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WRITTEN BY R. S. Hart SECTION Sodium Reactors GROUP Engineering UNIT Reactor Technology APPROVED BY: (SUPERVISOR)	**ATOMICS INTERNATIONAL** *A Division of North American Aviation, Inc.* **TECHNICAL DATA RECORD** PROGRAM Advanced Sodium Cooled Reactor Program PROJECT Sodium Cooled Reactor Design and Evaluation	TDR NO. 5817 GO 7519 LEDGER ACCT. 3621 SUB-ACCT. 4410 TWR DATE October 24, 1960 PAGE 1 OF 8

TO: K. W. Foster*

COPIES TO:

R. L. Ashley*
R. J. Beeley*
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S. Siegel

SUBJECT: Radiological Implications Concerning the Use of Argon as the Core Cover Gas in Sodium Graphite Reactors

CONTENTS:

I STATEMENT OF PROBLEM

The substitution of argon for helium as the inert core cover gas in sodium graphite reactors would result in an economic advantage in most areas of the world. The major objection usually raised to such a proposal is related to the induced argon radioactivity that would occur in the cover gas system. The purpose of this report is to; (1) calculate the actual amount of extra radioactivity which would occur, and (2) evaluate the radiological significance of this activity with reference to existing design and operating characteristics of sodium graphite reactors.

II(SUMMARY OF RESULTS AND RECOMMENDATIONS

Using HNPF parameters as a reference model,(4)(5) the levels of argon radioactivity in the cover gas were calculated as:

A^{41} - ($T_{1/2}$ = 110 min.) - 3.2×10^{-2} μc/cc

A^{37} - ($T_{1/2}$ = 34.1 days) - 1.2×10^{-3} μc/cc

These are saturation values at full reactor power (254 Mw). There is also an A^{39} isotope formed ($T_{1/2}$ = 260 years), but assuming a one year turn over rate for the cover gas, the maximum concentration is only 8.2×10^{-8} μc/cc. The radiological significance of these additional activities in the cover gas is evaluated by comparison with expected Xe and Kr levels under normal operating conditions. The results indicate that the Xe^{135} and Kr^{85} alone that could be released from the void space of one defective fuel element would produce rare gas concentrations in the cover gas exceeding those of A^{41} and A^{37}. Since it has been previously estimated that normally up to 1% of the total fuel rods (i.e., ~30 rods in the case of the U-10 Mo loading for HNPF) may be or become defective during reactor operation, the argon activity would not be a radiologically significant addition.

On this basis, there does not appear to be any radiological reason, design, operational or accident-wise why argon could not be substituted for helium as the cover gas in sodium-graphite reactors as presently conceived. Moreover, an increase of the thermal neutron flux in the cover gas region by at least a factor of ten could be tolerated without materially changing the situation. Some confirmation of these conclusions may be inferred from the selection of argon for the inert gas system for both EBR-II and the Fermi plant(1).

III METHOD USED, DESCRIPTION OF EQUIPMENT, SAMPLE CALCULATIONS

A. Radiological Properties of Argon and Calculation of Expected Concentrations

Five radioactive isotopes of argon are presently known.(2) Of these only four can be created through thermal neutron irradiation of naturally occurring argon. One of the four remaining isotopes (A^{42}) requires two successive n, γ reactions and in consideration of the cross-sections and neutron flux levels involved may be neglected here. Table I summarizes the pertinent radiological data on the three isotopes of interest.

TABLE I

Stable Isotope	Natural Abundance %	Activation Cross-Section (barns)	Resulting Radioactive Isotope	Half-Life	Principal Radiation Emitted	MPC(3) air (uc/cc) (*)	MPC(3) air (uc/cc) (**)
A^{36}	0.34	6	A^{37}	34.1 da	γ, 2.6 kev	6×10^{-3}	1×10^{-4}
A^{38}	0.063	0.8	A^{39}	260 yrs.	β, 0.56 mev	1×10^{-5}	3×10^{-7}
A^{40}	99.6	0.53	A^{41}	110 min	β, 1.2 mev γ, 1.3 mev	2×10^{-6}	4×10^{-8}

(*) Maximum permissible concentration in air - 40 hr week, occupational

(**) Maximum permissible concentration in air - 168 hr week, off-site

Although A^{41} is usually the isotope of primary interest, expected concentrations of A^{37} and A^{39} were also calculated because their longer half-lives could possibly make them a consideration in some instances.

