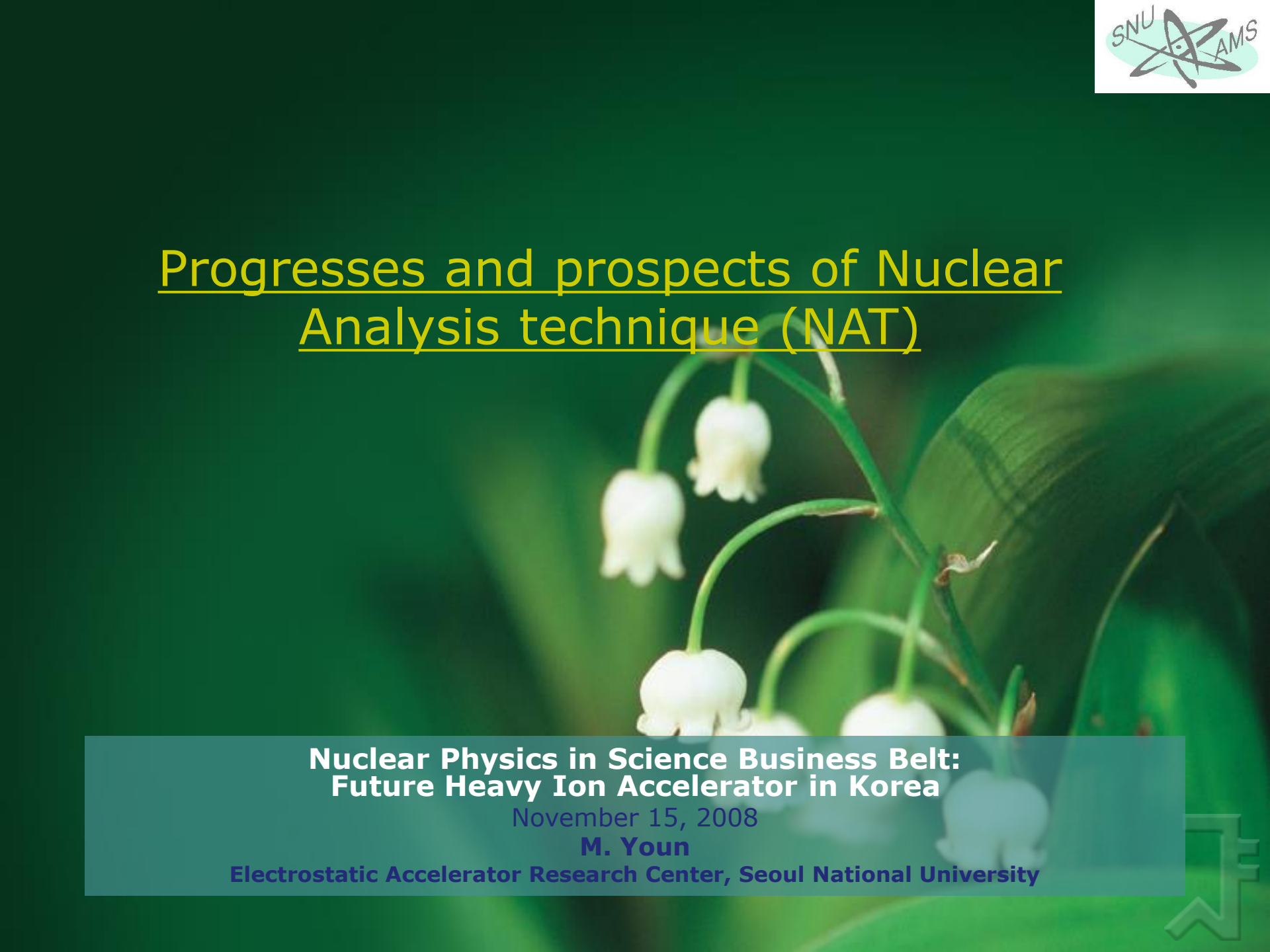
Progresses and prospects of Nuclear Analysis technique (NAT)



Nuclear Physics in Science Business Belt:
Future Heavy Ion Accelerator in Korea

November 15, 2008

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Electrostatic Accelerator Research Center, Seoul National University

Contents

I. Introduction

1. NAT (Nuclear-atomic analysis technique)

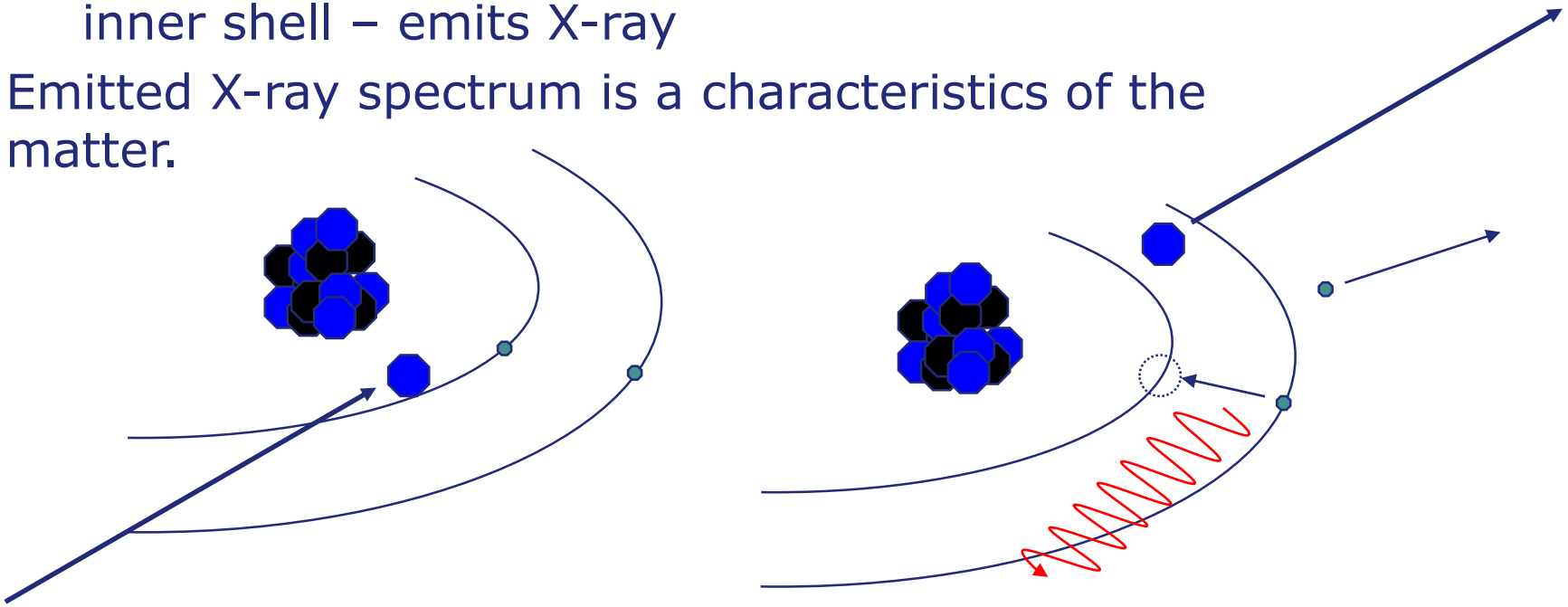
- Elemental or chemical analysis using radiation
- Useful radiations
  - ionbeam(neutron, proton, heavy-ion)
  - β-ray
  - γ-ray & x-ray
  - LASER
  - UV

  } Elemental analysis

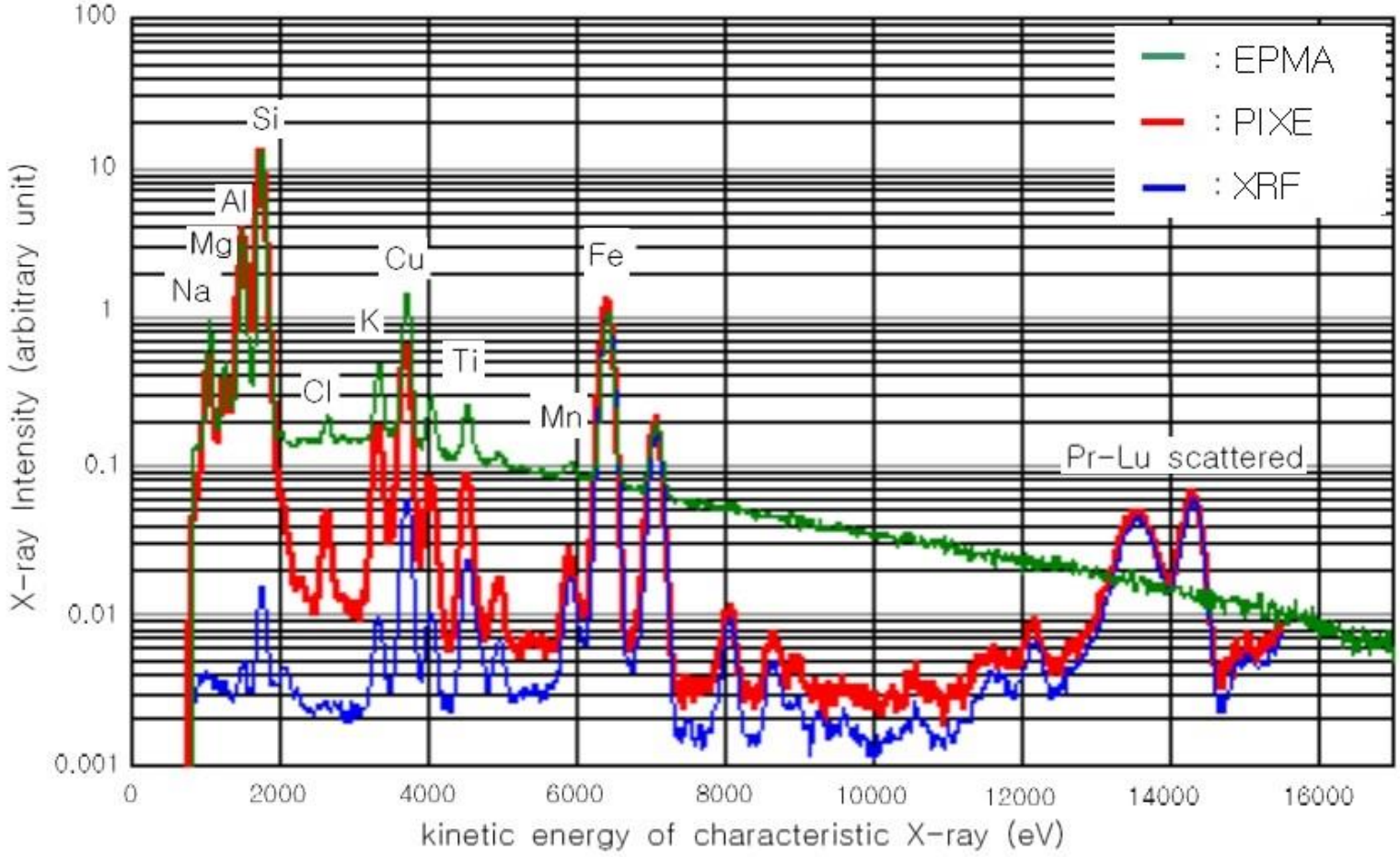
  } Chemical analysis
- Dedicated devices have been developed
  - Standalone devices : SEM, TEM, Raman spectroscopy, EPMA, ICP-MS, XRF, ····
  - Bigger size facility for nuclear radiation usage (PIXE, PIGmE, NAA, ····)
- Can be applied to various fields of human activity
- Present talk will concentrate on PIXE

2. PIXE(양성자유발 X선 방출)

- **PIXE**(proton induced X-ray emission)
  cf. **PIGmE**(proton induced gamma-ray emission)
- Model for underlying process
  - Accelerated proton kicks out electron in matter – creates a hole state
  - Outer shell orbital electron transits to the hole at inner shell – emits X-ray
- Emitted X-ray spectrum is a characteristics of the matter.



