IDENTIFICATION OF A LEAKING TRIGA FUEL ELEMENT AT THE NUCLEAR REACTOR FACILITY OF THE UNIVERSITY OF PAVIA.

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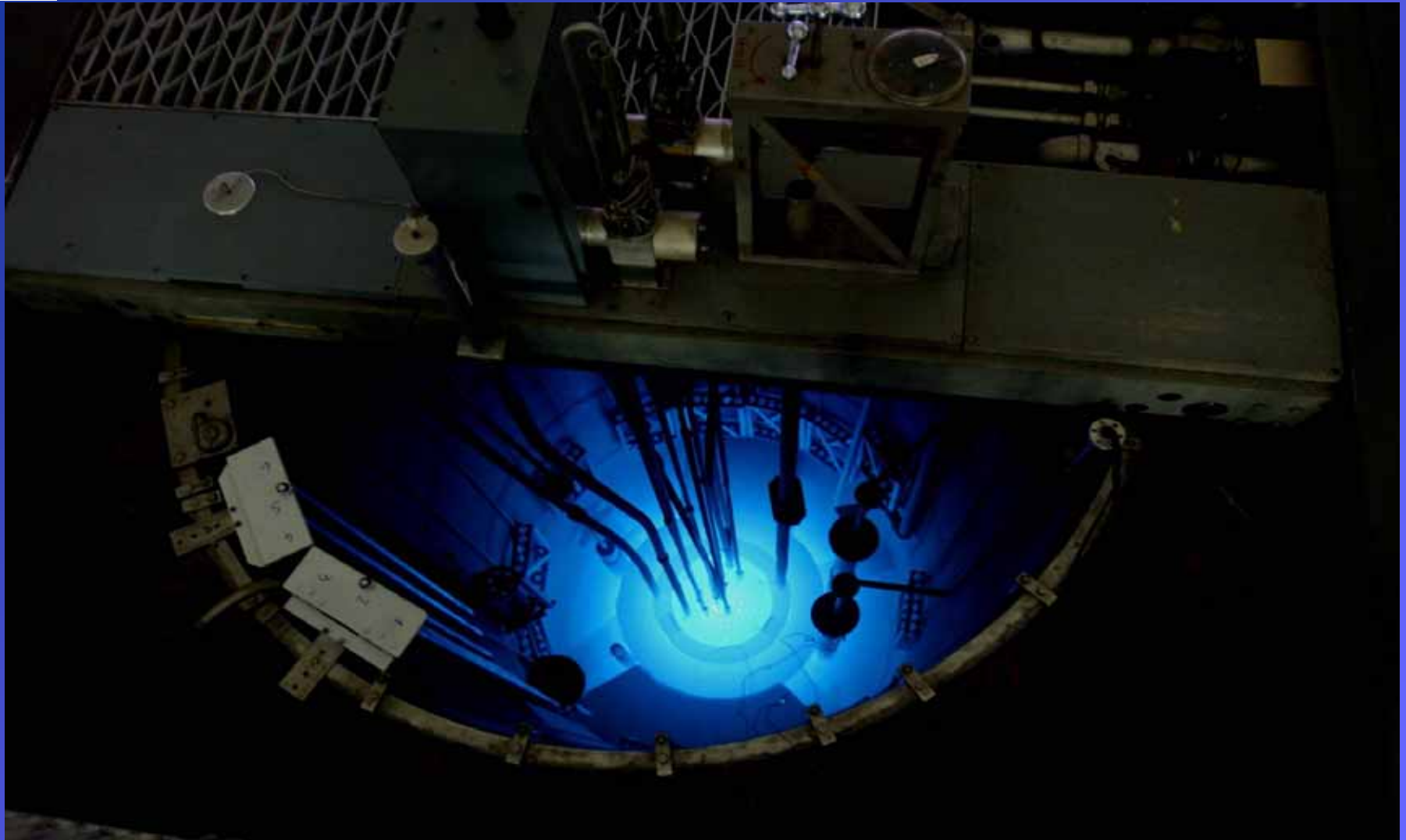*University of Pavia - Italy*

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THE 250 kW TRIGA REACTOR

THE TRIGA REACTOR POOL & CORE

INTRODUCTION

During a periodical activity of characterization of the ionic-exchange resins of the demineralizer of the primary cooling circuit of the TRIGA Mark II reactor of the University of Pavia a small but detectable amount of ^{137}Cs contamination was measured.

Since the reactor has been running for several hundreds of hours at full power without showing any anomaly in the radiometric and thermo-hydraulic parameters, the reactor was brought at the nominal power of 250 kW for one hour and a sample of water was collected from the reactor tank and analyzed in a low-background gamma-ray detector.

As a result a small amount of fission products were detected in the reactor pool water (few Bq/g) suggesting the existence of a possible clad defect in one ore more fuel elements.

