Criticality and Thermal Analyses of Separated Actinides

E. Bakker

University of Nevada, Las Vegas:4505 Maryland Parkway Box 4027, Las Vegas, NV, 89154,
eabakker@yahoo.com

Abstract *– Curium and americium pose special problems in the chemical preparation of spent fuel for transmutation. Once separated from the other actinides, the isotopes can lead to nuclear fission with the subsequent release of a large amount of radiation. A neutron criticality code was used to determine* k_{eff} *for varying quantities of* Cm_2O_3 *and* Am_2O_3 *held within spherical or cylindrical containers. These geometries were investigated both in air and in water. Recommendations are made on the maximum amount of* Cm_2O_3 *and* Am_2O_3 *that can be safely stored or handled before encountering criticality.*

Several isotopes of curium and americium also generate a significant amount of heat by radioactive decay. If kilogram quantities are stored in a container, for example, the material may heat to an equilibrium temperature that exceeds its melting temperature. The heat generation of curium and americium present even more restriction on the mass of that can safely be contained in one location.

INTRODUCTION

The objective of this thesis is to devise a suitable storage plan for several mixtures of the actinides found in spent nuclear fuel, specifically curium and americium oxide, by analyzing their criticality and heat transfer properties. We will determine the amount of material that can be safely stored in a container while the material awaits transmutation. This information will be based on the characteristics of the fuel including its initial enrichment, fuel burnup, and decay time. These three factors can be varied in order to investigate various scenarios of spent fuel storage. Once the components of the characteristic fuel are determined, criticality tendencies will be analyzed by modeling that specific fuel in various geometries and calculating the effective neutron multiplication factor (k_{eff}) for that configuration. The k_{eff} value determines whether or not the spent fuel can sustain a nuclear fission chain reaction. If a criticality accident occurred outside the carefully designed interior of a nuclear reactor, the results could be devastating. Great care must be taken in order to prevent this type of accident from happening.

Next, the heat transfer tendencies of the spent fuel were investigated by using the radioactive decay heat, as well as many other properties of the material, to calculate the maximum temperature achieved inside the storage container. Conduction, convection, and radiation heat transfer are considered. The purpose of this analysis is to avoid melting of the material, storage container, and surroundings. The completion of this analysis will show the maximum amount of curium or americium oxide that may be placed in a receptacle of certain geometry.

PREVIOUS RESEARCH

Americium is a man-made element. It was produced by Glen Seaborg and a group of scientists by neutron bombardment of plutonium (1). It is produced inside a nuclear reactor by beta decay of 241Pu. It is most commonly used in smoke detectors and is available for commercial sale by contacting Oak Ridge National Laboratory via the U.S. DOE Isotope Programs website. It is packaged in a welded stainless steel contain meeting special form requirements in units of grams and in americium dioxide powder form (2).

Curium is also a man-made element created from the beta decay of 242Pu. It, too, is shipped in a welded stainless steel container meeting special requirements in the form of curium oxide in milligrams (2). “Since only milligram amounts of curium have ever been produced, there are currently no commercial applications for it, although it might be used in radioisotope thermoelectric generators in the future. Curium is primarily used for basic scientific research (3).”

Opperman, E.K., et al, in 2001, outlines requirements for shipment and transportation of different mixtures of spent nuclear fuel including americium and curium. This report states that “americium and curium will require a packaging with gamma and neutron shielding, and heat removal capability” (4).

Methodology

The largest source of high level nuclear waste comes from commercial nuclear power

(5). Nuclear power is created in a reactor, an intricate assembly designed to sustain a nuclear fission chain reaction. Nuclear material inside the core of a reactor gets used up and must be removed and disposed of.

There are several types of nuclear reactors that have been tested and developed, but the most widely used in the world today is the light-water reactor (6). It uses water to moderate, reflect, and cool the reactor at high pressures. There are two types of LWRs in use today; the pressurized water reactor (PWR) and the boiling-water reactor (BWR).

Both of these types of reactors are fueled by a fission reaction. A neutron strikes the fissile material and releases fission fragments, radiation, and neutrons which go on to produce further fission reactions. The energy released by the fission reaction depends on the amount of neutrons traveling through the core. The amount of neutrons is controlled by control rods which should be positioned in the reactor very carefully at all times.

