $AgNO_3$ (soluble) # $AgCl_3$ (suspended)

- ➢ No significant difference between count obtained with dissolved and precipitated $^{110}Ag$ (24 sec)

- □ Stability of dead time

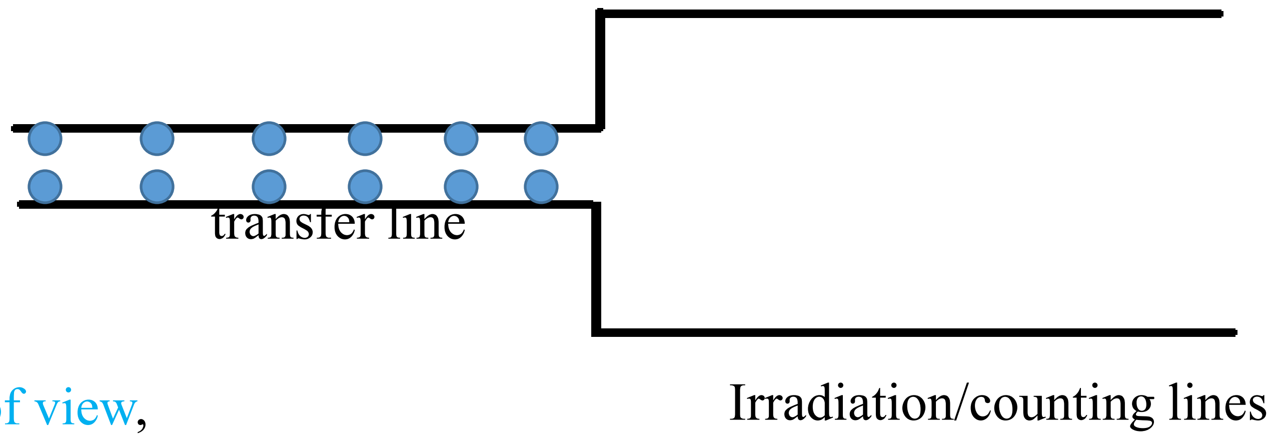
Measuring In solution (In-116m2 (2.2 sec))

- ➢ The pump continually feed the detector with radioactive sample maintaining a constant dead time



# FSNAA…
# What we did…….

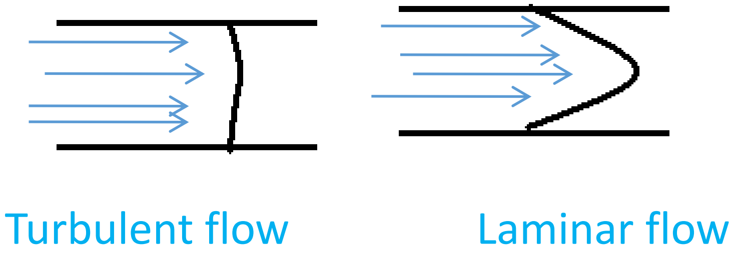
## Optimization irradiation/counting geometries




From fluid mechanics point of view,

- Extreme difference in tube diameters leads to an increase in the amount of eddies
- Tube diameter controls the type of flow: Laminar or Turbulent

Turbulent flow is preferred over Laminar one because it guarantees a homogenous irradiation and counting of the irradiated samples



