

NEUTRON ACTIVATION ANALYSIS AS A FOREIGN INTRUSION CAVITY DETECTION TOOL

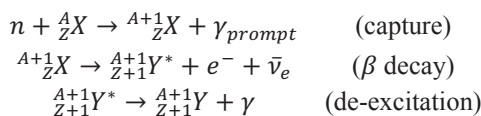
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Abstract

Neutron Activation Analysis (NAA) is one of the currently available techniques used to determine contaminants in Nb superconducting cavities, allowing a non-destructive determination of foreign materials, provided they have radioactive isotopes with a sufficiently long half-life. We present the NAA technique application with the goal of contaminants determination, identification and localization for the European XFEL 3rd harmonic cavities (3.9 GHz). Irradiation and analysis has been performed in collaboration with the LENA nuclear reactor (Pavia, Italy) and the University of Milano Bicocca. The main difference respect to the measurements performed in the past is the goal to apply of the NNA directly to entire cavities and not to material samples. Currently nine samples were exposed to thermal and fast neutron flux and the resulting activity was measured with HPGe detectors.

NEUTRON ACTIVATION ANALYSIS TECHNIQUE

Instrumental Neutron activation analysis is a technique of analysing (either qualitatively or quantitatively) the atomic composition of non-radioactive materials [1]. This process consists in the production of unstable isotopes by means of neutron absorption by the nuclei in the sample under study. In a neutron-irradiated material several nuclear reactions can take place, namely (n, p) , (n, α) , $(n, 2n)$ for fast neutrons and (n, γ) for thermal neutrons. Among these the most used is the last one for which n is the absorbed nucleus and γ is the prompt photon emitted when the nuclear reaction takes place, with typical energies in the 6-8 MeV range, according to the binding energy of the captured neutron. The newly formed nucleus has one neutron in excess and often undergoes β decay. The overall process can be summarized by the following reactions:



The β decay takes place according to the radioactive exponential decay law $N(t) = N_0 e^{-\lambda t}$. Usually the daughter nucleus of the decay is formed in an excited state, thus de-excitation gamma rays are emitted [2]. Thought the readjustment is governed by the electromagnetic force, the γ rays are emitted in pretty short times (usually of the

order of 10^{-12} s). It is thus acceptable to consider the β particles and potential γ emission synchronous.

The NAA can therefore be performed according the the following steps:

- the material sample exposition to a thermal neutron flux for a known time,
- the irradiated sample extraction and measurement of the induced γ radioactivity,
- the quantitative determination of each selected element, knowing the reaction cross section, the photon energy and detection efficiency.

To allow quantitative elemental composition it is necessary to know the irradiation neutron flux and the natural percentage of the precursor isotope, for background discrimination. In order to measure the number of activated atoms the gamma spectroscopy technique is typically used, because it is easy to identify the elements whose nuclear de-excitation γ are characteristics and monochromatic. β spectroscopy is not suitable due continuous spectrum and the inability to discriminate backgrounds.

The NAA offers several advantages:

- it is a multielemental analysis technique, because it allows to simultaneously identify the presence of several elements within the irradiated sample.
- it allows the measurements of independent parameters as the γ energy and the mean life of the radionuclide.
- it offers a high sensitivity for most the elements identifiable with this method.

There are some limits to the application of this technique: for example the element for which a quantitative determination is needed must appear in the sample with a sufficient abundance, and with a sufficient cross section for thermal neutron activation, otherwise the detectability of the few neutron activated atoms would be impossible. Moreover, the half-life should be at least enough long enough to allow the isotope not to decay during the time between the irradiation and the spectroscopic measurements. Finally the radioisotope can not be a pure β emitter, for which the gamma spectroscopy setup is useless.

Element Mass Calculation & Reference Standard SMR Usage

Quantitative elemental determination requires the use of standard samples, containing known amounts of the elements under study, used for relative measurements.

