176
12/30/68

586

SC-RR-68-832
December 1968

AEROSPACE NUCLEAR SAFETY

RADON PRODUCTION IN $^{238}PuO_2$ FUELS

M. A. Parsont, 9515

AEROSPACE NUCLEAR SAFETY DEPARTMENT, 9510

SANDIA LABORATORIES

OPERATED FOR THE U. S. ATOMIC ENERGY COMMISSION BY SANDIA CORPORATION | ALBUQUERQUE, NEW MEXICO, LIVERMORE, CALIFORNIA

DISCLAIMER

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Issued by Sandia Corporation,
a prime contractor to the
United States Atomic Energy Commission

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfilms $0.65

SC-RR-68-832

RADON PRODUCTION IN $^{238}PuO_2$ FUELS

M. A. Parsont, 9515
Sandia Laboratories, Albuquerque

Approved by: L. A. Hopkins, Jr., 9500

V. E. Blake, 9510

ABSTRACT

This report contains the results of calculations of the amounts of ^{220}Rn and ^{222}Rn produced from $^{238}PuO_2$ fuel having known impurities. Graphs are presented from which the approximate pressure in closed vessels resulting from these radon species may be easily determined. Radon-220 pressure from ^{236}Pu and ^{228}Th, and ^{222}Rn pressure from ^{238}Pu, are calculated. In addition, the radon release from a SNAP-19 is compared with present MPC values; the potential biological effect on a human is shown to be small.

December 1968

The cutoff date for information in this report is November 30, 1968.

SUMMARY

The production of radon species from Pu isotopes and impurities in $^{238}PuO_2$ fuel material has caused some concern from the health physics standpoint. The potential biological effect on a human was shown to be small based on comparison to current MPC values. Calculations showed that pressure from radon species would not exceed 10^{-7} atmospheres in SNAP-19 capsules in the range of operating temperatures.

Tables and Figures are presented which allow the simple computation of pressure and activity from radon produced in $^{238}PuO_2$ fuel having certain impurities. These tables and figures can be used for any system using $^{238}PuO_2$ fuel material.

BLANK

RADON PRODUCTION IN $^{238}PuO_2$ FUELS

Introduction

A potential biological hazard results from the radon gas species released from $^{238}PuO_2$ SNAP fuel after the fuel is stored in closed vessels for some period of time. In addition to the biological effect, the additional pressure in the fuel capsule may create undesirable mechanical stresses.

This report contains the results of calculations of the amounts of ^{220}Rn and ^{222}Rn produced from $^{238}PuO_2$ fuels containing known impurities. Graphs are presented from which the approximate pressure in closed vessels in general, and specifically in the SNAP-19, may easily be determined. Ingrowth of the gaseous species as a function of time and pressure buildup as a function of temperature and void volume are included.

In addition to the above physical considerations, assumptions were made in order to compare the release of radon from ^{238}Pu fuel material with the maximum permissible concentration (MPC) of radon in air.

General Considerations

The amount of Rn produced from a $^{238}PuO_2$ fuel material and the time over which the gas exerts its maximum pressure depend primarily on two factors. The first of these is the original isotope of Pu from which the Rn species is descended; this is a function of the isotopes which comprise the $^{238}PuO_2$ fuel. The second factor is the length of time for which the fuel is stored and the duration of the mission. The species of radon present in $^{238}PuO_2$ fuel change with time, as does the amount of any single radon species.

Pressure is directly proportional to the number of atoms and the temperature; it would then be expected to be directly proportional to the activity of the Rn produced. On the other hand, the gas void volume and the pressure are inversely proportional to one another.

Isotopic Composition of $^{238}PuO_2$ Fuel

In general, $^{238}PuO_2$ fuel is composed of the following amounts of materials.

