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F. T. Binford
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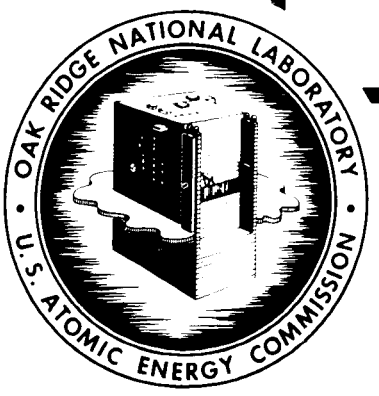

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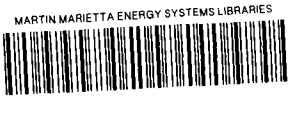

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A METHOD FOR THE DISPOSAL OF VOLATILE FISSION PRODUCTS FROM AN ACCIDENT IN THE OAK RIDGE RESEARCH REACTOR

F. T. Binford T. H. J. Burnett

ABSTRACT

The Oak Ridge Research Reactor is a fully enriched, heterogeneous, light-water-moderated and -cooled, beryllium-reflected reactor of the MTR-BSF type. It is designed to operate at a power level of 20 to 30 megawatts with a thermal-neutron flux of the order of 10^{14}.

In this paper the maximum credible accident is postulated to be that situation in which the reactor suffers from a deficiency in cooling capacity sufficient to cause melting of the fuel elements and subsequent release into the building of all the volatile fission products.

It is shown that with properly engineered ventilating and gas-scrubbing equipment the radioactive gas can be disposed of in such a way as to prevent harmful exposure to persons in the surrounding area.

Direct radiation from the large mass of gas initially present in the building will be extremely intense in the immediate vicinity of the building. It is possible, however, through the use of a suitable alarm system, to evacuate this area in a time short enough to prevent serious overexposure to personnel.

1. INTRODUCTION

One of the more important considerations which must be faced when choosing a site for a nuclear reactor is the problem of containment of dangerous substances which may be released as the result of an accident to, or a malfunction of, the machine.

The type of containment required will vary, depending, among other things, upon the nature and population density of the surrounding area, the kind of reactor under consideration, the proposed operating power level, and the maximum credible accident postulated.

In the following analysis the containment of fission gases resulting from the maximum credible accident to the Oak Ridge Research Reactor (ORR) is discussed. This reactor is a fully enriched, heterogeneous, light-water-moderated and -cooled, beryllium-reflected reactor of the MTR-BSF type, as illustrated in Fig. 1. It is designed to operate at a power level of 20 to 30 megawatts (Mw) with a thermal-neutron flux of the order of 10^{14} neutrons/cm^2·sec.

2. THE MAXIMUM CREDIBLE ACCIDENT

The maximum credible accident is taken to be the release of all volatile fission products contained in an ORR core which has been operated continuously for 39 days at a power level of 20 Mw. These gases are assumed to mix uniformly with the air in the building which houses the reactor, and it is further assumed that this structure, illustrated in Fig. 2, remains intact during and after the accident. The total volume of the building and hence of the mixed gases is 8×10^5 ft^3.

The significant nuclides which will be present, together with their initial activity, concentration, and other pertinent data, are listed in Table 1.

The basis on which the foregoing situation has been chosen to be the maximum credible accident in the case of the ORR has been discussed elsewhere[1] and will not be considered further at this time.

3. DISPOSAL OF THE RADIOACTIVE GAS

Elimination of the air-borne hazard resulting from an occurrence such as that described above is based on the following assertion: *If air is exhausted from a building at a rate such that all movement of air, other than movement through the exhaust system, is into the building, then that building is "gastight."*

Now if it is assumed that such an exhaust system is available on the building which houses the

[1] F. T. Binford, ORNL-1795 (Secret).

