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CALCULATION OF FUEL LOSS FROM VENTED NUCLEAR FUEL ELEMENTS FOR SPACE POWER REACTORS

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ABSTRACT

The rate of loss of uranium dioxide (UO_2) or of uranium mononitride (UN) from cylindrical nuclear fuel elements which are vented to prevent buildup of high fission gas pressures has been calculated for various conditions. For 10 000 hr operation at a cladding temperature of 1400° C and venting to a vacuum, the maximum fuel temperature allowable for UO_2 is about 2000° C and for UN about 1800° C. These temperatures may be increased to about 2500° and 2600° C, respectively, by venting to an argon atmosphere containing enough oxygen or nitrogen to prevent decomposition of the fuels.

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SUMMARY

An analytical study has been made of the rate of loss of uranium dioxide (UO_2) or uranium mononitride (UN) from a vented cylindrical nuclear fuel element 30-centimeters long by 1.25 centimeters in diameter including a 0.1-centimeter thick tungsten cladding. The vent is a cylindrical hole 0.1-centimeter long and 0.0125 centimeter in diameter and is provided to avoid buildup of high fission gas pressures. The fuel element is assumed to contain 20 cubic centimeters of fuel. The upper limit on allowable fuel loss was assumed to be 0.1 gram after 10 000 hours at a cladding temperature of 1400° C. This is believed to be a conservative estimate.

This study indicated that under these conditions UO_2 can be used at maximum fuel temperatures of about 2000° C when the fuel element is vented to vacuum and to about 2500° C when the fuel element is vented to a space containing 10^6 newtons per square meter of argon with about 2×10^{-1} newtons per square meter of oxygen to prevent decomposition of the UO_2. The corresponding fuel temperatures for UN are 1800° and 2600° C except that for 2600° C operation the argon must contain about 10^4 newtons per square meter partial pressure of nitrogen (instead of oxygen) to suppress decomposition of the UN.

Operating times longer than 10 000 hours can be achieved by decreasing the maximum allowable fuel temperature. For example, the maximum UN fuel temperature for 50 000-hour operation must be decreased by 200° C, to about 2400° C, for elements vented to a space pressurized with 10^6 newtons per square meter of argon and about 10^2 newtons per square meter of nitrogen.

INTRODUCTION

The feasibility of a fast spectrum, lithium-cooled nuclear reactor for space power generation is being studied at the Lewis Research Center. The reactor must heat the lithium to temperatures of 900° to 1400° C, and must operate for 10 000 to 50 000 hours. Uranium burnups of 5 atomic percent or more are expected under proposed operating conditions. Two methods for dealing with the resultant fission gases are being examined. One method is to contain the fission gas inside of the fuel element by providing a void space and a strong, creep-resistant cladding. The other method, which will be discussed here, is to allow the fission gas to escape from the fuel element through a vent. In either method, excessive amounts of fuel must not escape from the fuel elements - either through the cladding or through the vent.

The main purpose of this work is to find out if it is possible to use uranium dioxide (UO_2) or uranium nitride (UN) in vented, long-life fuel elements without excessive fuel loss. The approach is to calculate rates of high temperature fuel decomposition and fuel loss from a vented, tungsten-clad fuel element of a size and configuration considered for use in space power reactors. A complete parametric analysis is not the intent of this work, but the calculation method given could be applied to do such an analysis. The calculations are based on reasonable mechanisms and on available vapor pressure and thermodynamic data. The calculations are made assuming a cladding temperature of 1400° C, a selected fuel element size, fuel temperatures up to about 2700° C, and operating times of 10 000 hours or longer. The effects of venting to vacuum and to atmospheres containing combinations of argon, nitrogen, and oxygen are examined. It is hoped that the results will aid in the selection and design of necessary experimental tests.

Some recent unpublished investigations of mass transport mechanisms in vented thermionic emitters containing UO_2 fuel have been carried out by the General Electric Company under NASA contract. These investigations include both analytical and experimental studies. Although some of the analytical approaches are similar to those used here, the conditions are quite different.

FUEL ELEMENT MODEL

In this report the calculations of fuel decomposition and loss are based on a cylindrical fuel element that is 1.25 centimeters (~1/2 in.) in outside diameter and 30-centimeters (~12-in.) long (see fig. 1). These dimensions include a 0.1-centimeter (~0.040-in.) thick tungsten cladding. The fuel element consists of about 11 cubic centimeters of cladding, about 20 cubic centimeters of fuel (either UN or UO_2), about 5 cubic centimeters of void volume as a central void, and about 1 cubic centimeter of vent assembly. This reference fuel element is vented with a hole 0.1-centimeter long and 0.0125 centimeter in diameter. The element must operate for at least 10 000 hours with a maximum cladding temperature of 1400° C.

For this analysis, it is assumed that: (1) 50 cubic centimeters of fission gas are formed per gram of uranium fissioned (ref. 1 and appendix B), (2) burnup is linear with

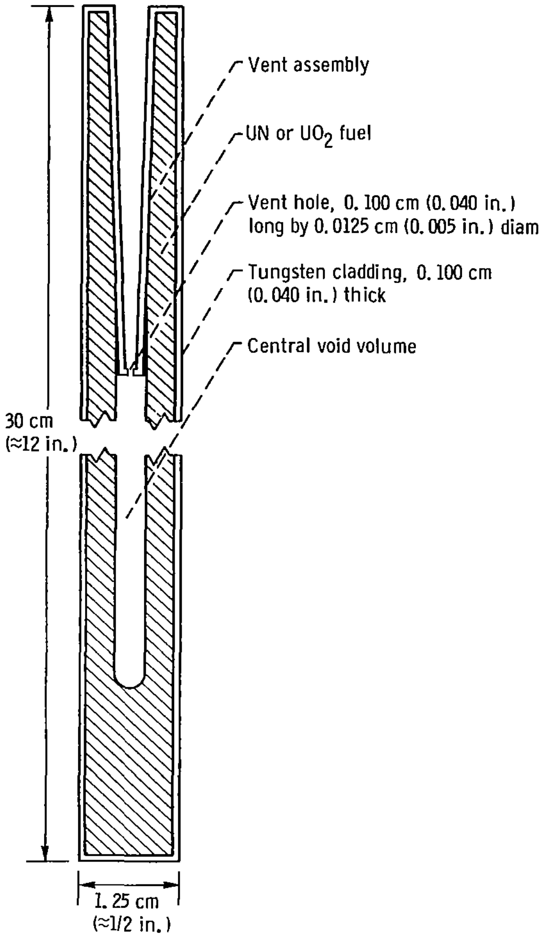

Figure 1. - Schematic drawing of cylindrical fuel element used in calculations. Fuel element is surrounded by lithium except at the ends.

time to 5 atom percent at 10 000 hours, (3) fission gas is released from the fuel immediately when it is formed,[1] (4) fission gas escapes through the vent by molecular flow, (5) locating the vent hole in the central void, where the temperature is highest, will avoid restriction or plugging the vent with fuel (ref. 2) and, (6) there are no thermal diffusion or surface transport effects.

For this fuel element model operating under these assumed conditions, the steady-state gas pressure inside the fuel element due to the fissioning of fuel is calculated to be

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[1] In terms of the rate of loss of fission gas through the vent, this is a conservative assumption. The problems resulting from incomplete fission gas release and fuel swelling are not considered in this report.

less than 8×10^{-4} atmosphere ($80 \ N/m^2$) at temperatures up to about 2700° C (see appendix B). This calculated pressure is consistent with the assumption of molecular flow and also shows that the vent is large enough to accomplish its purpose of preventing buildup of high fission gas pressures.

MODES OF FUEL LOSS

Fuel can be lost through the tungsten cladding even if the tungsten cladding initially is free of cracks or porosity. If enough free uranium is formed, uranium will penetrate grain boundaries in the tungsten and create defects through which fuel can be lost by evaporation or by being swept away by the lithium. In addition, fuel also can be lost by evaporation and flow through the vent.

It is not known how much fuel loss by evaporation through the vent and cladding is tolerable. But the loss of a maximum of 0.1 gram of fuel in 10 000 hours is believed to be a conservative estimate of the limit for the reference fuel element. In the following calculations it is assumed that a fuel element will fail if this limit is exceeded. Also, it is assumed that if more than 0.1 gram of liquid uranium is formed the cladding will be damaged so that more than 0.1 gram of fuel will be lost by evaporation through the cladding.

