VENTING OF FISSION PRODUCTS AND SHIELDING IN THERMIONIC NUCLEAR REACTOR SYSTEMS*

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Most thermionic reactors are designed to allow the fission gases to escape out of the emitter. There exist several design variations to allow the gas to escape from the reactor core. The final disposition of the gas has several interesting problems. If the gas is put into a chamber, its volume is too large to be contained in the shield. Fortunately the fission gas do not pose a radiation problem at 100 ft. Because of the long half-life the fission gases could prevent any close approach to the reactor after it has been shut down.

The fission gases could instead be stored in an absorption trap. An absorption trap is usually assumed to require very low temperatures, however, experimental data indicate that reservoirs at several hundred degrees C can be useful.

Another scheme is to simply allow the fission gases to escape. Again, because of the low activity of the fission products, this method should pose no radiation hazards.

INTRODUCTION

A thermionic reactor has an interesting radiation problem generally not associated with other nuclear systems, that is, the disposal of gaseous fission products. Almost all designers of thermionic reactors assume that the gaseous fission products must be removed from the core. This assumption is due to the very high operating temperatures of the fueled emitter in a thermionic diode. In order to illustrate the problem an idealized (not real) schematic of an emitter is shown in Fig. 1. This hypothetical emitter consists of a heavy (~ 1-mm-thick) tungsten cladding surrounding the fuel which usually consists of UO_2 or UC. If UO_2 is used, a void amounting to ~ 10% of the fuel volume is created in the fuel after prolonged high-temperature operation. A typical set of operating conditions is shown in Table I.

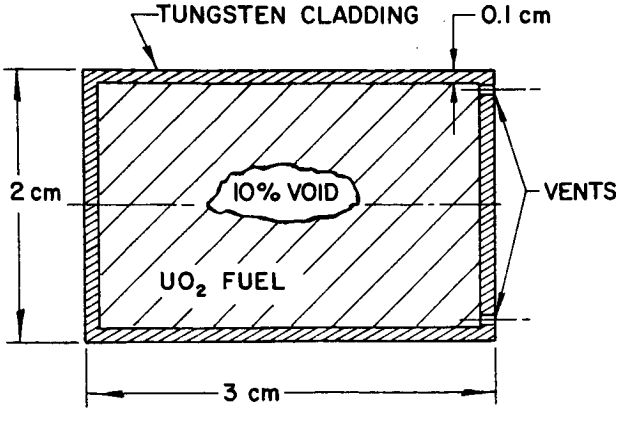

Fig. 1. Hypothetical tungsten-clad fuel emitter.

TABLE I. TYPICAL EMITTER OPERATING CONDITIONS

Emitter surface temperature, °C	1700
Emitter center-line temperature, °C	2500
Fuel power density, W/cm^3	50
Total fissions/cm^3 after 10^4 h	7.1×10^{19}
^{235}U burnup, %	~ 0.24
Final gaseous fission-product pressure, psi	~ 1000
Time to 2.5% creep of cladding, h	10

Experiments have shown that at an operating temperature of ~ 1700°C almost all gaseous fission products, e.g., xenon and krypton, escape from the solid fuel and accumulate in the central void. After 10,000 h of operation the gaseous fission product pressure in the void would be ~ 1000 psi. This high pressure would induce high hoop stresses in the tungsten cladding with resultant creep and eventual shorting of the emitter to the collector. The required creep time until shorting occurs is ~ 10 h. Obviously, this is unsatisfactory. Most emitter designs are affected by this difficulty and therefore allow the fission gases to escape. The design of emitters which would permit such escape before the cladding swells presents some problems. Solutions to these difficulties have apparently been found, but are not the subject of this paper.

RELEASE OF FISSION PRODUCTS FROM THE REACTOR

The fission products from the reactor core can be released by several methods. Two are illustrated in Fig. 2, which shows a typical thermionic fuel element of the flashlite design. The fission gases, on leaving the fueled emitter, enter the emitter support. In Method A, shown in the lower half of

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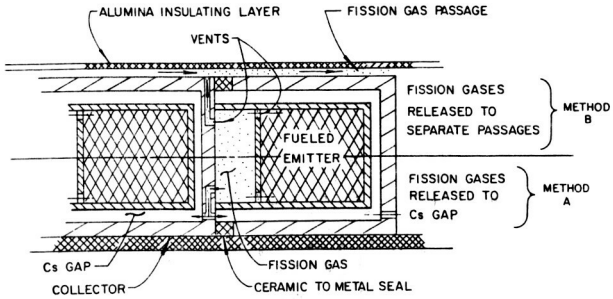

Fig. 2. Typical thermionic fuel element and possible methods of fission-gas release.

the illustration, vents are leading from the support structure to the cesium gap. The cesium gaps of the individual diodes are then connected by separate vents throughout the length of the fuel rod.