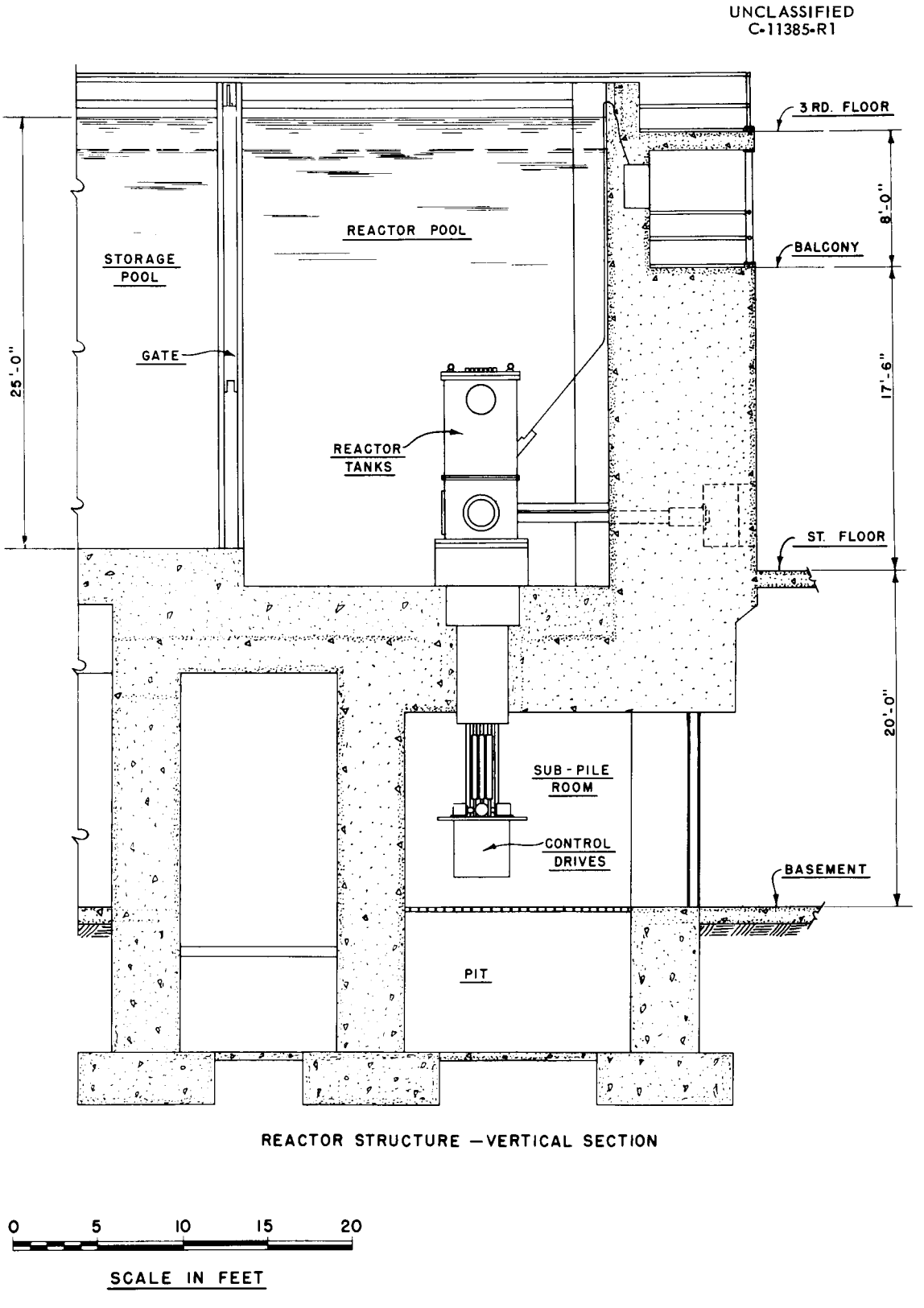

Fig. 1. Reactor Structure – Vertical Section of ORNL Research Reactor Building 3042, Oak Ridge National Laboratory.

