IRRADIATIONS PERFORMED AT CEN- SACLAY CONDITIONS AND METHODOLOGY DEVELOPED

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ABSTRACT

In this paper the irradiation facilities used to irradiate salt samples at CEN–SACLAY are described. With these facilities salt samples could be irradiated at temperatures upto 400 °C and dose rates upto 10^5 Gy.h^{-1}.

1. INTRODUCTION

The Osiris nuclear experimental reactor is capable of supplying γ radiation, which spectrum is similar to that of waste, using its own fuel as it undergoes deactivation while simultaneously allowing to speed up the production of radiolytic gases by an increase in the dose rate level (by way of example, an total dose of 10^8 Gy corresponds to the first one hundred years of disposal).

The selection of an irradiation installation inside a reactor to conduct the program offers several advantages, such as :

a) the best cost–effectiveness : the gamma sources are already available and do not have to be fabricated or shipped ; and

b) the best availability : the installation was dedicated to the research program and available sources had a wide range of activity, facilitating the achievement of high dose rate levels.

The existing irradiation installation as well as the operating procedures were gradually adapted to the evolution of program needs, especially for the parameters of dose rate, total dose and temperature.

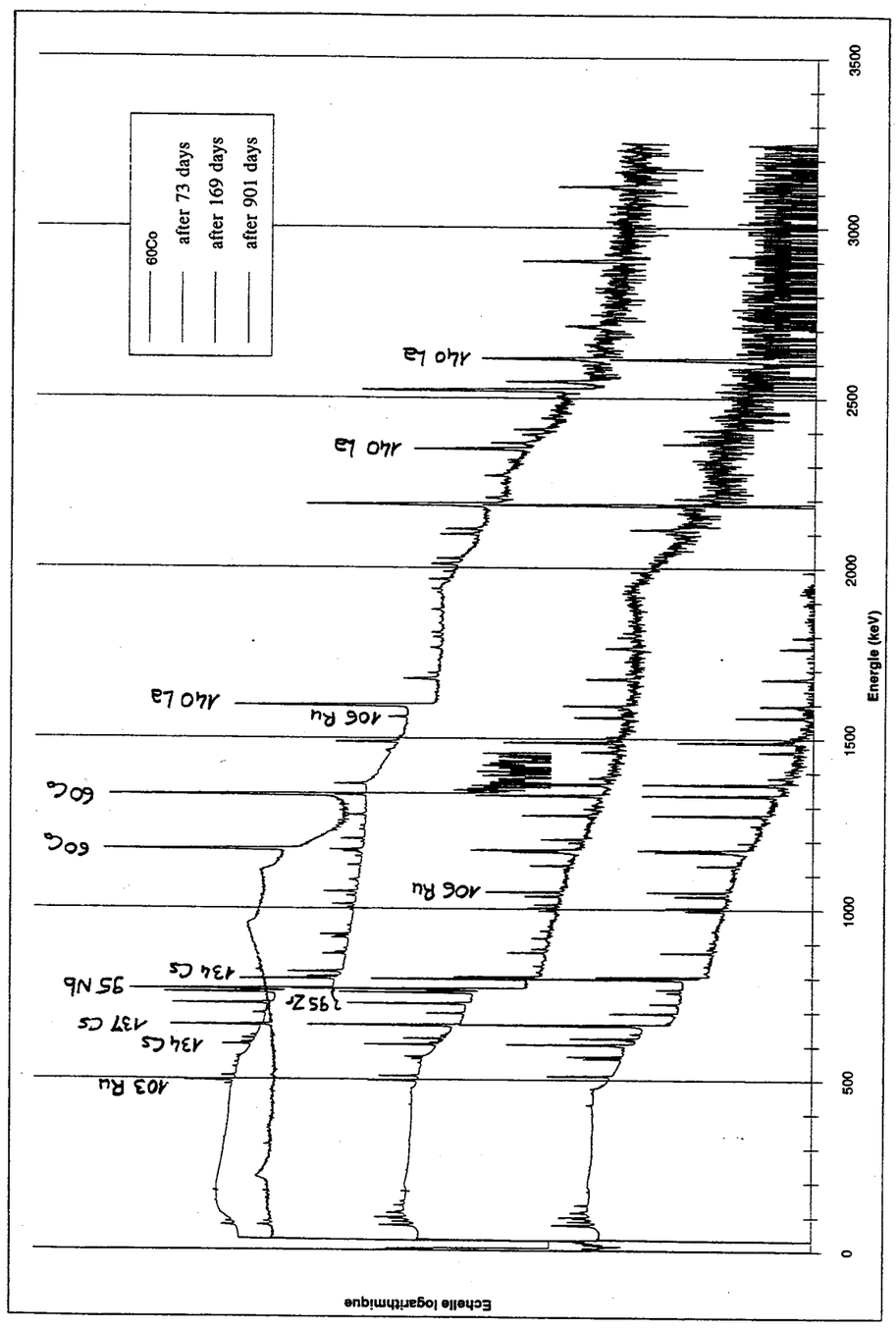

Figure 1: *Gamma spectrum emitted by Osiris fuel element and ^{60}Co*

2. THE USED RADIATION SOURCES

2.1. Gamma radiation

The γ radiation sources are primarily fuel elements unloaded from the Osiris reactor core and transferred underwater to the irradiator designed to accommodate them. The radiation spectrum is therefore determined by the fission products and daughter products present; its complexity diminishes as cooling increases (Fig. 1).

Low dose rates are achieved with elements that have been cooled for at least a few months (and that are no longer returned to the reactor core) and are positioned at a rather large distance from the samples (Fig. 4a).

High dose rates are achieved with elements unloaded from the core specifically for the purpose of γ irradiation. These elements are transferred into the irradiator after a reduced cooling time and are positioned close to the samples.

