THE "EQUIVALENT PLUTONIUM" CONCEPT AND ITS APPLICATION TO SYNERGETIC FUEL CYCLES CALCULATION

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ABSTRACT

The advanced fuel cycles are seen as very interesting alternatives to improve the utilization of Uranium resources in the middle term .

Among them, the synergetic cycles between different type of reactors, particularly PWR and CANDU are seen as very promising.

In the frame of the Argentinean-Brazilian cooperation agreement, a neutronic and economical study was done on a Tandem cycle between the Brazilian Pressurized Water Reactor Angra-I, and the Argentinean CANDU reactor Embalse.

The first calculations showed very interesting results regarding the obtainable savings in natural resources, the cost of the fuel cycle, and the lower quantity of wastes to be disposed.

To perform the initial calculations, two methods were mainly used: standard calculation codes, which use discrete ordinates or collision probabilities method to solve the neutronics of the cell, or an algorithm that from now on we will call EQUIVALENT PLUTONIUM.

The present work describes the concept in which the algorithm is based, the obtention of the coefficients needed for its determination, and, as an example, the results obtained applying the algorithm to two particular cases of Tandem cycles:

- CANDU MOX fuel fabricated from PWR fuel, diluted with natural uranium, and with depleted uranium.

The obtained results are compared with calculations performed with WIMS code.

It was verified that the methodology which makes use of the concepts of equivalent plutonium simplifies a lot burn-up and blending ratio calculations for preliminary fuel cycle analysis, giving results with very good approximation. (approximately 5%) and in a very simple way.

1. INTRODUCTION

The introduction of advanced fuel cycles in commercial reactor is seen as a very interesting alternative to improve the utilization of Uranium resources in the middle term, as well as a way to reduce the amount of highly radioactive wastes to be disposed, coming both from front and back end of the cycle.

Among them, the synergetic cycles between different type of reactors, particularly PWR and CANDU /1/ are seen as very promising.

In the frame of the Argentinean-Brazilian cooperation agreement, a neutronic and economical study was done of a Tandem cycle between the Brazilian PWR Angra-I, and the Argentinean CANDU reactor Embalse /2/, /3/.

The basic idea of the TANDEM cycle is to burn in heavy water reactors, especially of CANDU (Canadian-Deuterium-Uranium) type, the remaining fissile material from PWR's spent fuel elements, which still contain approximately 1.5% of ^{235}U and Plutoniums.

After its extraction from the PWR, the fuel is chemically decontaminated, removing the fission products and extracting uranium and plutonium together, without separating them into different streams; this material is afterwards blended with natural or depleted uranium, to be used in the CANDU reactor as mixed oxide fuel, MOX [$(U\text{-}Pu)O_2$].

Argentina is presently operating two nuclear power reactors, both in the heavy-water - natural uranium line. Between them, Embalse was chosen for the study, because CANDU is the most convenient technology to fulfill the requirements for the proposed advanced fuel cycles: its fuel elements are shorter, with lower fabrication costs, easier manipulation, better neutronic behavior and more flexible possibilities for fuel management strategies.

To perform the initial calculations, two methods were mainly used: standard calculation codes, which use discrete ordinates or collision probabilities method to solve the neutronics of the cell, or an algorithm that from now on we will call "EQUIVALENT PLUTONIUM".

A related concept was previously used only in a light-water advanced reactor /4/.

The present work describes the concept in which the algorithm is based, the obtention of the coefficients needed for its determination, and, as an example, the results obtained applying the algorithm to two particular cases of Tandem cycles:

- MOX fuel fabricated from PWR fuel after one year of cooling time, diluted with natural uranium, and with depleted uranium.

The obtained results are compared with calculations performed with WIMS code /5/.

2. "EQUIVALENT PLUTONIUM" CALCULATION

The definition of an "Equivalent Plutonium" factor is based in the fact that, for a given reactor, if the isotopic composition of structural material, coolant and moderator are fixed, the burn-up of the fuel element will be basically dependent of its isotopic composition.

Normally, burn-up is determined using calculation codes which solve the transport equation, by discrete ordinates or collision probabilities, and with the neutronic spectra so obtained, follow the evolution of the different isotopes of the system. It implies a use of a big amount of time and computational resources for each case to be calculated.