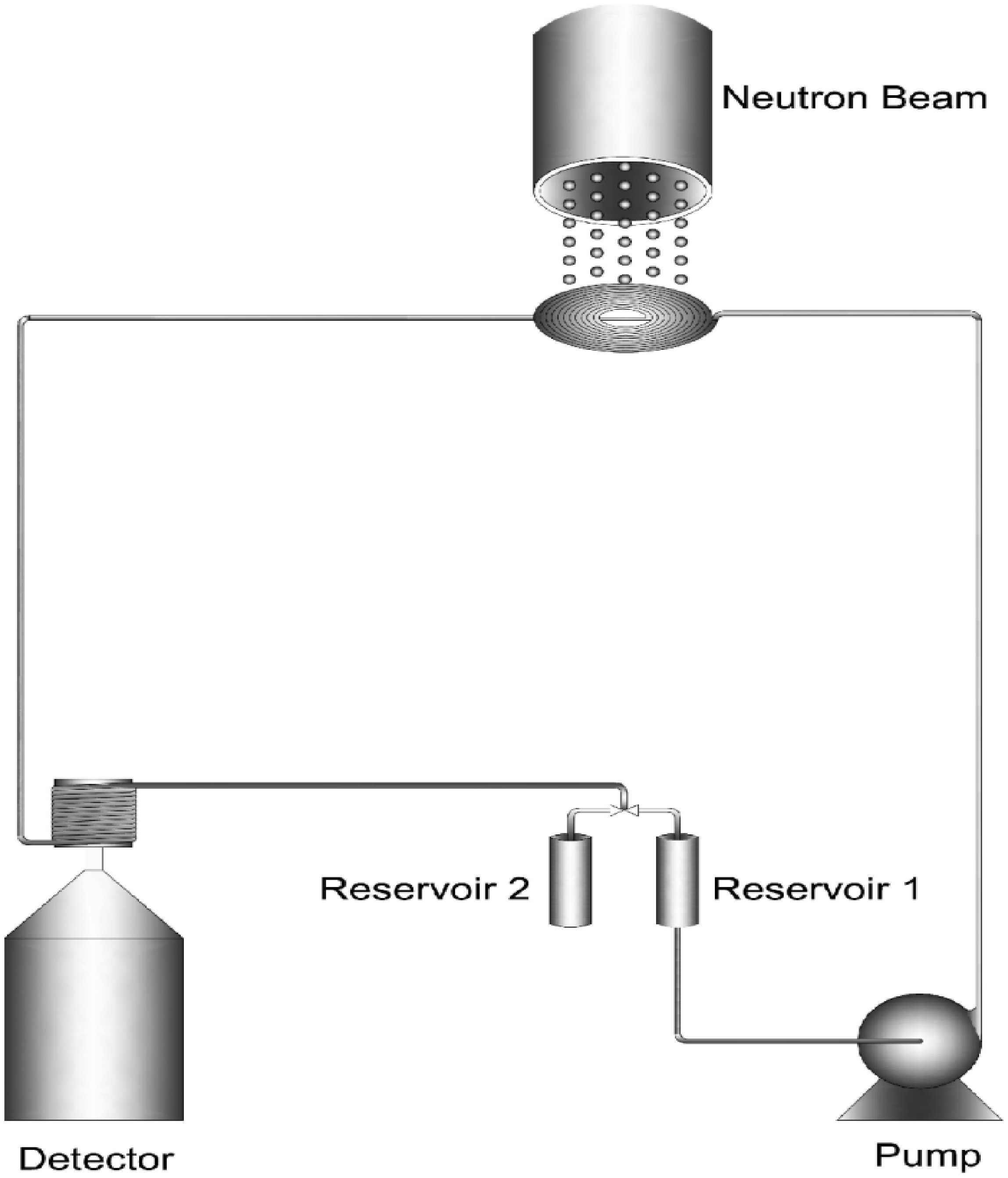
# FSNAA…
# What we did…….

## Installation in ETRR-2

In 2015, Installation of the FSNAA at the neutron radiography beam.

Neutron flux: ~ $10^7$ $cm^{-2}.s^{-1}$

Ph.D. Thesis by Fatma Abdo " Factors Affecting the Performance of Flowing Sample Neutron Activation Analysis", 2016, Helwan University, Egypt.



