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Corresponding Author: Dr. László Szentmiklósi, Ph.D.

Corresponding Author's Institution: Institute of Isotopes, Hungarian Academy of Sciences

First Author: László Szentmiklósi, Ph.D.

Order of Authors: László Szentmiklósi, Ph.D.; Tamás Belgya, Ph.D.; Zsolt Révay, D.Sc.; Zoltán Kis, Ph.D.

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Suggested Reviewers: Jan John
Professor, Department of Nuclear Chemistry, Czech Technical University in Prague
Jan.John@fjfi.cvut.cz
organizer of the 16th Radiochemical Conference

Jan Kucera
Associate Professor, Nuclear Physics Institute AS CR
kucera@ujf.cas.cz
organizer of the 16th Radiochemical Conference

Upgrade of the Prompt-Gamma Activation Analysis (PGAA) and the Neutron Induced Prompt-gamma Spectroscopy (NIPS) facilities at the Budapest Research Reactor

László Szentmiklósi*, Tamás Belgya, Zsolt Révay, Zoltán Kis

Institute of Isotopes, Hungarian Academy of Sciences, H-1525 Budapest, P.O. Box 77, Hungary

* Corresponding author.

E-mail: szentm@iki.kfki.hu

Tel: +36-1-392-2539

Fax: +36-1-392-2584

Abstract

The Budapest Research Reactor's Prompt-Gamma Activation Analysis (PGAA) and Neutron Induced Prompt-gamma Spectroscopy (NIPS) facilities were significantly upgraded during the last few years. The higher neutron flux, achieved by the partial replacement and realignment of the neutron guides, made feasible the automation and specialization of the two experimental stations. A new neutron flux monitor, computer-controlled beam shutters and a low-level counting chamber have been put into operation to assist with in-beam activation experiments. An automatic sample changer has been installed at the PGAA station, while the NIPS station was redesigned and upgraded with a Compton suppressor to use for the non-destructive analysis of bulky samples. In the near future the latter setup will be completed with a neutron tomograph and a moving table, to turn it into a Neutron Radiography/Tomography-driven PGAA equipment.

Keywords: prompt gamma activation analysis, neutron induced prompt gamma spectroscopy, low-level counting chamber, sample changer, neutron monitor, in-beam activation

Introduction

Since starting its operation in 1996, several major upgrades were accomplished at the Budapest prompt-γ facility in order to improve its performance and productivity. This is in line with the trends at other PGAA facilities [1-3]. From 1996 to 2000 a thermal neutron beam was available at the Budapest Research Reactor with a flux of about 2×10^6 $cm^{-2}s^{-1}$ [4,5]. Following the installation of the cold neutron source at one of the reactor's tangential channels [6] in 2000 and the modifications made in the neutron guide system, the beam flux increased substantially to 3×10^7 $cm^{-2}s^{-1}$. At the end of the neutron guide the rectangular beam was split into two square sub-beams to serve two independent experimental stations. The prompt-gamma activation analysis (PGAA) facility was dedicated to chemical analysis; while the neutron-induced prompt gamma-ray spectroscopy (NIPS) facility was used for advanced nuclear spectroscopic measurements, including γ-γ coincidence studies [7]. This configuration was operational between 2001 and 2006. The most important feature of that system was the exceptionally low beam-background [8], which provided ideal conditions to establish a library of radiative neutron capture gamma-rays for chemical analysis [9]. In 2003 a beam-chopper was added to the PGAA setup for studies on short-lived radionuclides [10-12] produced during the irradiation.

The last significant upgrade started in 2007, when the last section of the neutron guide No. 1 was changed to $2\theta_c$ supermirror elements [13], increasing the thermal equivalent neutron flux to 1.2×10^8 $cm^{-2}s^{-1}$ at the PGAA and 7×10^7 $cm^{-2}s^{-1}$ at the NIPS sample positions, respectively. A new 27-% relative efficiency HPGe detector with an elongated crystal was purchased that fit into the existing PGAA Compton suppressor. New components of the data acquisition electronics were also put into operation. A factor of 20–50 gain in the neutron flux, compared to the performance of the original thermal system, made it possible to significantly reduce the beamtime needed to measure a sample.