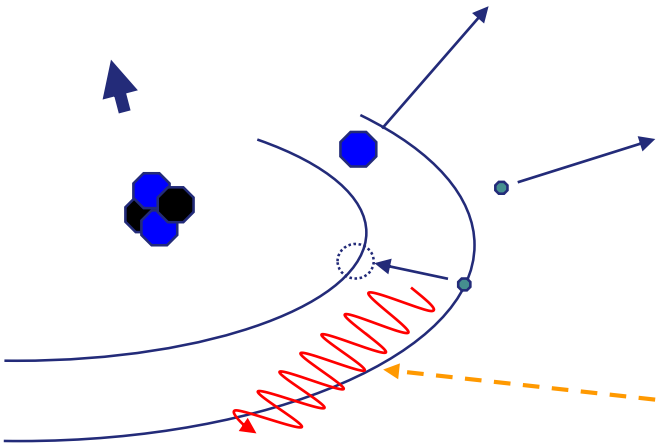
Typical PIXE spectrum



PIXE spectrum of a rock sample from MARS
(University of Guelph, Canada)

Limitations for PIXE

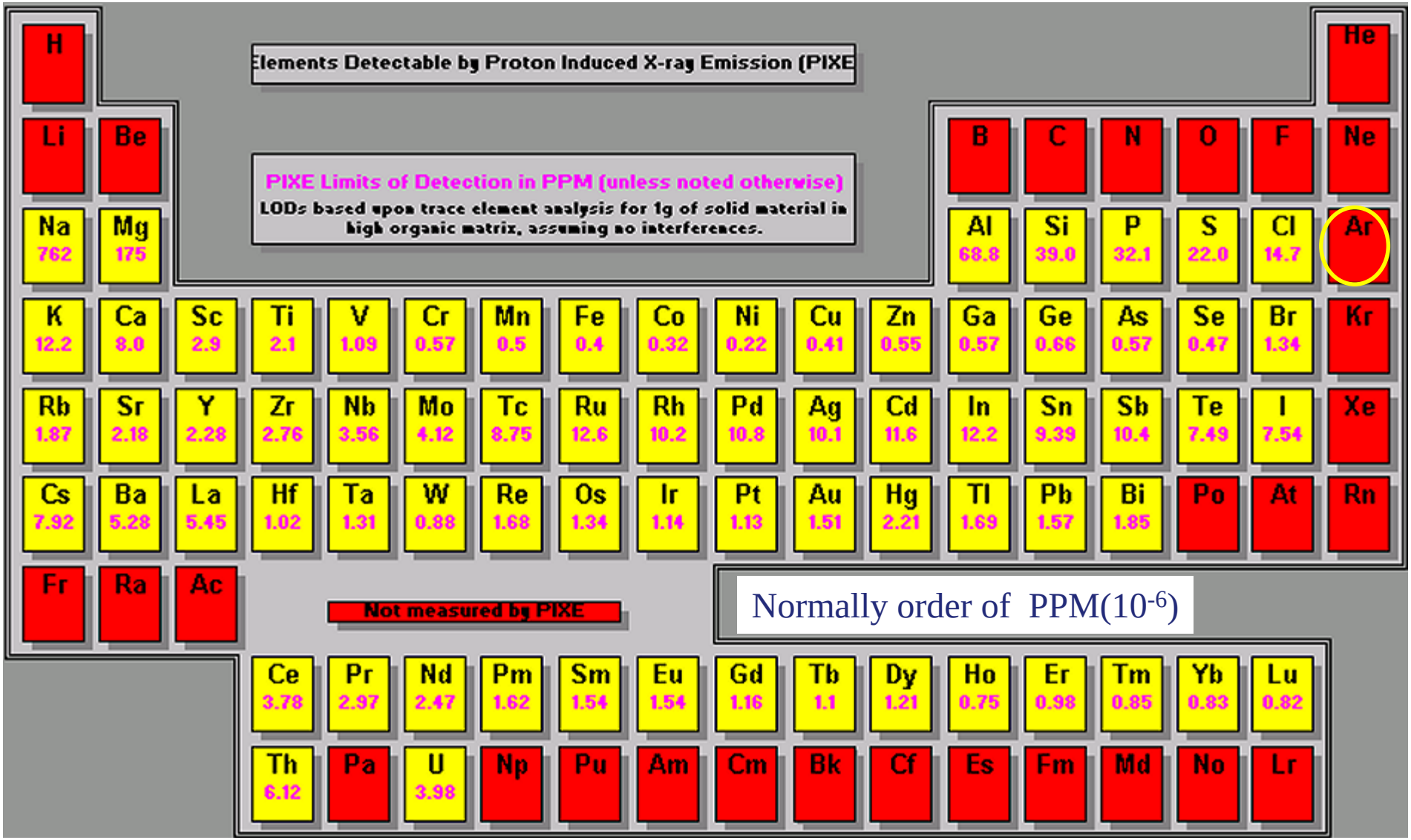
- Atomic recoil – Doppler broadening of X-ray for light atoms



For light nucleus, the atomic recoil results to a significant Doppler broadening

- Inert gases: closed orbital shell suppresses the orbital transition, very low X-ray emission probability (not always)
- Unstable nuclei can not last during PIXE process
- Traditional PIXE measurement : in vacuum
  - Due to ion-beam transport
  - Limits applicable sample material

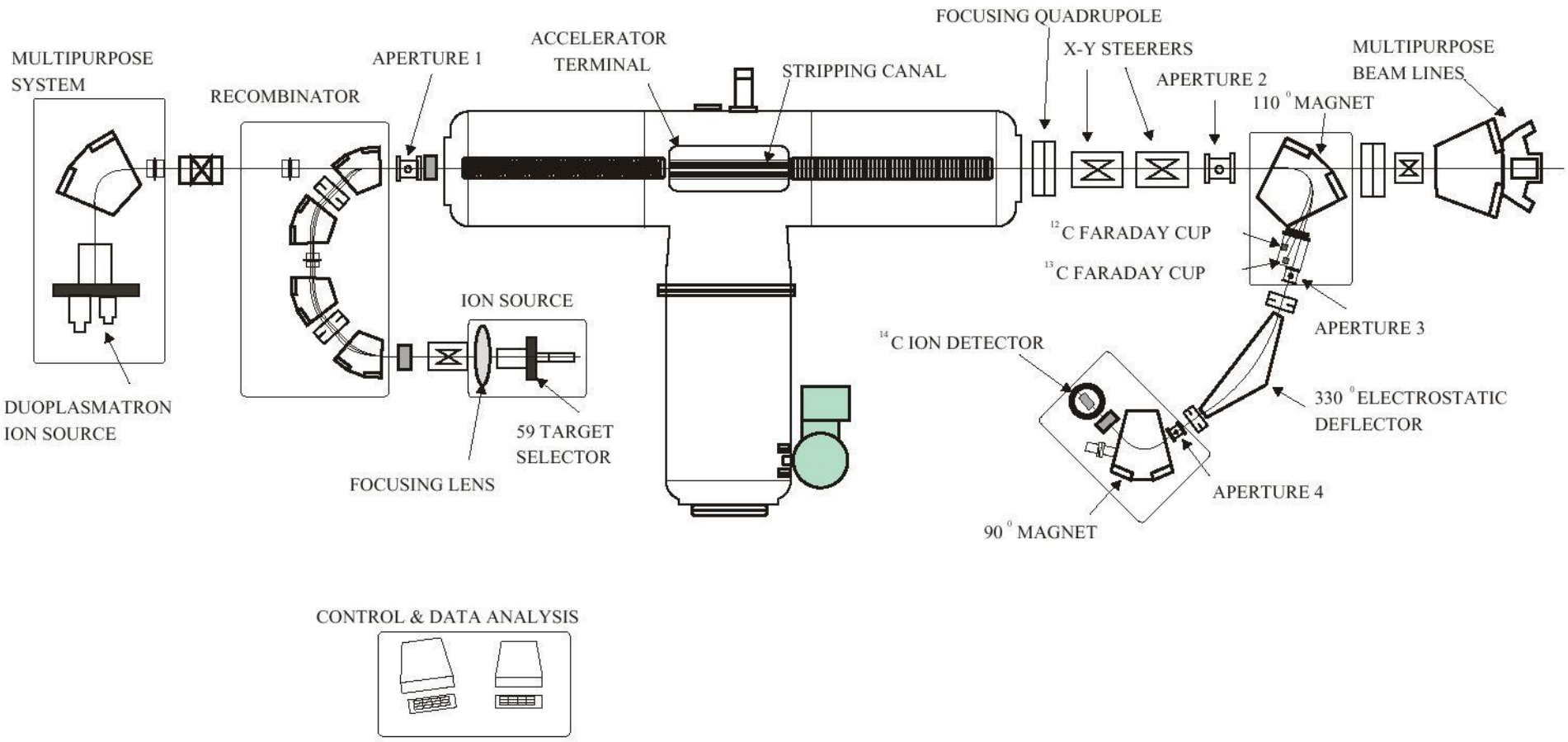
LOD(Limits of Detection) of PIXE analysis



