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CONTROL OF TRITIUM IN LMFBR SODIUM BY COLD TRAPPING

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ABSTRACT

Control of tritium in the sodium coolant of LMFBRs is important for achieving as low as practicable release of radioactivity. Cold-trapping has been shown to be an effective method for controlling hydrogen in sodium and should be effective for controlling tritium as well. Two mechanisms for removal of tritium from sodium have been studied: (1) coprecipitation of hydrogen and tritium from solution in sodium in a cold trap and (2) isotopic exchange of tritium in sodium with hydrogen in solid NaH in a cold trap.

Mathematical models have been developed to describe the two mechanisms and experimental runs have been made to determine their relative effectiveness. These experiments, together with the models, have indicated that the coprecipitation mechanism is, as expected, much more effective in removing tritium from sodium than the isotopic exchange mechanism. The model was used to calculate tritium levels in the EBR-II primary system, and agreement with measured levels was achieved within a factor of three.

The isotopic exchange mechanism by itself cannot be expected to adequately control tritium in LMFBRs. Hydrogen sources such as corrosion in the steam generators will be important factors in controlling the tritium level.

INTRODUCTION

One of the objectives of the Clinch River Breeder Reactor Plant (CRBRP) and future LMFBRs is to achieve as low as practicable radioactivity releases to the environment. One of the most difficult radioactive isotopes to contain in the LMFBR is tritium (^3H) because, as an isotope of hydrogen, it diffuses readily through structural materials at the LMFBR temperatures. Cold-trapping the sodium coolant is an effective method for removing hydrogen from sodium, and it is expected that the method should be effective in retaining tritium and preventing its release. The purpose of this paper is to present the results of studies with sodium cold traps and their effectiveness for removing tritium from sodium under various operating conditions.

Tritium is produced in the reactor core by three primary mechanisms: 1) three-particle fission of the fuel, 2) activation of boron in the B_4C control rods, and 3) activation of boron and lithium impurities in the fuel, sodium coolant, and structural materials. One fission in every 200-300 results in a third particle. These third particles are either alpha-particles or tritons; the alpha-particles being favored roughly ten to one over the tritons. The fission yield of tritium[1] from LMFBR fuel is expected to be of the order of 25 to 50 kd/s-Je (2 to $4x10^4$ Ci/1000 MWe-yr). Tritium production from the B_4C control rods is expected to be approximately 76 kd/s-Je (6.5 $x10^4$ Ci/1000 MWe-yr), whereas the combined production of the impurity sources should be less than 2 kd/s-Je ($2x10^3$ Ci/1000 MWe-yr).[2] Essentially all of the tritium generated in the fuel and from impurity sources is expected to be released to the coolant sodium,[3,4] whereas as little as 20 percent of that born in the control rods may be released.[5,6] Thus, the total tritium source, released into the sodium coolant is expected to be 40 to 90 kd/s-Je (3.5 to $7.5x10^4$ Ci/1000 MWe-yr). To keep the release as low as practicable, the cold traps will be required to remove at least 90 percent of this tritium burden.

MECHANISMS OF COLD-TRAP OPERATION

At least two mechanisms are available for control of tritium in sodium: 1) coprecipitation with hydrogen and 2) isotopic exchange with hydrogen in solid NaH. The coprecipitation mechanism is simple precipitation from a solution of hydrogen isotopes in sodium. Tritium should be precipitated with hydrogen in the same ratio as they exist in solution. The isotopic exchange mechanism consists of exchanging tritium in solution with hydrogen in the cold-trap deposit. The driving force for this exchange is proportional to the difference between the tritium specific activities in solution and in the solid deposit.