DESCRIPTION OF THE ACTUAL WORK

RADDB was used to determine the composition of the spent nuclear fuel. It is a light water reactor radiological database developed at Oak Ridge National Laboratory (ORNL) that breaks down the characteristics of spent nuclear fuel removed from a commercial reactor based on its initial enrichment, burnup, and decay time. This program provides interpolated data from ORIGEN, another code developed by ORNL. It lists the radioactive totals, elemental compositions, and individual isotope concentrations of all of the materials present within a characteristic fuel. For this research, a characteristic fuel of 50,000 MWd/MTHM burnup, 4.26% enrichment, and a 10 year decay time was used. Once the isotopic concentration of the fuel was calculated in terms of g/MTIHM and W/MTIHM, the neutron diffusion equation was solved using a neutron criticality code (SCALE4.4a) to determine the effective neutron multiplication factor (k_{eff}) for varying quantities of curium and americium oxide held within cylindrical containers. These geometries were investigated both in air and in water. Based on the value of k_{eff}, recommendations are made on the maximum amount of curium and americium oxide that can be safely stored or handled before encountering nuclear criticality.

Decay heat generation also presents significant problems if curium and americium oxide are to be separated from spent fuel. The melting temperature can be exceeded if kilogram quantities are stored. A heat transfer analysis of curium and americium containers can be used to determine the temperatures that will be attained and whether melting will occur. The heat generated by decay within a container of curium or americium 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 material. The analysis of a sphere provides a conservative view of how fast the container will heat up and will over predict the maximum temperature attained when compared to a cylindrical container.

The change in heat stored within the container is equal to the heat generated by decay, the heat lost by radiation heat transfer, and the heat lost by convection heat transfer. This balance is given by Eq. (1)

$$mc_p \frac{d[T]}{dt} = g'''V - \sigma \varepsilon A(T_s{}^4 - T_\infty{}^4) - hA(T_s - T_\infty) \qquad (1)$$

where T_s is the surface temperature of the sphere. 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 material will reach a maximum, or equilibrium, temperature at the center of the geometry.

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. Convection in this analysis will not be forced. 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.

The results of the both the criticality and the heat transfer analysis are compared and the analysis that most limits the maximum allowable radius of the container becomes the limiting analysis, providing recommendations for the maximum allowable container radius.

RESULTS

Each material was investigated under both the criticality and heat transfer analysis. Table I and III shows results from the criticality analysis for a Cm_2O_3 and Am_2O_3 mixture, respectively. The heat transfer results for these mixtures are shown in Tables II and IV. The heat transfer analysis proved to be more limiting and the recommendation for the size of the container was based on this analysis. This study concludes that materials such as americium and curium oxide are dangerous and great care must be taken in handling and storing them. The containment of a relatively small amount of these materials can create criticality and heat generation concerns. A mixture of 92% americium oxide and 8% curium oxide in air, for example, can have a semi-infinite cylinder radius of only 1.4 cm before it begins to melt. This corresponds to a mass of only 2.02 kg. The results of the 92% americium oxide and 8% curium oxide mixture for both analyses is shown in Tables V and VI. This work was sponsored by the Advanced Fuels Cycles Initiative through the Harry Reid Center at the University of Nevada, Las Vegas.

TABLE I. Criticality Radius and Mass for Cylindrical Waste Form

Cm2O3 Cylinder Criticality	Radius (cm)	Mass (kg)
Air	5.4	125.59
Water	3	21.53

TABLE II. Heat Transfer Radius and Mass for Cylindrical Waste Form

Cm2O3 Cylinder Heat Transfer	Radius (cm)	Mass (kg)
Air	1.55	2.74
Water	2.25	8.37

TABLE III. Criticality Radius and Mass for Cylindrical Waste Form

Am2O3 Cylinder Criticality	Radius (cm)	Mass (kg)
Air	11.2	1032.28
Water	10.5	850.58

TABLE IV. Heat Transfer Radius and Mass for Cylindrical Waste Form

Am2O3 Cylinder Heat Transfer	Radius (cm)	Mass (kg)
Air	1.6	3.01
Water	1.9	5.04

TABLE V. Criticality Radius and Mass for Cylindrical Waste Form

92%/8% Mix Cylinder Criticality	Radius (cm)	Mass (kg)
Air	10	737.9
Water	9.3	593.54

TABLE VI. Heat Transfer Radius and Mass for Cylindrical Waste Form

92%/8% Mix Cylinder Heat Transfer	Radius (cm)	Mass (kg)
Air	1.4	2.02
Water	2	5.88

REFERENCES

1. Foundation for Blood Research Home Page. http://www.fbr.org (accessed November 2003).
2. Department of Energy Isotope Program Home Page. http://www.ne.goe.gov (accessed November 2003).
3. Jefferson Lab Home Page. http://www.jlab.org (accessed November 2003).
4. Opperman, E.K. et al, *Transportation Packages to Support Savannah River Site Missions.* Westinghouse Savannah River Site Scientific and Technical Information (2001).
5. Nuclear Waster Disposal Home Page. http://carbon.cfr.washington.edu/esc110/2003Winter/projects/133/index.html (accessed October 2003).
6. Lamarsh, J.R., Baratta, A.J. *Introduction to Nuclear Engineering.* 3rd ed. Upper Saddle River, NJ: Prentice Hall (2001).