Burn up Measurements of LEU Fuel for short cooling Times

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Abstract. The measurements presented in this work were made essentially at in-pool gamma-spectrometric facility, installed inside of the secondary pool of the RECH-1 research reactor, where the measured fuel elements are under 2 meters of water. The main reason for using the in-pool facility was because of its capability to measure the burning of fuel elements without having to wait so long that is, with only 6 cooling days, which are the usual times between reactor operations. Regarding these short cooling times, this work confirms again the possibility of using the $^{95}$Zr as a promising burnup monitor, in spite of the rough approximations used to do it. These results are statistically reasonable within the range calculated using codes. The work corroborates previous results, presented in Santiago de Chile [1], and it suggests future improvements in that way.

1. Introduction
In the near future, the RECH-1 research reactor will be completely converted to the use of LEU (19.75% of $^{235}$U) fuel. The current reactor core loads 22 HEU (45% of $^{235}$U) fuel assemblies fabricated by the UKAEA in Deanery, Scotland, and 12 LEU fuel assemblies fabricated by the Chilean Fuel Fabrication Plant (PEC). The meat composition of the experimental LEU fuel assembly is $\text{U}_3\text{Si}_2\text{-Al}$, whereas the HEU fuel assemblies have a meat composed by $\text{UAl}_x\text{-Al}$. The first two LEU fuel assemblies were loaded in the reactor core in December 1998, and the second two in July 1999. LEU fuel assemblies have been gradually loaded in the core to replace HEU fuel assemblies which have reached the discharged burn-up. The total conversion of the RECH-1 reactor will be achieved during the first semester 2006. The first four LEU fuel assemblies loaded in the reactor core are supporting a local qualification program to know the behavior under irradiation of fuel assemblies fabricated by the PEC.

In order to measure the fuel burnup of irradiated fuel assemblies, the CCHEN has two completely independent facilities using gamma spectroscopy technique: a hot cell facility and an in-pool facility described in earlier works [1],[2]. The first facility is mainly used to measure burn up of spent fuel assemblies with decay periods larger than three months. With the purpose to measure burnup of fuel assemblies with shorter decay periods, it was decided to build an in-pool facility.

The measurement of burnup using gamma spectroscopy technique after long decay period is very well known and $^{137}$Cs as monitor gives reliable results [2],[3],[4],[5]. However, the same measurement with short decay periods (few days) produces serious difficulties in the treatment of the collected experimental data. The origin of these difficulties is the high activity generated by a large number of fission products of short life time, which increases the dead time and background reducing the quality of the statistics of the monitor [6], and in our experience submerging completely the $^{137}$Cs under the background radiation, even with 4 months operation and 6 cooling days. Monitors like $^{95}$Zr, $^{140}$La,
$^{103}$Ru, $^{95}$Nb, etc. have good statistics; however, they have too short life to keep memory of the accumulated burnup for long irradiation time. Remember that the maximum accepted burnup is 50%.

2. Measurements and Results

The burn up of one Chilean LEU fuel assembly, LR-04L of the RECH-1 research reactor with short decay period was measured at the in-pool facility using $^{95}$Zr (724 keV, peak) as monitor in 2003 [1], and in 2005 were measured the same assembly, plus its similar LR-047. The methodology presented in this paper should be taken as a verification of these first attempts to use $^{95}$Zr as a monitor. In figure 1, is showed in a qualitative way, the origin of a new algorithm to develop the F factor needed to compensate for cumulative decay of $^{95}$Zr occurring during the different irradiation periods between the measurement and the relevant initial core operation, and to take into account his mentioned short memory. In figure 1, the red and blues lines only tries to sketch the general behaviour of the relevant involved physics processes.

![Figure 1](image)

**Figure 1**

Generation-Decay Scheme of Radionuclides $^{95}$Zr during a succession of operations at RECH-1

To formulate the F factor three simple assumptions were taken into account and with the figure 1 notation, we have:

Since the RECH-1 is a thermal reactor, we assume the involved cross sections in fission’s processes are essentially thermals.[8]

The amount of $^{95}$Zr nuclides $n_{0i}$ measured is approximately equal to the amount of nuclides $N_{0i}$ generated during each operation. (See figure 1)

In each operation approximately the same net amount of $^{95}$Zr is generated. In agreement with the names of Figure 1, and under the previous assumptions, the spectrometric measured burnup in each operation of period $T$, is given essentially by the number,
(N_0 - n_{0-0.4})e^{-\lambda \tau}/y_{95}, where \( \lambda \) is the disintegration constant of \(^{95}\)Zr, and \( y_{95} \) is the fission yield of \(^{95}\)Zr, then the accumulated burnup in the time of measurement, which is the searched for value, turns out:

\[
F = \left[ k + e^{-\lambda \tau} (p + 1 - k) - p e^{-\lambda T'} - e^{-\lambda T''} \right]
\]

(1)

Where, \( k \): total numbers of reactor operations,

\( \tau \): Time between operations,

\( T' \): maintenance period, and

\( p \): number of maintenance periods while the fuel assembly was in the reactor core.