A simple cold-trap model was developed to describe the behavior of tritium and the operation of the two mechanisms described above. The model for the coprecipitation mechanism is quite simple in that it is assumed that tritium precipitates with hydrogen in the same ratio as the tritium/hydrogen ratio in solution. In a finite system with no significant hydrogen or tritium sources, the tritium would be removed by the cold trap along with the hydrogen, and the tritium specific activity, C_T, would decline according to the relationship

$$C_T = C_H \frac{C_{TO}}{C_{HO}} \tag{1}$$

where C_{TO} = the initial tritium specific activity, d/s-kg,
C_H = the hydrogen concentration at any time, mg/kg, and
C_{HO} = the initial hydrogen concentration, mg/kg.

The tritium/hyrogen ratio in solution would remain constant throughout the cold-trapping run.

The hydrogen concentration, C_H, changes during a cold-trapping run according to the equation

$$C_H = (C_{HO}-C_e)e^{-E_H \frac{W}{M}t} + C_e \tag{2}$$

where C_e = the equilibrium hydrogen concentration based on the minimum cold-trap temperature, mg/kg,
E_H = efficiency expressed as the fraction of hydrogen removed by the cold trap in a single pass,
W = the sodium flow rate through the cold trap, kg/s
M = sodium mass in the system, kg, and
t = elapsed time from the beginning of the cold trap run, s.

The decline of tritium specific activity in a closed sodium system with no significant hydrogen or tritium sources during a cold-trapping run can be described by combining equations 1 and 2.

$$C_T = (C_{TO} - \frac{C_{TO}\ C_e}{C_{HO}})e^{-E_H\frac{W}{M}t} + \frac{C_{TO}C_e}{C_{HO}} \qquad (3)$$

The model describing the isotopic exchange mechanism is more complex than the coprecipitation model. This complexity arises from the experimentally-determined fact that the isotopic exchange mechanism is so ineffective that hydrogen from even small sources in the system becomes important in coprcipitating with tritium. This model consists of two parts, and includes a coprecipitation term and an isotopic exchange term. The tritium mass balance in the system is expressed as follows:

$$M\frac{dC_T}{dt} = S_T - R_c - R_I \qquad (4)$$

where S_T = the tritium source term, d/s^2, (zero in these experiments),
R_c = the coprecipitation removal term, d/s^2, and
R_I = the isotopic exchange removal term, d/s^2.

If the tritium source term were not zero, it could be carried through the integration as a constant. This source term is used in models that describe the behavior of tritium in LMFBRs; however, during the cold-trapping experiments, the tritium source was zero.

The coprecipitation removal term was assumed to be the same as that described by the differential of equation 3. In describing the isotopic exchange term, R_I, it was assumed that the driving force for isotopic exchange is simply the difference between the tritium specific activity in solution and the specific activity that would be in equilibrium with the solid deposit.

$$R_I = kA\ [C_T(\text{ave}) - C_{Te}] \qquad (5)$$

where k = the tritium mass transfer coefficient, kg/m^2-s,
A = precipitation surface area, m^2
C_T(ave) = the average tritium specific activity in solution in the cold trap, d/s-kg.
C_{Te} = the equilibrium tritium specific activity based on the composition of the solid cold-trap deposit, d/s-kg.

If the isotopic exchange mechanism were solid-state-diffusion limited, the differences in specific activities above would occur in the solid deposit, and the mass-transfer coefficient would be independent of sodium flow rate.

R_I may be rewritten as

$$R_I = \alpha kA\ (C_{Tout} - C_{Te}) \qquad (6)$$

where $\alpha = \dfrac{C_T(\text{ave}) - C_{Te}}{C_{Tout} - C_{Te}}$

and C_{Tout} = the specific activity of tritium in the the cold trap effluent, d/s-kg. The constant, α, is used simply to relate the driving force to measurable quantities. Previous cold-trapping studies[7] have shown this constant to be in the range of 0.5 to 0.8.

From mass balance, R_I may also be written

$$R_I = W\ (C_T - C_{Tout}) \qquad (7)$$

Combining equations 4, 6, and 7, and integrating we obtain the equation that describes the combined isotopic exchange and coprecipitation mechanisms:

$$C_T = [C_{To} - \frac{E_T}{D}C_{Te}]\ e^{-\frac{W}{M}Dt} + \frac{E_t}{D}C_{Te} \qquad (8)$$

where $D = [E_H(1 - \frac{C_e}{C_H}) + E_T]$, and

$E_T = \dfrac{\alpha kA}{W + \alpha kA}$ = *efficiency of the cold trap for* removing tritium by isotopic exchange, fractional.