# FSNAA…
# What we get…….

- Applications

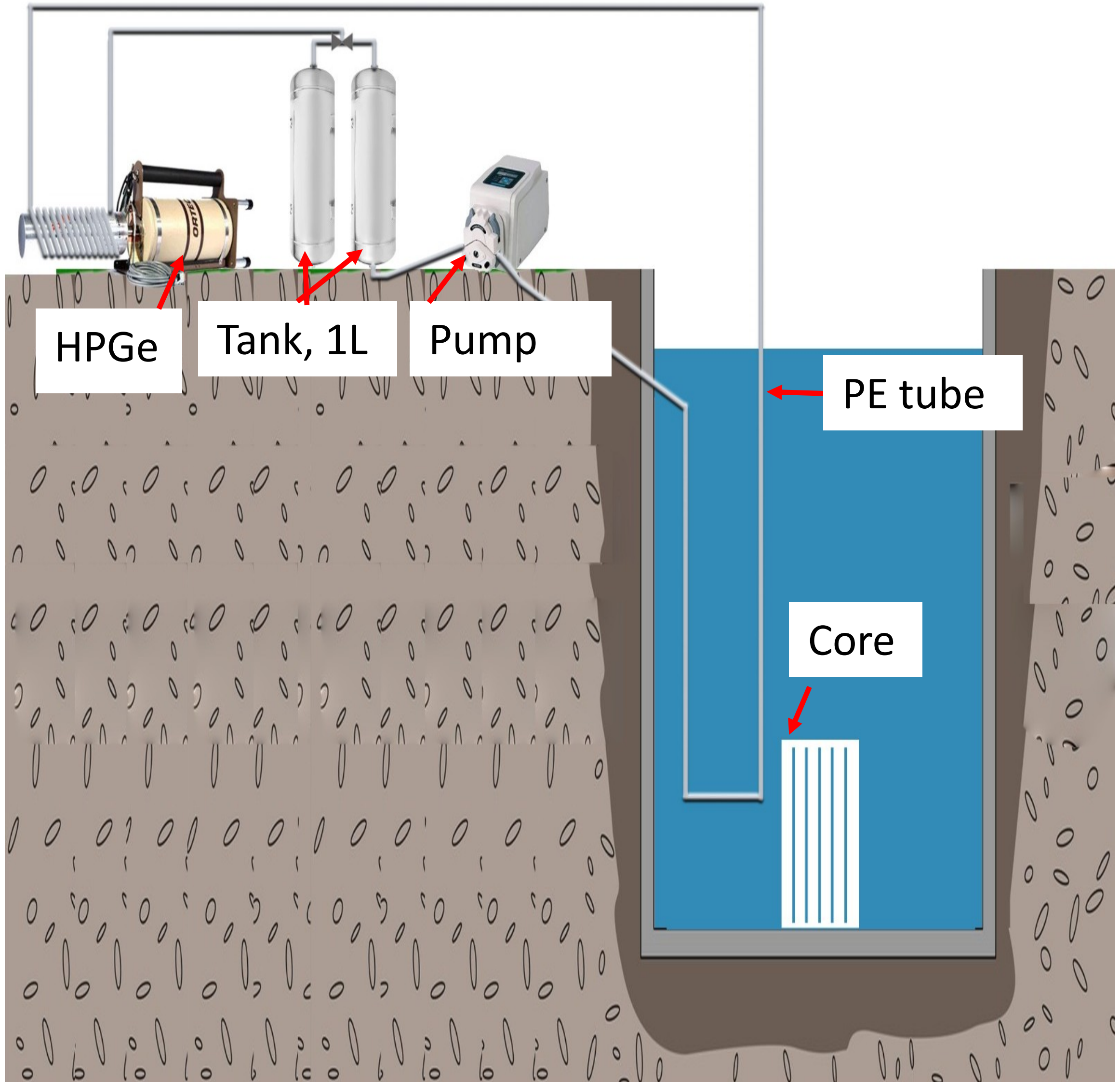
| Sample type | Detected radionuclides | Neutron source |
|---|---|---|
| Synthetic multi-elements standard | $^{116m2}$In (2.2 s), $^{110}$Ag (24 s), $^{165m}$Dy (75 s), $^{52}$V (3.75 m), $^{56}$Mn (2.58 h), $^{20}$F (11 s), and $^{77m}$Se (17 s) | Cf-252 and ETRR-2 |
| Sea water | $^{24}$Na (15 h), $^{38}$Cl (37 m), $^{42}$K (12 h), and $^{66}$Cu (5 m) | Cf-252 |
| Ismailia Cannel (near $Al_2(SO_4)_3$ factory) | $^{28}$Al (2.24 m) | ETRR-2 |
| Suspension of IAEA-soil-7 | $^{28}$Al (2.24 m) | ETRR-2 |
| Milk sample | $^{49}$Ca (8.7 m) | ETRR-2 |