For a given activity, the ratio between the dose rate obtained when the elements are placed in the innermost positions and the dose rate obtained when the elements are placed in the outermost position, is 10

Few other γ radiation sources consisted of piles of small ^{60}Co plates with a very high specific activity prepared in Osiris by the activation of metallic cobalt. While the geometry of the baskets of ^{60}Co is not strictly identical to that of the fuel elements (Fig. 4b), the overall configuration remains the same. Only the energy spectrum of the ^{60}Co plates is different from that of the fuel elements.

Figure 1 illustrates the spectral differences of the two types of γ sources (the results were achieved with a γ spectrometry bench in the Osiris reactor pool).

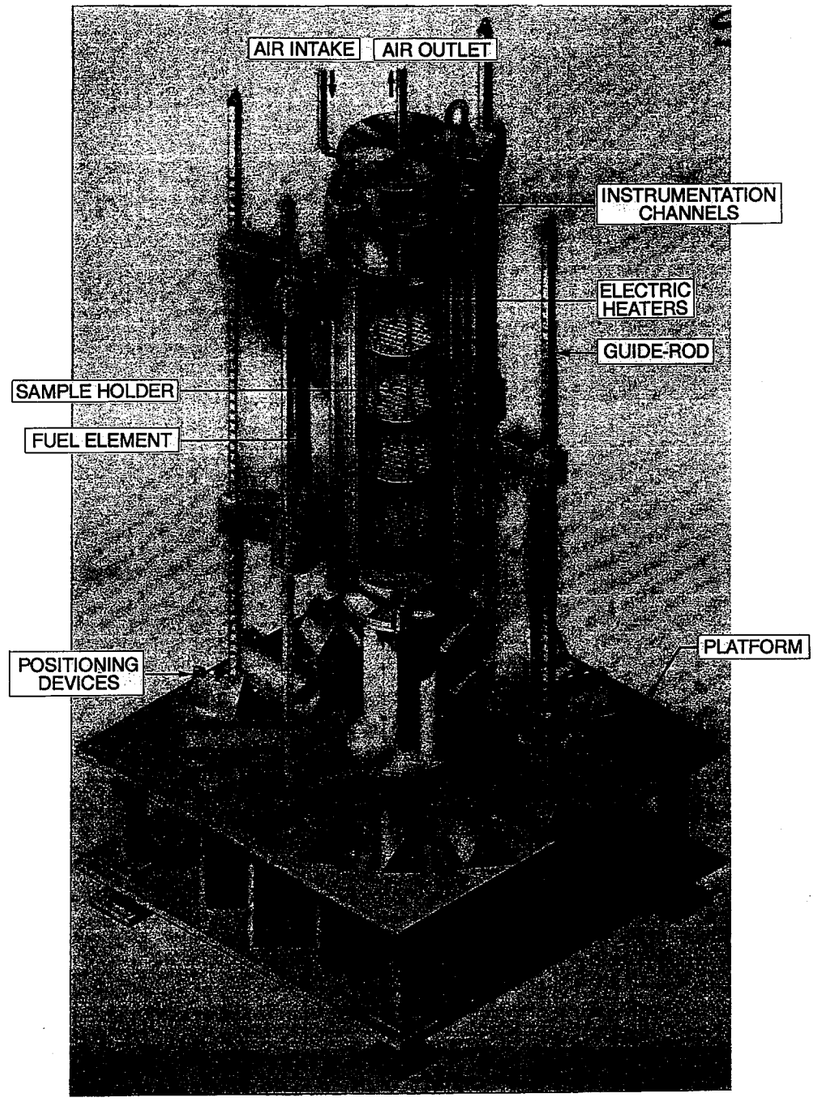

Figure 2: *View of the main irradiator*

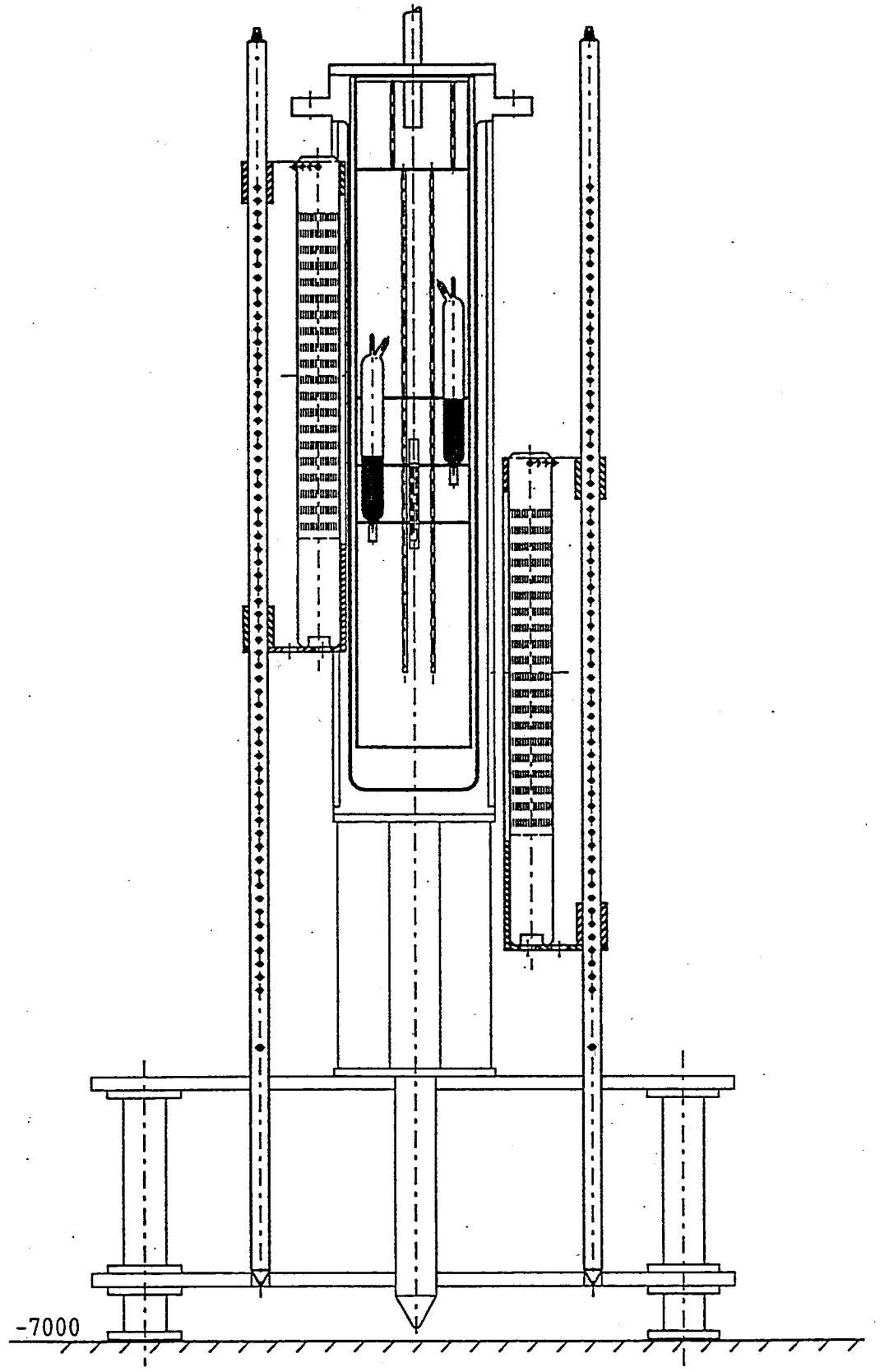

Figure 3: *Main irradiator (vertical cross section)*

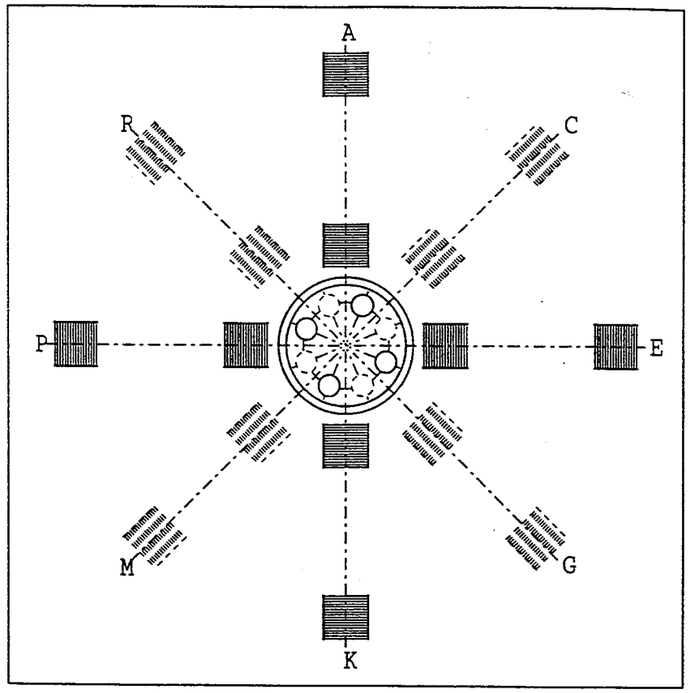

Figure 4a: *Horizontal cross section of gamma irradiator. Fuel elements are represented at the two extreme possible radial distances from the samples. Dashed lines = lower position, full = higher position.*

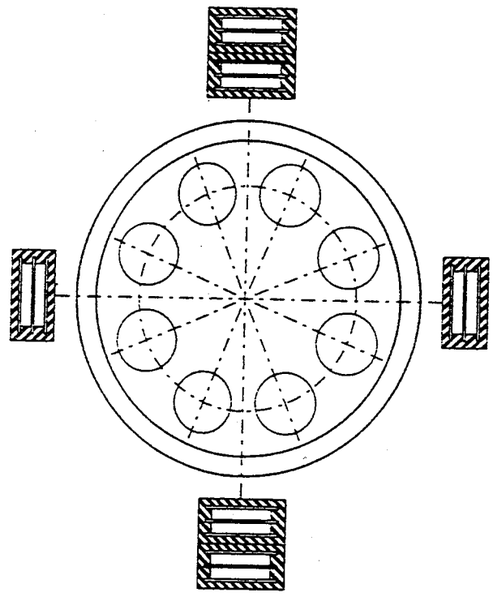

Figure 4 b: *Horizontal cross section of gamma irradiator with six ^{60}Co elements (sources are here located as close as possible to the samples)*