The ratio, C_e/C_H, depends upon the hydrogen source term; the larger the hydrogen source, the smaller the ratio. The term, D, is the sum of the tritium removal efficiencies of the two mechanisms. The term, $\frac{E_T}{D}C_{Te}$, is the final tritium concentration at $t \to \infty$. If there were no hydrogen source, D would reduce to E_T and the final tritium specific activity would be C_{Te}; however, with a hydrogen source, the final tritium specific activity is lower than C_{Te}. In the case where both tritium and hydrogen sources exist, the final tritium level may be either above or below C_{Te}, depending upon the relative magnitudes of the sources. The efficiencies were experimentally determined using the final tritium concentration, $\frac{E_T}{D}C_{Te}$, and equation 8; it was not necessary to determine C_{Te} experimentally.

EXPERIMENTAL

The cold-trapping experiments were conducted with the Apparatus for Monitoring and Purifying Sodium (AMPS). The AMPS is a .378 m^3 (100-gallon), forced convection sodium system capable of operation at 650°C (1200°F) and 340 kPa (50 psig) pressure. It is equipped with two cold traps: one for impurity control and one for experimental work. Impurity monitoring instrumentation, which includes an in-sodium hydrogen meter, a cover-gas hydrogen meter, an oxygen meter, two tritium meters and a multi-purpose sodium sampler are part of the AMPS system. Each of the hydrogen meters[8,9] consists of a nickel membrane inserted in the sodium (or in the cover gas) with a vacuum system attached to the nickel membrane. The vacuum system is capable of measuring either hydrogen flux (as an ion-pump current) or hydrogen pressure (with an ion gauge). The hydrogen pressure in the vacuum system reaches equilibrium quickly and can be interpreted as being in equilibrium with the hydrogen in the sodium.

The oxygen meter is an electrochemical cell,[10,11] the voltage of which is related to the oxygen activity of the sodium. The cell uses a solid ThO_2-15 wt % Y_2O_3 electrolyte tube and an air reference electrode. This type of cell has been used extensively in sodium systems with good results.

The tritium meters[12] are similar to the hydrogen meters in that they use nickel membranes to allow the tritium to diffuse through from the sodium. They differ in that an argon-1% hydrogen sweep gas is used to carry the tritium from the membrane to a counting chamber where the tritium activity is constantly monitored.

The multi-purpose sampler[13] was used both to take sodium samples for tritium analyses and to equilibrate scandium tabs and vanadium wires. The scandium tabs and vanadium wires were analyzed for hydrogen and oxygen content respectively, and these analyses were correlated with hydrogen and oxygen activities in sodium using previously determined distribution coefficients. The results of these analyses were compared with the hydrogen, oxygen and tritium meter readings for calibration purposes.

One of the tritium meters was used as a tritium-injection device to add tritium to the sodium. Argon containing 200 vppm hydrogen and tritium with an activity of 10×10^{10} d/s-m^3 was passed through the membrane region at a very low flow rate. The tritium entered the sodium quantitatively, so that the amount added could be accurately determined by integrating the gas flow.

The cold-trap used in these experiments was of an integral economizer-crystallizer design with the economizer located in the upper half of a vertical pipe 168 mm O.D. x 7.1 mm wall thickness, [6-in. schedule 40 (NPS)] and the crystallizer region in the lower half. Sodium flowed into the top of the cold trap, downward through the economizer into the annulus of the crystallizer region. The crystallizer was packed with stainless-steel wire mesh and was air-cooled on the outer surface. The sodium continued up the center section of the crystallizer, through the economizer, and out the top of the cold trap. Temperature control was maintained by varying the input to heaters on the external surface of the cold trap while the air coolant flow was maintained constant. The volume of the crystallizer region was 9300 cm^3 (2.46 gallons).