3. The Accelerator @ SNU-Acceleator Research Center

- 3MV Tandetron(electrostatic)
- 3 ion sources
- 5 beam ports
- Mass spectroscopy system for carbon AMS
- Home-made PIXE analysis beamline & other equipments at 10-degree MPS beamline

Schematic layout of SNU-AMS facility



Beam properties from SNU accelerator center

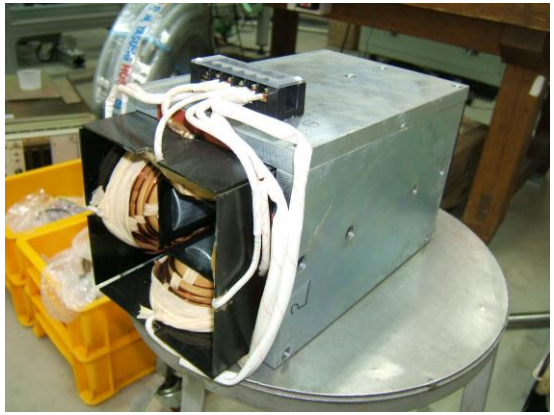
- Various accelerated ion beam are available
  - Using 3 ion sources
    - 1 Cs-sputtering source : Carbon AMS dedicated
    - 1 Cs-sputtering source @ MPS beamline
    - 1 duoplasmatron source @ MPS beamline
  - Only negative ion can be injected
  - Various charge state for accelerated beam
- Still more nuclides are to be tried
  - He, Cl, I, …

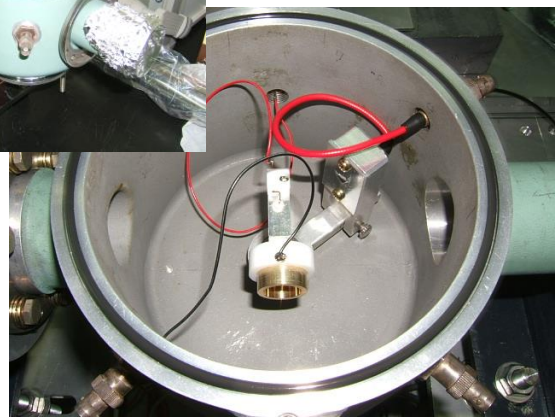
Ion	Current	comment
^{1}H	~1μA	Duoplasmatron 1~6MeV
^{10,11}Be	~100nA	Cs sputtering
^{12,13}C	~300nA	Cs sputtering; 1/97 chopping for ^{12}C

Ion	Current	comment
^{14,15}N	~100nA	Cs sputtering
^{16}O	~100nA	Cs sputtering
^{27}Al	~50nA	Cs sputtering
^{40}Ca	~20nA	Cs sputtering

External-beam PIXE development @ SNU

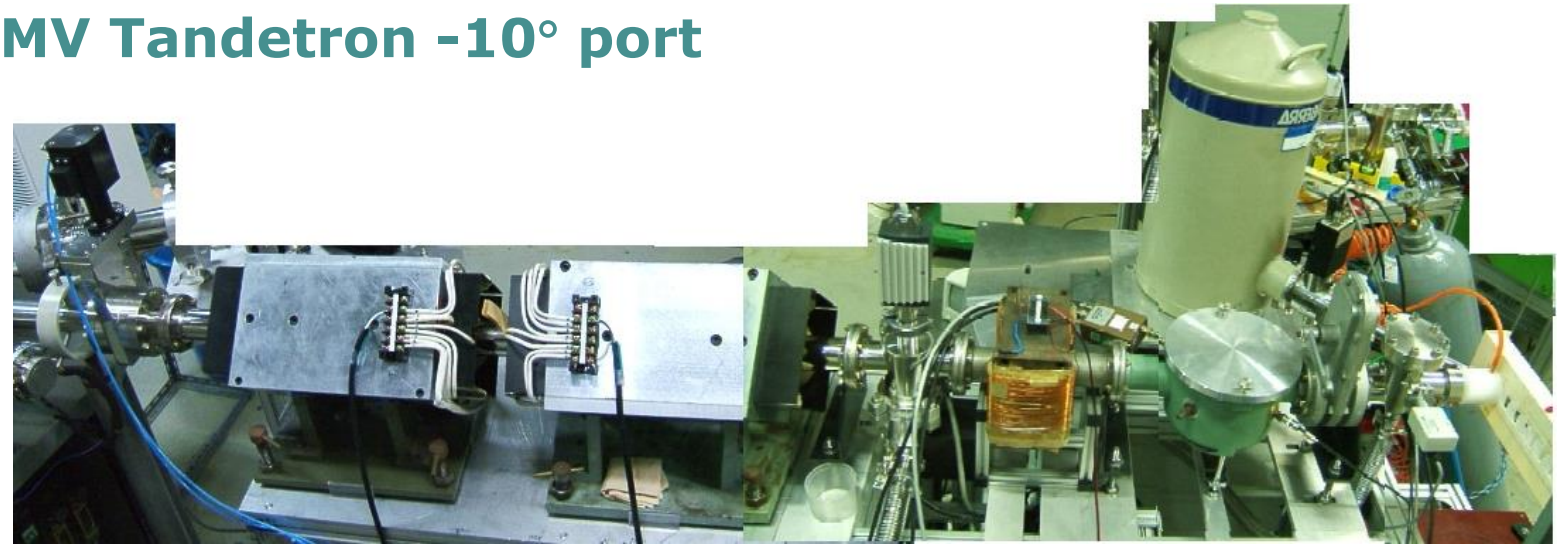
- Developments for beamline element
  - Visual BPM
  - Focusing elements – Doublet Quadrupole
  - Steering element
  - Auxiliary chamber
  - Vacuum elements
  - Beam window