It was soon realized that a more efficient way of operation under these new conditions requires a higher level of automation and specialization of the two facilities. This is dictated by the increasing demand from international scientific collaborations (e.g. EU FP6 ANCIENT CHARM [14]), EU facility access programs (such as EU FP7 NMI3, EFNUDAT, CHARISMA), and commercial

measurements. Therefore it was decided to turn the PGAA facility into a fully-automated measuring system for small samples (powders, slabs, liquids in vials), while NIPS would serve for the non-destructive analysis of bulky objects, including cultural heritage objects and material science samples. The option will be however kept open to perform here diverse experiments with custom setups.

In order to achieve these research aims, several existing components of the system have been replaced and a few new features were recently added. These will be discussed in the paper.

Instrumental

Improvement in the beamline

There is a primary (pneumatic) instrument shutter at the end of the neutron guide No. 1, followed by two (electrically actuated) secondary shutters for the two experimental stations. These secondary shutters have been completely rebuilt. The device is now computer-controlled and uses linear motors to achieve faster opening and closing times (about 350 ms). The shutter plates are coated with two neutron absorber layers (B_4C-loaded rubber plus a Cd sheet). Each plate has a 20×20 mm^2 rectangular aperture to define one of the two sub-beams from the originally 100×25 mm^2 beam spot. Additional ^{6}Li-poly collimators and a borated paraffin block are placed into the flight tube downstream to filter the divergent or scattered neutrons before they reach the sample chambers. This will be replaced with a lead block in the near future to better shield against the gamma-radiation propagating along the beamline.

The controller of the beam shutter can be commanded from either of the two data acquisition computers via an Ethernet network. In addition to the standard on-off use, another aim was to use the shutters as a “beam modulator”, i.e. to open and close the beam according to a predefined time pattern. This mode of operation is an extension of the chopped-beam PGAA concept [11, 12] towards longer, and not necessarily equal, irradiation and counting periods. The collection of gamma spectra can be synchronized with the status of the beam shutter. Thus – in combination with either conventional MCA event processing or with list-mode

data acquisition – the time dynamics of the growth and decay of radionuclides produced during the irradiation can be better investigated.

Whenever in-beam activation experiments were done so far, a constant incoming beam flux was assumed. In order to further improve the accuracy of our nuclear data and to test the validity of this assumption, a neutron flux monitor has been purchased and installed downstream of the beam shutters. The ORDELA Model 4511 N has an active surface of 110×25 mm^2, and it is filled with a mixture of N_2 and CF_4 gases. Its counting efficiency was chosen to be as low as 10^{-6}, to provide a count rate of about 1000 cps. The monitor TTL output is counted with a NI-6601 PCI counter/timer card, and is displayed and recorded with a time-resolution of one second over the whole 10-day long reactor cycle.

The biological protection for the personnel has also been improved by placing two more shielding blocks of concrete between the beam area and the personnel working area. This was necessary due to the increased dose rate produced by the upgraded beams. In parallel, the measurement cabin has been extended to offer more room for work and sample manipulation. The new layout of the experimental area is depicted in Figure 1.

Figure 1.

Prompt-gamma activation analysis (PGAA) station

The PGAA station uses the upper of the two neutron beams. A new automatic sample changer has been manufactured that is exchangeable with the existing (manually operated) sample chamber. It has a rotating cylindrical stage 40 cm above the beam with 16 radial slots for the sample frames. A vertical slider moves the selected sample down to the measurement position and removes it after the irradiation. The sample holders can accommodate objects with dimensions of about 70×70×5 mm^3. The whole chamber is lined with two layers of ^{6}Li-loaded polymer to keep the beam-background low. This double thickness of the material gives a neutron attenuation factor of about 1000. The lower beam passes undisturbed through the chamber towards the NIPS station, located about 1 m downstream of the PGAA sample position.

A user-friendly facility control program, "*Budapest PGAA Data Acquisition Software*", has been written for manual, semi-automatic, and unattended automatic batch measurements. This program controls the beam shutters, the sample changer and the data acquisition in a Canberra AIM 556 network acquisition module. The stopping condition of the batch run can be preset time or count; whichever is fulfilled earlier stops the measurement. The parameters of the batch run are described in a text file called 'Batch Input Table'. In order to comply with the recent quality management standards, a detailed book-keeping of the acquisition events and key instrument parameters is implemented.