The hydrogen source term for the AMPS was experimentally determined at two separate times. The primary sources of hydrogen were thought to be the two tritium meters and corrosion of the piping by atmospheric moisture. During all the runs, except run 1-15-76, both tritium meters were in operation using a sweep-gas composition of argon + 5 percent hydrogen. For run 1-15-76, one of the tritium meters was shut down and the other meter was operated with a sweep-gas composition of argon + 1 percent hydrogen. The hydrogen source was measured before this change and was found to be 140 ng/s. Following the change, the source dropped to 8.6 ng/s. The sources were measured over periods of three weeks and one week, respectively. The day-to-day variations in the source are not known.

The coprecipitation runs were initiated with high (above equilibrium) hydrogen and tritium concentrations. The cold-trap flow was started with a low cold-trap temperature, and the hydrogen and tritium concentrations were carefully monitored as the cold trap removed them from the system.

The isotopic exchange runs were conducted differently. Both hydrogen and tritium were cold-trapped to a low level initially and the cold trap was isolated from the system. Tritium was injected to increase the tritium/hydrogen ratio, and the hydrogen concentration was carefully measured. The cold trap was then started up at the hydrogen saturation temperature to avoid any transfer of hydrogen into or out of the cold trap. The tritium specific activity was then measured as it declined, and the hydrogen concentration was measured periodically to be assured that it was remaining constant.

RESULTS

Tritium Injections

A total of five coprecipitation and five isotopic exchange cold-trapping runs were completed. Tritium injections were made prior to each isotopic exchange run to increase the tritium/hydrogen ratio at the beginning of each run. The results of the tritium injections are summarized in Table I. The injections resulted in excellent recovery of the tritium, *i.e.*, the tritium concentrations observed with the tritium meter were, in all cases, within 27 percent of the quantities added. This discrepancy is thought to be an accumulation of uncertainties in gas flowmeter readings and statistical counting variations. The tritium measurement error is thought to be less than the injection error because the flowmeters were operating very near the lower end of their range. Uncertainties in reading flow rates could easily reach 20 percent. The data appear to have a negative error bias that could also be attributed to a bias in the flowmeter at the lower end of the scale.

Table I. Tritium Injections into AMPS Sodium

	Tritium (kd/s-kg)				Diff.
No.	Inj.	Init.	Total	Meas.	(%)
1	10.1	2.52	12.6	14.5	+15
2	11.8	2.48	14.3	14.2	-.8
3	23.6	2.78	26.4	26.1	-1.1
4	23.6	1.96	25.5	18.7	-26.8
5	23.6	21.5	45.1	38.8	-13.8

Coprecipitation Runs

The first assumption in the coprecipitation model is that tritium and hydrogen are precipitated in the same ratio as they exist in solution. This assumption leads to the conclusion that the tritium/hydrogen ratio should remain constant during a coprecipitation run. The tritium/hydrogen ratio was determined as a function of time during each of the five coprecipitation runs. Some variation was observed in the ratios, and the limits of these variations are shown in Table II. Also shown are the hydrogen removal efficiencies determined with the use of equation 2, and the uncertainties associated with the counting statistics for the different tritium activity levels of the various runs.

Table II. Variations of Tritium/Hydrogen Ratios During Coprecipitation Runs

Run No.	Residence Time, (s)	H Removal Efficiency (percent)	T/H Ratio (kd/s-mg)	Final T Conc. (kd/s-kg)
6-18-75	150	40	15 ± 31%	1.29 ± 11%
7-17-75	588	100	4.8 ± 23%	.78 ± 12%
7-30-75	588	100	28 ± 33%	1.89 ± 10%
9-10-75	264	63	30 ± 4%	25.9 ± 4%
9-25-75	192	67	17 ± 6%	10.0 ± 6%

The tritium/hydrogen ratio remained constant within statistical counting variations for the last two runs; however, larger variations were observed during three earlier runs. These first three runs were conducted at such low tritium levels that the variations in tritium/hydrogen ratio, although larger than the statistical counting variations, are not considered to be significant.