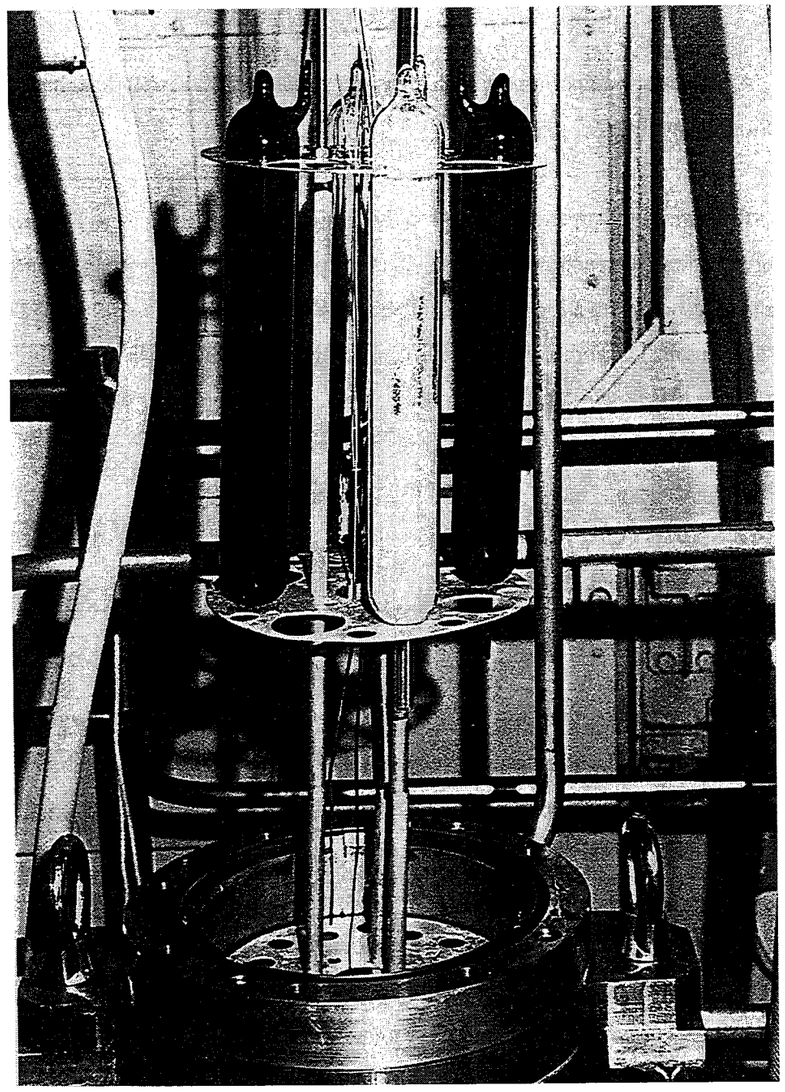

Figure 5: *Salt samples, sample holder and instrumentation*

2.2. X– Rays

A few tests were performed with an X–ray generator (300 keV) designed for radiographic inspection of mechanical parts.

The small size of the beam led to the design of small size vials. The anticathode being made of tungsten, the spectrum consists of X rays up to 300 keV with a peak around 72 keV. The dose rate, measured with a ionization chamber (PTW type), is approximately 600 $Gy.h^{-1}$ in air and 200 $Gy.h^{-1}$ in salt. The authorized duration of irradiations was limited to some hours, thus limiting the total dose to below 6.10^3 Gy.

3. IRRADIATORS USED

3.1. Principal irradiator

The irradiator is placed on a floor submerged beneath 7 meters of water in the n°3 canal of the Osiris reactor. It consists of a cylindrical irradiation chamber (useful diameter ~ 220 mm), weighted, submersible, surrounded by 8 supports for fuel elements (Fig. 2 and 3).

The irradiation chamber is sealed by a moveable cover which supports the sample carrier structure (Fig. 5). Electric furnaces enable a temperature of several hundred °C to be reached. To homogenize the temperature and evacuate any gases from radiolysis, the volume of the chamber is constantly swept with dry, dedusted, de–oiled air.

The chamber is instrumented with:

a) chromel–alumel thermocouples to monitor and regulate the temperature,

b) a central ionization chamber to monitor and regulate the dose rate (type CRGA11 manufactured by RTC). Its response vs. energy of the gammas is given in Fig. 6.

The measurement chain is periodically calibrated by a certified laboratory (LMRI). The total dose is calculated every minute using the measured dose rate values.

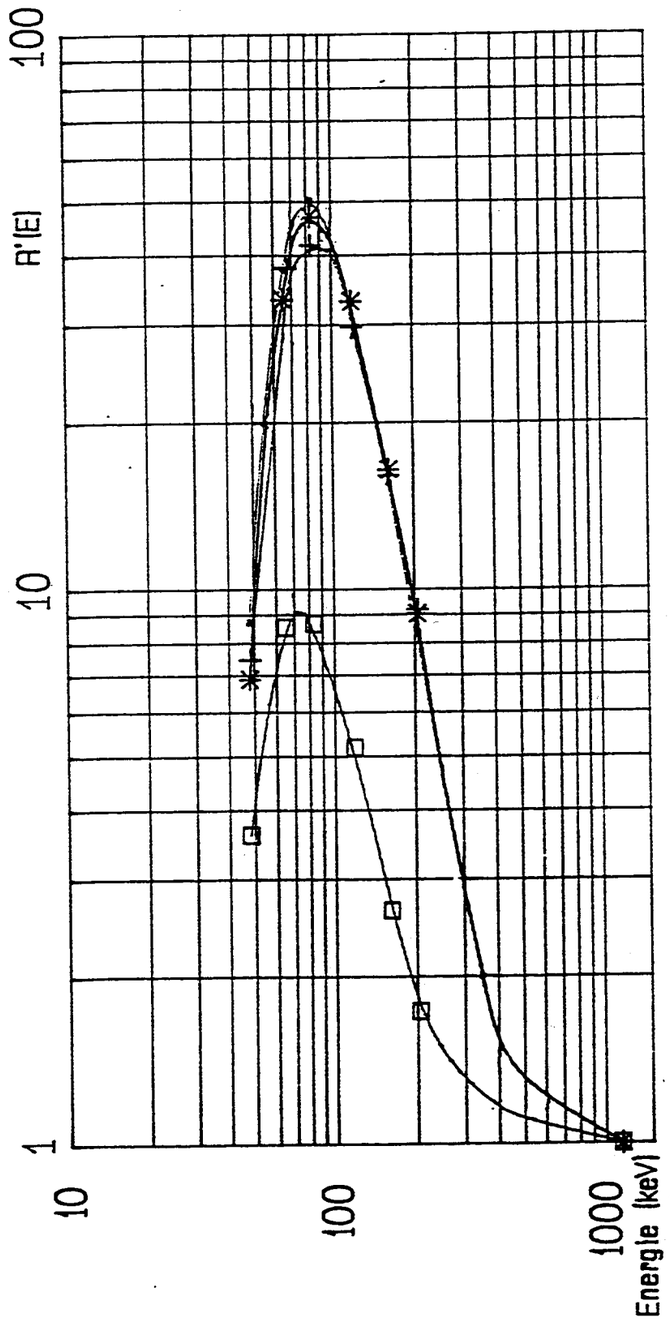

Figure 6: *Ion chambers for irradiation monitoring : response vs. energy of the gammas for 4 types of ion chambers*

