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EVALUATION OF OFF-GAS SYSTEM FOR THE CP-ORR CAPSULE EXPERIMENT

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ABSTRACT

The performance of the off-gas system of the CP-ORR (Coated Particle-Oak Ridge Research Reactor) Capsule Experiment is evaluated both with and without an activated charcoal trap by computing the possible atmospheric radiation doses that would result from a fission product release and comparing these doses with permissible atmospheric doses. Both continuous release and instantaneous release of accumulated fission products from the UC_2 fuel particles were considered. By the addition of the charcoal trap it is demonstrated that a possible total body radiation dose of 273 mr may be reduced to 3 mr which is 30% of the permissible ORNL atmospheric dose. It is concluded that the activated charcoal trap, as designed, will process released fission products satisfactorily and no undue atmospheric radiation hazard will be produced.

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I. Introduction

The purpose of this memorandum is to evaluate the performance of an activated charcoal trap designed for the off-gas system of the CP-ORR capsule experiment. The adequacy of design is to be determined by computing the possible atmospheric radiation doses that would result both with and without the charcoal trap and comparing these computed doses with permissible ORNL atmospheric doses.

These irradiation experiments are being conducted in support of the Pebble-Bed Reactor concept (8) and are designed to study the operating capabilities of UC_2 fuel specimens. A fuel specimen consists of pellets of fuel material stacked inside a loosely fitting graphite sleeve. The graphite sleeve is then enclosed by a capsule of stainless steel and the entire capsule is immersed in a NaK bath for temperature control. The fuel material consists of graphite-coated UC_2 particles distributed in a matrix of graphite. The annular space between the fuel pellets and the graphite sleeve is swept continuously by a flow of helium and/or argon gas. The experimental system will consist, basically, of four individually controlled capsules suspended in the core of the Oak Ridge Research Reactor. The sweep gas flow from each capsule assembly is to be sampled, processed through an activated charcoal trap, and then routed to the ORR experiment off-gas system which empties into the 3039 gas disposal stack. Provisions are included with which to terminate gas flow automatically upon indication of significant amounts of radioactivity being released from the irradiation capsule assembly.

II. Hazards of Fission Product Release

Release of fission products from these capsules is to be expected since a study of the characteristics of fission product release from this type of fuel

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matrix is one objective of the experiment. Fission products released by failure of a fuel specimen in this experiment could produce either of two hazards depending upon the severity of the failure. If the integrity of the graphite coatings is violated, then fission products will be present in the sweep gas stream and will be carried into the atmosphere via the off-gas system and the 3039 gas disposal stack. This release of radioactivity could produce an atmospheric radiation hazard to laboratory personnel as a whole. If graphite coating failure is accompanied by experiment containment rupture, then it may be possible for fission products to enter the working area of the reactor building producing a hazard to personnel concerned with operation of the reactor and the experiment. This type of accident would produce a hazard many times greater than would the release of fission products into the off-gas system where a much higher dilution factor is accomplished by release to the atmosphere through the gas disposal stack.

It is assumed that the probability of experiment containment failure is minimized by the design of the experimental system, and further, that the probability of graphite coating failure is much greater because of the nature of the experiment, i. e., to study the operating capabilities of graphite-coated UC_2 fuel specimens. Therefore, this evaluation will consider only the atmospheric radiation hazard that could result from graphite coating failure with subsequent release of fission products into the atmosphere via the experiment off-gas system and the 3039 gas disposal stack. While the possibility of a fission product release into the reactor building does exist, and proper steps should be taken to prevent this occurrence, a study of that type of accident is considered beyond the scope of this evaluation of the experiment off-gas system.

Disposal of gaseous or very finely divided fission products can be effected by discharge into the atmosphere through a gas disposal stack provided the meteorological conditions are favorable and the amount of radioactivity involved is not too large. This "safe" disposal is based upon the premise that the fission products are diluted by a sufficient volume of air so that the resulting air concentration is very low. If the release of radioactivity should occur under adverse meteorological conditions when the disposal efficiency of the gas disposal stack is low, then it is possible for the fission products to return to ground level without sufficient dilution and laboratory personnel could be exposed to both an internal and external radiation hazard.

