Reactor Simulations for Safeguards with the MCNP Utility for Reactor Evolution Code

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Abstract. To tackle nuclear material proliferation, we conducted several proliferation scenarios using the MURE (MCNP Utility for Reactor Evolution) code. The MURE code, developed by CNRS laboratories, is a precision, open-source code written in C++ that automates the preparation and computation of successive MCNP (Monte Carlo N-Particle) calculations and solves the Bateman equations in between, for burnup or thermal hydraulics purposes. Using MURE, we have developed a cell simulation of a typical CANDU reactor, which could be used to analyze the composition of fuel assemblies as a function of time or burnup. Diversion cases of a Pebble Bed Reactor loaded with UOx, PuOx and ThUOx fuels, and a Na-cooled Fast Breeder Reactor (FBR) with depleted Uranium or Minor Actinides in the blanket. The loading of Protected Plutonium Production (P^3) in the FBR was also simulated. The simulations of various reactor designs taking into account reactor physics constraints may bring valuable information to IAEA inspectors.

1. Introduction

It is necessary for the IAEA to study how much nuclear materials are consumed and generated in nuclear power plants. It is mandatory to study on proliferation scenarios for the IAEA inspectors to know:

- What kind of reactors do malicious actors seek?
- How many kilograms of plutonium are generated?
- How much time do they need to achieve it?
- What is the practical refueling plan?

To answer these questions above, we need a reactor simulation code coupled with several modules to test proliferation scenarios. It is also necessary that such code be customized according to the needs of users. In the following, we will first present the MCNP Utility for Reactor Evolution code that we have used to developed reactor core modeling. Then we will present selected examples of reactor core simulations performed for several reactor designs and fuels in the context of the development of the antineutrino probe, and we will attempt to show how these simulations could bring valuable information in themselves in order to help IAEA to meet its surveillance goal. We will then conclude and give possible perspectives.

2. MURE code and its capability

The MURE (MCNP Utility for Reactor Evolution) code [1], developed by CNRS laboratories, is a precision, open-source code written in C++ that automates the preparation and computation of successive MCNP (Monte Carlo N-Particle) [2] calculations and solves the Bateman equations in between, for burnup or thermal hydraulics purposes. It allows computing the time-evolution for various types of nuclear reactors and simplifies the elaboration of the input file containing the reactor configuration and fuel composition in MCNP. Figure 1 shows the evolution principle of MURE code [3].

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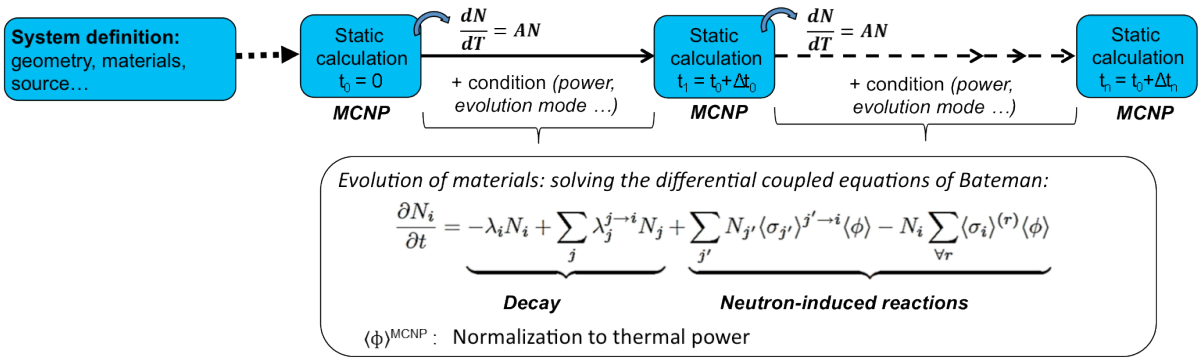

FIG. 1. MURE code principle [1,3].

A post treatment tool MureGui [3], with a C++ Graphical User Interface based on the ROOT graphical tools developed at CERN, is also available to read the results of a MURE evolution. Nuclei are sorted in different categories such as Actinides, Fission Products, Gas and users can plot the following quantities: inventory, microscopic /macroscopic cross sections and rates for the selected reactions, k-effective (multiplication coefficient), flux and breeding ratio. Each quantity can be plotted for a chosen nucleus or for a sum of selected nuclei, also with a spatial dependence if needed, according to time or burn-up steps defined for the evolution calculations.