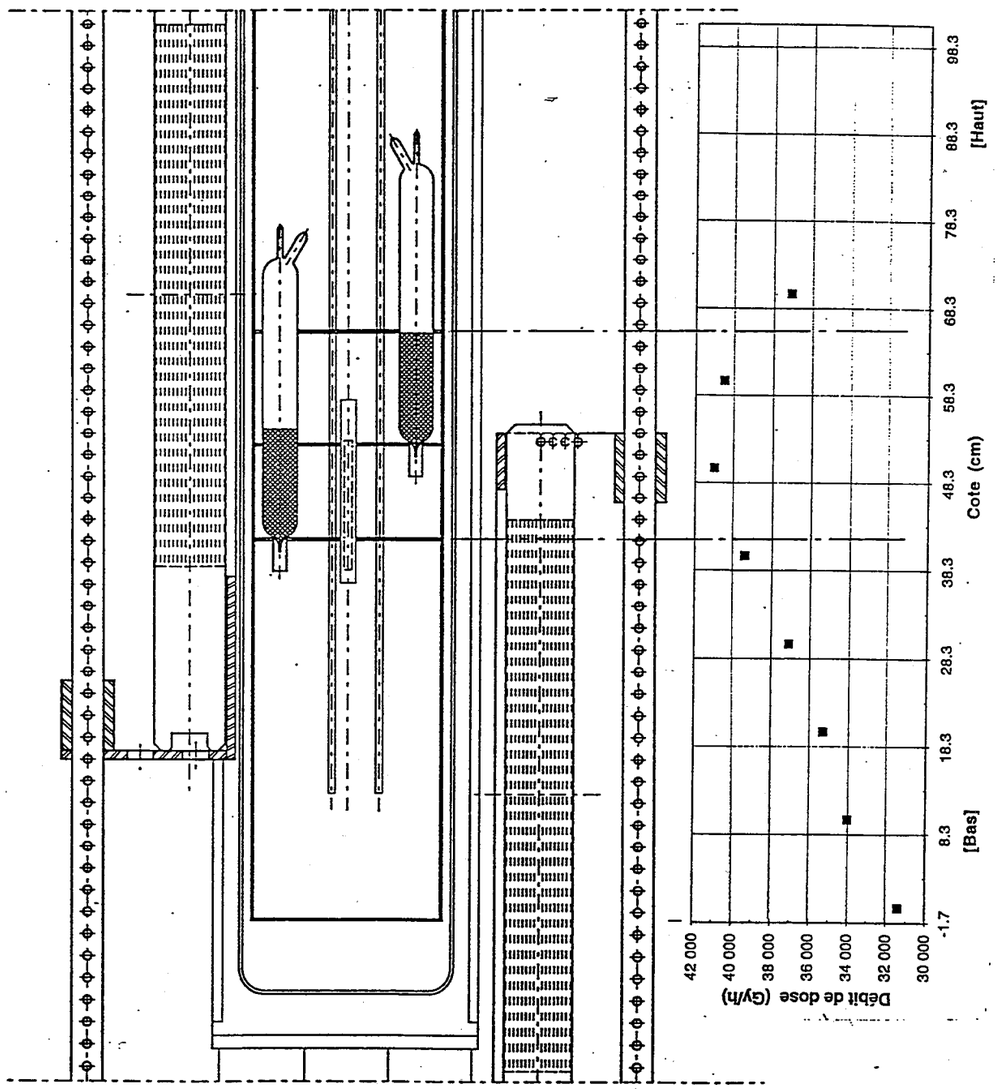

Figure 7: *Flux distribution in the irradiation chamber (irradiation Nr 79)*

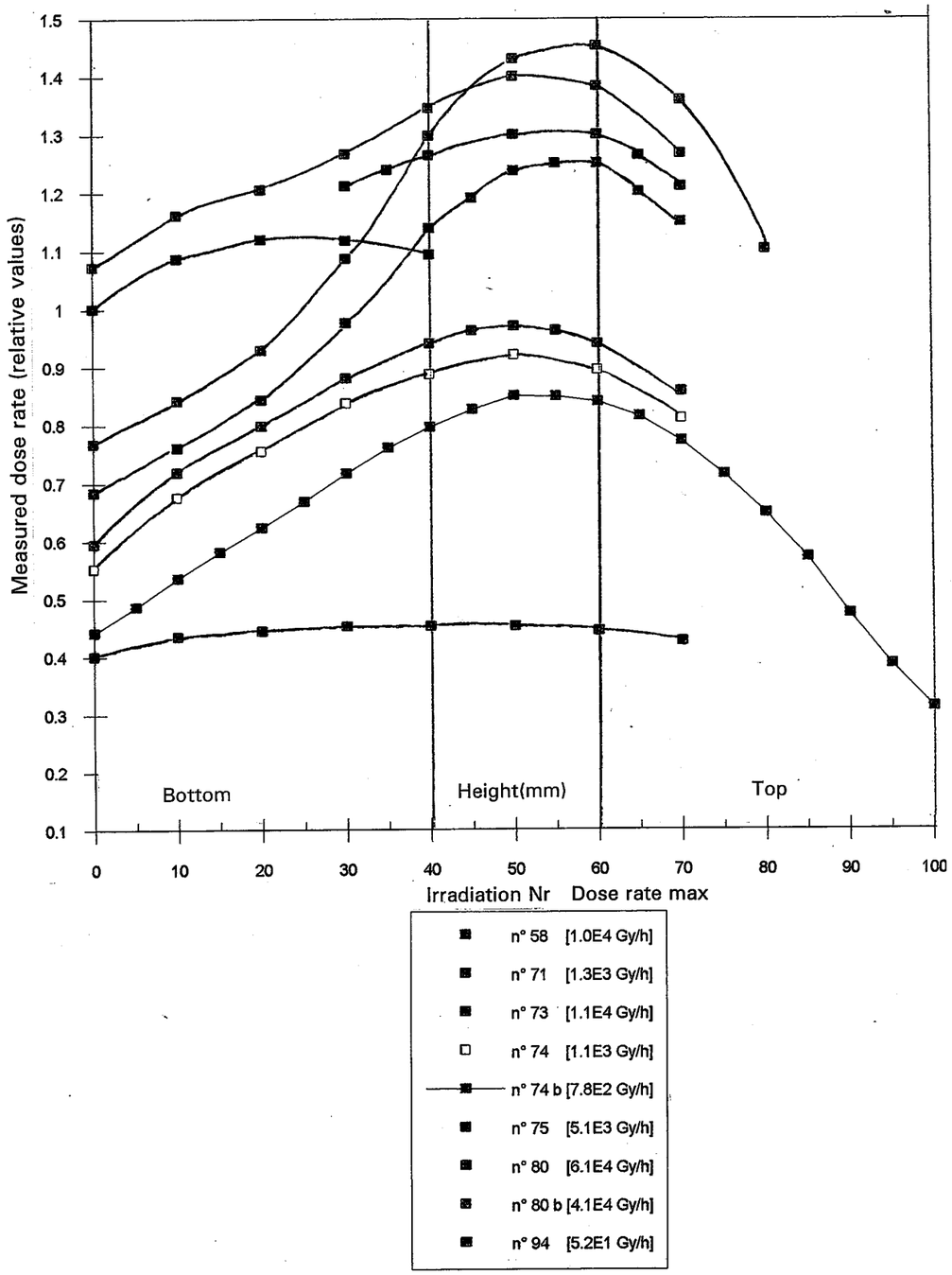

Figure 8: *Axial gamma flux distribution in the irradiator without salt (relative values). The samples are located between 40 and 60 cm.*

The homogeneity of the resulting gamma flux is a function of:

a) the number of γ sources;

b) the activity dispersion between the sources, which is minimized during selection; and

c) differences in radial position between the sources.

The volume where the dose rate is homogeneous may be increased along the vertical dimension by alternately dividing the γ sources between two levels. In addition, the temporal decrease in activity, and therefore of the dose rate, may be compensated by bringing the sources slightly closer together radially.

3.2. Additional irradiators

Three small size irradiators were constructed to be able to perform low dose irradiations of long duration at ambient temperature simultaneously with other experiments in the principal irradiator. An instrumented irradiation chamber capable of accommodating two vials is placed between two fuel elements in a structure adapted from fuel storage racks.

4. PROCEDURES DEVELOPED

Irradiations procedures aimed at complying as much as possible with the contractually fixed values of the following parameters: temperature, dose rate and total dose. According to these procedures, before each irradiation with a new configuration of γ sources, the gamma flux in the empty irradiation chamber is mapped (Fig. 7 and 8), enabling:

a) pre–adjustment of the radial position of the sources in relation to the nominal dose rate level and therefore reduction of the transient experienced by the samples when they are placed in or removed from the furnace;

b) verification of the central axial gradient of the dose rate and therefore of the setting of the sample carrier and the instrumentation.