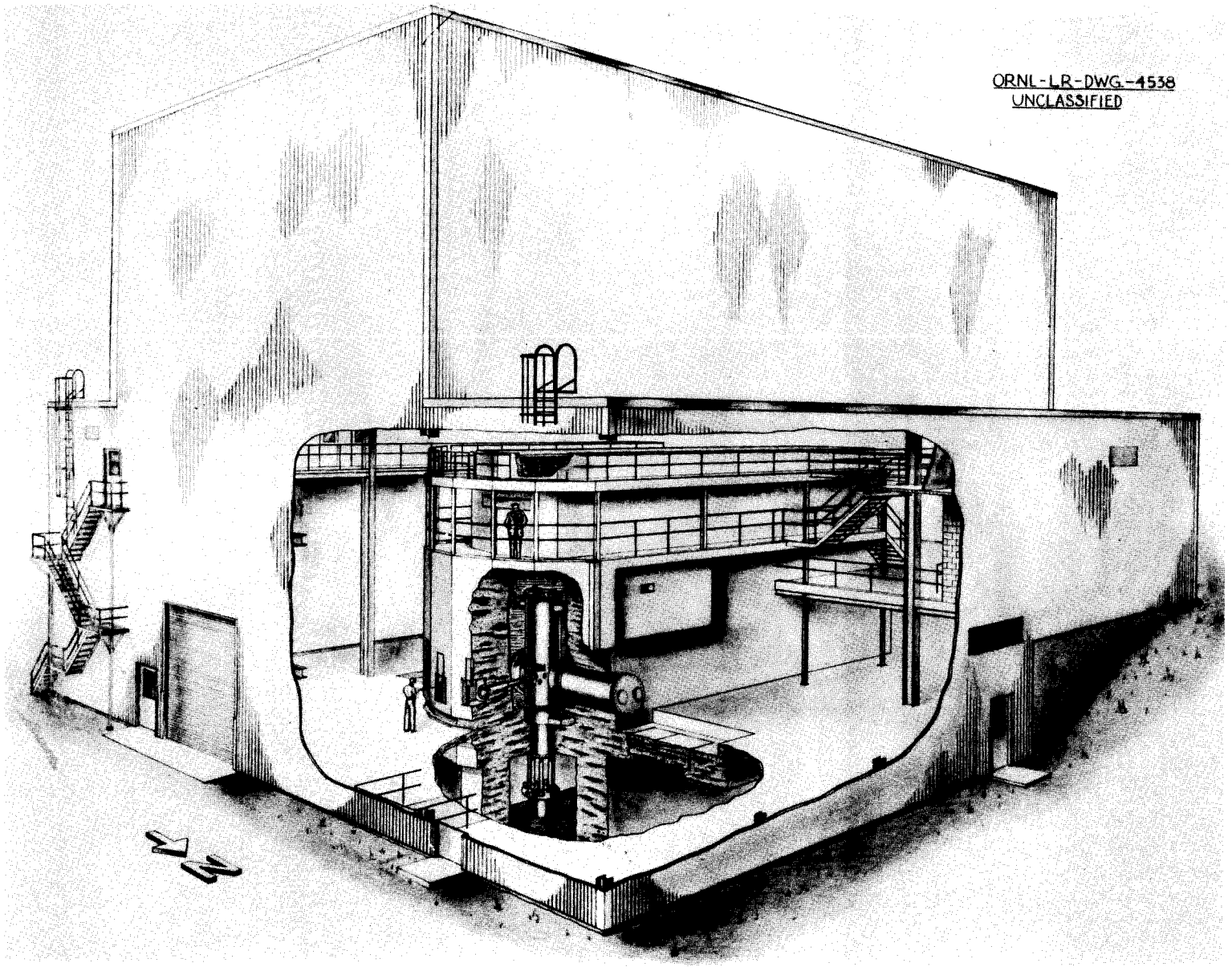

Fig. 2. ORNL Research Reactor, Building 3042.