The cases of UO_2 and UN as fuels are different in that UN decomposes to uranium and nitrogen on evaporation whereas UO_2 evaporates primarily as UO_2. Therefore, the mechanisms of escape of UO_2 and UN from vented fuel elements are discussed separately.

Uranium Mononitride Fuel

Loss through the cladding. - In the case of UN as the fuel, it is known that the UN will decompose at high temperatures to form nitrogen and free uranium (refs. 3, 4, and 5).

$$UN_{(s)} \rightarrow U(l) + \frac{1}{2} N_2$$

At equilibrium in an unvented fuel element at 2500° C the nitrogen pressure in the 5 cubic centimeters void would be only about 10^{-2} atmosphere ($10^3 \ N/m^2$) and the maximum amount of free uranium would be 2.6×10^{-5} gram. If liquid uranium is to accumulate in the fuel element, there must be a preferential escape of nitrogen. This can happen in two ways: (1) by permeation through the tungsten cladding, and (2) by flow through the vent.

The permeation mechanism for formation of free uranium is not very important at the maximum tungsten cladding temperature of 1400° C. The amount of uranium formed in the fuel element after 10 000 hours is 3×10^{-2} gram. This is calculated using a permeation coefficient of 2×10^{-7} $(cm^3)(mm)/(cm^2(min)(atm^{1/2})$ for nitrogen through tungsten at 1400° C (ref. 6), and a partial pressure of about 10^{-2} atmosphere (10^3 N/m^2) of nitrogen over a mixture of uranium and UN at 2530° C (ref. 3). The details of this calculation are given in appendix C.

For the composition of the UN inside the fuel element to remain constant, the number of moles of uranium (U) vapor lost through the vent must be twice the number of moles of nitrogen (N_2) lost through the vent. Assuming molecular flow through the vent and nitrogen and uranium at equal pressures, nitrogen will be lost 2.9 times faster than the uranium (see appendix D). Therefore, for constant composition of UN in the cell, the partial pressure of uranium must be 5.8 times that of the nitrogen. It can be shown (see appendix D) that at temperatures above about 1500° C liquid uranium will be present with the UN inside a vented fuel element. The calculated amount of liquid uranium formed in 10 000 hours against fuel temperature is shown in figure 2 (see appendix D for the calculation method). This plot shows that fuel temperature would have to be held at less than 1800° C to limit the amount of free uranium formed in 10 000 hours to less than 0.1 gram.

Fuel loss through the cladding can be prevented by avoiding the formation of liquid

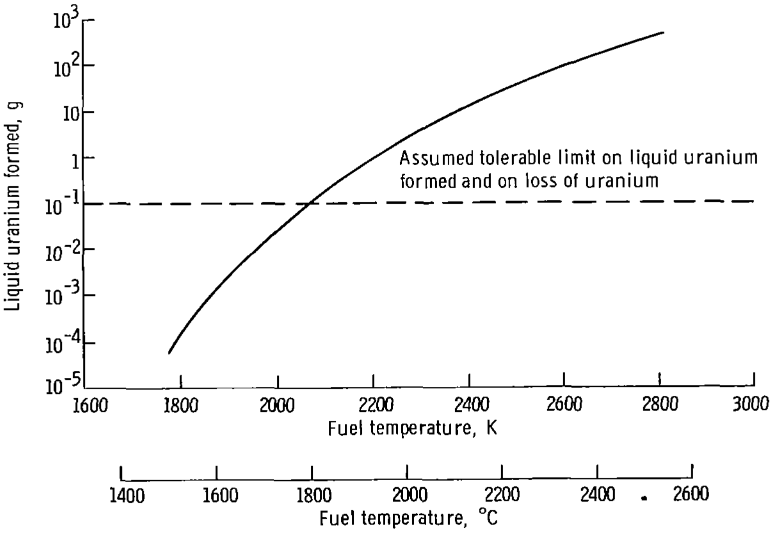


Figure 2. - Amount of liquid uranium formed in vented fuel element containing uranium nitride because of loss of nitrogen through the vent. Time, 10 000 hours; vent size, 0.100 centimeter (0.040 in.) long by 0.0125 centimeter (0.005 in.) in diameter; atmosphere outside of vent, vacuum.

uranium. In the case of UN this can be done by providing a pressure of nitrogen outside of the vent equal to the partial pressure of nitrogen over the fuel inside of the fuel element. This nitrogen pressure would range from about 10^{-8} atmosphere (10^{-3} N/m^2) at 1500° C to about 10^{-1} atmosphere (10^4 N/m^2) at 2600° C (see ref. 7). However, the maximum allowable fuel temperature also depends on the rate of uranium loss through the vent. This is discussed in the next section. It will be shown how much the operating temperature can be increased by means of a small nitrogen pressure.

Loss through the vent. - In this discussion it is assumed that the condensed phases present in the fuel element are solid UN and a trace of free uranium (i.e., < 0.1 g). This assumption will give the maximum possible partial pressure of uranium in the gas phase inside of the fuel element when compared to the case where UN is the only condensed phase. This pressure is determined by the vapor pressure of uranium. Therefore, the losses of uranium by flow through the vent as calculated in this section are upper limits.

The gases will escape through the vent into a surrounding vacuum by molecular flow, transition-range flow, or viscous flow depending on the total pressure of the gases inside the fuel element. But for the conditions and fuel element model used in this analysis, it can be shown (see appendix E) that U and N_2 will escape through the vent by the molecular flow mechanism.

The 10 000 hour loss of uranium by molecular flow through the reference vent to a

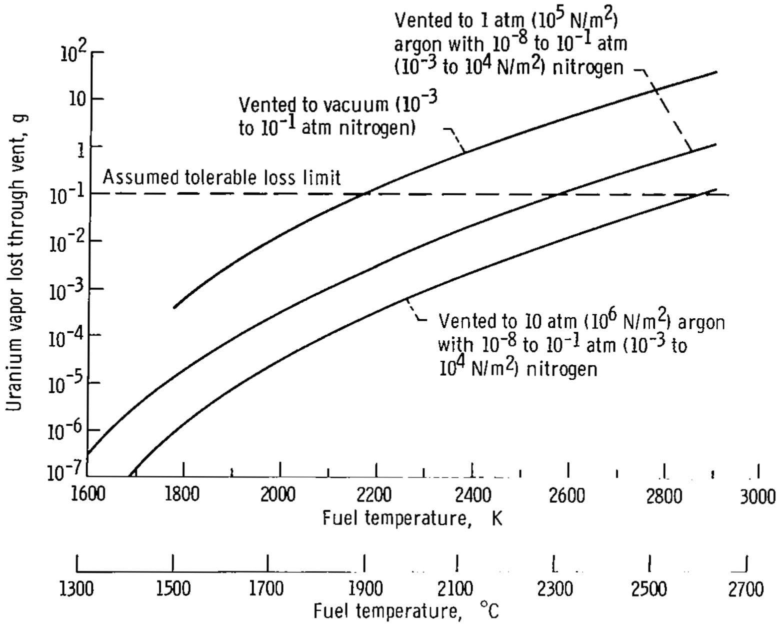


Figure 3. - Amount of uranium vapor lost through vent of fuel element containing uranium nitride. Time, 10 000 hours; vent size, 0.100 centimeter (0.040 in.) long by 0.0125 centimeter (0.005 in.) in diameter; cladding, 0.100-centimeter- (0.040-in.-) thick tungsten.

vacuum (actually to 10^{-8} to 10^{-1} atm (10^{-3} to 10^4 N/m^2) of N_2 depending on the temperature) is plotted against temperature on the upper curve in figure 3. The method of calculation for the values plotted is shown in appendix E. The upper curve in figure 3 shows that loss of 0.1 gram of uranium will not occur in 10 000 hours until fuel temperatures of about 1900° C are reached. Therefore, an increase of about 100° C in the fuel temperature can be achieved by using a small pressure of nitrogen (see preceding section and fig. 2).

