A STUDY OF THE NUCLEAR NATURAL RESOURCES UTILIZATION IN OPEN AND CLOSED FUEL CYCLES

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DOI: https://doi.org/10.47168/rbe.v25i2.416

RESUMO

Neste trabalho, o uso de recursos naturais foi analisado usando uma metodologia simplificada e assumindo condições de cálculo próximas às reais, a fim de avaliar a sustentabilidade da fonte nuclear e a eficiência no uso desses recursos. Para a análise de ciclos de combustível aberto foram selecionados quatro reatores, sendo os Reator de Água Pressurizada (PWR) e Reator de Água Pesada Pressurizada (PHWR), dois reatores de Geração II comumente usados até hoje, o reator avançado da Geração III AP1000 e o reator conceitual AP-Th 1000. Para ciclos fechados de combustível, avaliouse a variação da utilização do recurso natural com a variação do fator de conversão, parametrizado pela queima. Observou-se que os reatores de Geração II utilizam apenas 1% do recurso natural e, apesar dos avanços tecnológicos, o reator da Geração III não apresentou aumento significativo em comparação ao primeiro. O ciclo fechado de combustível, apesar de reciclar o combustível queimado de reatores térmicos, explora apenas cerca de 10% do recurso. Grandes melhorias são observadas nos Fast Breeder Reactors, podendo obter um uso próximo a 100% com o aumento da queima e a minimização de perdas. Embora tenha sido comprovada a viabilidade técnica do uso do tório como combustível nuclear, este seria mais bem usado em ciclo fechado, como no autossustentável Liquid Fluoride Thorium Reactor (LFTR), um reator de Geração IV que pode transformar a energia nuclear em uma fonte de energia sustentável e renovável.

Palavras-chave: Utilização de Recursos Naturais, Reatores Nucleares, Ciclos de Combustível, Tório.

ABSTRACT

In this work, the use of natural resources was analyzed using a simplified methodology and assuming calculation conditions close to the real ones, to assess the sustainability of the nuclear source and the efficiency in the use of these resources. For the analysis of open fuel cycles, four reactors were selected, these being the Pressurized Water Reactor (PWR) and Pressurized Heavy Water Reactor (PHWR), two Generation II reactors commonly used until today, the advanced Generation III reactor AP1000 and the conceptual reactor AP-Th 1000. For closed fuel cycles, the variation of the utilization of the natural resource alongside with the variation of the conversion factor were evaluated, parameterized by the burnup. It was observed that the Generation II reactors use only 1% of the natural resources and, despite technological advances, the Generation III reactor did not show a significant increase in comparison to the former. Although the closed fuel cycle includes recycling the burnt fuel from thermal reactors, it exploits only about 10% of the resources. Major improvements are observed in Fast Breeder Reactors, being able to obtain a use of almost 100% with the increase of the burning and the minimization of losses. Although the feasibility of using thorium as a nuclear fuel has been proven, it would be better used in a closed cycle, as in the self-sustainable Liquid Fluoride Thorium Reactor (LFTR), a Generation IV reactor that can transform the nuclear energy in a sustainable and renewable source of energy.

Keywords: Natural Resources Utilization, Nuclear Reactors, Fuel Cycle, Thorium.

1. INTRODUÇÃO: A REVIEW OF NUCLEAR REACTORS TOWARD A SUSTAINABLE AND RENEWABLE SOURCE OF ENERGY

Most of the commercial nuclear reactors in operation in the world are Generation II reactors, which were built in the 20th century. They use uranium as a primary source of energy and operate in a once-through cycle (OTC). The main representatives of this generation are the Pressurized Water Reactor (PWR), with a share of almost 70% of nuclear power plants in commercial operation, and the Boiling Water Reactor (BWR) and Pressurized Heavy Water Reactor (PHWR), with 15 and 11% participation, respectively (WNA, 2020). The extraction burnup (B), defined as the energy generated in an equilibrium cycle by the input mass of uranium in the reactor, is approximately 30 MWD/kg U for PWR and BWR reactors, and approximately 7 MWD/kg U for PHWR reactors, which uses natural uranium as input feed.

Figure 1 illustrates a typical mass balance for a PWR, with 1000 MWe and B=30 MWD/kg U, and an average enrichment of 3% in mass of the input UO2, as calculated by the code IAEA VISTA code (IAEA, 2007). We notice that in this reactor the spent fuel is stored in interim storage - usually a pool adjacent to the reactor building - until sent to a final storage (deep repository).



