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UNITED STATES ATOMIC ENERGY COMMISSION

**A HOMOGENEOUS-REACTOR GAMMA
IRRADIATION FACILITY**

By
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July 27, 1956

Oak Ridge National Laboratory
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Technical Information Service Extension, Oak Ridge, Tenn.

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By

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July 27, 1956

Work performed under Contract No. W-7405-Eng-26

OAK RIDGE NATIONAL LABORATORY

Operated by

Union Carbide Nuclear Company

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Oak Ridge, Tennessee

SUMMARY

A brief study was made of a meat-irradiation facility consisting of an aqueous homogeneous reactor and a source utilizing fission-product gases and their daughter products. It was estimated that a reactor power level of 15 Mw would be required to obtain a source that would irradiate 6 in. x 16 in. x 20 in. boxes of meat to a dose of 2×10^6 rep in 30 min and at a rate of 2800 lb/hr. Approximately 1/4% of the reactor power would be emitted as energy of gamma photons in the source. Based on experience with the Homogeneous Reactor Test, it is believed that the entire facility would cost about \$5.5 million.

INTRODUCTION

The use of gamma radiation from reactor fission products to sterilize medical products and to preserve foods has been widely discussed. Aqueous homogeneous reactors offer a unique opportunity for exploitation of this concept because of two circumstances: first, approximately two-thirds of the fission-product yield appears as xenon and krypton gases during the decay of the primary fission products to stable isotopes, and second, the fluid nature of the homogeneous reactor fuel makes it possible to remove these gases and to pipe them quickly and continuously to a nearby irradiation source. Such a method would involve no fuel-element processing and only a minimum of radiation-wasting delay.

Presented here are the results of a brief study of a homogeneous reactor irradiation facility that was made at the request of the Army Reactor Branch, Division of Reactor Development of the Atomic Energy Commission in Washington, D. C.⁽¹⁾ Although it was requested that utilization of radiation from the circulating fuel solution and from an indium sulfate solution irradiated in a blanket around the reactor core be considered also, the study was limited to the case of the gaseous fission products. Their use presents fewer obvious problems at the present time.

Radiation requirements that were specified by the Army Reactor Branch are contained in Appendix A. A source was required that would provide for irradiation of several types of packages to several different dosages. Consideration here is limited to a source that would irradiate 6 in. x 16 in. x 20 in. boxes of meat to a dose of 2×10^6 rep at a continual rate of 1000 tons per month or 2800 lb per hr. It is assumed that the facility can be designed to meet the other specifications presented in Appendix A.

FISSION-PRODUCT ISOTOPES OF INTEREST

Although two-thirds of the total fission-product yield appears as xenon or krypton during the decay of primary fission products to stable isotopes, in most instances the nuclei have half lives which are very short or very long, so that only a few of the gaseous fission products are useful in an irradiation facility.

Gaseous isotopes and radioactive daughter products which can be utilized are listed in Table 1. The important isotopes appear to be Kr^{87} , Kr^{88} and daughter Rb^{88} , the Rb^{89} daughter of Kr^{89} , Xe^{135} , and the Cs^{138} daughter of Xe^{138} .