Pu Isotope	Fraction of Fuel[1]
242	1×10^{-3}
241	8×10^{-3}
240	2.9×10^{-2}
239	0.15
238	0.81
236	1.2×10^{-6}

In addition, unsupported ^{228}Th may be present (2.3×10^{-8}) in the fuel material.[2]

Of the five Pu isotopes present, only ^{238}Pu and ^{236}Pu are important from the standpoint of Rn production over the period of time projected for a SNAP-19 mission. The amount of Rn produced from ^{242}Pu, ^{240}Pu, and ^{239}Pu is lower by more then 3 orders of magnitude than that produced from ^{236}Pu or ^{238}Pu during this time period. There is no gaseous daughter product from ^{241}Pu. There are, therefore, three possible sources of radon gas in the $^{238}PuO_2$ fuel: ^{238}Pu, ^{236}Pu, and unsupported ^{228}Th.

The Production of Rn Species of Interest

The decay schemes to Rn from ^{238}Pu and ^{236}Pu are as follows.

I.	II.	III.
^{238}Pu	^{236}Pu	
↓ 89 years	↓ 2.85 years	
^{234}U	^{232}U	
↓ 2.48×10^5 years	↓ 72 years	
^{230}Th	^{228}Th	
↓ 7.6×10^4 years	↓ 1.91 years	Unsupported ^{228}Th (1.91 years, 3.64 days)
^{226}Ra	^{224}Ra	
↓ 1.6×10^3 years	↓ 3.64 days	
^{222}Rn	^{220}Rn	
↓ 3.823 days	↓ 56 seconds	

We can now examine further the activities of radon from these sources. The activities at the various times were determined by a computer program which solved the Bateman Equation.

There are two distinct activity maxima attained by $^{238}PuO_2$ fuel containing ^{236}Pu and ^{228}Th impurities. The first activity maximum occurs at about 28 days and is the result of ^{220}Rn ingrowth from the unsupported ^{228}Th (Figure 1). The second activity maximum occurs at about 6500 days and arises from ^{220}Rn ingrowth from the ^{236}Pu (Figure 2). The ^{222}Rn produced from ^{238}Pu does not reach a maximum during the planned orbital lifetime of the SNAP-19 (600 years) (Figure 3).

The corresponding peak pressure curves are shown in Figures 4, 5, and 6. These are presented as functions of gas void volume and operating temperature. It is assumed that all the Rn is found in the gas void volume.

Figures 7, 8, and 9 give the ^{220}Rn and ^{222}Rn pressure calculated for the SNAP-19 and take into consideration the operating temperature of 850K and the peak reentry temperature of 1950K. It is assumed that all of the radon produced is found in the gas void volume and that, of the 2840 g of fuel, 1.2 x 10^{-4} percent is ^{236}Pu, 2.3 x 10^{-6} percent is ^{228}Th, and 80 percent is ^{238}Pu.

Table I gives the peak pressures expected from radon in the SNAP-19 at 850K and 1950K, given the above assumptions.

TABLE I

SNAP-19 Radon Pressure at Two Temperatures

	^{222}Rn (no equilibrium after 600 years)*		^{220}Rn From Initially Pure ^{236}Pu (equilibrium after 6500 days)		^{220}Rn From Initially Pure ^{228}Th (equilibrium after 28 days)	
Temperature (K)	Pressure per Capsule (atm)	Activity per Capsule (Ci)	Pressure per Capsule (atm)	Activity per Capsule (Ci)	Pressure per Capsule (atm)	Activity per Capsule (Ci)
850	3.9 x 10^{-8}	2.9 x 10^{-3}	8.0 x 10^{-12}	3.6 x 10^{-2}	6.0 x 10^{-12}	2.6 x 10^{-2}
1950	9.0 x 10^{-8}	2.9 x 10^{-3}	1.8 x 10^{-11}	3.6 x 10^{-2}	1.3 x 10^{-11}	2.6 x 10^{-2}

*Computed after 600 years.

Comparison of Radon Release from a SNAP-19 with the Maximum Permissible Concentration in Air (MPC_A)

The release of ^{220}Rn from the ^{228}Th decay chain has the most biological significance because it would most likely occur during storage and transportation (peak activity at about 30 days).