Poor limits of detection

# FSNAA…
# What we did…….

## Installation in Kyoto University Research Reactor

In 2019, Installation of the FSNAA at the slant irradiation tube, flux: 4.5 x $10^{12}$ $cm^{-2}s^{-1}$




Safety considerations

1. Radiation durability of the tubing materials:

   - Irradiation of polyethylene, TYGON, and Ethylene tetrafluoroethylene tubes for different times followed by simple bending test.

2. HPGe:

   - n-type HPGe was covered with Cd-sheet to avoid any possible damage caused by neutrons

3. Leakage test

   - Cold run with deionized water (reactor shutdown)

   - Hot run with deionized water (reactor in operation) - $^{41}Ar$ and $^{16}N$ radioactivity.

# FSNAA…
# What we did…….

## Installation in Kyoto University Research Reactor

### Experimental steps:

1- Background was recorded.

2- With de-ionized water:

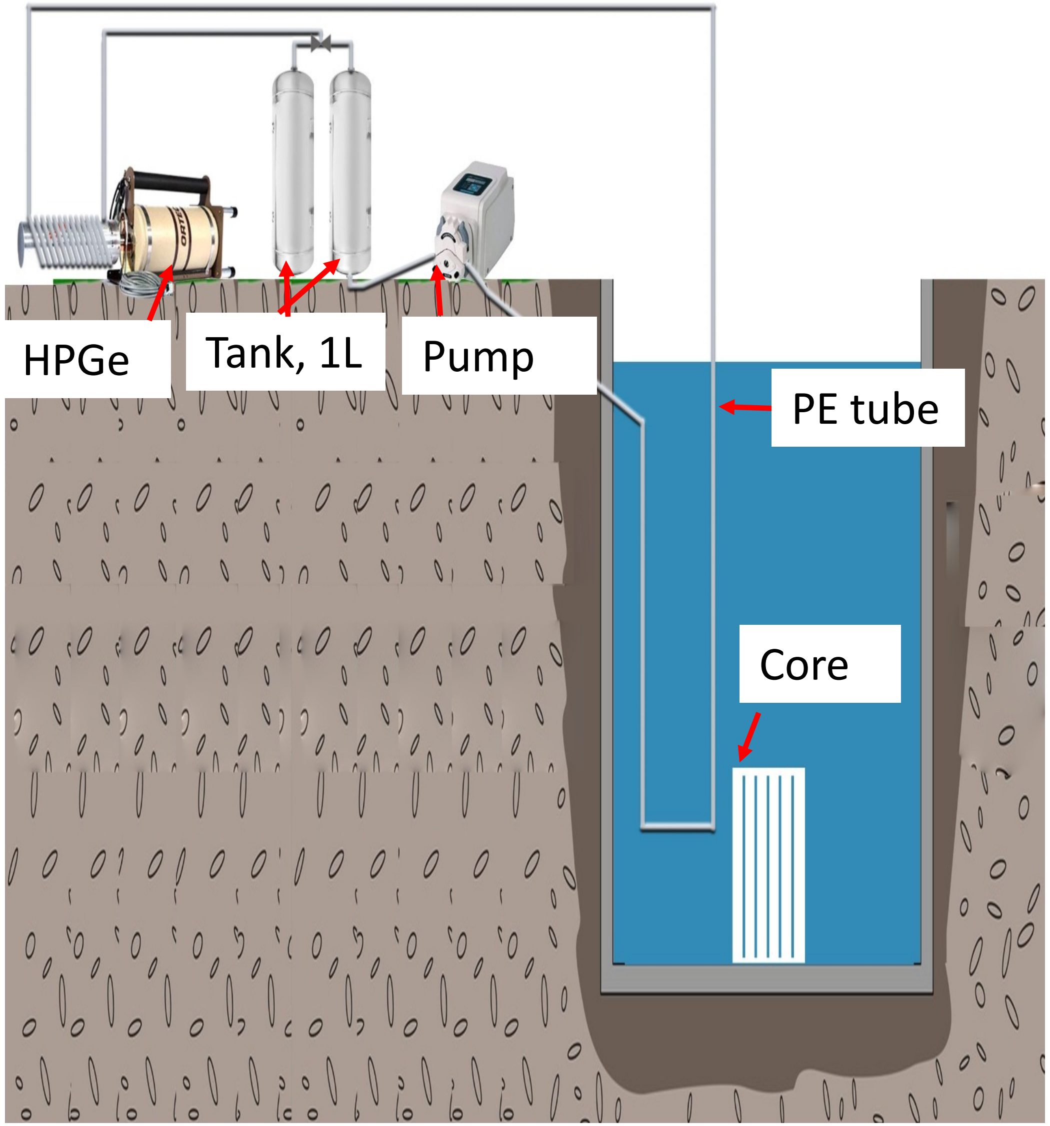
- $^{16}N$ ($^{16}O(n, p)$ $^{16}N$) was not detected- well thermalized neutron.
- Purging $N_2$ gas into samples minimizes Ar-41.
- Calibration of pump flow-rate