Method for decreasing the rate of loss of uranium through the vent. - The loss of uranium from UN through the vent can be decreased further by introducing an inert gas into the fuel element and the venting space. The inert gas must contain enough nitrogen to prevent decomposition of the UN (i.e., 10^{-8} to 10^{-1} atm for fuel temperatures from about 1500° to 2600° C but not enough nitrogen to allow formation of higher nitrides like U_2N_3. The pressure of the inert gas must be high enough so that the mechanism of flow through the vent is changed from molecular to diffusive flow.

The effect of argon at 1 and 10 atmospheres pressure has been calculated (see appendix F), and the results are plotted on figure 3. This plot shows that the reference fuel element can operate in argon at 1 atmosphere pressure up to about 2300° C for 10 000 hours before 0.1 gram of uranium is lost through the vent. Ten atmospheres of argon allow fuel temperatures up to about 2600° C for 10 000 hours before 0.1 gram of uranium is lost. Where no argon is used the top curve shows that the maximum allowable temperature for 10 000 hour operation is about 1900° C. Or, viewed in another way, the use of an argon cover gas can give a 2 to 3 order of magnitude decrease in uranium loss at a given temperature.

Uranium Dioxide Fuel

Loss through the cladding. - The rate of loss of UO_2 through the tungsten cladding depends on the following reactions (refs. 8 and 9):

$$UO_2 \rightarrow UO_{2-x} + \left(\frac{x}{2}\right)O_2 \tag{1}$$

$$UO_{2-x} \rightarrow \left(1 - \frac{x}{2}\right)UO_2 + \frac{x}{2}\,U \tag{2}$$

The oxygen in reaction (1) can be lost by permeation through the tungsten cladding and by molecular flow through the vent. Reaction (2) shows the disproportionation of the substoichiometric UO_{2-x} formed in reaction (1) to form free uranium. this reaction occurs on cooling from the temperature at which reaction (1) occurs. The amount of uranium which can be formed in 10 000 hours because of permeation of oxygen through the tungsten cladding can be calculated similarly to the UN case. This was done using data on partial pressure of oxygen over UO_2 against temperature (from ref. 10) and a permeation coefficient for oxygen through tungsten at 1400° C of 1.4×10^{-3} $(cm^3)(mm)/(cm^2)(min)(atm^{1/2})$ (ref. 11). The resulting plot of grams of uranium formed in 10 000 hours against fuel temperature is shown on figure 4. This shows that the fuel temperature cannot exceed about 2200° C if the free uranium formed is to be kept less than 0.1 gram in 10 000 hours.

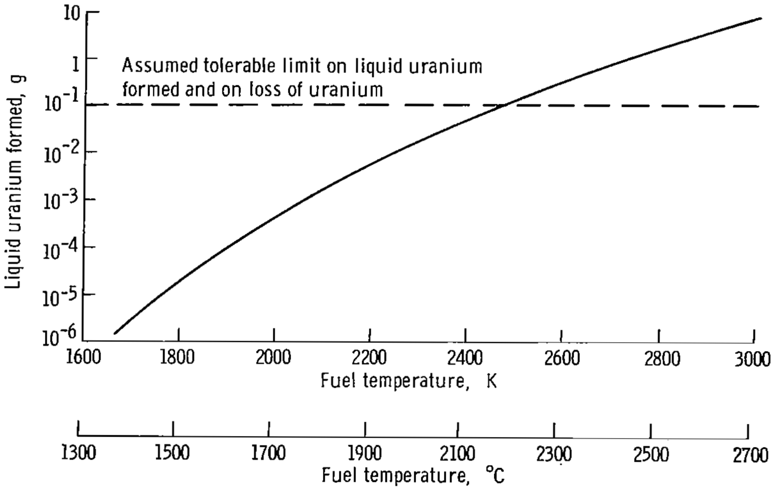


Figure 4. - Amount of liquid uranium formed in vented fuel element containing uranium dioxide because of permeation of oxygen through the cladding. Time, 10 000 hours; vent size, 0.100 centimeter (0.040 in.) long by 0.0125 centimeter (0.005 in.) in diameter; cladding, 0.100-centimeter- (0.040-in.-) thick tungsten; cladding temperature, 1400° C.

From a partial pressure of oxygen of about 10^{-8} atmosphere (10^{-3} N/m^2) at 2200° C (ref. 10) and the equation for molecular flow, it can be shown that the amount of uranium formed because of effusion of oxygen through the vent is about 2×10^{-4} gram in 10 000 hours (see appendix D). Therefore, the contribution of the vent to the formation of free uranium from UO_2 is small compared to permeation through the cladding (i.e., 10^{-4} g $<<$ 0.1 g).

Loss through the vent. - The main constituent of the vapor over solid UO_2 at high temperatures is gaseous UO_2 (ref. 12). Therefore, fuel is lost through the vent primarily as UO_2. This UO_2 is lost through the vent into a surrounding vacuum by molecular flow at fuel temperatures to about 2500° C (see appendix E). The loss of UO_2 in grams after 10 000 hours is plotted against temperature on the upper curve in figure 5. The method of calculation is shown in appendix E. The maximum allowable temperature

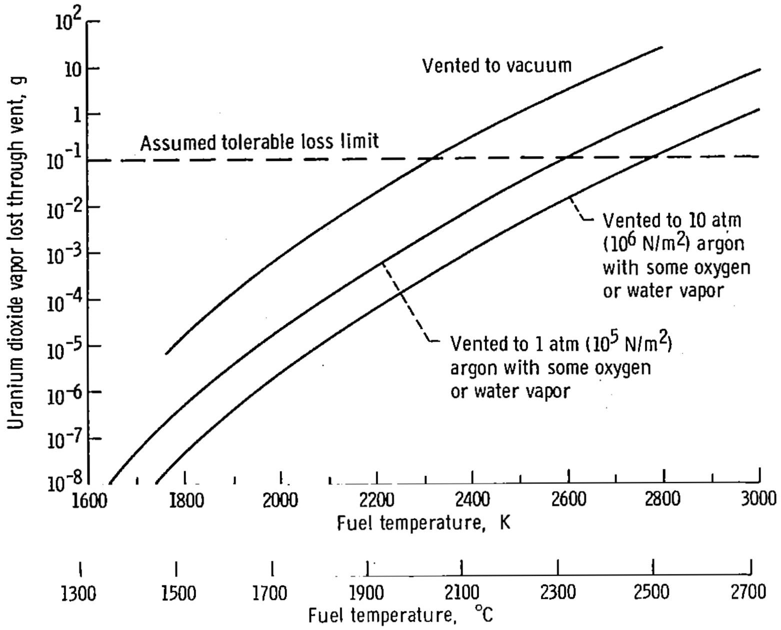


Figure 5. - Amount of uranium dioxide vapor lost through vent of fuel element. Time, 10 000 hours; vent size, 0.100 centimeter (0.040 in.) long by 0.0125 centimeter (0.005 in.) in diameter; cladding, 0.100-centimeter- (0.040-in. -) thick tungsten.

for 10 000 hours operation is about 2000° C.

Method for decreasing the rate of UO_2 loss through the vent. - The loss of UO_2 by flow through the vent can be decreased by changing the mechanism of flow from molecular flow to diffusive flow by blanketing the fuel element with argon. But the decomposition of UO_2 to form uranium because of permeation of oxygen through the tungsten cladding also must be prevented. One way of doing this would be to add a small partial pressure of oxygen or water to the argon to prevent decomposition of the UO_2 (ref. 10).

The effect of argon at pressures of 1 and 10 atmospheres (10^5 and 10^6 N/m^2) has been calculated (appendix F) and the 10 000 hour weight loss of UO_2 is plotted against temperature on figure 5. One atmosphere (10^5 N/m^2) of argon allows operation up to about 2300° C for 10 000 hours to 0.1 gram of UO_2 loss. Ten atmospheres (10^6 N/m^2) of argon allows operation at near 2500° C before 0.1 gram of UO_2 is lost. These temperatures may be compared to the 2000° C maximum where argon is not used (see preceding section).