Once the sample and the standard undergo the same irradiation and detection conditions the irradiation time,

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the neutron flux (ϕ) and the detector efficiency at a particular energy (ϵ) are equal for the measurements. Then the analysis consists of comparing the peaks in the standard and in the sample spectra, considering some parameter equal for both spectra (as the branching ratio BR, the radioactive decay constant λ , etc...). Moreover, the irradiation and measurement time are the same for the sample and the standard. The overall considerations lead to the following expression for m , the mass concentration of the unknown material:

$$m = m_{STD} \frac{n_{\gamma}^{riv}}{n_{\gamma,STD}^{riv}} e^{-\lambda(t_{a,STD}-t_a)}$$

where n_{γ}^{riv} is the number of detected gamma, t_a the time elapsed from the irradiation to the measurements and the *STD* suffix indicates the corresponding quantities measured for the standard sample of known concentration m_{STD} .

TRIGA MARK II REACTOR AS A NEUTRON SOURCE

As discussed before for the NAA to be feasible it is necessary to induce radioactivity in the nuclei of some isotopes as a result of a thermal neutron capture process. It is thus necessary a thermal neutron source, which usually is an expensive tool. A possible solution is the use of a nuclear reactor as a source: after the fission reactions the produced neutrons are moderated down to thermal energy (0.25 eV), so the fission process which produces the neutrons autofeeds itself. To achieve this goal the reactor *Triga Mark II* operated by L.E.N.A. (Nuclear Applied Energy Laboratory) of the University of Pavia (Fig. 1) was used. LENA is a 250 kW research reactor in a stationary regime.

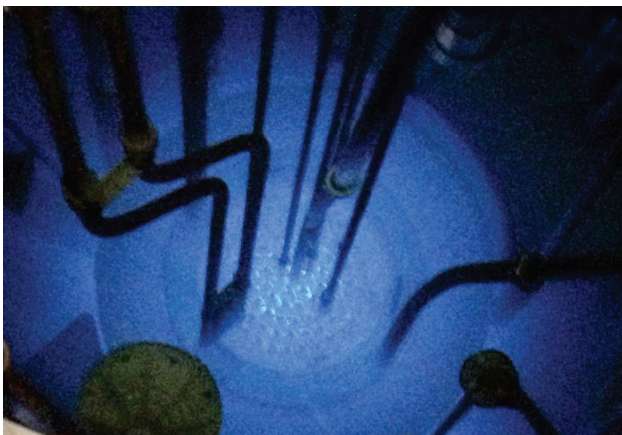


Figure 1: The LENA reactor core, when active.

The reactor *core* (Fig. 2) is contained in an aluminium recipient (*tank*) and installed on the pool floor, full of water, with a 2 m diameter and a 6.25 m depth. The fuel is a mixture of ZrH and enriched uranium ^{235}U at 19.75%, in the form of stainless steel clad bars. In each bar the fuel is 38 cm long, at the beginning and at the end of each

element there is graphite neutron reflector. The core is equipped with control bars, whose aim is to absorb fission produced neutrons. When these are fully inside the reactor it is operated in a *subcritical* regime (neutrons used for the fission are less than the ones produced). The reactor is said to be operated in overcritical regime when the opposite configuration just described takes place. The source used to activate the reactor is a Ra-Be source with an activity of 10^8 n/s.

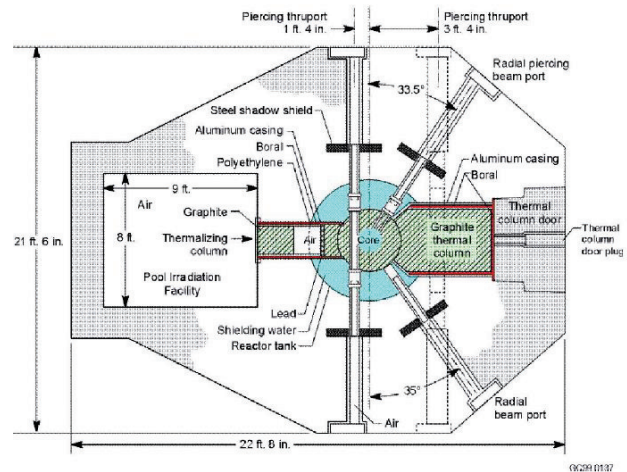


Figure 2: LENA reactor, transversal section.