If the relationship between burn-up and isotopic composition is unique, it can be interesting to determine if a general curve or analytical expression can be defined, to permit the obtention of an approximate extraction burn-up value for a given reactor, if a modification in the fuel composition is done, for instance if MOX fuel is included, without new and expensive transport calculations.

The possibility to define such an analytical expression relating the isotopic composition of the fuel with reactor parameters like burn-up or multiplication factor is very important, because in this way relevant fuel cycle parameters would be estimated in an approximate and immediate way, without additional calculations.

The "Equivalent Plutonium" can be thought as an equivalent composition, in which the weight percentages of the most important isotopes are multiplied by an importance or worth factor.

To calculate these factors, a series of calculations with WIMS code is performed.

Maintaining the characteristics of the natural uranium cycle, a small percentage of the isotope to be considered in the mixture is introduced, and the system is calculated by a transport code.

The ^{235}U composition is afterwards modified, and the system recalculated, until the obtention of a burn-up value similar to the one obtained before the perturbation.

In this way, it is possible to determine, for each isotope, a worth or relative value, as ratio of numerical densities, referred to ^{235}U.

The relevant isotopes for this case are Uranium 235, Plutoniums, and in the case of a long time between irradiation in PWR and use of the new assembled fuel element, Americium 241, not considered in the present case.

Based in the factors so obtained, it is possible to define, for CANDU reactors, a formula to calculate the Equivalent Plutonium as a function of the fuel isotopic composition.

For our case, the resulting expression is:

$$Pu\ eq = 0.9\ U5 + Pu9 - 0.3\ Pu0 + 1.3\ Pu1 - 0.2\ Pu2$$

where U5, Pu9, Pu0, Pu1 y Pu2 are the different isotopes' contribution to the fuel, expressed in weight percentages, and the importance factors were normalized to 1 for ^{239}Pu.

It should be mentioned that, because of the way in which they were calculated, they maintain a constant burn-up relationship for different isotopic compositions with equal Equivalent Plutonium.

If results of burn-up calculations done with WIMS code in configurations with different fuel composition are plotted against Equivalent Plutonium, a monotonously growing tendency can be observed, as can be seen in Figure 1.

Results of calculated burn-up values and its Equivalent Plutonium factors for the cases of natural uranium, LEU, four different MOX compositions and Tandem options are included, as well as some data taken from Reference /1/, together with the best fitting curve.

The points referenced as "integral criteria" or "2*Medium BU" correspond to the same WIMS calculation, but different criteria for the determination of the extraction burn-up (Reference /2/).

Making a statistical regression of these data, an analytical expression can be obtained for burn-up calculation from equivalent plutonium factors.

For our case, the points can be fitted by an exponential curve, with a correlation factor of 0.999

$$Pu\ eq = 0.488247 \exp.\ (3.69424E\text{-}05 * Q)$$

where Q is the burn-up value expressed in MWd/ton and Pu eq are the factors calculated from the isotopic compositions.

This analytical expression permits the estimation of the extraction burn-up value obtainable from a fuel, knowing only the isotopic composition of the initial MOX, in a very simple and immediate way.

This result is obviously particular for CANDU reactors, and will be valid in a restricted burn-up range, because it depends of the framework in which the perturbations to the fuel were done, the isotopes that are present , and from the criteria adopted for the extraction burn-up calculation.

Regarding the accuracy of the method, for the cases of the present work, which as was previously said include natural uranium fuel, LEU and different MOX compositions, the maximum deviation from the predictions obtained by equivalent plutonium with respect to values from transport calculations, was 11%.

Particularly, for the range of burn-ups obtainable in advanced fuel cycles in CANDU, an error of 5% is predictable when it is calculated through the Equivalent Plutonium formulation, which is a very good approximation for initial or parametric calculations.

The maximum difference obtained with the values of reference /1/ is 2%.

Regarding the curve, it should be noted that, as it has a growing exponential behavior, it reaches a saturation burn-up value with the fissile material composition, result that agrees with the physical characteristics of the problem.

3. EQUIVALENT PEAK FACTOR

To consider in an estimate way the limitations imposed by the fuel channel when an element with different neutronic characteristics is included, the effective multiplication factor is a good parameter to evaluate.

Through a procedure similar to the one explained in the previous section to obtain the importance factors for each isotope for Equivalent Plutonium calculations, worth can be defined for each isotope for fresh fuel multiplication factor calculation.