Let us take the maximum buildup from ^{228}Th, 5.2 x 10^{-2} Ci ^{220}Rn, and allow it to be released under the following conditions:

1. Release into a room $3m^3$ (27 x 10^6 cm^3),
2. Release 1 percent of the total Rn from both capsules (5.2 x 10^{-4} Ci),

3. MPC_A (40-hour week)^3 = 3×10^{-7} μCi cm^{-3}, and

4. An individual is exposed to this concentration for 1 hour.

The ratio of ^{220}Rn concentration in the room to the MPC_A would then be:

$$\left(\frac{5.2 \times 10^2 \ \mu Ci}{27 \times 10^6 \ cm^3}\right)\left(\frac{cm^3}{3 \times 10^{-7} \ \mu Ci}\right) \cong 60$$

The fraction of the maximum permissible lifetime dose would then be about 6×10^{-4}.

From the standpoint of a potential biological effect, the risk to a human exposed to radon gas species from $^{238}PuO_2$ fuel is small.

References

1. Holley, W. L., Methods of Assessing the Lung Dose from Inhaled $^{238}PuO_2$, SC-TM-67-594, Sandia Laboratories, Albuquerque, New Mexico, October 1967.

2. Weber, W. B., and Jicha, Jr., J. J., SNAP-19 Operational Safety Review IRHS Addendum, MND-3607-118A, Martin Marietta Company, Middle River, Maryland.

