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## Combined PIXE/PIGE with the high-energy beams of the ARRONAX cyclotron

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## summary and outlook

A high energy PIXE platform is now available at ARRONAX. Its is well suited for in-air multi-elemental analysis (at the ppm level for medium and heavy elements) and bulk analysis (stoichiometry, density, etc.) of thick and multilayer samples. For the analysis of low atomic number elements, an additional Silicon Detector can be used closer to the samples and a large LaBr detector will be soon installed to develop the complementary PIGE method.

## Ion Beam Analysis with the ARRONAX cyclotron beams

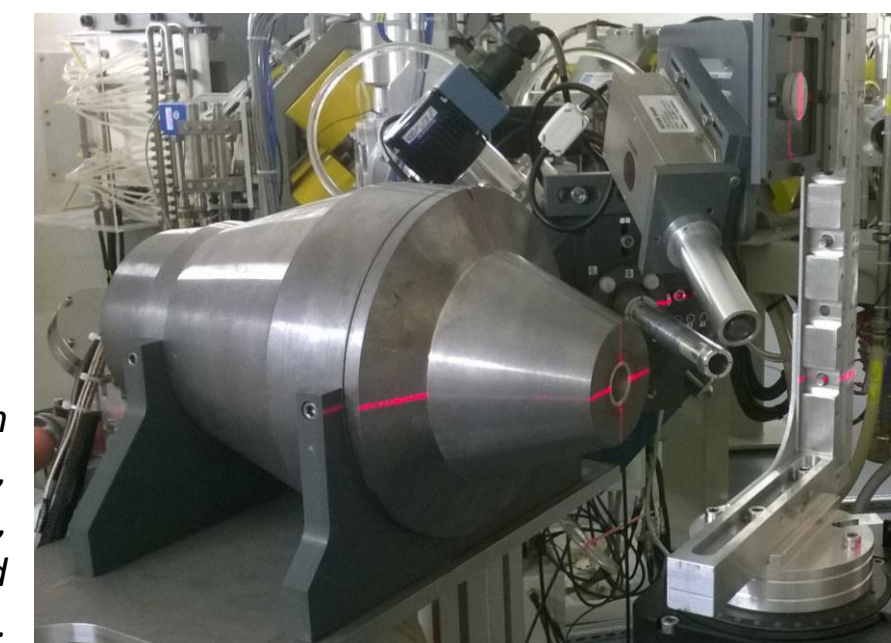
ARRONAX (Nantes, France) is a multi-particle cyclotron (Cyclone®70): **protons** can be accelerated from 30 MeV up to 70 MeV, **deuterons** from 15 MeV up to 35 MeV and **alpha** particles at fixed 68 MeV.

For ion beam analysis, protons are selected for their **higher range** in the matter, alpha for their **higher PIXE sensitivity** (K X-ray production cross section) and low energy deuterons essentially for **PIGE**.

Main features:

- multi-elemental analysis (ppm) thanks to K X-ray detection
- fast, non-destructive, in normal air
- thick / multilayer samples
- density and stoichiometry measurement

Fig. 1 – high energy PIXE set-up with the beam extraction window and collimators (center), the shielded X-ray LEGe detector (left), the X-ray silicon drift detector (top) and the automated sample holder (right).



## Multi elemental analysis

D.Ragheb et al., J Radioanal Nucl Chem (2014) 302:895-901

X-rays are detected in normal air with a LEGe detector (Canberra GL005P, Be window) at an angle of detection  $\theta = 135^\circ$  (see Fig. 1). The distance between sample and detector varies from 5 to 25 cm (dependent on experimental requirements). The number of incident particles is measured either with a 2  $\mu\text{m}$  monitor copper foil or the argon (in air) peak (previously calibrated). The beam diameter is few mm and the intensity is <1nA.

The limit of detection (LoD) reaches the ppm for medium and heavy elements in light matrix.