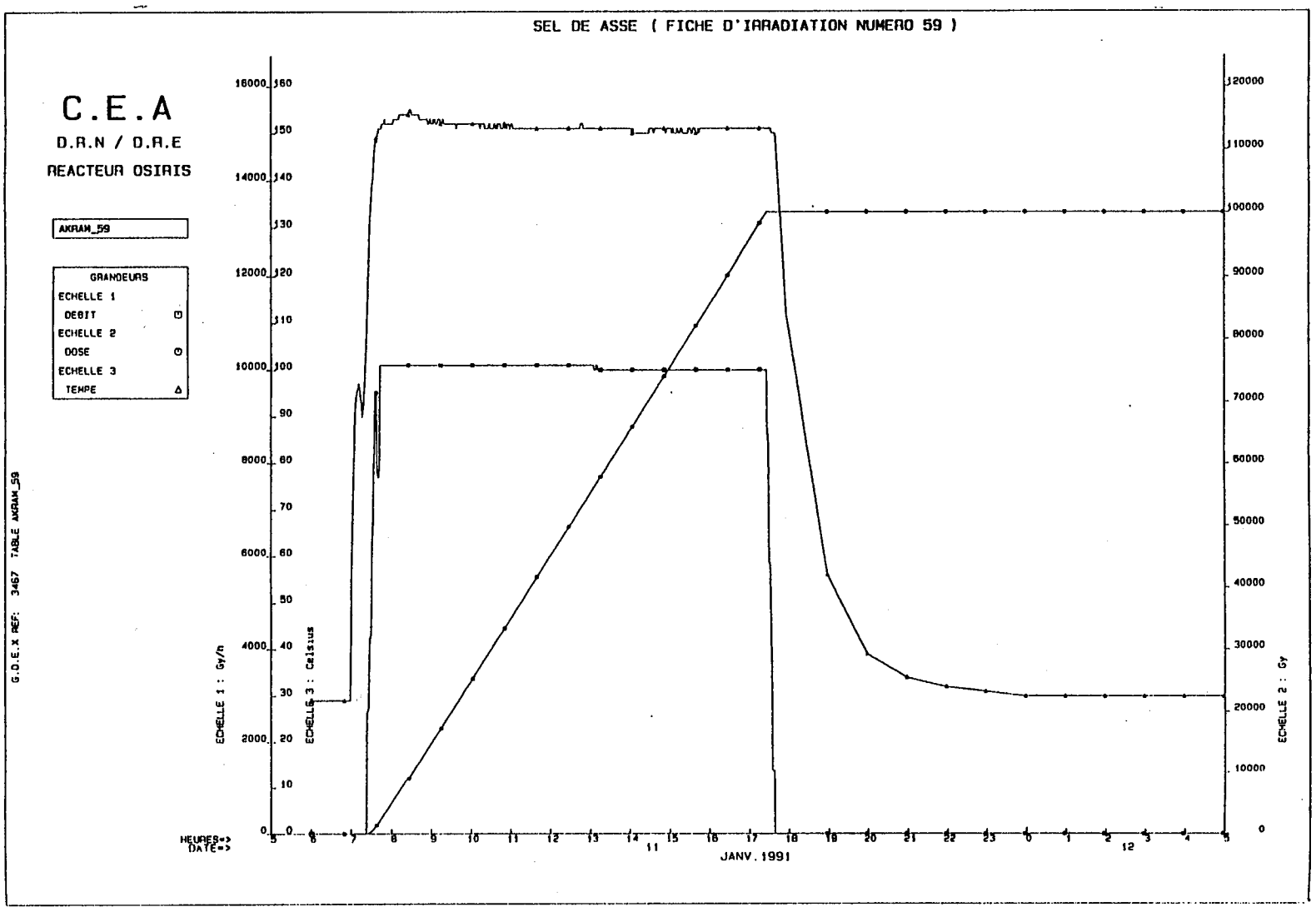

Figure 9a: *Example of graphic data report for an irradiation (Irr. nr 59) : dose rate, total dose and temperature vs. time*

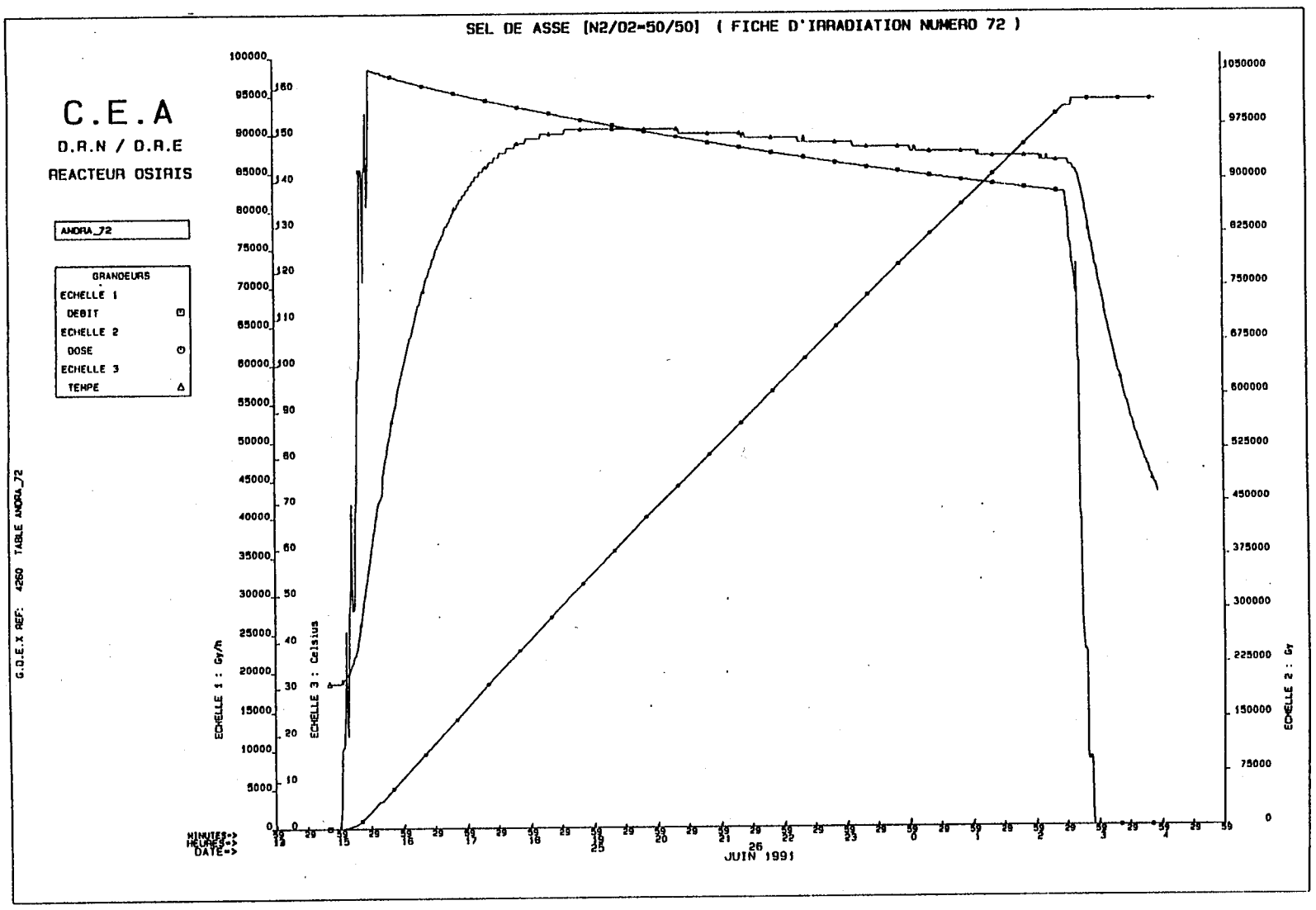

Figure 9b: *Example of graphic data report for an irradiation (Irr. nr 72) : dose rate, total dose and temperature vs. time*

From a safety standpoint, the gamma irradiation installation is the responsibility of the Nuclear Facility Safety Division of the Ministry of Industry; the various authorizations for operation and operating amendments (high dose rates, etc.) had to be procured from this entity following examination of the various design, construction and operation procedures by commissions of experts.