To calculate the air concentration of the various isotopes, at ground level, the efficiency of the gas disposal stack must be estimated. Under adverse meteorological conditions, as will be assumed for this evaluation, a stack factor of 1.8×10^{-5} sec/M^3 for the 3039 gas disposal stack will be used (5). The product of the stack factor and the number of curies of each isotope released gives an estimation of the integrated ground concentration in the units, curie·sec/M^3.

It has been proposed that the maximum release of radioisotopes to the atmosphere from the entire laboratory operation be limited to such an amount that the resulting radiation dose to laboratory personnel be not greater than 10% of the maximum permissible dose over a 40 hour work week (4). This 10% dose will be referred to as the ORNL permissible atmospheric radiation dose. Since the release is assumed to be composed of numerous isotopes, it is not immediately evident whether the body as a whole or some individual organ will be considered the critical organ, that is, the organ most affected by the release. Therefore, the following organs were selected for individual dose calculations: total body,

lung, kidney, GI tract, thyroid, and bone. The ORNL atmospheric radiation doses (10% of the weekly permissible dose) upon which this hazard evaluation is based are taken to be: (1) 10 mr for total body (external and internal dose), (2) 30 mr for the lung, kidney, bone, or GI tract, (3) 60 mr for the thyroid. On this basis, it is possible to obtain a calculated integrated ground concentration (curie.sec/M^3) for each isotope in air from which a person would receive the permissible ORNL atmospheric dose. These values were calculated from maximum permissible concentration in air (MPCa) data for the various isotopes as listed in NBS Handbook No. 69 (7), and have been reported in a previous evaluation of this type (1). By comparing the ground concentration of each isotope resulting from the release with the ground concentration of each isotope required to produce the permissible ORNL atmospheric radiation dose to an organ, a fractional dose from each isotope is obtained. These fractional doses from individual isotopes are then summed and a total dose is determined in terms of the permissible atmospheric dose for each of the above listed organs assumed to be the critical organ for the release.

III. Experiment Off-Gas System

Transport of fission products released from the fuel specimen will be accomplished by passage of helium and/or argon through the annular space existing between the fuel pellets and the loosely fitting graphite sleeve. Upon leaving the capsule assembly the sweep gas passes through a gas holdup volume, a sampling facility, a gas flow restrictor, a charcoal trap and then is discharged to the ORR experiment off-gas system. The off-gas system must therefore be capable of processing (1) a continuous release of small quantities of fission products and (2) a sudden release of fission product content of one fuel specimen capsule as

a result of, for example, catastrophic oxidation of the graphite matrix. To process a sudden release of fission product gases the charcoal trap will have a capability of containing these gases until the sweep gas flow can be automatically terminated or, in the event of mechanical failure, can be manually terminated.

For maximum flexibility it was decided to provide each capsule assembly with an individual charcoal trap rather than having one large trap to process the off-gas from the four separate capsules. Preliminary calculations indicated that a charcoal trap containing approximately 2 lbs. of activated charcoal should be sufficient to process either a continuous or an instantaneous release of fission products from one fuel specimen capsule. The adsorption characteristics of activated charcoal for krypton, xenon, and iodine have been demonstrated (2,6). The charcoal trap will also serve to remove particulate matter from the gas stream with high efficiency by virtue of the small charcoal mesh size and the low linear velocity of the gas stream. Either Pittsburgh PCB or Columbia HCC, 12/30 mesh, activated charcoal is suggested for this trap.