Using the CHARS (CHAracterization of Radioactive Sources) package in addition [4], the MureGui interface computes gamma, alpha, beta, and neutron energy spectra of any spent fuel after irradiation, as shown in Fig 2. Decay calculations can be performed after a MURE evolution or directly from a user input composition without a prior evolution. Activities, total and partial radiotoxocities, heat released as a function of time can be computed for a given system. This includes the cooling phase in core and the before/after reprocessing phases out of the core. For example, in collaboration with an industrial partner last year and to study material damages induced by gammas, a container of intermediate level long lived wastes was simulated and CHARS used to compute the gamma energy spectrum and activity as a function of the cooling time. The CHARS package could be then used to better characterize a spent fuel radiation, according to the IAEA needs.

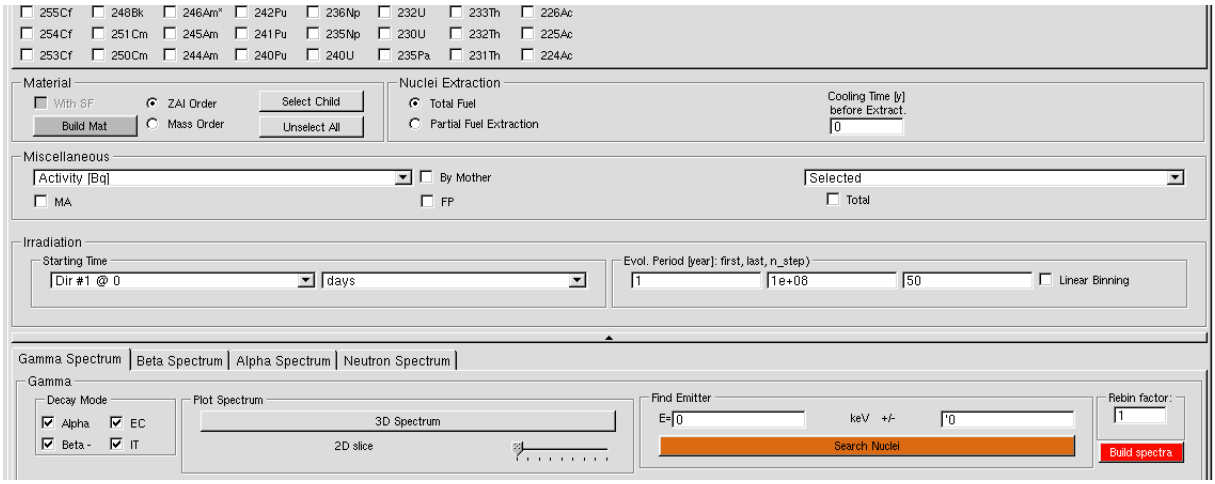

FIG. 2. View of the CHARS module included in the MureGui interface [4].

3. Selected Reactor Core Studies

3.1. Pressurized Water Reactor core model and benchmarks

Two 4.25 GWth Pressurized Water Reactor (PWR) cores from the Chooz power plant in France have been simulated with the MURE code, based on the loading maps given by the EDF company [5,6], and a complete study of the sensitivity to most important reactor parameters was able to be performed. The quality of our simulations has been evaluated beforehand through various benchmarks, as the Takahama benchmark [7] and a benchmark of the MURE code and of the DRAGON [8] code with results from the APOLLO [9] code provided by EDF on assembly simulations. In this way, important input parameters of the codes for fission rate calculation could remain under control.
The simulations have been performed in the conditions of the Double Chooz reactor antineutrino experiment i.e. with a follow-up of the thermal power and the boron concentration along the operation of the cores. These parameters are retrieved from the EDF database. The design of the various assembly types loaded in the cores was given by EDF as well as material compositions, temperature and density information. Each year, around one third of burnt assemblies are discharged and replaced by fresh fuel assemblies. The other two thirds are shuffled in order to ensure a homogeneous neutron flux in the core. A new core simulation was developed after each core refueling.

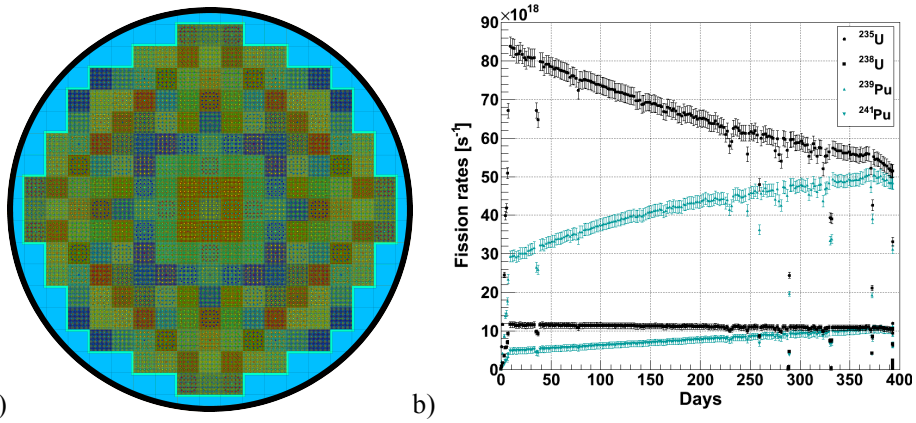

FIG. 3. a) The simulated PWR core model [6]. b) Fission rates of a PWR from Chooz [6], along with their errors, computed with MURE.