Since no halogens such as iodine and bromine were detected in the sampled water, the more probable hypothesis, also supported by literature, seemed to be a micro-fissure in the neck of an instrumented fuel element.

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As a consequence of the unusual fission products activity detected in the water of the reactor pool a campaign of gamma-ray spectrometry was implemented in order to evaluate the importance of the release.

Using a HGe (1.72 keV FWHM – 31.3% efficiency – 58.5 Photo Peak/Compton) many analysis were performed and some of them, the most significant ones, are presented below.

Notice that all the results of the measurements presented have a relative error less than 10% and that the MDAs were evaluated according to the common formula:

$$MDA = 4{,}65 * \frac{\sqrt{Background}}{LiveTime}$$

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PRELIMINARY MEASUREMENTS

Measurement of a sample of the ionic-exchange resins of the demineralizer of the primary cooling circuit

- Counting Time = 850 s;
- Activity reported to last day of reactor operation at nominal power

Radioisotope	$T_{1/2}$	Bq/g
Co-60	5.26 y	129,8
Zn-65	244 d	17,4
Mn-54	312 d	123,8
Co-58	70.8 d	86,4
Cs-134	2 y	2,86
Cs-137	30.2 y	245
Cr-51	27,7 d	265

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PRELIMINARY MEASUREMENTS

Measurement of a sample of a new charge of ionic-exchange resins
No ^{137}Cs or other fission product were detected in the sample.

Smear-Test of the external surface of a fresh fuel element.
The measure was performed in order to exclude the hypothesis that the clad of the new SST fuel elements was contaminated by a small amount of fission products.
The measure didn't show any presence of such contamination (MDA = 2,75 10^{-04} Bq/cm^2 at 662 keV).

PRELIMINARY MEASUREMENTS

Measurement of a sample of the water filter of the primary cooling circuit.

- Counting Time = 50.000 s
- Activity reported to last day of reactor operation at nominal power

Radio-isotope	$T_{1/2}$	Bq/g
Eu-152	13,6 y	10,5
Co-60	5,26 y	36,6
Fe-59	44 d	11,6
Eu-154	8,8 y	1,5
Zn-65	244 d	4,9
Sc-46	83,8 d	3,5
Mn-54	312 d	2,5
Co-58	70,8 d	1,04
Cs-134	2,0 y	0,38
Cs-137	30,2 y	1,19
Cr-51	27,7 d	204

Measurement of a sample of water collected from the reactor pool before to operate the reactor test at nominal power.

- Counting Time = 25.700 s
- Activity reported to last day of reactor operation at nominal power

Radio-isotope	$T_{1/2}$	Bq/g
Co-60	5,26 y	5,60E-04
Zn-65	244 d	4,10E-04
Mn-54	312 d	2,80E-04
Co-58	70,8 d	3,50E-04
Cs-134	2,0 y	< 9,33E-05
Cs-137	30,2 y	1,50E-03

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PRELIMINARY MEASUREMENTS

Measurement of a sample of water collected from the reactor pool after one hour of operation at 250 kW nominal power.

The sample was collected 30 cm below the pool water surface with the primary cooling system of the reactor off. The analysis was performed after 90 min from the collection of the sample.

- **Counting Time = 1032 s**
- **Activity reported at reactor shut-down**

Radio-isotope	$T_{1/2}$	Bq/g
Cs-138	32,2 m	7,08
Xe-138	14,13 m	5,51
Ar-41	109 m	14,82
Na-24	15 h	3,58
Mn-56	2,58 h	0,53
Kr-88	2,84 h	0,70
Rb-88	17,8 m	21,67
Kr-85m	4,48 h	0,14
Kr-87	76,3 m	1,04
Cl-38	37,21 m	0,57
Xe-135	9,11 h	0,04

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PRELIMINARY MEASUREMENTS

The same sample was analysed after 96 hours and 41 min in order to look for long life radio-isotopes.
- Counting Time = 50.000 s
- Activity reported at reactor shut down

Radio-isotope	$T_{1/2}$	Bq/g
La-140	40,22 h	4,46E-03
Na-24	15 h	3,68
Co-60	5,26 y	5,63E-04
Mn-54	312 d	6,66E-04
Co-58	70,58 d	7,69E-04
Cs-134	2,0 y	< 1,74E-04
Cs-137	30,2 y	1,84E-03
Cr-51	27,7 d	2,13E-03

Measurement of aerosol sample collected above the water pool surface.

The measurement was performed sampling 3 m^3 of air on a active-carbon and absolute filter (porosity 4,5 μm). The spectrometry of the filter was performed 60 min after the sampling (Counting Time = 408 s) and besides natural radio-isotopes only 22 Bq/m^3 of ^{138}Cs was detected.

