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Multi-pass Monte Carlo simulation method in nuclear transmutations

Liviu Mateescu[a,*], N. Prasad Kadambi[b], Nuggehalli M. Ravindra[a]

[a] New Jersey Institute of Technology, United States
[b] Kadambi Engineering Consultants, United States

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ABSTRACT

Monte Carlo methods, in their direct brute simulation incarnation, bring realistic results if the involved probabilities, be they geometrical or otherwise, remain constant for the duration of the simulation.

However, there are physical setups where the evolution of the simulation represents a modification of the simulated system itself.

Chief among such evolving simulated systems are the activation/transmutation setups. That is, the simulation starts with a given set of probabilities, which are determined by the geometry of the system, the components and by the microscopic interaction cross-sections. However, the relative weight of the components of the system changes along with the steps of the simulation. A natural measure would be adjusting probabilities after every step of the simulation.

On the other hand, the physical system has typically a number of components of the order of Avogadro's number, usually 10^{25} or 10^{26} members. A simulation step changes the characteristics for just a few of these members; a probability will therefore shift by a quantity of $1/10^{25}$. Such a change cannot be accounted for within a simulation, because then the simulation should have then a number of at least 10^{28} steps in order to have some significance. This is not feasible, of course. For our computing devices, a simulation of one million steps is comfortable, but a further order of magnitude becomes too big a stretch for the computing resources.

We propose here a method of dealing with the changing probabilities, leading to the increasing of the precision. This method is intended as a fast approximating approach, and also as a simple introduction (for the benefit of students) in the very branched subject of Monte Carlo simulations vis-à-vis nuclear reactors.

1. Introduction

No simulation can be discussed *in abstractum*, therefore we studied the particular case of current great interest, of Np-237 to Pu-238, without impeding the generality of the subject.

We will illustrate here a direct simulation, following mainly the transmutation of atoms in a lattice of Np-237 in atoms of Pu-238. We picked this particular transmutation for the obvious reason: it is of great interest nowadays, in relation with the energy needs for space probes.

US Plutonium-238 production has just restarted, with the first 50 g at Oak Ridge in December 2015 (Walli). We do not enter here in the discussions regarding opportunity of using other radioisotopes than Pu-238; we consider that this discussion was settled once and forever in (Ralph and McNutt, 2014).

The quantities necessary for space applications mainly beyond Mars orbit are in excess of 2 kg per year. Besides the Oak Ridge facility, which can produce Pu-238 using a particular methodology, other propositions have been advanced for obtaining significant quantities of this isotope. They mainly include using the Advanced Test Reactor (ATR) in Idaho, or the same High Flux Isotope Reactor at Oak Ridge, see (Caponiti, 2011). An interesting proposal calls for using exotic reactors, such as Triga (Steven et al., 2013), and a different setup than the usual Np-237 oxide encapsulated in Aluminum.

Our simulation involves a run-of-the-mill thermal neutrons reactor, with a rod of Np inserted between the fuel rods. We bring (back) the idea of thermal neutrons because of the considerable reserve of such neutrons in the existing reactors.

We propose using a thermal neutrons reactor as the source for transmuting neutrons. The usual counter-argument for this proposition is that the neutron flux at the target will be about 2×10^{12} neutrons/s/cm^2, in comparison with fluxes of 10^{16} or even 4×10^{14}, as in (Steven et al., 2013). This is why our standard production time is one year, as opposed with a few weeks in all the proposals. We posit that the slower yield is more than compensated by a possible syndication of production, since thermal neutrons reactors are the norm in today's world, and they are ubiquitous.

* Correspondence to: 600 Broad Street, Newark, NJ 07102, United States.
E-mail address: liviu.mateescu@gmail.com (L. Mateescu).

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2. Principle of the method

We devise a simple reactor, with cylindrical fuel rods, and the usual production of 5×10^{15} neutrons/s. Then we pose a cylindrical target between the rods, and proceed to a Monte Carlo simulation. We preferred a home-made program rather than one of the multitude of the existing MC packages, because it can accommodate easily our multi-pass method.

All the nuclear data used in the simulations pertaining to this paper come from (Korean_Atomic_Energy_Comission) or (Brookhaven_National_Laboratory).

We used a direct simulation, as a direct consequence of the principles in the seminal book of Sobol (Sobol, 1994), with refinements from (Bielajew, 1998; Rubinstein and Dirk, 2008).

Simplifying assumptions are unavoidable in simple models; however, we consider we avoided exaggerations.

First assumption, which no doubt will be disputed, is that the production of neutrons in the fuel rods stays constant. We are perfectly aware that the presence of the target may influence the neutron flux, but we consider that the control bars have command of much more reactivity reserve than a slim Np bar can steal.

The second assumption is of a logistic order: our simulation cannot modify on the fly probabilities with the accumulation of transmutation products, especially Np-238. This is why we conceived the simulation in two steps. The rate of production of Np-238 is established by a first simulation, and then, once this rate is found, a new simulation, with variable probabilities, takes place. The second step takes into account not only the alpha decay of Pu-238, but also the very large fission cross-section of Np-238.

