



Neutron Activation Analysis (NAA), Radioisotope Production via Neutron Activation (PNA) and Fission Product Gas-Jet (GJA)

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ABSTRACT

Three different non-diffractive applications of neutrons are outlined: neutron activation analysis, production of radionuclides, mostly for medical applications, and production of short-lived fission nuclides with a so-called gas-jet. It is shown that all three devices may be incorporated into one single insert at SINQ due to their different requests with respect to thermal neutron flux. Some applications of these three facilities are summarized.

1. Introduction

Neutron activation analysis (NAA) is a powerful technique in analytical chemistry. It bases on the well known fact that via neutron capture stable nuclides transform to neighbouring, mostly radioactive isotopes. A measurement of the nuclear decay of these isotopes, usually via γ -spectroscopy, can then be used for a quantitative mass determination of the corresponding element. For NAA without any chemical separation the acronym INAA (Instrumental-NAA) is used, and if chemical separations are required after irradiation but prior to a measurement of the radioactive species the label RNAA (Radiochemical-NAA) is applied.

A clear advantage of NAA is the fact that it is a non-destructive method. This strongly reduces contamination problems. NAA is therefore a reference technique in analytical chemistry and often *the* preferred method in ultra-trace analysis. Most of the alternative techniques such as inductively coupled plasma - mass spectrometry (ICP-MS) and - optical emission spectrometry (ICP-OES) as well as atomic absorption spectrometry (AAS) and others require dissolution of the sample prior to injection into the corresponding device.

Formation of radioactive nuclides via neutron-capture (PNA) is also frequently applied for production of radiotracers for widespread applications. Examples are optimizations in chemical synthesis or developments of chemical analytical separations. Important is also the production of radioisotopes for radio-pharmaceutical and nuclear medical applications. In this case, relatively long-lived radionuclides (half-lives \geq hours) are needed since production facility and application site (i.e. hospital) are usually not within short distance. Moreover, for therapy applications highly radioactive compounds are needed. The irradiation facility has then to fulfill some special requirements to safely handle such a production site.

Another application of thermal neutrons in radiochemistry are the so-called gas-jet facilities for fission products [1]. Here, a thin ^{235}U target is placed in a closed chamber exposed to neutrons. This chamber is continuously flushed with a carrier gas containing aerosol particles. From the target recoiling fission fragments are thermalized in the gas and then attached to the aerosol particles. With high efficiency such particles can then be swept out of the production chamber and continuously transported along thin capillaries to a laboratory. With this technique short-lived nuclides with half-lives down to about one second are available for further applications such as e.g. chemical studies. Typical examples are modeling experiments for heaviest elements or studies of surface chemical interactions in an aerosol. A special option is to use the nuclear decay time as a clock to measure a time of a chemical reaction [2].

2. The concept of „Insert 60“ at SINQ

All three different techniques described above have been included into one single insert (see [3]). This was possible due to their different requests with respect to thermal neutron flux. The position of RPN is closest to the core, with the highest beam intensity, followed by the position of NAA, and, finally, by the gas-jet facility. Expected thermal neutron fluxes are $4 \cdot 10^{13}$, 10^{13} and $10^9 \text{ cm}^{-2} \text{ s}^{-1}$ at the positions of PNA, NAA and GJA, respectively.

3. Neutron activation analysis (NAA)

Neutron activation analysis is especially attractive if short-lived isotopes are formed by the neutron capture process. This requires a fast transportation of the samples, mounted in a suitable container, from a laboratory to the irradiation position and back again. Usually the containers are moved via pneumatic transport („rabbit“ systems). Such a device was operational for many years at the SAPHIR reactor („BLASIUS“ system). With this system the samples were irradiated between 20 s and 1 h at a flux of $2.2 \cdot 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$ and then transported along a 100 m pneumatic transfer tube to the radiochemistry laboratory in a neighbouring building.

Generally, the production of an isotope via NAA may be described by the following equation:

$$A = N\sigma\phi[1-\exp(-\lambda t_b)]\exp(-\lambda\Delta t)[\lambda t_m / \{1-\exp(-\lambda t_m)\}] \quad (1),$$

with

A: Activity of the produced isotope at the beginning of the counting time, in Becquerel (Bq)

N: Number of target atoms, in cm^{-2}

σ : Thermal neutron capture cross section, in cm^2 (see e.g. [4])

λ : Decay constant of the produced isotope, in s^{-1} (see e.g. [4])

t_b : Irradiation time, in s

Δt : Time difference between end of bombardment and start counting, in s

t_m : Measuring time, in s

ϕ : Neutron flux, in $\text{cm}^{-2} \text{ s}^{-1}$

As a general rule, an activity of 10 Bq can be defined as detection limit.

