Decay Interval Effect on Targets Containing Curium Upon Subsequent Irradiation

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INTRODUCTION

The Radiochemical Engineering Development Center (REDC) at Oak Ridge National Laboratory (ORNL) irradiates curium oxide targets in the High Flux Isotope Reactor (HFIR) to produce ^{252}Cf and other transuranium isotopes for research, industrial, and medical applications. These materials are produced in the flux trap of the HFIR, where the peak thermal-neutron flux is about 2.3×10^{15} neutrons/(cm^2·s). The curium oxide is pressed with aluminum powder into pellets and loaded into aluminum target tubes. There are typically 35 pellets per target and 9–13 targets per "campaign." The oxide material, rich in the heavier isotopes of curium, also has small amounts of some isotopes of plutonium and americium. The composition of the oxide is determined through calorimetry and other analyses performed on the pellet and oxide batches. Time-dependent fission heat calculations, using the FORTRAN programs TCOMP and FCOMP [1, 2], on the target with the greatest loading of curium oxide provide a thermal qualification for irradiation in the HFIR. The actinide composition and expected reactor power history are input, and the fission rates are output for each time step, which is then used to determine the peak fission rate. The reactor's central position is assumed for the target qualification. This "hottest" position, applied to a pellet, is mathematically extended to the whole target. For the current set of targets, the expected peak fission heating rate was found to occur on the first day of irradiation and to be well below (about 65% of) the allowable maximum [3, 4].

DESCRIPTION

The recent extended downtime of the HFIR to change the reactor's beryllium reflector raised concerns about what effect on fission heating that fissionable decay daughters, grown in

during the reactor downtime, would have upon subsequent irradiation of the targets. The current set of targets previously underwent three irradiation cycles and has been decaying in the reactor pool for a little over a year. Their known irradiation history was used to calculate the effect of the reactor downtime on the expected fission heating when irradiation resumes. Table 1 shows the original gram amounts of the various nuclides per target before they were first irradiated; the amounts of the various nuclides after three irradiation cycles; and the amounts after 1.25, 2, 3, and 5 years of decay.

Some of the decay daughters growing in have large capture cross sections, which lead to nuclides with large fission cross sections that must be considered when calculating the expected peak in fission heat upon resuming irradiation. The current set of targets is expected to undergo about 1.25 years of decay before the HFIR restarts.

RESULTS

Figure 1 shows the calculated peak fission rate for the fresh targets, experienced within the first day of the initial irradiation cycle, and the expected peaks for subsequent irradiation if the targets undergo various decay intervals. The peaks for decayed targets occur later (6 or 7 days) into the first irradiation cycle as the ingrowth of the plutonium nuclides becomes more dominant in the fission heat production.

The maximum allowable fission rate is different for fresh targets than for decayed targets because oxide film developing on the aluminum housing impedes heat transfer.

Although, for this set of targets, no allowable fission heating rates were exceeded, it can be seen that decay interval can certainly affect the heat production. This concern could also apply to some power reactors in the event of extended shutdown if significant ^{244}Cm inventories have been accumulated.