3- Mixed standard solution was prepared for calibration – relative method (Na, Al, Ca, Cu, K, Mg, Mn, S, and V).

4- Rain, river and tap water samples were collected.

5- Sample irradiation/counting

6- Washing between samples



# FSNAA…
# What we did…….

## Installation in Kyoto University Research Reactor

## Results

Excellent limits of detection for heavy elements

| | Tap water | | River water | |
|---|---|---|---|---|
| Element | mg/l ± % | DL | mg/l ± % | DL |
| Al | 0.255 ± 6 | 0.0022 | 0.240 ± 1.3 | 0.002 |
| Ca | 10.1 ± 4 | 0.280 | 15.9 ± 2.5 | 0.240 |
| Cu | 0.028 ± 35 | 0.022 | ND | |
| K | 3.35 ± 23 | 0.980 | 2.935 ± 25 | 1.180 |
| Mg | 1.300 ± 5 | 0.135 | 3.720 ± 3 | 0.200 |
| Mn | 0.0017 ± 30 | 0.001 | 0.132 ± 2 | 0.002 |
| Na | 9.700 ± 2 | 0.110 | 8.800 ± 2 | 0.105 |
| S | 8.000 ± 25 | 0.740 | 10.500 ± 20 | 6.600 |
| V | 0.0004 ± 14 | 0.0002 | 0.00037 ± 15 | 0.0002 |
| O ($^{18}O$) | Qualitative – no available standard | | | |
| Cl | | | | |
| Br | | | | |
| I | | | | |

$^{18}O$ is important in:

- isotope hydrology
- climate change studies

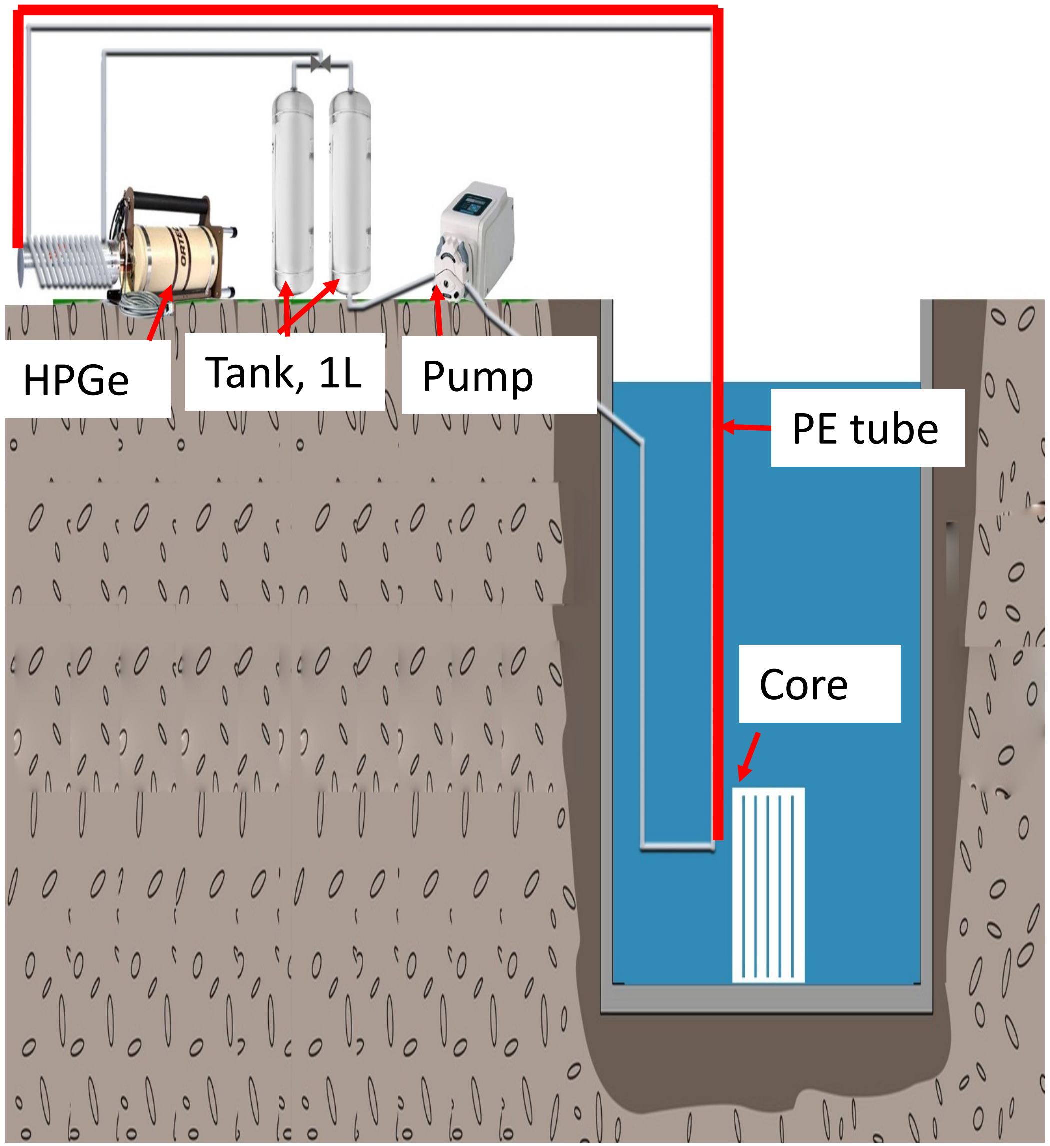
Halogens are important for water quality

# FSNAA…
# We can get more…….

Irradiation-to-HPGe transfer time was 3 min.