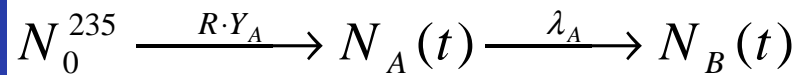**Measurement of aerosol sample collected in the off-gas channel of the reactor.**

The measurement was performed sampling 50 m^3 of air on a active-carbon and absolute filter (porosity 4,5 μm) and only natural radio-isotopes were detected

- MDA for ^{138}Cs = 7,5 10^{-02} Bq/m^3
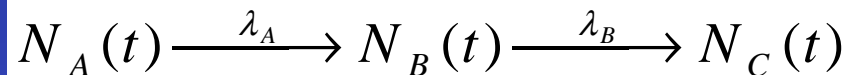- MDA for ^{137}Cs = 5,8 10^{-04} Bq/m^3

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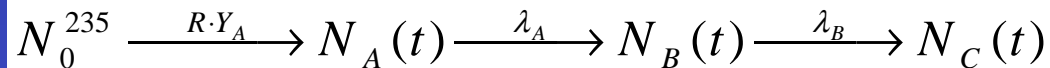
PRELIMINARY EVALUATION OF THE RELEASE

In order to evaluate the typology of the release of the fission products the relative abundance of the noble gas was calculated and compared with the specific activities measured in the sampled water.

$$N_0^{235} \xrightarrow{R \cdot Y_A} N_A(t) \xrightarrow{\lambda_A} N_B(t)$$

$$\lambda_A \cdot N_A(t) = R \cdot Y_A \cdot (1 - e^{-\lambda_A \cdot t})$$

$$N_A(t) \xrightarrow{\lambda_A} N_B(t) \xrightarrow{\lambda_B} N_C(t)$$

$$\lambda_B \cdot N_B(t) = \frac{\lambda_A \cdot \lambda_B}{\lambda_B - \lambda_A} N_{A0} \cdot (e^{-\lambda_A \cdot t} - e^{-\lambda_B \cdot t})$$

$$N_0^{235} \xrightarrow{R \cdot Y_A} N_A(t) \xrightarrow{\lambda_A} N_B(t) \xrightarrow{\lambda_B} N_C(t)$$

λ = Radioisotope Decay Constant
R = Fission Reaction Rate = ($\approx$ 7.75 10^{15} fission·s^{-1} at 250 kW)
ϕ = Average Reactor Thermal Flux ($\approx$ 2 10^{12} n·cm^{-2}·s^{-1} at 250 kW)
σf = Fission Microscopic Cross Section for ^{235}U (583 barn)
N = Number of atoms
Y = Fission Yield

$$\lambda_B \cdot N_B(t) = R \cdot Y_A \cdot \left(\frac{\lambda_B \cdot \lambda_A \cdot e^{-\sigma_f \cdot \phi \cdot t}}{(\lambda_A - \sigma_f \phi) \cdot (\lambda_B - \sigma_f \phi)} + \frac{\lambda_B \cdot \lambda_A \cdot e^{-\lambda_A \cdot t}}{(\sigma_f \phi - \lambda_A) \cdot (\lambda_B - \lambda_A)} + \frac{\lambda_B \cdot \lambda_A \cdot e^{-\lambda_B \cdot t}}{(\lambda_A - \lambda_B) \cdot (\sigma_f \phi - \lambda_B)} \right)$$

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PRELIMINARY EVALUATION OF THE RELEASE

The results of the calculation as a function of the Fission Reaction Rate (R) are reported below:

^{138}Xe   [$\lambda_{Xe}N_{Xe}$(1h) = 5.97 10^{-2} R]
direct production (Y= 4.81 %) $\lambda_{Xe}N_{Xe}$(1h) = 4.56 10^{-2} R
production from ^{138}I (Y = 1.49 %) $\lambda_{Xe}N_{Xe}$(1h) = 1.41 10^{-2} R

^{135}Xe   [$\lambda_{Xe}N_{Xe}$(1h) = 3.79 10^{-4} R]
direct production (Y= 0.0785 %) $\lambda_{Xe}N_{Xe}$(1h) = 5.70 10^{-5} R
production from ^{135}I (Y = 6.28 %) $\lambda_{Xe}N_{Xe}$(1h) = 2.36 10^{-4} R
production from ^{135m}Xe (Y = 0.178 %) $\lambda_{Xe}N_{Xe}$(1h) = 8.57 10^{-5} R

^{88}Kr   [$\lambda_{Kr}N_{Kr}$(1h) = 7.69 10^{-3} R]
direct production (Y= 3.55 %)

^{87}Kr   [$\lambda_{Kr}N_{Kr}$(1h) = 1.075 10^{-2} R]
direct production (Y= 2.56 %)

^{85m}Kr   [$\lambda_{Kr}N_{Kr}$(1h) = 1.85 10^{-3} R]
direct production (Y= 1.29 %)

PRELIMINARY EVALUATION OF THE RELEASE

The ratios between the calculated activities and the measured specific activities in the reactor pool water for noble gas are the following:

Radio-isotopes	Calculated Ratio	Measured Ratio
$^{138}Xe/^{135}Xe$	157	138
$^{87}Kr/^{88}Kr$	1.40	1.48
$^{138}Xe/^{87}Kr$	5.55	5.30

From this evaluation it seemed clear that the noble gas were release promptly in coincidence with the reactor operation at the power of 250 kW.