In Method B, illustrated in the upper half of Fig. 2, the vents bypass the cesium gap and lead to separate fission-gas passages outside the collector in the Al_2O_3 insulating region. These passages also extend throughout the full length of the fuel rod.

Fission Gases Enter the Cesium Gap - Method A

Both methods have their disadvantages. Experiments with Method A show that the gaseous fission products degrade the electrical power output of the diode even after only a short operating time: the presence of xenon or krypton in the cesium vapor at more than 10 or 20 Torr can increase the electrical resistivity across the cesium vapor gap and thereby reduce the electrical power output. Also, the presence of foreign gases increases the thermal conductivity across the gap, which, in turn, results in a reduction of the emitter temperature. The final result is the fact that only a very small fraction of the gaseous fission products can be retained in the cesium vapor space.

Fission Gases Bypass the Cesium Gap - Method B

Method B offers the advantage of not restricting the fission-product pressure to 10 or 20 Torr. This pressure restriction can influence the fission gas storage problem outside the reactor core. There are some disadvantages to the method. The small passages running through the alumina outer sheath might contain cracks connecting the collector and the outer sheath. The gaseous fission products are not purely xenon and krypton but also contain many other elements. In particular, cesium fission products are about 70% as numerous as the xenon and krypton products. Therefore, cesium at high pressure is present in any cracks in the separate venting system. With approximately 10 V possible across the small passage, there may be electrical shorting between the collector and the outer sheath resulting in very low electrical power output of the fuel element.

Another disadvantage of Method B is the presence of many ceramic-to-metal seals in the fuel element (Fig. 2). Should any of these break, the high-pressure gaseous fission products could enter the cesium vapor gap and reduce or terminate the power output. Because of this latter possibility and because of the large number of seals, the separate venting system should preferably be limited to the 10- or 20-Torr pressure regime. From these considerations, a separate fission-gas venting system does not appear to offer major advantages.

FISSION-GAS STORAGE PROBLEMS

Assume that the fission products, after venting, are finally stored in an outside reservoir. Fission products that may be gaseous at an emitter temperature of ~ 2000°C are listed, together with their atomic percentages, in Table II. Note that the rare gases represent ~ 12% of the fission products while the total high-temperature gases amount

TABLE II. GASEOUS FISSION PRODUCTS AT 2000°C AND THEIR PART OF TOTAL GAS RELEASE

Fission Products	Amount of Species, At. %
Kr and Xe	12.2
Br and I	2.5
Rb and Cs	9.0
As, Se, Te	4.3
Total	28.0

to almost 30%. In assessing whether all these gaseous products can escape from the emitter, some understanding of the escape mechanism may be useful. A microphotograph of UO_2 fuel after several thousands of hours of operation at ~ 2000°C is shown in Fig. 3; the dark spots are voids and the gray material is UO_2. Many dark spots have a lenticular shape and seem to have developed a trail through the UO_2.

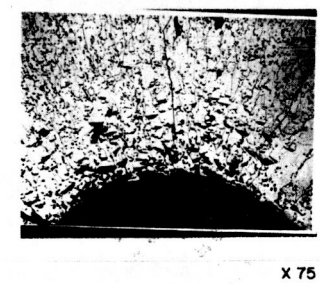

Fig. 3. Microphotograph of UO_2 fuel after prolonged operation at 2000°C.

According to fission-product escape theory voids are formed during operation, move up the temperature gradient through the fuel, sweep up the gaseous fission products, and eventually deposit the gases in the center of the fuel. In other words, the fission products do not have to diffuse long distances. This would imply that all fission products with a substantial vapor pressure at ~ 2000°C could be swept up by the lenticular voids and escape from

the fuel as readily as either xenon or krypton. Experimental data on these problems at present are not abundant.

The postulated escape mechanism out of the emitter can be considered to have a half life, which has been estimated to be about one week to one month for all fission products that can escape. The value of one week is used throughout this study.