DISCUSSION OF RESULTS

Comparison of Uranium Nitride and Uranium Dioxide

The reference fuel element containing UN vented to vacuum will be limited to a maximum fuel temperature of about 1800° C for 10 000 hours of operation. For the same condition with UO_2, a maximum fuel temperature of about 2000° C can be allowed. The UN temperature is limited by the rate of accumulation of liquid uranium in the fuel element because of preferential escape of nitrogen through the vent. The UO_2 temperature limit is set by the rate of effusion of UO_2 vapor through the vent and to a lesser degree by decomposition of UO_2 because of oxygen permeation through the cladding.

Introduction of a small nitrogen pressure, corresponding to the dissociation pressure of nitrogen over UN, into the vacuum outside of the vent will allow a maximum UN temperature of 1900° C for 10 000 hours. This temperature limit is set by the rate of effusion of uranium vapor through the vent. The rate of loss of uranium through the vent can be decreased by changing the loss mechanism from molecular flow to diffusive flow. This can be done by using an inert gas in place of the vacuum outside of the vent. For example, a pressure of 10 atmospheres of argon (containing about 10^{-1} atm (10^4 N/m^2) of nitrogen to prevent accumulation of liquid uranium) will allow a maximum UN temperature of 2600° C for 10 000 hours.

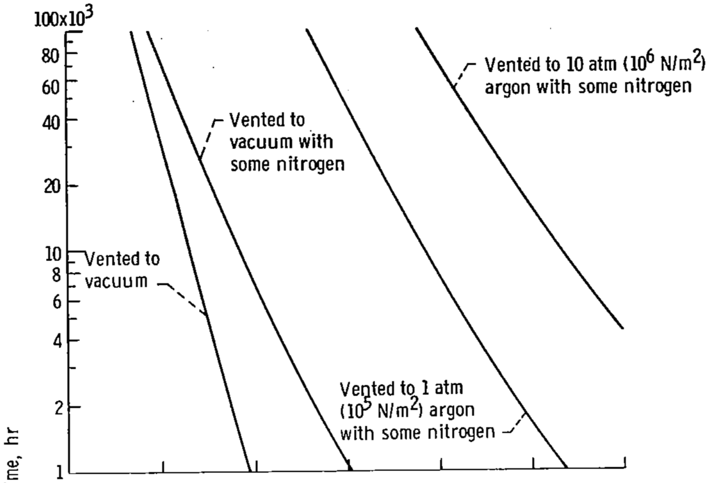
The use of an argon cover gas also will repress the loss of UO_2 vapor through the vent and will allow an increase in maximum fuel temperature to 2200° C. This limit is set by the accumulation of liquid uranium in the fuel element because of permeation of oxygen through the tungsten cladding. If the accumulation of uranium can be prevented by use of metal oxide additives to the UO_2 (ref. 13) or by addition of a small partial pressure of oxygen (or water vapor) to the argon (ref. 10), the maximum UO_2 temperature could be raised to about 2500° C in the presence of 10 atmospheres (10^6 N/m^2) of argon.

Therefore, it appears feasible to use UN at fuel temperatures up to 1800° C in vacuum and up to about 2600° C in argon. And UO_2 can be used at temperatures up to 2000° C in vacuum taking both permeation and effusion into account and up to about 2500° C in argon.

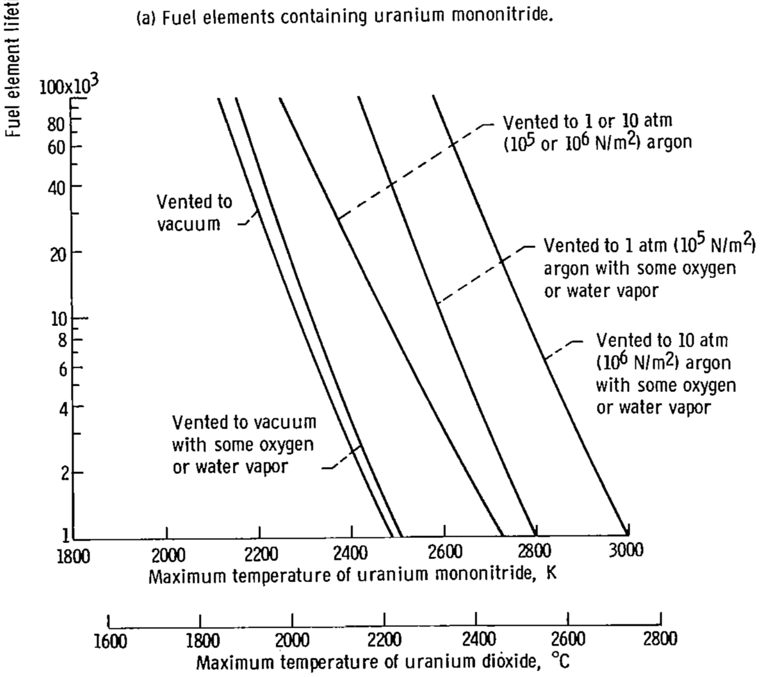
Effect of Longer Operating Times

The calculations and discussions presented so far in this report are centered around a 10 000 hour fuel element life. If longer lifetimes are required, the maximum fuel temperature must be decreased for the same total amount of fuel loss. For example, the fuel loss in 10 000 hours is plotted in figure 3 against fuel temperature for fuel elements containing UN vented to vacuum, to 1 atmosphere of argon, and to 10 atmospheres of argon. The curve for the case of venting to 10 atmospheres of argon shows that at a fuel temperature of about 2600° C the fuel element will operate for 10 000 hours before 0.1 gram of fuel is lost. If 50 000 hours of life are required, then the fuel loss in 10 000 hours can be only 0.02 gram. Therefore, the maximum fuel temperature can be only about 2400° C.

Figures 6(a) and (b) show the predicted maximum allowable life expected at different



(a) Fuel elements containing uranium mononitride.



(b) Fuel elements containing uranium dioxide.

Figure 6. - Predicted lifetime as function of maximum fuel temperature for reference fuel elements vented to selected atmospheres. Failure criteria: formation of 0.1 gram of uranium inside fuel element or loss of 0.1 gram of uranium through vent.

fuel temperatures in different environments for fuel elements containing either UN or UO_2, respectively. Comparison of the plots in figures 6(a) and (b) show that in an environment of 10 atmospheres (10^6 N/m^2) of argon, UN and UO_2 can be operated to about the same maximum temperature levels. Under a vacuum, however, UO_2 can operate at much higher temperatures than UN. This appears to be an advantage of UO_2 for use in vented fuel elements. But this potential advantage is negated by the poor thermal conductivity of UO_2 compared to UN (refs. 14 and 15). The use of UN instead of UO_2 would allow fuel elements to operate at a much lower maximum fuel temperature for the same power rating. Therefore, vented fuel elements containing UN fuel might actually be able to operate for longer times than fuel elements containing UO_2 at the same cladding temperature and power rating.

Experimental Tests of Results

The results of these calculations indicate the feasibility of venting long-life fuel elements containing either UN or UO_2. However, some long-term experimental tests to confirm the predicted lifetimes would be desirable. For the longest possible lifetimes and highest heat fluxes, a cover gas like argon containing oxygen or water vapor (for UO_2 fuel) or nitrogen (for UN fuel) should be used. Long term tests in a reactor would be necessary. If, for some reason, argon with water vapor or nitrogen cannot be used, then tests of fuel elements vented to vacuum will be required. In this case the tests can be accelerated to obtain results in less than 10 000 hours by starting with substoichiometric fuel or by operating at fuel temperature over 1800° C for UN and over 2000° C for UO_2.

CONCLUSIONS

The decomposition and loss of fuel from a vented, tungsten-clad, cylindrical fuel element containing either uranium nitride or uranium dioxide have been calculated for a cladding temperature of 1400° C as a function of fuel temperature, operating time, and the gaseous environment. It has been concluded that:

1. Either uranium mononitride or uranium dioxide can be used in vented fuel elements. Such fuel elements can operate for 10 000 hours with fuel losses less than 0.1 gram out of about 250 grams of fuel.

2. Uranium mononitride can be used at fuel temperatures up to about 2600° C for 10 000 hours if the fuel element is vented to a space pressurized with 10 atmospheres

(10^6 N/m^2) of argon containing about 0.1 atmosphere (10^4 N/m^2) partial pressure of nitrogen.