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PRELIMINARY EVALUATION OF THE RELEASE

For what concerns the specific activity in air of ^{138}Cs (22 Bq/m^3) measured above the pool water surface, it was consistent with the specific concentration in water of the radionuclide, presuming an evaporation coefficient of ~10^{-3}, realistic for the reactor pool water temperature of 40 °C.

On the contrary, the increase of the specific activity in water of ^{137}Cs after the operation of the reactor for 1 hour at the power of 250 kW, it was not consistent with the hypothesis of a prompt release unless about 30% of the fuel elements of the core were fissured.

A possible explanation of this anomalous increase was that ^{137}Cs could be dissolved into the moisture that could be accumulated inside the damage fuel element when the reactor was not in operation and which could be released all at once when the fuel element was heated up .

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A sampling and detection apparatus was realised with the following components:

• an aluminium anticorodal tube (length 6 m $\varnothing$ 25,4 mm) with a funnel terminal ($\varnothing$ 55 mm)

•a hydraulic pomp OMAN mod. ALM25 in SST (max flow 43 lt/min – Prevalence 12 m)

•a rubber tube for water circulation ($\varnothing$ 25.4 mm)

•a HPGe Ortec GMX (n-type, Coaxial Detector, Be Window) - FWHM 2.57 keV at 1.33 MeV 60Co – Efficiency 30% - Photopeak/Compton = 46/1

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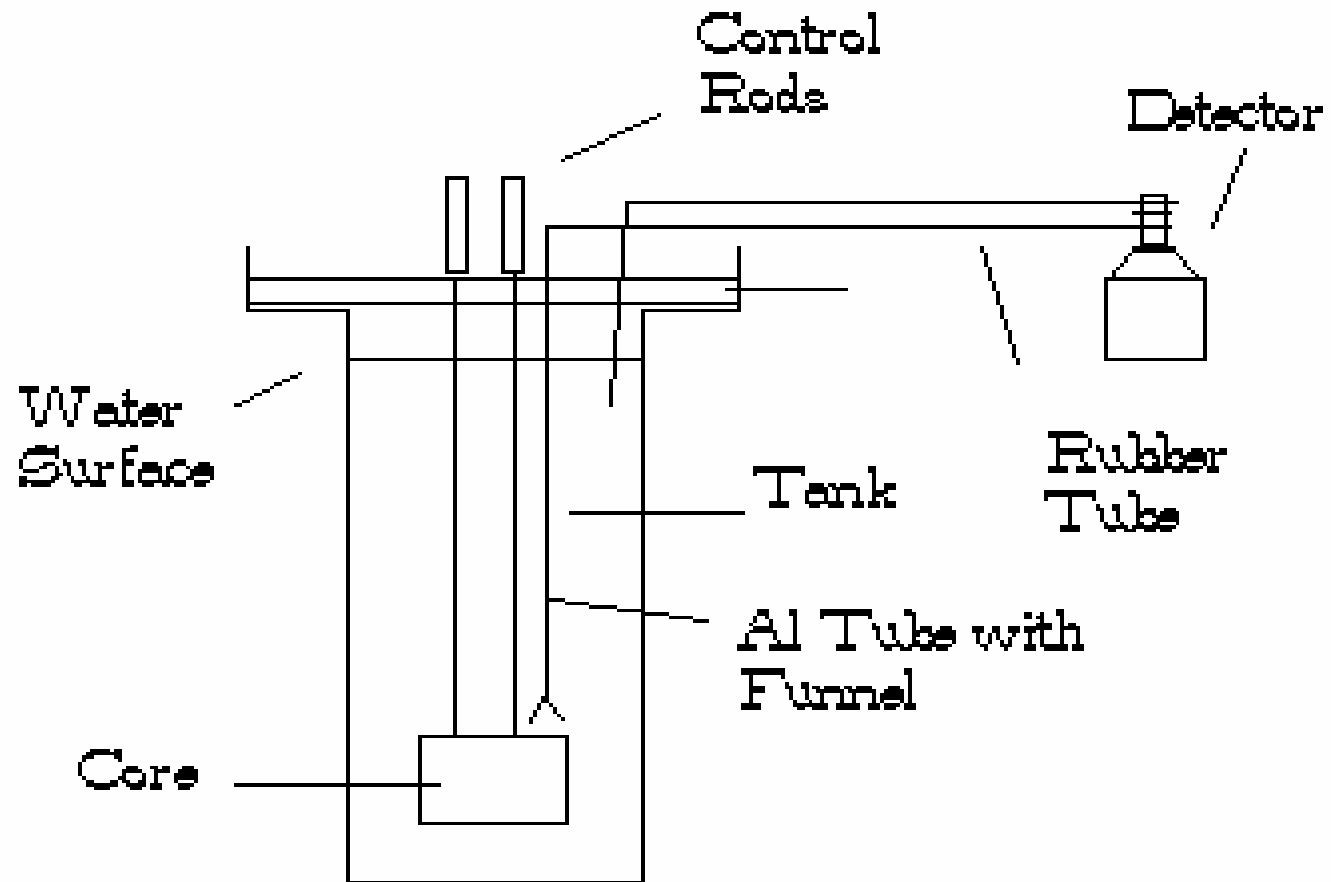
SAMPLING APPARATUS SET-UP

