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SUBJECT: Additional Calculations of the Distribution of Tritium in the MSRE

TO: Distribution

FROM: R. B. Briggs

ABSTRACT

Some of the calculations reported in CF 70-7-13 were repeated, taking into account recent information on the solubility of hydrogen in molten salts and the sorption of tritium by graphite. Reasonable agreement was obtained between the measured and calculated distributions of tritium in the MSRE. Additional experimental data are needed to reduce the uncertainties in the calculations.

Key Words: tritium, MSRE, fused salts, reactors, operation.

ADDITIONAL CALCULATIONS OF THE DISTRIBUTION OF TRITIUM IN THE MSRE

Results of calculations of the tritium distribution in the MSRE and discussion of the methods used in the calculations were reported in CF 70-7-13, "Calculation of the Tritium Distribution in the MSRE." Since the time of publication of those results, several changes have occurred:

1. Measurements by Malinauskas and Savolainen have indicated that the solubility of hydrogen in molten salt is about 1/3 the values used previously.

2. Only a small amount of lithium was found by chemical analyses of samples of insulation from the MSRE reactor furnace, so we conclude that the tritium in the reactor cell was produced in the fuel salt and diffused through the metal walls of the reactor system into the cell.

3. The rate of production of tritium in the fuel salt during the time that the tritium distribution was being measured is now estimated to be 54 curies per day.

4. Tritium was found in graphite removed from the reactor core in a quantity equivalent to a deposition rate of about 8 curies per day.

Items 2-4 are discussed by P. N. Haubenreich in a memorandum now in preparation.

The effect on the distribution of tritium of hydrogen solubility and of sorption of tritium by graphite were considered in the previous memorandum. The calculation of the solubility effect was, however, not entirely correct (reducing the solubility causes more tritium to enter the off-gas than was reported), and sorption by graphite was considered in only two cases. Because of these differences, it seemed desirable to make some additional calculations. They were made and the results are summarized in Table 1.

Values of the reference parameters in the equations that describe the tritium distribution were listed in Appendix A of CF 70-7-13. The same values were used in these calculations except that k_A, the solubility coefficient for T_2 in fuel salt, was reduced from 0.06 to 0.02, and k_B, the solubility coefficient for T_2 in coolant salt, was reduced from 0.04 to 0.02. The complete set of calculations involves cases for the following conditions.

I. With $UF_4/UF_3 = 1000$

   A. Reference condition without graphite, with graphite, and with graphite and hydrogen.

Table 1. Summary of Results of Calculations of Tritium Distribution in MSRE

Case No.	UF_3/UF_4	Condition: Mass Transfer Coefficient (x Ref.)	Condition: Metal Permeability (x Ref.)	Condition: Hydrogen	Tritium Distribution - Percent of Production: Cooling Air	Coolant Cell	Coolant Pump Off-Gas	Reactor Cell	Fuel Pump Off-Gas: T_2	Fuel Pump Off-Gas: TF	Fuel Pump Off-Gas: Total	Graphite: T_2	Graphite: TF	Graphite: Total	Concentrations, molecules/$cm^3 \times 10^{-11}$: T_2 in Fuel Salt	T_2 in Coolant Salt	TF in Fuel Salt
1	1000	1	1		8	2	0.1	17	41	31	72	0	0	0	6	0.9	130
2					2	0.4	0	3	8	14	22	3	70	73	1	0.2	57
3				*	8	2	0.1	15	37	4	41	13	20	33	310	50	940
5			0.01	*	7	2	0.2	11	41	4	45	14	21	35	340	83	990
7			0.001	*	2	0.7	0.8	3	8	14	22	3	70	73	420	320	1100
8		0.5	1		5	1	0.2	10	49	34	83	0	0	0	7	1	140
9					2	0.4	0.1	4	18	20	38	3	53	56	3	0.4	85
10				*	6	1	0.2	11	55	5	60	10	12	22	470	74	1100
12			0.01	*	5	1	0.2	10	57	5	62	10	12	22	480	91	1200
14			0.001	*	2	0.7	0.8	3	65	5	70	12	13	25	550	350	1200
15	100	1	1		12	3	0.2	24	57	4	61	0	0	0	8	1	15
16					8	2	0.1	17	40	3	43	14	16	30	6	0.9	13
17				*	10	2	0.2	20	48	0.4	48	17	2	19	400	65	110
19			0.01	*	9	3	0.3	14	53	0.5	54	19	2	21	450	120	110
20			0.001		7	2	0.3	11	44	3	48	16	17	32	6	2	13
21				*	3	0.8	1	3	66	0.5	66	23	3	26	560	440	130
22		0.5			7	2	0.2	15	72	4	76	0	0	0	10	2	17
23					6	1	0.2	12	58	4	61	10	10	20	8	1	15
24				*	7	2	0.2	13	65	0.5	65	11	1	13	550	86	120
26			0.01	*	6	2	0.3	11	67	0.5	68	12	1	13	570	110	130
27			0.001		6	2	0.3	9	60	4	64	11	10	20	9	2	16
28				*	2	0.7	1	3	77	0.6	77	14	1	15	650	420	140
Measured distribution					5-9	-	1	5-9			46-56			15			