TABLE 1
RADIATIONS FROM SOME OF THE RADIOACTIVE GASEOUS
FISSION PRODUCTS AND THEIR DAUGHTERS

Nuclide	Mass No.	Half Life ⁽¹⁾	Decay Const. ⁽¹⁾ (sec ⁻¹)	Yield ⁽¹⁾ (%)	Beta Particles			Gamma Transitions			Fission-Product Generation Rate (atoms/sec-MW)	Potential Gamma- Energy Release Rate (Mev/sec-MW)
					E _β (Mev)	Yield (%)	Av E _β (Mev)	E _γ (Mev)	Yield (%)	Total E _γ (Mev)		
³⁶ Kr	83m	114m	1.01 × 10 ⁻⁴	0.458	0 ⁽¹⁾	0 ⁽¹⁾	0 ⁽¹⁾	0.032 ⁽¹⁾	100 ⁽¹⁾	0.04	1.5 × 10 ¹⁴	0.06 × 10 ¹⁴
	85m	4.36h	4.41 × 10 ⁻⁵	0.848	0.824 ⁽²⁾	75 ⁽²⁾	0.2	0.305 ⁽²⁾	25 ⁽²⁾	0.19	2.7 × 10 ¹⁴	0.51 × 10 ¹⁴
	87	78m	1.46 × 10 ⁻⁴	2.7	1.27 ⁽²⁾	25 ⁽²⁾	1.0	2.57 ⁽²⁾	20 ⁽²⁾	1.1	8.6 × 10 ¹⁴	9.4 × 10 ¹⁴
					3.3	≤ 10		2.05	5			
					3.8	65		0.847	17			
	88	2.77h	6.95 × 10 ⁻⁵	3.78	0.52 ⁽²⁾	70 ⁽²⁾	0.3	0.403	82			
					0.9	~ 10		2.40 ⁽²⁾	35 ⁽²⁾	1.7		
					2.7	20		2.19	≤ 18			
								1.85(?)	≤ 15			
								1.55	14			
³⁷ Rb	88	17.8m	6.49 × 10 ⁻⁴		5.30 ⁽³⁾	76 ⁽³⁾	1.5	1.20 ⁽⁷⁾	4			
					3.45	4		0.85	29			
					2.54	14		0.355 ⁽⁷⁾	5			
					2.06	2		0.191	35			
					1.78	0.2		0.163	7			
					1.62	0.08		0.028	7			
					0.77	2		4.87 ⁽³⁾	0.3 ⁽³⁾	0.70	12 × 10 ¹⁴	29 × 10 ¹⁴
					0.43	2		3.68	0.1			
								3.52	0.3			
								3.24	0.3			
³⁶ Kr ³⁷ Rb	89	3.18m	3.63 × 10 ⁻³	4.6	4 ⁽¹⁾	100 ⁽¹⁾	1.3 ⁽¹⁾	3.01	0.3			
	89	15.4m	7.50 × 10 ⁻⁴		3.92 ⁽⁴⁾	7	1.5	2.68	2.5			
					2.87	5		2.11	1			
					1.61	53		1.85	23			
					1.33	2		1.39	1.4			
					1.17	3		0.91	14			
					0.67	28		0 ⁽¹⁾	0 ⁽¹⁾	0 ⁽¹⁾		
					0.40	2		3.62 ⁽⁴⁾	2 ⁽⁴⁾	2.4		
								2.75	3			
								2.59	12			
								2.20	14			
								1.35	4			
								1.26	63			
Sub Total											15 × 10 ¹⁴	36 × 10 ¹⁴
												75 × 10 ¹⁴

Nuclide	Mass No.	Half Life ⁽¹⁾	Decay Const. ⁽¹⁾ (sec ⁻¹)	Yield ⁽¹⁾ (%)	Beta Particles			Gamma Transitions			Fission-Product Generation Rate (atoms/sec.MW)	Potential Gamma- Energy Release Rate (Mev/sec.MW)
					E _β (Mev)	Yield (%)	Av E _β (Mev)	E _γ (Mev)	Yield (%)	Total E _γ (Mev)		
⁵⁴ Xe	133m	2.3d	3.49 × 10 ⁻⁶	0.16	0 ⁽¹⁾	0 ⁽¹⁾	0 ⁽¹⁾	0.23 ⁽¹⁾	100 ⁽¹⁾	0.23	0.51 × 10 ¹⁴	0.01 × 10 ¹⁴
	133	5.27d	1.52 × 10 ⁻⁶	6.62	0.345 ⁽¹⁾	100	0.115 ⁽¹⁾	0.081 ⁽¹⁾	100 ⁽¹⁾	0.081	21 × 10 ¹⁴	1.7 × 10 ¹⁴
	135m	18.6m	7.40 × 10 ⁻⁴	1.7	0 ⁽¹⁾	0 ⁽¹⁾	0 ⁽¹⁾	0.82 ⁽¹⁾	100 ⁽¹⁾	0.82	5.4 × 10 ¹⁴	2.8 × 10 ¹⁴
	135	9.13h	2.11 × 10 ⁻⁵	6.58	0.91 ⁽²⁾	97 ⁽²⁾	0.3	0.804 ⁽²⁾	9 ⁽²⁾	0.26	21 × 10 ¹⁴	5.5 × 10 ¹⁴
					0.55	3		0.360	0.1			
								0.250	97			
	137	3.9m	2.96 × 10 ⁻³	6.19	4.0 ⁽¹⁾	100	1.33	0 ⁽¹⁾	0	0		
	138	17m	6.79 × 10 ⁻⁴	6.12	3.0 ⁽¹⁾	100	1.0	2.01 ⁽²⁾	(?)	0 ⁽¹⁾		
								1.78	(?)			
								0.51	(?)			
⁵⁵ Cs	138	32m	3.62 × 10 ⁻⁴		3.4 ⁽²⁾	50 ⁽²⁾	0.8	3.34 ⁽⁵⁾	1 ⁽⁵⁾	3.0	20 × 10 ¹⁴	60 × 10 ¹⁴
					2.4	25		2.64	11			
					0.7	25		2.20	21			
								1.44	100			
								1.02	33			
								0.88	11			
								0.55	17			
								0.46	37			
										Sub Total		70 × 10 ¹⁴