3. Uranium dioxide can be used at fuel temperatures up to about 2500° C for 10 000 hours if the fuel element is vented to a space pressurized with 10 atmospheres (10^6 N/m^2) of argon containing about 2×10^{-6} atmosphere (2×10^{-1} N/m^2) partial pressure of oxygen.

4. In case the fuel element is vented to vacuum, the maximum fuel temperatures for 10 000-hour operation are about 2000° C for UO_2 and about 1800° C for UN.

5. Operating times longer than 10 000 hours can be achieved by decreasing the maximum allowable fuel temperature. For example, the maximum UN fuel temperature for 50 000-hour operation must be decreased by about 200° C, to about 2400° C for elements vented to a space pressurized with 10 atmospheres (10^6 N/m^2) of argon and about 10^{-3} atmosphere (10^2 N/m^2) of nitrogen.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, July 11, 1968,
120-27-04-06-22.

APPENDIX A

SYMBOLS

A	cross-sectional area of vent (cm^2)
a	radius of vent (cm)
C_i	molar concentration ($mole/cm^3$)
D_{ij}	binary diffusion constant between species i and j (cm^2/sec)
F	rate of flow (cm^3/sec)
$\mathscr{F}$	rate of molecular flow (g-mole/sec)
ΔG	change in Gibbs free energy (cal/g-mole)
J	rate of diffusive flow (g-mole/sec)
K	a dimensionless function of l/a (equal to 0.1367)
k	Boltzmann constant (erg/(molecule)(K))
L	mean free path (cm)
l	length of vent (cm)
M	gram molecular weight
N_o	Avogadro's number (molecule/g-mole)
P	permeation coefficient ((cm^3)(mm)/(cm^2)(min)($atm^{1/2}$))
p_i	partial pressure of gas i (atm)
R	gas constant (usually ((cm^3)(atm)/(g-mole)(K)); in some instances cal/(g-mole)(K))
S_{ij}	collision radius (cm)
T	temperature (K)
V	volume of gas (cm^3)
$\tilde{V}_{b,liq}$	volume at boiling point of liquid (cm^3/g-mole)
$\tilde{V}_{m,sol}$	volume at melting point of solid (cm^3/g-mole)
ϵ	maximum energy of attraction between a pair of molecules (ergs)
ν	number of molecules per unit volume ($molecule/cm^3$)
ρ	density (g/cm^3)
σ_{ij}	distance of separation between species i and j on collision where $\varphi_{ij}(r)$ is zero (Å)
$\varphi_{ij}(r)$	intermolecular potential field (ergs)
$\Omega_{D,ij}$	dimensionless collision integral characteristic of diffusion

APPENDIX B

STEADY STATE PRESSURE OF GASES GENERATED BY FISSION IN A VENTED FUEL ELEMENT

It is important to know the steady state fission gas pressure inside the fuel element. The vent must be large enough to keep the gas pressure below a preset design point and it must be small enough to prevent excessive fuel loss. Also the gas pressure determines the mechanism of flow through the vent. The mechanism of flow must be known to carry out calculations of fuel loss rates through the vent.

The purpose in this appendix is to calculate the steady state gas pressure in the reference fuel element. Fuel loss is discussed elsewhere in this report. It is assumed that 5 atom percent burnup occurs in 10 000 hours and that about 50 cubic centimeters (STP) of fission gas are released per gram of uranium fissioned. This fission gas includes about 25 cubic centimeters of xenon and krypton and about 25 cubic centimeters of cesium and rubidium which are assumed to be gaseous at the fuel temperatures considered (see refs. 1 and 15). For these calculations it is assumed that the gas is all xenon and that it is released from the fuel immediately upon formation. Complete release is the worst conceivable case insofar as adequacy of venting is concerned. Problems arising from incomplete gas release and fuel swelling are not considered in this report. The amount of fission gas formed will be greater for UN than for UO_2 for the same volume of fuel and the same percent burnup because of the greater uranium density in UN. Therefore, this calculation is made for UN. In addition to the fission gas, it is expected that about 8.2 cubic centimeters (STP) of nitrogen will be released from UN per gram of uranium fissioned (ref. 16).

Assuming molecular flow the rate of flow of xenon (F_{Xe}) out through the short cylindrical vent (0.0125-cm diam by 0.10-cm long) is given by ref. 17):

$$F_{Xe} = 3638\ KA\left(\frac{T}{M}\right)^{1/2}\ \mathrm{cm^3/sec}$$

K	a dimensionless function of l/a (equal to 0.1367)
l	length of vent, cm
a	radius of vent, cm
A	cross-sectional area of vent, cm^2
T	absolute temperature, K
M	gram-molecular weight of gas (in this case xenon)

$$F_{Xe} = 5.3\times10^{-3}\sqrt{T}\ (\mathrm{cm}^3/\mathrm{sec})$$

Similarly in the case of nitrogen

$$F_{N_2} = 1.15\times10^{-2}\sqrt{T}\ \mathrm{cm}^3/\mathrm{sec}$$

The rate of formation of xenon is $676/3.6\times10^7$ cubic centimeter per second at standard conditions of temperature and pressure and the rate of formation of nitrogen is $111/3.6\times10^7$ cubic centimeters per second at STP.

At temperature T (K) and pressure p_{Xe} (atm), the rate of formation of xenon is $676/3.6\times10^7 \times T/273 \times 1/p_{Xe}$ cubic centimeters per second assuming perfect gas law behavior. And, similarly for nitrogen, the rate is $111/3.6\times10^7 \times T/273 \times 1/p_{N_2}$ cubic centimeters per second. Under steady state conditions the rate of formation of each gas must be equal to its rate of loss so:

$$F_{Xe} = \frac{676T}{273 \times 3.6\times10^7 \times p_{Xe}}$$

$$p_{Xe} = 6.9\times10^{-8}\frac{T}{F_{Xe}}$$

And

$$p_{N_2} = 1.13\times10^{-8}\frac{T}{F_{N_2}}$$

If the gas had been assumed to be all krypton instead of xenon the pressure of krypton would be $1.03\times10^{-5}\sqrt{T}$ compared to a xenon pressure of $1.28\times10^{-5}\sqrt{T}$. Thus, higher pressures will be calculated for the assumption of all xenon and, therefore, will be conservative estimates of the total pressure. The molecular weight of cesium is close to that of xenon, so cesium and xenon should behave similarly. The same applies to rubidium and krypton. Therefore, steady state pressures of Xe and N_2 have been calculated and are listed as a function of temperature in table I. Thus, the total steady state pressure is less than 8×10^{-4} atmosphere at all temperatures up through 3000 K. The reader is reminded that the purpose here is to show that the vent is capable of keeping the gases resulting from fissioning at low pressures. Actually, the nitrogen

TABLE I. - STEADY STATE PRESSURES OF

XENON AND NITROGEN

T, K	p_{Xe} (atm)	p_{N_2} (atm)	($p_{Xe} + p_{N_2}$) (atm)
1200	4.4×10^{-4}	3.5×10^{-5}	4.8×10^{-4}
1400	4.8	3.7	5.2
1600	5.1	4.0	5.5
1800	5.4	4.2	5.8
2000	5.7	4.5	6.1
2200	6.0	4.7	6.5
2400	6.3	4.9	6.8
2600	6.5	5.1	7.0
2800	6.8	5.3	7.3
3000	7.0	5.5	7.6

pressures will be higher than those given in table I because of thermal decomposition of the fuel (see appendix D).

The pressures calculated here are consistent with the assumption of molecular flow because the mean free path (L) is such that $L/a > 1.00$ where a is the radius of the vent (ref. 17). For example, at 2800 K this can be shown as follows (ref. 18):

$$L_{Xe} = \frac{1}{\sqrt{2}\,\pi \nu_{Xe} S_{Xe}^2 + 2.4\,\pi \nu_{N_2} S_{XeN_2}^2}$$

ν_{Xe}	number of molecules of Xe per unit volume
S_{Xe}	collision radius of Xe ($\simeq 2.03\times10^{-8}$ cm) (ref. 19)
ν_{N_2}	number of molecules of N_2 per unit volume
S_{XeN_2}	one-half of distance between Xe and N_2 upon collision, cm
S_{N_2}	collision radius of $N_2 \simeq 1.84\times10^{-8}$ cm) (ref. 19)

$$L_{Xe} = \frac{10^{16}}{18.3 \times \nu_{Xe} + 28.3 \times \nu_{N_2}}$$

$$L_{Xe} = \frac{10^{16}}{18.3 \times \frac{N_o p_{Xe}}{RT} + 28.3 \times \frac{N_o p_{N_2}}{RT}}$$

N_o	Avogadro's number
R	gas constant, (cm^3)(atm)/(mole)(deg)
L_{Xe}	0.27 cm

Therefore $L/a = 0.27/(0.0125/2) = 43$ which is > 1.