In reality NAA is mostly used as a relative method since the neutron flux at the irradiation position is not known with sufficient accuracy and usually not constant along the irradiation tube. This has the consequence that standards have to be irradiated simultaneously with the real samples.

Detection limits which may be reached with NAA for a 10 min irradiation with 10^{13} n/cm^2 s are summarized in Table I. They base on an activity of 10 Bq after a Δt of 20 s (a typical minimum time expected for transportation and mounting of the samples in front of a detector at the new device NAA/SINQ). However, it has to be kept in mind that the detection limit might be higher if no convenient γ -line is available for detection. Moreover, if less than 1g of material is used, the detection limit has to be corrected accordingly.

Table I: Detection limits for several elements in a 1g sample under typical SINQ conditions and an irradiation time of 10 min (see text).

Element	Detection limit in ppb	Element	Detection limit in ppb
Mg	20	Rh	0.002
Al	0.2	Ag	0.01
Cl	3	Cd	100
Ca	70	In	0.03
Sc	0.02	Sn	50
Ti	20	I	0.2
V	0.02	Ba	10
Cr	10	Pr	0.2
Co	0.01	Nd	7
Cu	0.2	Sm	0.2
Ge	3	Gd	2
Se	0.2	Dy	0.001
Br	0.2	Hf	0.04
Rb	15	Re	1
Nb	2	Pt	5
Mo	20	Hg	10

1 ppb = 1 part per billion = 10^{-9} g/g

At BLASIUS typical applications of NAA were

- trace analysis of heavy metals in the system atmosphere-biosphere(forest)-soil,
- determinations of several essential elements in food,
- Se concentrations in blood plasma or Cu in liver tissues (Wilson disease),
- absolute mass determinations of silver aerosols,
- studies of halogen profiles in meteorites,

- irradiations of single crystals doped with ^{235}U for diffusion studies of fission products,
- La determinations for industry (EMS company) etc.

With the new NAA/SINQ facility the samples will be transported within less than 10 s to the radiochemistry laboratory at PSI West (WBGA/C41). There the tubes (polyethylene, dia. 25 mm, length 48 mm) containing the samples will be collected in a shielded container placed inside a hood. After dismounting, the samples will be measured with HPGe detectors placed at a distance of a few meter from the hood. For informations on γ -spectroscopy see [5]. Nuclides with half-lives as short as about 10 s may be determined.

With this new facility similar applications are scheduled as for the previous BLASIUS device. The emphasis will most probably lie on aerosol studies, since NAA as a non-destructive multi-element detection method is especially powerful as a „finger print“ technique to investigate source regions of such particles. Other fields of future interest may be heavy metal trace analysis in samples from solid state chemistry or from environmental samples as well as rare earth determinations in e.g. meteorites. It's also planned to increase NAA applications for external users, both for research (e.g. earth sciences or materials sciences at universities and research centres) as well as for industry. Last but not least applications of this facility for educational purposes should be mentioned.

4. Radioisotope Production via Neutron Activation (PNA)

This chapter bases on informations made available to the author by P. Bläuenstein [6]. Production of labelled radiopharmaceuticals requires access to radionuclides with high specific activity, in some cases of carrier-free species. Of great interest are nuclides which can be incorporated into molecules with high tumor affinity. Optimum are nuclides with half-lives of similar values as the time periods these molecules are biologically active, usually about 0.5 to 5 days. For therapy applications weak β^- , Auger electron or even α -emitters are used due to their high local energy deposition. For monitoring purpose it is useful if these nuclides emit γ -rays, however with low probability, to reduce whole body irradiation. Total activities of up to 20 GBq may be applied to a single patient (e.g. of ^{131}I -meta-iodbenzylguanidin for neuroblastom therapy).

In case of diagnostic applications much lower activities are used, typically on the order of 10 to 100 MBq per patient.

Typical radionuclides used at PSI and to be produced with SINQ/PNA are ^{131}I ($T_{1/2} = 8.0$ d) from nuclear fission of ^{235}U , and ^{111}Ag ($T_{1/2} = 7.45$ d), produced via neutron activation of ^{110}Pd and subsequent β^- -decay and ^{186}Re ($T_{1/2} = 3.8$ d) from neutron activation of ^{185}Re .

5. Fission product gas-jet system (GJA)

Thermal neutron induced fission of ^{235}U produces a well known double-humped distribution of fission nuclides (Fig. 1). The so-called light fission fragments are centered around mass numbers (isobars) $A = 95$ and the heavy ones around $A = 138.5$ (the difference of about 2.5 mass units between the sum of both values and the mass of the fissioning nucleus, ^{236}U , is caused by evaporation of prompt neutrons). The corresponding second

moments of both mass peaks are rather high with 15 and 22 mass units for the light and heavy mass peak, respectively, which leads to a broad range of elements being produced as primary fission fragments. They are, at a level of $\geq 1\%$ independent fission yield [4], Se, Br, Kr, Rb, Sr, Y, Zr, Nb, Mo and Tc in the light peak and Sb, Te, I, Xe, Cs, Ba, La, Ce, Pr and Nd in the heavy peak. The primary products are neutron rich isotopes of the corresponding elements which have short half-lives of typically a few seconds to minutes. Via β -decay chains long lived secondary fission products are formed. Well known examples of such secondary products are ^{137}Cs , ^{90}Sr or ^{131}I .

