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Nuclear Criticality Analyses of Separations Processes for the Transmutation Fuel Cycle: Quarterly Report

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Quarterly Report
(Second Quarter)
AAA/UNLV University Participation Program

Title:

Nuclear Criticality Analyses of Separations Processes for the Transmutation Fuel Cycle

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1. Project Description

The success of the ATW program will rely upon the ability of radiochemists to separate spent nuclear fuel into uranium, fission products, and transuranic wastes. The Chemical Technology Division at the Argonne National Laboratory is actively involved in the development of pyrochemical separation technology that minimizes the usage of strong acids with the subsequent problems involved in disposing of the acidic residue.

Small scale experiments are being validated at ANL to separate spent fuel, but they must be scaled up to accommodate the large amount of commercial spent fuel that must be treated. As the volume of waste to be treated is increased, there is a higher probability that fissionable isotopes of plutonium, americium, and curium can accumulate and form a critical mass. Criticality events can be avoided by ensuring that the effective neutron multiplication factor, k_{eff}, remains below a safe level. NRC regulations normally allow an upper value of 0.95 for k_{eff}. This parameter can be computed for any combination of fuel and geometry using Monte Carlo neutron transport codes. SCALE 4.4a from the Oak Ridge National Laboratory and MCNP4C2 from the Los Alamos National Laboratory are two codes that are regularly used to assess criticality.

In this project, students at the University of Nevada were trained in the use of KENO and SCALE 4.4a to assist Dr. Laidler and his team at ANL in criticality safety assessments.

2. Review of Tasks

The proposed tasks for this project are listed in the timetable shown below:

Task	9/01	10/01	11/01	12/01	1/02	2/02	3/02	4/02	5/02	6/02	7/02	8/02
Train Students in use of Monte Carlo Codes												
Student and Faculty Visits to ANL/CMT												
Simulation of Criticality for ANL Designs												
Integrate Criticality Codes into Excel Model												

This Quarterly Report covers progress to this point.

During the first two quarters of the work, the tasks included training students in the use of Monte Carlo codes used in radiation transport studies and the assessment of neutron multiplication factors for specific problems outlined by ANL-East through Drs. Laidler and Vandegrift.

The proposal also included objectives for the first year of work on this project, as listed below. The work conducted in the second quarter of the project was in partial completion of these objectives.

- Train UNLV students in the use of SCALE and/or MCNP for the assessment of nuclear criticality.

- Assess neutron multiplication factor, k_{eff}, for geometries and material concentrations as defined by the collaborating team from ANL-CMT for the ATW project.

- Provide software, extrapolation tables, or other methods to incorporate criticality estimates into the existing ANL Excel model of the pyrochemical treatment process to be used for ATW.

3. Progress in the Second Quarter

- Student Training

The following students are continuing their work from the first quarter.

- Jason Viggato – doctoral student in mechanical engineering.
- Elizabeth Bakker – senior in mechanical engineering.
- Daniel Lowe – sophomore in mechanical engineering

Mr. Lowe and Mr. Viggato are also involved with another AAA project involving radiation transport calculations for neutron spallation target studies at LANSCE. During the first quarter of both projects, students were taught to use KENO and SCALE 4.4a in preparation for their work in radiation transport and criticality.

In January 2002, all three students also completed the introductory course in MCNPX offered by Dr. Laurie Waters from LANL. The course was taught at UNLV and was supported by Tony Hechanova and the UNLV/AAA University Participation Program. In addition to radiation transport modeling, the students also did a criticality problem in class. The nuclear criticality program, MCNP4C, is included in the current distribution of MCNPX.

In April, 2002, Mr. Viggato accepted a job with Bechtel, SAIC working on the Yucca Mountain Project. His job involves the analysis of thermal and criticality problems for the proposed national nuclear waste repository. He is continuing his work to finish his doctoral project.

Computational Resources

The students working on the project have been trained in the use of SCALE 4.4a, a Monte Carlo simulation code that simulates the scattering and absorption of neutrons in nuclear fuel. Students began their training with simple problems using KENO IV and are now involved in preparing CSPAN input files. Danny Lowe and Elizabeth Bakker are also involved in writing BASIC programs that automate the process of preparing CSAS and KENO-VI input.

Through the project, each student was equipped with a 1.8 GHz Gateway computer with 512 MB of memory. The students have desks and workspace allocated in TBE B-113 of the Engineering Complex at UNLV.

- Simulation of Criticality for ANL Designs

Dr. Laidler requested information about the potential for samples of curium to become critical. The curium will be separated from spent nuclear fuel and will contain fissionable isotopes. The problem was analyzed using SCALE 4.4a by Ms. Bakker and an extensive report titled: "Fission and Thermal Effects in Curium Separated from Spent Nuclear Fuel" was prepared and submitted to ANL.

Curium separated from spent fuel also contains isotopes that generate a great deal of decay heat. This heat generation also creates safety problems in the handling and storage of curium. The decay heat can cause samples to melt very quickly if excessive quantities of curium are created. The report also outlines the thermal problem and the maximum mass of curium that can be safely stored is documented.

- Integrate Criticality Codes into Excel Model

An Excel model was created in the first quarter for the analysis of mixtures of fission products, process salts, and TRU. In the second quarter, MathCad models were used to analyze thermal problems in curium.

4. Work Scheduled for Third Quarter

With completion of the second quarter work on curium, we are seeking additional criticality problems to analyze. Dr. Laidler previously expressed an interest in us looking at criticality safety in pyrochemical cells and we will pursue this analysis.

Appendix A

Report to ANL

Fission and Thermal Effects in Curium Separated from Spent Nuclear Fuel

Report

Fission and Thermal Effects in Curium Separated from Spent Nuclear Fuel

UNLV

University of Nevada, Las Vegas

April 19, 2002

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Abstract

Curium poses special problems in the chemical preparation of spent nuclear fuel for transmutation. Once separated from the other minor actinides, the seven curium isotopes in spent fuel can lead to nuclear fission with the subsequent release of a large amount of radiation. Several isotopes of curium also generate a significant amount of heat by radioactive decay. Sustained fission can be avoided by preventing the accumulation by more that a critical mass of curium. The heat generation of curium presents even more restriction on the mass of curium that can safely be contained in one location.

To analyze the nuclear and thermal properties of curium, the curium isotopes within spent fuel were quantified using RADDB, a light water reactor radiological database. The critical mass of curium was analyzed for shielded and unshielded cylindrical and spherical geometries. The criticality studies were completed using SCALE 4.4a, a Monte Carlo code approved by the Nuclear Regulatory Commission for the analysis of critical nuclear reactors. The results include recommendations on the maximum mass of curium that may be safely handled.

Finally, a conservative case for the buildup of decay heat was analyzed to determine the equilibrium temperature of a curium-filled container. Both natural convection and radiation heat transfer were considered. The equilibrium temperature was used to recommend the maximum mass of curium that can be safely handled or stored before melting occurs.

The Fission Properties of Curium Separated from Spent Nuclear Fuel

1. Why is Curium a Concern?

Curium presents special problems in the treatment of spent nuclear fuel. It is desirable to separate out the isotopes of curium from other transuranic elements to allow for further treatment. Curium, however, has several isotopes that are fissionable and care must be taken to prevent the accumulation of a critical mass. Curium isotopes also generate a significant amount of decay heat that may lead to high temperatures within containers used to process curium.

In this report, some of the nuclear and thermal properties of curium are reviewed. The critical mass of curium was investigated for cylindrical and spherical containers filled with curium to assess the minimum quantity of the element that may lead to criticality. Recommendations are made on critical mass and heat generation to assist the scientists and engineers working on the pyrochemical treatment of nuclear waste. Decay heat generation was also investigated to determine the maximum quantities of curium that can safely be handled and stored.

1.1 Fission

Seven isotopes of curium[1] are found in significant quantities within spent nuclear fuel and several of these isotopes are fissionable. A neutron criticality code was used to determine the effective neutron multiplication factor for varying quantities of curium held within cylindrical containers or stored in a spherical shape. Recommendations are made on the maximum amount of curium that can be safely stored or handled before encountering nuclear criticality.

1.2 Decay Heat Generation

Curium generates a great deal of decay heat. For spent nuclear fuel, 2.6 watts of heat may be generated for every gram of curium. This will present special problems when curium is separated from the other minor actinides. If kilogram-quantities of curium are stored in a container, for example, the curium may heat to an equilibrium temperature that exceeds the melting temperature of this actinide.

To study the effects of decay heat on the time-dependent temperature of curium, a heat transfer analysis was completed to determine the equilibrium temperature and additional thermal properties.

2. Properties of Curium

2.1 General Properties

Curium with an atomic number of 96 is a minor actinide present in spent nuclear fuel. It is created by the bombardment of plutonium nuclei by neutrons within the reactor core[2]. It has a density of 13,511 kg/m^3 and it melts at 1613.15 K. A summary of some of its other properties are included in Table 2.1[3].

Table 2.1 Properties of Curium

Property	Value	Units
Electron Configuration	(Rn)5f^{7}6d7s^2	
Atomic Number	96	
Atomic Mass	247	amu
Melting Point	1613.15	K
Boiling Point	unknown	K
Atomic Radius	119	pm
Density	13511	kg/m^3
Pauling Electronegativity	1.3	
State at Room Temperature	Solid	
Oxidation States	3	
Thermal Conductivity	10	W/m*K
Heat of Fusion	15	kJ/mol

Curium has a low thermal conductivity (10 W/m-K) when compared with metals, however it is comparable with UO_2 fuel (7.3 W/m-K at 400 K)[4].

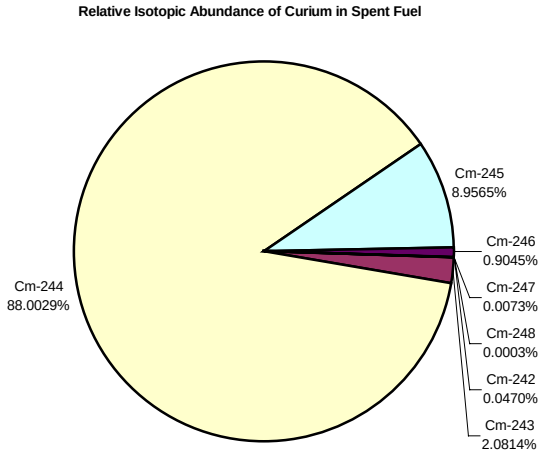

Figure 2.1 Typical Isotopic Abundance of Curium in Spent Fuel (30 GWD/MTIHM, 3.11% Initial Enrichment, 30 Year Old Fuel)

Spent fuel contains significant quantities of seven isotopes of curium. As shown in Figure 2.1, the most abundant isotope is Cm-244 at 88% with Cm-245 following at 9%.

The isotopic abundance is strongly dependent upon fuel burnup, initial enrichment, and the time since the fuel was removed from the reactor. Isotopic abundance is discussed further in Section 3.

2.2 Neutron Cross Sections

Neutrons entering a sample of curium will be scattered, captured with the resulting emission of a gamma ray, or captured with the nucleus subsequently undergoing fission. The probability of each interaction occurring is expressed in terms of the microscopic cross section, σ. The microscopic cross section varies with incident neutron energy and the target isotope. Table 2.2 lists cross section values for the important isotopes of curium exposed to "thermal" or 0.025 eV neutrons[5].

Table 2.2 Microscopic Cross Sections for Curium Isotopes

Microscopic Cross Sections (barns/atom)

Isotope	fission	scattering	capture	absorption
242	< 5.00		16	16
243	617		130	747
244	1.04	11.6	15.2	16.24
245	2145		369	2514
246	0.14	11.1	1.22	1.36
247	81.9		57	138.9
248	0.37		2.63	3

Curium 245 has a very high fission cross section of 2145 barns. The fission cross section for Pu-239, for comparison, is 742.5 barns per atom[6]. Cm-243 and 247 also contribute to fission. The high fission cross sections lead to criticality concerns when more than 5 kilograms of curium is collected in one

Table 2.3 Curium Halflives and Decay Schemes

Isotope	Atomic Mass (amu)	Specific Activity (Ci/g)	Halflife	Decay Scheme	% Decay by Scheme	Decay Energy (MeV)	Emitted Radiation Energy (MeV) alpha	beta	gamma
242	242.0588	3400	162.8 d	SF	6.20E-06		6.1	0.01	0.0018
				alpha		6.216			
metastable			180 ns	IT		2.8			
243	243.0614	52	29.1 y	alpha	99.71	6.169	5.8	0.14	0.13
				E.C.	0.29	0.009			
				SF	5.30E-09				
244	244.0627	82	18.1 y	SF	1.30E-04		5.8	0.086	0.0017
				alpha		5.902			
metastable			34 ms	SF		1.04			
245	245.0655	0.17	8500 y	SF	6.10E-07		5.4	0.065	0.096
				alpha		5.623			
246	246.0672	0.31	4760 y	alpha	99.97	5.475	5.4	0.008	0.0015
				SF	0.03				
247	247.0704	9.40E-05	1.56e7 y	alpha		5.353	4.9	0.021	0.32
248	248.0723	4.30E-03	3.48e5 y	alpha	91.74	5.162	4.7	0.006	0.0012
				SF	8.26				

location. By handling and storing smaller quantities of curium, sustained fission can be avoided. This problem is discussed extensively in Sections 4 and 5.

2.3 Nuclear Properties

The halflives for the isotopes found in spent fuel are listed in Table 2.3[7]. All seven isotopes can decay by alpha emission resulting in the production of a 4 to 6 MeV helium nucleus. Most decays are accompanied by the emission of low energy gamma rays.

Cm-244 (18.1 year halflife) and Cm-245 (8500 year halflife) are the most abundant isotopes of curium found in spent fuel. They have specific activities of 84 and 0.17 curies per gram, respectively. For the curium contained within the spent fuel shown in Figure 2.1, the overall specific radioactivity is 75 curies per gram of curium[1].

2.4 Decay Heat Generated by Curium

The emission of approximately 5 MeV per alpha decay results in the production of considerable decay heat by the various isotopes of curium. This is discussed in detail in Section 6 since the resulting heat generation rate of approximately 2.5 watts per gram creates some significant problems in handling and storing curium samples. Equilibrium temperatures and estimates of safe quantities of curium are discussed in this subsequent section.

3. Isotopic Abundance in Spent Nuclear Fuel

3.1 Definition of Terms

There are seven curium isotopes that appear in significant quantities in spent nuclear fuel. The relative concentration of each isotope is highly dependent upon the fuel burnup, the initial enrichment of the fuel, and the age of the fuel.

The total energy generated as heat from nuclear fuel is expressed as the *fuel burnup* in units of megawatt-days or gigawatt-days of thermal energy produced per metric ton in heavy metal, or MWd/MTIHM. Each fuel assembly composed of roughly 200 fuel pins weighs about ½ of a metric ton. The majority of this weight is composed of the uranium within the fuel. An entire reactor core may hold 200 fuel assemblies. The maximum fuel burnup that commercial fuel may be exposed to is based on the amount of fissile uranium available in the fuel and upon the amount of radiation-induced damage allowed in the fuel and cladding. For commercial fuel, the burnup ranges from 30 to 60 GWd/MTIHM.

The *initial fuel enrichment* represents the ratio of fissile U-235 to total uranium within the fuel. The initial enrichment for PWR fuel is on the order of 3%. The enrichment of natural uranium as mined from ore is 0.714%. Greater initial enrichment allows increased fuel burnup.

The *age of the fuel* is defined as the time that has expired since the fuel was removed from its reactor. Spent fuel resides for at least a year at the reactor site in a water-filled pool to allow decay heat and radiation to be dissipated. Spent fuel radioactivity and heat generation decreases roughly as:

$$P / P_o = A / A_o = t(\text{years})^{-1.2} \qquad (3.1)$$

3.2 Changes with Fuel Burnup

The relative concentration of curium isotopes in spent fuel changes significantly with fuel burnup. To study the change in curium content with fuel burnup, the computer database RADDB[1] was used to compute the isotopic concentrations for four levels of fuel burnup. The nominal level of initial fuel enrichment was used for each case:

- 30,000 MWd/MTIHM, 3.11% initial enrichment.
- 40,000 MWd/MTIHM, 3.72% initial enrichment.
- 50,000 MWd/MTIHM, 4.26% initial enrichment.
- 60,000 MWd/MTIHM, 4.73% initial enrichment.