The hydrogen and tritium reductions during run 9-10-75 are shown in Fig. 1. The data points are the concentration and specific activity readings, whereas the curves represent equations 2 and 3 plotted with a hydrogen removal efficiency of 30 percent. As can be

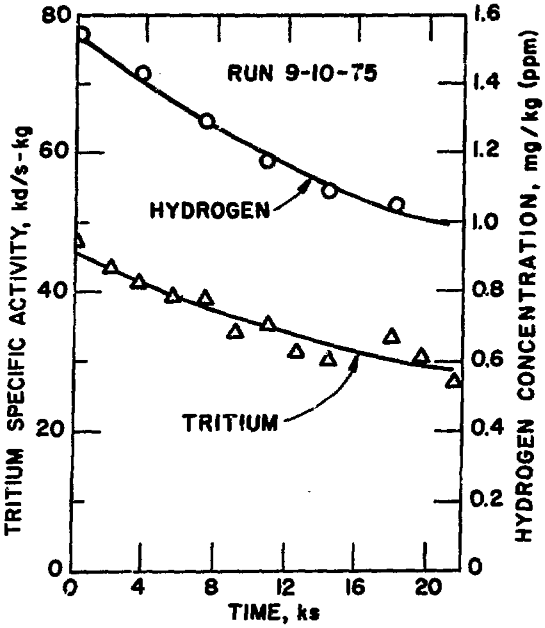

Fig. 1. Tritium and hydrogen removal by the cold trap during run number 9-10-75.

seen from this figure, the coprecipitation model expresses the tritium and hydrogen reductions very well.

Isotopic Exchange Runs

The isotopic exchange model (equation 8) was used to analyze each of the five runs of this type. One of these runs (run 1-15-76) is shown in Fig. 2 where the tritium specific activity and hydrogen concentration are shown as a function of elapsed time following the startup of the cold trap. Equation 8 is plotted using a tritium removal efficiency of 22 percent. The fit of the model with the tritium specific activity data is fair, but not as good as desired. The tritium level drops more rapidly in the first 50 ks of the run than the model predicts; also the decline continues more rapidly than the model predicts after 400 ks of elapsed time. Some slight variations in the hydrogen source term during these runs could cause these deviations from the predicted behavior.

Summary of Coprecipitation and Isotopic Exchange Runs

Equations 3 and 8 were used to determine the cold-trapping efficiencies of all the runs conducted in this study. Efficiencies were chosen that resulted in the best fit with the experimental data. The results of this analysis are shown in Table III where the sodium residence times in the cold trap, the sodium flow rate and the resulting tritium removal efficiencies are listed for each run.

The data in Table III are shown graphically in Fig. 3, where cold-trap efficiencies are plotted *vs.* sodium residence times. In general, efficiencies

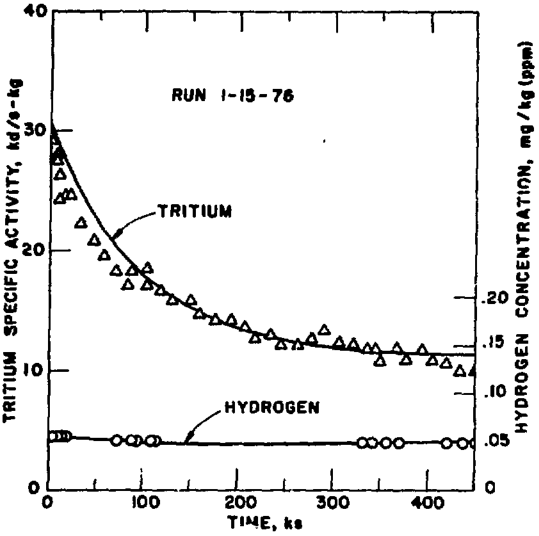

Fig. 2. Tritium removal by the cold trap during run number 1-15-75.