TABLE 1. INITIAL PROPERTIES OF THE GAS CLOUD

	Nuclide	Decay Constant (hr^{-1})	A_{j_0}, Initial Activity (curies $\times 10^{-5}$)	C_{j_0}, Initial Concentration* (curies/ft^3)	E_j, Effective Energy** (Mev)	$C_{j_0}E_j$	ϵ_j, Effective Gamma Energy (Mev)	$A_{j_0}\epsilon_j \times 10^{-5}$
1	Kr^{85}	0.159	2.724	0.340	0.412	0.140	0.18	0.49
2	Kr^{87}	0.533	4.903	0.613	2.386	1.463	1.088	5.34
3	Kr^{88}, Rb^{88}	0.250	6.719	0.840	4.923	4.136	2.45	16.46
4	Kr^{89}	13.08	8.354	1.044	3.377	3.526	2.0	16.71
5	Xe^{133m}	0.0126	0.291	0.036	0.233	0.0085	0.05	0.01
6	Xe^{133}	0.0055	11.733	1.467	0.190	0.279	0.03	0.35
7	Xe^{135m}	2.67	3.269	0.409	0.520	0.213	0.520	1.70
8	Xe^{135}	0.076	11.259	1.407	0.566	0.797	0.262	2.95
9	I^{131}	0.00355	5.077	0.635	0.583	0.370	0.386	1.96
10	I^{132}	0.289	7.99	0.999	2.706	2.703	2.241	17.90
11	I^{133}	0.033	11.804	1.476	1.027	1.516	0.60	7.08
12	I^{134}	0.792	13.802	1.725	2.298	3.964	1.635	22.50
13	I^{135}	0.104	10.714	1.339	1.456	1.950	1.14	12.20

*Assumed building volume 8×10^5 ft^3.

** Total gamma energy plus one-third maximum beta energy.

ORR, then this building is gastight under the above definition. The radioactive gas so exhausted can be disposed of by scrubbing it and then diluting it with large amounts of air and passing it up one of the stacks available in the ORNL area.

The rate at which gas can be disposed of in this manner will depend upon the hazard resulting from the emission of the radioactive gas from the stack. Control of this hazard will dictate the maximum rcte of exhaust from the building, and this, in turn, will determine the type of building construction required. In effect, the proposal here is to replace random, uncontrolled leakage of concentrated gas from the building at the surface of the ground with controlled leakage of diluted gas at an elevation of 200 to 250 ft.

For the purpose of illustration, the 3039 stack at the Oak Ridge National Laboratory will be considered. This is a 250-ft brick stack through which, under normal operating conditions, air is forced at a rate of 120,000 cfm. The stack is shown in Fig. 3. Purdy and Meyers[2] have calculated that the maximum ground concentration resulting from an emission rate of 1 curie/min from this stack occurs at a wind velocity of 1 mph and is 3×10^{-7} μc per cubic centimeter of air.

In the treatment which follows, it is assumed that a suitable filter is placed in the exhaust system so that all particulate matter will be removed and that a scrubber which will remove 99.90% of the iodine is also incorporated in the system. The cost of such a scrubber is estimated to be $2.00 per cfm of throughput.[3]

3.1 The Internal Hazard

The internal hazard is associated only with the presence of the iodine isotopes and the Sr^{89}, which decays from Kr^{89} via Rb^{89}. A total dose of 25 μc of iodine is considered to be acceptable. This is somewhat less than the usual tracer dose

[2]D. R. Purdy and R. F. Meyers, *Calculations for Unit Emissions of Air-Borne Contaminants*, 1954 (unpublished).

[3]O. T. Zimmerman and I. Lavine, *Chemical Engineering Costs*, Industrial Research Service, Dover, N.H., 1950.