After the high-temperature gaseous fission products escape from the emitter they may condense on cool surfaces both inside and outside the reactor. The escaped products can be bled through various filters or absorption beds, which could either react with or condense these products into a relatively small volume located within the reactor radiation shield. The rare gases, however, may not be trapped by these devices and may require some other method of disposal. To illustrate the problem, consider Table III which shows the results of a few simple calculations. A 100-kWe thermionic reactor with

TABLE III. GENERATION OF RARE GASES IN TYPICAL FUEL EMITTERS

Reactor thermal power, MW	1
Fissions produced during 10^4 h	1.1×10^{24}
Total number of xenon and krypton atoms	2.6×10^{23}
Gas volume at STP, liter	9.8
Gas volume at 20 Torr and 600°C, liter	1.2×10^3
Diameter of equivalent sphere, m	1.32

10% efficiency would have a thermal power of 1 MW. In 10,000 h of operation there would have been 1.1×10^{24} fissions. Assuming that 12% of the fission products are rare gases, there would be 2.6×10^{23} atoms of xenon and krypton or 0.44 mole. At standard temperature and pressure the gas volume would be 9.8 liters. However, in most reactor system designs a large radiator operates at ~ 600°C in the immediate vicinity of the reactor so that a large volume at room temperature may be difficult to locate. Also, as mentioned earlier, a rare-gas pressure greater than ~ 20 Torr may not be acceptable because of restrictions imposed by the thermionic diodes. Assuming the two worst conditions, i.e., a pressure of 20 Torr and a temperature of 600°C, the gas volume becomes 1190 liters rather than ~ 10 liters. A sphere of this large volume would have a diameter of 132 cm or of nearly 4.5 ft. Such a gas volume is almost an order of magnitude larger than the reactor and obviously could not be accommodated inside the reactor radiation shield.

POSSIBLE SOLUTIONS TO STORAGE PROBLEM

Three solutions to the fission-product storage problems exist:

- Remove the gaseous products from the main shield to a special shielded tank.
- Absorb the gases in a charcoal trap.
- Let the fission gas escape to space.

Remove the Gases to a Special Shielded Tank

In examining this method, the question of the existing radiation level arises immediately. To investigate the problem an example was postulated. Assume again a 1-MW thermal reactor and a half-life of one week for gaseous fission products to leave the emitter and calculate the activity of the various fission products. Decays per second as a function of operating time are plotted in Fig. 4. The top curve shows the fission-product activity remaining in the reactor; the middle curve plots the activity of the fission products which escape from the emitter but are assumed to be collected in a trap; and the bottom curve shows the activity of the rare gases which have escaped from the emitter and are assumed not to be caught in the trap.

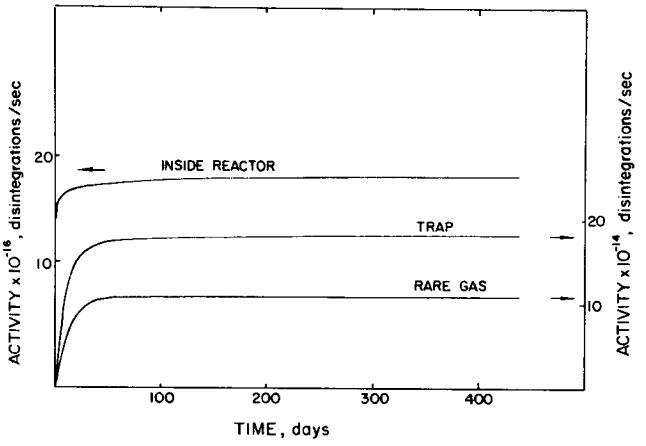

Fig. 4. Activity of various fission products as a function of reactor operating time.

The curves show that a steady state is achieved after two months of reactor operation. The activity in the trap would be 1.8×10^{15} disintegrations/sec or ~ 50,000 Ci. However, this trap was assumed to be small and located within the reactor shield. The rare gas value is 1.1×10^{15} disintegrations/sec which represents 30,000 Ci. To calculate the roentgens this source represents, the activity and decay schemes of the various isotopes of xenon and krypton were investigated. More than 90% of the activity of the rare gases is due to ^{133}Xe and ^{135}Xe (Fig. 5). In each case there is a metastable state; however, a detailed investigation of the decay chain shows that the metastable states contribute little to the activity. The ^{133}Xe is about six times more active than ^{135}Xe; however, the 81-keV γ-ray is strongly internally converted and because of the high absorption cross section it may be readily absorbed in a gas container wall. The main contribution to the outside γ-radiation therefore probably comes from ^{135}Xe. If one assumes that the β-rays do not leave the fission-gas container, and taking into account the internal conversion of the γ-rays, this rare-gas source would produce about 500 R/h at one meter or ~ 500 mR/h at 100 ft.