\( \lambda \): \(^{95}\)Zr Disintegration Constant.

\( T'' \): non-routine decay time.

Fig. 2 shows a spectrum of LR-04L assembly after a decay period of 51 days, and Fig. 3 shows the spectrum of the same assembly obtained after a decay period of 5 days. The spectra shows the great differences in the peaks of present radionuclides between the two very diverse physical situations.

### Table 1

| Fuel Assembly Identification | Date of Measurement | Decay Period [d] | Measured Burnup [\(^{95}\)Zr] % | Calculated Burnup [Codes] % | (M-C)/C % |
|----------------------------|---------------------|------------------|---------------------------------|----------------------------|------------|
| LR-4L                      | 30/01/03            | 6                | 27.88                           | 22.78                      | 22.4       |
| LR-04L                     | 13/03/03            | 6                | 23.68                           | 23.00                      | 2.96       |
| LR-04L                     | 12/06/03            | 6                | 25.08                           | 24.52                      | 2.28       |
| LR-04L                     | 13/01/05            | 6                | 25.16                           | 29.98                      | -16.01     |
| LR-04L                     | 20/01/05            | 6                | 29.07                           | 29.98                      | -3.03      |
| LR-47                      | 27/01/05            | 6                | 2.44                            | 1.95                       | 25.12      |

The table 1 above, shows measured burnup results obtained for the LR-04L and LR-47 fuel assemblies using \(^{95}\)Zr (at fourth row) as monitors and the algorithmic method in (1), compared with Citation code results (fifth column). The sixth column shows the percentage difference between measured (M) and calculated (C) values, referred to this last one.
3. Conclusions
Considering the simplicity of the physical assumptions used to formulate F, and the approximately supposed periodical operation of RECH-1 used on the algorithm to calculate F, the results are promising. Nevertheless the Table 1 shows that the percentage differences between the measured burnup and the calculated burnup have visible instabilities. However given the rude approximation
used for the $^{95}$Zr, and the common error associated as much to the spectrometric measurements as to the codes results, (15 and 10 % respectively [3],[8]) it seems reasonable to follow with these works to refine it.

Three possible improvements (in increasing order of importance) are suggested in algorithmic and experimental fields:

(i) Algorithmic: Take into account the actual power history of RECH-1 reactor in a very much detailed way, as with the $^{137}$Cs monitor. It is possible that the hypothetical benefit in precision could be smaller than the loss in the simplicity of the present F.

(ii) Experimental: The HPGe detector must be fixed to the mechanical measurement system, due to the fact that it produces important variations in activity measurements, which only contributes to increase the uncertainty in the efficiency of system.

(iii) Experimental and Algorithmic: It is necessary to make continuous measurements during a sufficient time (3 or 4 months) for a specific LEU assembly, in order to improve the empirical knowledge of $^{95}$Zr behavior, between and in routine reactor operations.

The second improvement (experimental) has been made during the four last month, and it is absolutely desirable to evaluate experimentally, from now on, the associated error of the three mentioned assumptions used for the generation of factor F, mainly the third of them (assumption c) because it is probably the most relevant and the main error source in final results.

**References**

[1] C. Pereda, C. Henríquez, J. Medel, J. Klein, and A. Kestelman, “Fuel Burnup Measurements Using Gamma Spectrometry Technique”, ICRR 2003, Santiago de Chile, November 2003.

[2] C. Henríquez, G. Navarro, C. Pereda, H. Torres, L. Peña, J. Klein, D. Calderón, and A. J. Kestelman, “Burnup Measurements at the Rech-1 Research Reactor”, RETR 2002, November 2002, Bariloche, Argentina.

[3] C. Henríquez, G. Navarro, C. Pereda, and G. Steinman, “Medición de Quemado de un Elemento Combustible Experimental Mediante Espectroscopía Gamma”, Nucleotécnica, 35, 71-83, (2001).

[4] L.A.A. Terremoto, C.A. Zeituni, J.A. Perrota, and J.E.R. Da Silva, “Gamma-ray spectroscopy on irradiated MTR fuel elements”. Nucl. Instr. Meth. A 450 (200) 495-514.

[5] A. J. Kestelman and S. Ribeiro Guevara, “Determinación del Quemado en Combustible tipo MTR Mediante Espectrometría Gamma con Cristal de INa (Tl)”, Informe Técnico CNEA-CAB 88/034.

[6] Debertin K. and Helmer R., “Gamma and X-Ray Spectrometry with Semiconductor Detectors”, North Holland, (1988).

[7] P. Suárez and A. Kestelman, “Determinación no destructiva del quemado en elementos combustible tipo MTR mediante espectroscopía gamma de alta resolución”, Memoria de Título, Instituto Balseiro Universidad Nacional de Cuyo, CNEA, (Junio 1989).

[8] J. Medel R., Private Communication. RECH-1 (2005).