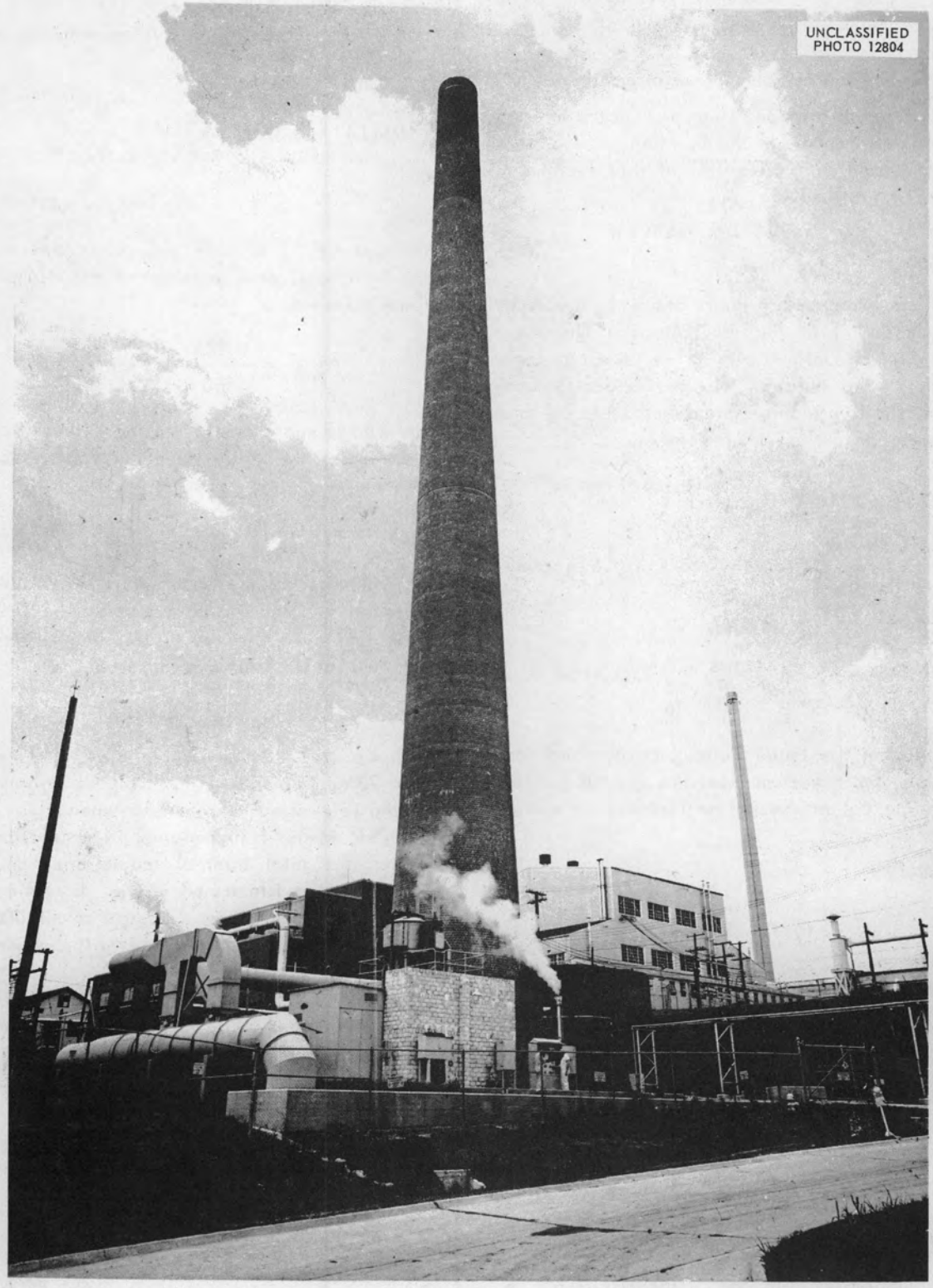

Fig. 3. Stack 3039 and Decontamination Equipment for Exhaust Gases.

of I^{131} given for diagnostic purposes.[4] The maximum permissible concentration of Sr^{89} (for continuous inhalation) is[5] 2×10^{-8} μc/cc.

Let C_{j_0} be the initial concentration (curies/ft^3) of the jth iodine isotope. Then, if P is the exhaust rate (ft^3/min) and V is the building volume (ft^3), the concentration (curies/ft^3) of this isotope in the building at time t is

$$(1) \qquad C_j(t,P) = C_{j_0} e^{-(\lambda_j + 60P/V)t} ,$$

where t is expressed in hours and λ_j is the decay constant (hr^{-1}) of the jth isotope. The basic assumption of uniform mixing is taken to apply throughout the duration of the incident. Considering the five iodine isotopes present, the total concentration (curies/ft^3) becomes

$$(2) \qquad C(t,P) = \sum_{1}^{j=5} C_{j_0} e^{-(\lambda_j + 60P/V)t} .$$

The emission rate from the stack, $E(t,P)$ (curies/min), is just

$$(3) \quad E(t,P) = PC(t,P) = P \sum_{1}^{j=5} C_{j_0} e^{-(\lambda_j + 60P/V)t} .$$

Introducing the factor determined by Purdy and Meyers,[2] the maximum possible ground concentration (μc/cc) of the iodine isotopes in the unscrubbed gas is

$$(4) \quad G(t,P) = 3 \times 10^{-7} P \sum_{1}^{j=5} C_{j_0} e^{-(\lambda_j + 60P/V)t} ,$$

where $G(t,P)$ is expressed in μc/cc.