The resulting concentrations expressed in grams per metric ton in heavy metal of spent fuel are shown in Table 3.1.

Table 3.1 Isotopic Concentration of Curium in Spent Nuclear Fuel

Burnup	Curium Isotope (g/MTIHM)							Total
(MWD/	242	243	244	245	246	247	248	Curium
MTIHM)								g/MTIHM
30,000	3.10E-03	1.37E-01	5.80E+00	5.90E-01	5.96E-02	4.84E-04	2.28E-05	6.59E+00
40,000	5.68E-03	3.23E-01	1.66E+01	2.37E+00	2.80E-01	3.27E-03	2.10E-04	1.95E+01
50,000	6.54E-03	4.74E-01	3.12E+01	5.09E+00	7.84E-01	1.14E-02	9.29E-04	3.75E+01
60,000	7.03E-03	6.25E-01	5.26E+01	9.54E+00	1.87E+00	3.26E-02	3.32E-03	6.47E+01

Total curium content increases as fuel burnup is increased. By increasing the burnup from 30,000 to 60,000 MWd/MTIHM, the total curium content increases by a factor of 10.

The concentration of Cm-244 dominates the total curium content at 88% for low burnup. The heavier isotopes make up an increasing fraction of the total curium at higher levels of burnup. Cm-245 increases from 9% of the total to 15% of the total over the range of burnup investigated. It should be noted that Cm-245 is the most fissionable curium isotope with a cross section of 2145 barns. The relative concentrations are also shown in Figure 3.1 as a function of fuel burnup.

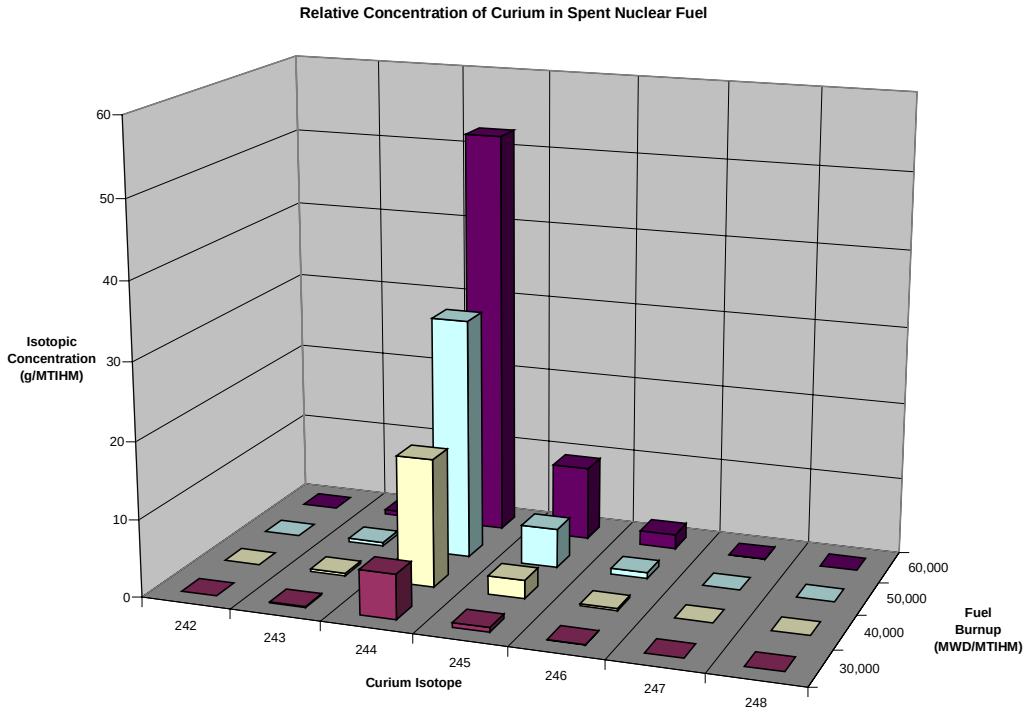

Figure 3.1 Relative Concentration of Curium Isotopes in Spent Nuclear Fuel As a Function of Fuel Burnup

3.3 Change with Initial Fuel Enrichment

The isotopic concentrations also change with initial fuel enrichment. For 30 year old fuel with a burnup of 30,000 MWd/MTIHM, the resulting concentrations in spent fuel are tabulated in Table 3.2.

Table 3.2 Changes in Isotopic Concentration with Initial Uranium Enrichment

PWR Fuel, 30,000 MWD/MTIHM, 30 Year Old								
Enrichment	Curium Isotope (g/MTIHM)							
(%)	Cm-242	Cm-243	Cm-244	Cm-245	Cm-246	Cm-247	Cm-248	Total Cm
2.5	3.52E-03	2.00E-01	1.03E+01	1.15E+00	1.37E-01	1.28E-03	6.99E-05	1.18E+01
2.75	3.33E-03	1.70E-01	8.03E+00	8.60E-01	9.52E-02	8.35E-04	4.29E-05	9.15E+00
3	3.17E-03	1.46E-01	6.38E+00	6.59E-01	6.84E-02	5.68E-04	2.74E-05	7.26E+00
3.25	2.98E-03	1.24E-01	5.01E+00	4.98E-01	4.83E-02	3.79E-04	1.72E-05	5.68E+00
3.5	2.79E-03	1.05E-01	3.91E+00	3.73E-01	3.39E-02	2.52E-04	1.07E-05	4.42E+00
3.75	2.63E-03	8.93E-02	3.10E+00	2.86E-01	2.44E-02	1.72E-04	6.92E-06	3.50E+00

Total curium content decreases as initial enrichment is increased as shown in Figure 3.2. Although total curium concentration in spent fuel decreases with enrichment, the fraction of curium that is fissionable (mainly Cm-245) drops from 9.7% to 8.2%. The isotope Cm-242 is responsible for much of the decay heat generated by curium. Its

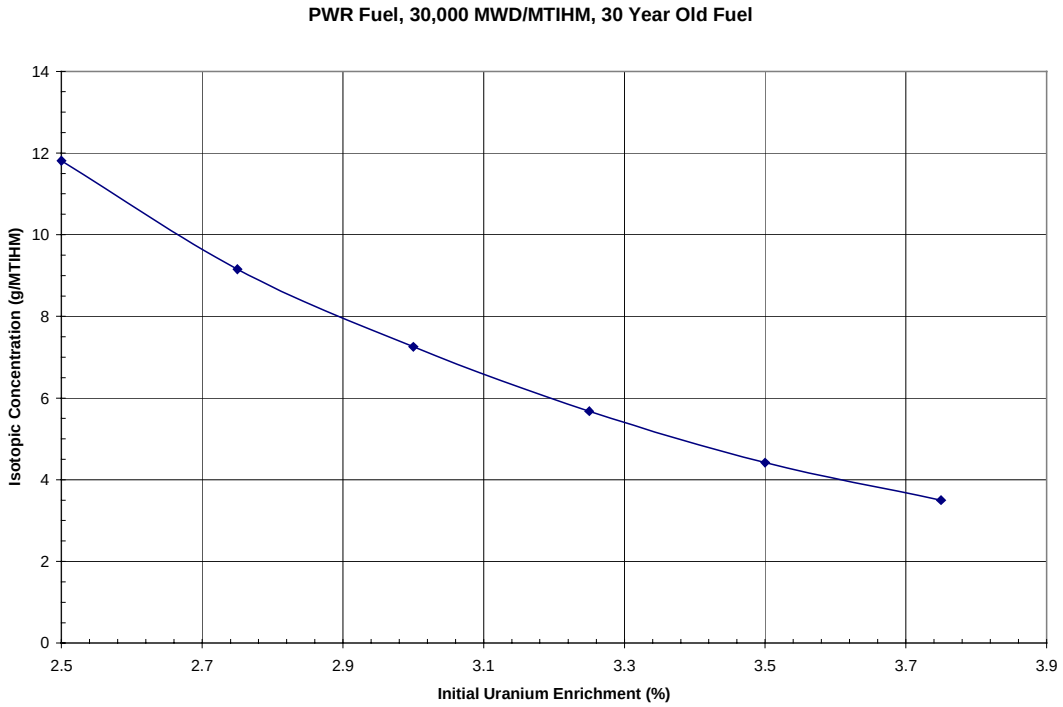

Figure 3.2 Total Curium Concentration in Spent Fuel as a Function of Initial Uranium Enrichment

fraction increases from 0.03% to 0.075% with increasing enrichment. A bar graph of curium concentration with initial uranium enrichment is included in Figure 3.4.

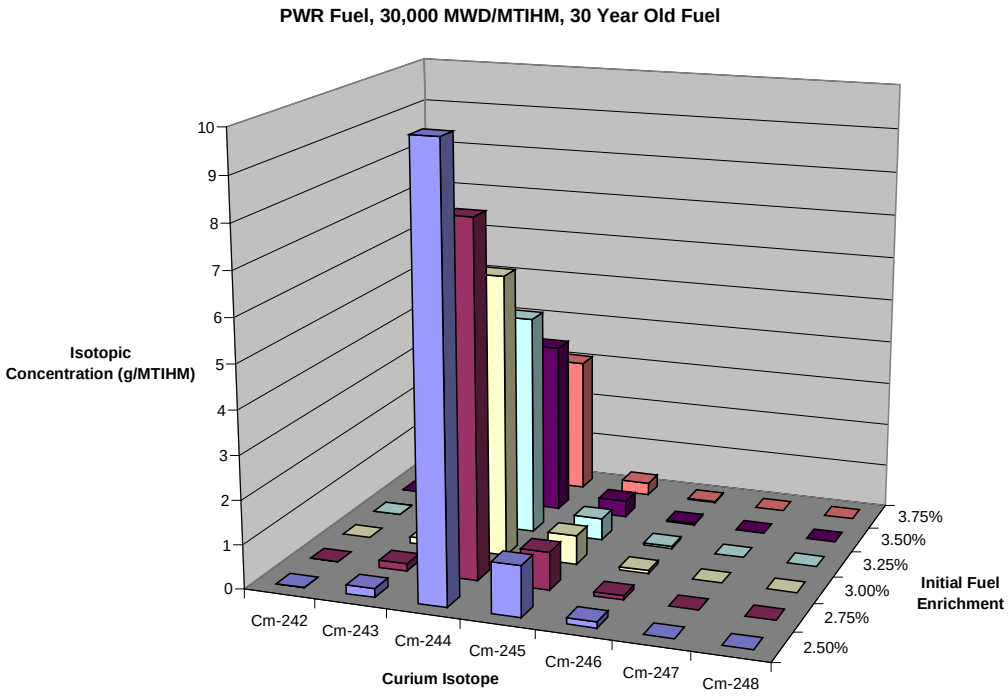

Figure 3.4 Relative Concentration of Curium Isotopes in Spent Fuel As a Function of Initial Fuel Enrichment

3.4 Change with the Age of Spent Fuel

The last case to be considered involves the age of the fuel. This is based on the number of years since the fuel was removed from the nuclear reactor. Total curium content decreases rapidly as fuel ages from 10 years to 1000 years. A plot of the total curium in Figure 3.5 shows that the concentration decreases to 5% of its 10-year value. Data in Table 3.3 and Figure 3.6 provide the isotopic concentrations.

Table 3.3 Changes in Isotopic Concentration with the Age of the Fuel

PWR Fuel, 30,000 MWD/MTIHM, 3.11% Initial Enrichment								
Age	Curium Isotope (g/MTIHM)							
(years)	Cm-242	Cm-243	Cm-244	Cm-245	Cm-246	Cm-247	Cm-248	Total Cm
10	3.40E-03	2.23E-01	1.25E+01	5.91E-01	5.98E-02	4.84E-04	2.28E-05	1.33E+01
30	3.10E-03	1.37E-01	5.80E+00	5.90E-01	5.96E-02	4.84E-04	2.28E-05	6.59E+00
50	2.83E-03	8.44E-02	2.68E+00	5.89E-01	5.94E-02	4.84E-04	2.28E-05	3.42E+00
100	2.25E-03	2.50E-02	3.98E-01	5.87E-01	5.90E-02	4.84E-04	2.80E-05	1.07E+00
1,000	3.72E-05	7.80E-12	3.79E-13	5.46E-01	5.17E-02	4.84E-04	2.28E-05	5.98E-01

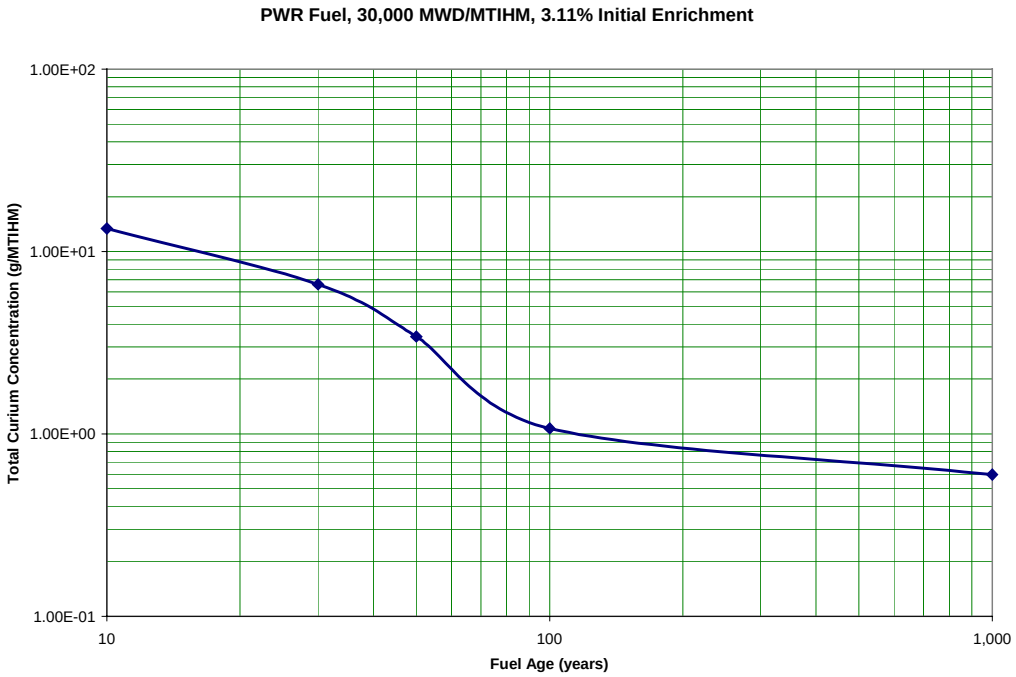

Figure 3.5 Total Curium Concentration in Spent Fuel as a Function of Fuel Age

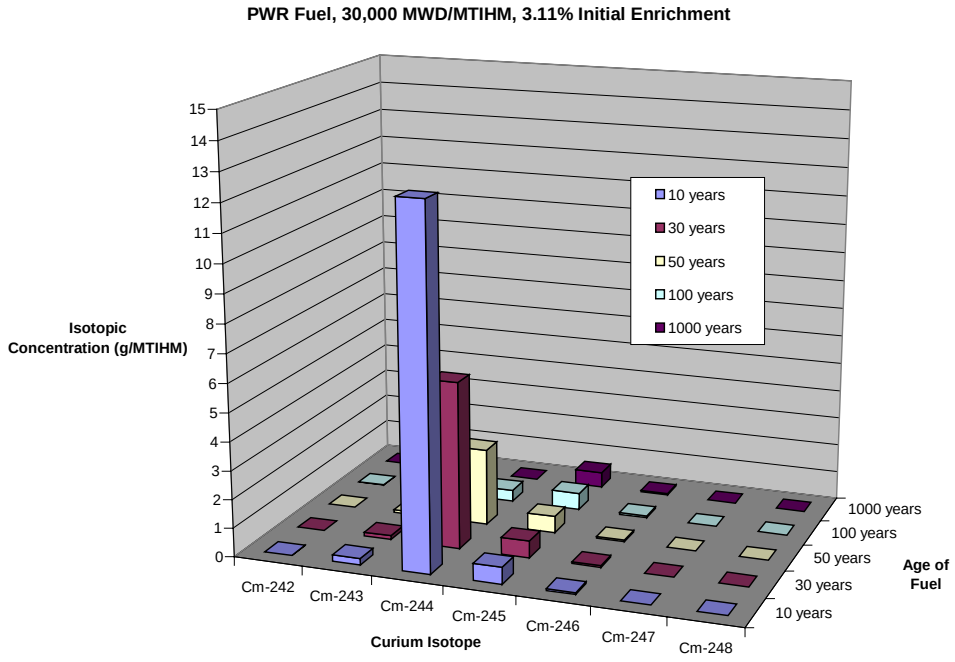

Figure 3.6 Relative Concentration of Curium Isotopes in Spent Fuel As a Function of Fuel Age

The concentration of heat producing Cm-244 decreases rapidly with time, however, highly fissionable Cm-245 concentrations remain steady over 1,000 years. As a fraction of curium, this fissionable isotope increases from 4% to 91% of all curium in a sample over a period of 1,000 years.