Neutron-induced prompt gamma spectrometry (NIPS) station

The NIPS facility had a small sample chamber in order to allow coincidence measurements with close sample-to-detector geometry. Now it is being replaced with a dismountable and larger chamber with dimensions of 200×200×200 mm^3, similar to one successfully used in our PGAI/NT experiments [14]. The top, the bottom and one side of the chamber are all removable, so that oversized objects can also be easily irradiated here. The redesigned NIPS facility is expected to be completed with a moving table and a neutron tomograph by the end of year 2010, to turn it once again into a PGAI/NT-capable experimental station.

The vacuum system of the NIPS flight tube section is also going to be separated from the upper part, making the operation of the two facilities fully independent. The NIPS detector and the shielding are moved to the other side of the flight tube (relative to the PGAA detector) to allow easier access to the NIPS sample chamber and to reduce the cross-talk between the two facilities.

Both active and passive components of gamma shielding are now employed to achieve a low background at the NIPS station. The active component is a new bismuth germanate (BGO) Compton suppressor. It was designed using extensive Monte Carlo calculations, using the computer program MCNP-CP [15]. The shape of the BGO suppressor was optimized to reach the best peak-to-total ratio in the gamma spectrum, up to 10 MeV. It was concluded that the optimum length of the BGO side element is 170 mm with a well depth of 150 mm and a well diameter of 78 mm. The minimum thickness of the BGO side element is

found to be about 40-45 mm, whereas 30 mm is sufficient at the front region, where – due to the nature of Compton scattering – only low-energy gammas have to be detected. The passive shielding is made up using standard lead bricks. The installation of the upgraded NIPS detector system is still in progress.

Low-level counting facility

To better assist the in-beam activation measurements, to perform the off-line counting of samples [16] on a routine basis and to enable the measurement of environmental samples with low activities during the reactor shutdown periods, a permanent low-level counting facility (referred as DÖME) has been established (see also in Figure 1), in cooperation with the KFKI Atomic Energy Research Institute. This consists of an iron chamber manufactured from pre-WW2 steel, and therefore is free of any man-made radioactivity. The internal dimensions of the chamber are 800×800×800 mm^3, so that a Canberra GR1319 HPGe detector with a Big MAC cryostat can be placed along its horizontal diagonal. This geometry allows sample-to-detector distances up to 250 mm. The wall thickness of the chamber is 155 mm and has a graded shielding inside (Cd and Cu layers). The weight of the chamber and its support exceeds 7 metric tons. As an option, a Canberra Low Energy HPGe detector is also available if the main emphasis is not the detection limits, but low-energy lines are to be measured with a better energy resolution.

Results and Discussion

The intensity profile of the beam after reactor startups

During the first 6 hours following any reactor startup, only a thermal beam is available and its flux increases by about a factor of six when the cold neutron source becomes fully operational. This known time behavior could be recorded now with a proper time resolution, using the recently installed neutron monitor, and is depicted in Figure 2. The curve is in general agreement with the earlier measurements (using the gamma-peak count rate from a Ti foil) [17], but is able to reveal all fine details, e.g. the jumps due to changes in the reactor power. After the steady-state condition is reached, the beam intensity becomes almost constant

over longer periods of time, and the monitor counts follow closely the Poisson distribution. For a high-accuracy activation measurement, one can use the pointwise data stream and solve numerically the *Bateman-Rubinson* differential equations at each time bin, rather than using a time-averaged flux.

Figure 2.

Pilot experiment with the programmable beam shutter

To demonstrate the capabilities of the new computer-controlled beam shutter and the concept of the "beam modulation", a pilot experiment has been carried out. A sample containing Al and V was irradiated in nine activation and counting cycles with successively increasing cycle times, from 1 minute to 9 minutes. The gamma-rays from the radiative neutron capture and from the radioactive decay were recorded with the list-mode feature of our XIA Pixie4 digital spectrometer. The events were processed with a home-made computer program implementing the procedure described in Ref. [18]. The measured peak areas of the ^{28}Al and ^{52}V radionuclides are plotted as a function of time in Figure 3, together with the calculated responses based on the activation equations and literature half-lives. A good agreement between the experimental and the expected curves was found that confirms the feasibility of such measurements.

Figure 3.