Inside the core there are ninety sample housings, cylindrically located around the *central channel* (with a diameter of 3.38 cm). The central channel is the most intense irradiation environment and provides a flux of $1.7 \cdot 10^{13}$ n/(s cm²). The other channel used for our study, is the *Rabbit* one, whose flux is of $7.5 \cdot 10^{12}$ n/(s cm²) in position F24. Figure 3 illustrates the LENA core.

The core is surrounded by a graphite reflector whose goal is to minimize the neutron radial escape. On the top of this reflector there is a granular cavity inside which there is a rotating sample holder, made of forty tubular containers, also available for the irradiations. The *Lazy Susan* channel (the second used in our study) provides a neutron flux of $2.4 \cdot 10^{12}$ n/(s cm²).

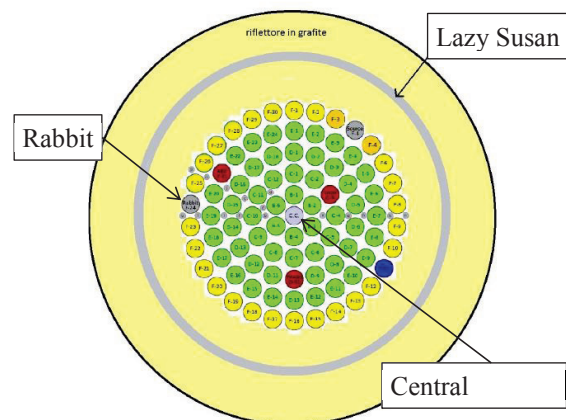


Figure 3: Layout of the LENA core and irradiation channels.

The Nb samples were inserted in the irradiation channels and exposed to the neutron fluxes mentioned above, continuously produced by the fission reactions. Fission produces promptly fast neutrons which, after undergoing thermalization, reach a high cross section for capture. In the NAA only the thermal neutrons are considered, because they are the ones involved in the neutron capture process.

SAMPLES PREPARATION

For our purpose eight Niobium samples were prepared to be inserted in the irradiation reactor channels, along with several reference standards in order to perform a quantitative elemental concentration in the samples.

The eight samples needed to be small enough to fit the channel limited dimension (few centimeters), their masses ranged from about 60 to 80 mg. The reference standard used were chosen depending on their elemental composition. In detail we are interested in material inclusions potentially taking place during cavity fabrication, as Al, Fe, Cu, Mn, Mg, W, so standards containing these elements were used. The samples and the standards were inserted in polyethylene vials (with an external diameter of 6 mm and height of 32 mm). In order to avoid to use vials with unwanted and misleading impurities they were cleaned with nitric acid and washed with high purity deionized water. The eight samples were both used for the *Rabbit* and *Lazy Susan* channels irradiations. The vials were sealed up to avoid losing the samples during the irradiation.

RABBIT CHANNEL MEASUREMENTS

The irradiation in the *Rabbit* channel took place on April the 3rd 2013 for both the samples and the SMRs. The irradiation and measurement time were chosen to be 60 s and 300 s, while the delay time from irradiation and measurement varied from few minutes to approximately ten minutes, depending on the activity of the irradiated samples. A dedicated attention was given to the geometrical configuration during the measurements, because it needs to be the same for all the samples, to keep the same irradiation conditions.

In the *Rabbit* irradiation channel each sample is individually irradiated at different times so that the samples are not exposed to exactly the same neutron flux. To account for this detail several SMR were exposed in the channel in order to quantify the neutron flux stability. After the irradiation the measurements of the samples were performed directly at the laboratory in Pavia and the spectra are now under study in order to identify the interesting elements in the samples, by relative comparison with the SMR spectra. Figures 4 and 5 show the overlap of Niobium samples spectra and SMR spectra to assess the measurement and irradiation repeatability and the relative comparison.

In order to evaluate the counts per unity of time and mass, the delay time between the extraction and the measurements and the measuring life time, considering

that some isotopes decay with a time constant lower than the measuring time, were properly taken into account.

Standard Reference Materials (SRM) irradiated were: 1632a, 1633, 1645, 71A, 71B, 71C, CCS6.

