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CRITERIA FOR NOBLE-GAS REMOVAL USING PERMSELECTIVE MEMBRANES

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ABSTRACT

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Criteria have been defined for experiments that are designed to develop a process for removing noble gases from a reactor containment shell under accident conditions. The effect of these criteria on the processing rate of the gas in the containment shell was calculated. When the characteristics of the permselective membranes (silicone rubber) to be used in this program have been determined, the processing rates will be used to establish equipment size, and processing costs. The total cost will then be compared with the cost of the downtime of the reactor.

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CRITERIA FOR NOBLE-GAS REMOVAL USING PERMSELECTIVE MEMBRANES

R. H. Rainey

In the initial part of the program for removing noble gases from a reactor containment shell by a permselective membrane, we used calculations that were based on conditions in the Dresden Reactor to establish criteria since this reactor had been used previously as the basis of an iodine removal study.¹ We chose a processing rate that would produce a decontamination factor of 100 for krypton in one week.² The purpose of the present report is to outline an experiment that will demonstrate the feasibility and the economic advantage of using permselective membranes (silicone rubber) to remove noble gases from the containment shell of an advanced water-cooled reactor, which was described in the letter "Request for Review by Water-Reactors Safety Program Office of Proposals on Filter Tests and Noble Gas Removal," written by Joseph A. Lieberman, Assistant Director of Nuclear Safety, Division of Reactor Development and Technology, to the Manager, Idaho Operations Office. The characteristics of this advanced reactor are as follows:

Size	3200 Mw (thermal) 1000 Mw (electrical)
Flux	3×10^{13} n/cm ² sec
Fuel life	625 days (5.4×10^7 sec)
Fuel load	100 metric tons (10^8 g) of uranium
Fuel enrichment	2.3% ²³⁵ U (10^4 moles, or 6×10^{27} atoms, of ²³⁵ U)
Fuel irradiation	20,000 Mwd/ton
Containment shell volume	3×10^6 ft ³

The concentrations of krypton and xenon, and the total curies and total watts of radioactive krypton and xenon, have been calculated, assuming a total release of noble gas as a result of an accident at the

end of the fuel cycle. The concentrations of krypton and xenon in the containment shell will be about 100 ppm and 650 ppm, respectively; and, since most of the gas will be nonradioactive, these concentrations will not decrease appreciably with time. After a period of one day, the activity from xenon will be about 150 times that from krypton (Fig. 1) and the total activity in the containment shell will be more than 2×10^8 curies (3.5×10^5 w) of activity. However, because of the relatively short half-life of the ^{133}Xe (5 days) and the long half-life of the ^{85}Kr (10 years), the total activity will decrease rapidly until, after about 45 days, the activity of the krypton will become the limiting factor and will remain at about 10^6 curies (10^3 w) for several years thereafter. Thus, if no permselective-membrane gas cleanup system is provided, it will be necessary to hold the gases in the containment shell for about two months to allow the xenon to decay; then the krypton can be vented slowly to the atmosphere. If the discharge rate is restricted to 10^4 curies per day, the removal will require at least 100 additional days and the cost attributable to the downtime of the reactor will be very large. To decrease the downtime, it will be necessary to use a noble-gas concentration system, which consists of a cascade of permselective membranes through which the gases in the containment shell will be pumped; with this system, the concentrated radioactive gases can be stored external to the reactor shell for decay while the reactor resumes its normal function.

Part of the noble-gas processing study will consist of establishing an economic balance between the cost of removing the noble gases from the containment shell and the cost of the downtime of the reactor. To aid this study, an evaluation has been made of the effect of processing rate on the time required to remove the gases.

The activity remaining in the containment shell after any processing time may be calculated from the following equations which were developed by Arnold and Lyness:³

$$X(t) = X_0 e^{-\alpha t} \left[1 - \mu \int_0^t \frac{dt}{X_0 e^{-\alpha t} + Y_0 e^{-\beta t}} \right] \quad 0 \leq t \leq \tau$$

Fig. 1. Decay of Xenon and Krypton in Containment Shell Following a Reactor Accident.

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$$Y(t) = Y_0 e^{-\beta t} \left[1 - \mu \int_0^t \frac{dt}{X_0 e^{-\alpha t} + Y_0 e^{-\beta t}} \right] \quad 0 \leq t \leq \tau,$$

where

X_0 and $X(t)$ are the activities of xenon when the processing starts and at time (t) , respectively,

Y_0 and $Y(t)$ are the activities of krypton when the processing starts and at time (t) , respectively,

α is the xenon decay constant plus the processing removal constant for xenon,

β is the krypton decay constant plus the process removal constant for krypton,

τ is the maximum value of t ,

μ is the rate at which the gases are vented to the atmosphere.

Since these equations could not be integrated analytically, solutions were obtained by a numerical method which used a computer code, called RARGAS, programmed for the CDC 1604 computer.³

Calculations showed that, at a discharge rate of 10^4 curies per day, the time required to reduce the activity in the containment shell to 10^4 curies will increase from 7 days at a processing rate of 1382 ft³/min to 26 days at a rate of 276 ft³/min (Fig. 2). These values are based on the ratio of the permeabilities of krypton and xenon through an unsupported membrane.⁴ In spite of the greater activity of xenon in the containment shell at the time processing starts, the faster decay and the higher removal rate of xenon through the permselective membrane will cause the processing time to be limited by the removal of krypton at all processing rates.

When the permeabilities of nitrogen, oxygen, and the noble gases have been measured under processing conditions, the processing rates will be used to establish equipment size; also, processing costs will be calculated and then compared with the cost of the downtime of the reactor.

Fig. 2. Activity Removed from Reactor Containment Shell vs Processing Rate.

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