Figure 1 - Mass balance of a typical Generation II PWR (1000 MWe) operating in an OTC (Maiorino, D'Auria and Akbari-Jeyhouni, 2018)

Given the loss of competitiveness of the nuclear industry, the public opinion against nuclear generation, and safety issues, since the beginning of the century the industry has launched new innovative designs to be competitive and safety improvement. These reactors, denominated as Generation III, are already in an advanced stage of projects, many of them in construction and operation, as AP1000, that soon will replace the previous generation (World Nuclear Association, 2017a).

These are large reactors designed to operate with powers in the range of 1000 MWe, although, more recently, compact reactors called Small Modular Reactors (SMR) are also being developed, built to generate up to 300 MWe (IAEA, 2016). Even though they still use the same type of fuel, i.e., UO2, and the main characteristics remain almost the same as the reactors of the previous generation, Generation III has advanced improvements related to safety, economy and operational performance, such as:

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- Design standardization to expedite licensing, diminishing construction time and implying in reducing the capital cost (economics criteria);

- Simplified design that reduce the operational faults;

- Greater availability increases the time between refueling and, also, the plant lifetime (60 years);

- Minimization of the possibility of core meltdown;

- Passive emergency coolant system;

- Higher burnup rates (60 MWD/kg U) and reduction of waste production;

- Utilization of advanced fixed burnup poison to increase the fuel lifetime.

In addition to the improvements mentioned, there is a small advance in the natural resources utilization by a factor of approximately 2 (IAEA, 2007), as illustrated in the mass balance of Figure 2 for the thermal reactor Advanced PWR (1000 MWe), with a burn up of 60 MWD/kg U and an average enrichment of approximately 4% in mass of ²³⁵U. The operation in an OTC was calculated by VISTA.



Figure 2 - Mass balance of a Generation III 1000 MWe PWR operating in an OTC (Maiorino, D'Auria and Akbari-Jeyhouni, 2018) Several countries are working on the reprocessing of spent fuel from thermal reactors aiming at the extraction of uranium and plutonium for MOX (mixed oxide fuel) production in a closed fuel cycle, thus increasing the utilization of the energy content in the natural resources.

Depending on the conversion ratio (*C*), which is defined as the ratio of the mass of fissile nuclides at the end of cycle (EOC) by the mass of fissile nuclides at the beginning of cycle (BOC), the utilization of natural resources could achieve meaningful results. For thermal reactors, the conversion ratio is less than 1 (*C*<1), but for a Fast Breeder Reactor (FBR) the conversion ratio is greater than 1 (*C*>1), i.e., produces more fissile material over time than it consumes and, therefore, turning, in principle, nuclear energy independent of natural resources (renewable). Although the technical feasibility of the FBR has already been demonstrated, it is not yet economically competitive, and it is only now as the Generation IV reactors are being considered (World Nuclear Association, 2017b).

Another option is to utilize thorium as a primary source of energy. Although not fissile, upon thermal neutron interaction ²³²Th produces ²³³U, which is one of the best fissile nuclides (number of neutrons produced per neutron absorbed). Its use aims at the reduction of high-level waste (minor actinides), improving the nuclear power sustainability and fuel utilization. Also, thorium is three times more abundant than uranium in nature and the fuel (U-Th)O₂ (mixed thorium-uranium oxide) presents better thermal and physical properties than traditional UO₂ (Ashley et al., 2014; Lindley et al., 2014; Baldova and Shwageraus, 2016).

Several Th/U fuel cycles using thermal and fast reactors were proposed and are still under investigation (OECD, 2015). The technical feasibility to use thorium was proven in PWR Indian Point Reactor, utilizing a core load with (Th0-0.9/U0.1)O₂, with highly enriched U (93w/o) and achieving a maximum burnup of 32 MWD/kg HM. Recently, a study to convert the Advanced PWR AP1000 for (U-Th)O₂ fuel use was performed, called AP-Th 1000. Although the feasibility of the concept was proved, its utilization in an OTC is not attractive, mainly due to the need for use of ²³⁵U enriched to 20%. However, by optimizing the production of ²³⁵U and conversion ratio, it could operate in a closed fuel cycle as a first step towards using thorium in nuclear reactors (Maiorino, Stefani and D'Auria, 2017).

Despite efforts, controversial issues related to high-level waste solutions (HLW) and the safety of nuclear facilities undermine the political and public acceptance of nuclear energy (Maiorino and Moreira, 2014). With the purpose of making nuclear energy sustainable, a longterm deployment of innovative reactors is underway. These reactors and their associated fuel cycles are old concepts but incorporate modern technological improvements, to become the next generation of nuclear reactors.