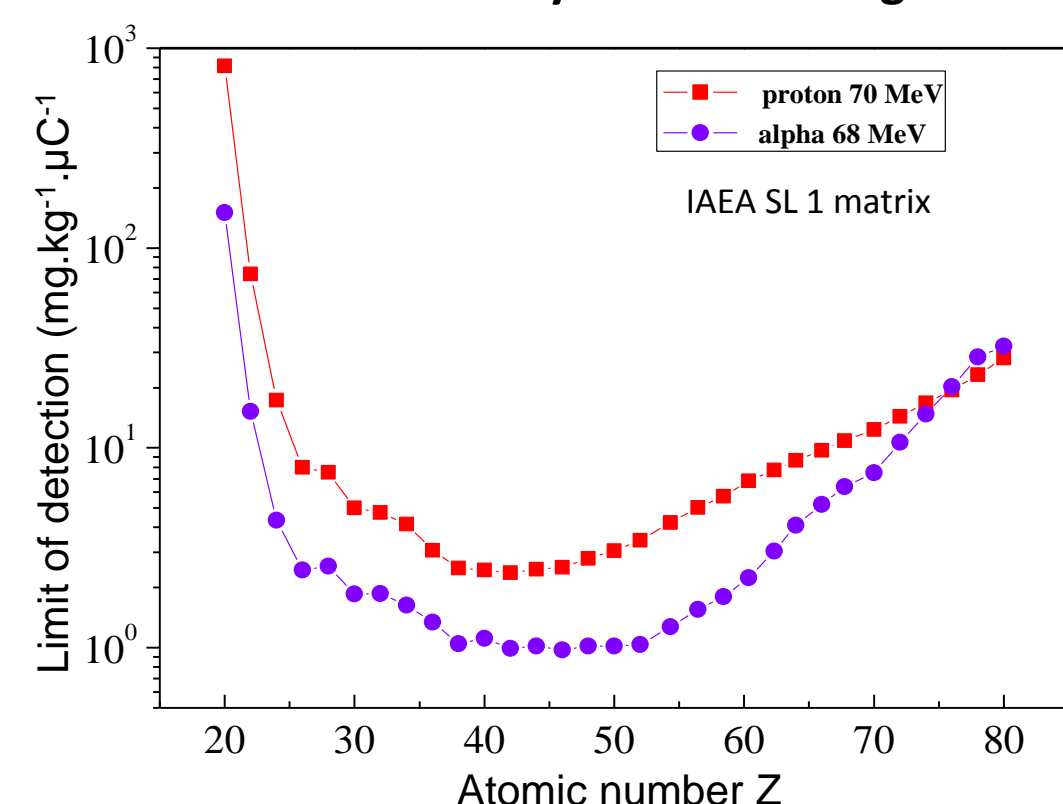


Fig. 2 – limit of detection in lake sediment IAEA RM SL-1 matrix (500 $\mu\text{m}$  thick pellet; detector at 16 cm away).

Ex: composition of natural and synthetic sodalite (photochromic material)

- 68 MeV alpha particle beam
- Relative quantification (ref. IAEA RM SL-1):

$$N_i = N_p C_i \omega_{K_i} b_{ij} T_{abs} \varepsilon_i \frac{\Omega}{4\pi} \int_{E_{in}}^{E_{out}} \frac{\sigma_i(E) T_{target}(E)}{S(E)} dE$$

Sample	Natural sodalite	Synthetic sodalite	IAEA RM SL-1
Form	Pellet	Pellet	Pellet
Mass	216.9 mg	94.2 mg	400 mg
Thickness	1200.74 $\mu\text{m}$	521.74 $\mu\text{m}$	552 $\mu\text{m}$
Diameter	1 cm	1 cm	2.2 cm
Density	2.3 g.cm <sup>-3</sup>	2.3 g.cm <sup>-3</sup>	2.1 g.cm <sup>-3</sup>

Table 1 – characteristics of the sodalite samples and the reference one (IAEA RM SL-1).