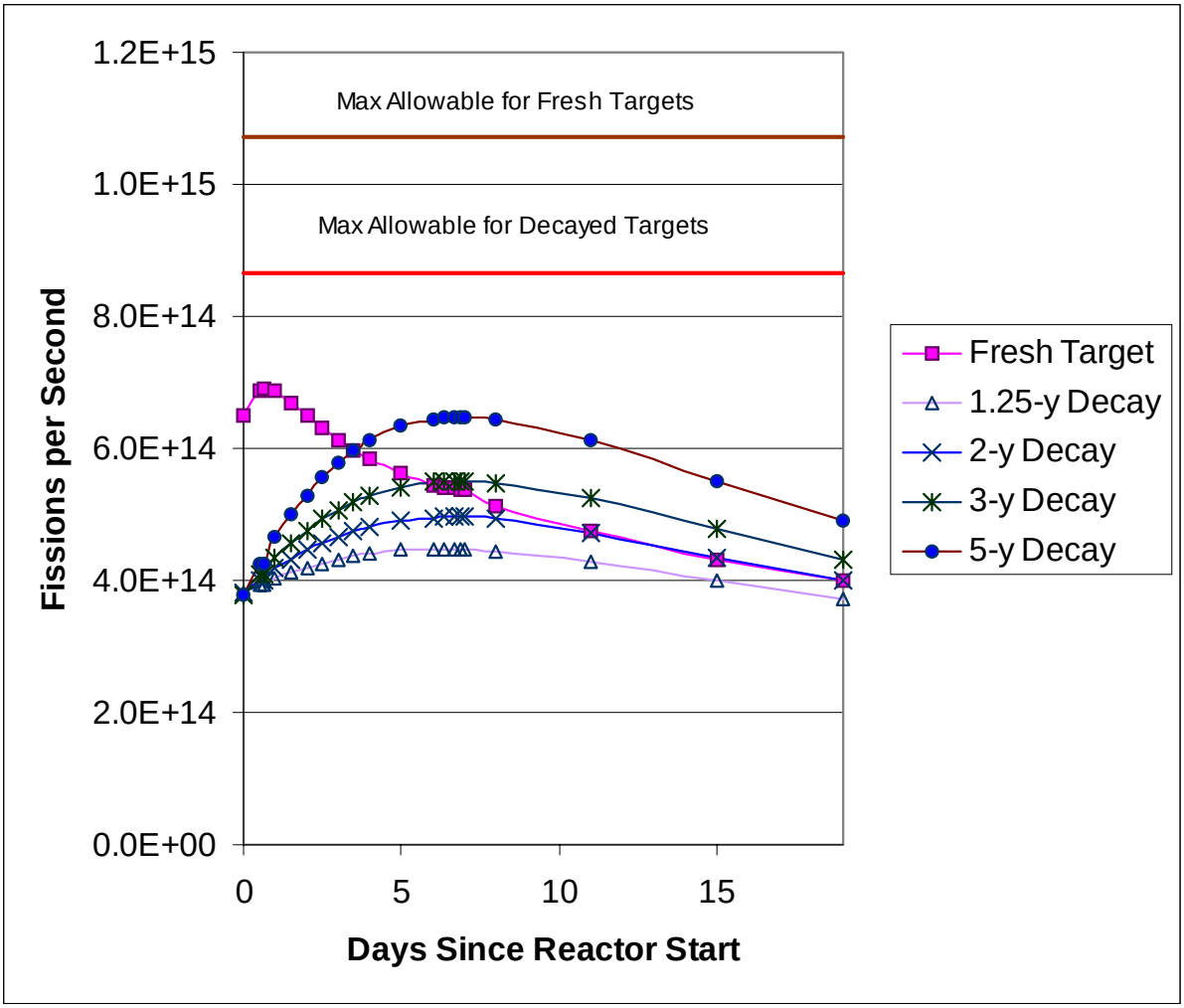
TABLE 1

Gram Amounts of Nuclides/Curium Target at Various Decay Intervals

Nuclide	g@Start	g After 3 cyc	g@1.25-y Decay	g@2-y Decay	g@3-y Decay	g@5-y Decay
Pu-238	3.33300E-03	2.71910E-05	5.25160E-02	5.82631E-02	5.99640E-02	5.95728E-02
Pu-239	1.56000E-04	3.35570E-05	8.21971E-05	1.10693E-04	1.47903E-04	2.19713E-04
Pu-240	1.16329E-01	1.35390E-03	9.12446E-02	1.43144E-01	2.10059E-01	3.36419E-01
Pu-241	4.20000E-05	1.19490E-03	1.12693E-03	1.08808E-03	1.03841E-03	9.45994E-04
Pu-242	2.36200E-03	2.17870E-02	2.25696E-02	2.30339E-02	2.36528E-02	2.48903E-02
Pu-243		7.32320E-05	3.99973E-12	3.99973E-12	3.99973E-12	3.99973E-12
Am-241	1.74520E-01	6.54350E-04	7.22966E-04	7.62087E-04	8.11972E-04	9.04392E-04
Am-242*m*	6.08000E-04	4.80460E-06	4.77517E-06	4.75760E-06	4.73428E-06	4.68797E-06
Am-242*g*	4.91943E-01	5.06280E-05	6.15990E-11	6.13724E-11	6.10715E-11	6.04741E-11
Am-243		1.95014E-01	1.94991E-01	1.94978E-01	1.94959E-01	1.94923E-01
Am-244		1.95612E-04	0.00000E+00	0.00000E+00	0.00000E+00	0.00000E+00
Cm-242	3.65000E-04	6.26970E-02	8.98494E-03	2.79964E-03	5.91403E-04	2.63905E-05
Cm-243	1.20300E-03	1.69120E-03	1.64160E-03	1.61254E-03	1.57459E-03	1.50135E-03
Cm-244	2.04011E+00	1.95580E+00	1.86458E+00	1.81180E+00	1.74375E+00	1.61520E+00
Cm-245	4.54900E-02	2.08517E-02	2.08496E-02	2.08483E-02	2.08466E-02	2.08432E-02
Cm-246	4.39595E+00	4.29748E+00	4.29699E+00	4.29668E+00	4.29627E+00	4.29541E+00
Cm-247	1.21545E-01	1.12181E-01	1.12181E-01	1.12181E-01	1.12181E-01	1.12181E-01
Cm-248	9.07993E-01	8.90380E-01	8.96008E-01	8.98598E-01	9.01347E-01	9.05090E-01
Cm-249			6.97877E-07	6.33219E-07	5.56231E-07	4.29197E-07
Cf-249			3.96959E-03	5.01677E-03	5.71743E-03	6.16249E-03
Cf-250		4.72980E-03	4.42671E-03	4.25425E-03	4.03472E-03	3.62906E-03
Cf-251		1.31900E-03	1.31773E-03	1.31697E-03	1.31595E-03	1.31392E-03
Cf-252		2.11370E-02	1.52339E-02	1.25161E-02	9.63126E-03	5.70308E-03
Cf-253		2.66530E-04	5.13561E-12	1.20642E-16	8.11665E-23	3.67393E-35
Bk-249		6.33060E-03	2.38984E-03	1.32048E-03	5.98706E-04	1.23078E-04
Es-253		8.82640E-05	3.74019E-10	3.78743E-14	1.64859E-19	3.00991E-30
Es-254		4.63390E-07	1.47080E-07	7.38785E-08	2.94988E-08	4.70301E-09

FIGURE 1.

Fission Rates for Various Target Decay Intervals



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