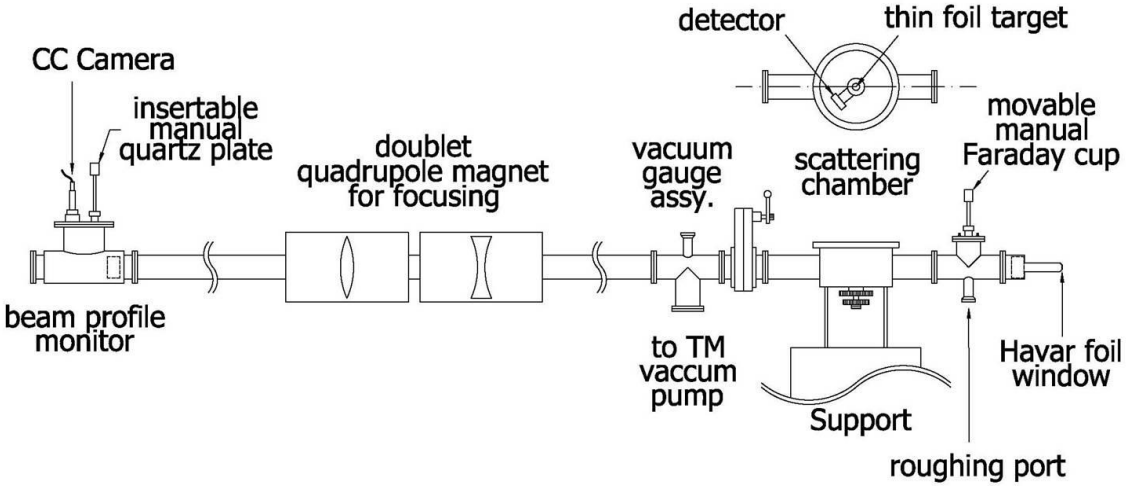




Installation

- **@ SNU IRF Accelerator Research Center**
  3MV Tandetron -10° port





양성자빔을
공기중으로
인출하여
실험

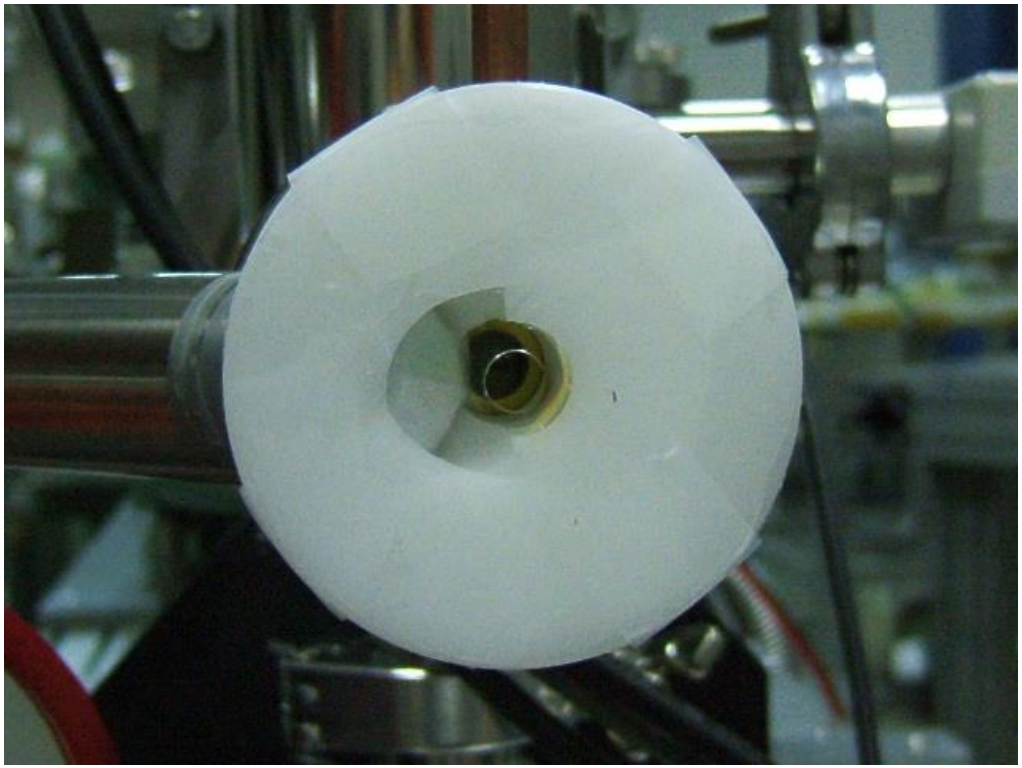
Characteristics of our beamline

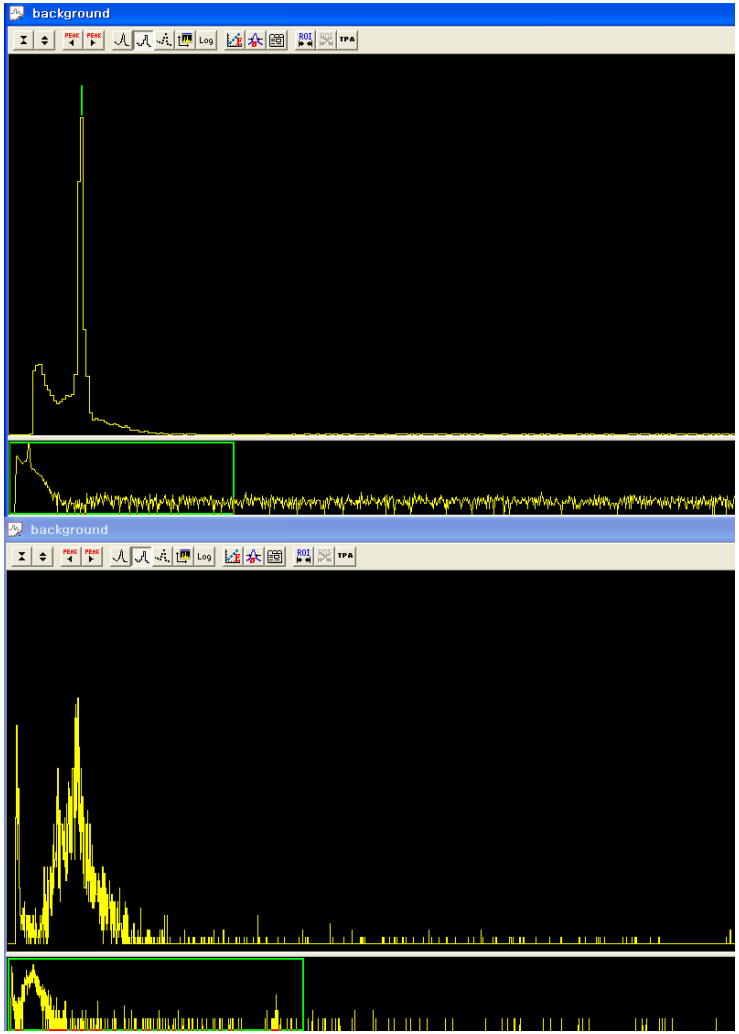
- Beam intensity
  - 0.5~20nA variable
  - To guarantee the endurance of the beam extraction window : 2~3nA @ 3MeV
- Beam size
  - Ø1mm ~ Ø5mm adjustable
  - Beam extraction window can last longer as the beam size gets bigger
- Proton beam extraction window
  - 4μm Harvar foil
    - Expensive
    - generates background
    - heavy duty
  - 10μm Kapton
    - Cheap
    - Background-free
    - Wears out at high beam current

Improvements for detection methods

Background suppresion (Ar peak)

- Using He gas
- Background reduced to 1/63





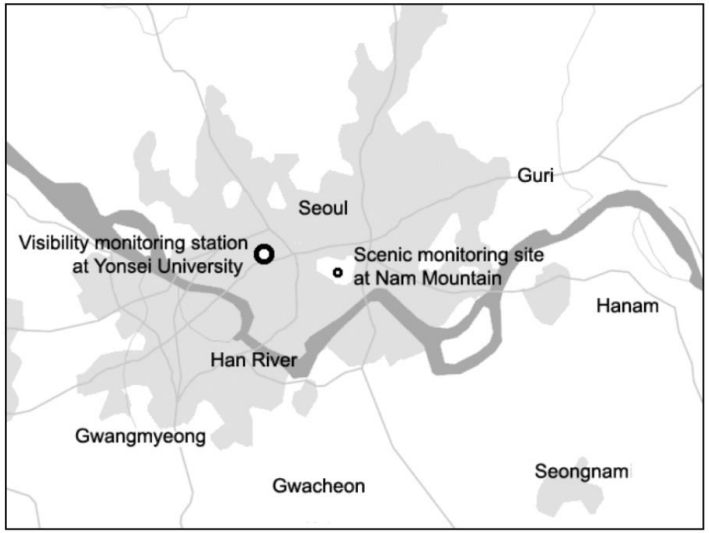
II. Case studies for the PIXE applications

1. Aerosol analysis
2. Dangerous heavy-metal in green tea?
3. 영양제 (biomedical application)
4. Discriminating forgery artifacts
5. Applications to legal documents

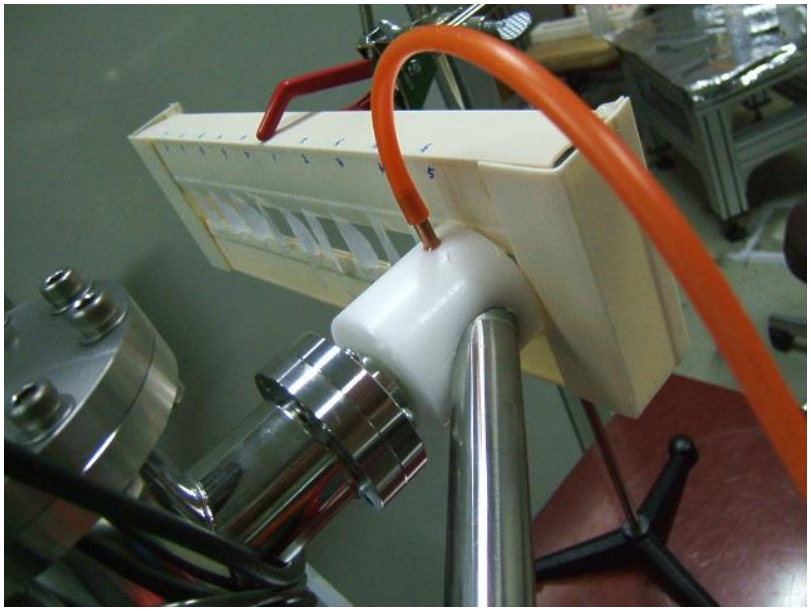
Case 1. Aerosol analysis

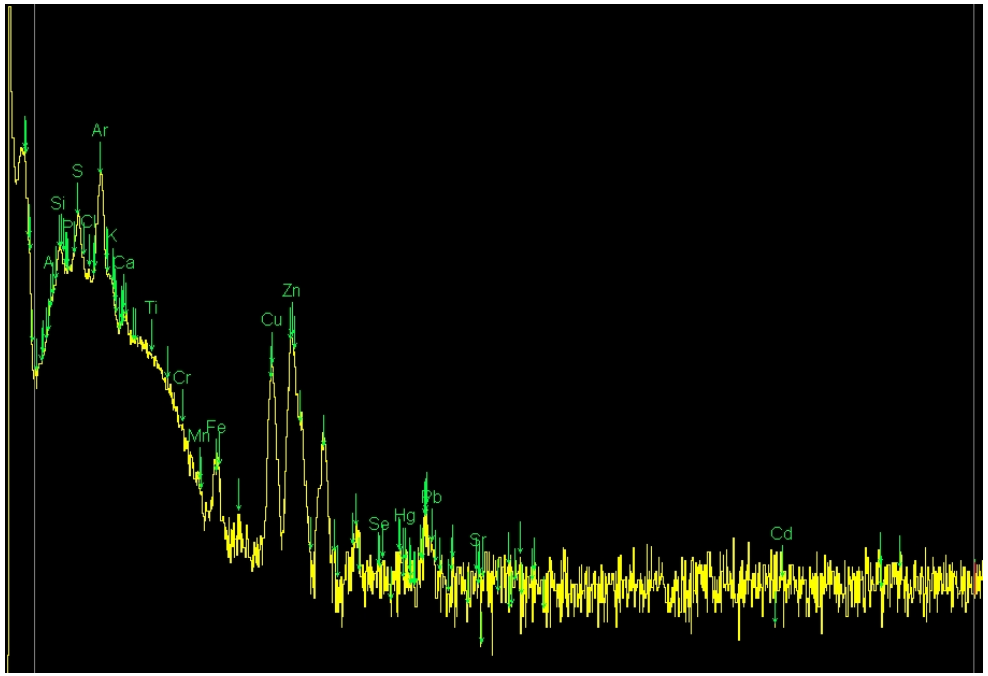
Co-work 경주대학교 환경공학과
JKPS 52 No 3 p898 (2008)
JKPS 52 No 4 p 1143 (2008)

- Aerosol(부유미세먼지)
  - Sub-micron size airborne dust
  - Strong influences on climate, health & environment
  - Collected using nucleopore filter
    @ Yonsei Univ.
  - Elemental analysis using PIXE
    as well as other analyses