Using the well-known expression for activation we have,

$$A_i\,(T,t) = \frac{N_i\,\sigma_i\,\phi\,(1 - e^{-\lambda_i T})\,e^{-\lambda_i t}}{K} \qquad (1)$$

where,

$A_i\,(T,t)$ = concentration of ith isotope of argon after an irradiation of time T and a decay of time t, uc/cc

N_i = number of atoms per cc of the ith isotope of argon,

σ_i = thermal cross section of the ith isotope of argon

ϕ = thermal neutron flux, n/cm^2 - sec

λ_i = decay constant for the ith isotope of argon, time $^{-1}$

T = irradiation time

t = decay time

K = disintegration per second per $\mu c (3.7 \times 10^4)$

Using HNPF parameters as a reference, the average gas temperature at full power assumed to be 500°C (932°F) and the pressure as 720 mm Hg (absolute). Taking the natural abundance of the isotopes as listed in Table 1,

$$N_{36} = \frac{6.02 \times 10^{23}}{22.4 \times 10^3} \times 3.4 \times 10^{-3} \times \frac{273}{773} \times \frac{720}{760} = 3.05 \times 10^{16} \text{ atoms/cc}$$

similarly,

$$N_{38} = 5.66 \times 10^{15} \text{ atoms/cc}$$

$$N_{40} = 8.95 \times 10^{18} \text{ atoms/cc}$$

The thermal neutron flux at the top of the sodium pool at HNPF has recently been calculated as 4×10^8 $n/cm^2/sec$ (at 254 Mw).(4) The temperature adjusted cross sections as listed in Table 1 are: 3.7, 0.5, and 0.33 barns, respectively, for A^{37}, A^{39}, and A^{41}. During continuous reactor operation, saturation equilibrium will normally occur for A^{37} and A^{41}. However, the very long half-life of A^{39} necessitates the choice of some effective irradiation time. One year will be assumed here as the turn-over period for one volume of cover gas. If the actual turn-over period is somewhat longer, the A^{39} concentration will be simple multiples of the calculated concentration (eg., two years, multiply by two, etc.). On these bases, the maximum argon conentrations in the cover gas will be,

$$A^{37} = \frac{3.05 \times 10^{16} \times 3.7 \times 10^{-24} \times 4 \times 10^8}{3.7 \times 10^4} = 1.2 \times 10^{-3} \mu c/cc$$

$$A^{39} = \frac{5.66 \times 10^{15} \times 0.5 \times 10^{-24} \times 4 \times 10^8}{3.7 \times 10^4} \times \left(1 - e^{\frac{-.69 \times 1}{260}}\right) = 8.2 \times 10^{-8} \mu c/cc$$

$$A^{41} = \frac{8.95 \times 10^{18} \times 0.33 \times 10^{-24} \times 4 \times 10^8}{3.7 \times 10^4} = 3.2 \times 10^{-2} \mu c/cc$$

An inspection of these concentrations permits the elimination of A^{39} from further consideration since the activity level in the cover gas itself is about a factor of four less than the permissible continuous concentration for unrestricted (off-site) areas. The saturated A^{37} concentration in the cover gas is actually less than permissible for a 40 hour/week occupational exposure, but about a factor of eight above that used for the off-site criterion. The A^{41} component would require a decay period of about 1 day and 1½ days, respectively, to reduce the concentrations to the permissible levels for on-site and off-site exposures. Also of interest, may be the total saturation inventories of these activities in the cover gas. Using the HNPF value of 3.8×10^7 cc (1340 ft^3, excluding the primary fill tank), we have,

$A^{37} = 1.2 \times 10^{-3} \times 3.8 \times 10^7 \times 10^{-6} = 4.6 \times 10^{-2}$ curies

and similarly

$A^{39} = 3.1 \times 10^{-6}$ curies

$A^{41} = 1.2$ curies

B. Areas of Potential Design or Operational Concern

In order to evaluate any new or increased radiological problems due to the presence of argon activity in the cover gas, the following aspects were considered:

1. Normal gas leakage from top shield area during reactor operation.
2. Radioactive gas storage or disposal problems (eg., in event of necessary cover gas purges)
3. Radioactive gas transfer during fuel or moderator handling.
4. Addition radiological hazard in event of major accident.
5. Interference with cover gas fission monitoring system.