*Indicates hydrogen added to salt at rate of 60 times the production of tritium. Addition rate is approximately that which would be produced by complete decomposition of 0.5 g/day of oil in pump bowl.

B. Permeability of metal reduced by factor of 100 with graphite and with graphite and hydrogen.

C. Permeability of metal reduced by factor of 1000 with graphite and with graphite and hydrogen.

II. Repeat I with all mass transfer coefficients reduced by factor of 2.

III. Repeat I and II with $UF_4/UF_3 = 100$.

Not all the results are reported in Table 1. The salt is the major barrier to the transport of tritium and reducing the permeability of the metal by a factor as large as 1000 had no significant effect in some cases, so the results are not included in the table. For cases with hydrogen, the hydrogen was added to the fuel salt at a rate of 3×10^{17} molecules/sec. This is 60 times the rate of tritium production and results in a concentration of hydrogen in the salt that is about what should be obtained from complete decomposition of 0.5 g per day of oil in the pump bowl.

The measured distribution is also given in Table 1 and does not account for 10–28% of the production of tritium. Some of this tritium was dissolved in the metal, some of it was held in deposits in the reactor system, but I believe that most of it must have left the fuel pump bowl in the off-gas. The percentages assigned to the cooling air, to the reactor cell, and to sorption by the graphite could not be too low by such a large amount. In my consideration of the data I assign 66 to 74% of the tritium to the fuel pump off-gas.

Examination of the data in Table 1 leads me to conclude that case 27 is in best agreement with the measured distribution. In this case, $UF_3/UF_4 = 100$, the mass transfer coefficients were reduced by a factor of 2, and the permeability of the metal was reduced by a factor of 1000 from the reference values. The uncertainty in the mass transfer coefficients is at least a factor of 2. Oxide on the metal surfaces or a change from Q proportional to $p^{1/2}$ to Q proportional to p at very low pressure might produce such a reduction in the effective permeability of the metal. In case 27 the amount of tritium sorbed by the graphite is too high, but T_2 or TF might not be sorbed as efficiently as was assumed in the calculations. The distributions in other cases, such as 10, 12, 16, in which the hydrogen has an effect, agree about as well with the measurements except that the flow rate to the reactor cell is too high, and that measurement seems to me to be our most reliable one.

Two factors deserve additional attention:

1. The distribution is sensitive to the values assigned to the mass transfer coefficients. In all calculations to date, we have changed all mass transfer coefficients simultaneously and by the same multiplier, on the basis that an uncertainty in one parameter in the calculation would apply to all the mass transfer coefficients. It

appears that the distribution could be shifted considerably by adjusting individual mass transfer coefficients. A more careful analysis of the mass transfer coefficients in each of the regions might justify changing only some of the coefficients.

2. The assumption that graphite retains both tritium and tritium fluoride might not be correct and could markedly affect the amount held by the graphite. Experimental data are needed to confirm this assumption or to provide the basis for a better one.

The conclusions and recommendations of CF 70-7-13 are not changed much by the more recent data and the calculations reported here. The graphite did indeed prove to be a reservoir of tritium and to have a greater capacity than had been anticipated. Preliminary measurements of the solubility of hydrogen in salt and analyses of the lithium in the insulation in the reactor furnace have reduced some of the uncertainties in the calculations and in the measured distribution. In calculations that give the best agreement with the measured distribution, the permeability of the metal still seems to be much lower than one might expect, although the same effect might be obtained by adjusting the mass transfer coefficients. More of the experimental data outlined in CF 70-7-13 are needed to provide an adequate understanding of the behavior of tritium in molten-salt reactors.

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