In Section 5, the concentration of the various isotopes of curium will be used to assess the ability of curium to sustain a chain reaction. In Section 6, the heat production of curium will be explored based on the isotopic distribution.

4. Fissionability of Curium Isotopes

4.1 Fissionable Curium Isotopes

Spent fuel contains three isotopes of curium that have significant fission cross sections. These include:

- Cm-243, σ_f = 617 barns/atom.
- Cm-245, σ_f = 2145 barns/atom.
- Cm-247, σ_f = 81.9 barns/atom.

The relative concentration of these isotopes change with fuel burnup, initial enrichment, and fuel age. As shown in Figure 4.1, the chief isotope responsible for fission is Cm-245. Cm-243 plays some role, but its shorter halflife (29.1 years) when compared to Cm-245 (8500 years) cause its importance to fission to decrease with fuel age and fuel burnup. Estimates of the critical mass of curium that may lead to sustained fission are documented in the next section.

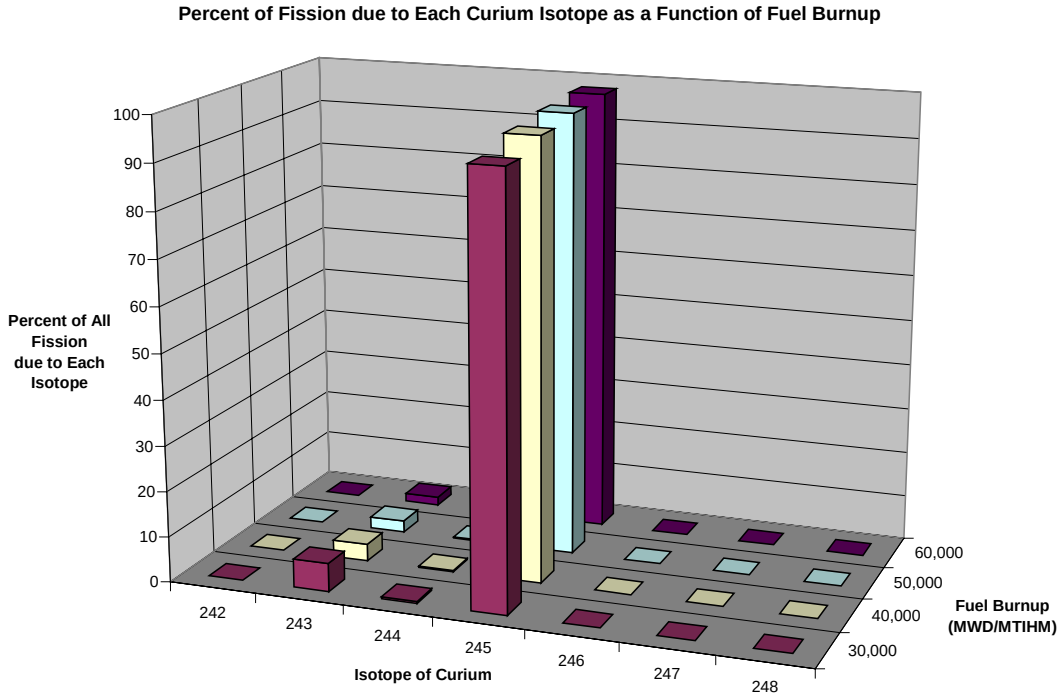

Figure 4.1 Relative Fissionability of Curium Isotopes in Spent Fuel As a Function of Fuel Burnup

5. Critical Geometries and Masses

5.1 Definition of Terms

Three of the curium isotopes in spent nuclear fuel have significant fission cross sections as documented in Section 4. If a large enough mass of curium obtained from spent commercial nuclear fuel is placed in a container, it can lead to a sustained fission reaction. The consequences of sustained fission can be devastating and includes the emission of a large flux of neutrons, gamma rays, heat, other subatomic particles, and the generation of highly radioactive fission products. Sustained fission accidents have occurred in 1945 at the Los Alamos National Laboratory involving a plutonium sphere[8] and, more recently, in Tokaimura, Japan on September 30, 1999 at the JCO Company, Limited uranium conversion facility[9]. There have been 60 reported criticality accidents around the globe since 1945.

To avoid a sustained fission accident or *criticality event*, the accumulation of more than the critical mass of curium or any other fissionable actinide must be avoided. Several criticality assessment codes are available to estimate the propensity of actinides to sustain fission. These include the Monte Carlo neutron transport code SCALE 4.4a[10] developed at the Oak Ridge National Laboratory and MCNP 4C developed at Los Alamos. Both codes have been approved by the Nuclear Regulatory Commission for criticality safety assessment.

Based on the concentration of curium and the geometry and material of its container, the ability of curium to sustain a chain reaction is given in terms of the effective neutron multiplication factor[11], k_{eff}. If k_{eff} is less than 1, the fuel is subcritical and cannot sustain a chain reaction. For k_{eff} 1, the fuel is critical or supercritical and poses a dangerous situation. Any material surrounding the fuel container also has an impact on k_{eff}. Hydrogen-bearing material, including water, paraffin, and plastics, will tend to moderate neutrons and can greatly increase k_{eff}.

For this study, various container geometries and curium concentrations were analyzed using SCALE 4.4a. The input programs, CSPAN and CSAS4, were used to generate the data input files. Output included an estimate of k_{eff} and its statistical uncertainty. The data is presented in a series of plots with the uncertainty expressed as error bars. In most cases, the error bars were too small to be visible on the graphs.

There are two sources of error in using Monte Carlo neutron transport codes to estimate k_{eff}. Since the codes can only handle a finite number of neutrons (typically 50,000 for one simulation), there is a statistical uncertainty introduced by the Monte Carlo method. There is also a small variation between the results of the code and specific criticality experiments referred to as “benchmark tests.” For safety, the NRC requires that k_{eff} be maintained at less than 0.95 when all other sources of error are taken into account. This factor of safety is observed in the critical masses of curium recommended in this report.

Two curium container geometries were considered. First, a spherical shape was assumed for the separated curium. A sphere has minimal surface area for a given volume of curium and its shape minimizes the loss of neutrons through the surface. This is considered the "conservative" or worst case shape. The second shape analyzed was a square cylinder where the diameter and height are equal. This is the conservative case of a cylinder since it minimizes surface area for a given volume.

Both "bare" and "shielded" masses of curium were considered. A bare mass is surrounded by air with no container walls. A shielded mass involves curium placed in a 1/8 inch thick steel wall container. To estimate the extreme or "conservative" case, we also studied the impact of placing curium in water, a significant neutron moderator. Results are explained in the following subsections.

5.2 Critical Bare Sphere

For a spherical mass of curium, the ability of the material to sustain fission increases as the diameter of the sphere increases. Results of the Monte Carlo computer simulations are presented in Table 5.1. A bare sphere becomes critical when its radius exceeds 70 mm corresponding to a mass of over 19 kilograms.

Table 5.1 Bare Curium Sphere Criticality

Bare Curium Sphere			
radius (mm)	k-eff	Volume (cm^3)	Mass (kg)
0	0	0	0.000
40	0.6074	268	3.622
50	0.741	524	7.074
55	0.8123	697	9.415
60	0.8657	905	12.224
65	0.9217	1150	15.541
70	0.9818	1437	19.411
75	1.0398	1767	23.874
80	1.0937	2145	28.974
90	1.1964	3054	41.255

5.3 Critical Shielded Sphere

By adding a 1/8" thick steel liner to the outside of the curium sphere, the behavior of a container wall can be modeled. The changes to critical mass are small as shown in Table 5.2. The steel wall acts as a neutron reflector and slightly decreases the mass of curium necessary to form a critical mass.

Table 5.2 Shielded Curium-Filled Sphere

Shielded Curium Sphere			
radius (mm)	k-eff	Volume (cm^3)	Mass (kg)
0	0	0	0
45	0.6832	382	5.157
50	0.7497	524	7.074
55	0.8123	697	9.415
60	0.8705	905	12.224
65	0.9325	1150	15.541
70	0.9926	1437	19.411
75	1.0431	1767	23.874
80	1.1046	2145	28.974
85	1.1545	2572	34.754
90	1.2105	3054	41.255

5.4 Bare Curium Sphere in Water

Hydrogen serves as a moderator of neutrons and its presence has a significant impact on critical mass. During collisions with hydrogen nuclei, neutrons tend to loose quite a bit of energy per collision with minimal chance of being lost through absorption. The most common source of hydrogen during an accident involving curium will likely be through immersion in water. As seen in Table 5.3, if a curium sphere is placed in water, its critical mass decreases to 7 kilograms corresponding to a radius of 50 mm.

Table 5.3 Curium Sphere Immersed in Water

Bare Curium Sphere in Water			
radius (mm)	k-eff	Volume (cm^3)	Mass (kg)
0	0	0	0.000
40	0.84	268	3.622
42.5	0.88	322	4.344
45	0.93	382	5.157
47.5	0.965	449	6.065
50	1	524	7.074
52.5	1.04	606	8.189
55	1.075	697	9.415
57.5	1.105	796	10.758
60	1.14	905	12.224

5.5 Critical Bare Cylinder

The same studies were conducted on a square cylinder (D = H). It is more likely that curium would be stored in a cylindrical container and a square cylinder will minimize neutron leakage through its walls. For a bare curium cylinder, 23 kilograms of curium will cause criticality corresponding to D = 130 mm. Results are included in Table 5.4.

5.6 Critical Shielded Cylinder

With 1/8" shielding, the critical mass changes slightly as shown in Table 5.5.

Table 5.5 Shielded Square Cylinder of Curium

Shielded Curium Cylinder			
radius (mm)	k-eff	Volume (cm^3)	Mass (kg)
40	0.6779	402	5.433
45	0.7526	573	7.735
50	0.8269	785	10.611
55	0.8944	1045	14.123
60	0.9593	1357	18.335
65	1.0207	1726	23.312
70	1.0851	2155	29.116
75	1.1407	2651	35.811
80	1.1988	3217	43.462

5.7 Bare Cylinder in Water

A curium cylinder immersed in water will have a critical mass of approximately 7 kilograms corresponding to a diameter of 86 mm. Once again, water has a very significant impact on critical mass. Table 5.6 contains the results of these SCALE 4.4a runs.

Table 5.6 Curium Cylinder Placed in Water

Bare Curium Cylinder in Water			
radius (mm)	k-eff	Volume (cm^3)	Mass (kg)
30	0.73	170	2.292
32.5	0.79	216	2.914
35	0.84	269	3.639
37.5	0.89	331	4.476
40	0.94	402	5.433
42.5	0.985	482	6.516
45	1.03	573	7.735
47.5	1.07	673	9.097
52.5	1.15	909	12.283

5.8 Summary of Criticality Data

Figures 5.1 and 5.3 contain provide information on the values of the effective neutron multiplication factor, keff, as a function of sphere radius and square cylinder radius. Figures 5.2 and 5.4 represent the same information in terms of the mass of curium.

Figure 5.1 compares values for bare curium spheres, shielded curium spheres, and a bare sphere immersed in water. A curium sphere placed in air will experience a criticality event if the radius of the sphere exceeds 71 mm or includes more than 20 kg of curium. To maintain safe level of operation, the conservative values based on a sphere immersed in water should be employed. To maintain keff < 0.95, the maximum sphere radius must not exceed 46 mm corresponding to 5.2 kg of curium.

Figures 5.3 and 5.4 present data for a square cylinder. The maximum safe cylinder radius is 40 mm and a height of 80 mm corresponding to 5.1 kg of curium.