Over the course of this experimental program some factors will vary over rather wide limits. For example, the U-235 content of the capsules may vary from 1.2 to 6.5 grams; the coolant gas may be helium, argon, or mixtures thereof; the purge gas flow rate may vary from 1 to 10 cc/sec; and the transit time from fuel capsule to charcoal trap can vary from 38 sec to 35 min depending upon the purge gas flow rate and the gas holdup volume utilized. Depending upon the various combinations of these factors, the charcoal trap may have many various holdup times. For example, when operating under normal conditions, with the helium flow of 1 cc/sec at a temperature of 80°F, the trap is designed to provide a holdup of krypton and xenon for 6.9 and 69 hours, respectively. With an argon

flow rate of 1 cc/sec these holdup times will be reduced to 5.1 and 51 hours, respectively. Holdup time is defined as the length of time required for fission gases to first be detected in the effluent gases from the charcoal trap. The time required for all of a given quantity of gaseous activity to pass through the trap would be approximately 21 and 210 hours for Kr and Xe with the trap operating with a helium purge gas rate of 1 cc/sec. For this evaluation it will be assumed that all fission gases emerge at the holdup time. To evaluate the operation of charcoal traps using all possible combinations of the experimental variables would be impossible. Therefore, the combination of variables that would produce the more extreme demands on charcoal trap operation will be selected for this evaluation. If it can be demonstrated that the trap will operate satisfactorily under extreme conditions, then it should perform satisfactorily under the less severe conditions.

III. A. Sudden Release of Accumulated Fission Products

The radiation hazards involved and the function of the charcoal trap in reducing these hazards will be demonstrated by first examining the radiation doses to the various body organs produced by a direct release of fission products into the atmosphere by way of the gas disposal stack, then by a release processed through a charcoal trap operating under the more extreme conditions and finally, a release processed under the less extreme conditions or "normal" conditions. All releases are assumed to occur during adverse meteorological conditions when the disposal efficiency of the 3039 gas disposal stack is at a minimum. An ORACLE computer program developed for a previous evaluation of this type was used to facilitate dose calculations.

The maximum accident for this type of release is postulated to occur in the form of a gross oxidation of a fuel specimen containing 6.5 grams of U-235 after having operated for a period of six months in a thermal neutron flux of 8×10^{13}

$n/cm^2/sec$. It is assumed that fission products have accumulated in the fuel specimen and none have escaped prior to the accident. It is assumed that all of the fission gases, 50% of the iodine, barium, cesium, ruthenium, strontium, tellurium, and yttrium isotopes, and 5% of the remaining isotopes are released from the fuel specimen by oxidation of the graphite matrix (8). The number of curies of each isotope assumed to be released from the fuel specimen was calculated from data of Blomeke and Todd (3) and is contained in Table I.

The amounts of various fission products which will deposit or plate-out on the metal surfaces of the capsule assembly or the metal surfaces of the off-gas system are difficult to estimate with any reasonable degree of accuracy. So, for the purpose of this evaluation, it will be assumed that no deposition occurs and that any reduction in concentration will be accomplished solely by the clean-up provisions of the off-gas system.

Dose calculations assuming releases as described below were made and the results together with the percentage of the permissible ORNL atmospheric radiation dose are contained in Table II.

Case I - Direct release to the atmosphere via the ORR experiment off-gas system and the 3039 gas disposal stack. No provisions in the experiment off-gas system for fission product trapping.

Case II - Fission product release is processed through the activated charcoal trap. Purge gas is argon at a flow rate of 10 cc/sec and a transit time of 38 sec exists between fuel specimen and charcoal trap. Holdup of krypton and xenon is 0.51 and 5.1 hours, respectively. Iodine removal efficiency is 99.99% and particulate removal efficiency is 99.9%.

Case III - Fission product release passed through the activated charcoal trap. Purge gas is helium at 1 cc/sec, transit time between fuel specimen and charcoal trap is 35 min., holdup time for Kr and Xe is 6.9 and 69 hours, respectively. Iodine and particulate efficiencies identical to those in Case II.