Hydraulic pomp OMAN mod. ALM25 in SST (max flow 43 lt/min – Prevalence 12 m)

Aluminium anticorodal tube (length 6 m $\varnothing$ 25,4 mm)

SAMPLING APPARATUS SET-UP

Funnel terminal ($\varnothing$ 55 mm) of the aluminium anticorodal tube

SAMPLING APPARATUS SET-UP

**HPGe Ortec GMX (n-type, Coaxial Detector, Be Window)
FWHM 2.57 keV at 1.33 MeV 60Co – Efficiency 30% - Photo-
peak/Compton = 46/1**

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Water was collected from the superior grid of the reactor core by means of the aluminium tube in different sectors and in different fuel element positions and was counted on-line using the HPGe detector positioned in the Radiochemistry Laboratory (about 20 m distance from the reactor top).

In order to allowed the decay of short half-life radioisotopes such as ^{19}O, ^{16}N, the pump suction flow was reduced to 5 lt/min, that means that the sampled water took at least 2 minutes to reach the detector.

The radioisotope considered for the measurements was ^{138}Xe that presents three well defined and clean gamma-peak at 258 keV, 434 keV, 1768 keV.

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Reactor Pool Water Sampling
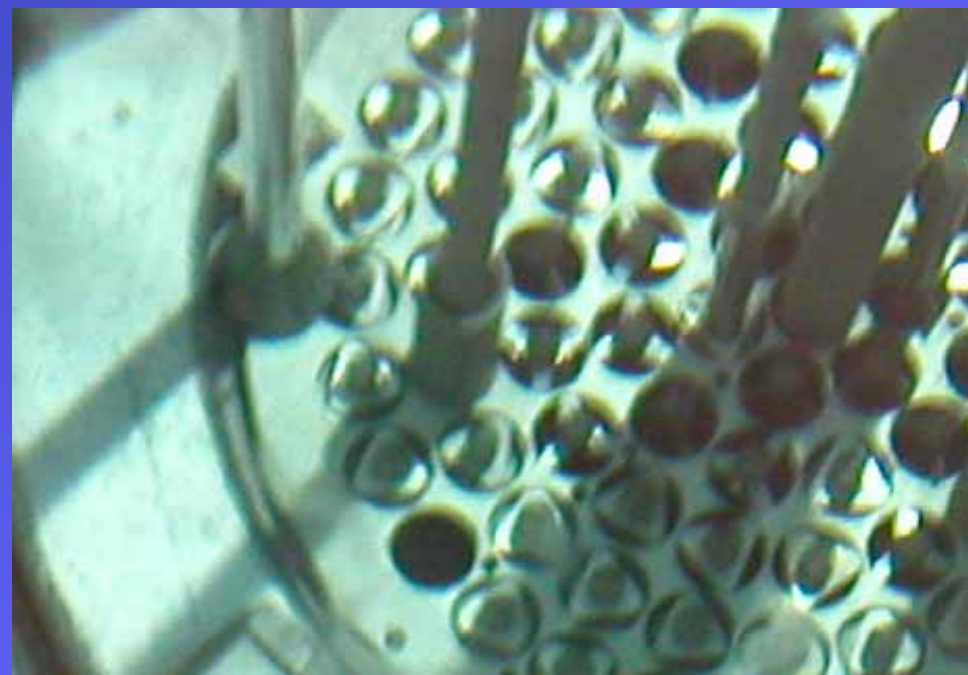
The reactor core was virtually divided into 4 sector:
N°1 (Sud-Est)
N°2 (Sud-West)
N°3 (Nord-West)
N°4 (Nord-Est)

After testing the sampling apparatus before to start the reactor, the reactor was operated at the power of 1 kW and the water was sampled in all four sector at a distance of about 15 cm from the superior grid. In this condition no fission products ware detected in the water.

Thus the reactor power was raised up to 50 kW (i.e. a fuel temperature about 45 °C) and the same investigation in all four sectors was repeated, but no fission product released was detected either.