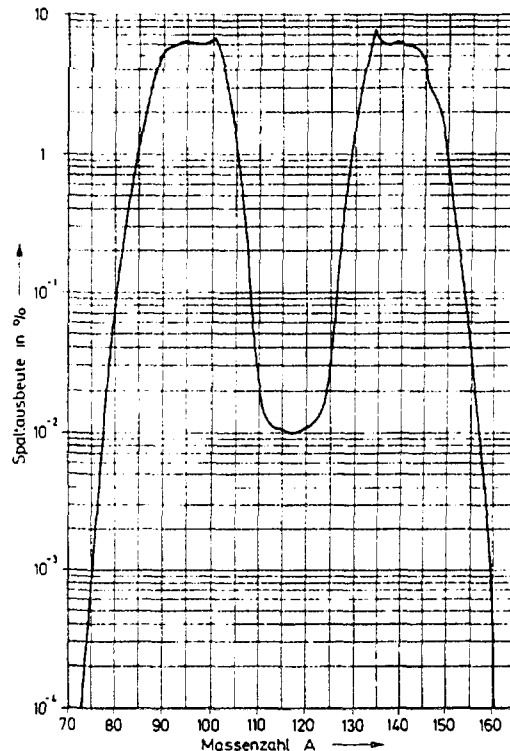


Fig. 1: Mass distribution from thermal neutron induced fission of ^{235}U [7].

The average total kinetic energy (TKE) released to both fission fragments is, for thermal neutron induced fission of ^{235}U , 168.3 ± 1.7 MeV. Due to moment conservation, this TKE is divided to the fragments indirectly proportional to the fragment masses, i.e. the light fission fragments have average kinetic energies of about 100 MeV and the heavy ones of about 69 MeV. This has the consequence that fission products from the light mass peak have higher recoil ranges in a material than those of the heavy mass peak.

Fission product gas-jet facilities use ^{235}U targets of ≤ 1 mg/cm² thickness. In this case most of the fission products escape from the target. They can then be thermalized in a gas volume. Typical ranges for fission products in gases - under NTP condition - are about 1 to 2 cm in Ar and N₂ and about 15 cm in He. To design a device which should make available fission products from the light mass peak only the target has to be covered by a degrader foil of appropriate thickness.

Usually, the primary products attach via diffusional processes and adsorption to aerosol particles contained in the gas. Mostly salt or graphite particles of about 20 to 100 nm size are used for this purpose. If the carrier gas containing the aerosol particles is continuously flushed through such a collection chamber, the fission products can be continuously transported from

the production site along thin capillaries (typically with 1 - 2 mm diameter) over long distances to a laboratory. The relatively large aerosol particles have low diffusion coefficients which prevents losses caused by surface interactions with the wall of the capillary. On the other hand, gravitational sedimentation velocities are still small, preventing losses due to this effect.

At SINQ a set of three different production chambers will be installed (Fig. 2). All three chambers contain two ^{235}U targets, one at the front side and one at the rear side. Each target contains 2.5 to 3 mg ^{235}U . This yields a fission rate of about 10^7 s^{-1} in each chamber, assuming a thermal neutron flux of 10^9 s^{-1} and a fission cross section of 584 barn. To adjust the real fission rate to the needs of the experiment, a tunable mechanical shutter will be placed in front of the chamber array which will allow to vary the neutron beam impinging onto the targets between 0 and 100 %.

The sizes of the chambers are adjusted to the following applications:

Chamber 1 : All fission products in Ar or N₂. No cover foil.

Chamber 2 : All fission products in He. Target covered by a 3 μm thick Ni cover foil to pre-degrade fission fragment energy. This is required to reduce the total recoil range in He to the dimensions of the chamber.

Chamber 3: Light fission products in Ar or N₂. Target covered by a 6 μm thick Ni cover foil to stop entirely the fission fragments from the heavy mass peak.