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Whether Kr^{89} , Xe^{137} , and Xe^{138} are potentially important contributors of gamma radiation is not clear. Essentially no data concerning the photons emitted during the decay of those isotopes were found. Their possible contributions are neglected in this study.

It is concluded that the total amount of gamma energy available from the decay of those gases which have favorable half lives and from their daughters is about 0.5 Mev per fission or 1/4% of the total reactor power. As much as 70% of the energy appears to be associated with the decay of the rubidium and cesium daughters.

REMOVAL OF FISSION-PRODUCT GASES FROM THE REACTOR SYSTEM

Maximum utilization of the fission-product gases is obtained if they are stripped rapidly from the reactor fuel and delivered almost immediately to the irradiation source. With aqueous homogeneous reactors this can be accomplished by operating the reactor under conditions where hydrogen and oxygen gases are produced by decomposition of some of the water in the reactor. These radiolytic gases, saturated with water vapor and containing xenon and krypton, which distribute themselves between the liquid and vapor phases, are throttled with some entrained fuel solution from a high-temperature, high-pressure circulating system to a low-pressure system. Gases and liquid are separated as they enter the low-pressure system; the gases are diluted with steam so that the hydrogen concentration is below the explosive limit and are passed through a bed of platinum catalyst, where the hydrogen and oxygen are recombined. The vapor then is superheated steam containing traces of fission-product gases and oxygen, which is added continuously to the reactor to replace the oxygen consumed in corrosion reactions. After these vapors leave the recombiner, they are desuperheated and most of the steam is condensed. The oxygen and the remainder of the steam are used to carry the fission products to the irradiation source.

The fraction of the fission-product gases that can be extracted from the fuel stream in this manner can be calculated from the equation

$$F = \frac{(fv/V)H}{\lambda + \sigma\phi + (fv/V)H}$$

where F = fraction of a particular fission-product isotope in the letdown stream,

f = void fraction or ratio of gas volume to total volume of reactor fuel stream at gas separator,

v = fuel circulation rate, liters/sec,

V = total volume of fuel in reactor system, liters,

H = Henry's law constant, $\frac{\text{atoms of Xe or Kr per cm}^3 \text{ of gas}}{\text{atoms of Xe or Kr per cm}^3 \text{ of liquid}}$,

λ = radioactive decay constant, sec^{-1} ,

σ = absorption cross section, cm^2/atom ,

ϕ = thermal-neutron flux, $\text{neutrons}/\text{cm}^2 \cdot \text{sec}$.

In typical aqueous homogeneous reactor design studies and in the Homogeneous Reactor Experiment⁽²⁾ and the Homogeneous Reactor Test,⁽³⁾ the fuel volumes and circulation rates respectively have been 0.05 to 0.1 liters/kw and about 0.06 liters/(sec)(kw), so $0.6 \leq v/v \leq 1.2$. The Henry's law constant for xenon has been determined to be 1 to 2×10^5 psi/mole fraction or 4 to 8 $\frac{\text{atoms of Xe per cm}^3 \text{ of gas}}{\text{atoms of Xe per cm}^3 \text{ of liquid}}$ at 250 to 300°C.⁽⁴⁾ The constant for krypton should be equal to or greater than that for xenon. Although reactor safety and control considerations dictate that f should be kept small, values as large as 0.02 are certainly permissible. A representative value for $(fv/V)H$ would be 0.05.