The assemblies have been grouped as a function of the following criteria: assembly type defined by its geometry, initial fuel enrichment and Gd proportion, same position in the core under a 1/4th or 1/8th rotation, closest initial burnup. This classification gave rise to 50 groups containing each from 1 to 8 assemblies and was the result of a study to obtain the best compromise between a realistic CPU time without affecting the target accuracy. The fuel composition of the assemblies loaded in the core at start of cycle was computed using single assembly simulations of each type. The resulting geometry of the modeled core is displayed in Fig. 3 a), while obtained fission rates with their error bars are displayed in Fig. 3b).
This work represents an important step because it is the first academic example of detailed simulation of a reactor following the core history at a realistic scale. This shows that it could be possible to perform such simulations in order to confront hypothetical diversion scenarios with reactor safety aspects (criticality, reactivity, delayed neutron fraction…), or to deduce fuel compositions after various core operations, if proven to be useful to the IAEA. This has been performed in the case of a PWR but could be contemplated for other designs. In the following paragraphs, we present examples of simpler models of Generation IV reactors with which we attempted to elaborate diversion scenarios that we found to be limited by reactor safety constraints.

3.2. On-load reactor examples: CANDU and Pebble Bed Reactor Simulation

We have studied a diversion scenario for several on-load reactor designs: CANDU reactors [10] and Pebble Bed Reactors (PBR) [11].
A simplified model of CANDU-600 reactor has been elaborated with MURE. We have simulated one single channel surrounded by a mirror in order to simplify the calculations, with various refuelling periods by third of the bundles. The studied scenarios consist in a change of the loading plan of the core, with a part of the channels refuelled faster w.r.t the normal refueling period, and another part refueled less often in order to maintain the number of movements per day as constant. Reactor physics is used to deduce the refuelling period corresponding to a standard (hereafter "legitimate") operation of the core. A core would be the super-imposition of 400 channels and refuelled every 200 days in average, with 2 channels refueled per day. Then the principle of a diversion scenario is the following: refuel part of the core to extract higher quality plutonium and in parallel refuel less often the remaining part of the core so as to still have only 2 channels refueled per day, while maintaining the reactivity constraints in-core. In a first scenario 1/4th of the reactor are discharged every 100 days, and 3/4th every 300 days. With such a scenario, the plutonium produced would be fuel-grade [10, 12]. In case of a refuelling plan that would contain 1/8th of the channels discharged every 50 days and 7/8, every 350 days, the plutonium produced would be almost weapon-grade (>90% in ^{239}Pu) [10, 12]. Of course these scenarios are not realistic, but following the same principle, a detailed complete core model of a CANDU reactor such as the ones used by the Canadian electricity company would help the IAEA to identify the diversion scenarios allowed by the reactor physics (and thus safety) constraints.
Pebble Bed Reactors (PBR) are a design of very high temperature reactors with an on-load refueling mode contemplated in the frame of the Generation IV forum. We have simulated a single pebble surrounded by a mirror, thus simulating an infinite reactor, with a packing fraction of 51%. The first simulation was confronted to a benchmark in order to validate the reactor physics. The reactor of the benchmark was of 200 MWth power, filled with UOx pebbles, PuOx pebbles or ThUOx pebbles. Our comparison to the benchmark shows that our results are within the predictions of the participants to the benchmark. Using the simulation, we could determine the quality of the Pu isotopic vector of the pebbles as a function of burnup, and the normal residency time of the pebbles in-core under normal operation. From that we can deduce the number of pebbles having the same burnup in-core. A scenario of diversion for this reactor could consist in the withdrawal of pebbles of a given burnup replaced by a mix of new pebbles and pebbles already at the maximum burnup. But the reactivity variation induced in the core should be compatible with the efficiency of the control rods that should compensate it, for safety aspects. The reactor physics shows that the a diversion of almost weapon-grade plutonium [11, 12] would only be possible if the operator could arbitrarily add more fresh pebbles than expected in a "legitimate" scenario in the core. Such calculations could be used to determine with which accuracy margin the number of pebble movements should be controlled to prevent such diversion.