Installation and operating procedures were discussed and approved by the reactor operator.

5. RESULTS OBTAINED

The records in Fig. 9a to 9b show the evolution of the mean temperature of the air surrounding the vials, of the dose rate measured by the ionization chamber and of the resulting total dose during irradiation.

The time to establish or to stop irradiation was kept as brief as possible. However, the return to ambient temperature is not as rapid due to the thermal inertia of the irradiation chamber, a consequence of its design allowing the achievement of high temperatures (400°C).

In the absence of additional electrical heating, the temperature level is determined by that of canal water (measurably constant at some 30°C) and by the absorbed γ energy; In this way, the temperature rises with the dose rate to reach 110°C at 10^5 Gy.h^{-1}. For the same reason, temperature fluctuations are correlated to the small radial displacements that are applied to the fuel elements to regulate the dose rate.

The evolution of spatial and temporal gradients has already been mentioned during the description of the irradiators. It may nonetheless be specified that the maximum dose rate level that can be achieved, taking into account the diameter of the irradiation chamber and the duration of cooling required before the transfer of the elements can take place (radiation protection requirement), is 10^5 Gy.h^{-1}. Since the sources are closest as possible to the samples, it is not possible to maintain a constant rate and their natural decay is observed.

5.1. Accuracy of the results

Temperatures

Ambient air temperatures in the irradiation chamber are measured and appear homogeneous regardless of the temperature and dose rate levels. However, the existence of spatial and temporal temperature gradients in the samples is not to be excluded; they could be estimated through calculations performed using computer simulation of the irradiation chamber and instrumentation of the samples.

Dose Rates and Total Doses

The dose rate levels are estimated by application of the calibration factors determined by irradiation of the ^{60}Co in a range of a few Gy.h^{-1} to some 20 Gy.h^{-1}.

Two additional dosimetries performed at 10^4 Gy.h^{-1} with the alanine at around 10^4 Gy and 35°C (CEA–DTA–DAMRI–LMRI Laboratory) and with calcium sulfate (approximately 10^6–10^5 Gy and 45–200°C) (CEA–IPSN–DPHD–SDOS Laboratory). They show an apparent gross difference of less than 30% with the monitoring system of the irradiator.

A better concordance could be achieved by a better understanding of the spatial gradients of the dose rate, but also of temperature, which can influence sensor response. However, irradiations performed in the framework of the HAW program show that the response of ion chambers type CRGA11 is not influenced by a variation in the level of ambient air temperature, whether the amplitude is low (less than 10°C) or high (greater than 100°C).

One notes that the central axial gradient in the absence of samples is less than 15% over a height of 30 centimetres (Fig. 8); the horizontal gradient is much lower (Fig. 7).

In the framework of another irradiation program, a discordance was noted between the CRGA chambers on the one hand and the PTW–type chambers and alanine dosimeters on the other; the latter estimated a rate and a dose of about a factor two lower than the CRGA chambers, at a dose rate level of on the order of 40 to 70 Gy.h^{-1}.

At these low dose rate levels, the contribution of the radiation diffused may no longer be insignificant and the sensor may be more sensitive to low energies (Fig. 5); it was not possible to determine the energy response of the PTW chambers, nor to go into more depth on the various factors involved.

We note, however, that although there may be a relative uncertainty of a few tenths of a percent in the dose rate level and hence of the dose, the relative values are correct. In fact, measurements have shown the right response of chamber CRGA in the dose rate range : 10^2 to 4.10^3 $Gy.h^{-1}$.

The approach undertaken nonetheless enabled several causes of systematic uncertainties to be identified. To reduce these uncertainties, the following must be done:

a) modification of the irradiator to reduce the flux gradients (rotation of sample carrier, coordinated displacement of fuel elements) and to better understand them (improved instrumentation of the irradiation chamber, filing the data of position of fuel elements and samples, etc.);
b) identification and implementation of other dosimetric processes;
c) performance of several irradiations dedicated to dosimeter under increased surveillance;
d) performance of more irradiations to increase the level of confidence.

6. FURTHER STRATEGIES

The entire salt research program was conducted from 1988 to 1993. Aside from the preliminary preparation of test facilities and their gradual adaptation during the implementation of the program, a large volume of samples was subjected to γ radiation under various spectrum, dose rate, total dose and temperature conditions. The irradiation program went well overall. Good mastery of irradiation planning, preparation and operation was acquired, allowing the principal objectives of the program to be met.

While the accuracy of knowledge of dose rate levels and, subsequently, of total dose is better than an order of magnitude, error values remain nonetheless greater than the 15% that would have been preferred. It is possible that the doses are overestimated. The relative values are most assuredly correct.

It nonetheless appears that miscellaneous improvements could still be made to reduce the systematic uncertainties identified:

a) establishment of preliminary calibration covering the entire field explored (spectrum, dose rate, temperature);
b) identification and implementation of other dosimetric processes in addition to the coverage of the field explored;
c) modifications to the irradiator to reduce residual flux gradients (rotation of sample carrier, coordinated displacement of γ sources, etc.) and to better understand flux gradients (*in situ* cartography, archiving of position data);
d) characterization of thermal gradients inside the samples;
e) modelling of energy deposits and of the associated thermal behaviour; and
f) performance of several irradiations dedicated to dosimetry under heightened surveillance.