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The reactor power was then raised up to 100 kW (i.e. a fuel temperature about 90°C) and finally fission product were detected starting from sector N°3, giving the following results:

Isotope	E_n Peak (keV)	Peak Area(cps)
Xe-138	258	9993
	434	3543
	1768	898

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IDENTIFICATION OF THE LEAKING FUEL ELEMENT

Unfortunately the specific activity measured in each sector ended up to be of the same magnitude suggesting two possible explanations:

1) there were more than one fuel element fissured in the core positioned in different sectors

2) the water of the pool mixed up very fast in proximity of the superior grid preventing the possibility of identifying the sector of origin of the release.

Anyway it was clear that a more systematic analysis, fuel element per fuel element, should have been performed.

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IDENTIFICATION OF THE LEAKING FUEL ELEMENT

Thus the aluminium tube was lowered down towards the grid in such a way that the funnel covered just one fuel element position.

The water sampling was repeated until when two SST clad fuel element, close one to the other in sector N°3 (position C5 and C6 of the core), seemed to be the possible origin of the release.

These two elements, though, were close to three instrumented fuel elements and, knowing from literature that these kind of elements are more likely to undergo fissure, the hypothesis of their involvement in the release was still the more probable.

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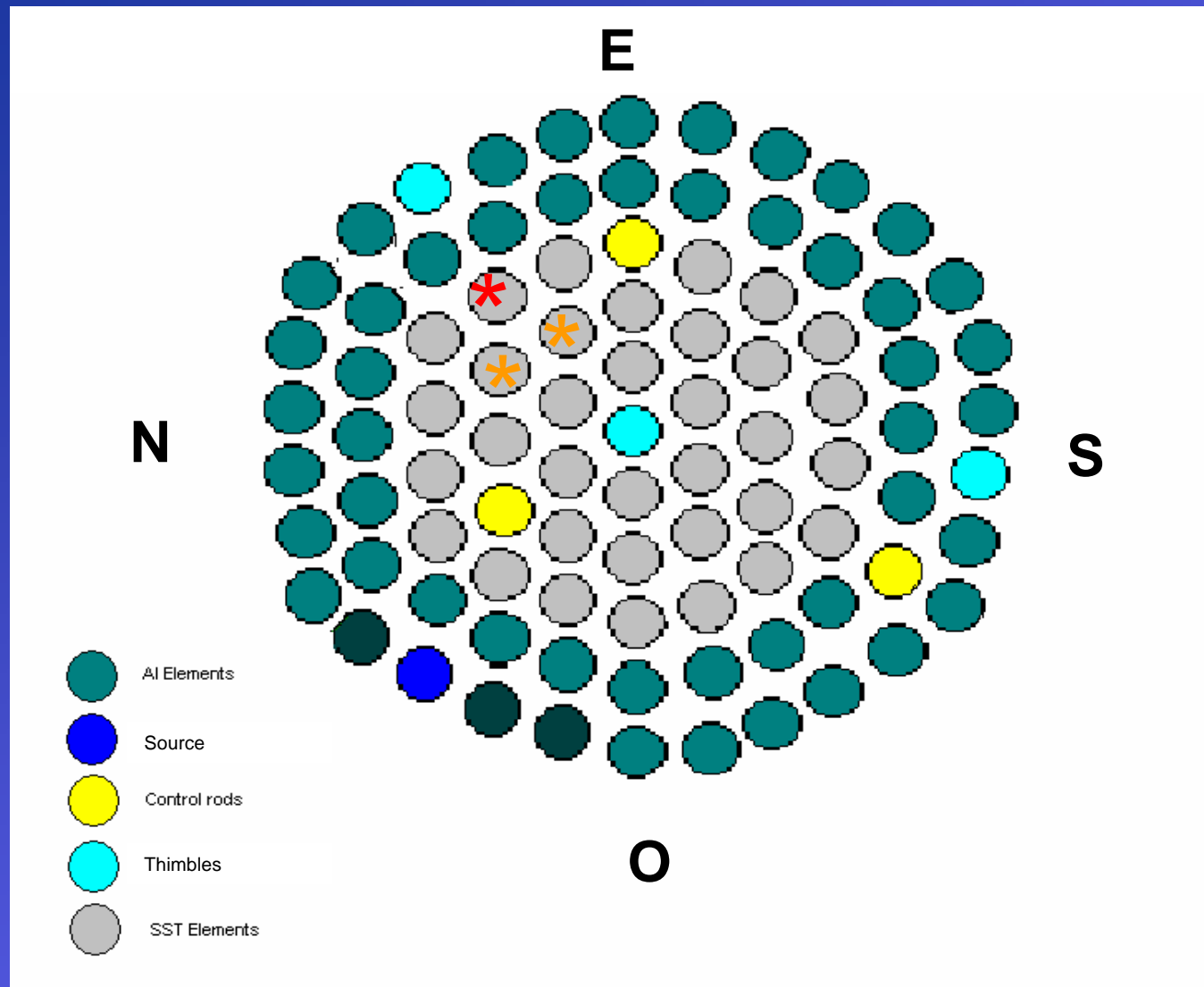# IDENTIFICATION OF THE LEAKING FUEL ELEMENT

In order to verify this last hypothesis, the two SST clad fuel elements were moved to another position of the core where they were measured again.