3.3. Sodium Fast Breeder simulation, loaded with fuels with high proliferation resistance

3.3.1. Proliferation resistance and protected plutonium production

Saito proposed the Protected Plutonium Production (P^3) concept [14-16], which aims to produce high-proliferation-resistant Pu by increasing ^{238}Pu isotopic composition through transmutation of minor actinides (MAs), such as Np and Am. Plutonium-238 has a high decay heat and spontaneous fission neutron rate that prevents Pu from being used in nuclear

explosive devices. Several criteria with respect to the indicators for the proliferation resistance of Pu have been proposed so far. Kimura et al. suggested that Pu more than 15% of ^{238}Pu is regarded as Pu with a high proliferation resistance in the light of decay heat [17].

3.3.2. Methodology

We have adopted an Indian Prototype Fast Breeder Reactor as a reference [11,13, 18]. Pu content in inner core was 21 wt.% and in outer core 28 wt.%, respectively. Core configuration is shown in Fig. 4. Reloading was planned once every 180 EFPDs, when 1/3 of the core and 1/8 of the radial blanket were exchanged. The thermal power is 1.25GWth. In the MURE code we can choose several nuclear databases to use with a priority order; we used JEFF 3.1. as the primary nuclear database.

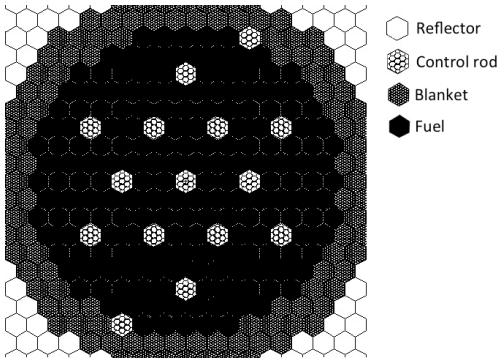

FIG. 4. Core configuration (X-Y)

The loaded nuclear fuels are a normal MOX fuels and P^3 fuels. First of all, a spent fuel of 3.5% ^{235}U-enriched UOx fuel burnt in PWR until 40GWd/tHM and a 5 years' cooling time were assumed. Then Pu from this spent fuel was used for the normal MOX fuel (Case 1). As for P^3 fuels, first we used the MA from the spent fuel above (^{237}Np: ^{241}Am: ^{242m}Am: ^{243}Am: ^{243}Cm: ^{244}Cm: ^{245}Cm=47.5: 35.53: 0.07: 12.86: 0.03: 3.74: 0.22) and put this MA in 5% ^{235}U-enriched UOx fuel. This U-MA fuel was burnt in PWR until 30 GWd/tHM. Then the Pu isotopic vector of this spent fuel was used for the P^3 fuels. Table I shows the isotopic composition of each fuel. Since we projected the P^3 using scenario, 5% ^{241}Am (Case 2) and 5% ^{237}Np (Case 3) were added to ^{238}U in blanket area. ^{238}Pu isotopic composition of the P^3 case was over 15% and met the criterion of Kimura et al. [17].

Table I. Isotopic compositions of the analyzed cases

	Isotopic composition of Pu in driver fuel [%]					Isotopic composition in blanket fuel [%]		
	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	U-238	Am-241	Np-237
Case 1	1.9	56.8	22.9	12	6.4	100	0	0
Case 2	21.42	53.3	12.54	9.24	3.49	95	5	0
Case 3							0	5

3.3.3. Results

The behavior of the multiplication coefficient, k-effective, according to the burn-up is shown in Fig. 5 [11,13]. The red line represents the normal MOX case (case 1) and the black line represents the P^3 case (case 3). It can be seen that the reactivity drastically increases every

180 EFPDs because of the refueling. It can be seen that the initial excess reactivity in P^3 case (black line, case 3) is a little more suppressed than the normal MOX case (red line, case 1) because the MAs in the blanket capture neutrons and ^{238}Pu in the driver fuel area is less fissionable than ^{239}Pu. However, ^{238}Pu is still nicely fissile in fast neutron energy region and contributes for criticality, which can remain k-eff over 1.