3. Report of Committee II on Permissible Dose from Internal Radiation, Pergamon Press, London, 1959.

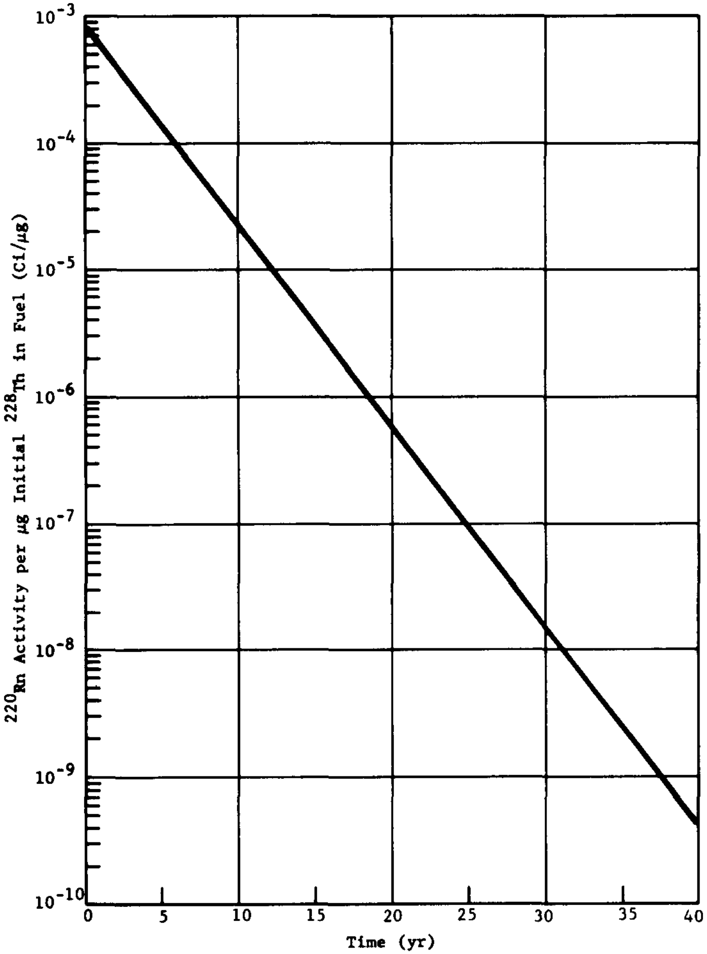

Figure 1. Activity of ^{220}Rn from ^{228}Th parent as a function of time

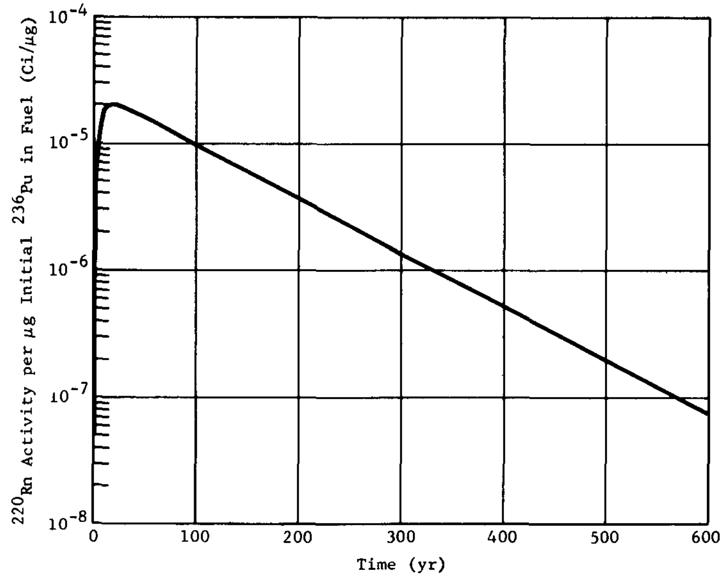

Figure 2. Activity of ^{220}Rn from ^{236}Pu parent as a function of time

