^{99}Mo Cumulative and Individual fission yield from ^{235}U: a simple FLUKA exercise.

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The problem

The traditional production of ^{99}Mo/^{99m}Tc for medical application is based on the fission of ^{235}U in nuclear reactor and in the subsequent extraction of ^{99}Mo from the Uranium target. This production route yield in a no-carrier added, high specific activity, ^{99}Mo. In the literature the fission yield for ^{99}Mo is quoted to be 0.061 atom per fission. In order to check the FLUKA capability to simulate the ^{99}Mo production from the neutron induced fission of ^{235}U a simple model has been implemented (see the fig 1).

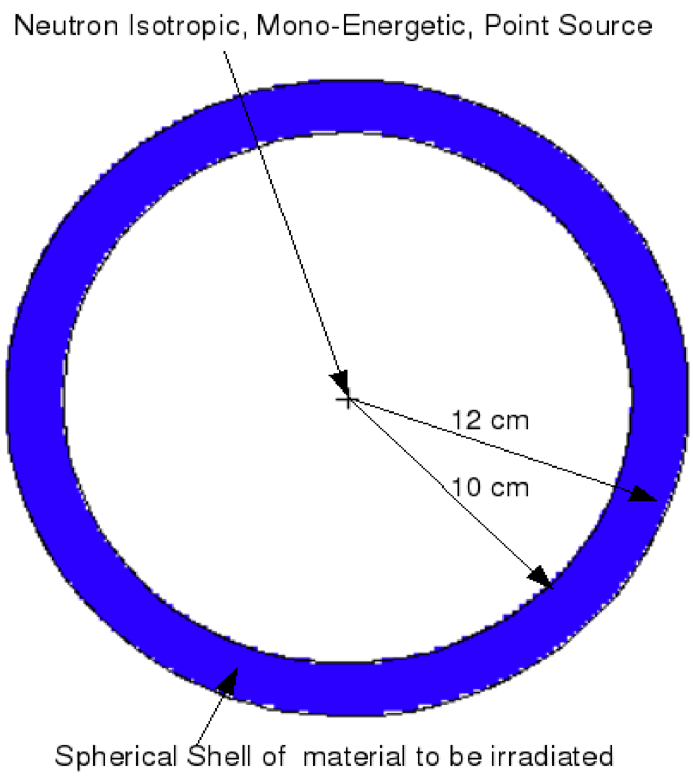

Figure 1: Schematic view of the problem geometry.

The geometry is constituted by a first void spherical shell that host in its center an isotropic point source (neutron energy 0.1 eV) and by a subsequent spherical shell filled with metallic Uranium (100% ^{235}U, bulk density 19.00 g/cm3). The number of fissions induced in the Uranium layer has been estimated by using the SCORE estimator in the FISSIONS modality. The saturation activity of ^{99}Mo has been estimated by using the RESNUCLEI estimator. The results have been averaged on a batch of 4 runs with different initialization of the random number generator. The fissions in ^{235}U has been estimated to be 2.09 per primary neutron and the saturation activity of ^{99}Mo is 2.3058E-04 atom/fission (4.75E-04 atom/pr). This results is at least 2 order of magnitude below the expected results of 0.06 atom/fission. Evidently something was missed in the simulations and I try to post the problem to the FLUKA-DISCUSS mailing list. Unfortunately, I send the mail in HTML format making unreadable the attached files and turning this simple exercise in an unsolvable problem. As usual, with a little bit of concentration, I found the solution of the problem a couple of days after I have posted my request of support.

The solution

It is well known that the fission fragments are generated directly (Individual fission yield) during the fission event or indirectly from the decays of previously generated fission fragments (decay chains). The overall yield of a given fission fragment is also called cumulative yield. In the case of ^{99}Mo exists the ^{99}Y decay chain (see Table 1).

Table 1: 99Y decay chain and the relative half life of all parents.

Nuclide	Half life
^{99}Y	1.470(7) s
^{99}Zr	2.1(1) s
^{99}Nb	15.0(2) s
^{99}Mo	2.7489(6) d
^{99}Tc	2.111(12)E+5 y
^{99}Ru	Stable

It is clear that a great contribution to the ^{99}Mo cumulative yield is given by the Yttrium decay chain after few minutes of irradiation. As the previous run has been executed in a stationary time conditions the only contribution that can be estimated is the individual saturation yield of ^{99}Mo (I.e. the ^{99}Mo produced directly in the fission events). In order to verify the FLUKA capability (or is better to say my capability) to estimate the ^{99}Mo cumulative yield the problem has been executed again turning on decays on the RADDECAY card and allowing an irradiation profile (IRRPROFI card) of 10 days with a neutron source intensity (4.18e+17 n/s) equivalent to the forward neutron current crossing the inner surface of the uranium spherical shell of the problem when the neutron flux is 1e+14 n cm^{-2} s^{-1} (the order of magnitude of the flux in a High Flux Reactor). By using the DECAYTIMES, DECAYSCORES and RESNUCLEI cards the ^{99}Mo amount has been estimated at given times during the irradiation. In order to check the consistency of the FLUKA results the cumulative and the individual activities of 99Mo at the given irradiation time have been calculated by using the following irradiation kinetic equation:

$$A = A_o \left(1 - e^{\frac{-\ln(2)}{t_{1/2}} t_{irr}}\right)$$

where
A_0 is the specific saturation activity [Bq/g]
$t_{1/2}$ is ^{99}Mo Halflife [s]
t_{irr} is the given irradiation time.

In the case of the Cumulative Yield the Ao has been calculated has follow

A_0= Source Intensity [n/s] * Fission [fission/n] * ^{99}Mo Cumulative Saturation Activity [atoms/fission] / Shell Volume [cm^3] /bulk density [g/cm^3] = 2.09 * 4.18880017E+17 * 0.062/3049.43994 / 19.

Where the ^{99}Mo cumulative Saturation activity per fission is the one quoted in the literature (0.062 atom/fission) and the total fission has been obtained from the SCORE estimator of the first problem.

In the case of ^{99}Mo individual fission yield the Ao has been calculate as follows
A_0= ^{99}Mo Individual Saturation activity [atoms/cm^3/n] * Source Intensity [n/s] / bulk density [g/cm3] = 1.5413e-07*4.18880017E+17*0.062/19.

The value of ^{99}Mo Individual Saturation activity has been obtained by the RESNUCLEI output of the first run. The results have been reported Table 2 :

Table : Comparison of the ^{99}Mo Cumulative yield obtained directly from a FLUKA simulation with the IRRPROFI procedure and the ones obtained by post processing of a time independent run

Irradiation Time [Hours]	Estimated ^{99}Mo Cumulative activity [Bq/g]		Estimated ^{99}Mo Individual activity [Bq/g]
	FLUKA	* Hand made Calculation	* Hand made Calculation
24	1.93E+11	1.98E+11	7.19E+08
48	3.44E+11	3.53E+11	1.28E+09
72	4.61E+11	4.72E+11	1.71E+09
96	5.50E+11	5.65E+11	2.05E+09
120	6.20E+11	6.38E+11	2.31E+09
144	6.75E+11	6.94E+11	2.52E+09
168	7.20E+11	7.38E+11	2.68E+09
192	7.50E+11	7.72E+11	2.80E+09
216	7.80E+11	7.98E+11	2.89E+09
240	8.00E+11	8.18E+11	2.97E+09

*Hand made calculation have been based on the post processing of fission data from the first run.

The good agreement between FLUKA and the hand made calculation in the case of the ^{99}Mo Cumulative activity confirms that FLUKA simulations is well aligned with the values reported in the literature.