The term $\sigma\phi$ can be neglected since it has a value of only 0.003 for Xe^{135} at 10^{15} neutrons/cm².sec. Then

$$F \approx \frac{0.05}{\lambda + 0.05}$$

and is greater than 0.85 for isotopes of half life greater than 1 min. It can be assumed that at least 95% of the yield of the gases listed in Table 1 can be separated from the circulating fuel stream.

RELATIONSHIP BETWEEN TRANSIT TIME AND ENERGY AVAILABLE AT SOURCE

The effect of transit time between reactor and source on the energy available from the decay of the fission-product gases and their daughters is shown in Fig. 1. If the reactor and irradiation facility are operated under steady-state conditions, gases are delivered to the source continuously with a transit time A and pass through the source in time B ; the energy emitted in the source is the difference in the potential energy values at times A and $(A + B)$. This is true if the solid daughter products are retained and decay where they are formed. There is little incentive to hold the gases in the source for more than 7 hr. The relationship between fraction of the radiation emitted in the source and delay time is presented in Table 2.

TABLE 2. EFFECT OF TRANSIT TIME ON ENERGY AVAILABLE IN SOURCE

Transit Time (min)	Energy Release Rate in Source* (Mev/sec.Mw)	Fraction of Total Energy* Emitted in Source
1	12×10^{15}	0.86
2	11×10^{15}	0.79
3	10×10^{15}	0.72
5	9×10^{15}	0.65
10	7.3×10^{15}	0.53
30	4.6×10^{15}	0.33

* Assumes that gas is retained in source for 7 hr.

Fig. 1. Effect of Decay Time on Gamma Energy Available from the Decay of the Fission Product Gases and Their Daughters

REQUIRED REACTOR POWER LEVEL

Specification of the reactor power level necessary to obtain the source power for a fixed production rate and irradiation time requires at least a preliminary design of the source and conveyor arrangement. A simple slab was selected as the most useful, although not necessarily the most efficient, source geometry. Figure 2 is a diagram showing dimensions of the slab and the path that the cartons would take. Dimension "b" was fixed at 28 in. to provide sufficient overlap for a variation of 10%⁽⁵⁾ or less in the energy flux at the surface of the packages. Dimension "a" was determined by the irradiation time and the production rate of 2800 lb/hr. Although the package arrangement was selected so that the radiation would pass through a minimum thickness, the dosage at the center would be only about 80% of that at the surface. Thinner packages would be required to obtain greater uniformity.

Source lengths and reactor power levels were calculated for irradiation times of 10, 30, and 60 min, a thin slab with no self-absorption, a space of 6 in. between the slab and the packages, and an energy release rate of 12×10^{15} Mev/sec.Mw in the source. Results of the calculations are presented in Table 3.

TABLE 3. EFFECT OF IRRADIATION TIME AND SOURCE LENGTH
ON REQUIRED REACTOR POWER

Irradiation Time* (min)	Source Length (in.)	Source Power (Mev/cm ² .sec)	Reactor Power (Mw)
10	67	5.0×10^{12}	10
30	200	1.3×10^{12}	7.9
60	400	0.62×10^{12}	7.4

* Total dose = 2×10^6 rep.

The reactor power would have to be greater than the values presented in Table 3 to compensate for edge losses from a slab of finite thickness, for absorption within the slab, and for inefficiencies in delivery of radioactive material to and retention in the source. Some increases in efficiency could be realized by a careful study of source and conveyor arrangements. A firm value for the power level requires a detailed design of the reactor and irradiation facility. At the present, there are only suggestions for parts of the design. For purposes of this study 15 Mw and 30 min, respectively, were selected as probable values for the reactor power level and irradiation time. The gamma energy absorbed in the meat would be 18% of the total available from the decay of the radioactive gases and their daughters and 0.045% of the reactor power.

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Fig. 2. Source and Carton Geometry

THE REACTOR SYSTEM

The flow diagram of a 15-Mw aqueous homogeneous reactor for the irradiation facility is shown in Fig. 3. The reactor consists of a high-temperature, high-pressure circulating system in which the nuclear reaction is sustained and a low-pressure system in which fuels are stored, radiolytic gases are recombined, and the fission-product gases are finally separated from the reactor system for delivery to the source. Operation of the reactor at high temperature is required to prevent the uranium from precipitating as uranyl peroxide.

