BURN-UP MEASUREMENTS OF SPENT FUEL USING GAMMA SPECTROMETRY TECHNIQUE

C. Pereda, C. Henríquez, J. Klein and J. Medel

Comisión Chilena de Energía Nuclear, Amunátegui 95, Santiago, Chile.

ABSTRACT

Burn-up results obtained for HEU (45% of ^{235}U) fuel assemblies of the RECH-1 Research Reactor using gamma spectrometry technique are presented. The spectra were got from an in-pool facility built in the reactor to be mainly used to measure the burn-up of irradiated fuel assemblies with short cooling time, where ^{95}Zr is being evaluated as possible fission monitor. A program to measure all spent fuel assemblies of the RECH-1 reactor was initiated in the frame of the Regional Project RLA/4/018: "Management of Spent Fuel from Research Reactors".

The results presented here were obtained from HEU spent fuel assemblies with cooling time greater than 100 days and ^{137}Cs was used as fission monitor. The efficiency of the in-pool system was determined using a slightly burnt experimental fuel assembly, which has one fuel plate (one of the outer plates) and the rest are dummy plates. An average burn-up of 2.8% of ^{235}U was previously measured for the experimental fuel assembly utilizing a facility installed in a hot cell and ^{137}Cs was used as monitor.

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 Dounreay, Scotland, and 12 LEU fuel assemblies fabricated by the Chilean Fuel Fabrication Plant (PEC). The meat composition of the experimental LEU fuel assembly is U_3Si_2-Al, whereas the HEU fuel assemblies have a meat composed by UAl_x-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 burn-up of irradiated fuel assemblies, the CCHEN has two completely independent facilities using gamma spectrometry technique: a hot cell facility and an in-pool facility. The first facility is mainly used to measure the burn-up of spent fuel assemblies with cooling time larger than three months, while the second is mainly used to measure the burn-up of fuel assemblies with shorter cooling time.

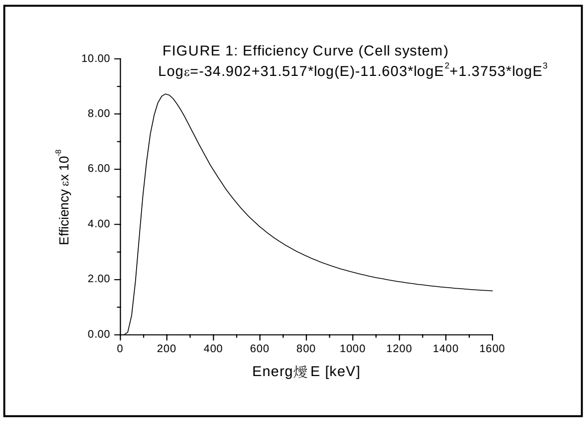
For long cooling time, ^{137}Cs is an excellent fission monitor [1,4]; however, this monitor could be used up to cooling time of about 90 days with good reproductiveness of the measurements and reliable results [2,3,5,6,8]. The results here presented are confirming the quality of ^{137}Cs as monitor for irradiated fuel assemblies with cooling time larger than 100 days.

There are great experience in the measurement of burn-up in several places using gamma spectroscopy technique and ^{137}Cs as fission monitor; however, the use of this technique for irradiated fuel assemblies with short cooling time (few days) produces serious difficulties in the treatment of the collected data. The origin of these difficulties is the high activity generated by a large number of fission products of short half life, which increases the dead time of the measuring system and the background, both effects reduce the quality of the statistics of the monitor. Monitors like ^{95}Zr, ^{140}La, ^{103}Ru, and ^{95}Nb have good statistics; however, they have too short half life to keep the accumulated burn-up for long irradiation time.

2. Measurements

Before collecting the spectra it is necessary to determine the absolute efficiency, ε, of the measuring system, which includes a set of careful measurements performed outside of the reactor pool and a recreation of the proper conditions once the system is placed in the reactor pool. With the purpose to determine the absolute efficiency, a fuel assembly with removable LEU fuel plates was fabricated by the PEC. In the experimental arrangement, the attenuation factors due to the water between the plates of the fuel assembly and the 7.4 mm water gap between the outer plate and the lower end of the collimators scanning tube were considered. The measurements were performed using a ^{152}Eu source with an initial activity of 3.66 mCi ± 3.5% determined on 28 January, 1995 in accordance with the calibration certificate. The efficiency obtained with that source was satisfactory; however, measurements later performed demonstrated that this source does not represent neither the geometry nor the material of a fuel assembly. Additionally, the collimators system is difficult to align and taking in account that the detector is detachable some instabilities in the results were found.