Background measurements with the low-level counting chamber

It was found that the shielding of the newly established low-level counting facility reduces the energy-integrated γ-ray background by more than two orders of magnitude compared to the room background (211 cps). It is as low as 1.305±0.002 cps while the reactor is on and 1.116±0.002 cps if the reactor is shut down, over the energy range of 7-3150 keV. Only the count rate of the annihilation peak and the level of the continuous background differ significantly (approx. 0.2 cps) between beam-on and beam-off periods. The count rates of the characteristic background peaks are listed in the Table 1. The values for the bare

HPGe detector and for the PGAA Compton-suppressed spectrometer are also given as a comparison:

Table 1.

Conclusions

After the significant upgrade, the Budapest PGAA and NIPS facilities offer improved possibilities for routine element analysis and also for advanced research. The intense neutron beams having no epithermal component, the neutron flux monitor, the computer-controlled beam shutter and the low-level counting chamber make us able to study better the activation and decay of the in-beam produced radionuclides. The new Compton suppressor at the NIPS station makes the spectroscopic conditions similarly favorable to what was previously available only at the PGAA station. Due to the higher level of automation and the rewritten acquisition softwares, the productivity of each experimental station increased substantially. Both facilities will continue to serve national and international collaborations within the EU FP7 NMI3 and many other projects.

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Table caption:

Table 1. The comparison of the beam-off background conditions inside the DÖME counting chamber with the data for the same bare HPGe detector outside, and also with the PGAA Compton-suppressed spectrometer [5]. (mcps = 10^{-3} cps) The values marked with * could not be reliably evaluated due to a gigantic background bump.

Figure captions:

Fig. 1. The 3D sketch of the Budapest PGAA-NIPS and DÖME facilities.

Fig. 2. The change in the incoming neutron flux at the PGAA station, following the reactor startup.

Fig. 3. The response of the V and Al radionuclides to successive irradiations with increasing cycle times.

Table 1

Energy (keV)	Origin	DÖME bkg (mcps)	Bare HPGe room bkg (mcps)	PGAA room bkg (mcps)
46.2539(1)	^{210}Pb(β^-)^{210}Bi [^{238}U]	8.1	–*	16.4
53.2275(21)	^{214}Pb(β^-)^{214}Bi [^{238}U]	0.7	–*	1.3
63.29(2)	^{234}Th(β^-)^{234}Pa [^{238}U]	11.2	–*	9.3
74.9	Pb-X $K_{\alpha1}$, Bi-X $K_{\alpha2}$	1.7	327	14.3
77.107	Bi-X $K_{\alpha1}$	2.3	176	18.3
84.373 (3)	^{228}Th(α)^{224}Rn [^{232}Th]	1.3	119	2.8
92.38 (1) + 92.80 (2)	^{234}Th(β^-)^{234}Pa [^{238}U]	15.8	207	13.3
143.76 (2)	^{235}U(α)^{231}Th [^{235}U]	1.3	7	1.2
185.715(5) + 186.211(13)	^{235}U(α)^{231}Th [^{235}U]; ^{226}Ra(α)^{222}Rn [^{238}U]	7.3	87	5.7
238.632 (2)	^{212}Pb(β^-)^{212}Bi [^{232}Th]	2.0	675	3.5
295.997 (3)	^{214}Pb(β^-)^{214}Bi [^{238}U]	1.5	112	0.3
352.932 (2)	^{214}Pb(β^-)^{214}Bi [^{238}U]	2.8	206	0.7

511 (beam on)	Annihilation	17.6	–	–
511 (beam off)		13.1	342	0.6
583.191 (2)	$^{208}Tl(\beta^-)^{208}Pb$ [^{232}Th]	0.6	489	0.4
609.312 (7)	$^{214}Bi(\beta^-)^{214}Po$ [^{238}U]	2.0	190	0.6
661.660 (3)	$^{137}Cs(\beta^-)^{137}Ba$	0.4	47	0.3
767.36 (2)	$^{234m}Pa(\beta^-)^{234}U$ [^{238}U]	0.3	17	0.2
911.205 (4)	$^{228}Ac(\beta^-)^{228}Th$ [^{232}Th]	0.4	381	0.7
968.971 (10)	$^{228}Ac(\beta^-)^{228}Th$ [^{232}Th]	0.3	228	0.5
1001.03 (3)	$^{234m}Pa(\beta^-)^{234}U$ [^{238}U]	0.3	17	0.5
1120.287 (10)	$^{214}Bi(\beta^-)^{214}Po$ [^{238}U]	0.4	51	0.3
1460.83 (5)	$^{40}K(\beta^+ + EC)$	1.0	590	4.9
1764.494 (14)	$^{214}Bi(\beta^-)^{214}Po$ [^{238}U]	0.3	45	0.5
2614.551 (13)	$^{208}Tl(\beta^-)^{208}Pb$ [^{232}Th]; $^{212}Po(\alpha)^{208}Pb$ [^{232}Th]	0.6	312	1.1