An important sidelight of these calculations is that the pressure of the nitrogen liberated from UN during the fission process may be greater than the thermal decomposition pressure of nitrogen over UN at temperatures up to about 2200 K (see appendix D). This effect is important in relation to the formation of free uranium in UN and could mean that the liberation of free uranium is not an important factor until the fuel temperature exceeds 2200 K (not 1800 K as calculated in appendix C of this report). But because the amount of nitrogen liberated is based on preliminary studies only, the effect of this nitrogen is not considered further in this report.

APPENDIX C

RATE OF FORMATION OF URANIUM FROM UN AS CALCULATED FROM RATE OF PERMEATION OF NITROGEN THROUGH TUNGSTEN

The loss of nitrogen by permeation through 1 square centimeter of 0.1-centimeter thick tungsten cladding during 10 000 hours is $6\times10^{5}\ P(p_{N_2})^{1/2}$ cubic centimeters (STP) (ref. 6). Substituting 2×10^{-7} $((\mathrm{cm}^3)(\mathrm{mm})/(\mathrm{cm}^2)(\mathrm{min})(\mathrm{atm}^{1/2}))$ for the permeation coefficient (P) of nitrogen through tungsten at 1673 K and 10^{-2} atmosphere for the pressure (p_{N_2}) of nitrogen over UN at 2800 K gives a loss of 1.2×10^{-2} cubic centimeter of nitrogen per square centimeter of cladding. The pressure of nitrogen outside of the cladding is assumed to be zero. The surface area of the reference fuel element is 120 square centimeters. So, the amount of uranium formed in 286 grams of UN after 10 000 hours because of permeation is about 3×10^{-2} gram. This is less than the arbitrary 0.1 gram limit and would be even less for lower cladding and fuel temperatures.

APPENDIX D

FORMATION OF FREE URANIUM IN VENTED FUEL ELEMENTS

Calculation of the Amount of Uranium Formed in 10 000 Hours Against UN Temperature Because of Loss of Nitrogen Through Vent

The rate of loss of nitrogen by molecular flow through the vent is $\mathscr{F}_{N_2}$ moles per second. For uranium vapor the rate of loss is $\mathscr{F}_U$ moles per second. If UN decomposes on evaporation to give U(g) and $N_2(g)$:

$$UN(s) \rightarrow U(g) + \frac{1}{2} N_2(g)$$

then for each mole of N_2 lost through the vent two moles of U(g) will be formed. Therefore, the net rate of accumulation of uranium in the fuel element is $(2\mathscr{F}_{N_2} - \mathscr{F}_U)$ moles per second. From the equation:

$$F = 3638\ KA \left(\frac{T}{M}\right)^{1/2}\ cm^3/sec$$

(see appendix B and ref. 17), it can be shown that

$$2\mathscr{F}_{N_2} - \mathscr{F}_U = \frac{7.39\times10^{-4}}{\sqrt{T}}\left(\frac{p_{N_2}}{2.65} - \frac{p_U}{15.4}\right)\frac{\text{moles of uranium}}{\text{sec}}$$

The partial pressure of uranium (p_U) as a function of T was obtained from reference 20. The partial pressure of nitrogen was obtained from reference 7. Values for the amount of uranium accumulated in the fuel element in 10 000 hours as a function of temperature are listed in table II and are plotted in figure 2. It is clear that, at fuel temperatures over about 2000 K, more than 0.1 gram of liquid uranium will accumulate in the fuel element in 10 000 hours.

TABLE II. - URANIUM ACCUMULATION IN VENTED FUEL ELEMENT CONTAINING UN

T, K	p_{N_2} (atm)	p_U (atm)	Uranium accumulated (g/10 000 hr)
1800	1.5×10^{-8}	6.8×10^{-8}	1.9×10^{-4}
2000	9×10^{-7}	1.3×10^{-6}	3.6×10^{-2}
2200	2.5×10^{-5}	1.4×10^{-5}	1.1
2400	3×10^{-4}	1.1×10^{-4}	1.3×10^{1}
2600	3×10^{-3}	5.6×10^{-4}	1.4×10^{2}
2800	1×10^{-2}	2.4×10^{-3}	4.2×10^{2}

Calculation of the Amount of Uranium Formed in 10 000 Hours at 2480 K For UO_2 Because of Loss of Oxygen Through Vent

For the overall decomposition reaction

$$UO_2 \rightarrow U + O_2$$

it can be seen that for each mole of O_2 lost one mole of uranium is formed. Supposing none of the uranium is lost through the vent, then the maximum possible amount of uranium accumulating in the fuel element can be calculated similarly to the case for UN except that $\mathscr{F}_U$ is set equal to zero and that only one mole of U is formed for each mole of O_2 lost through the vent:

$$F = 3638\ KA\left(\frac{T}{M}\right)^{1/2}\ \mathrm{cm}^3/\mathrm{sec}$$

$$\mathscr{F}_{O_2} = 3638 \times 0.1367 \times 1.2\times10^{-4}\left(\frac{2473}{32}\right)^{1/2}\left(\frac{273}{2473} \times \frac{10^{-8}}{22\ 400}\right)$$

$$= 2.6\times10^{-14}\ \frac{\text{mole of oxygen}}{\text{sec}}$$

or

$$\mathscr{F}_{O_2} = 2.2\times10^{-4}\ \frac{\text{gram of uranium}}{10\ 000\ \text{hours}}$$

This is much less than the amount of uranium formed because of oxygen permeation through the cladding and can be neglected.

Lowest Fuel Temperature at Which Liquid Uranium Will Be Present With Solid UN in the Vented Fuel Element

The relative rates of loss of nitrogen and uranium ($\mathscr{F}_{N_2}/\mathscr{F}_U$) through a vent by molecular flow will be equal to the ratio of the square roots of the molecular weights $\left(M_U^{1/2}/M_{N_2}^{1/2}\right)$ or 2.9. Because the loss of U atoms must be twice that of N_2 molecules to maintain a constant composition of the UN in the fuel element, p_U must be equal to 5.8 p_{N_2}.

For the reaction:

$$UN(s) \rightarrow U(g) + \frac{1}{2} N_2$$

the free energy change is:

$$\Delta G = -RT \ln p_U p_{N_2}^{1/2}$$

$$= -RT \ln \frac{p_U^{3/2}}{5.8^{1/2}} \qquad \text{(D1)}$$

From reference 20:

$$\Delta G = 181\ 400 - 68.7\ T + 5.75\ T \log T\ (1400 - 2000\ K) \qquad \text{(D2)}$$

and

$$\log p_U = 10.32 - \frac{24\ 090}{T} - 1.26 \log T \qquad \text{(D3)}$$

Substituting equations (D2) and (D3) into (D1) and solving for T, the lowest temperature at which liquid uranium phase can appear is found to be about 1800 K. This is the temperature at which the partial pressure of uranium in the vapor over the UN becomes equal to the vapor pressure of liquid uranium. So, at fuel temperatures above 1800 K liquid uranium will be present with the UN in the fuel element vented to vacuum.

APPENDIX E

LOSS OF FUEL THROUGH VENT INTO A VACUUM

Mechanism of Flow of Uranium Vapor from a Gaseous Mixture of Uranium and Nitrogen Through the Vent into a Vacuum

The purpose of this section is to find out the mechanism of flow of the uranium vapor in equilibrium with UN through the reference vent into a surrounding vacuum. Once the mechanism of flow is known, the rate of loss of uranium through the vent can be calculated as a function of the fuel temperature.