In the high-pressure circulating system an aqueous solution of uranyl sulfate is circulated through a 4-ft-dia reactor vessel at a rate of 1500 gpm. It enters at 440°F and leaves at 525°F. The pressure in the system is maintained at 1250 psia. Gas generation in the reactor is controlled, by the use of copper sulfate catalyst in the fuel, to a production rate of 4 cfm containing 30 vol % of hydrogen and oxygen, 70 vol % of steam, and traces of xenon and krypton. The amount of hydrogen and oxygen formed is equivalent to operation at a power level of 5 Mw in the absence of a catalyst.

Fuel leaving the reactor passes through an axial-flow gas separator where the gas and 2 to 3 gpm of fuel are withdrawn from the circulating stream. Then the recirculating fuel is cooled in heat exchangers and returned to the reactor by canned-motor pumps. Steam is produced in the heat exchangers at 420°F and 320 psia and can be supplied to turbines to generate about 3000 kw of electricity.

Connected to the high-pressure piping is a surge chamber to contain liquid expelled from the reactor in the event of a rapid increase in reactivity and reactor power. The circulating system is pressurized through the surge chamber by steam produced in an attached boiler which evaporates condensate obtained from the low-pressure system.

The mixture of gas and liquid from the gas separator passes through a throttling valve and into the fuel storage tanks of the low-pressure system. There the hydrogen and oxygen are diluted with steam to a concentration below the explosive limit, entrained liquid is removed in a separator, and the hydrogen and oxygen are recombined by passing the mixture over a platinum catalyst. The superheated steam leaving the recombiner at 1000°F flows into a condensing and degassifying column where the volume is reduced to about 5 cfm of steam and noncondensables which are vented to the irradiation source. The pressure in the low-pressure system is held at about 30 psia by control of the condenser and the vent valves.

Condensate from the condenser is returned partially to the dump tanks and partially to feed pumps which are used to purge the rotor cavity of the circulating pumps and to feed the pressurizer boiler. A storage tank is provided for holding condensate when the reactor fuel concentration is changed to adjust the operating temperature.

Feed pumps are provided to return fuel solution to the reactor to maintain the inventory in the high-pressure system.

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Fig. 3. Reactor System Flow Diagram

Either an H_2O or a D_2O solution of uranyl sulfate and copper sulfate would be used in the reactor. The critical concentrations at the operating temperature would be 17 g and 3.7 g of U^{235} per liter, respectively, in H_2O and in D_2O . There would be 0.02 to 0.03 m copper sulfate and some excess sulfuric acid added to either solution. The volume of the high-pressure system would be about 2500 liters, and there would be an additional 100 to 500 liters in the low-pressure system. Type 347 stainless steel would be used for construction of the entire reactor system. In determining whether to use H_2O or D_2O , the disadvantages of a 50-kg inventory of U^{235} and the increase in corrosion problems associated with the higher uranyl sulfate concentration required with H_2O would have to be balanced against the inconvenience of handling and conserving D_2O and the problems of neutron production by the (γ, n) reaction with deuterium in the source.

THE SOURCE

The delay between production of gas in the reactor and departure from the condenser in the low-pressure system is estimated to be about 45 sec. Delivery to a source building, as much as 400 ft away, could be accomplished in 15 sec or less. Concentrating the activity in the source and retaining it there present the major design problems.

Adsorbing the radioactive nuclei on activated carbon is a doubtful solution to the problem because of difficulties in controlling the distribution of activity and in cooling the carbon. A gaseous source appears more attractive. The flow diagram for such a source is presented in Fig. 4.

The source for a 30-min irradiation time is 28 in. high, 200 in. long, and 1-1/2 to 3 in. thick and has a volume of 5 to 10 ft³. It consists of a number of tubes of rectangular or circular cross section through which the gas is circulated continuously, at essentially atmospheric pressure and at a temperature as high as 200°F.