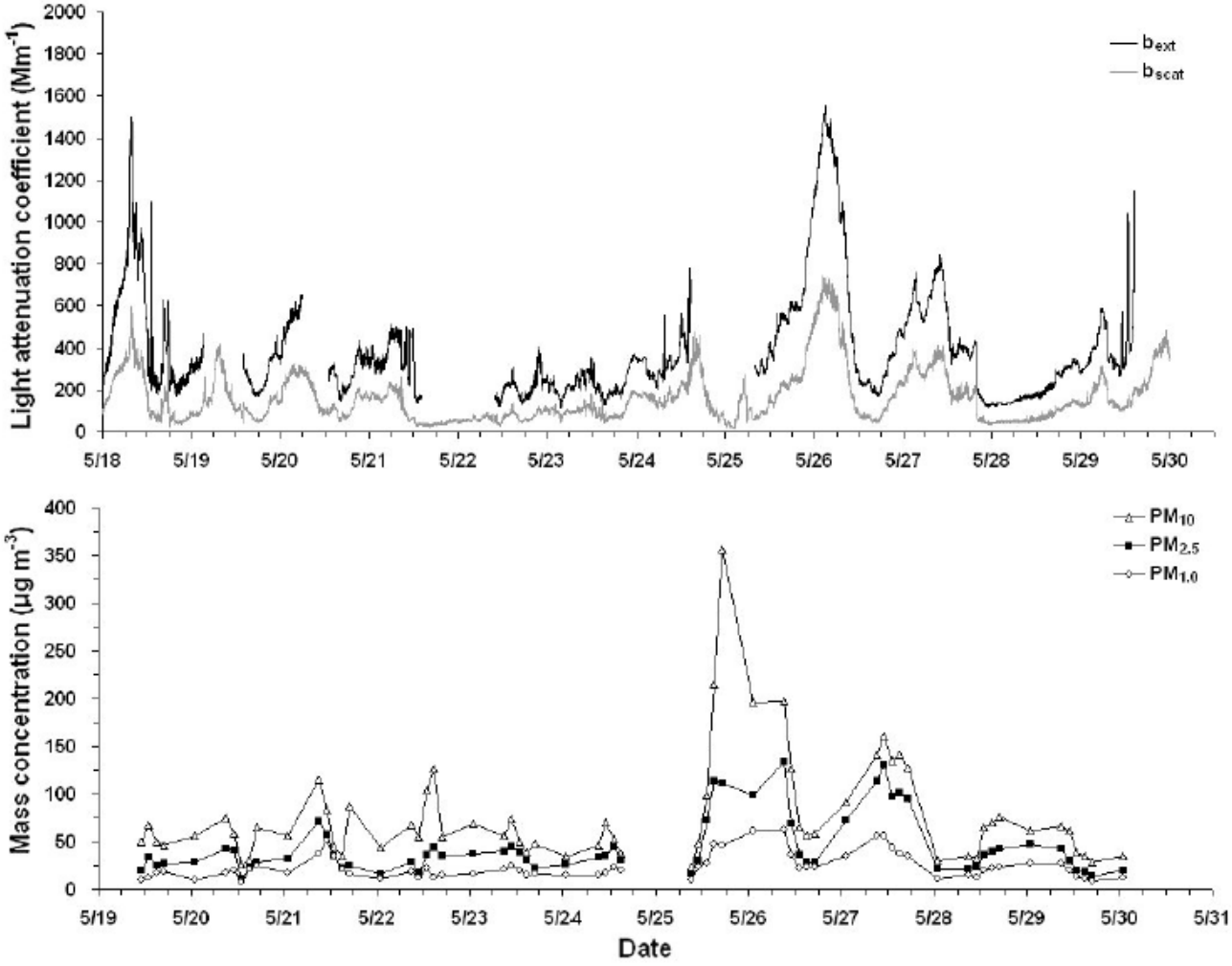


- Elemental analysis for Aerosol
  - collaboration with Gyeongju Univ.
  - Collected airbourne particle using 1.0 μm & 2.5 μm nuclepore filter
- Minimum collection : ***2 hrs***
  - Big improvements compared to the usual collection time of 10 hrs.
- Typical PIXE measurement duration : 10 min.
- Over 3000 samples have been measured so far





- Visibility & Mass concentration



- Elemental concentration
  - AD : Asian dust event (황사)
  - P : after precipitation – 비 온 후 맑은 날

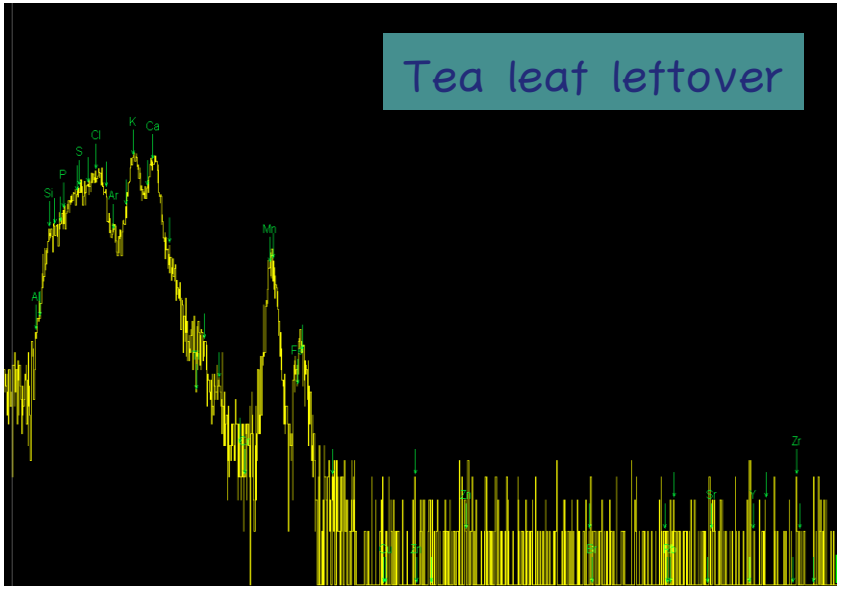
Table 2. Variation of the physical and the chemical properties of the aerosol observed at the monitoring sites of Seoul and Gyeongju. The mass concentrations of elements and ions were obtained from particles below 1.0 μm collected with $PM_{1.0}$ filter.

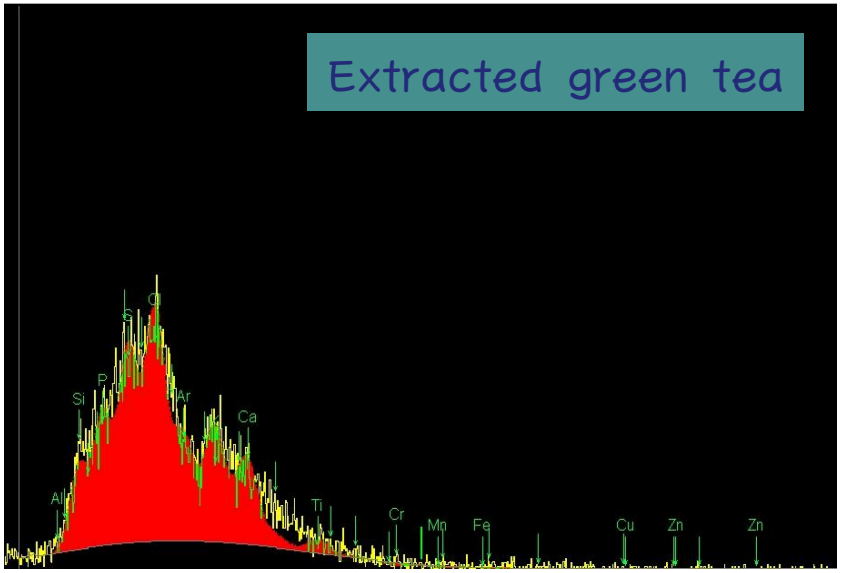
Area	Type		Mass		Element														
			$PM_{2.5}$	$PM_{1.0}$	Al	Si	P	S	K	Ca	Fe	Pb	Ti	Cr	Mn	Cu	Zn	Se	Cd
			μg m^{-3}		μg m^{-3}									ng m^{-3}					
Seoul	Total	Avg.	46.0	24.6	0.1	0.1	0.3	1.0	0.0	0.1	0.1	0.2	7.8	13.7	4.9	16.9	2.4	1.8	9.8
		Std.	30.8	13.4	0.1	0.2	0.5	1.1	0.1	0.1	0.3	0.2	9.4						
	AD[a]	Avg.	99.8	46.5	0.3	0.1	0.2	2.3	0.0	0.1	0.2	0.1	4.1	10.0	7.2	13.4	1.3	0.6	6.4
		Std.	25.2	13.8	0.2	0.1	0.2	1.7	0.0	0.1	0.3	0.2	4.8	10.8	12.1	12.9	1.5	0.5	4.3
	Non-AD[b]	Avg.	38.8	21.6	0.1	0.1	0.3	0.8	0.0	0.0	0.1	0.2	7.8	13.5	4.5	16.6	2.5	1.9	10.0
		Std.	24.9	11.1	0.1	0.2	0.5	0.9	0.1	0.1	0.3	0.2	9.6	17.5	2.9	15.5	2.8	1.8	9.6
	P[c]	Avg.	17.0	10.7	0.1	0.0	0.2	0.3	0.0	0.0	0.0	0.3	6.3	6.8	4.2	8.8	4.0	1.4	9.1
		Std.	4.2	3.1	0.1	0.0	0.3	0.2	0.0	0.0	0.0	0.5	8.0	5.5	2.5	4.3	3.8	1.0	7.1
Gye-ongju	Total	Avg.	18.1	7.7	0.5	1.2	0.4	0.2	0.1	0.1	0.1	0.1	45.2	41.9	5.9	7.5	8.8	1.6	6.5
		Std.	11.4	5.2	0.6	1.5	0.6	0.2	0.3	0.2	0.2	0.1	37.2	48.0	13.2	7.0	10.1	0.8	7.6
	AD	Avg.	53.8	25.5	1.4	6.0	1.2	0.5	0.8	0.4	0.8	0.3	138.3	107.5	50.1	11.3	30.0	0.7	35.7
		Std.	13.3	5.3	0.8	0.9	0.5	0.5	1.1	0.6	1.1	0.2	69.3	85.4	64.5	11.6	11.4	0.2	2.0
	Non-AD	Avg.	16.8	7.1	0.5	1.0	0.3	0.2	0.1	0.1	0.1	0.1	41.9	39.6	4.3	7.4	8.0	1.6	5.4
		Std.	9.2	3.9	0.5	1.2	0.6	0.2	0.2	0.2	0.1	0.1	32.0	45.8	5.9	6.9	9.3	0.8	5.3
	P	Avg.	9.1	4.0	0.3	0.3	0.1	0.1	0.1	0.0	0.1	0.1	25.1	36.9	3.0	6.3	3.6	1.7	4.5
		Std.	4.8	2.2	0.5	0.5	0.2	0.1	0.1	0.1	0.1	0.1	24.4	60.9	3.3	6.3	4.5	0.7	3.5

Case 2. Dangerous heavy-metal in green tea?