The Generation IV International Forum selected six reactor technologies for further research and development: Very High-Temperature Reactors, Gas, Sodium or Lead Fast Reactors, Molten Salt Reactors, and Supercritical Water Reactor, designed for to be efficient, insurance, burners or breeders and, in some cases, HLW burners (World Nuclear Association, 2017c). Among the concepts of Generation IV reactor, the most promising is the LFTR (Liquid Fluoride Thorium Reactor), a self-sustainable Molten Salt Reactor with a promise of bringing sustainability to fission-generated nuclear energy. The utilization of the natural resources is of 100% since it recycles the fissile ²³³U online, with zero losses in the process (Sorensen and Dorius, 2011; Serp et al., 2014; Aniza et al., 2015), as illustrated in Figure 3.

This paper, after presenting a brief review of the evolution of nuclear reactors towards sustainability, will present in the next sessions a simplified analysis of the utilization of natural resources (session 2), results (session 3), and conclusions (session 4) on the possibility of maki



How does a fluoride reactor use thorium?

Figure 3 - Schematic view of the LFTR - Liquid Fluoride Thorium MSR (Sorensen and Dorius, 2011)

2. METHODOLOGY

To compare different reactors and their fuel cycle, an important parameter is the efficiency by which they use the energy content in the primary source (uranium or thorium) to generate a secondary source (heat or electricity) for the final use. As demonstrated in Lamarsh and Baratta (2001), the utilization of the natural resource, *U*, is defined quantitatively as the ratio of the amount of fuel that fissions in a given nuclear system to the amount of natural uranium or thorium input required to provide those fissions - that is

$$U = \frac{\text{Mass of fissile fuel burned}}{\text{Input mass of the primary source}} = \frac{F}{M_a}$$
(1)

In open cycles, as those illustrated in Figures 1 and 2, the parameter "burnup", B, gives the energy generated by the fissile material, usually expressed in megawatts day (MWD), by the input mass of enriched uranium (Me) in the reactor. Given the fact that 1 MWD is approximately 1g of the material burned (Lamarsh and Baratta, 2001), the specific burnup (b) may be defined as the fraction of the input feed mass which is fissionable. Therefore, the mass of fissile fuel burned (F), can be calculated by:

$$F = bM_e \tag{2}$$

To calculate the input feed mass, the loss in the conversion process from the front of the fuel cycle is neglect and considers that the only process which modifies significantly the input mass of natural primary resource is the isotopic enrichment process. In this process, to obtain the mass of enriched uranium at the reactor inlet, with a specified enrichment (x_e), is necessary an input of mass of natural resource given by:

$$M_{a} = (\frac{x_{e} - x_{d}}{x_{a} - x_{d}})M_{e} = \xi M_{e}$$
(3)

where x_e , x_d and x_a are the isotopic enrichment of the input fissile, depleted, and natural material, respectively. As assumed in (Lamarsh and Baratta, 2001), in the case of natural uranium, x_a =0.0071, and the depleted enrichment depends on the enrichment process, assuming typical values of 0.002-0.003. Thus, the natural resource utilization can easily be calculated for:

$$U = \frac{b}{\xi} \tag{4}$$

For closed fuel cycles using thermal reactors, in which the conversion ratio is less than 1 (C<1), the calculation of the utilization of the natural resource involves a mass balance in a closed cycle, between fissile and fertile material, and could be given by:

$$U = \frac{b}{\xi [b(1+\alpha)(1-\beta)(1-\gamma)(1-C) + \gamma x_e]}$$
(5)

where γ is the fraction of losses in reprocessing and fabrication process, α is a factor which depends on the ratio capture to fission, and β is the fraction of fissions occurring in fissile nuclide originated from the fertile material (²³⁹Pu in U-Pu cycles or ²³³U in U-Th cycles).

Assuming $(1+\alpha)$ $(1-\beta)\sim 1$, i.e., independent of the closed fuel cycle, U-Pu or U-Th, the utilization of the natural resource could be easily calculated knowing the burnup, conversion factor, the enrichment factor of the input uranium, and the losses in the reprocessing and fabrication process.

In closed fuel cycles for Fast Breeder Reactors using U-Pu or U-Th cycles, where C>1, the utilization of natural resources could be calculated by:

$$U = \frac{b}{b(1-\gamma) + \gamma - (\xi - 1)[b(1-\gamma)(C-1) + \gamma x_e]}$$
(6)

In the case of a self-sustaining cycle, which depends on the regeneration gain, G=C-1, or regardless of the mass of incoming fissile material, $\xi=1$, the use of the natural resource will depend only on burnup and losses, thus:

In closed fuel cycles for Fast Breeder Reactors using U-Pu or U-Th cycles, where C>1, the utilization of natural resources could be calculated by:

$$U = \frac{b}{b(1-\gamma)+\gamma} \tag{7}$$

So, neglecting the losses, with $\gamma=0$, the utilization of the natural resource would be 100%, as is the case of the self-sustainable Molten Salt Reactor LFTR.