Table III. Cold-Trapping Efficiencies for All Runs

Run No.	Sodium Flow Rate (cm^3/s)	Sodium Residence Time (s)	Tritium Removal Efficiency (Percent)
Coprecipitation Runs			
6-18-75	62	150	40
7-17-75	16	580	100
7-30-75	16	580	100
9-10-75	35	270	63
9-25-75	47	200	67
Isotopic Exchange Runs			
11-6-75	32	290	7.0
11-11-75	32	290	7.5
11-19-75	60	160	2.4
12-8-75	17	550	30
1-15-76	16	580	22

increase with increasing residence times, as expected. The effectiveness of the isotopic exchange mechanism is much lower than that for the coprecipitation mechanism; however, the efficiencies increase with residence time in both cases.

DISCUSSION AND CONCLUSIONS

The specific cold trap used in this study has demonstrated impurity removal characteristics similar to some cold traps of similar design that have been tested in the past.[14] Hydrogen removal efficiencies tend to increase with increasing sodium residence times up to 300 to 600 s. Tritium removal efficiencies were found to increase similarly in the coprecipitation experiments.

The data obtained in this work are applicable to a specific cold trap and sodium system, and caution should be used in applying the numbers to systems that are significantly different. Large variations in cold trap efficiencies from system to system are not expected; however, the scaling factors are not well understood and are the subject of continuing work.

The general conclusions are considered to be valid for most cold-trap systems; however, the uncertainties in the efficiencies applied to other systems are not known.

To assess the applicability of the mathematical model developed in this work, a modified version of equation 8 was compared with tritium data from the EBR-II primary sodium system. It was necessary to modify equation 8 to include the tritium source term and an overall system-boundary permeation term. The modified equation is:

$$C_T = \left(C_{TO} - \frac{E_T}{D} C_{Te} - \frac{S_T}{DW}\right) e^{-D\frac{W}{M}t} + \frac{E_T}{D} C_{Te} + \frac{S_T}{DW} \qquad (9)$$

where D is redefined as $\left[E_H\left(1 - \frac{C_e}{C_H}\right) + E_T + \frac{P}{W}\right]$, and P = an overall permeation constant, kg/s. In the calculations, Kumar's[15] values for the permeation constant, 0.032 kg/s, and tritium source term, 288 kd/s^2, were used. The "equilibrium" tritium concentration for C_{Te} (if no hydrogen source were present, C_{Te} would be the final tritium concentration) was determined using the following reasoning: The rate of hydrogen deposition in the cold trap was calculated from the hydrogen concentration in the EBR-II system, the equilibrium concentration based on the cold-trap temperature, and the cold-trap flow. The rate of deposition was calculated to be 19 μg/s. The rate of tritium transfer to the cold trap was calculated using Kumar's source term and the permeation constant. This transfer rate was 231 kd/s^2. The ratio of these rates should be equal to the tritium/hydrogen ratio in the cold-trap deposit. This tritium/hydrogen ratio is 12.3 Md/s-mgH. The equilibrium hydrogen concentration is known based on the cold trap temperature, and the equilibrium tritium concentration should be in the same ratio as that in the solid deposit, *i.e.*, $C_{Te} = C_e \times 12.3$Md/s-mgH so that C_{Te} was calculated to be 737 kd/s-kg.

The hydrogen and tritium removal efficiencies were taken directly from the data in Fig. 3 so that $E_H = 1$ and $E_T = 0.3$. Figure 4 is a plot of equation 9 showing the predicted rise of the tritium specific activity in the EBR-II primary system from an initial valve of zero to equilibrium at ~1000 kd/s-kg (27nCi/g) after one month of operation. Also shown in figure 4 are data from three reactor-startup periods over the last three years. In general, the predicted specific activity is within a factor of three of the observed values, and the activities appear to rise to equilibrium in approximately one month of operation. (The vertical bar represents the range of equilibrium values that have been observed in EBR-II primary sodium in recent years). Considering the uncertainties in some of the values used in this analysis (source term, permeation data, and correlation between AMPS cold trap and the EBR-II cold trap), the agreement appears to be fairly good. Additional tritium data from EBR-II, along with improved permeation data and additional AMPS cold-trapping data, should be helpful in improving the LMFBR tritium behavior model.