Integration of Eq. 4 from zero to t and division by t yield the maximum ground concentration (μc/cc) averaged over the time t; this concentration is denoted by $\overline{G(t,P)}$.

$$(5) \quad \overline{G(t,P)} = \frac{3 \times 10^{-7}}{t} P \sum_{1}^{j=5} \frac{C_{j_0}}{\lambda_j + 60P/V} \times \{1 - \exp[-(\lambda_j + 60P/V)t]\} .$$

The inhalation rate for "standard man" is[6] 1.25×10^6 cc/hr. Thus the total maximum internal iodine dose received from breathing this concentration $\overline{G(t,P)}$ during the time t becomes

$$(6) \qquad D(t,P) = 1.25 \times 10^6 \overline{G(t,P)} t ,$$

where $D(t,P)$ is the dose expressed in microcuries. Thus

$$(7) \quad D(t,P) = 0.375P \sum_{1}^{j=5} \frac{C_{j_0}}{\lambda_j + 60P/V} \times \{1 - \exp[-(\lambda_j + 60P/V)t]\} .$$

The upper limit of this maximum dose cannot exceed that delivered when both t and P approach infinity. Therefore

$$(8) \qquad D_{max} \leqq \frac{0.375V}{60} \sum_{1}^{j=5} C_{j_0} ,$$

expressed in microcuries. Taking V to be 8×10^5 ft^3 and using the values given in Table 1 for the C_{j_0}, then

$$(9) \qquad D_{max} \leqq 30.7 \times 10^3 \ \mu c .$$

This value is for the case where no removal of the iodine by scrubbing is accomplished. If 99.9% of the iodine is removed by scrubbing, then the upper limit of this maximum dose becomes

$$(10) \qquad D^s_{max} \leqq 30.7 \ \mu c ,$$

which exceeds the acceptable dose of 25 μc by about 23%. This situation obtains only if the building is emptied nearly instantaneously. This, of course, is not contemplated. The relationship between the total internal iodine dose and the pumping rate is illustrated in Fig. 4. As can be seen, if the exhaust rate is kept below 10,000 cfm the total internal iodine dose will not exceed 25 μc.

The internal hazard associated with Kr^{89} is due to the formation of Sr^{89} by means of the chain

$$Kr^{89} \xrightarrow[2.6 \text{ min}]{\beta^-} Rb^{89} \xrightarrow[15 \text{ min}]{\beta^-} Sr^{89} \xrightarrow[53 \text{ day}]{\beta^-} Y^{89}$$

---

[4] M. Brucer, Oak Ridge Institute of Nuclear Studies, private communication.

[5] Bureau of Standards Handbook 52, *Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water.*

[6] *Recommendations of the International Commission on Radiological Protection (Revised December 1, 1954)*; published as *Supplement No. 6, British Journal of Radiology, Section C, Permissible Dose for Internal Radiation,* Table C III.

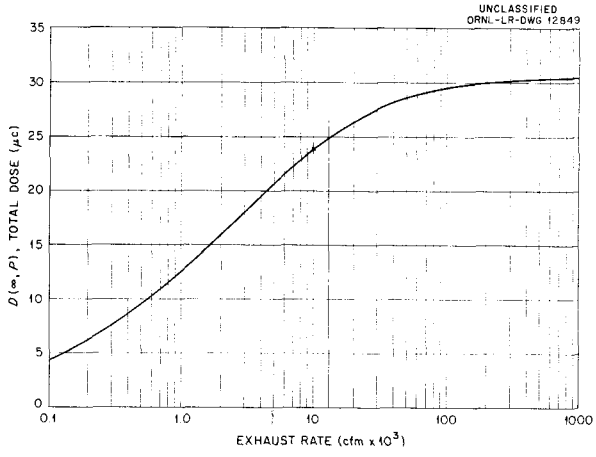

Fig. 4. Maximum Internal Iodine Dose vs Building Exhaust Rate (Scrubbed Gas).