It should be noted that the above three cases assume complete purging of fission products through the charcoal trap. This is to be considered abnormal operation and normally termination of the experiment will occur in such a length of time that very little, if any, of the fission product inventory will escape into the atmosphere via the off-gas system.

III. B. Continuous Release of Fission Products

Since one purpose of this experiment is to determine leak rates or characteristics of the various fission products from graphite-clad UC_2 particles, accurate fractional release data are not available for use in this evaluation. Therefore, release data for alumina-coated UO_2 particles as listed in ORNL-CF-60-10-63 will be used (8). These are listed in Table III as R/B values, defined as rate of release divided by rate of buildup of individual fission products.

To adapt the ORACLE computer code to the continuous release case it was necessary to assume that equilibrium had been achieved in the production of all isotopes of interest. Also it is assumed that the recipient of the atmospheric dose is exposed for the 40 hour week and that the adverse meteorological conditions exist for the 40 hour period. All these assumptions will tend to produce a larger calculated radiation dose than would actually be produced in practice.

Dose calculations assuming releases as described below were made and the results together with percentages of the permissible ORNL atmospheric radiation doses are contained in Table IV.

Case A - Off-gas conditions identical to Case I.

Case B - Conditions identical to Case II. Dose produced by A^{41} also considered in this case.

Case C - Conditions identical to Case III.

IV. Conclusions

It is evident from examination of Table II that a fission product trapping system is required for this experiment to process a sudden release of fission products if it is not possible to terminate the sweep gas flow. The doses computed for Cases II and III may appear to represent a large fraction of the permissible atmospheric dose, but it should be noted that rather unlikely conditions are necessary to produce such doses. It is anticipated that the situation covered by Case B and C of Table IV will be considered the range of normal operating conditions for this experiment. A charcoal trap as designed should process fission gas releases of this type satisfactorily.

Table I. Fission Products Assumed Released thru Failure of One CP-ORR Capsule*

Isotope	Half-life	Curies	Isotope	Half-life	Curies
Kr-83m	114m	72.8	Zr-97	17h	65.0
Kr-85m	4.36h	308.7	Nb-95	35d	50.5
Kr-85	10.27y	2.0	Nb-97	72.1m	46.6
Kr-87	78m	565.9	Mo-99	67h	64.8
Kr-88	2.77h	752.6	Te-99m	6.04h	6.1
Kr-89	3.18m	857.9	Ru-103	41d	29.8
Xe-131m	12d	6.0	Ru-105	4.5h	8.4
Xe-133m	2.3d	31.6	Rh-103m	54m	17.8
Xe-133	5.27d	1375.1	Rh-105	36.5h	9.4
Xe-135m	15.6m	374.3	Te-127m	9.3h	23.2
Xe-135	9.13h	109.7	Te-127	9.3h	4.4
Xe-137	3.8m	1076.4	Te-129m	33d	34.7
Xe-138	17m	1137.4	Te-129	72m	72.5
I-131	8.05d	292.5	Te-131m	30h	61.0
I-132	2.4h	398.9	Te-132	77h	450.3
I-133	20.8h	657.2	Cs-137	26.6y	8.1
I-134	52.5m	502.1	Ba-140	12.8d	652.0
I-135	6.68h	584.9	La-140	40.2h	64.4
Sr-89	54d	431.3	Ce-141	32d	59.4
Sr-90	28y	8.2	Ce-143	32h	64.5
Y-90	64.5h	7.8	Ce-144	290d	21.2
Y-91m	51m	159.2	Pr-143	13.7d	66.2
Y-93	10h	629.3	Pm-147	2.6y	2.9
Zr-95	63d	56.2	Pm-149	54.h	13.6

*Containing 6.5 gms U^{235}.