In some directions around a reactor power supply this radiation level is acceptable; however, in the direction of the crew compartment it is excessive. The activity calculations assumed an escape lifetime from the emitter of one week. Because the two

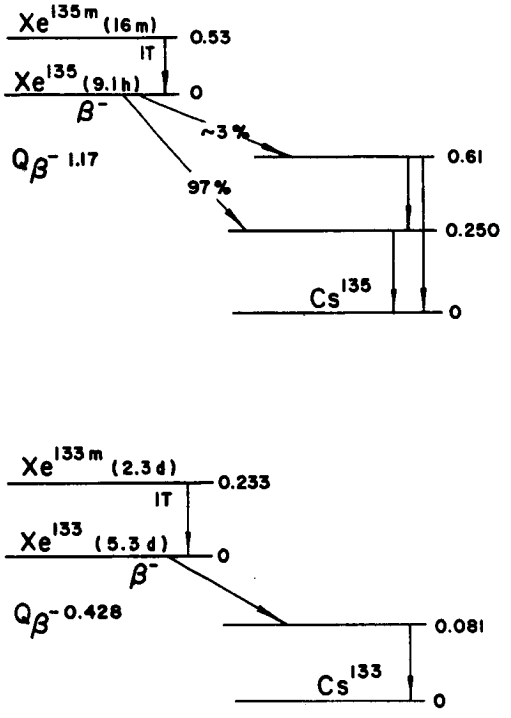

Fig. 5. Decay schemes for ^{133}Xe and ^{135}Xe.

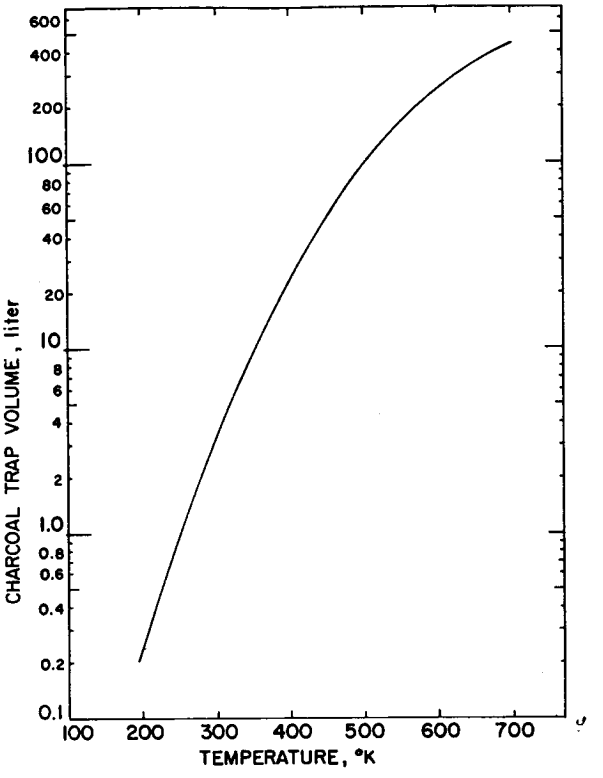

Fig. 6. Amount of xenon absorbed in charcoal as a function of temperature.

isotopes of greatest interest, ^{133}Xe and ^{135}Xe, have half-lives of 9 h and 5.3 days, respectively, the large uncertainty in the value of the escape lifetime can significantly affect the calculated radiation level. Experimental investigations of escape rates out of emitters are required before a satisfactory value can be assigned to the radiation level. In any case, the storage of fission gases in a lightly shielded gas chamber can pose design problems.

Absorption in a Charcoal Trap

Normally, absorption traps are thought to operate at very low temperatures; however, at pressures of 20 Torr a charcoal trap can be useful even at moderate temperatures. For the example of a 1 MW thermal reactor, the volume of charcoal required to store the xenon and krypton at 20 Torr can be calculated as a function of temperature.[1] As shown in Fig. 6, this charcoal volume is considerably smaller than the original volume of 1.2×10^3 liters without charcoal as shown in Table III. At a temperature of 100°C the charcoal volume is about 15 liters. Although 15 liters is not a trivial volume to shield, it may represent a compromise solution. At 100°C, the use of heat pipes may prove feasible for controlling the charcoal trap temperature.