For molecular flow the mean free path (L_U) of the uranium atoms must be greater than the radius (a) of the vent (see ref. 17). The mean free path for uranium in a gaseous mixture of uranium and nitrogen can be calculated from the expression (see ref. 18):

$$L_U = \frac{1}{\sqrt{2}\,\pi\nu_U S_U^2 + \sqrt{9.4}\,\pi\nu_{N_2} S_{UN_2}^2}$$

ν_U	number of molecules of U per cubic centimeter
ν_{N_2}	number of molecules of N_2 per cubic centimeter
S_U	collision radius of U $\simeq 1.4\times10^{-8}$ cm
S_{N_2}	collision radius of $N_2 \simeq 1.84\times10^{-8}$ cm
S_{UN_2}	one-half of the distance between U and N_2 upon collision, $1/2\,(S_U + S_{N_2}) \simeq 1.6\times10^{-8}$ cm
L_U	mean free path of uranium molecules (cm)

The values for S_U and S_{N_2} were estimated from information from references 19 and 21 using the approximate empirical relation:

$$2S_U = 1.166\times10^{-8}\,\tilde{V}_{b,liq}^{1/3}$$

$\tilde{V}_{b,liq}$ is the volume in cubic centimeters of one gram-mole of liquid uranium at its boiling point. The density of uranium at its boiling point is estimated as 15.8 gram per cubic centimeter assuming a linear decrease in density with increasing temperature.

$$\tilde{V}_{b,liq} \cong 15 \text{ cm}^3/\text{g-mole}$$

$$S_U \cong 1.4\times10^{-8} \text{ cm}$$

$$S_{N_2} = 1.84\times10^{-8} \text{ cm (from table in ref. 19)}$$

Substituting values for S_U and S_{UN_2} gives:

$$L_U = \frac{10^{16}}{8.7\ \nu_U + 25\ \nu_{N_2}}$$

From the perfect gas law $PV = (\nu/N_o)RT$, ν_U and ν_{N_2} can be expressed in terms of T, the partial pressures of U and N_2 and the constants R and N_o.

$$L_U = \frac{10^{16}\ RT}{N_o(8.7\ p_U + 25\ p_{N_2})} = \frac{1.3\times10^{-6}\ T}{8.7\ p_U + 25\ p_{N_2}}$$

The partial pressures, p_U and p_{N_2}, are obtained as a function of temperature from references 20 and 7.

For molecular flow L_U must be greater than a so:

$$L_U = \frac{1.3\times10^{-6}\ T}{8.7\ p_U + 25\ p_{N_2}} > 0.00625$$

This can be shown to be true for temperatures up through 2800 K.

T, K	p_U (atm)	p_{N_2} (atm)	Mean free path, L_U, cm
2800	2.4×10^{-3}	10^{-2}	0.013
2900	5	5×10^{-2}	.003
3000	8	10^{-1}	.002

Therefore the molecular flow mechanism holds for temperatures up through about 2800 K. At any temperature above 2800 K calculations based on molecular flow will give high results because gas will escape faster by molecular flow than by transition flow or viscous flow.

Loss of Uranium Vapor From UN Fuel by Molecular Flow Through Vent in 10 000 Hours

For molecular flow the rate of loss can be calculated from reference 17.

$$F = 3638\ KA\left(\frac{T}{M}\right)^{1/2}\ \text{cm}^3/\text{sec}$$

for $M = 235$

$$F = 3.98\times10^{-3}\sqrt{T}\ \text{cm}^3/\text{sec}$$

$$\mathscr{F} = 3.98\times10^{-3}\sqrt{T}\times\frac{273}{T}\ \frac{p_U}{22\ 400}\ \text{moles of U/sec}$$

where p_U is the vapor pressure of uranium at temperature T (ref. 20). The loss in grams of uranium in 10 000 hours is:

$$\mathscr{F}t + 4.1\times10^5\ \frac{p_U}{\sqrt{T}}\ \frac{\text{grams of uranium}}{10\ 000\ \text{hours}}$$

Values of $\mathscr{F}t$ have been calculated for temperatures from 1800 to 2800 K and are tabulated below (also see fig. 3):

T, K	p_U (atm)	$\mathscr{F}t\left(\frac{g}{10\ 000\ hr}\right)$
1800	6.8×10^{-8}	6.6×10^{-4}
2000	1.3×10^{-6}	1.2×10^{-2}
2200	1.4×10^{-5}	1.2×10^{-1}
2400	1.1×10^{-4}	9.1×10^{-1}
2600	5.6×10^{-4}	4.4
2800	2.4×10^{-3}	1.8×10^{1}

Calculation of Maximum Temperature for Molecular Flow of UO_2 Vapor in Equilibrium with UO_2 Solid

From reference 18:

$$L_{UO_2} = \frac{1}{\sqrt{2}\,\pi\nu\left(2S_{UO_2}\right)^2}$$

where

L_{UO_2}	mean free path of UO_2 molecules, cm
ν	number of molecules of UO_2 per cm^3
S_{UO_2}	estimated collision radius of UO_2, cm

A value for S_{UO_2} can be estimated (ref. 19):

$$2S_{UO_2} = 1.222\times10^{-8}\,\tilde{V}^{1/2}_{m,solid}$$

where: $\tilde{V}_{m,solid}$ is the molar volume of UO_2 at its melting point ≅ 29 cubic centimeters

$$S_{UO_2} = 1.9\times10^{-8}\ \text{cm}$$

Assuming the perfect gas law for the UO_2 vapor:

$$\nu = \frac{N_o p_{UO_2}}{RT}$$

N_o	Avogadro's number
p_{UO_2}	vapor pressure of UO_2
R	gas constant in cm^3 atm/^oC g-mole

Then:

$$L_{UO_2} = \frac{2.1\times10^{-8}\ T}{p_{UO_2}}$$

For molecular flow

$$L_{UO_2} = \frac{2.1\times10^{-8}\ T}{p_{UO_2}} > 0.00625$$

Calculating L_{UO_2} at several temperatures gives:

T, K	p_{UO_2} (atm)	L_{UO_2}, cm
2800	4×10^{-3}	0.015
2900	10^{-2}	.006
3000	3.5×10^{-2}	.002

Therefore molecular flow of UO_2 through the vent into a vacuum occurs at temperatures up to nearly 2900 K.

Loss of UO_2 Vapor from UO_2 Fuel by Molecular Flow Through Vent in 10 000 Hours

From the equation for molecular flow (ref. 17):

$$F = 3638\ KA\left(\frac{T}{M}\right)^{2}\ cm^{3}/sec$$

where $M = 267$. The rate of loss of UO_2 in grams per 10 000 hours ($\mathcal{F}t$) can be shown to be:

$$\mathcal{F}t = 2.8\times10^{5}\ \frac{p_{UO_2}}{\sqrt{T}}\left(\frac{\text{grams of } UO_2}{\text{10 000 hours}}\right)$$

Values of $\mathscr{F}t$ are tabulated for a series of temperatures below (also see fig. 5):

T, K	p_{UO_2} (atm) (ref. 15)	$\mathscr{F}t\left(\frac{\text{g of } UO_2}{10\ 000\ \text{hr}}\right)$
1800	2×10^{-9}	1.3×10^{-5}
2000	10^{-7}	6.1×10^{-4}
2200	3×10^{-6}	1.8×10^{-2}
2400	5×10^{-5}	2.8×10^{-1}
2600	5×10^{-4}	2.8
2800	4×10^{-3}	2.1×10^{1}

APPENDIX F

THE EFFECT OF AN ARGON ATMOSPHERE ON THE LOSS OF FUEL THROUGH A VENT

Loss of Uranium Vapor From UN Fuel in 10 000 Hours by Diffusion Through a Vent Under an Argon Cover Gas

It is expected that the loss of uranium through the vent of the fuel element can be decreased by using an inert gas like argon to change the mechanism of flow from molecular flow to diffusive flow. The purpose in this section is to calculate the loss of uranium through the vent in 10 000 hours as a function of fuel temperature for pressures of 1 and 10 atmospheres of argon.

A linear concentration gradient of uranium vapor from the inside end of the vent to the outside end is assumed. Also a constant composition mixture of uranium in argon inside the fuel element and pure argon outside the fuel element are assumed.