Table II. Calculated Atmospheric Radiation Doses Produced by Disposal of Fission Products from One CP-ORR Capsule*

	Doses (mr)					
	Krypton	Xenon	Iodine	Solids	Total	% of Permissible[c]
Case I						
Total Body	21.5	14.0	7.8	229.7	273.0	2730.0
GIT (insol)[a]	21.5	14.0	236.3	1202.1	1473.9	4913.0
GIT (sol)[b]	21.5	14.0	24.7	580.5	640.7	2135.7
Bone	21.5	14.0	8.0	2457.9	2501.4	8338.0
Kidney	21.5	14.0	-	128.7	164.2	547.3
Lung	21.5	14.0	72.5	2237.0	2345.0	7816.7
Thyroid	21.5	14.0	4751.9	-	4787.4	7979.0
Case II						
Total Body	13.5	2.0	-	0.2	15.7	157.0
GIT (insol)	13.5	2.0	-	1.2	16.7	55.7
GIT (sol)	13.5	2.0	-	0.6	16.1	53.7
Bone	13.5	2.0	-	2.5	18.0	60.0
Kidney	13.5	2.0	-	0.1	15.6	52.0
Lung	13.5	2.0	-	2.2	17.7	59.0
Thyroid	13.5	2.0	0.5	-	16.0	26.7

NOTE: A^{41} produced in argon sweep gas contributes an additional dose of 0.04mr.

Case III						
Total Body	1.6	1.2	-	0.2	3.0	30.0
GIT (insol)	1.6	1.2	-	1.2	4.0	13.3
GIT (sol)	1.6	1.2	-	0.6	3.4	11.3
Bone	1.6	1.2	-	2.5	5.3	17.7
Kidney	1.6	1.2	-	0.1	2.9	9.7
Lung	1.6	1.2	-	2.2	5.0	16.7
Thyroid	1.6	1.2	0.5	-	3.3	5.5

a Assuming fission products are insoluble.
b Assuming fission products are soluble.
c Permissible ORNL atmospheric radiation dose per 40 hour week.
* Containing 6.5 gms U^{235}.

Table III. Values for Continuous Fission Product Release from Coated UC_2 Particles

Isotope	R/B*	Isotope	R/B
Kr-83m	6.0×10^{-5}	Sr-89	1.5×10^{-6}
Kr-85m	6.1×10^{-5}	Sr-90	1.9×10^{-7}
Kr-85	5.0×10^{-2}	Y-90	1.9×10^{-7}
Kr-87	1.3×10^{-5}	Y-91m	1.3×10^{-6}
Kr-88	3.4×10^{-5}	Y-91	1.3×10^{-6}
Kr-89	1.3×10^{-7}	Y-93	$<1.0 \times 10^{-9}$
		Zr-95	$<1.0 \times 10^{-9}$
Xe-131m	5.0×10^{-2}	Nb-95	$<1.0 \times 10^{-9}$
Xe-133m	2.6×10^{-3}	Te-127m	5.0×10^{-2}
Xe-133	5.8×10^{-3}	Te-127	1.6×10^{-4}
Xe-135m	1.5×10^{-6}	Te-129m	5.0×10^{-2}
Xe-135	3.3×10^{-4}	Te-129	1.1×10^{-5}
Xe-137	2.8×10^{-7}	Te-131m	3.7×10^{-5}
Xe-138	1.7×10^{-6}	Te-132	2.6×10^{-3}
		Cs-137	5.0×10^{-2}
I-131	8.6×10^{-3}	Ba-140	3.9×10^{-8}
I-132	2.7×10^{-3}	La-140	3.9×10^{-8}
I-133	4.3×10^{-4}		
I-134	1.7×10^{-5}		
I-135	1.1×10^{-4}		

*R/B defined as rate of release/rate of buildup.

Ref. CF-60-10-63, pages 19.7-19.9.