The two methods discussed above of handling fission gases have their disadvantages. If the charcoal-trap temperature rose appreciably, the fission-gas pressure could increase and pose a hazard to reactor operation. On the other hand the lightly shielded gas-chamber system could pose a radiation hazard after reactor shutdown. Figure 7

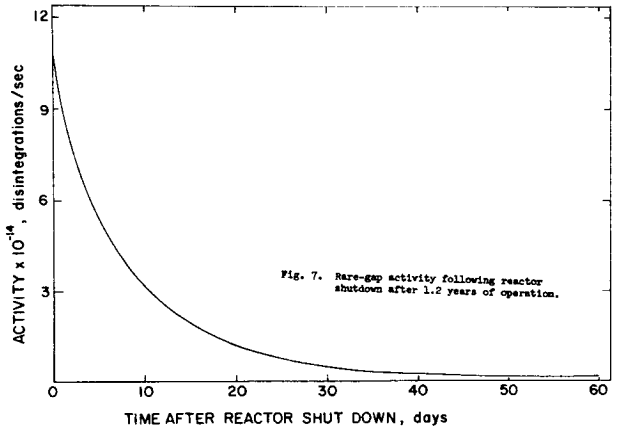

Fig. 7. Rare-gap activity following reactor shutdown after 1.2 years of operation.

shows the rare-gas activity expected following reactor shutdown after 1.2 years of operation. The curve shows that after one month the activity has fallen by a factor of 100. At this time the activity is due mainly to ^{85}Kr which has a ten-year half-life. Although this activity is small compared to the previously cited values, it is a 5-R/h radiation source at a distance of 1 m. With a ten-year half-life it does not disappear suddenly. If poorly shielded, it might pose a hazard for any crew working near the reactor after shutdown.

Escape of Gases to Outer Space

Values for fission gases escaping to space have been calculated, Table IV, and show that the number of rare-gas atoms escaping per second would be 7.5×10^{15}. These escaping atoms have an activity of 1.5×10^{10} disintegrations/sec, which represent 0.5 Ci leaving the reactor per second. Although at first glance this value seems high, the radiation hazard is actually small. Assume that the gases are emitted by an orifice to form a radioactive cloud filling 2π radians. As the cloud expands, the atom density at a distance of 1 m from this low-density expanding radioactive cloud, the radiation dose would be $\sim 10^{-8}$ R/h and certainly should pose no radiation hazard.

TABLE IV. ACTIVITY OF RARE GASES RELEASED TO SPACE

Atom escape rate, atom/sec	7.5×10^{15}
Activity of escaping atoms, disintegration/sec	1.5×10^{10}
Radiation leaving reactor, Ci/sec	0.5
Atom density at 1 m, atom/cm^3	1.5×10^{7}
Atom mean free path, km	200
Dose rate a 1 m from cloud, R/h	$\sim 10^{-8}$
Moles of ^{85}Kr released per year, mole/yr	7×10^{-3}
1975 projection for ^{85}Kr release, mole/yr	~ 500

Another aspect of this method of disposal of fission gases is simply that radioactivity is being released. The isotope that is worrisome is the long-lived ^{85}Kr. This 1-MW reactor would release about 7×10^{-3} moles of ^{85}Kr per year. This value must be contrasted with other sources of ^{85}Kr. At present, the ^{85}Kr obtained from the reprocessing of power reactor fuel elements is allowed to escape out of the air vents. No attempt is made, at present, to prevent their escape. Assuming that 100% of the ^{85}Kr is released, the escape rate in a few years is projected to be about 500 moles per year. By contrast, the 1-MW reactor is only a very small source.

Conclusions

This study led to the following conclusions:

- Release of rare-gas fission products to space is desirable, and
- Their release does not constitute a safety hazard.

If however, for some reason, the gases should not be released to space, design variations are possible to store the rare-gas fission products and to solve the associated radiation problems.

REFERENCE:

1. "Gas-Cooled Fast Breeder Reactor," GA-9359, Quarterly Progress Report, Feb. 1 - April 30, 1969.