As expected no fission product were detected. On the contrary, fission products were detected again in position C5 and C6 where two different fuel elements, previously verified for the absence of leakage, were inserted.

At that point it was clear that the release was caused not by the SST fuel elements in position C5 and C6 but by one or more fuel elements positioned nearby.

Since three instrumented fuel elements were positioned close to positions C5 and C6 (in position D7, D8 and B3), the oldest of the three (in position D8) was removed from the core.

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IDENTIFICATION OF THE LEAKING FUEL ELEMENT

The measurements in position C5 and C6 were repeated and no fission products were detected.

The reactor power was raised up to 250 kW and the measurement of the water sampled in all four sector was repeated showing no presence of fission products.

A sample of water was collected from the reactor pool after one hour of operation of the reactor at the power of 250 kW with the primary cooling system off and it was measured in a low-background gamma detector. No fission products were revealed in the water.

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CONCLUSION

As expected the fission products leakage was due to a micro-fissure of a fuel element that released only noble gas only when the fuel element was heated up to a temperature around 90 °C, i.e. at the reactor power of about 100 kW.

The fuel element identified as the origin of the release was the oldest SST clad instrumented fuel element present in the core. It was removed and stored in a rack of the reactor pool. In this condition the element will not release any fission product any more but it will be necessary to condition it in a proper way after few years of cooling down.

The reactor was back in regular operation within two months since the leakage detection.

As a routine operation, the reactor pool water is now sampled and measured with a low-background gamma-ray detector every month before the reactor start-up and after 30 minutes of operation of the reactor at full nominal power.