The most interesting SRM used in the channel is 71A which contains the following elements in known concentrations:

Ag, Al, As, B, Ba, Be, Ca, Cd, Ce, Co, Cr3, Cs, Cu, Dy, Er, Eu, Fe, Ga, Gd, Ho, K, La, Lu, Mg, Mn, Na, Nd, Ni, P, Pb, Pr, Rb, S, Se, Sm, Sr, Th, Tl, Tm, U, V, Yb, Zn.

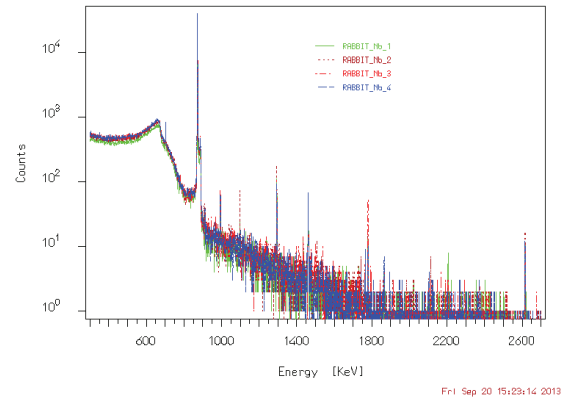


Figure 4: Overlap of several Nb samples spectra (acquired with a HPGe detector) after the *Rabbit* channel irradiation. It is possible to notice the reproducibility of the different spectra.

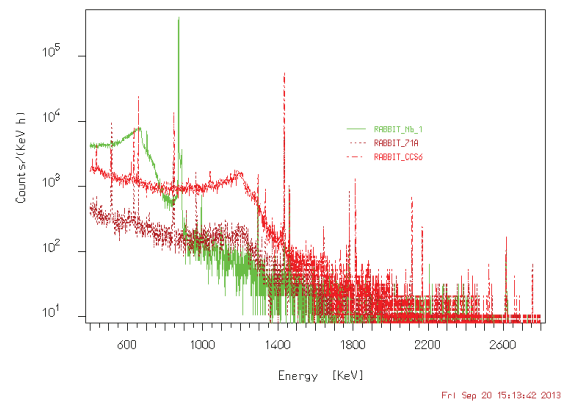


Figure 5: Overlap of Nb sample spectrum (green, acquired with a HPGe detector) and two different SMR spectra (dark red SMR 71A and light red CCS6).

LAZY SUSAN CHANNEL

The 2nd irradiation took place on April 10th 2013 and was dedicated to analyze and study isotopes with longer half-life than the ones studied using the *Rabbit* channel. Using this channel it is possible to irradiate all the samples at the same time (as they are held in a rotating sample holder) and thus there is no need to consider potential neutron flux fluctuations between samples. Even in this step it is important to reproduce the same experimental and measuring conditions for the SMR and samples in order to keep the same irradiation parameters

as for the Nb samples, in order to properly calculate the elemental concentrations.

These data are currently under analysis in order to estimate the Nb sample composition or the statistical limits of the elements in the Nb samples. Figure 6 and 7 show the overlap between Nb and SMR spectra.

Standard Reference Materials (SRM) irradiated were: 1632a, 1633, 71B, 71C, CCS6

The most interesting SRM used in the channel is 71B which contains the following elements in known concentrations:

Ge, Hf, Mo, Nb, Sb, Si, Sn, Ta, Te, Ti, W, Zr

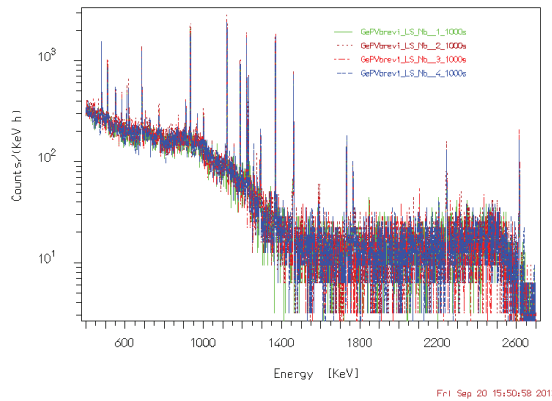


Figure 6: Overlap of several Nb samples spectra (acquired with a HPGe detector) after the *Lazy Susan* channel irradiation. It is possible to notice the reproducibility of the different spectra.