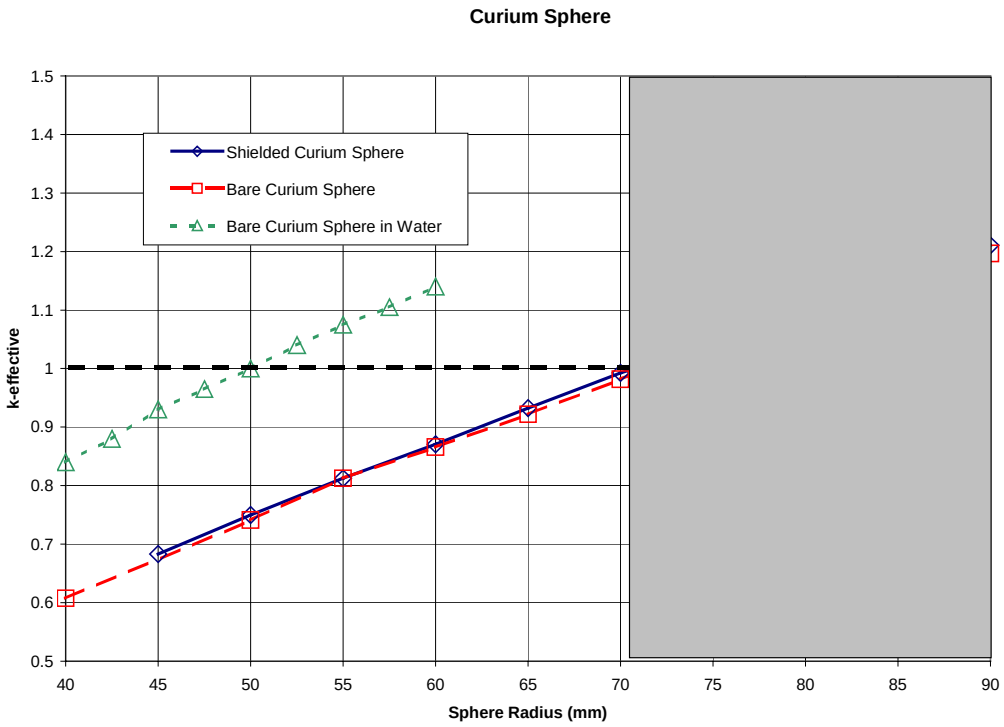

Figure 5.1 Curium-Filled Sphere Criticality As a Function of Sphere Radius

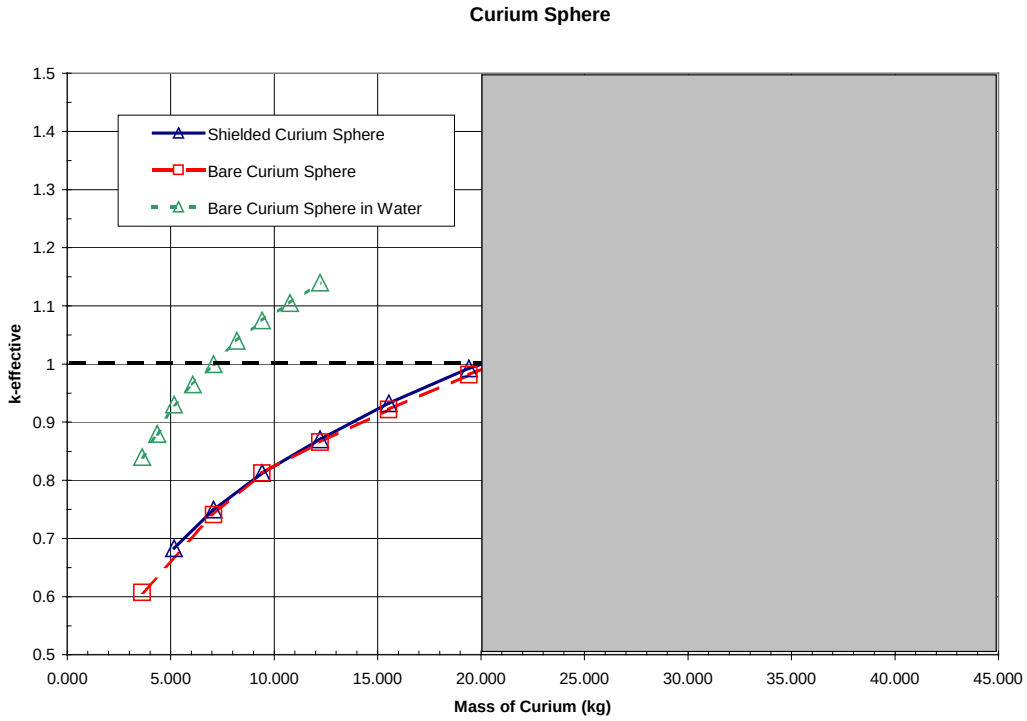

Figure 5.2 Curium-Filled Sphere Criticality As a Function of Curium Mass

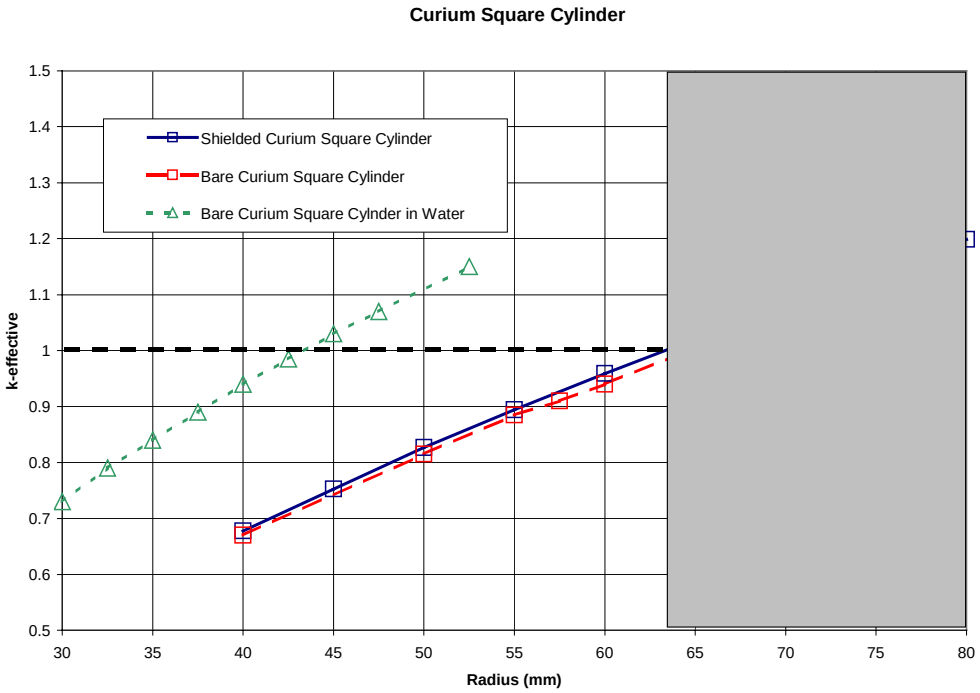

Figure 5.3 Curium-Filled Square Cylinder Criticality As a Function of Cylinder Radius

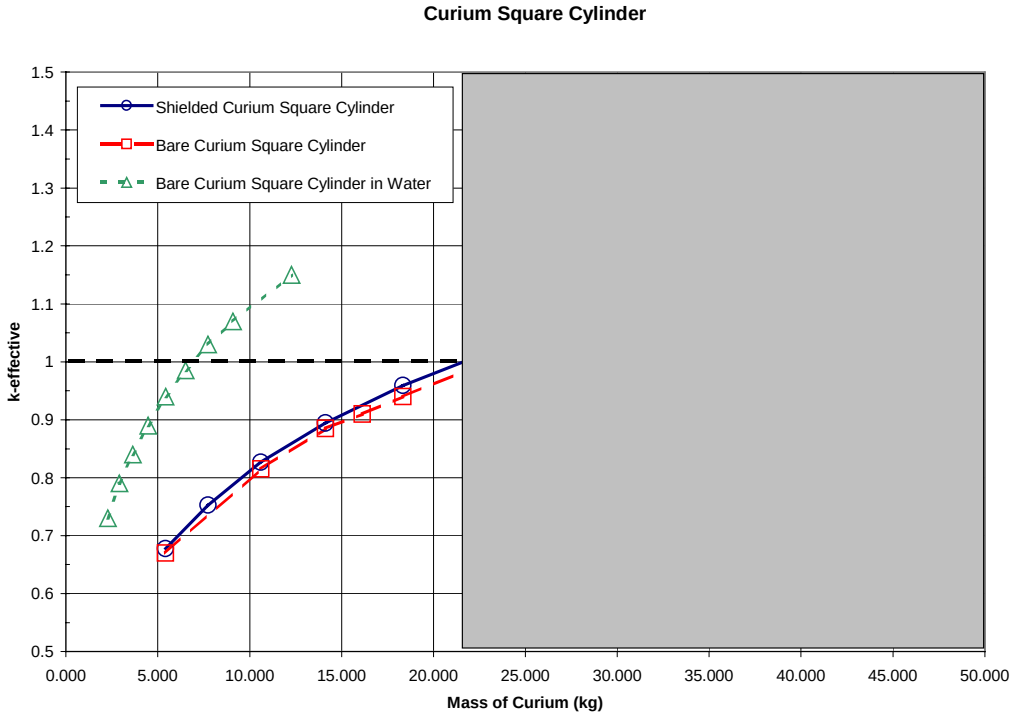

Figure 5.4 Curium-Filled Square Cylinder Criticality As a Function of Curium Mass

6. Decay Heat Generation

6.1 Sources of Heat

Curium poses special problems due to the amount of heat generated by radioactive decay. Curium sources have been employed as an energy source for use in space or in other remote applications with a heat generation rate of up to 10 watts per gram of curium. The heat generation rate of curium in spent nuclear fuel varies with its isotopic abundance. This varies with fuel burnup, initial fuel enrichment, and the time that the spent fuel has been out of the reactor. Tables of heat generation are listed in Appendix B for burnup of 20 to 40 GWd/MTIHM.

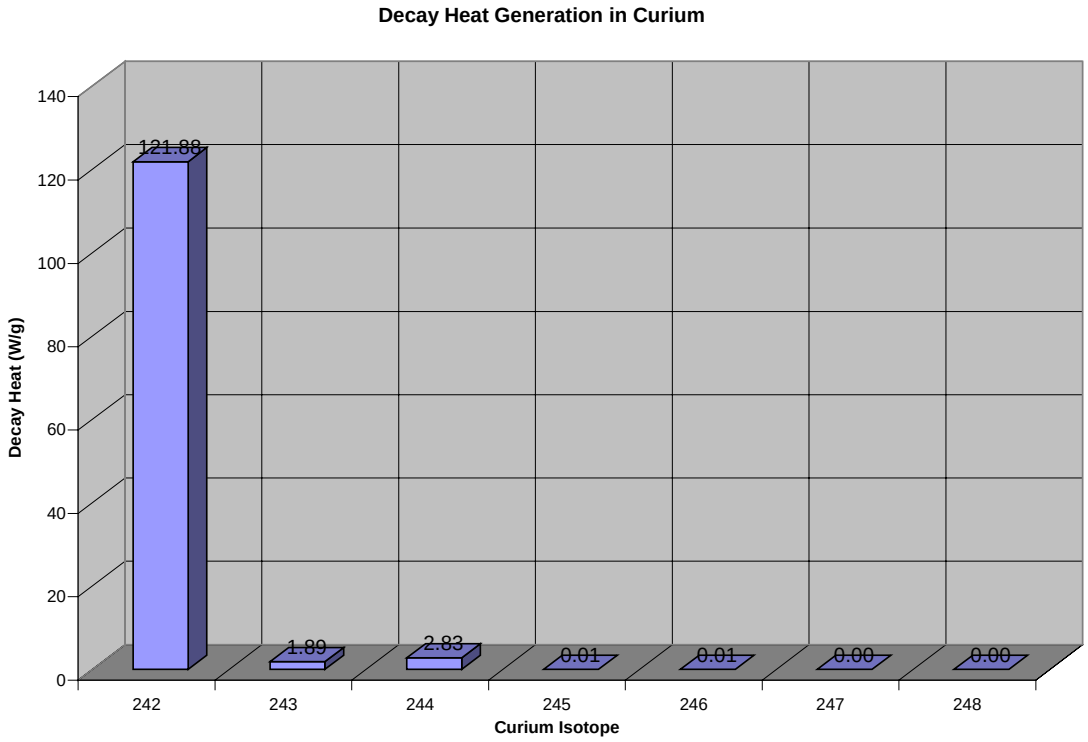

Figure 6.1 Decay Heat Generation by the Isotopes of Curium Found in Spent Nuclear Fuel

The thermal output varies significantly by isotope as shown in Figure 6.1. The highest thermal output occurs within Cm-242 at 122 W/g. Cm-244, however, also generates a significant amount of heat at 2.83 W/g. The heat generated by curium in spent fuel is also affected by the relative abundance of each of these isotopes. Figure 6.2 shows the relative abundance. Although the relative abundance changes with fuel burnup, age, and initial enrichment, the largest producer of heat is Cm-244 representing over 88% of all curium present in spent fuel.

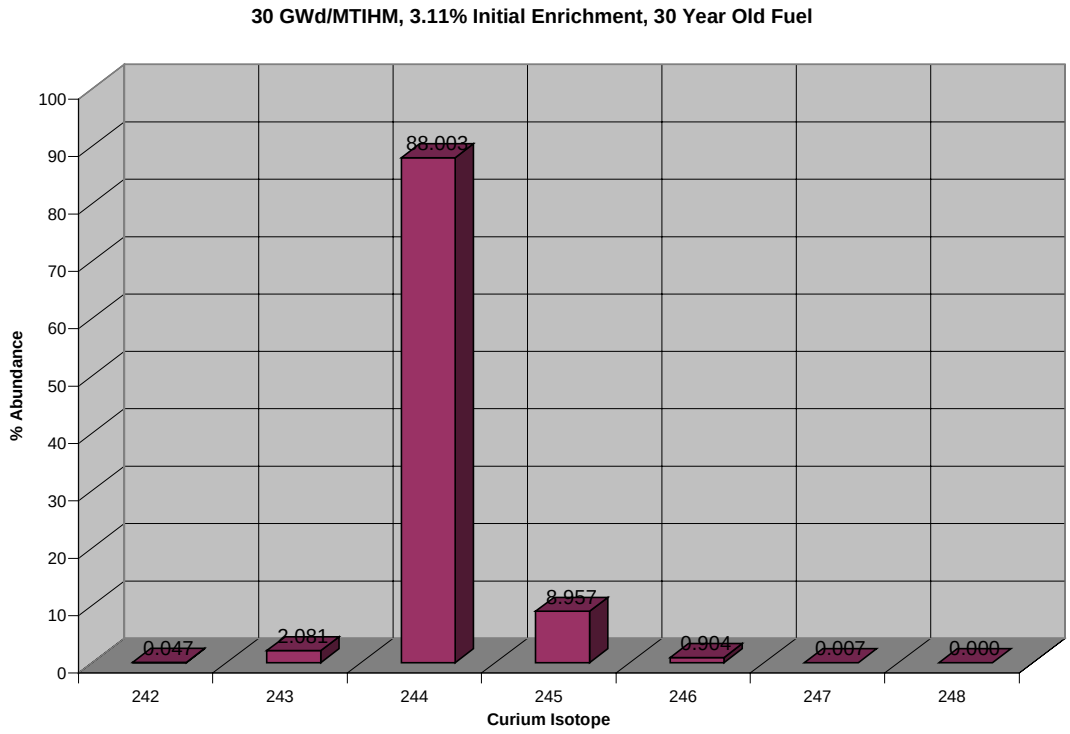

Figure 6.2 Relative Abundance of Curium Isotopes within Spent Nuclear Fuel

The total heat generation with a sample of curium varies from 2.37 to 2.69 watts per gram for fuel burnup that ranges from 20 to 40 GWd/MTIHM. Appendix B shows variation in heat output as a function of burnup and fuel age.

Decay heat generation presents significant problems if curium is to be separated from other minor actinides. The melting temperature of 1613.15 K can be exceeded rather quickly if kilogram quantities of curium are stored. A heat transfer analysis of curium containers can be used to determine the temperatures that will be attained and whether melting will occur.

6.2 Heat Transfer Analysis

The heat generated by decay within a container of curium will be lost by heat transfer from the surface of the container. Eventually, the container will reach an equilibrium temperature where the generated heat will balance the heat lost from the container surface. The "worst case" container is a sphere. For a sphere, the surface available for heat transfer is minimal for a given volume of curium. The analysis of a sphere provides a conservative view of how fast the container will heat up and will overpredict the maximum temperature attained when compared to a cylindrical container.

An analysis of the heat transfer from a container may be based on Figure 6.3. The change in heat stored within the container is equal to the heat generated by decay, the

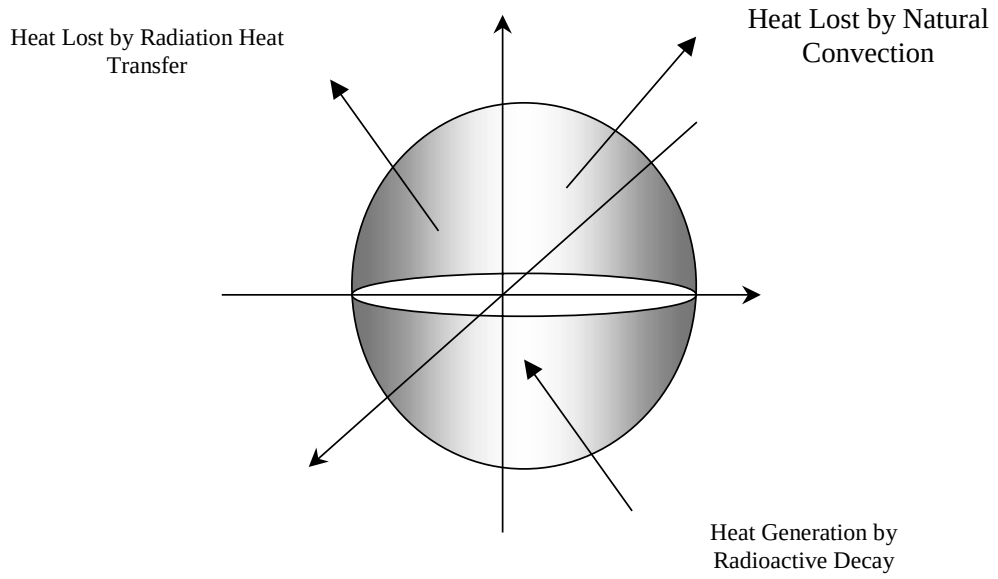

Figure 6.3 Heat Generated and Lost by a Spherical Container

heat lost by radiation heat transfer, and the heat lost by convection heat transfer. This balance is given by equation 6.1.

$$m\, c_p \frac{dT}{dt} = \dot{g}'''V - \sigma\, \varepsilon\, A\,(T^4 - T_\infty{}^4) - h\, A(T - T_\infty) \qquad (6.1)$$

The temperature will rise until radiation heat transfer and convection removes as much heat from the surface of the container as is generated by decay. At this point, the temperature of the curium will reach a maximum or equilibrium temperature.

Convection may be due to a forced air flow over the container or it may be due to the natural currents that arise in stagnant air flow. The second case, natural convection, produces very little heat transfer. Convection heat transfer is defined in terms of the Nusselt number, a dimensionless group that provides the ratio of heat lost by convection to the heat transferred by conduction within the curium. Natural convection from a sphere is given by Churchill[12].

$$\mathrm{Nu}(T) := 2 + \frac{0.589\,\mathrm{Ra}(T)^{\frac{1}{4}}}{\left[1 + \left(\frac{0.469}{\mathrm{Pr}_{\mathrm{air}}(T)}\right)^{\frac{9}{16}}\right]^{\frac{4}{9}}} \qquad (6.2)$$

The transient heat transfer equation (6.1) may be solved through integration, but radiation heat transfer changes as T^4 resulting in a nonlinear ordinary differential equation. Several numerical methods are available for solving this equation, including the 4th order Runge-Kutta method.