Krypton itself is considered to be unaffected by the scrubbing process, although the filtration and scrubbing might remove some of the Rb^{89} and Sr^{89}. To be conservative, the Kr^{89}–Sr^{89} hazard is calculated as if no removal of members of the chain is accomplished.

The initial concentration of Kr^{89} is 1.044 curies/ft^3. This will decay to

$$1.044 \frac{2.6}{53 \times 24 \times 60} = 3.57 \times 10^{-5} \text{ curies/ft}^3$$

of Sr^{89}. The maximum possible ground concentration (μc/cc) then is

$$G_{max} = 1.07 \times 10^{-11} P \quad . \tag{11}$$

Thus the maximum permissible concentration of 2×10^{-8} μc/cc is never exceeded for exhaust rates less than 1870 cfm. For pumping rates higher than this, the maximum permissible concentration may be exceeded for periods of 2.6 hr or less, as illustrated in Fig. 5. For example, at an exhaust rate of 6000 cfm the initial maximum ground concentration would exceed the maximum permissible level by a factor of 3.2 and would fall exponentially to that level in about 2.58 hr. During this period the average maximum ground concentration would exceed the maximum permissible concentration by a factor of 1.98.

3.2 The External Hazard

The external hazard is considered to arise from all the isotopes listed in Table 1. The iodine isotopes are assumed to be reduced in concentration by a factor of 1000 as a result of the scrubbing procedure. The external dose which should not be exceeded is 25 rep for persons not occupationally exposed to radioactive material and 10 rep for plant personnel.[7]

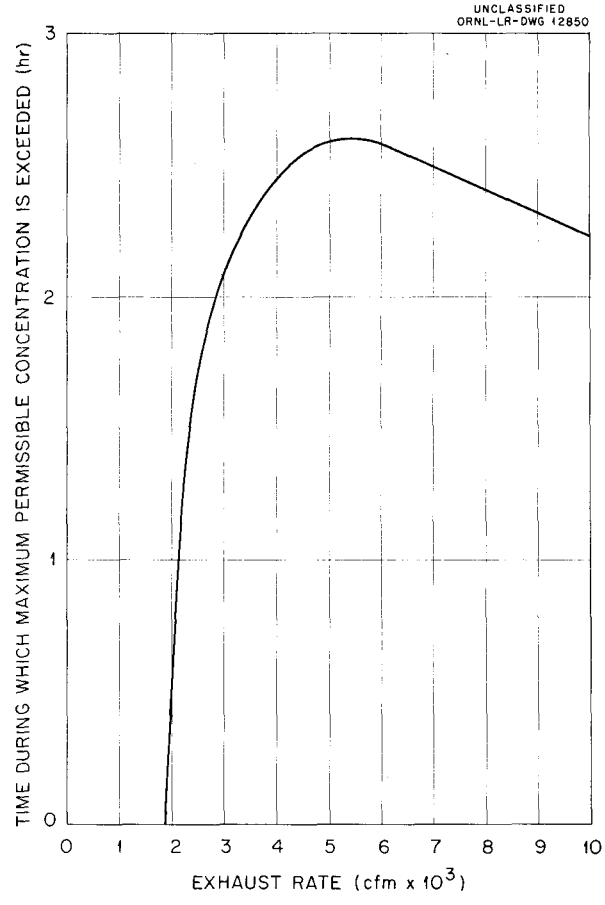

Fig. 5. Time During Which Maximum Sr^{89} Concentration May Exceed 2×10^{-8} μc/cc at Ground Level as a Function of Exhaust Rate.