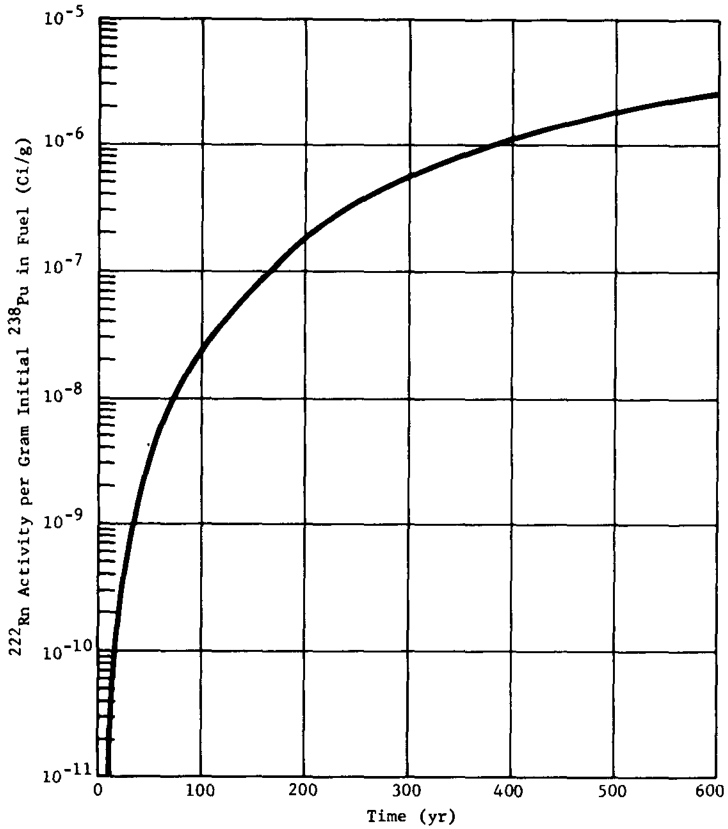

Figure 3. Activity of ^{222}Rn from ^{238}Pu parent as a function of time

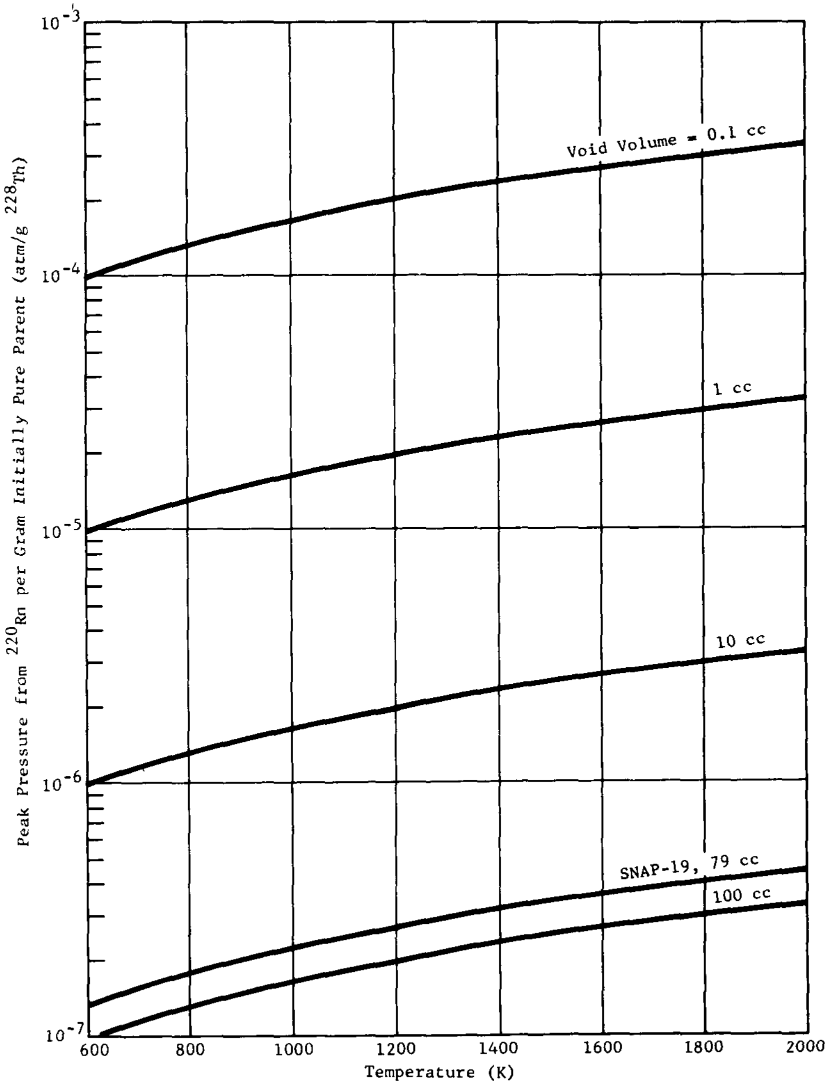

Figure 4. Peak pressure from ^{220}Rn from an initially pure ^{228}Th parent as a function of temperature, mass of ^{228}Th and void volume