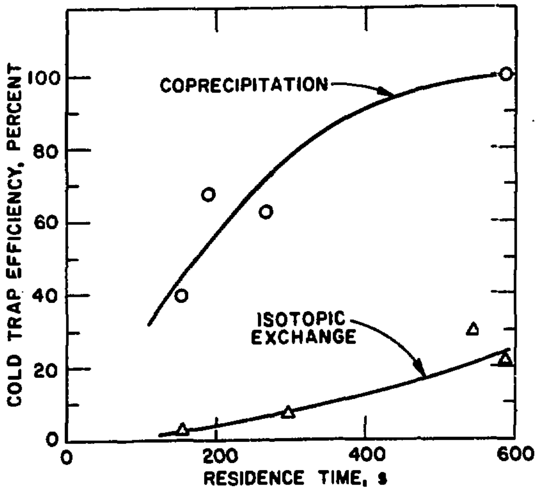

Fig. 3. Cold-trapping efficiencies for coprecipitation and isotopic exchange experiments.

Several additional conclusions may be drawn from the results of this study with the AMPS system:

A. The isotopic exchange mechanism is much less effective than the coprecipitation mechanism for removing tritium from sodium. The data produced in this study indicate that for any given sodium flow rate, the isotopic exchange mechanism is only 10 to 20 percent as effective as the coprecipitation mechanism.
B. The isotopic exchange mechanism by itself was not considered a candidate mechanism for controlling tritium in future LMFBRs. This work has confirmed the previous thinking and has allowed order-of-magnitude prediction of expected efficiencies of the mechanism. At typical LMFBR cold-trap residence times (approximately 5 min.) the tritium removal efficiency with no hydrogen source will be less than 10 percent. Normal hydrogen sources in the system will increase the overall tritium removal efficiency; the extent of removal will depend on

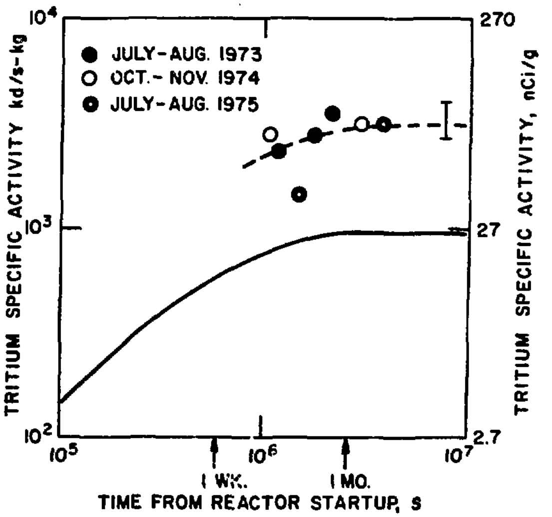

Fig. 4. Tritium specific activities observed in EBR-II primary sodium compared with the activities predicted by the mathematical model.

the magnitude of the hydrogen source.

C. Models designed to describe the behavior of tritium in LMFBRs should account for hydrogen sources, including corrosion in the steam generators and permeation through the IHX from the secondary to the primary system. These hydrogen sources may constitute the primary mechanism for removal of tritium with the primary and secondary cold traps.

D. The equations developed in this work are useful in determining the respective hydrogen and tritium removal efficiencies of a cold trap. The equations also allow separate determinations of the coprecipitation efficiency and the isotopic exchange efficiency. Additional work is needed (and is planned) to describe the hydrogen and tritium mass-transfer coefficients and the composition and location of the impurity deposits. This work will allow development of a more sophisticated model for tritium cold-trapping.

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