- the news concerning harmful heavy metal made my life tough - Decided to examine myself
- Samples prepared
  1. Drinking water (background)
  2. Coffee (results not shown here)
  3. Extracted green tea
  4. Tea leaf after extraction

- Result
  - Drinking water contains almost nothing (not shown here)
  - Extracted tea contains almost no heavy metal
  - Tea leaf keeps Mn, Cr, & Fe – Non water-soluble

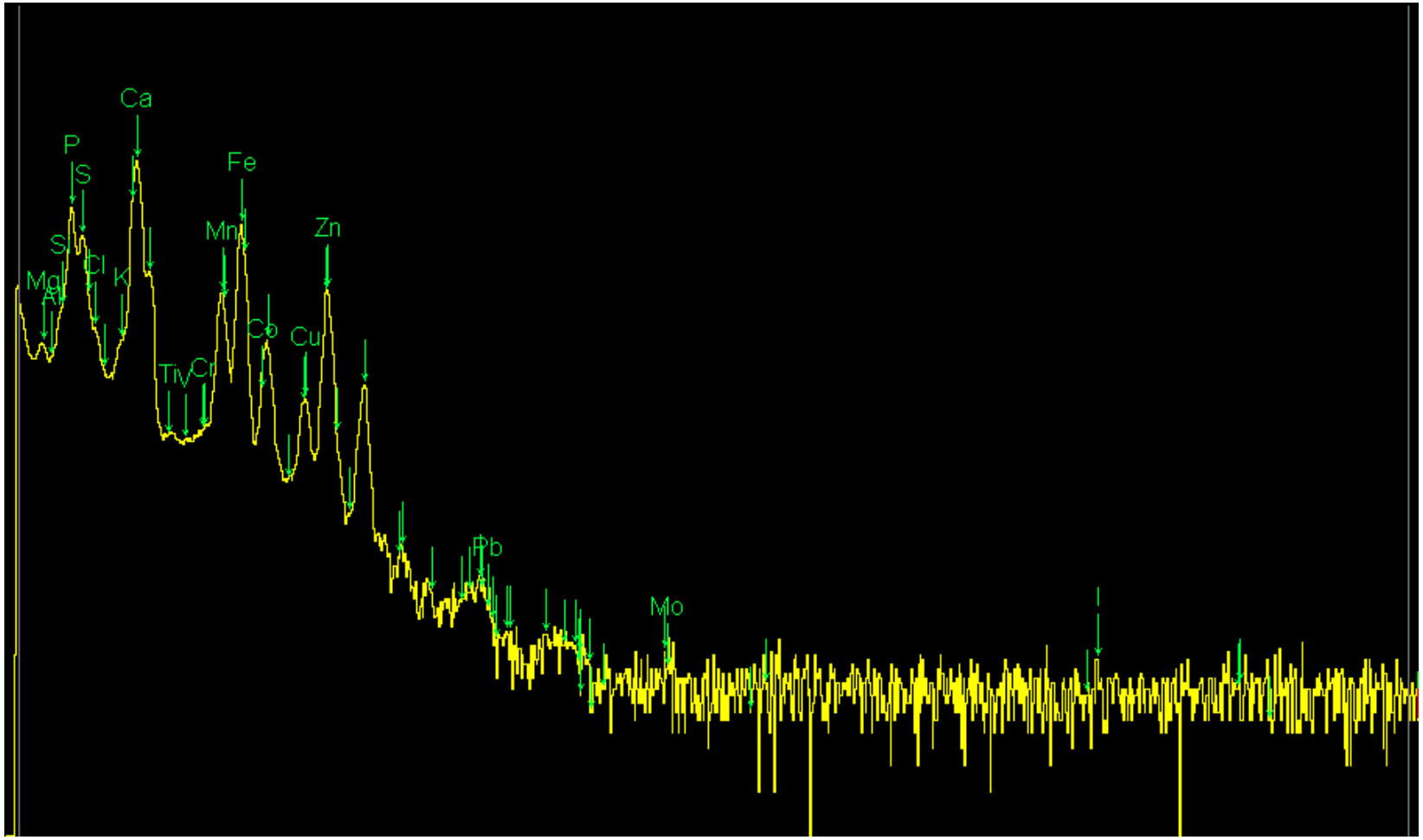




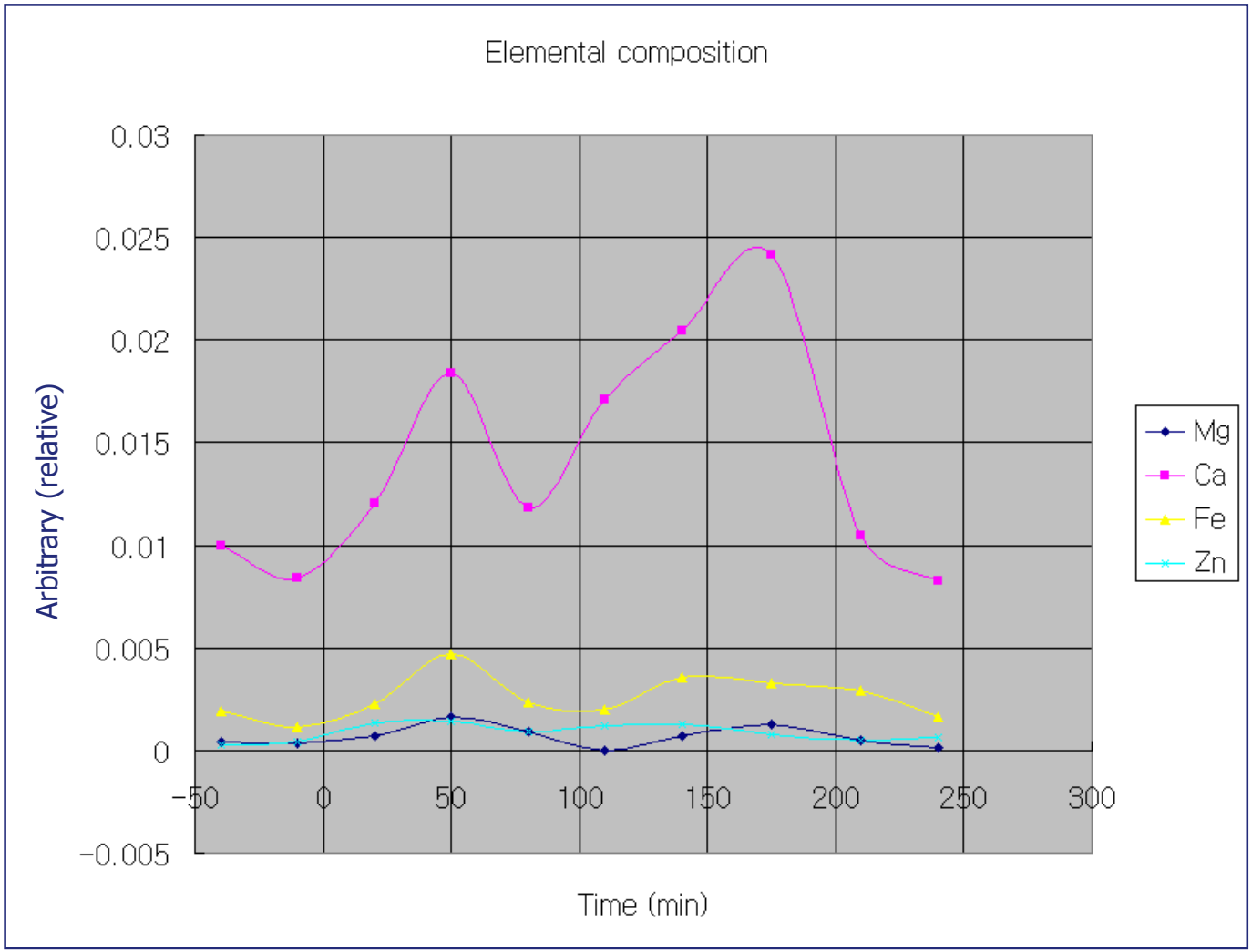
Case 3. 영양제 (biomedical application)

- Q : 영양제를 먹으면 몸에 뭐라도 좀 좋은가?
- A : 안 먹으면 괴롭다 (아내와 딸이 생일선물로 사 주었기 때문에)
- Q : 먹으면 몸에 무슨 효과가 있을까?
- A : 모르겠는데 – 한 번 알아보자
  - 공복에 영양제 **2**알 **+** 물
  - 매 **30**분 가량 마다 소변 채취
  - **1~2** 방울을 **filter paper**에 떨구어 건조
  - **PIXE**로 원소 성분 검사

- 영양제의 원소 구성 - Mg, Al, Si, P, S, Cl, K, Ca, Ti, Mn, Fe, Co, Cu, Zn, Mo, I, Pb,….

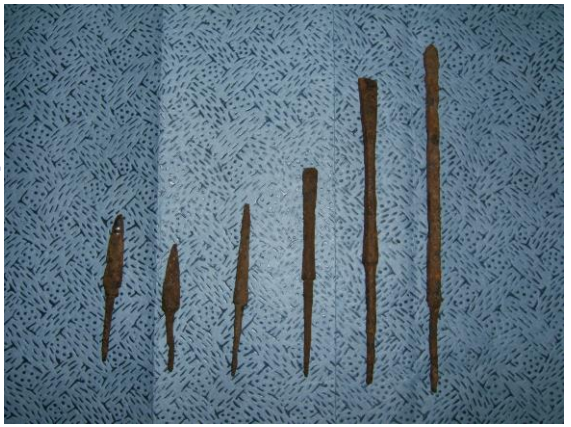


Result for the case



아마도 약 먹고 1.5시간 내에 전부 배설되는 것 같다.
- 꼭 좋은 것 같지 않음 (Ca, Fe, Mg).

Case 4. Discriminating forgery artifacts



- 태왕신검(太王神劍)?
  - Iron artifacts, as well as pottery artifacts claimed as excavations from a royal tomb of Goguryeo (高句麗: 37BC ~ 668AD) period
  - Arrived to the Lab for radiocarbon dating
- Several suspicious facts
  - Almost no carbon from weaponry artifacts : For sword, body is steel & blade is cast iron – should contain lots of carbon → radiocarbon-dating failed
  - Stain layer was too uniform for 1000 years old artifact
  - Iron under the stain layer is quite clean
- tried PIXE analysis