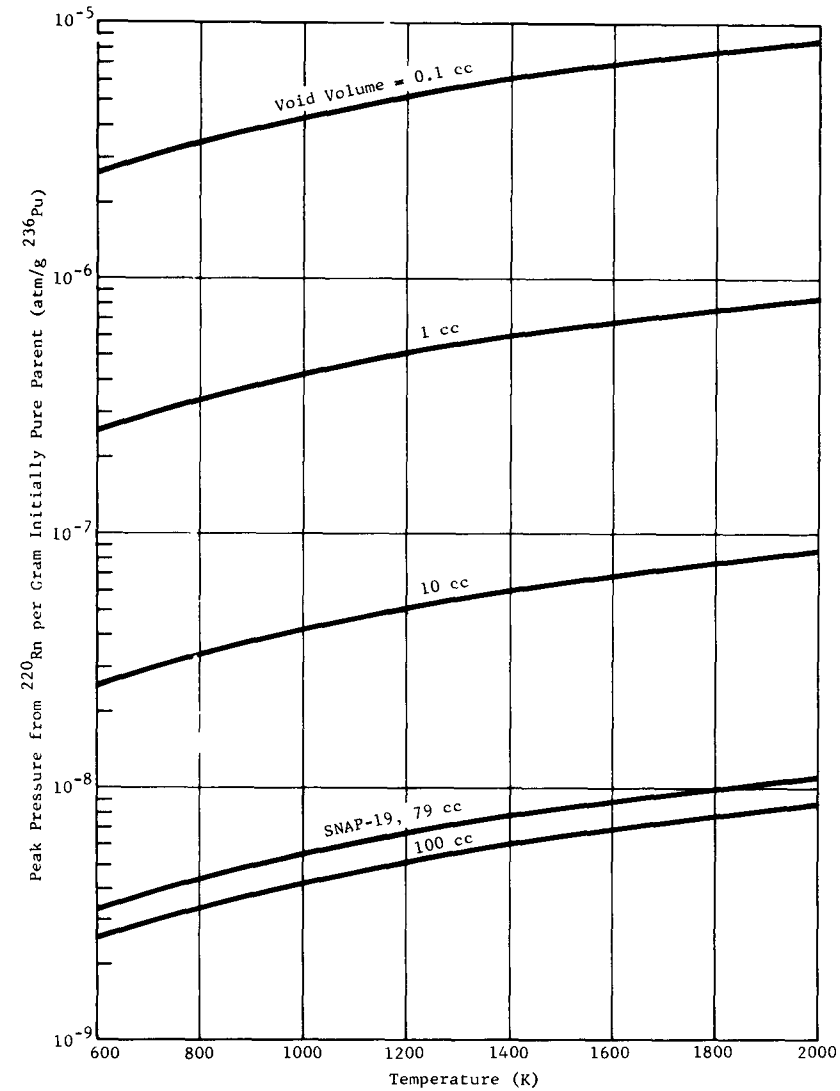

Figure 5. Peak pressure from ^{220}Rn from an initially pure ^{236}Pu parent as a function of temperature, mass of ^{238}Pu and void volume

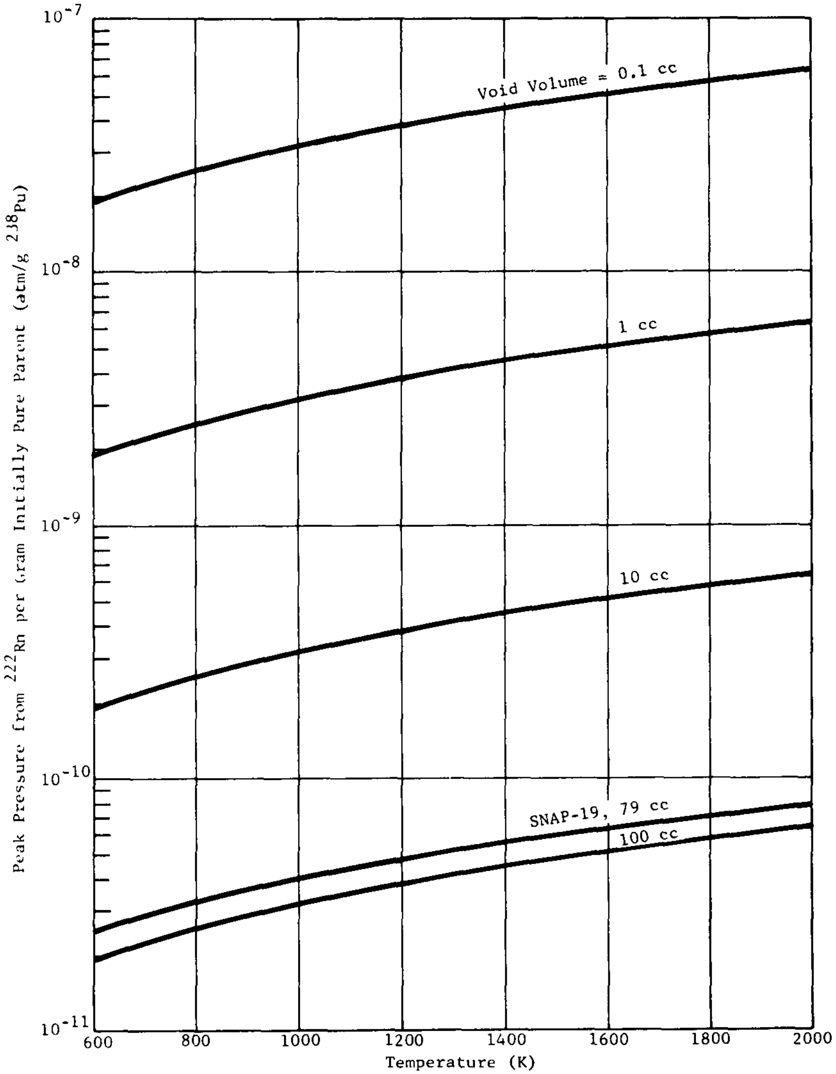

Figure 6. Peak pressure from ^{222}Rn from an initially pure ^{238}Pu parent as a function of temperature, mass of ^{238}Pu and void volume