Only elastic collisions were taken into account; the justification of this choice is clear from Fig. 1.

A third assumption, which in fact is not limitative, is the use of metallic Np-237, instead of its oxide. We consider from here on that all precautions were taken so that no undesired chemical reaction will take place. For example, the Np is separated from the water by a layer of Al or Au, which does not affect the neutron count.

The fission neutrons were simulated based on the energy spectrum found in, for example, (Hill, 1952).

The modification in-situ of the spectrum was followed during all the steps of the simulation, and the corresponding changes in the cross-sections were observed.

3. Setup

A reactor cooled and moderated by H_2O is used, with the rods disposed in a triangle/hexagonal pattern. Again, a setup which does not affect the generality of the method. Each fuel bar produces 5×10^{15} neutrons/s.

Fig. 2 represents the setup, with a cylindrical target. Other shapes (concave section, triangular section) were considered and simulations were performed in a previous phase of our study, and the conclusion was that circular section is the most effective for transmutation.

The minimum distance between the peripheries of the fuel rods is 2 cm, and a fuel element has the dimensions: 0.91 cm diameter, and 4 m length.

No matter what shape we assign to it, the target has a length of 4 m and a cross-section area of 0.8 cm^2.

4. First pass[1] (first simulation)

We generated one million neutrons in the fuel bars of order 1, 2, 3, and 4, respecting the energy distribution from (Hill, 1952). Each neutron was followed via simulation until it was absorbed in either water, or target, or a fuel bar.

Trying to separate the influence of the resonances around 1 eV, first we used in the resonances region a linear interpolation of the tabled cross-sections, results being seen in the first set of numbers in Table 1, then we followed a very fine grouping of the cross-sections in the interval between 0.1 and 10 eV. The results can be seen in the second row of Table 1.

The results are presented in Table 1.

We have to remark the fact that following the structure of the resonances meant a jump in the necessary computing power, which meant a much longer time for our one-million-neutron simulation. We decided that, since the method is meant from the beginning to be a first approximation, we will use the interpolated variant.

All the same, we took into consideration the spatial self-shielding, due to the accumulation of Np-238 and Pu-237, and we adjusted the probabilities/cross-sections accordingly.

A separate simulation (although it could have done within the first pass simulation) was performed for the production of Np-236, via the reaction ${}^{237}_{93}Np + {}^{1}_{0}n \rightarrow {}^{236}_{93}Np + 2{}^{1}_{0}n$. The 236 isotope is an undesired possibility, and must be kept under 6 ppm (Steven et al., 2013).

The results were under the sensitivity of the method. One million neutrons failed to bring one atom of the Np-236 isotope, which is to be expected, given that the average energy of a fission neutron is 0.8 Mev, and the (n,2n) reaction channel opens only at about 7 MeV (Korean_Atomic_Energy_Comission) (see also Fig. 1, the red graph). We stress the fact that this simulation is not a separate pass in our algorithm.

5. Second pass (second simulation)

Aware that the quantity of Np-238 generated is not automatically reflected in the final quantity of Pu-238, we proceeded to a second simulation, which takes into account the two main phenomena that can affect the quantity of the desired isotope. For second simulation, we adjusted the probabilities with the evolution in time of the Np-238. The general idea is to count out of the results in Table 1 the atoms of Np238 which will fission along the year. For that, we calculated the existing number of atoms of Np-238 at any moment of the year, then we adjusted the interaction probabilities accordingly.

For the second step, we used just the circular cylinder, which the first step showed to be the ideal shape. We start from a rate of production of 11.8 g/year, and from there we calculate the modifications brought by decay and fission. We consider, as in (Steven et al., 2013), that any atom of Np-238 undergoing fission is lost for the cause of Pu-238. As in the above-mentioned paper, we did not follow the fate of fission neutrons generated by Np-238, due to the lack of necessary information (how many neutrons per fission, fission spectrum, etc.). We are content with just the information that the atoms of Np-238 were “destroyed”.

Before the second simulation, we corrected the number of atoms of Np-238 existing at the end of every day of irradiation. We took into consideration the mass production rate of 11.8 g/year, and also the decay with a period of 2.1 days.

In what follows, the notations are as follows: N = number of Np-238 atoms; r_N = rate of production of Np-238 atoms, r_m = rate of mass production of Np-238, $T_{1/2}$ = half-life of Np-238 decay.

r_m =11.8 g/year, according to Step 1.

Then, $r_N = \frac{r_m}{m_n A}$, where m_n is the mass of a nucleon, and A is the atomic mass number.

The contribution to the increase of N is given by $dN_+ = r_N dt$, and the diminishing of N is given by $dN_- = -\lambda N dt$.

By solving the equation $dN = (r_N - \lambda N)dt$, with $\lambda = \ln 2 / T_{1/2}$, we obtain a corrected number of Np-238 atoms:

[1] We use the term “pass” because the more natural “step” is used in too many papers to indicate one instance of one and the same simulation, whereas pass indicates a separate simulation.