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Before …

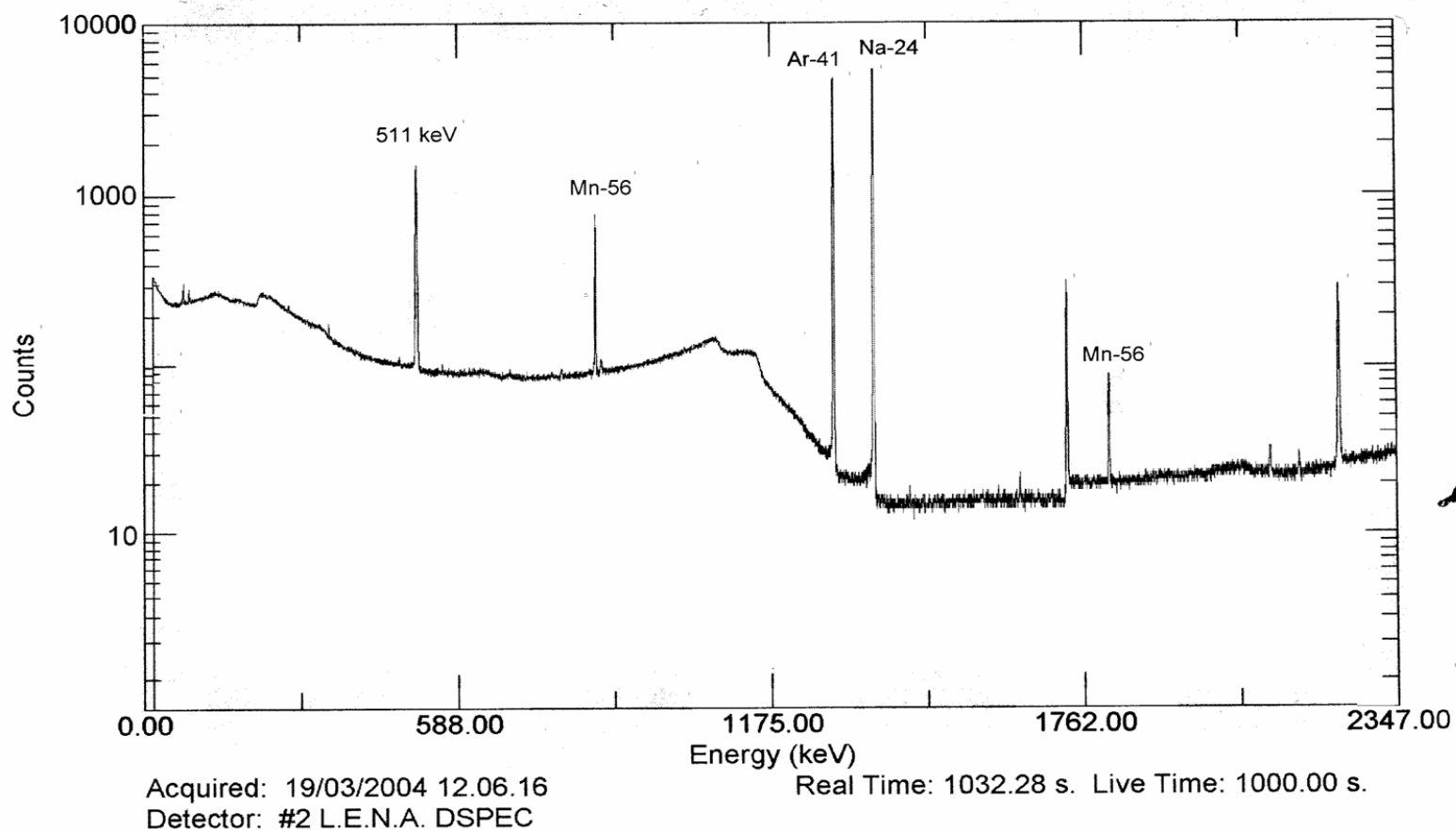Fig. 4 Reactor pool water after 1 hour of operation at 250 kW power
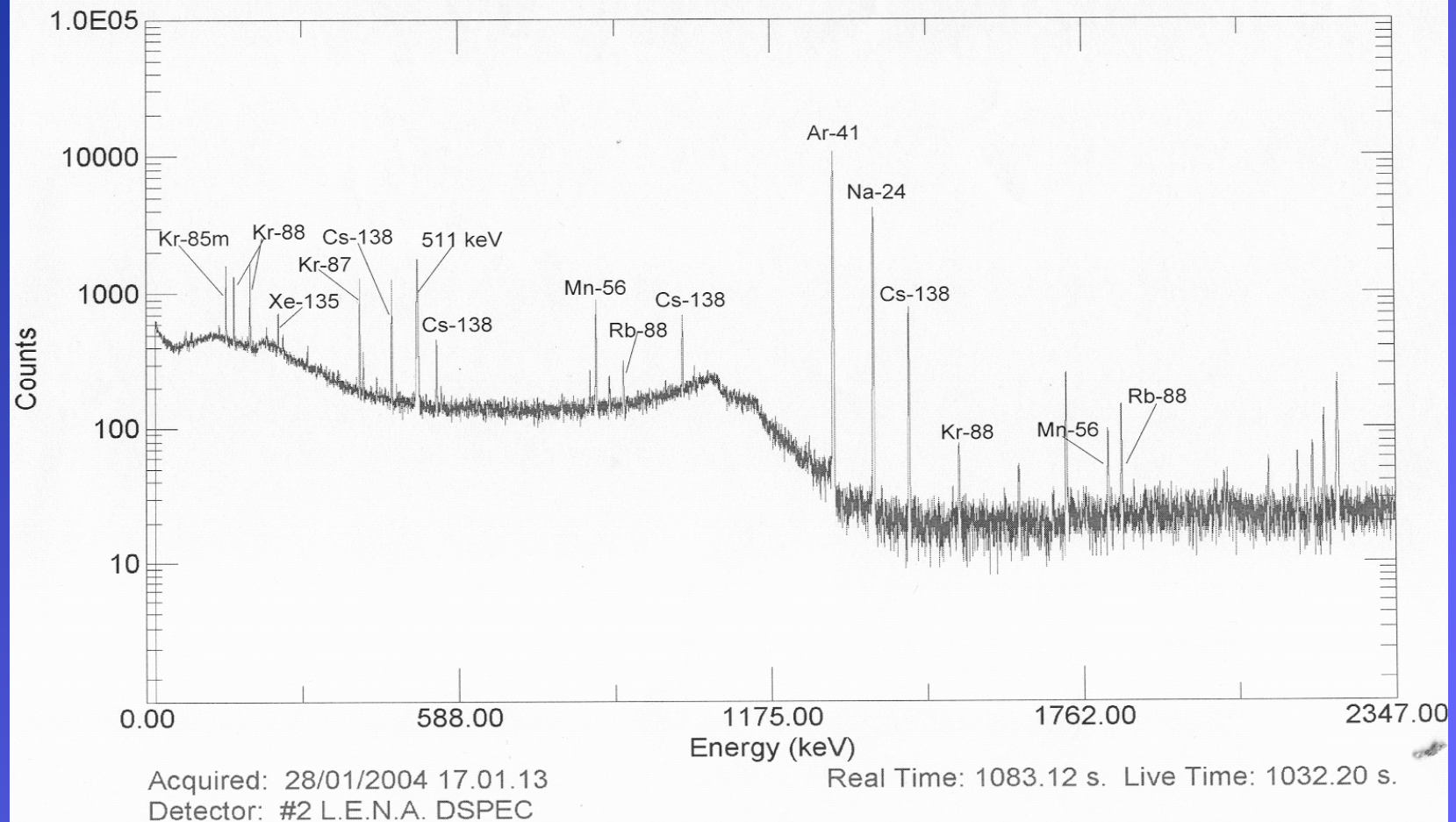
After …

Fig.5 Reactor pool water after 1 hour of operation at 250 kW power