The external dose is calculated in a manner similar to that used to compute the internal dose. The maximum ground concentration (μc/cc) is

$$G(t,P) = 3 \times 10^{-7} P \sum_{1}^{j=13} C_{j_0} e^{-(\lambda_j + 60P/V)t} \quad , \tag{12}$$

where j_0 refers to all the nuclides listed in Table 1 and where no scrubbing is considered. The external dose from a large cloud of radioactive

[7] *9th Semiannual Report of AEC*, p 19, January 1951.

material may be estimated by using the relation[8]

$$I = 10^3 GE \quad , \tag{13}$$

where I is the dose rate in rep/hr, G is the maximum ground concentration in the cloud in $\mu c/cc$, and E is the effective energy in Mev. Upon introducing the factor f_j by which scrubbing reduces the concentration of each constituent, then for the total maximum dose rate

$$I(t,P) = 3 \times 10^{-4} P \sum_{1}^{j=13} C_{j_0} E_j f_j e^{-(\lambda_j + 60P/V)t} \tag{14}$$

in rep/hr, and the total maximum dose (rep) received during the time t becomes

$$D(t,P) = 3 \times 10^{-4} P \sum_{1}^{j=13} \frac{C_{j_0} E_j f_j}{\lambda_j + 60P/V} \times \{1 - \exp[-(\lambda_j + 60P/V)t]\} \quad . \tag{15}$$

In the case considered, $f_j = 1$ for all constituents except the iodine, and $f_j = 10^{-3}$ for the iodine isotopes. If D_{max} is the upper limit of this maximum dose, clearly

$$D_{max} \leqq 5 \times 10^{-6} V \sum_{1}^{j=13} C_{j_0} E_j f_j \quad . \tag{16}$$

Thus, using $V = 8 \times 10^5$ ft^3 and the values of $C_{j_0} E_j$ from Table 1,

$$D_{max} \leqq 42.3 \text{ rep} \quad . \tag{17}$$

This value is reached only if the building is emptied instantaneously. The dependence of total external dose on exhaust rate is illustrated in Fig. 6. The maximum permissible dose of 25 rep is not exceeded unless the building is exhausted at rates greater than 20,000 cfm.

[8] T. H. J. Burnett, *Sampling Methods and Requirements for Estimating Airborne Radioparticulate Hazards*, ORNL CF-52-11-1.

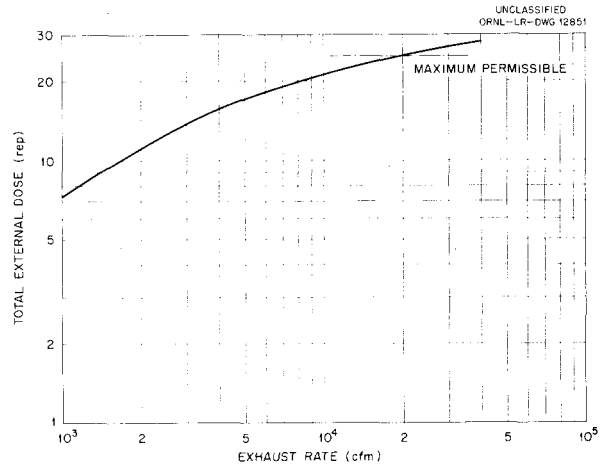

Fig. 6. Maximum External Dose vs Exhaust Rate.

4. DIRECT RADIATION FROM THE BUILDING

Immediately following the release of the gaseous fission products, the building itself becomes a massive radiation source containing nearly 10^7 curies. This source constitutes a severe hazard to those in the immediate vicinity of the building at the time of the accident.

For the purpose of estimating the order of magnitude of this hazard, it is assumed that the gaseous activity leaves the reactor in the form of a bubble before dispersing throughout the building. The radiation intensity from this bubble located immediately above the reactor core is computed by using the effective gamma energies listed in Table 1 and by considering the bubble as a point source.

At a distance of 100 ft from the building, the radiation intensity is initially 47 r/min. This means that personnel in the vicinity of the building at the time of the accident will have 4 to 6 min in which to evacuate the area before receiving the lowest of the range of median lethal dose values. Evacuation can be quite easily accomplished in this time if a suitable alarm system is provided.

Further analysis of the direct-radiation hazard is considered to be beyond the scope of this paper, since this hazard is essentially independent of the type of "gas containment" attempted.