BURNUP MEASUREMENTS AT THE RECH-1 RESEARCH REACTOR

C. Henríquez, G. Navarro, C. Pereda, H. Torres, L. Peña, J. Klein, D. Calderón. Unidad de Reactores, Reactor La Reina (RECH-1), Comisión Chilena de Energía Nuclear, Amunategui 95, Casilla 188-D, Santiago de Chile. E-mail: chenriqu@cchen.cl, Fone: 56 2 364 6154, Fax: 56 2 273 1827 A. J. Kestelman

Laboratorio de Análisis por Activación Neutrónica, Centro Atómico Bariloche e Instituto Balseiro. 8400 Bariloche - Argentina

ABSTRACT

The Chilean Nuclear Energy Commission has decided to produce LEU fuel elements for the RECH-1 research reactor. During December 1998, the Fuel Fabrication Plant delivered the first four fuel elements, called leaders, to the RECH-1 reactor. The set was introduced into the reactor's core, following the normal routine, but performing a special follow-up on their behavior inside and outside the core. In order to measure the burn-up of the leader fuel elements, it was decided to develop a burn-up measurements system to be installed into the RECH-1 reactor pool, and to decline the use of a similar system, which operates in a hot cell. The main reason to build this facility was to have the capability to measure the burn-up of fuel elements without waiting for long decay period. This paper gives a brief description of the facility to measure the burn-up of spent fuel elements installed into the reactor pool, showing the preliminary obtained spectra and briefly discussing them.

INTRODUCTION

As it is well known, the disponibility of a burnup data set is an essential first step in any systematic approach to knowledge of the physical behavior and consequent optimize of the fuel elements performance in the core of a research reactor. From the beginning of RECH-1 operations, the burnup evaluation was made through neutronic calculations, which needs experimental verification. Gamma Spectrometry allows a non-destructive determination of the fission and activation product contained in spent fuel reactor. The concentration of these products depends significantly on the fuel parameters which describes the irradiation history and the burnup fuel composition. Therefore, performing gamma spectrometry for the reactor fuel, its burnup can be determined experimentally. This gamma ray spectrometry method relies on the assumption that some specific gamma active nuclides are produced at well known and definite rates when heavy elements nuclei undergo fission. Among these, it has to be selected the burnup monitor, which ought to satisfy the following fundamental requirements:

- High fission yields.
- Negligible neutron captures cross section
- Long half-life with regard to fuel elements irradiation time
- High-energy gamma peaks in order to minimize attenuation's correction.
- Low migration in fuel.

In agreement with these criteria, usually recommended radioactive monitoring isotopes are: 95 Zr (64.03d), 106 Ru- 106 Rh(373.59d), 134 Cs(2.065y), 137 Cs(30.07)y, 144 Ce- 144 Pr(284.89y), 154 Eu(8.593y), 140 Ba- 140 La(12.75d).

The first measurements were made in a Hot Cell, using an experimental Chilean LEU fuel element with only one active plate. The experimental results, utilizing Cs-137 like monitor, were comparables with those obtained using codes, taking into account the measured error estimated in about 10%. Hot Cell measurements, however, are very difficult to carry out in the case of new LEU operational elements because of their very high activity (with consequent transport problems) and their very short cooling time(< 7 days).

In order to satisfy previous needs, an in-pool measurement device was designed and constructed. During last year, it was finally adjusted and set up to work inside the primary pool of the reactor. A graphical description of the complete structure to perform these measurements is shown in Figure 1. The equipment is composed essentially of two parts: (1) the mechanical measurements system, which includes a lead shielding HPGe detector of 30% relative efficiency and 1.8 keV resolution, and a 3 meter dry tube carrying a double set of collimators of various diameters (2.8 to 8 mm), everything carried by a little cart, with x-y movements.

(2) Stainless steel support structure with a fuel element upholder table in its lower end.

The associated electronic and computational data acquisition system consists of an ADC, an AFT Amplifier 2024, specially designed for high-count rates, and the Genie 2000 Software for spectral analysis.



EXPERIMENTS AND RESULTS

Before burnup measurements, it is necessary to have accurately determined the absolute efficiency ε of the measuring system, which compel to a very careful set of measurements performed outside of the reactor wall, trying to recreate identical geometric final conditions of the in-pool measurements system. To evaluate the absolute efficiency, a simile of a LEU fuel element was constructed and used. The attenuation factors due to water between plates; and the 7.4 mm water between the upper of the element plate and the lower end of the collimators scanning tube, were always considered in the experimental device

To perform all these measurements, a high activity Eu-152 source was used. Its initial activity was 3.66 mCi \pm 3.5%, on January 28 1995, as it is shown on its calibration certificate. The Eu-152 active isotope, is inside a ceramic matrix, and covered by a 15.9-mm long stainless steel cylinder.

Figure 2 and 3, shows two representative efficiencies curves got in two different times: the first in February 2002 and the second in July 2002. The radical different behaviors, above all, at high-energy range, have origin in the inherent alignment difficulties, because of the long distances involved. Another reason is the geometric differences between the upper and lower in-tube collimator pair, which was solved, by increasing the lead thickness between the lower face of the detector and the top collimator, from 2.5 cm to 10 cm, as seen in the second case. The measuring time in both cases was 12 hours.