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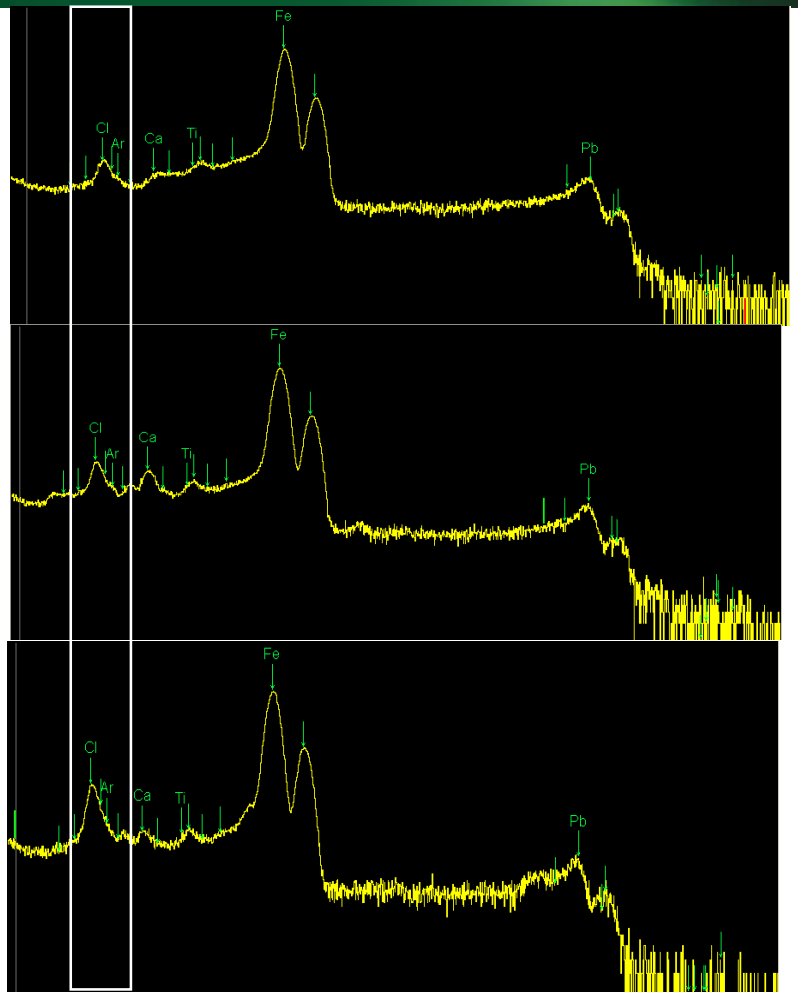
[1] Seoul National University
[2] Chungcheong Cultural Properties Research Institute

PIXE Measurements & analysis

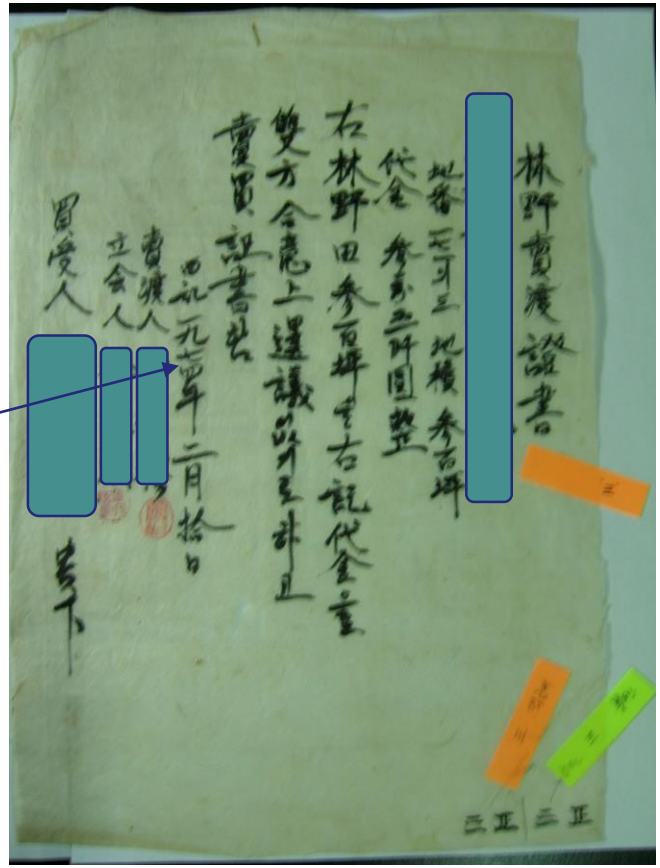


- Too low Cl component (that diffuses into the artifact from air) – less than several years old nail
- Conclusion : **the stain layer was recently formed artificially**

Case 5. Application to legal documents

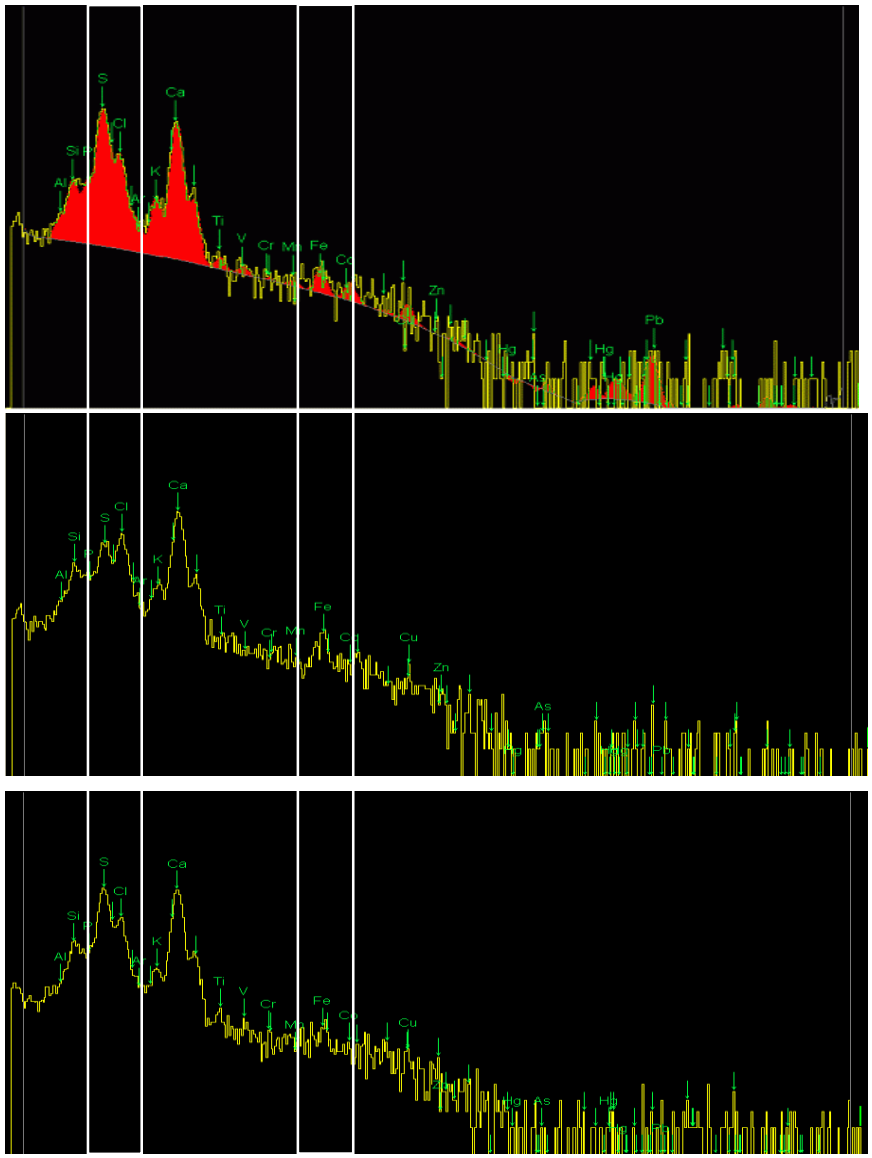
Confidential – No references, please

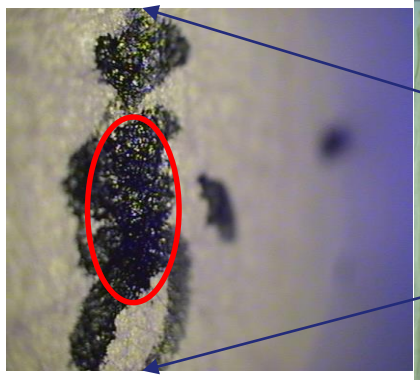
- Non-destructive elemental analysis using PIXE to evaluate the authenticity of legal documents
- Example : the contract
  - Dated to 10 February, 1974
  - Written on Korean paper using brush & Indian (Chinese) ink
- Is this a genuine document or a forgery?



Co-works : Institute of Korea Document Authentication

Analysis & conclusion



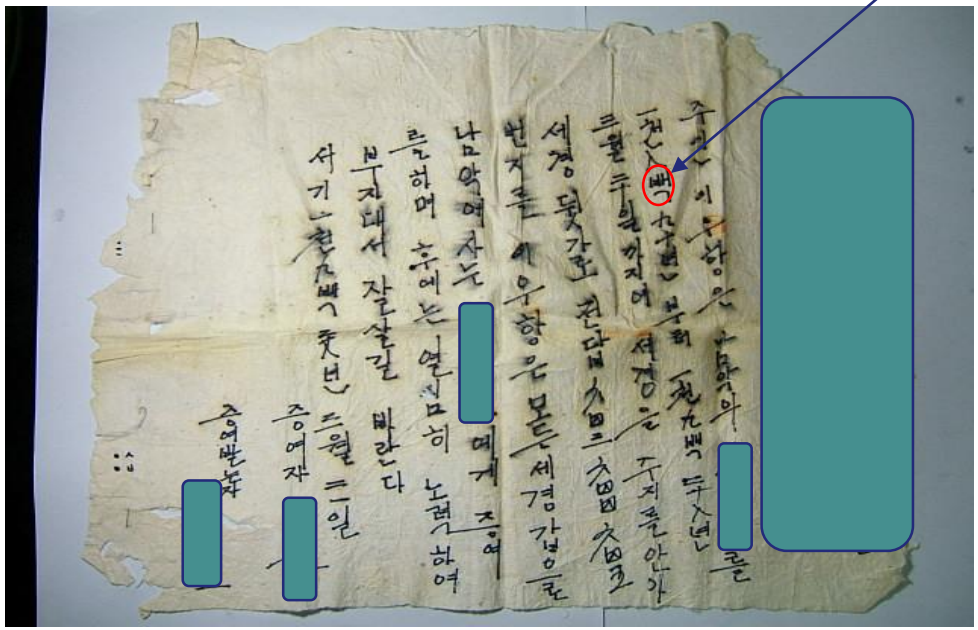


Conventional
brush & indian ink

Modern brush pen
(after 1980's)

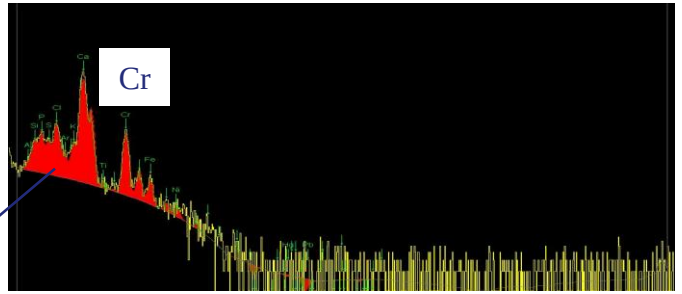
Another legal document

- 1930년 3월 3일자 증여증명서
- 당시까지 평생 머슴 산 대가를 주지 않았으므로 그 대가로 전답을 증여
  (문서제목 및 실명은 19禁 처리하였음)
- 진짜?