Element	K X-ray energy (keV)	natural sodalite (mg.kg <sup>-1</sup> )	synthetic sodalite (mg.kg <sup>-1</sup> )
Ca	3.7	9193 $\pm$ 1747	4655 $\pm$ 1303
Mn	5.9	373 $\pm$ 104	667 $\pm$ 113
Fe	6.4	8128 $\pm$ 1138	943 $\pm$ 132
Cu	8.0	3498 $\pm$ 514	1709 $\pm$ 290
Zn	8.6	247 $\pm$ 44	492 $\pm$ 83
Ga	9.2	18 $\pm$ 5	
Br	11.9	123 $\pm$ 20	
Ag	22.1	39 $\pm$ 15	
Sn	25.2		53 $\pm$ 14
I	28.6	97 $\pm$ 25	

Table 2 – quantification of the detected elements in both samples of sodalites by high energy PIXE.

## Bulk analysis (density and stoichiometry)

Determination of the thickness, the density and the mass fraction of a Ni/Ga alloy deposited on a gold substrate (to be used as target at ARRONAX for production of a <sup>68</sup>Ge/<sup>68</sup>Ga generator for medical imaging).

T. Sounalet et al., Nuclear Data Sheets, Vol. 119(2014)261-266

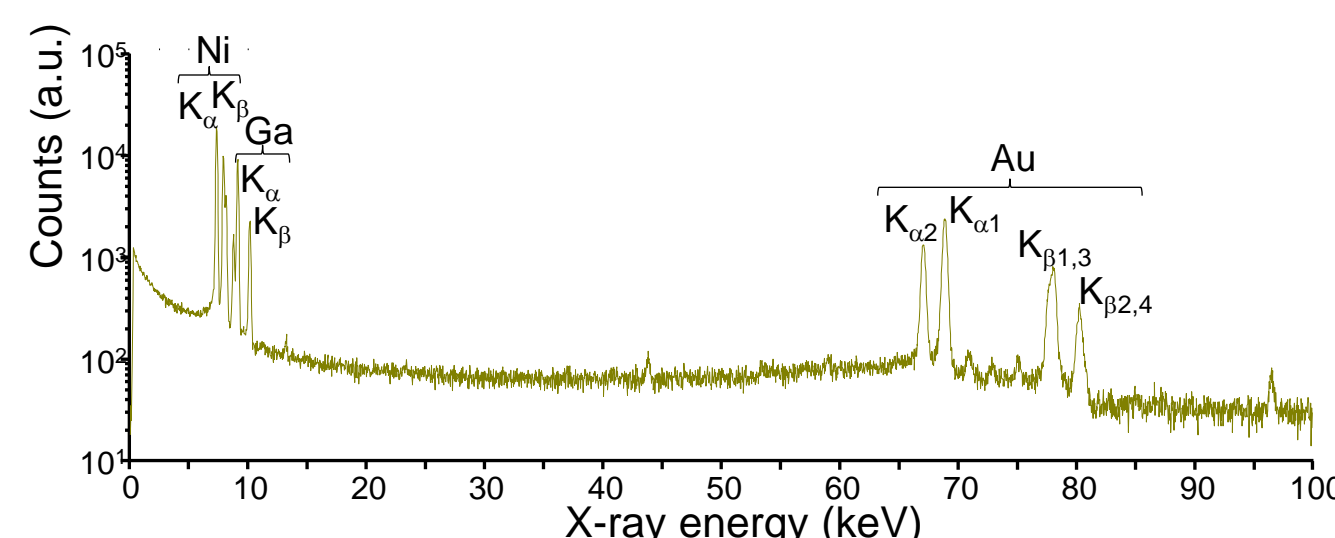


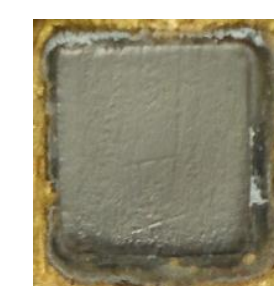
Fig. 4 – X-ray spectrum from irradiation of the Ni/Ga(gold substrate) target with a 70 MeV proton beam (LEGe detector at 25 cm).