Now, the magnitude to be kept invariable in the perturbation transport calculations is the k-effective instead of the burn-up.

Defining the weighting factors in such a way, the expression obtained for what we called equivalent peak factor is:

$$EPF = 0.5\ U5 + Pu9 - 0.75\ Pu0 + 1.47\ Pu1 - 0.07\ Pu2$$

Plotting this equivalent peak factor (EPF) against the effective multiplication factor of the fresh fuel (keff(0)) calculated with WIMS, again a monotonously growing curve is obtained, that can be fitted with an exponential curve with a correlation factor of 0.989, as can be seen in figure 2.

The analytical expression obtained is:

$$EPF = 1.28861E\text{-}02 \exp.\ (\ 3.0577 * keff(0)\ )$$

It should be noted that the sensitivity to the changes in the initial composition would be greater in this case, because it is not an integral magnitude, as burn-up is.

Even though, it is an interesting tool to evaluate limits in fissile material in the fuel, based in a limiting fresh fuel multiplication factor criterion.

4. APPLICATION TO TANDEM CYCLE CALCULATION

Typical time-consuming cases in this kind of studies are the parametric analysis of the problems, often done to find an optimum point in several variables, to make afterwards final and more refined calculations in that point.

The expressions obtained for equivalent plutonium and equivalent peak factor presented in this paper, are a very interesting tool to perform these parametric calculations for the introduction of advanced fuel cycles in CANDU reactors, because they give approximate values in an immediate and simple way.

As examples of applications of the algorithm, partial results of a more complete study on a Tandem fuel cycle between Angra-I and Embalse reactors will be presented here.

They were published in a complete way in reference /3/.

Figure 3 shows a comparison between burn-ups obtained with equivalent plutonium formula and with WIMS calculations, for different compositions of MOX fuel, obtained varying the percentage of natural uranium used in the dilution.

As it can be seen there, the agreement is very good. The maximum differences obtained in this case are of 8%.

As it was previously said, in a synergistic fuel cycle like TANDEM, the fissile material coming from the PWR burnt fuel should be diluted to fabricate the MOX.

For each dilution material, the optimal blending should be determined, taking first into account which of the cycle parameters will be optimized.

In the work, the initial goal was to optimize the utilization of the uranium resources of the binational system.

Figure 4 shows an example of the consumption of uranium for the binational system normalized to 1 GWe-year (where it has been assumed that both reactors have the same capacity factor), as a function of the initial multiplication factor chosen for the CANDU.

All the values of Burn-Up and fresh fuel multiplication factors for the different blendings were calculated through Equivalent Plutonium and Equivalent Peak Factors algorithms.

The minimum value of the bi-national uranium utilization curve indicates an optimum initial multiplication factor of 1.32-1.33, which agrees with the value reported in the literature for similar cases.

Therefore, using this value of Keff(0), the blending ratio for one year of PWR's fuel cooling time and natural uranium as dilution material should be 0.38 (38% of natural uranium)

With this blending ratio, the composition of the fuel and its correspondent Equivalent Plutonium can be calculated, as well as the obtainable extraction burn-up, through the analytical expression shown in section 2.

The value obtained for the CANDU is 22,525 MWd/ton U, which shows a very important increase (of about 3 times) over the actual CANDU extraction burn-up with natural uranium (7,500 MWd/ton U).

The values obtained calculating with WIMS code the composition with 38% of natural uranium are keff(0)=1.31032 and burn-up 22500MWd/ton.

It should be noted that the multiplication factor calculated by WIMS has a difference of approximately 1800 pcm (1.5 %) with the value obtained with the methodology of Equivalent Parameters, while in burn-ups the results are almost equal.

These differences are very low and completely acceptable considering the simplicity of the method; this result confirms the adequacy of the method of Equivalent Plutonium for this case, if the purpose is the estimation of the main fuel cycle parameters.

If depleted uranium coming from tail from enrichment plant is used as blending material instead of natural uranium, a similar study can be done (with the Equivalent Plutonium methodology).

Typical enrichments of these tails are between 0.2 and 0.3% in U-235. Between those limits, and with blendings up to 40%, the multiplication factor at the beginning of the cycle and burn-up is not sensitive to the initial enrichment of the tail, and the percentage that minimizes the Uranium consumption is 26% of material coming from the tail.