Table IV. Calculated Atmospheric Radiation Dose Rate Produced by Continuous Release of Fission Products from One CP-ORR Capsule*

	Dose (mr/hr)					% of
	Krypton	Xenon	Iodine	Solids	Total	Permissible[c]
Case A						
Total Body	1.8×10^{-4}	1.2×10^{-4}	7.6×10^{-4}	3.5×10^{-4}	1.4×10^{-3}	5.6×10^{-1}
GIT (insol)[a]	1.8×10^{-4}	1.2×10^{-4}	3.3×10^{-2}	9.3×10^{-3}	4.3×10^{-2}	5.7
GIT (sol)[b]	1.8×10^{-4}	1.2×10^{-4}	8.3×10^{-3}	4.6×10^{-3}	1.3×10^{-2}	1.7
Bone	1.8×10^{-4}	1.2×10^{-4}	-	4.1×10^{-4}	7.1×10^{-4}	9.5×10^{-2}
Kidney	1.8×10^{-4}	1.2×10^{-4}	-	3.6×10^{-3}	3.9×10^{-3}	5.2×10^{-1}
Lung	1.8×10^{-4}	1.2×10^{-4}	6.5×10^{-3}	8.1×10^{-3}	1.5×10^{-2}	2.0
Thyroid	1.8×10^{-4}	1.2×10^{-4}	4.4×10^{-1}	4.5×10^{-4}	4.4×10^{-1}	29.3
Case B**						
Total Body	1.1×10^{-4}	5.9×10^{-5}	8.2×10^{-8}	3.5×10^{-7}	1.1×10^{-3}**	4.4×10^{-1}
GIT (insol)	1.1×10^{-4}	5.9×10^{-5}	3.8×10^{-6}	9.4×10^{-6}	1.1×10^{-3}	1.5×10^{-1}
GIT (sol)	1.1×10^{-4}	5.9×10^{-5}	9.7×10^{-7}	4.6×10^{-6}	1.1×10^{-3}	1.5×10^{-1}
Bone	1.1×10^{-4}	5.9×10^{-5}	-	4.1×10^{-7}	1.1×10^{-3}	1.5×10^{-1}
Kidney	1.1×10^{-4}	5.9×10^{-5}	-	3.6×10^{-6}	1.1×10^{-3}	1.5×10^{-1}
Lung	1.1×10^{-4}	5.9×10^{-5}	7.1×10^{-7}	8.1×10^{-6}	1.1×10^{-3}	1.5×10^{-1}
Thyroid	1.1×10^{-4}	5.9×10^{-5}	4.8×10^{-5}	4.5×10^{-7}	1.1×10^{-3}	7.3×10^{-2}
Case C						
Total Body	1.5×10^{-5}	3.8×10^{-5}	7.6×10^{-8}	3.5×10^{-7}	5.3×10^{-5}	2.1×10^{-2}
GIT (insol)	1.5×10^{-5}	3.8×10^{-5}	3.3×10^{-6}	9.3×10^{-6}	6.6×10^{-5}	9×10^{-3}
GIT (sol)	1.5×10^{-5}	3.8×10^{-5}	8.3×10^{-7}	4.6×10^{-6}	5.8×10^{-5}	8×10^{-3}
Bone	1.5×10^{-5}	3.8×10^{-5}	-	4.1×10^{-7}	5.3×10^{-5}	7×10^{-3}
Kidney	1.5×10^{-5}	3.8×10^{-5}	-	3.6×10^{-6}	5.7×10^{-5}	7×10^{-3}
Lung	1.5×10^{-5}	3.8×10^{-5}	6.5×10^{-7}	8.1×10^{-6}	6.2×10^{-5}	8×10^{-3}
Thyroid	1.5×10^{-5}	3.8×10^{-5}	4.4×10^{-5}	4.5×10^{-7}	9.7×10^{-5}	6×10^{-3}

a Assuming fission products are insoluble.
b Assuming fission products are soluble.
c Permissible ORNL atmospheric radiation dose per 40 hr week.
* Containing 6.5 gms U235.
** Includes dose produced by A^{41} (8.8×10^{-4} mr/hr).

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