- the mass fraction and the density of the Ni/Ga alloy are calculated through an iterative process based on the K X-ray intensities of Ni and Ga.
- the thickness of the alloy is then determined thanks to the attenuation of the K X-ray coming from the gold carrier.

Element	HE PIXE wt%	ICP-AES wt%	SEM/EDX wt%
Ni	34,18 $\pm$ 0,7 %	34,15 $\pm$ 0,4 %	34,04 $\pm$ 0,3 %
Ga	65,19 $\pm$ 0,8 %	65,85 $\pm$ 1,6 %	65,96 $\pm$ 1,3 %

Table 3 – mass fraction (in percent by mass, wt%) of the Ni/Ga alloy measured by high energy PIXE and two comparative destructive methods.

Fig. 3 – electrodeposited Ni/Ga alloy on gold substrate



Measured (by PIXE) Ni/Ga alloy:  
• thickness: 317  $\pm$  32  $\mu\text{m}$   
• density: 6.9 g.cm<sup>-3</sup>

## Light element detection: complementary PIGE and SDD X-ray detector

As part of improving the detection of light elements, two additional detectors have been used: a HPGe detector (from Canberra, 50 mm diameter and 57 mm depth) gamma-rays and a Silicon Drift Detector (SDD from Ketek) for low energy X-rays. Two different types of sand have been irradiated with a 16 MeV deuteron beam; the HPGe gamma detector was located at 100 cm from the target and the SDD for low energy X-rays at 2.3 cm (see Fig. 1).