colour figure 1

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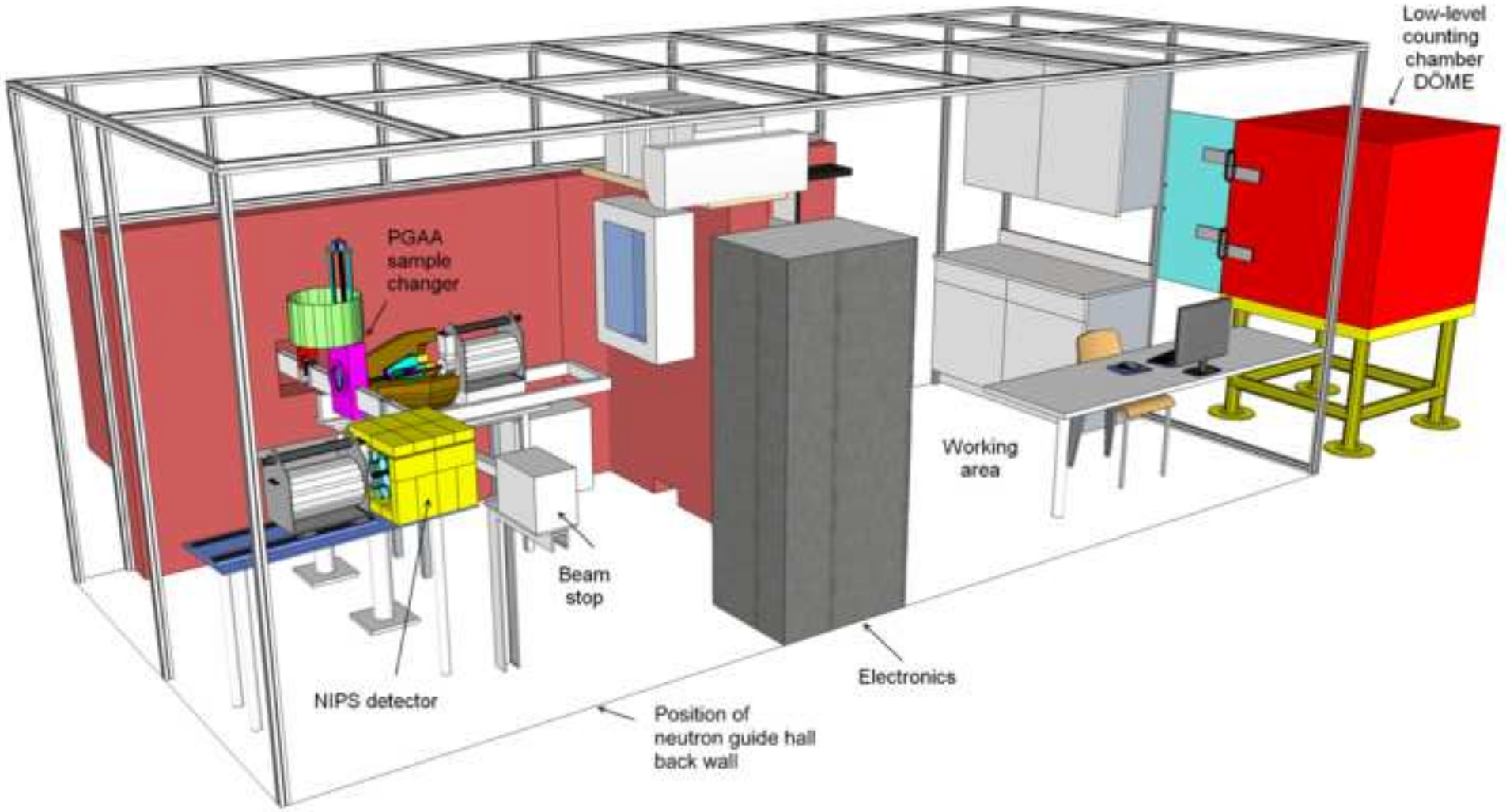