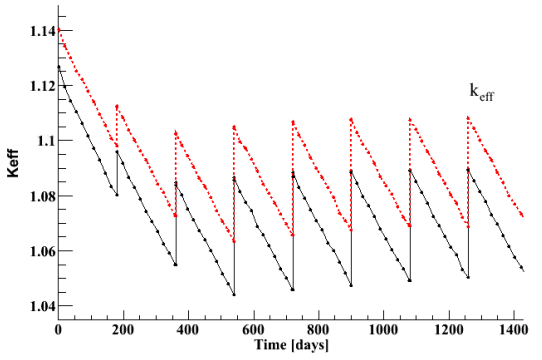

FIG. 5. Time evolution of the multiplication coefficient K-effective: in red the FBR is loaded with the normal MOX fuel; in black with P^3 fuel (case 3).

Table II shows the Pu isotopic composition of the total region of full core at 900 EFPDs. It is confirmed that ^{238}Pu isotopic compositions for case 2 and case 3 are still over 15% and meet the Kimura's criterion. As we refuel fuels of the reactor, the spent fuels are normally stored in a storage pool, and MURE enables us to access various information of the discharged Pu. In the normal MOX case, as the percentage of ^{238}Pu is few, it doesn't meet the Kimura's criterion. On the other hand, in the P^3 case, as the ^{238}Pu isotopic composition of the spent fuel is high, the Pu isotopic composition keeps meeting the Kimura's criterion during the storage period.

Table II. Pu isotopic compositions in the core at 900 EFPDs

	Pu isotopic composition [wt.%]				
	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
Case 1	0.04	61.11	24.29	10.53	4.04
Case 2	17.94	57.73	14.16	6.78	3.4
Case 3	18.95	56.98	13.98	6.72	3.36

4. Potential Case Study with Research Reactor Simulations

The MURE code is highly flexible to simulate reactors with a refueling scheme, an operation history or a core geometry that does not follow the standards used in reactors dedicated to electricity production. For instance, in the context of the development of the antineutrino detector Nucifer [20], a 3D simulation of the research reactor OSIRIS (CEA-Saclay, France) was performed taking into account the initial burnup of the fuel elements plus a follow up of the thermal power and the control rod positions during the operation of the reactor [10]. Based on the MURE features developed by the SUBATECH team, the simulation of another research reactor BR2 (SCK-CEN, Mol) is actually under development within the SoLid collaboration [21, 22].
In addition, the MURE code was used to simulate the energy spectrum of the gammas emitted

by the research reactor OSIRIS and to propagate them in the pool up to a bunker located behind the concrete walls. This dedicated simulation, very detailed in the processes involved in the gamma transport, allowed to better understand the gamma background measured [10].

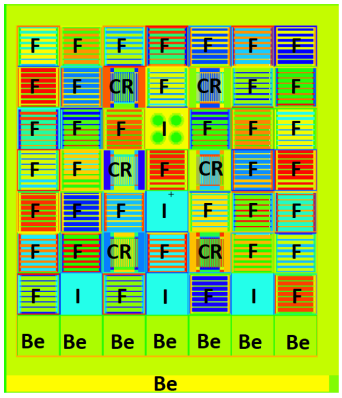

FIG. 5. Geometry simulated with the MURE code of the OSIRIS research reactor (Saclay, France) [10]. The elements are labeled according to their role in the core: F stands for fuel elements, I for irradiation cells and CR for control rods.

A case study could be performed involving a research reactor, in order to determine what kind of operation of the reactor or irradiations in the dedicated cells would be visible with the existing inspection devices and what would not be. The MURE-CHARS software would allow in addition to compute the gamma-rays to be detected in fresh and used fuel assemblies.

5. Conclusion

To tackle nuclear material proliferation, we conducted several proliferation scenarios using the MURE code. We have both developed a cell simulation of a CANDU reactor, which could be used to analyze the composition of fuel assemblies as a function of time or burnup. Diversion cases of Pebble Bed Reactor loaded with UOx, PuOx and ThUOx fuels, and a Na-cooled Fast Breeder Reactor with depleted Uranium or Minor Actinides in the blanket are studied. The loading of Protected Plutonium Production (P^3) in the FBR was also simulated. A realistic simulation of two French PWRs from the Chooz power plant was performed following the operation parameters (power and boron concentration) of the reactor cores in the frame of the Double Chooz reactor experiment. This work shows that, in principle, it is possible to perform a core simulation following a reactor cycle history, which could be used by inspectors to compute the awaited fuel composition, and what would be this composition in assumed cases of diversions. The MURE CHARS tool could be used to predict what would be the radioactive emission from diverse fuel compositions and irradiation histories, which could be compared with measurements. Research reactors have been simulated as well and could constitute a case study in collaboration with the IAEA.
Overall, the simulations of various reactor designs taking into account reactor physics constraints may bring valuable information to the IAEA safeguards, as it is easy to know the Pu production inside reactors loaded with varied types of fuels.

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