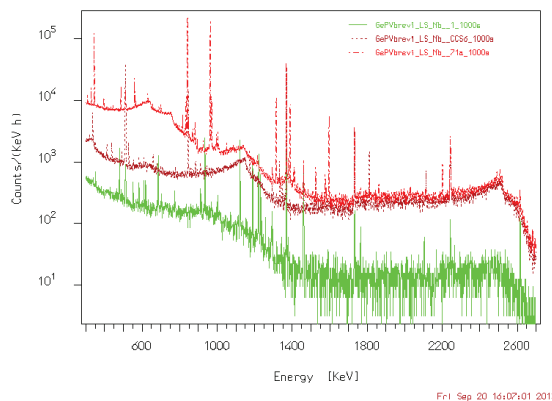


Figure 7: Overlap of Nb sample spectrum (green, acquired with a HPGe detector) and two different SMR spectra (dark red CCS6 and light red 71A).

Long Measurement Times: 1,000 s and 10,000 s

The 1,000 s measurements were performed at the L.E.N.A. laboratory in Pavia after one week, once the irradiation finished and they allow the investigation of elements with half-life up to some months. Acquisitions of 10,000 s were performed in the radioactivity laboratory at the University of Milano Bicocca (Physics Department B. Occhialini) and these allow analysing elements with half-lives up to a few years. Figure 8 and 9 show the overlap between Nb and SMR spectra.

For the different irradiations several HPGe detectors were used:

- A HPGe detector in Pavia after the *Rabbit* irradiation and for *Lazy Susan* 1,000 s long measurements.
- A 4π coverage detector of increased resolution for the measurements at 10,000 s performed in the Radioactivity Laboratory at the University of Milano Bicocca.

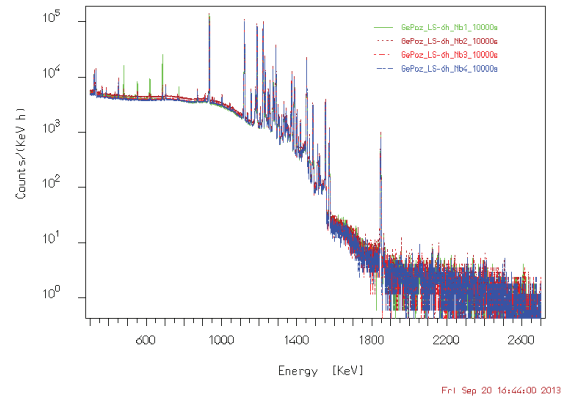


Figure 8: Overlap of several samples Nb spectra (acquired with a HPGe detector) after the *Lazy Susan* channel irradiation. It is possible to notice the reproducibility of the different spectra (including the 10,000s measurements).

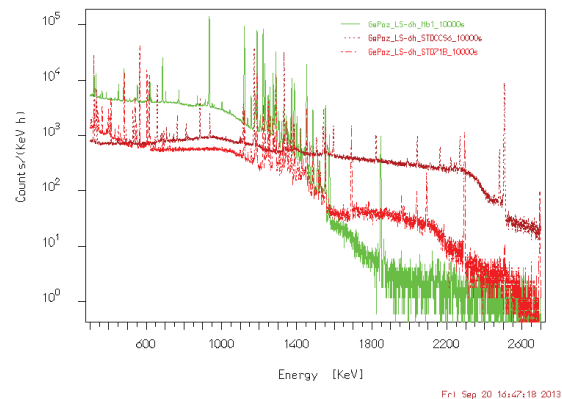


Figure 9: Overlap of Nb sample spectrum (green, acquired with a HPGe detector) and two different SMR spectra (dark red SMR CCS6 and light red 71B).

CONCLUSIONS

A number of Nb samples were irradiated in the LENA research reactor, along with SMR standard in order to evaluate elemental composition. The acquired spectra are now under study for the quantitative evaluation.

REFERENCES

- [1] G.F. Knoll, "Radiation Detection and Measurements", Wiley, 4th edition, 2010.
- [2] R.H. Philby et al., "Gamma Ray Energy Tables for Neutron Activation Analysis", Washington State University, 1970.