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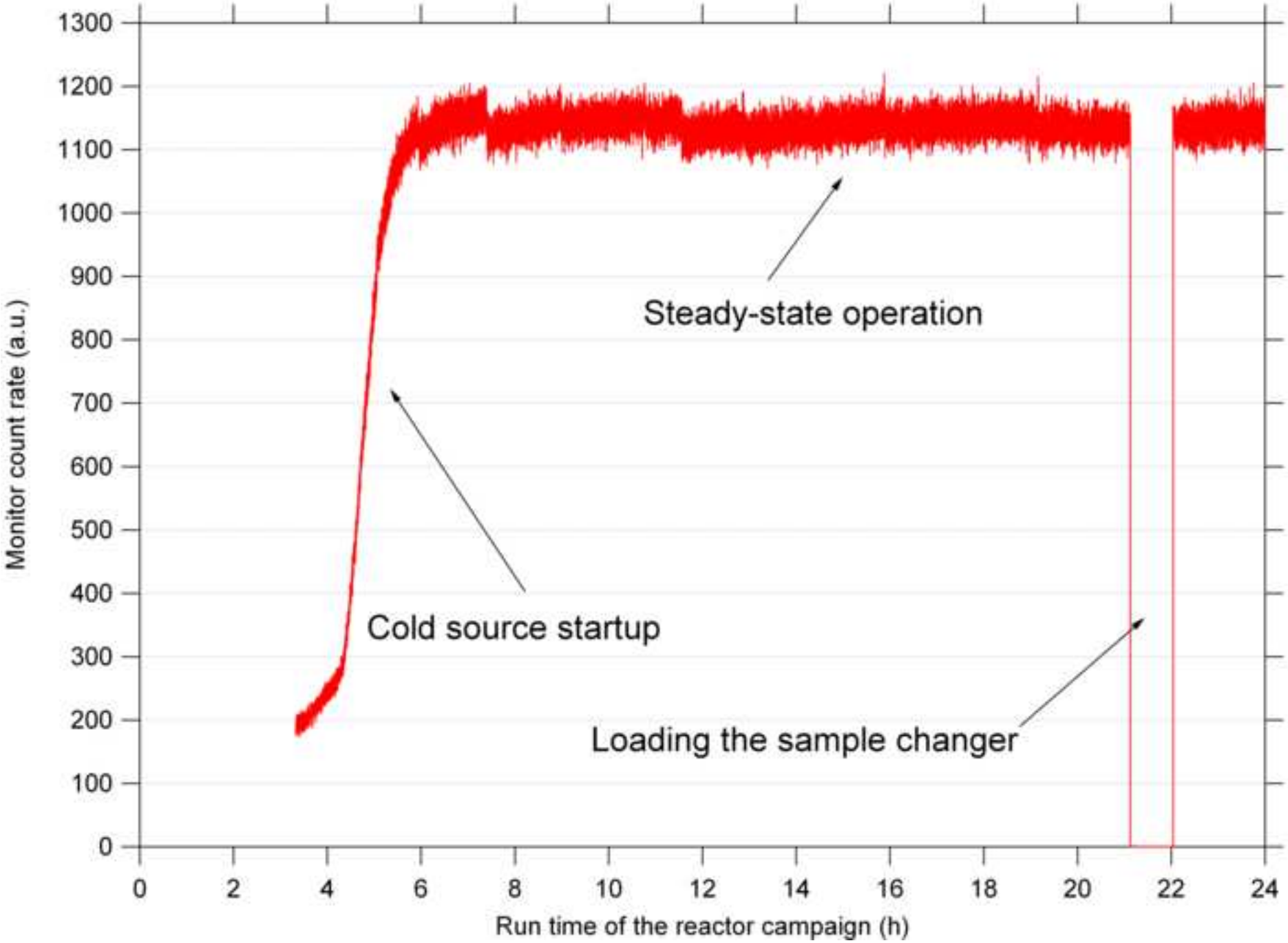