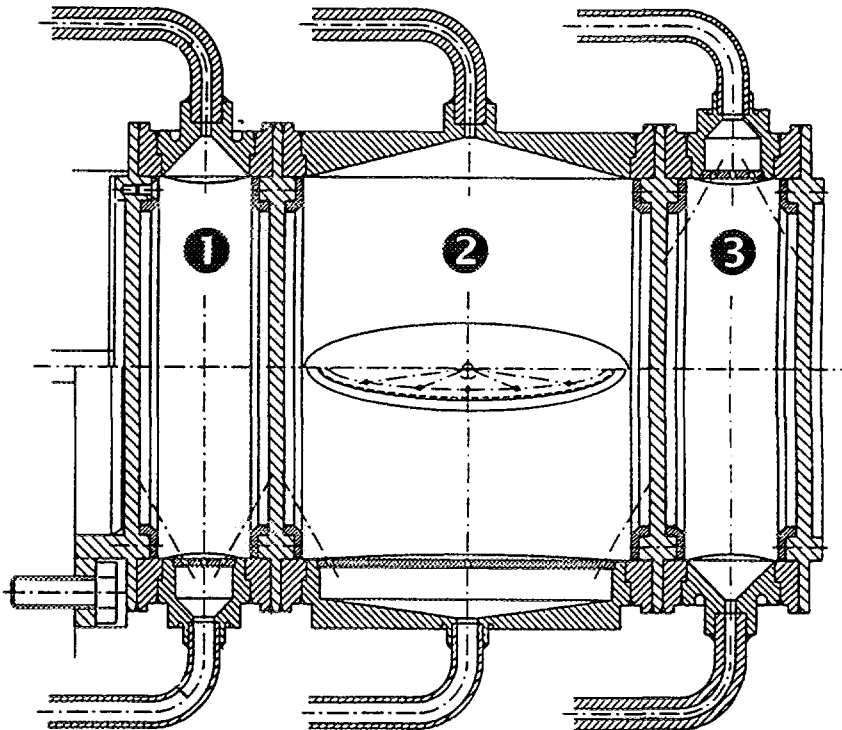


Fig. 2: Layout of the array of the 3 SINQ gas-jet chambers (see text)

The reason for including chamber 3 is initiated by the fact that some fission products from the heavy mass peak have very complicated γ -spectra. This may hamper detection of some nuclides from the light mass peak. An example of a γ -spectrum measured in a gas-jet device with degrader foil is depicted in Fig. 3 [8].

In a first series of experiments it is planned to run modeling experiments with the nuclides ^{105}Mo ($T_{1/2} = 37$ s) and ^{106}Tc ($T_{1/2} = 36$ s) to develop a fast separation device for studying the chemistry of element 106 (Seaborgium, Sg) and, for the first time, of element 107 (Nielsbohrium, Ns). Mo and Tc belong to the same groups in the periodic table as Sg and Ns, respectively.

Another planned application intends to produce brominated organic molecules, labelled e.g. with ^{87}Br ($T_{1/2} = 56$ s), to investigate heterogeneous surface interactions with supercooled droplets, a topic of great current interest in the context of the ozone depletion problem in the stratosphere.

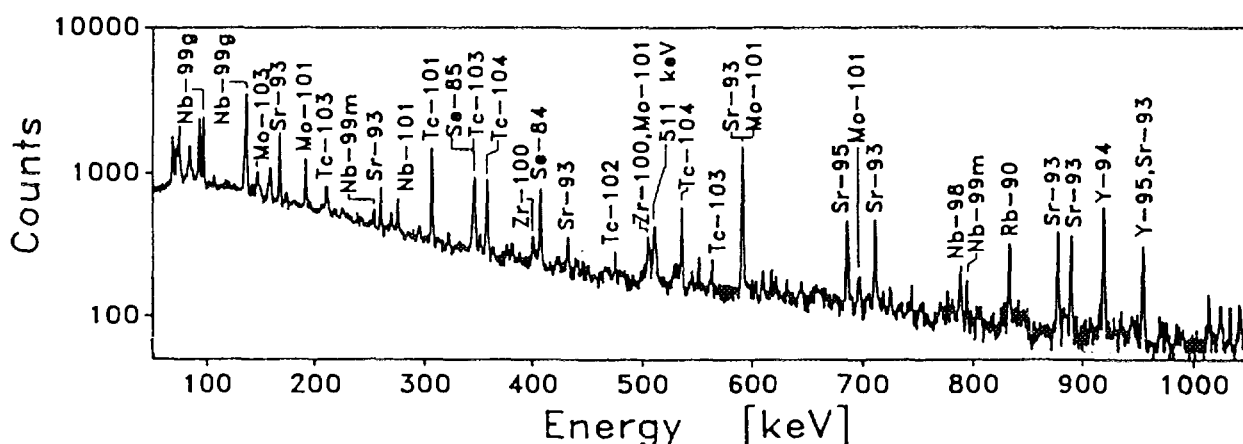


Fig 3: γ -spectra of fission products from a gas-jet device containing a target covered by a 12 μm thick Al foil (from ref. 8). Only fission products from the light mass peak are visible.

References

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