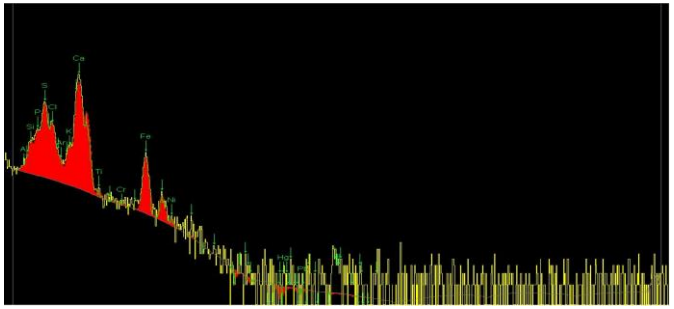


가짜 !!

Co-works : Inst. of Oriental Document Authentication



문서상의 글자 : 먹은 도저히 아님



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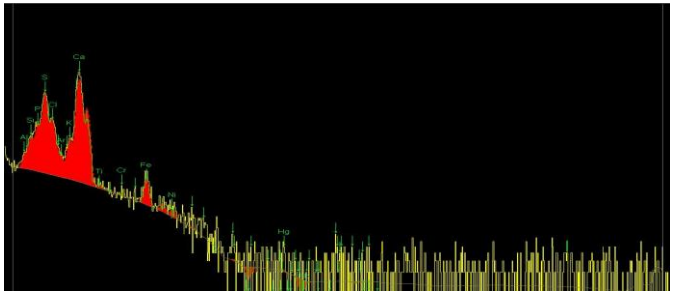
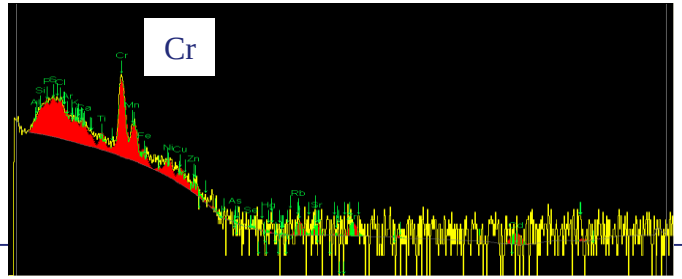


사인펜



네임펜

III. Limitations and prospects for improvements

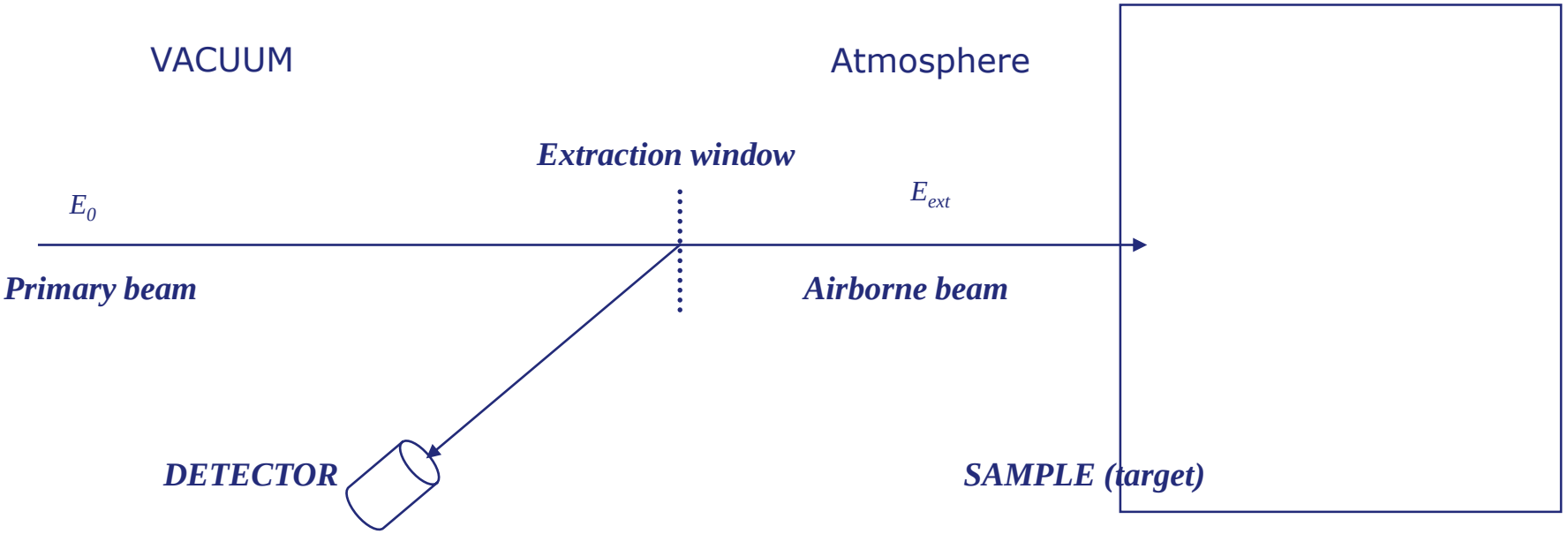
Basically VERY thick target experiment

1. Hard to perform an absolute measurement
   - ✓ Incident beam charge measurement & beam current monitoring
   - ✓ Hard to calculate an accurate cross-section : Calculating cross-section needs feedback from
     1. Stopping power
     2. Mass attenuation coefficient
     3. Concentration (!!!!!)

2. Relative measurement using of SRM(standard reference material) – possible

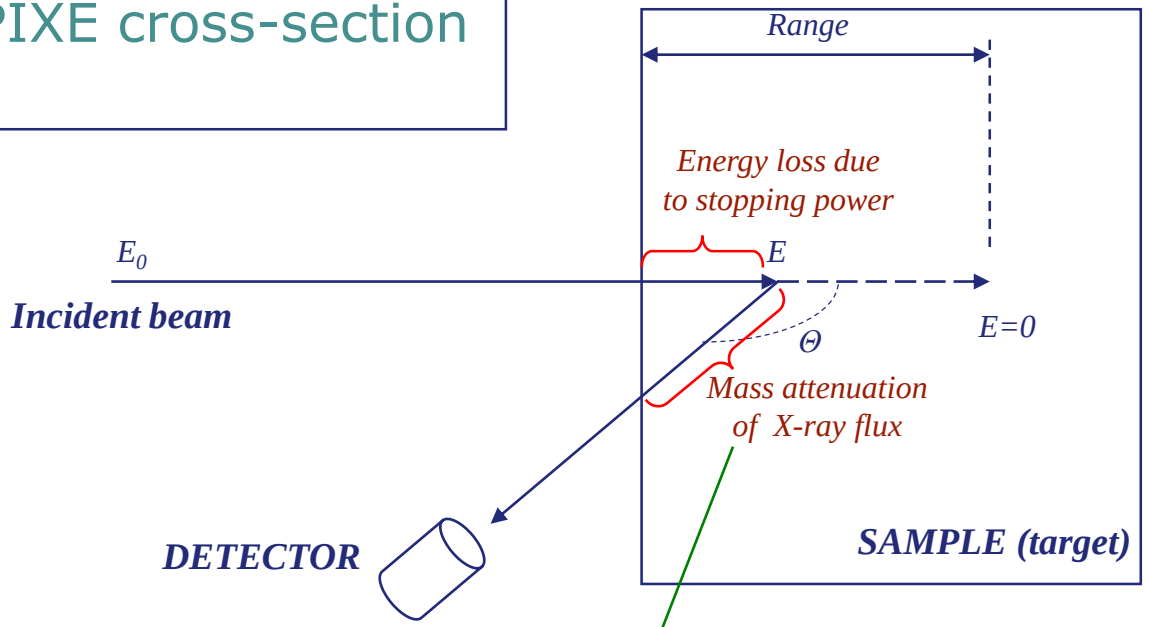
   However, what happens if
   - ✓ an appropriate SRM is hard to get
   - ✓ Sample contains new elements compared to SRM

Incident beam current monitoring



- Counting elastic-scattered proton from window
  - Window acts as a (relatively) thin uniform target
  - Proportional to primary beam (& extracted beam)
- *Solves the problem concerning the beam charge*

Description for a PIXE cross-section for a thick target



Yield

$$Y = \frac{CN_0}{W}\frac{\Omega Q_\varepsilon}{4\pi\epsilon}\int_{E_0}^{0}\sigma_X\frac{dE}{S(E)}exp\left[-\mu\int_{E_0}^{E}\frac{dE^*}{S(E^*)}cos\theta\right]$$

- Calculating variables in red circle need concentration – ambiguous feedback chain
- That's why traditionally PIXE does not pursue absolute measurement

Using SRM – relative measurement

- VERY many SRMs from various institution
  - SRM means a material of which the concentration has been measured with sufficient accuracy & statistics
- Ambiguity possibly develops
  - **What for a new material?**
  - **What if the sample contains more element?**
  - **What if the target is not thick enough?**
- Brings back the necessity for an absolute measurement
- Is there any way to go overcome?
  - R/D for Analysis method
  - New measurement method.

Using multiple, arbitrary SRMs (for thin sample)

- If the sample is thin enough, (such as writing or stamp on a thin paper)
  - **stopping power or attenuation plays no great role**
  - **Preparing compatible SRM sample of comparable thickness is enough**
  - **Usually less than 10% difference w.r.t. SRM difference**
  - **Currently used**

- If the sample is thick
  - **Still under development**
  - **Counting the backscattered proton from the sample is being considered (hard to detect airborne proton, though)**
  - **Hope this method may cancel some contributions from the stopping power**

On-going development

- Reducing the beam size
  - Ø 1~ Ø 5mm variable (current)
    - Adequate for large, uniform sample
    - Inadequate for small, inhomogeneous sample (such as stamp, hand-writing, sediment, rock)
  - Goal : Ø 0.2mm or less (next year)
    - Stamp or printed hand writings will be safely measured
    - Core-bored sediment sample can be measured
  - Necessary works
    - Defining actual beam-optical axis precisely
    - Fine tune steering of the axial beam position to optical axis
    - Introducing more quadruple magnets (1 doublet pair)
  - Hope we can report a good achievements.

Thank you