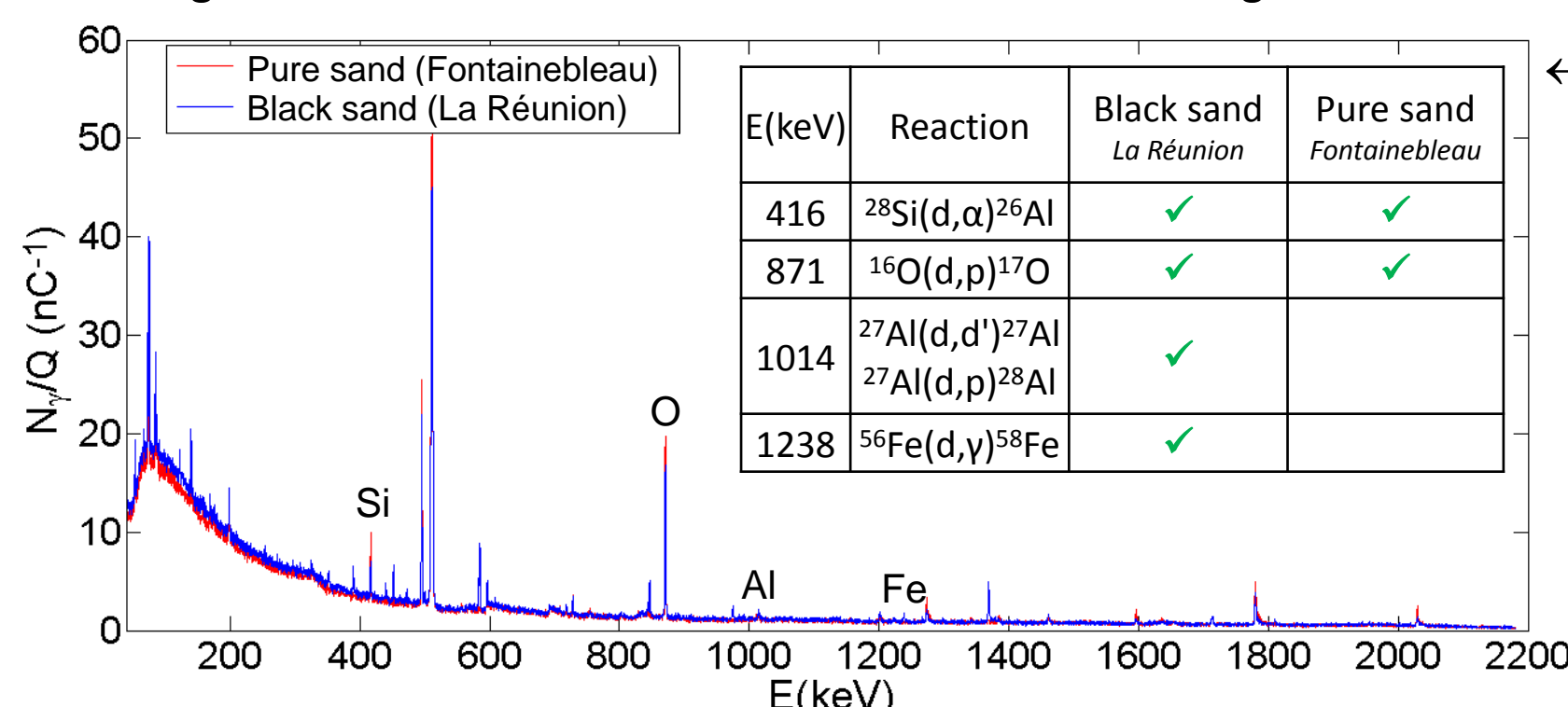
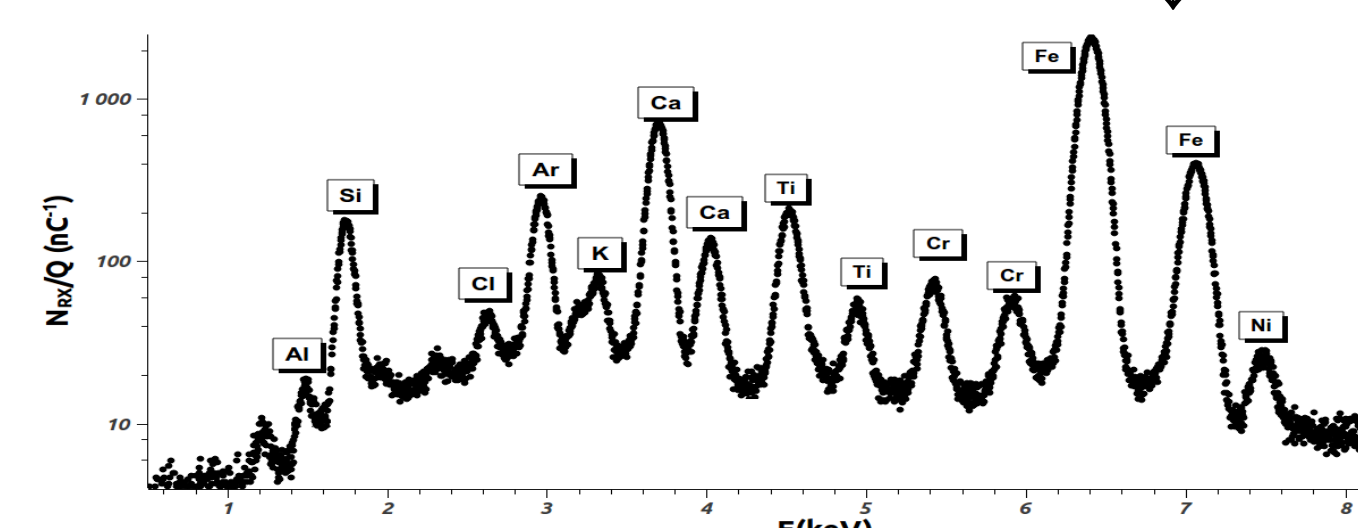


Fig. 5 – gamma-ray spectra obtained for 2 samples of different sands.

Fig. 6 – X-ray spectra (SDD detector) for black sand (from La Réunion).



- the **limit of detection** with X-rays is **improved for light elements** (down to Al, Z=13, at least) with the additional detector (compact SDD);
- lighter elements (such as **Oxygen** in this case) can be analyzed thanks to **gamma rays** (PIGE);
- a method to determine the water content of soil samples using gamma rays is being developed (from this study with sands).