With these values, the obtainable keff(0) is 1.33, and the extraction burn-up is 22000 MWd/ton

Calculating with WIMS, the keff(0) is 1.3112 and the extraction burn-up 21000 MWd/ton.

4. CONCLUSIONS

Analytical expressions were found to predict burn-up and fresh fuel multiplication factor of different kind of advanced fuels potentially useful for CANDU reactors, starting only from its isotopic composition.

It was verified that the methodology, which makes use of the concepts of equivalent plutonium and equivalent peak factor, greatly simplifies burn-up and blending ratio calculations for preliminary fuel cycle analysis, giving results with very good accuracy (approximately 5%) and in a very simple way.

Based in those results, it is possible to assert that these analytical formulas permit a simple and estimate analysis of advanced fuel cycles to be applied to CANDU reactors, because the variables needed to optimize the cycle, (masses, costs and resources use), can be obtained in a very direct and simple way, with very reasonable approximation.

The accuracy and simplicity of the method, make it highly recommendable for its application to preliminary calculations.

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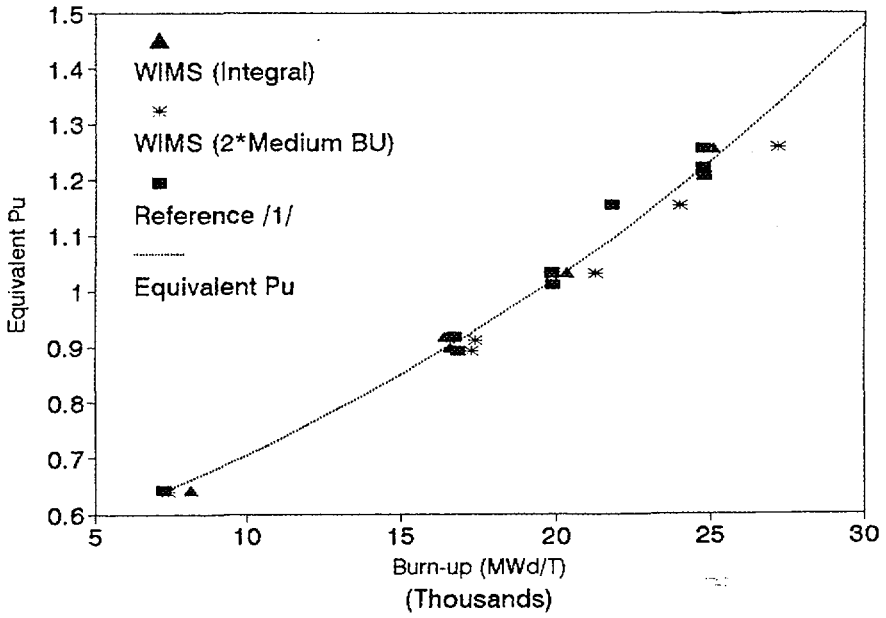