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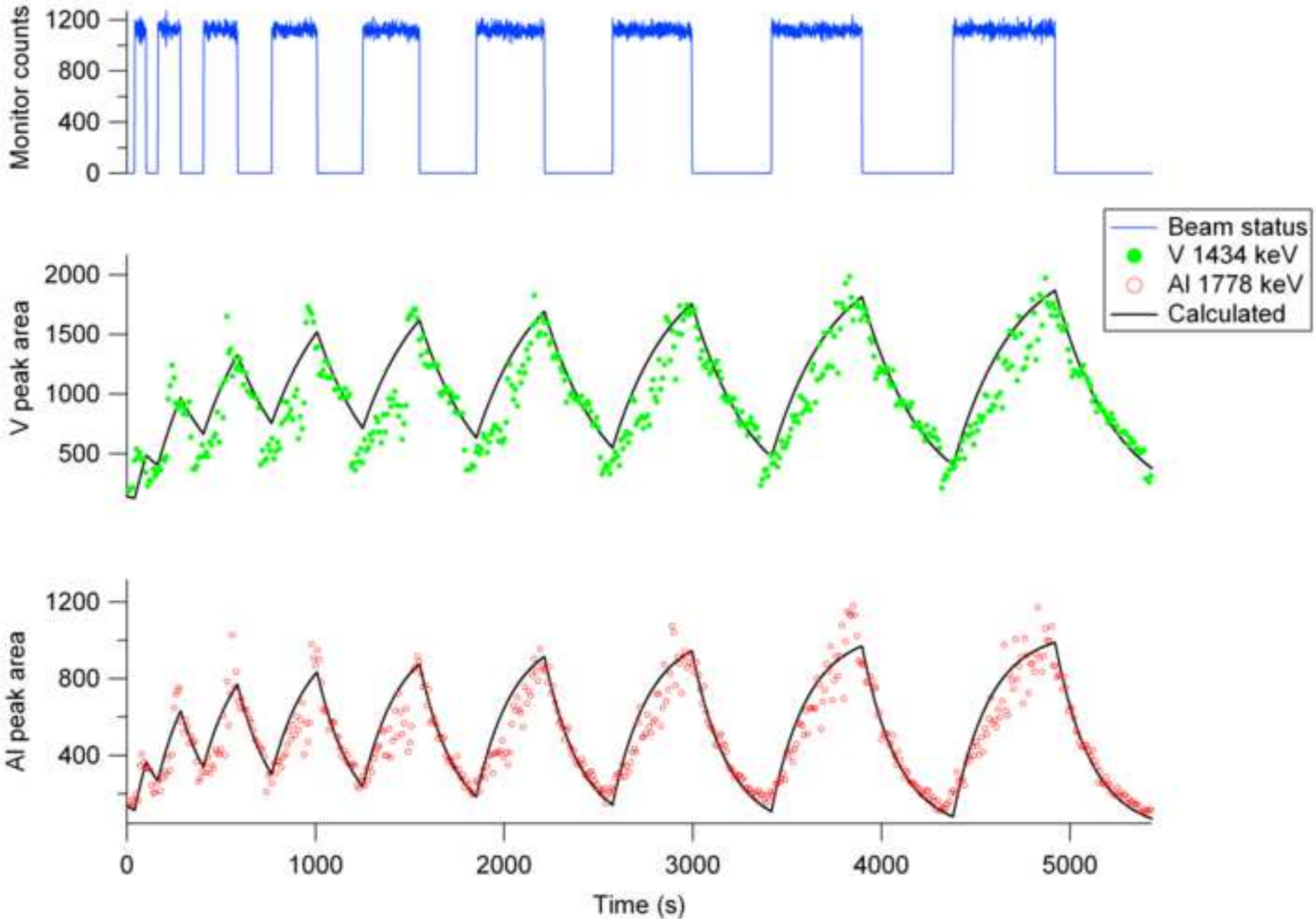

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Name: LÁSZLÓ SZENTMIKLÓSI
Address: 29-33 KONKOLY-THEGE STREET, BUDAPEST, HUNGARY
E-mail address: SZENTM@IKI.KFKI.HU

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