A "lumped capacity" analysis was used to analyze this problem. This method assumes that the thermal conductivity of the material within the sphere is very high when compared to the heat removal rate by convection and radiation heat transfer. The temperature within the curium sphere may then be represented by a single temperature. The lumped capacity analysis is valid if the Biot number[12], a dimensionless group representing the ratio of convection to conduction, is less than 0.1.

$$Bi = h\,(V/A) / k \qquad (6.2)$$

where, h = heat transfer coefficient including the effects of radiation and convection heat transfer.
V = volume of the sphere.
A = surface area of the sphere.
k = thermal conductivity of curium within the sphere.

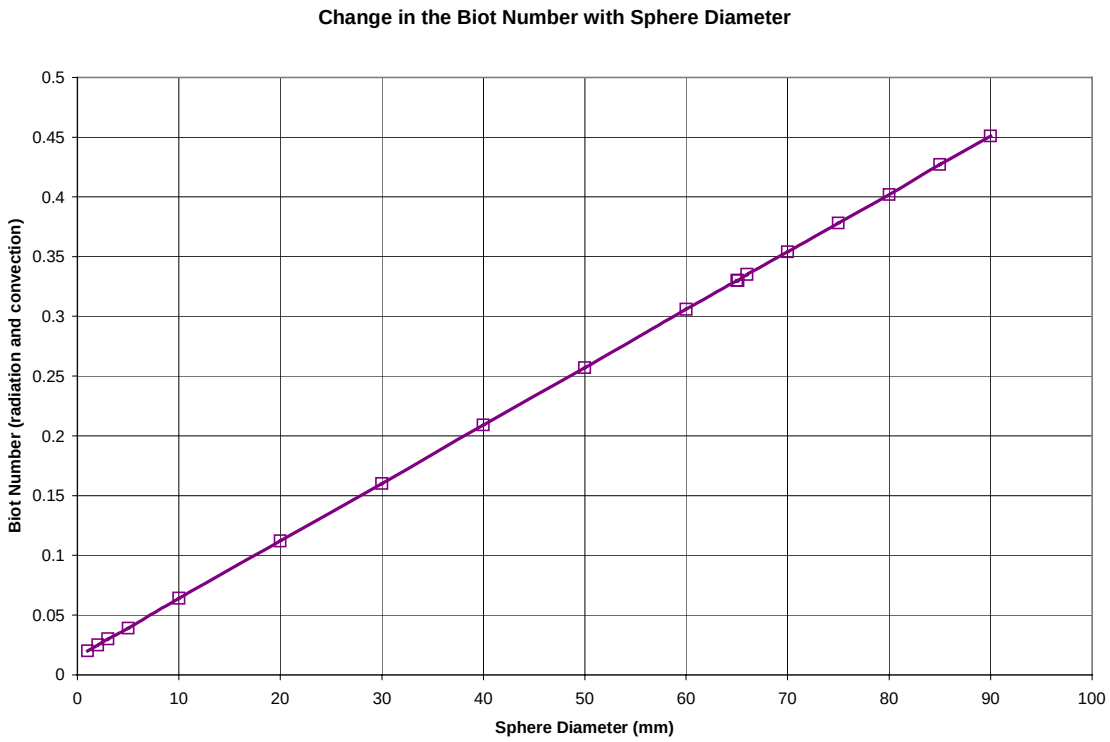

Figure 6.4 Change in the Biot Number with Varying Sphere Diameter

The Biot number was analyzed for a wide range of sphere diameters up to the melting temperature of curium. The properties of the air surrounding the sphere were based on the film temperature surrounding the sphere. The results are shown in figure 6.4.

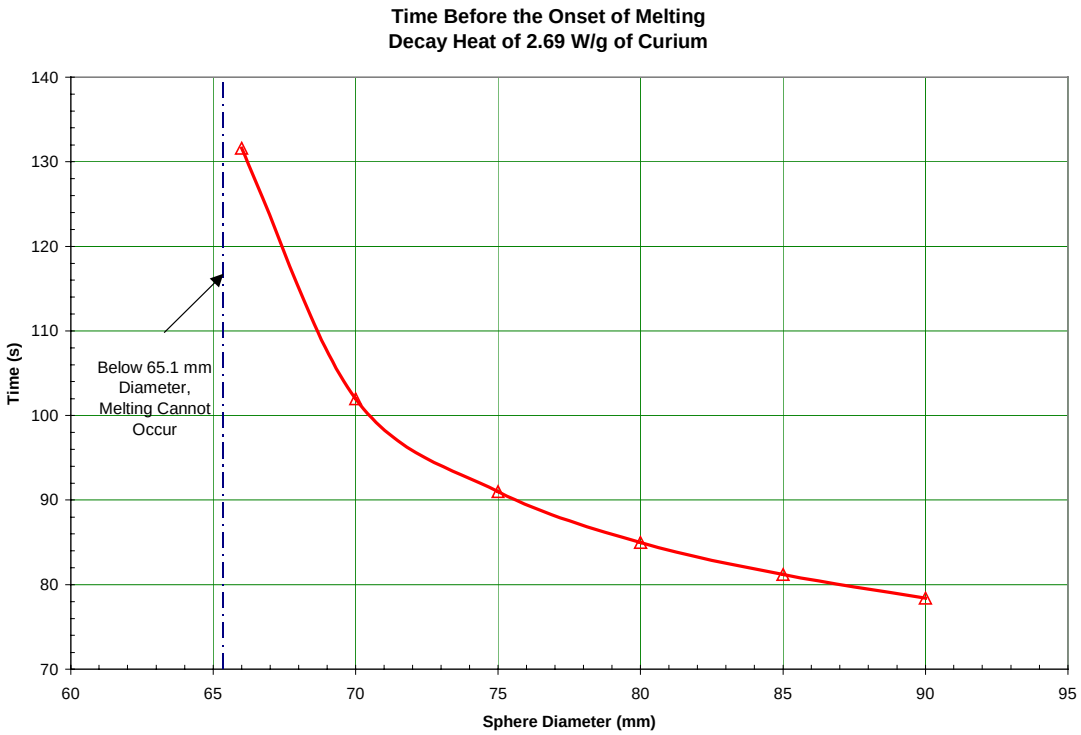

Figure 6.5 Time Required Before Onset of Melting in a Curium Sphere

The data in Figure 6.4 shows that the lumped capacity analysis is valid for spheres of less than about 20 mm up to the melting temperature of curium. The method will have some validity up to the diameter of a sphere that constitutes a critical mass of curium.

6.3 Heat Transfer Results

The analysis is based on an initial temperature of 20°C with an ambient air temperature of 20°C. The heat generation rate of 2.69 watts per gram of curium was used. A mathematical spreadsheet program was written in MathCad 2001 Professional[13] to calculate the equilibrium temperatures and time as well as the time required to begin melting the curium within the sphere. A copy of the MathCad program is attached in Appendix D. MathCad contains built-in routines to numerically solve ordinary differential equations. The Runge-Kutta method was used for this problem. The MathCad program also contains plots of various temperature-dependent properties for air.

Table 6.1
Heat Transfer Data for 2.69 W/g of Curium

Radiation and Convective Heat Transfer from a Curium Sphere					
Decay Heat Generation = 2.69 W/g of Curium					
Sphere Diameter (mm)	Sphere Mass (kg)	Equilibrium Temperature (K)	Time to Equilibrium (s)	Time to Melt (s)	Biot Number at Melting
90	5.233	1756.9	179.8	78.4	0.451
85	4.409	1730.9	176.8	81.2	0.427
80	3.675	1703.6	173.8	85	0.402
75	3.028	1674.9	170.6	91	0.378
70	2.462	1644.6	167.4	102	0.354
66	2.064	1619.1	164.6	131.6	0.335
65.1	1.981	1613.1	164		0.33
65	1.971	1612.5	163.8		0.33
60	1.551	1578.1	160.2		0.306
50	0.897	1501.3	152.2		0.257
40	0.459	1409.4	143		0.209
30	0.194	1292.4	132.4		0.16
20	0.057	1124.9	119		0.112
10	0.007179	807.8	92.4		0.064
5	8.97E-04	508.6	42.4		0.039
3	1.94E-04	389.5	16.6		0.03
2	5.74E-05	342.9	7.4		0.025
1	7.18E-06	309	1.6		0.02

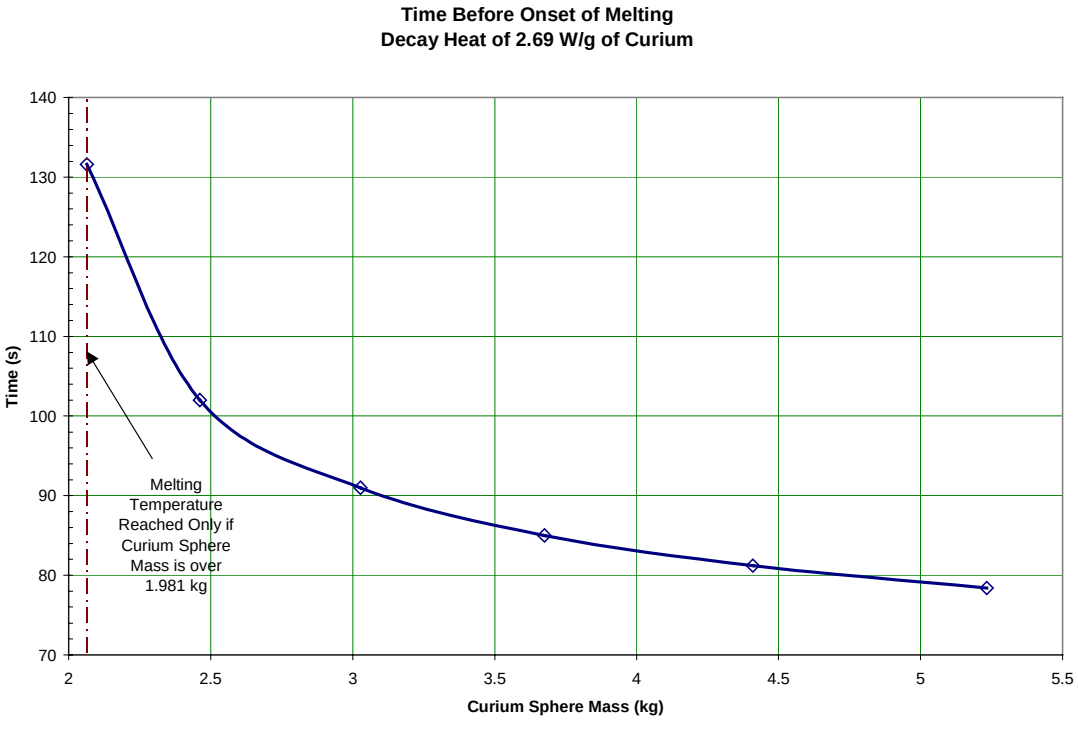

Figure 6.6 Time Required to Attain Equilibrium Temperature

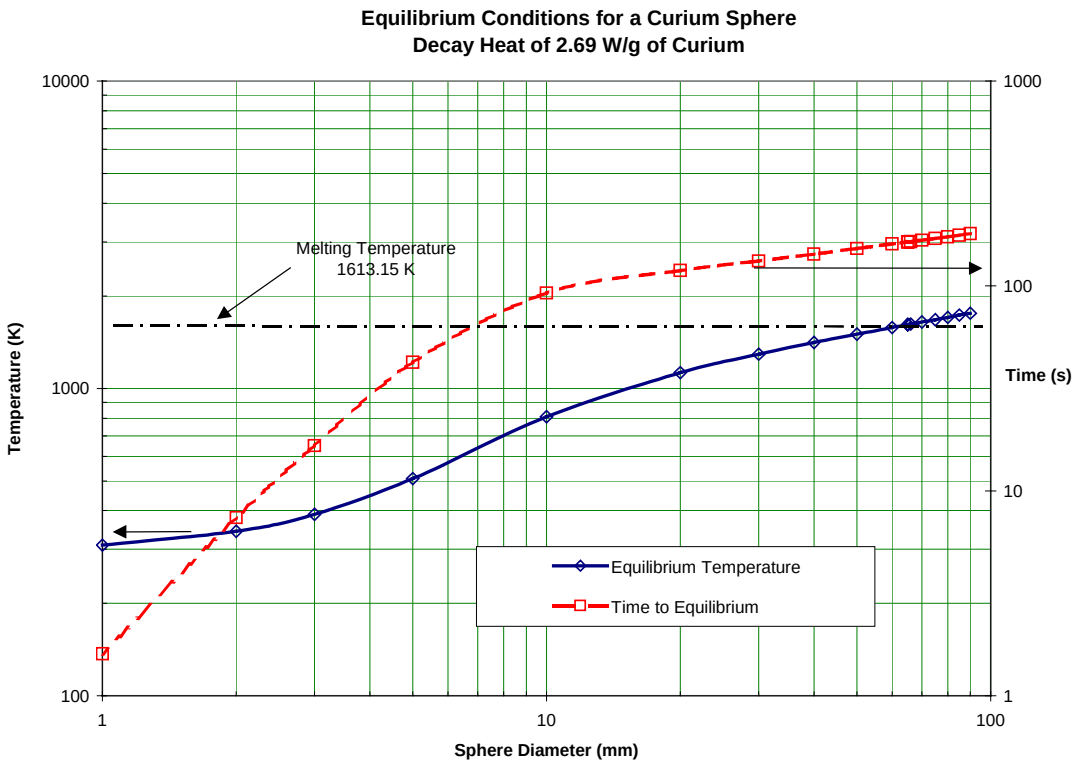

Figure 6.7 Equilibrium Temperature of a Curium Sphere

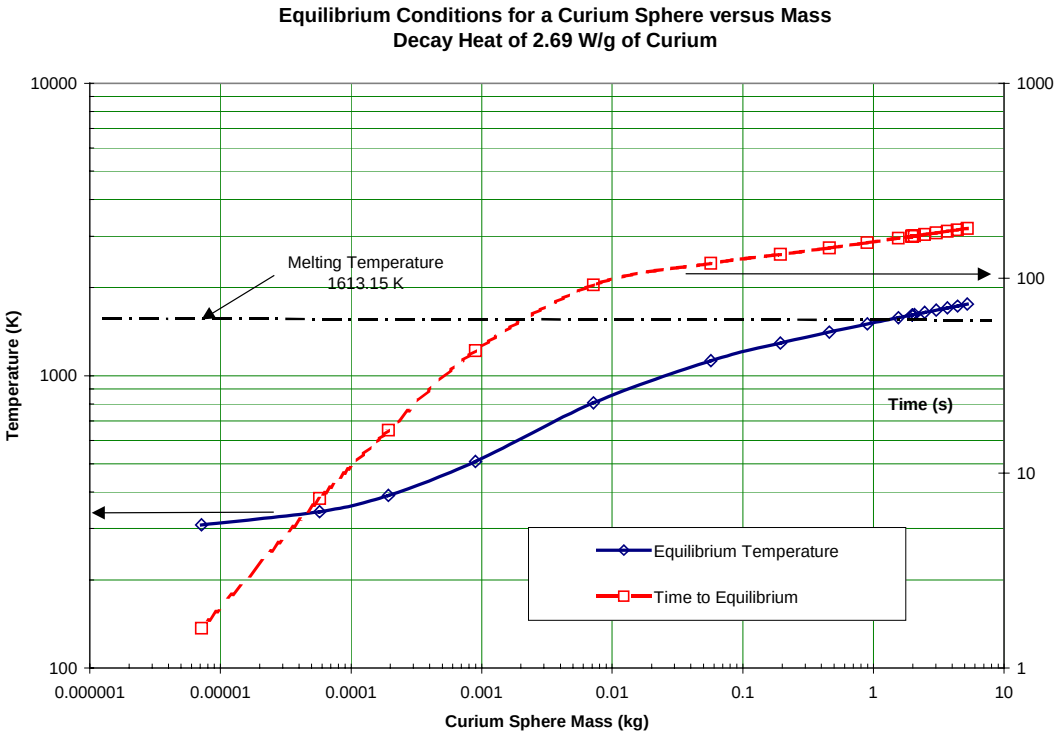

Figure 6.8 Equilibrium Temperature as a Function of Sphere Mass

The time that it takes for curium spheres of various sizes to begin to melt is shown in Figure 6.5. If the diameter of a curium sphere is less than 65.1 mm or 1.981 kg, the sphere will not reach the melting temperature.