Approximately 30 kw of heat is produced in the source by the collision of beta particles with the walls. This heat is removed most conveniently by circulating water over the outer surfaces of the source either in an integral jacket or as a falling film. The source is separated and possibly insulated from the rest of the irradiation cell to minimize the transfer of heat to frozen foods. Condensation within the source is prevented so that solids will not be washed from the walls.

Fresh fission-product gases are introduced into a part of the source gas which is pumped by a blower through an external piping and heat exchanger circuit. The steam carrier for the fresh gas is condensed and returned to the reactor, and the fission-product gases are circulated to the source. The volume of the external system is 1 ft³ or less.

An amount of gas equal to the fresh gas is bled from the source and passed through cold traps to remove traces of moisture. Then the gases are sent to carbon beds where the krypton and xenon are retained until most of the radioactive isotopes decay before the stable gases are discharged to the atmosphere.

Fig. 4. Flow Diagram for Source

FEASIBILITY AND COST

The reactor for the irradiation facility resembles the Homogeneous Reactor Test. It has about the same total volume and requires equipment of similar size. The HRT is more complicated in that it is a two-region reactor with separate fuel and reflector systems. Although the HRT is designed for operation at a power level of 5 Mw, the circulating and heat-removal equipment are capable of removing 10 Mw. The low-pressure evaporators and recombiner equipment can handle radiolytic gas from a reactor operating without an internal catalyst at a power level of at least 3 Mw. Satisfactory operation of the HRT should provide a convincing demonstration of the feasibility of the 15-Mw reactor.

A detailed design of the degassifying condenser and the entire irradiation facility is required to establish their feasibility and a firm power level for the reactor. Before constructing an irradiation facility an analysis should be made of radiations from the HRT off-gases after short delay to confirm the calculated relationship between reactor power and source power. A method for retaining the Rb and Cs in the source must be demonstrated.

Duplicating the HRT, except for power-generating or steam-condensing equipment for disposing of the heat, is estimated to cost \$3.5 million. Design, additional reactor equipment, and power-generating equipment for the 15-Mw reactor should cost about \$1.5 million. The cost of a minimum source and conveyor system has been suggested to be \$100,00 to \$175,000.⁽⁵⁾⁽⁶⁾ A reasonable total cost for the entire homogeneous-reactor irradiation facility is believed to be about \$5.5 million.

APPENDIX A

Radiation Requirements*

1. It is assumed that items to be exposed in the irradiation facility will require doses from 10^4 to 10^7 rep. Doses required for foods will be under 6×10^6 rep, with most items being given 2×10^6 rep or less.
2. Maximum dose rate is to exceed 1×10^6 rep/hr in order to minimize the size of the irradiation volume and the time of exposure.
3. The source should be capable of delivering a dose of 2 megarep to 1000 tons of food or other substances of unit density per month assuming operation on a continual basis. However, it is assumed that there will be frequent shutdowns to permit modifications in food processing lines, thus reducing the actual output of the radiation source.
4. The radiation volume should be capable of handling continuously:
 - a. items of unit density of sizes from No. 2 cans (3-7/16 in. in diameter by 4-9/16 in. high) up to V3 boxes (6 in. x 16 in. x 20 in.).
 - b. sacks of flour, potatoes, etc.

An overhead trolley should be provided to permit treatment of carcasses, military equipment, etc.

5. Temperature of radiation volume should be capable of being controlled within 5°C over a range of -20 to $+75^\circ\text{C}$.
6. Dosage delivered is to be within 15% of the required amount. This is an outside figure, with a maximum variation in dose being within 10% if this can be obtained.
7. Investigation at the MTR has shown that foods irradiated in the canal have been exposed to neutron fluxes of about 10^4 neutrons/cm²·sec for periods of as long as 1/2 hr with the production of no detectable radioactivity in either food or container. (Since each fuel element in the irradiation facility was placed in a cadmium-lined slot, it can be assumed that most of this flux was due to the (γ , n) reaction of the deuterium in the canal water with gamma photons of over 2.23 Mev energy.) Based on these observations, it would appear that a total maximum dose of 1.8×10^7 neutrons/cm² is acceptable. This figure corresponds closely with the maximum allowable personnel dose of neutrons as determined from data listed in NBS Handbook 52.

* Provided by Army Reactor Branch (AFB), Division of Reactor Development, Washington, as part of reference 4.

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