With the purpose to improve the methodology of measurement and to have a better representation of the behavior of a spent fuel assembly, it was decided to use an experimental LEU fuel assembly slightly burnt (2.8 %), which has one fuel plate while the others plates are dummy. The burn-up reported for the experimental fuel assembly was achieved from reliable measurements, which were done in the hot cell system; thus, it was decided to use that fuel assembly as calibration source. The fuel plate of the experimental assembly was divided in 11 zones of equal length. Consequently, counts of a total of 11 points were collected.



The efficiency curve given in Figure 1 was obtained in the hot cell system. This curve will be used to evaluate the absolute efficiency for the in-pool system using ^{137}Cs as fission monitor.

To obtain an expression which gives the absolute efficiency of the in-pool system, it will use the same hypothesis that it was used in the burn-up algorithm; i.e. the density of activity from a small area of the fuel plate is taken on equal independently if the measurement of that position was done in the hot cell system or in the in-pool system. Consequently, for the cell system:

$$\rho_C \cdot s \cdot \left[\frac{\pi b^2}{4}\right] \cdot S_C(E) P(E) \varepsilon_C^{abs}(E) = \left[\frac{Cts(x)}{t_m}\right]_C \qquad (1a)$$

and similarly for the in-pool system:

$$\rho_P \cdot s \cdot \left[\frac{\pi a^2}{4}\right] \cdot S_P(E) \cdot P(E) \varepsilon_P^{abs}(E) = \left[\frac{Cts.(x)}{t_m}\right]_P \qquad (1b)$$

where, ρ_p and ρ_c is the density of activity at the position x; s is the meat thickness; a y b are the main collimators diameters in the cell and in-pool system; $S_p(E)$ and $S_c(E)$ are the attenuation factors; P(E) is the fission yield of the monitor at the energy E; $\varepsilon_p(E)$ and $\varepsilon_c(E)$ are the absolute efficiency; t_m is the time of measurement; and Cts(x) are the counts at the position x. The sub-indices P y C mean pool and cell, respectively.

Taking into account that,

$$\rho_P = \rho_C e^{-\lambda \Delta t} \qquad (2)$$

and from equations (1a) and (1b), it obtains the expression,

$$\varepsilon_P^{abs}(E) = \frac{\left[\frac{Cts}{t_m}\right]_P}{\left[\frac{Cts}{t_m}\right]_C} \cdot \left[\frac{b}{a}\right]^2 \cdot \frac{S_C}{S_P}(E) \cdot e^{\lambda \Delta t} \cdot \varepsilon_C^{abs}(E) \qquad (3)$$

where, Δt is the elapsed time between cell and in-pool measurements, and λ is the half life of ^{137}Cs.

It is useful to define the factor, G(E), as follows:

$$G(E) = \left[\frac{b}{a}\right]^2 \frac{S_c}{S_P}(E)\, e^{\lambda \Delta t} \qquad (4)$$

For the energy peak of ^{137}Cs (661.66 keV), and with a = 0.28 cm, b = 0.40 cm, S_c = 0.96629, S_p = 0.87508, $\lambda = 6.2777\times 10^{-5}\ d^{-1}$, and Δt = 1435 d, the value of the factor G(E) turns out to be 2.46579.

The absolute efficiency found for the cell system was 3.76×10^{-8}, which did not depend on the position where the measurement was done. From equation (3), it was found that the efficiency for the in-pool system depends on the measuring position as it is shown in Table 1.

Table 1.- Absolute efficiency for cell and in-pool system

Position, cm	ε_C	$(Cts/t_m)_P$	$(Cts/t_m)_C$	ε_p
27.5	3.76×10^{-8}	1.94×10^{-1}	2.92×10^{-1}	6.17×10^{-8}
58.5	3.76×10^{-8}	2.74×10^{-1}	4.43×10^{-1}	5.73×10^{-8}

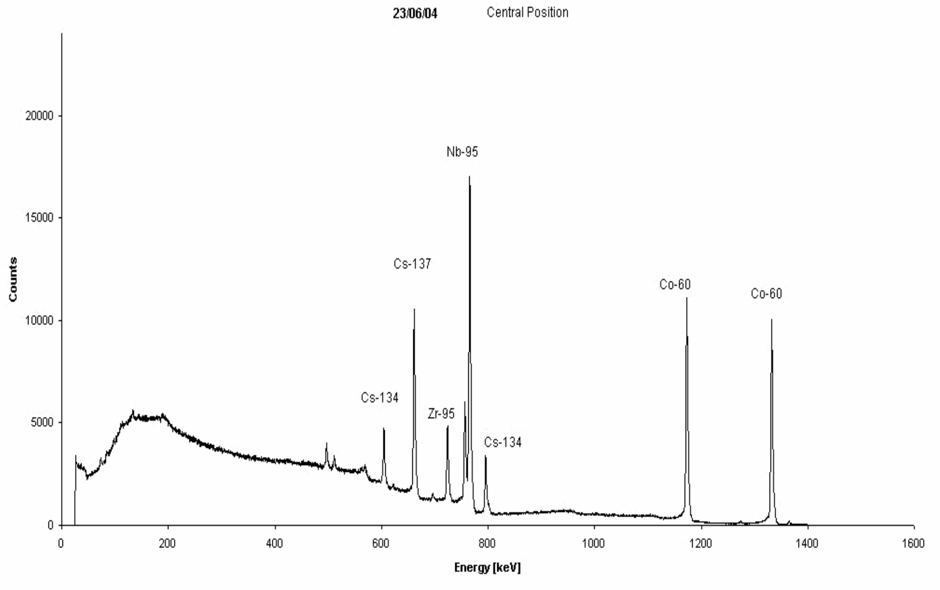
As a first approximation, it was decided to take the average value of the in-pool efficiencies given in Table 1 as the absolute efficiency for the system, whose value is:

$$\overline{\varepsilon}_P^{abs}(^{137}Cs) = 5.95 \times 10^{-8} \qquad (5)$$

Therefore, the efficiency given by equation (5) will be used to evaluate the burn-up of HEU fuel assemblies. Figure 2 shows the spectrum obtained from the LR-17 fuel assembly, which was loaded in the reactor core on 21 March, 1989 and unloaded on 24 January, 2004. During this period, the assembly was irradiated 520 days.

The spectrum was obtained counting during 1800 s, which shows well defined peaks for ^{95}Nb, ^{95}Zr, ^{134}Cs, and ^{137}Cs. In order to correct for possible instabilities of the measuring system, and to check on dead time and pile-up corrections, all measurements were referred to the 1332 keV peak produced by a low intensity ^{60}Co (5.27 y) source, which was simply taped to the HPGe detector. The ^{60}Co 1173 and 1332 keV peaks are clearly shown in the spectrum of the Figure 2.

Figure 2



3. Results

Table 2 and 3 show the burn-up values determined in two independents set of measurements during 2003-04. In total, 11 HEU fuel assemblies were measured. Table 2 shows burn-up values obtained using a source of ^{152}Eu to determine the efficiency of the system, and Table 3 shows burn-up results obtained after determining the efficiency using the experimental fuel assembly as was mentioned before. The burn-up calculated values were obtained using neutronic codes (WIMS-CITATION).

Table 2: Burn-up results using a source of ^{152}Eu to determine efficiency

Fuel Assembly	Date of Measure	Cooling Time, d	Measured Burnup, %	Calculated Burnup, %	Δ, %
LR-01	09/09/03	139	33.50	44.25	24.19
LR-15(1st)	26/11/03	882	48.00	41.30	16.22
LR-04	07/01/04	1226	34.33	39.13	12.26
LR-23	08/01/04	259	38.62	44.15	12.52
LR-19	13/01/04	1357	39.75	38.28	3.84
LR-21	14/01/04	588	38.09	41.39	7.97
LR-15(2nd)	15/01/04	932	31.46	41.30	23.83

Table 3: Burn-up results using an experimental fuel assembly to determine efficiency

Fuel Assembly	Date of Measure	Cooling Time, d	Measured Burnup, %	Calculated Burnup, %	Δ, %
LR-17	23/06/04	118	44.74	46.22	1.01
LR-27	24/06/04	119	35.96	46.88	19.94
LR-35	28/09/04	696	35.87	43.03	12.99
LR-10	29/09/04	697	35.58	43.01	13.67
LR-03	06/10/04	223	41.45	45.19	8.27

In general, the measured values of burn-up turned out to be lower than the calculated values, behavior that was shown in references [2,3,7]. The source of experimental error is attributed to the efficiency, algorithm, geometry, statistic factors, calibration, quality of the collected data, alignment of the system, etc. It recognizes that a representative error could be very difficult to estimate due to the correlation among the factors involved. In accordance with the previous burn-up measurements, it could establish that the greatest effect has its origin in the first two factors; i.e. efficiency and algorithm. Thus, at a rough estimate it could give an error of about 15%.

4. Conclusions

The burn-up measurements for HEU fuel elements with cooling time larger than 100 days have confirmed that the ^{137}Cs is an excellent fission monitor and, taking into account the results obtained, the algorithm used will be maintained to measure the whole spent fuel assemblies of the RECH-1 research reactor.

The efficiency for the in-pool system, which was obtained using the experimental LEU fuel assembly slightly burnt, gives results of burn-up statistically reasonable, representing an improvement in comparison with those obtained with the efficiency determined using a ^{152}Eu source.

It is recommended to improve the procedure to align the set of collimators and to modify the way to recharge nitrogen into the detachable detector, both with the purpose to avoid changes, which could perturb the measurements.

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6. References

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