Other heat transfer data is shown in Table 6.1. Figure 6.6 provides data on the time that it takes for the equilibrium temperature to be reached. This is dependent upon sphere radius.

7. Conclusions

7.1 Recommendations on Critical Mass

To avoid the accumulation of a critical mass of curium, it is recommended that no more than 5.0 kg of curium from spent fuel be allowed to accumulated in one container. This corresponds to a sphere of 90 mm diameter or a 80 mm high, 80 mm diameter cylinder. The 5.0 kg limit meets the NRC requirement that k_{eff} remain less than 0.95 after numerical uncertainties in the Monte Carlo code are incorporated.

Care must be taken to avoid placing materials containing much hydrogen in the vicinity of significant quantities of curium. This includes water, plastics, acrylic, hydrocarbon-based solvents, and paraffin wax.

7.2 Recommendations on Thermal Output

Isotopes of curium generate a significant amount of heat through radioactive decay. This thermal generation can lead to high temperatures within samples of spent fuel curium. The equilibrium temperature is a function of the container geometry, insulation, and the air flow past the container. As a conservative case, a spherical mass of curium was analyzed where radiation and natural convection heat transfer were the only mechanisms removing heat from the curium. If the diameter of a curium sphere is less than 65.1 mm or 1.981 kg in mass, the sphere will not reach the melting temperature.

Thermal considerations require a smaller mass of curium than criticality and becomes the limiting case. The mass of spent fuel curium should not be allowed to exceed 1.981 kg.

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Appendix A

Sample SCALE 4.4a Input and Output Files

A sample input file is included below. The CSPAN preprocessor was used in the analysis. Each run took approximately 15 seconds on a 1.8 GHz PC with 512 megabytes of memory.

```
=csas25
bil1.inp
44groupndf5 infhommedium
cm-242 1 0  .0000158     293   end
cm-243 1 0  .000697     293   end
cm-244 1 0  .0293     293   end
cm-245 1 0  .00297     293   end
cm-246 1 0  .000299     293   end
cm-247 1 0  2.42E-06     293   end
cm-248 1 0  1.13E-07     293   end
ss316s    3  1  293.  42092  14.23   42094  9.053
          42095  15.748  42096  16.673  42097  9.646
          42098  24.623  42100  10.028  end
 end comp
cellmix 1
bil1.inp
read geom
'
unit 1
com='bil1.inp'
cylinder  1 1  .5            .25          -.25          origin 00 0
cuboid    0 1  200 -200 200 -200 200 -200
 end geom
end data
end
```

Appendix B

Decay Heat Generation by Isotopes of Curium

20 GWD/MTIHM				
2.44% initial enrichment				
30 year old fuel				
Curium	Heat	% Heat	Mass	Heat/mass
Isotope	Output	Output		
	(W/MTIHM)		(g/MTIHM)	(W/g)
242	1.97E-01	3.900175	1.62E-03	1.22E+02
243	9.31E-02	1.841705	4.92E-02	1.89E+00
244	4.77E+00	94.24103	1.68E+00	2.83E+00
245	7.70E-04	0.015229	1.35E-01	5.70E-03
246	9.40E-05	0.00186	9.35E-03	1.01E-02
247	1.61E-10	3.19E-09	5.43E-05	2.97E-06
248	9.49E-10	1.88E-08	1.79E-06	5.29E-04
Total (Cm)	5.056184	100	1.878269	2.69E+00

30 GWD/MTIHM				
3.11% initial enrichment				
30 year old fuel				
Curium	Heat	% Heat	Mass	Heat/mass
Isotope	Output	Output		
	(W/MTIHM)		(g/MTIHM)	(W/g)
242	0.3777	2.213748	3.10E-03	1.22E+02
243	0.2599	1.523307	1.37E-01	1.89E+00
244	16.42	96.23971	5.801	2.83E+00
245	3.37E-03	0.019723	5.90E-01	5.70E-03
246	6.00E-04	0.003516	5.96E-02	1.01E-02
247	1.44E-09	8.41E-09	4.84E-04	2.96E-06
248	1.21E-08	7.07E-08	2.28E-05	5.29E-04
Total (Cm)	17.06156	100	6.591826	2.59E+00

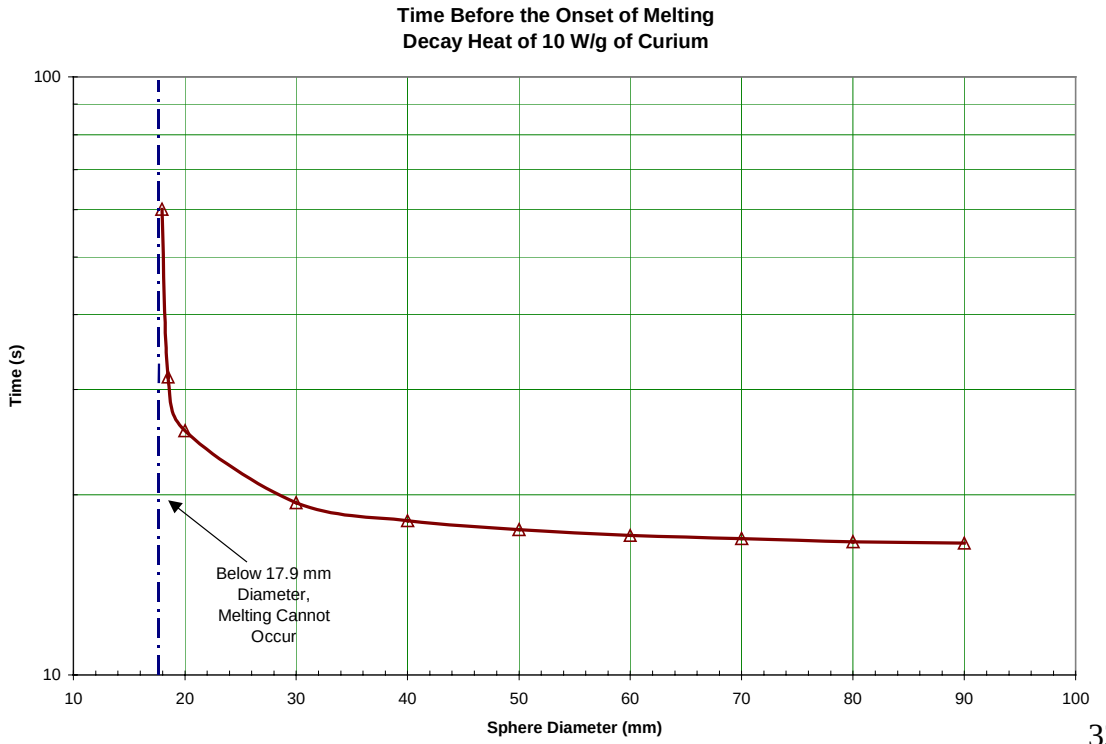
30 GWD/MTIHM				
3.11% initial enrichment				
50 year old fuel				
Curium	Heat	% Heat	Mass	Heat/mass
Isotope	Output	Output		
	(W/MTIHM)		(g/MTIHM)	(W/g)
242	3.45E-01	4.232463	2.83E-03	1.22E+02
243	1.60E-01	1.961565	8.44E-02	1.89E+00
244	7.64E+00	93.7574	2.70E+00	2.83E+00
245	3.36E-03	0.041232	5.89E-01	5.70E-03
246	5.98E-04	0.007341	5.94E-02	1.01E-02
247	1.44E-09	1.76E-08	4.84E-04	2.96E-06
248	1.21E-08	1.48E-07	2.28E-05	5.29E-04
Total (Cm)	8.146557	100	3.434526	2.37E+00

40 GWD/MTIHM				
3.72% initial enrichment				
30 year old fuel				
Curium	Heat	% Heat	Mass	Heat/mass
Isotope	Output	Output		
	(W/MTIHM)		(g/MTIHM)	(W/g)
242	6.92E-01	1.435892	5.68E-03	1.22E+02
243	6.12E-01	1.26888	3.23E-01	1.89E+00
244	4.69E+01	97.26142	1.66E+01	2.83E+00
245	1.35E-02	0.027967	2.37E+00	5.70E-03
246	2.82E-03	0.00584	2.80E-01	1.01E-02
247	9.70E-09	2.01E-08	3.27E-03	2.97E-06
248	1.11E-07	2.31E-07	2.01E-04	5.53E-04
Total (Cm)	48.2	100	19.53675	2.47E+00

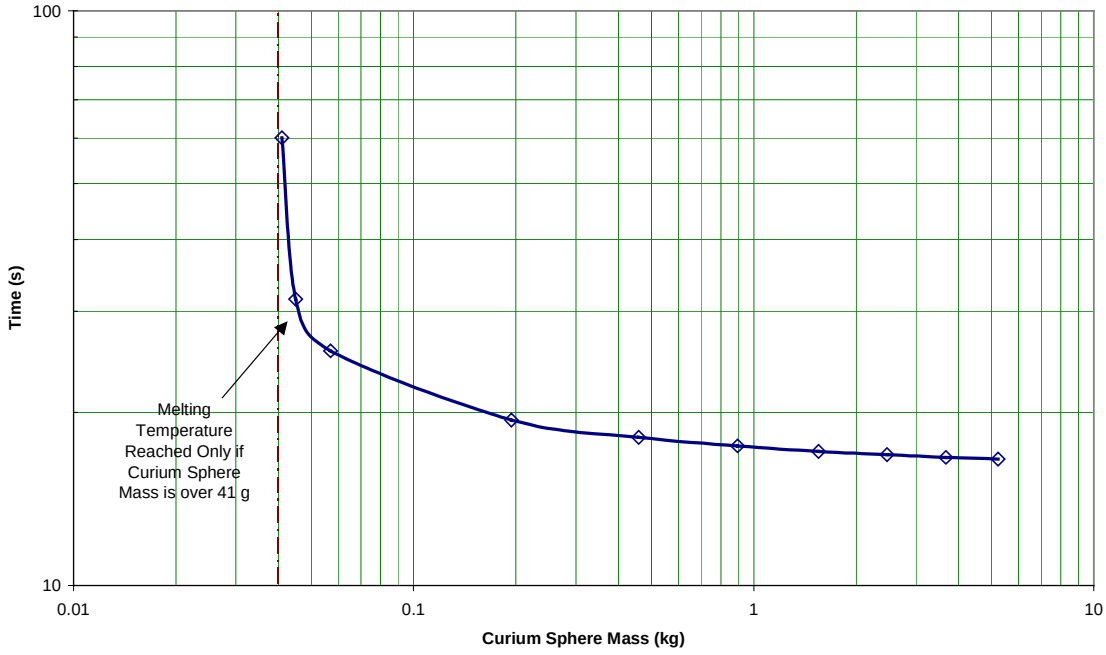
Appendix C

Heat Transfer Analysis for A Curium Sphere with a Generation Rate of 10 W/g

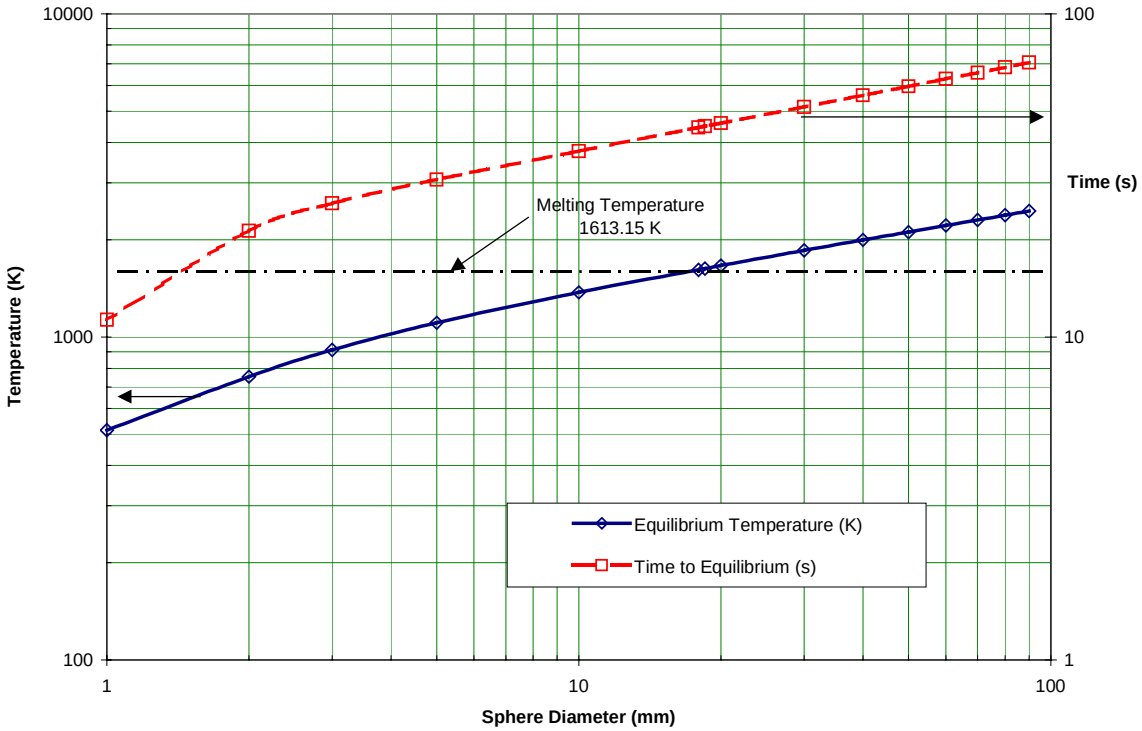
Radiation and Convective Heat Transfer from a Curium Sphere				
Diameter (mm)	Equilibrium Temperature (K)	Time to Equilibrium (s)	Time to Melt (s)	Sphere Mass (kg)
90	2451.4	70.6	16.6	5.233
80	2379.7	68.3	16.7	3.675
70	2300.8	65.7	16.9	2.462
60	2212.8	62.9	17.1	1.551
50	2112.5	59.6	17.5	0.897
40	1995.2	55.9	18.1	0.459
30	1851.9	51.5	19.4	0.194
20	1662.3	45.9	25.6	0.057
18.5	1627.2	44.9	31.5	0.045
17.93	1613.2	44.5	60.1	0.041
10	1373.4	37.6		0.007179
5	1106.6	30.7		0.0008973
3	911.2	25.9		0.0001938
2	754.2	21.3		5.74E-05
1	514.6	11.3		0.000007179