The diffusion coefficient (D_{UAr}) can be expressed (ref. 19):

$$D_{UAr} = 0.0018583 \frac{\sqrt{T^3\left(\frac{1}{M_U} + \frac{1}{M_{Ar}}\right)}}{p\sigma^2_{UAr}\,\Omega_{D,UAr}}$$

where

D_{UAr}	binary diffusion coefficient between uranium and argon, cm^2/sec
M_U	atomic weight of uranium, g
M_{Ar}	atomic weight of argon, g
T	temperature, K
p	total gas pressure $\cong p_{Ar}$, atm
σ_{UAr}	separation between uranium and argon atom on collision, Å
$\Omega_{D,UAr}$	a dimensionless function of the temperature and the intermolecular potential field, $\varphi_{UAr}(r)$ characteristic of diffusion, which is approximated by Lennard-Jones potential for the interaction between an atom of uranium and an atom of argon

$\Omega_{D,UAr}$ is given as a function of kT/ϵ_{UAr} in a table in reference 19. To make use of this table a value for k/ϵ_{UAr} is needed. An estimate of ϵ_U/k can be obtained from the empirical relationship, $\epsilon_U/k = 1.15\ T_h$, where T_b is the boiling point of uranium (4200 K) and k is the Boltzmann constant. And $\epsilon_{Ar}/k = 124$ K can be obtained from a table in reference 19. Then k/ϵ_{UAr} can be calculated from the expression, $\epsilon_{UAr} = \sqrt{\epsilon_U \epsilon_{Ar}}$. Values of $\Omega_{D,UAr}$ for some temperatures of interest are listed in table III.

TABLE III. - DIFFUSION OF U THROUGH VENT

T, K	$\Omega_{D,UAr}$	pD_{UAr}	p_U (atm)	Jt(p_{Ar} = 1 atm) (g/10 000 hr)	Jt(p_{Ar} = 10 atm) (g/10 000 hr)
1600	1.06	2.0	1.7×10^{-9}	2.8×10^{-7}	2.8×10^{-8}
1800	1.02	2.5	6.8×10^{-8}	1.2×10^{-5}	1.2×10^{-6}
2000	.99	2.6	1.3×10^{-6}	2.2×10^{-4}	2.2×10^{-5}
2200	.96	3.3	1.4×10^{-5}	2.7×10^{-3}	2.7×10^{-4}
2400	.94	4.1	1.1×10^{-4}	2.3×10^{-2}	2.3×10^{-3}
2600	.92	4.6	5.6×10^{-4}	1.3×10^{-1}	1.3×10^{-2}
2800	.91	5.3	2.4×10^{-3}	5.7×10^{-1}	5.7×10^{-2}
3000	.89	5.8	8.1×10^{-3}	2.0	2.0×10^{-1}

Now, $\sigma_{UAr} = 1/2\ (\sigma_U + \sigma_{Ar})$ where σ_U is the collision diameter of uranium in Å and σ_{Ar} is the collision diameter of argon in Å. The value of σ_U can be estimated from (see ref. 19).

$$\sigma_U = 1.166 \times \tilde{V}_{b,liq}^{1/3}$$

$$\tilde{V}_{b,liq} \cong 15\ \text{cm}^3/\text{g-mole}$$

$$\sigma_U \simeq 2.9\ \text{Å}$$

and

$$\sigma_{Ar} = 3.42\ \text{Å}$$

Therefore

$$\sigma_{UAr} \cong 3.2\ \text{Å}$$

Values for pD_{UAr} have been calculated as a function of temperature and are listed in the table.

Now the flux (J) in grams of uranium atoms per second through the vent is:

$$J = D_{UAr} (dC_U/dZ) A \times M_U$$

where (dC_U/dZ) is the concentration gradient in moles of uranium per cubic centimeter per centimeter along the length l of the vent and M_U is the molecular weight of uranium. Using the relation $(dC_U/dZ) = (p_U/l RT)$ then J can be expressed:

$$J = -pD_{UAr} \frac{p_U A M_U}{p l RT}$$

Where:

J grams of uranium through vent per sec

p total pressure of gas in atmospheres, ≅ argon partial pressure (p_{Ar}) because $p_U << p_{Ar}$

p_U partial pressure of uranium vapor inside of fuel element and pressure drop of uranium through the vent

A area of vent, cm^2

l length of vent, cm

R gas constant, $(cm^3)(atm)/(g\text{-mole})(^oC)$

M_U molecular weight of U

From known values of p_U, p_{Ar}, pD_{UAr}, etc. the amount of uranium diffusing through the vent in 10 000 hours (Jt where t is the number of seconds in 10 000 hours) has been calculated for argon pressures of 1 and 10 atmospheres and for a series of temperatures. The results are listed in table III and are plotted on figure 3.

Loss of UO_2 Vapor From UO_2 Fuel in 10 000 Hours by Diffusion Through A Vent Under an Argon Cover Gas

In this section methods and assumptions similar to those of the preceding section are applied to the calculation of the effects of 1 and 10 atmospheres of argon on the loss of UO_2 from a vented fuel element. First,

$$D_{UO_2,Ar} = 0.0018583 \frac{\sqrt{T^3\left(\frac{1}{M_{UO_2}} + \frac{1}{M_{Ar}}\right)}}{p\left(\sigma_{UO_2Ar}\right)^2 \Omega_{D,UO_2Ar}}$$

$$\sigma_{UO_2} = 1.222\ \tilde{V}_{m,sol}^{1/3}$$

where

$\tilde{V}_{m,sol}$	29.1 cm^3 for UO_2
σ_{UO_2}	3.74 Å
σ_{Ar}	3.42 Å
M_{UO_2}	molecular weight of UO_2

Ω_{D,UO_2Ar} is given in reference 19 as a function of kT/ϵ_{UO_2Ar}. ϵ_{UO_2}/k is obtained from the empirical relation, $\epsilon_{UO_2}/k = 1.15\ T_m$, where T_m is the melting point of UO_2 (3103 K). $\epsilon_{Ar}/k = 124$ K is obtained from a table in reference 19. Then k/ϵ_{UO_2Ar} can be calculated from $\epsilon_{UO_2Ar} = \sqrt{\epsilon_{UO_2}\epsilon_{Ar}}$. Values of Ω_{D,UO_2Ar} are listed in table IV.

TABLE IV. - DIFFUSION OF UO_2 THROUGH VENT

T, K	Ω_{D,UO_2Ar}	pD_{UO_2Ar}	p_{UO_2} (atm)	Jt(p = 1 atm), g/10 000 hr	Jt(p = 10 atm), g/10 000 hr
1600	1.10	1.5	2×10^{-11}	2.5×10^{-9}	2.5×10^{-10}
1800	1.06	1.8	2×10^{-9}	2.8×10^{-7}	2.8×10^{-8}
2000	1.02	2.2	10^{-7}	1.6×10^{-5}	1.6×10^{-6}
2200	.99	2.6	3×10^{-6}	5.3×10^{-4}	5.3×10^{-5}
2400	.97	3.0	5×10^{-5}	9.0×10^{-3}	9.0×10^{-4}
2600	.95	3.5	5×10^{-4}	9.6×10^{-2}	9.6×10^{-3}
2800	.95	4.0	4×10^{-3}	8.0×10^{-1}	8.0×10^{-2}
3000	.91	4.5	3×10^{-2}	6.5	6.5×10^{-1}

Values of pD_{UO_2Ar} have been calculated and listed in the table. Then the flux (J) in grams of UO_2 molecules through the vent per second is:

$$J = -D_{UO_2Ar} \frac{dC_{UO_2}}{dZ} A \times M_{UO_2}$$

where: dC_{UO_2}/dZ is the concentration gradient in moles of UO_2 per cubic centimeter of gas per centimeter along the length l of the vent; and M_{UO_2} is the molecular weight of UO_2. Putting

$$\frac{dC_{UO_2}}{dZ} = \frac{p_{UO_2}}{l RT}$$

then J can be expressed:

$$J = pD_{UO_2Ar} \frac{p_{UO_2} A M_{UO_2}}{p l RT}$$

The calculated values of Jt as a function of temperature for $t = 3.6 \times 10^7$ seconds and $p = 1$ and 10 atmospheres are listed in the table and are plotted on figure 5.

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