In order to perform the gamma spectrometry measurements, two irradiated fuel elements were selected. The first was a chilean made LEU leader element of 19.75 % enrichment, and the second was a english made MEU element of 45% enrichment. The leader element was measured between two reactor's operations, that is, with 5 days of cooling time, while the MEU had a cooling time of 94 days after its second time of permanence in core. Both irradiation histories are well known. Two representative spectra of each one are shown in figures 4 and 5. In the figure 4, it is shown a gamma ray spectrum obtained from measurements of 100 s of live time, at the central point of the irradiated fuel element, and the figure 5 shows a gamma ray spectrum from a MEU element, with 1800 s of live time.





In order to correct for possible instabilities of the measuring system, and also to check on dead time and pile-up corrections, all measurements were referred to the 1332 [keV] produced by a low intensity ⁶⁰Co (1925.1d) monitor source that was simply taped to the HPGe detector. The ⁶⁰Co peaks due to this source are clearly observed in the spectra shown in Figures 4 and 5. As it is known, ⁹⁵Zr is a primary fission product and ⁹⁵Nb comes essentially from the beta decay of ⁹⁵Zr. The ⁹⁵Zr high fission yield, and its half-lives related to the cooling times in both cases, explain its abundance. The fundamental difference between both spectra, from the point of view of the analysis results, is the strong presence of the characteristics fission monitors ¹³⁴Cs, ¹³⁷Cs in MEU spectrum, and the great decrease of 140La in the same case. The reason of this behavior is, naturally, the very high activity in the LEU spectrum and the very different respective cooling times.





2002 International Meeting on Reduced Enrichment for Research and Test Reactors, Bariloche, Argentina, November 3-8, 2002



Fuel Element LR04L 19,75% Activity Profile of Zr-95 September 2002



Figures 6, 7 shows the burnup profiles of LR-04L LEU element, measured on a interval of about 6 months.. These measurements were performed along the central line of the active length in the first plate of this element. 13 steps of 5 cm long, were selected starting from our zero, which corresponds to 11 cm on the rule joined to the fixed observation position point, where the activity starts to be measured in our spectrometry system. The selected isotope, for this case, was ⁹⁵Zr, because of its very suitable peaks, which are of high energies and abundant intensity.

Figure 8, shows an analog burnup profile, based on the areas under full energy peaks of 661.6 [keV] from ¹³⁷Cs. Again, the activity profile was measured along the longitudinal

central line on the first plate of LR-06 MEU fuel element, in identical steps of the preceding described situation. A burnup calculation using the measurements for this element was made using the ¹³⁷Cs monitor and the efficiency curve showed in figure 3. The fuel composition is UAl_x-Al and the fuel element is made up with 14 internal plates with $12.21[g]^{235}$ U and two external plates, with half uranium mass aforementioned.

CONCLUSIONS

- The technique to measure burnup through gamma spectrometry provides, by nondestructive analysis, a considerable amount of reliable information which permits to have a method, with necessary future improvement, to make burnup measurements and the following up of this fuel element behavior.
- The necessary improvements mentioned before refer, above all, to the adequate selection of a method to evaluate the short cooling times, involved in the measurements performed in pool.
- Making use of an analytical algorithm to treat the collected experimental data via gamma spectrometry, similar burnup results have been obtained, compared with code calculations based on Reactor Physics, taking into account the experimental error associated with this kind of measurements. The MEU fuel element burnup aforementioned happens to be in agreement with our estimations, 38.8% versus 40.7% get via codes.
- Throughout these experimental works, we have verified that ¹³⁷Cs is an excellent fission monitor, even for cooling times less to those considered in related literature. According to our case, ¹³⁷Cs gave excellent results, with cooling times of about three months and it wasn't perceived the influence of the Compton contribution of the close peaks coming from ⁹⁵Zr and ⁹⁵Nb.

ACKNOWLEDGEMENTS

The authors would like to express their gratitude to RECH-1 Operation Group, and specially to Mr. Octavio Mutis for his kind cooperation in the final version of this poster

BIBLIOGRAPHY

- [1] Kestelman A.J., Ribeiro Guevara S., "Determinación del Quemado en Combustibles tipo MTR Mediante Espectrometría Gamma con Cristal de INa (Tl)", Informe Técnico CNEA-CAB 88/034.
- [2] C Henríquez, G. Navarro, C. Pereda, G Steinman, "Medición de Quemado de un Elemento Combustible Experimental Mediante Espectroscopía Gamma", Nucleotécnica, 35, 71-83, 2001.
- [3] Suarez P., Kestelman A., "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.

- [4] Debertin K., Helmer R. "Gamma- and X-Ray Spectrometry with Semiconductor Detectors" North-Holland, 1988.
- [5] Knoll, G. "Radiation Detection and Measurements", John Wiley & Sons, 1979.
- [6] Mutis O., Medel J., Klein J, "Conversión del RECH-1 a combustible de bajo enriquecimiento". Departamento de Aplicaciones Nucleares, CCHEN, (1994).
- [7] "Determination of Research Reactor fuel Burnup", IAEA-TECDOC-633., January 1992.