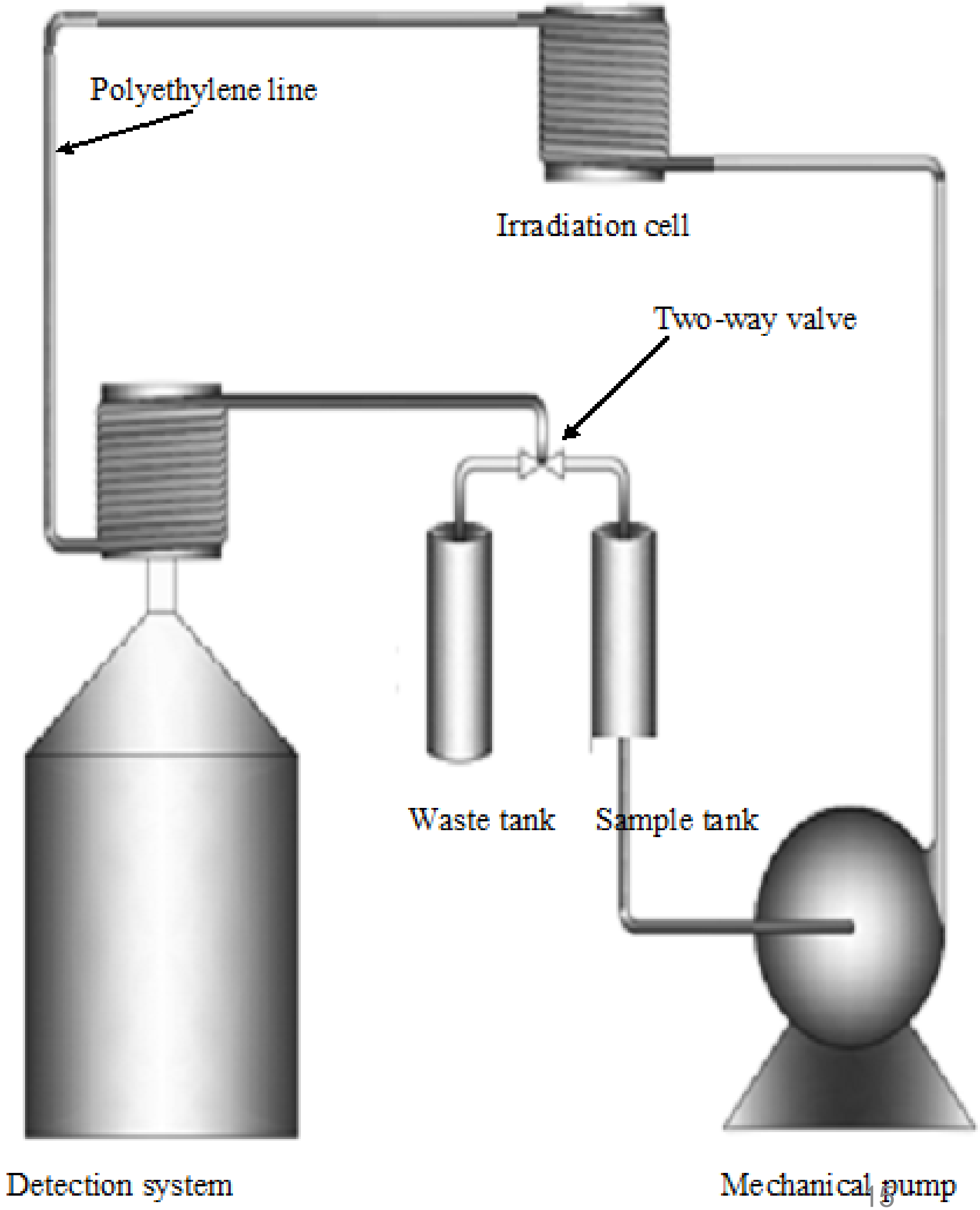
Use of a pump with higher flow-rate will shorten transfer time, leading to:

- ✓ Better limits of detection.
- ✓ Increasing the scope of the technique with more shorter haf-lives radionuclides e.g $^{20}$F(11 s), $^{46m}$Sc (18 s), $^{77m}$Se (17 s)



# FSNAA…
# Conclusions

- ❑Simple and non-destructive
  - ➢ No pre-treatment and/or radio-chemical process
- ❑Analysis of large volume
  - ➢ Excellent limits of detection
  - ➢ No pressure build-up
  - ➢ Measuring of short-lived isotopes
  - ➢ Can be installed with the currently available irradiation sites
- ❑Analysis of solutions contained suspended matters
  - ➢ No need for filtration and/or dissolution steps
- ❑Constant dead time
  - ➢ More accurate results
- ❑No blank correction



# FSNAA…
# Future Plan…….

- Installation at one of the irradiation sites inside the ETRR-2 reactor tank.
- Studying the motion of the suspended matters inside the solution and its effect on the accuracy of the results
- Implementation of $k_0$-method.
- More applications with real samples

# Thank you

# شكراً