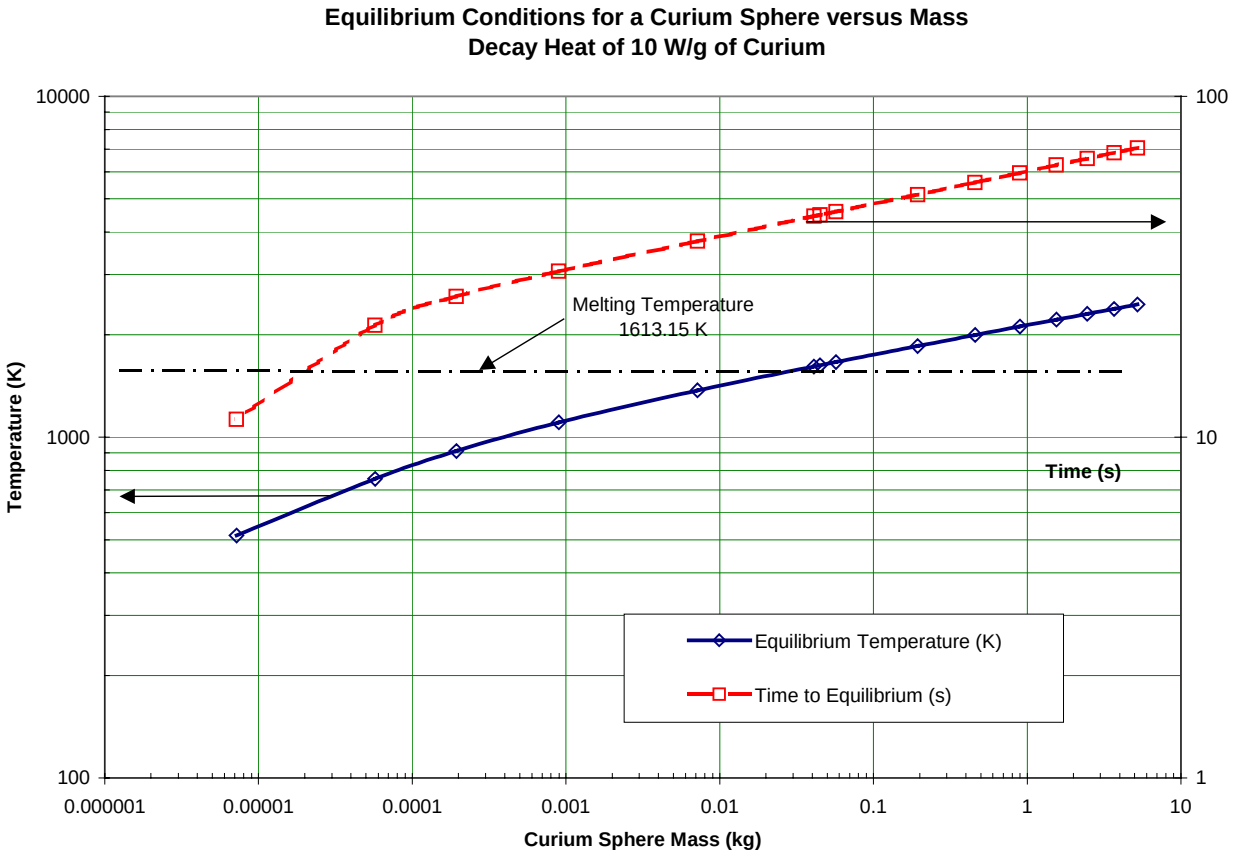


Time Before Onset of Melting
Decay Heat of 10 W/g of Curium



Equilibrium Conditions for a Curium Sphere
Decay Heat of 10 W/g of Curium




Equilibrium Conditions for a Curium Sphere versus Mass
Decay Heat of 10 W/g of Curium
10000
1000
100
100
10
1
Temperature (K)
Time (s)
Melting Temperature
1613.15 K
Equilibrium Temperature (K)
Time to Equilibrium (s)
0.000001
0.00001
0.0001
0.001
0.01
0.1
1
10
Curium Sphere Mass (kg)

Appendix D

MathCad Program

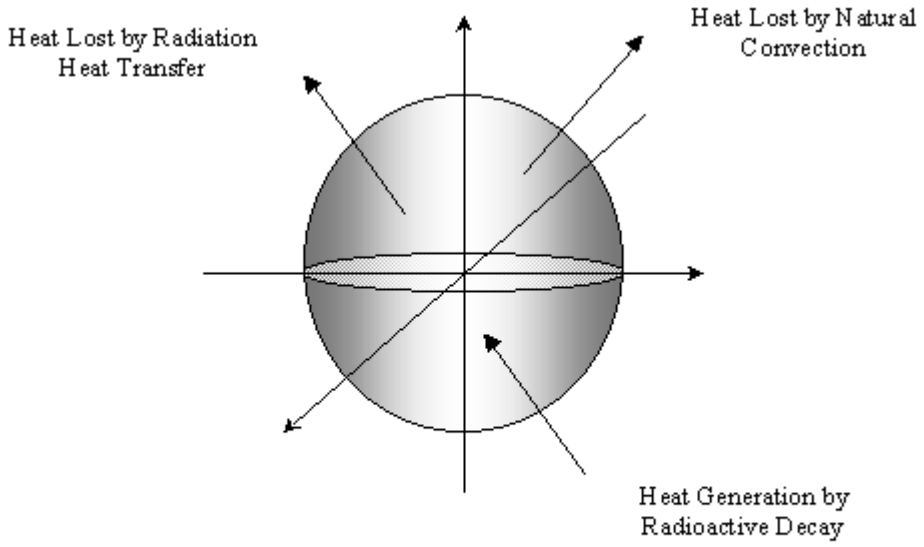
"Transient Temperature of a Curium Sphere using a Lumped Capacity Analysis"

Transient Temperature of a Curium Sphere Using a Lumped Capacity Analysis

Dr. William Culbreth
University of Nevada, Las Vegas

A. Introduction

Curium generates a great deal of decay heat (~ 10 W/g). This property has led to its limited use as an energy source for potential applications in space. In fuel fabrication, the accumulation of excessive quantities of curium can lead to extreme temperature increases within a short period of time. The conservative or "worst case" to study involves curium in the shape of a sphere. A sphere combines maximum volume of material for the minimum possible surface area available for heat transfer to the environment. A sample of curium in the shape of a sphere will loose heat by convection and by radiation heat transfer. In the absence of forced air convection, natural convection plays a limited role in removing heat from a curium sphere.



$$m\,c_p\,\frac{dT}{dt} = \dot{g}'''V - \sigma\,\varepsilon\,A(T^4 - T_\infty^4) - h\,A(T - T_\infty)$$

In this study, the temperature rise in a sphere of curium is analyzed. Both natural convection and radiation heat transfer are used to remove heat from the sphere. A "lumped capacity analysis" is used, as justified through the Biot number graphically presented in section 6b. A 4th order Runge-Kutta routine is used to solve the non-linear ordinary differential equation. The results include a graph of sphere temperature as a function of time, the time it takes to begin melting, the equilibrium temperature, and the time that it takes to reach within 1

B. Calculations

1. Define the Geometry of the Sphere

$D_{sphere} := 90 \cdot mm$ *Sphere diameter.*

$r := \frac{D_{sphere}}{2}$ *Sphere radius.*

2. Define the Properties of Curium

$\rho_{Cm} := 13710 \frac{kg}{m^3}$ *Density of curium.*

$g''' := 2.64 \frac{W}{g} \cdot \rho_{Cm}$ *Thermal generation rate of curium due to radioactive decay.*

$g''' = 3.619 \times 10^7 \frac{W}{m^3}$

$c_{p.Cm} := 120 \cdot \frac{J}{kg \cdot K}$ *Specific heat of curium.*

$k_{Cm} := 10 \cdot \frac{W}{m \cdot K}$ *Thermal conductivity of curium.*

$T_{fusion.Cm} := 1613.15K$ *Melting temperature of curium.*

3. Define the Properties of Air

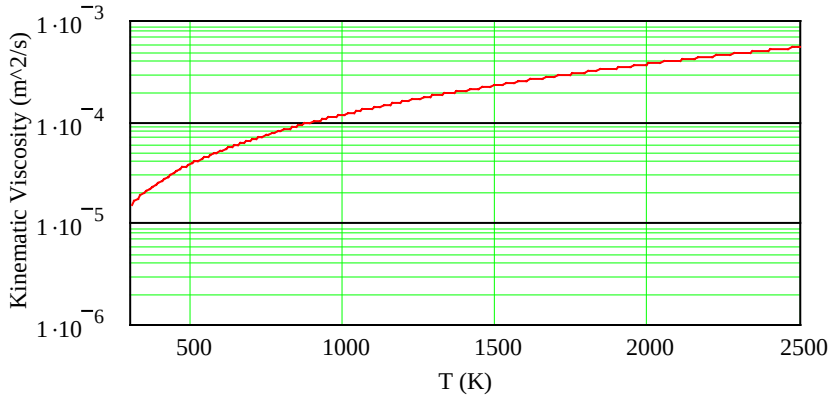
a. Kinematic Viscosity for air.

$a_0 := -10.42734575127361$ $a_1 := 6.740747047260812 \cdot 10^{-2}$

$a_2 := 6.031554524186101 \cdot 10^{-5}$ $a_3 := 5.245158141057499 \cdot 10^{-10}$

$\nu_{air}(u) := \left[\sum_{i=0}^{3} a_i \cdot \left(\frac{u}{1 \cdot K} \right)^i \right] \cdot 10^{-6} \cdot \frac{m^2}{s}$ $T := 300 \cdot K, 310 \cdot K .. 2500 \cdot K$

Plotting limits.

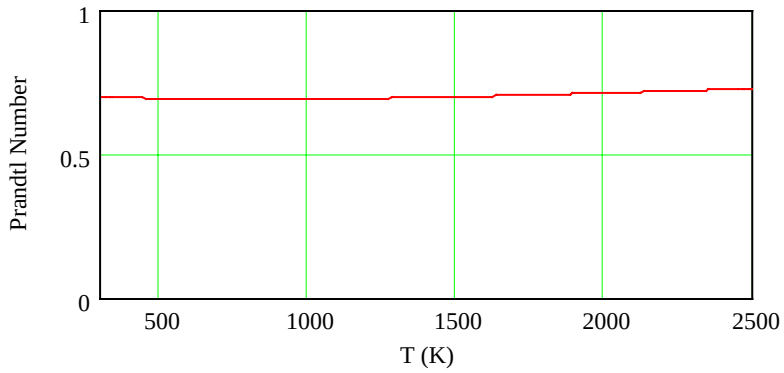


b. Prandtl Number for air.

$b_0 := 0.7082729262465852$ $b_1 := -4.4547541358749184 \cdot 10^{-5}$

$b_2 := 3.2281455976027584 \cdot 10^{-8}$ $b_3 := -4.5920179062775991 \cdot 10^{-12}$

$$Pr_{air}(u) := \left[\sum_{i=0}^{3} b_i \cdot \left(\frac{u}{1 \cdot K} \right)^i \right]$$

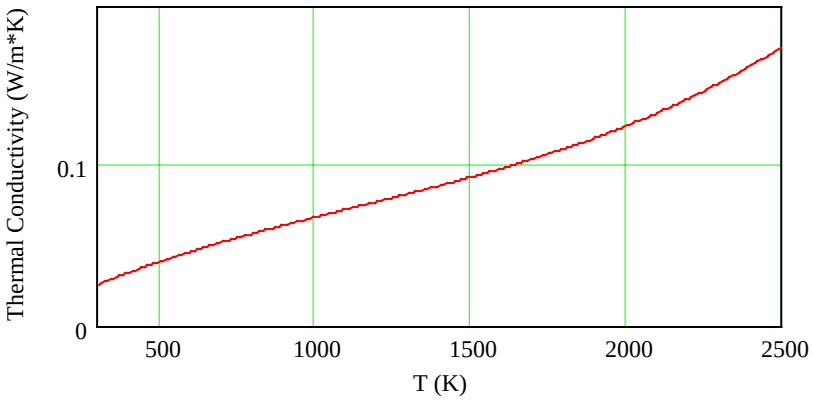


c. Thermal Conductivity for air.

$c_0 := -8.2062994960629966 \cdot 10^{-4}$ $c_1 := 1.0248007248940491 \cdot 10^{-4}$

$c_2 := -4.7123893906600231 \cdot 10^{-8}$ $c_3 := 1.3686233806514731 \cdot 10^{-11}$

$$k_{air}(u) := \left[\sum_{i=0}^{3} c_i \cdot \left(\frac{u}{1 \cdot K} \right)^i \right] \cdot \frac{W}{m \cdot K}$$



d. Expansion Coefficient.

$$beta_{air}(u) := \frac{1}{u}$$

4. Calculate the Mass of Curium in the Sphere

$A := 4 \cdot \pi \cdot r^2$ $\quad A = 0.025 m^2$ *Surface area of the sphere.*

$V := \frac{4}{3} \cdot \pi \cdot r^3$ $\quad V = 3.817 \times 10^{-4} m^3$ *Volume of the sphere.*

$m_{Cm} := \rho_{Cm} \cdot V$ $\quad m_{Cm} = 5.233 kg$ *Mass of curium in the sphere.*

5. Calculate the Heat Transfer Properties

$\varepsilon := 1$ *Emissivity of the surface of the sphere.*

6. Compute the Transient Temperature of the Sphere

a. Define the initial and ambient temperatures.

$$T_{initial} := (20 + 273) \cdot K$$

$$T_{infinity} := (20 + 273) \cdot K$$

b. Define the heat transfer coefficient.

$$Gr(T) := g_{gravity} \cdot beta_{air}(T) \cdot \left(T - T_{infinity}\right) \cdot D_{sphere}^{3} \cdot \frac{1}{\nu_{air}(T)^{2}}$$

Grashof number.

$$Ra(T) := Gr(T) \cdot Pr_{air}(T)$$

Rayleigh number.

$$Nu(T) := 2 + \frac{0.589\, Ra(T)^{\frac{1}{4}}}{\left[1 + \left(\frac{0.469}{Pr_{air}(T)}\right)^{\frac{9}{16}}\right]^{\frac{4}{9}}}$$

Reference from Holman, equation by Churchill, Ra < 10^11 and Pr > 0.5.

$$h(T) := \frac{Nu(T) \cdot k_{air}(T)}{D_{sphere}}$$

Convective heat transfer coefficient.

$$s_{sphere} := \frac{V}{A} \qquad s_{sphere} = 15\,mm$$

This is the effective diameter to be used in calculating the Biot number.

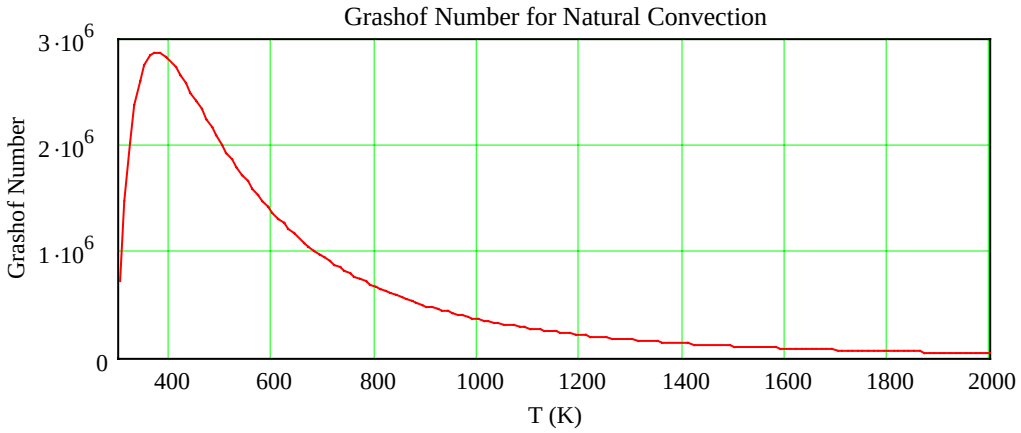
$$Bi(T) := \frac{h(T) \cdot s_{sphere}}{k_{Cm}}$$

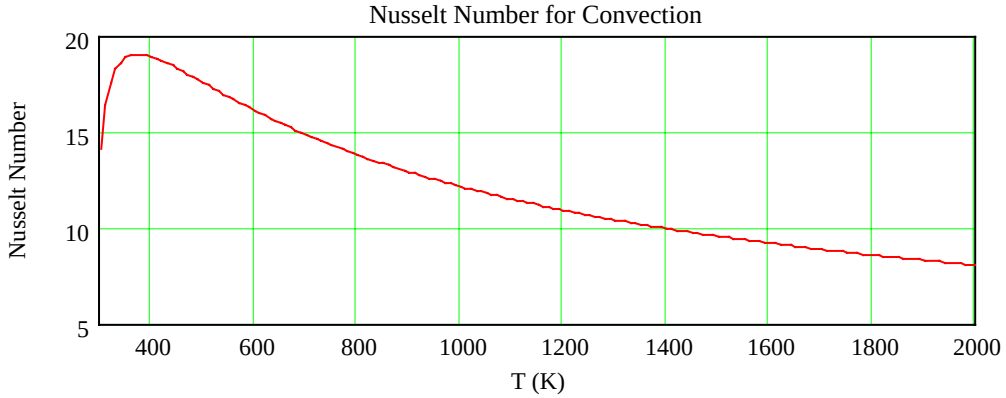
This is the Biot number. It represents the ratio of convected heat transfer to conductive heat transfer within the sphere. If Bi < 0.1.