Figure 1
EQUIVALENT PLUTONIUM VS BURN-UP

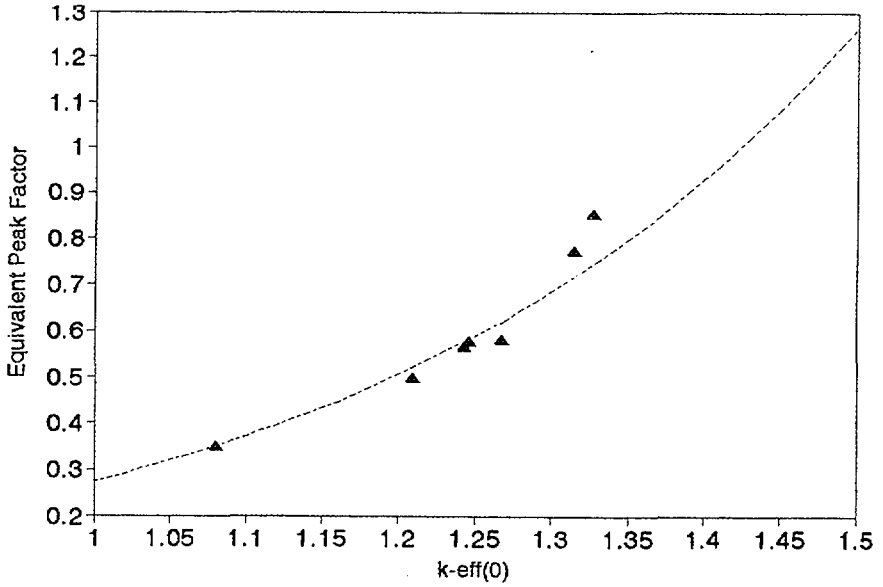

Figure 2
EQUIVALENT PEAK FACTOR VS K-EFFECTIVE

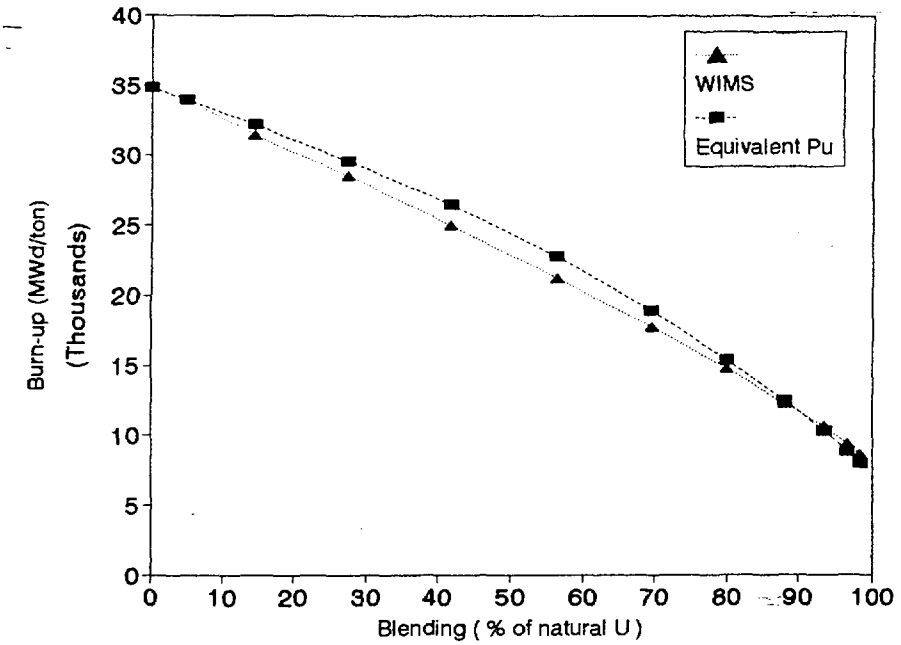

Figure 3
BURN-UP VS. BLENDING RATIO

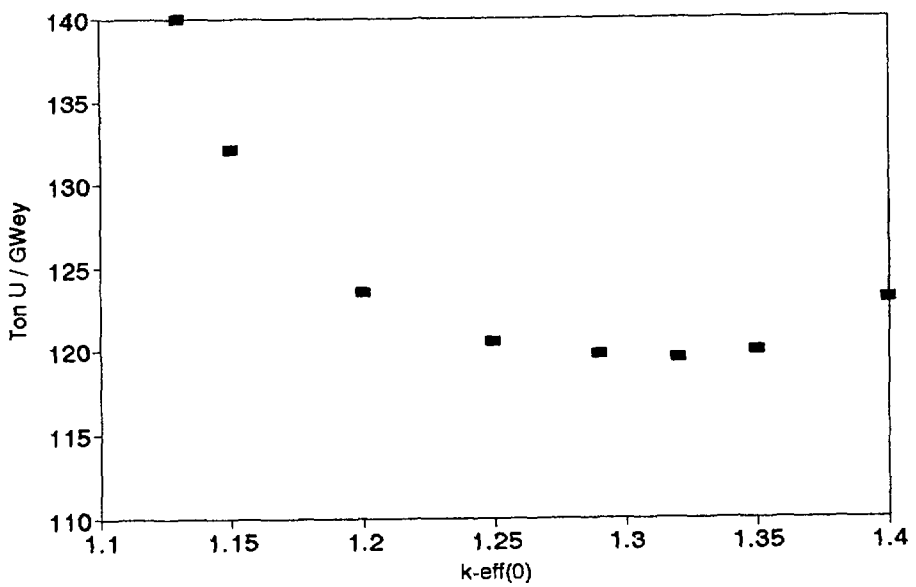

Figure 4
U BINATIONAL CONSUMPTION VS. KEFF(0)