The most direct and convenient evaluation would seem to be a comparison with the expected levels of fission gases in the cover gas (i.e., Xe, and Kr). These gases, being chemically inert as argon, should appear in the same places and evoke similar radiological considerations. In HNPF, Xe and Kr activities up to 620 μc/cc have been estimated.(5) This is about 2×10^4 greater than the saturated concentration of A^{41} as previously calculated. However the Xe and Kr value includes many short half-life isotopes, the MPC's vary somewhat for rare gases and is based on a pessimistic viewpoint (1% gas release from 1% of the total reactor fuel rods). To make a more direct and conservative comparison, the following fission gas isotopes were selected:

Xe^{135} - T½ = 9.2 hours

Kr^{85} - T½ = 10.4 years

Xe^{135} has MPC's about twice those of A^{41} (4×10^{-6} μc/cc on site; 1×10^{-7} μc/cc off site), but any acute radiation problems (i.e., exposure from leakage) would be similar. Kr^{85} was chosen to compare to A^{37} on a storage or disposal basis, since the comparatively long half-life of A^{37} is its only feature of possible concern.

The total inventories of the two fission gas isotopes may be calculated as (again using HNPF parameters), Xe^{135} (atoms/sec) = 254 Mw x 3.1×10^{16} fissions/sec-Mw x 6.4×10^{-2} atoms/fission; at equilibrium, atoms/sec = disintegrations/sec,

$$\therefore \quad Xe^{135} = \frac{5.04 \times 10^{17}}{3.7 \times 10^{10}} = 1.36 \times 10^{7} \text{ curies}$$

Similarly,

$$Kr^{85} = \frac{254 \times 3.1 \times 10^{6} \times 0.26 \times 10^{-2}}{3.7 \times 10^{10}} = 5.5 \times 10^{6} \text{ curies}$$

But considering 1 year irradiation, the Kr^{85} inventory is only 7% of saturation, so

$$Kr^{85} = 5.5 \times 10^{6} \times 0.07 = 3.85 \times 10^{5} \text{ curies}$$

Approximately 0.05% of the Xe and Kr formed in the fuel will be released to the bonding fluid by the recoil process and hence available for further release in event of a cladding failure or defect. (The previous value of 1% included a liberal allowance for other release mechanisms such as diffusion. Recent experiments indicate that the recoil process is the only important one for metallic fuels under normal operating conditions). Therefore, the concentration of Xe^{135} and Kr^{85} in the cover gas may be estimated as,

$$Xe^{135} (\mu c/cc) = \frac{1.36 \times 10^{7} \text{ curies} \times 10^{6} \mu c/\text{curie} \times 5 \times 10^{-4} \times F}{3.82 \times 10^{7} \text{ cc}}$$

$$= 1.78 \times 10^{2} F$$

$$Kr^{85} (\mu c/cc) = \frac{3.85 \times 10^{5} \times 10^{6} \times 5 \times 10^{-4} \times F}{3.82 \times 10^{7}} = 5.05 \times 10^{\circ} F$$

where F = fraction of total fuel rods leaking.

If the Xe^{135} concentration is to be equal to or less than the A^{41},

$$F \leq \frac{3.2 \times 10^{-2}}{1.78 \times 10^{2}} \leq 1.8 \times 10^{-4}$$

and if the Kr^{85} concentration is to be equal to or less than the A^{37},

$$F \leq \frac{1.2 \times 10^{-3}}{5.05 \times 10^{\circ}} \leq 2.4 \times 10^{-4}$$

Since there are about 3000 fuel rods in HNPF (U-10 Mo) this means that the expected release of Xe^{135} and Kr^{85} from less than one defective rod would create cover gas concentrations and associated radiological problems equal to the A^{41} and A^{37}. Actually, we should double the F factor for Xe^{135} to allow for its higher MPC and the Kr^{85} inventory will build up more slowly than the A^{37}. However, these considerations are minor in view of the fact that the other potentially important Xe and Kr isotopes were completely neglected (eg., Xe^{133}, Kr^{85m}, Kr^{87}, Kr^{88} etc.).

Therefore, the addition of argon activity in the cover gas would not appear to present any new or increased radiological problems, since sodium graphite reactors are presently designed to handle much greater concentrations of fission gases alone. Interference with a cover gas monitor (for Xe^{133} or Xe^{135} usually) is of course possible at low fission gas concentrations. But if a gamma spectrometer type unit is used, then the fission gas isotope usually followed should be separable from any argon activity down to levels of at least ten percent of the argon present. That is, about 10^{-3} uc/cc. It is unlikely that the Xe and Kr concentrations in the cover gas will remain less than these levels very long after the reactor reaches full power operation. Certainly the capability to detect sudden changes in the cover gas activity, which is the main purpose of the monitor, would be retained. It is also possible that the presence of fairly constant A^{41} activity in the cover gas during normal operation might be adaptable to some type of a gross top shield gas leakage monitor. Such a system would probably require covering the entire top shield area with a collection reservior (eg. plastic "tent") from which a continuous gas sample could be taken.

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