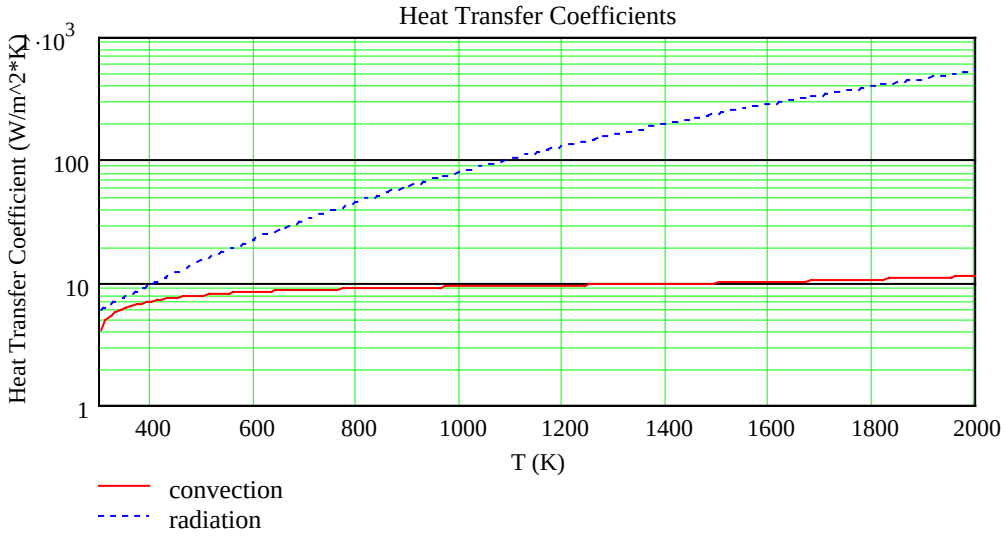
$$h_{radiation}(T) := \sigma \cdot \varepsilon \cdot \left(T^{2} + T_{infinity}^{2}\right) \cdot \left(T + T_{infinity}\right)$$

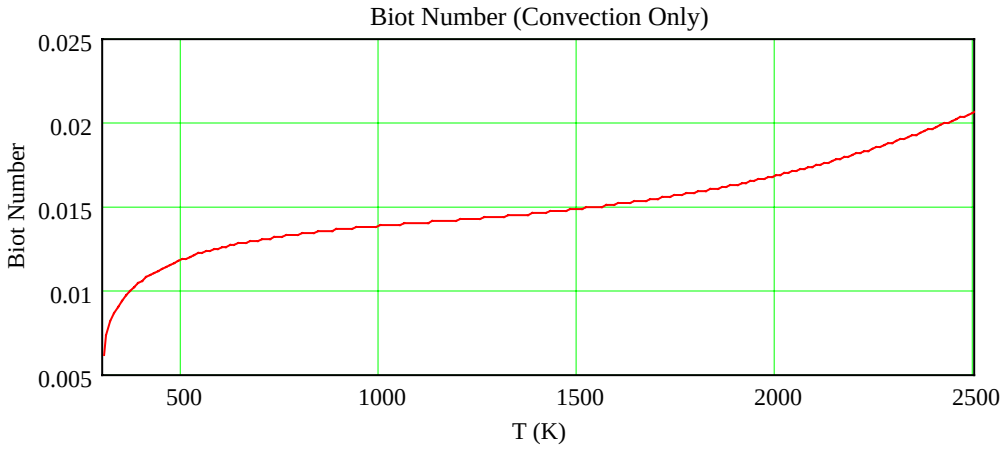
Convection is assisted by radiation heat transfer. This can be written approximately in terms of a "convection" coefficient for radiation. A combined Biot number is also computed.

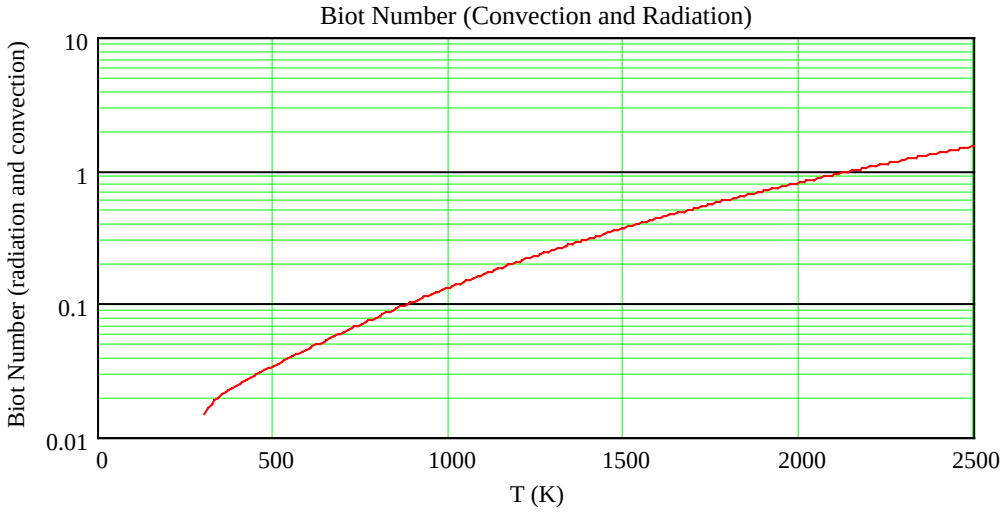
$$Bi_{radiation.and.convection}(T) := \frac{\left(h(T) + h_{radiation}(T)\right) \cdot s_{sphere}}{k_{Cm}}$$


Grashof Number for Natural Convection
Grashof Number
3·10^6
2·10^6
1·10^6
0
400
600
800
1000
1200
1400
1600
1800
2000
T (K)


Nusselt Number for Convection
Nusselt Number
20
15
10
5
400
600
800
1000
1200
1400
1600
1800
2000
T (K)


Heat Transfer Coefficients
Heat Transfer Coefficient (W/m^2*K)
·10^3
100
10
1
400
600
800
1000
1200
1400
1600
1800
2000
T (K)
convection
radiation





c. Define the differential equation for temperature as a function f time.

1) Set up the initial conditions.

$$T := T_{initial}$$

$$time_{stop} := 100 \cdot s$$

2) Analyze the case of an adiabatic sphere, first.

$$dTdt_{generated}(u) := \frac{g''' \cdot V}{m_{Cm} \cdot c_{p.Cm}}$$

$q_{generated}(T) := g'''\cdot V$ *Heat generated in watts.*

$D_{adiabatic}(t,u) := dTdt_{generated}(u)$

$answer_{adiabatic} := rkfixed\left(T_{initial}, 0\cdot s, time_{stop}, 1000, D_{adiabatic}\right)$

$t := answer_{adiabatic}^{\langle 0\rangle}$

$T_{adiabatic} := answer_{adiabatic}^{\langle 1\rangle}$

3) Next, add in the effect of radiation heat transfer.

$$dTdt_{radiation}(u) := \frac{\sigma\cdot\varepsilon\cdot A}{m_{Cm}\cdot c_{p.Cm}}\cdot\left(u^4 - T_{infinity}^4\right)$$

$q_{radiation}(T) := \sigma\cdot\varepsilon\cdot A\cdot\left(T^4 - T_{infinity}^4\right)$ *Heat radiated away from the sphere.*

$D_{radiation}(t,u) := dTdt_{generated}(u) - dTdt_{radiation}(u)$

$answer_{radiation} := rkfixed\left(T_{initial}, 0\cdot s, time_{stop}, 1000, D_{radiation}\right)$

$T_{radiation} := answer_{radiation}^{\langle 1\rangle}$

4) Now, combine the effects of radiation and convective heat transfer.

$$dTdt_{convection}(u) := \frac{h\left(\frac{u + T_{infinity}}{2}\right)\cdot A}{m_{Cm}\cdot c_{p.Cm}}\cdot\left(u - T_{infinity}\right)$$

$q_{convection}(T) := h\left(\frac{T + T_{infinity}}{2}\right)\cdot A\cdot\left(T - T_{infinity}\right)$ *Heat convected from the sphere.*

$D_{conv.and.rad}(t,u) := dTdt_{generated}\left(u_0\right) - dTdt_{radiation}\left(u_0\right) - dTdt_{convection}\left(u_0\right)$

$answer_{conv.and.rad} := rkfixed\left(T_{initial}, 0\cdot s, time_{stop}, 1000, D_{conv.and.rad}\right)$

$$T_{conv.and.rad} := answer_{conv.and.rad}^{\langle 1 \rangle}$$

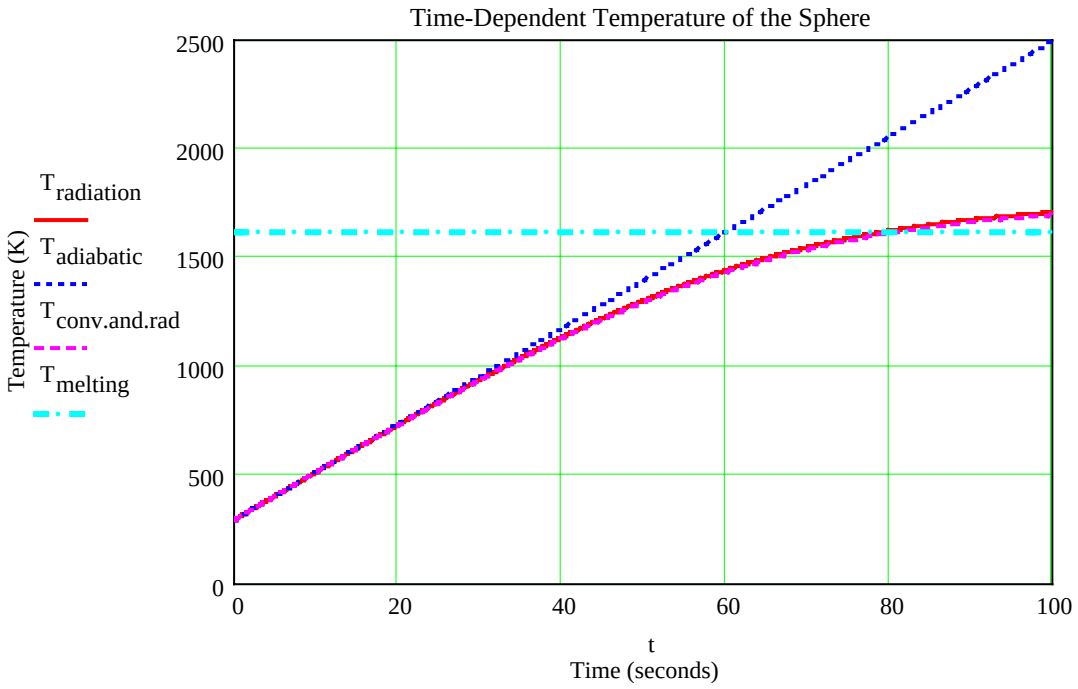
5) Define an array to display the melting temperature of curium.

$$D_{melting}(t, u) := 0$$

$$answer_{melting} := rkfixed\left(T_{fusion.Cm}, 0 \cdot s, time_{stop}, 1000, D_{melting}\right)$$

$$T_{melting} := answer_{melting}^{\langle 1 \rangle}$$

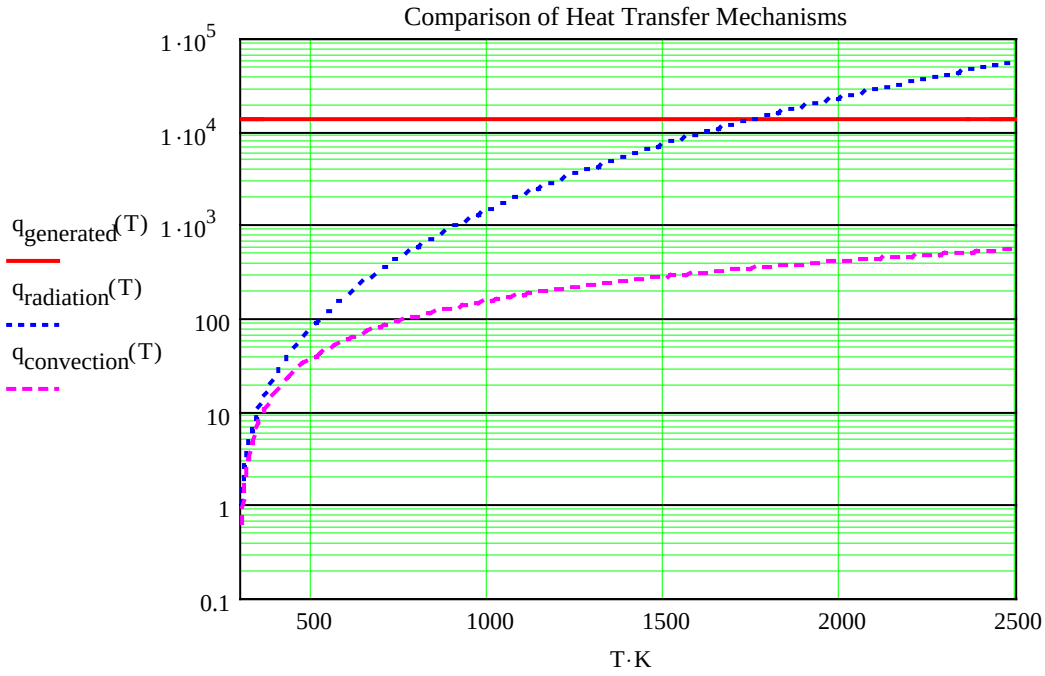
6) Plot all results for comparison.



6) Compare the heat generated to the heat lost by radiation and convection heat transfer.

a) Plot the heat transfer rates.

$$T := 300, 310 .. 2500$$



b) Show some sample values.

$q_{generated}(1200\,K) = 1.382 \times 10^4\,W$

$q_{radiation}(1200\,K) = 2.981 \times 10^3\,W$

$q_{convection}(1200\,K) = 202.873\,W$

$q_{generated}(2500\,K) = 1.382 \times 10^4\,W$

$q_{radiation}(2500\,K) = 5.634 \times 10^4\,W$

$q_{convection}(2500\,K) = 545.817\,W$

7. Compute the Equilibrium Temperature

a. When does the sphere temperature reach equilibrium and how long does it take?

$T := T_{initial}$

$T_{equilibrium} := root\left[\left(dTdt_{radiation}(T) + dTdt_{convection}(T) - dTdt_{generated}(T)\right), T\right]$

$T_{equilibrium} = 1748.5K$ *Equilibrium temperature.*

1) Find the time when the equilibrium temperature is attained.

$cc(x) := x \cdot K \le \left(\left|T_{equilibrium}\right| - 1 \cdot K\right)$ *Find out how long it takes to get within one degree of equilibrium.*

$$Locate(V, cc) := \left| \begin{array}{l} \text{for } i \in 0..rows(V) - 1 \\ \quad value \leftarrow i \text{ if } cc\left(V_i\right) = 1 \\ value \end{array} \right.$$

$Locate\left(T_{conv.and.rad}, cc\right) = 1 \times 10^3$

$equilibrium := Locate\left(T_{conv.and.rad}, cc\right)$

$t_{equilibrium} = 100$ *Time in seconds before the temperature reaches within 1 degree of the equilibrium temperature.*

b. Find the time when the curium begins to melt.

$c(x) := x \cdot K \le T_{fusion.Cm}$

$$Locate(V, c) := \left| \begin{array}{l} \text{for } i \in 0..rows(V) - 1 \\ \quad value \leftarrow i \text{ if } c\left(V_i\right) = 1 \\ value \end{array} \right.$$

$Locate\left(T_{conv.and.rad}, c\right) = 808$

$melt := Locate\left(T_{conv.and.rad}, c\right)$

$t_{melt} = 80.8$ *Time in seconds before the curium begins to melt.*

$Bi_{radiation.and.convection}\left(T_{fusion.Cm}\right) = 0.451$

Appendix A Constants

$g_{gravity} \equiv 9.81 \cdot \frac{m}{s^2}$ $\sigma \equiv 5.669\,10^{-8} \cdot \frac{W}{m^2 \cdot K^4}$ $g \equiv 10^{-3} \cdot kg$