In order to carry out a study to quantitatively assess the use of the natural resource in open cycles, four reactors were considered: the 2nd generation reactors, HWR and PWR, the AP1000 3rd generation reactor, and the AP-Th 1000 conceptual reactor that, as previously described, is an Advanced PWR using $U_{0.25}Th_{0.75}$. The enrichment and burnup values used for the selected reactors are shown in Table 1.

Reactor	Abbreviation	Generation	Enrichment [w/o]	Burnup [MWD/kg U]	Source
Heavy Water Reactor	HWR	2	0.0071 (Natural)	7.5	Maiorino, D'Auria and Akbari-Jeyhou- ni, 2018
Pressurized Water Reactor	PWR	2	0.03	30	Maiorino, D'Auria and Akbari-Jeyhou- ni, 2018
Advanced PWR - AP1000	AP 1000	3	0.034	62	Westinghouse, 2011
Advanced PWR using U0,25Th0,75 - AP-Th 1000	AP-Th 1000	Conceptual	0.2	65	Maiorino et al, 2017

Table 1 - Enrichment and burnup values for the selected reactors

For closed fuel cycles, instead of analyzing a specific type of reactor, it was preferred to evaluate the variation of the utilization of the natural resource (U) with the variation of the conversion factor (C) parameterized by the burnup (B).

3. RESULTS AND DISCUSSION

The results for the utilization of natural resources for the selected reactors operating in an OTC, using the methodology previously described and the data in Table 1, are illustrated in Table 2.

Table 2 -Results for the utilization of natural resources (U) for the selected reactors operating in OTC

Reactor	Generation	Utilization of Natural Resource (U)
HWR	2	0.0075
PWR	2	0.0055
AP 1000	3	0.0105
AP-Th 1000 Conceptual		0.0072

Comparing the results obtained between both Generation II reactors, it is observed that the HWR, despite its small burnup, had a better utilization of natural resources compared with PWR. This fact occurs, mainly, because it uses natural uranium as nuclear fuel, and does not need enrichment like PWR. In the AP1000 reactors, although there is an improvement in the utilization of natural resources about 1%, it is not yet significant. For the AP-Th 1000 conceptual reactor, the utilization of natural resources is of the same order of the HWR, and thus not very attractive for operation in an OTC. However, as discussed by reference (Maiorino et al, 2017), since this concept optimizes the production of ²³³U, it is of interest to operate in a closed fuel cycle of U-Th.

Figure 4 features the variation of the utilization of the natural resources with the variation of the conversion factors for different values of burnup, assuming losses of 2%, and Figure 5 for losses of 1%. From these results, it is observed that even for closed fuel cycles, using thermal reactors with the recycling of plutonium and which the conversion factor is close to 1, as those already used commercially, the utilization of natural resources assumes values close of 10% Despite a big improvement in the utilization factor, taking advantage of about 10 times more compared to open cycles, the results are still very low. For Fast Breeders Reactors, expected to operate commercially by this mid-century, depending on the burnup, could achieve values close to 1. For typical flare values of 60 MWD/kg HM, the use of natural resources, even considering high losses (1-2%), will reach values close to 75-80%. For the advanced Generation IV, such as LFTR, in which reprocessing online may have practically zero losses, resource utilization could reach values of 100% (U=1).



Figure 4 - Natural resources utilization (%) versus Conversion factor parameterized by the burnup for 2% of losses



Figure 5 - Natural resources utilization (%) versus Conversion factor parameterized by the burnup for 1% of losses

4. CONCLUSIONS

The evolution of nuclear reactors shows that, with each generation, improvements were developed with the purpose of meliorating safety and efficiency and reducing waste generated during burning. Currently, Generation II thermal reactors, operating in open cycles, have only 1% use of the energy content of the primary source (uranium or thorium). By recycling the spent fuel, burned in the thermal reactors, its utilization in the same type of nuclear reactors has an increase of about 10%. This result shows the importance of taking advantage of an energy resource that would be wasted.

With the near-term deployment of Fast Breeder Reactors, an increase of the burnup and improvements in the recycling processes to minimize the losses, the utilization of natural resources is going to be very close to 100%. For the future, the Generation IV reactors, mainly the Liquid Fluoride Thorium Reactor, which economically enables the use of thorium as a nuclear fuel and allow online reprocessing where losses are minimized, turning the nuclear energy in a sustainable and renewable source of energy.

5. ACKNOWLEDGMENTS

Ms. Leticia C. Gonçalves thanks for the scholarship provided by UFABC to realize this work.

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