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TITLE

N-REACTOR CO-PRODUCT TARGET ELEMENT RUPTURE DETECTION STUDY

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N-REACTOR CO-PRODUCT
TARGET ELEMENT RUPTURE
DETECTION STUDY

By

Duane O. Allred

Controls, Instruments, and Systems Analysis
Research and Engineering Section
N-REACTOR DEPARTMENT

April 27, 1964

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N-REACTOR CO-PRODUCT TARGET ELEMENT
RUPTURE DETECTION STUDY

1.0 INTRODUCTION

1.1 Purpose and Scope

At present there seem to be some uncertainties concerning the needs of rupture detection for the co-producer program. Also in question is the method by which ruptures of the target elements should be detected and located. The purpose of this report is to discuss the waste and hazard control of tritium (H^3) (the product of the co-producer program), which will determine rupture detection needs, and to discuss methods by which rupture detection and location may be accomplished.

The scope of the report will consider first, adaptation of the present rupture monitoring system, and second, monitoring systems using H^3 analyzers, together with the costs and time required to develop and use each method of rupture detection.

1.2 Problem Statement

The target elements are an alloy of lithium and aluminum which is clad in aluminum. This in turn is slipped into a zirconium can with a mechanical bond between the two metals. The zirconium can is somewhat longer than the aluminum cladding to allow for the greater expansion of aluminum.

The element is roughly two feet in length by 1/2-inch in diameter.

If the zirconium jacket should rupture, ^{calc?} it has been shown that the release rate of Al^{28} from the aluminum jacket will not be great enough for the present rupture monitor to detect above normal background. ⁽¹⁾

The problem of immediately detecting and/or locating a rupture where only the zirconium cladding has failed is more complex than the problem of rupture location only when the H^3 concentration within the loop exceeds certain limits. The latter problem would provide a relatively long period of time in which to detect and locate a rupture. This would mean simpler methods and less costly instrumentation.

The criteria for target element failure detection have not yet been clearly defined. Questions that must be answered are:

1. What will be the maximum permissible concentration of H^3 in the primary loop?
2. What will be the maximum allowable release rate of H^3 into the river?
3. What must be the response of the monitoring system?

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4. How soon must a monitoring system be available and at what maximum cost?
5. Is a monitoring system actually needed? *Why bother? It is only tedious*

2.0 SUMMARY OF RESULTS AND CONCLUSIONS

note: 5 tubes all
A previous study ⁽¹⁾ has shown that the present rupture monitor will be useless for detection of target element failures unless the elements are modified with a gamma emitting tracer which would be released by a rupture at a higher rate than Al^{28} will be. This survey shows that many questions concerning target element failure detection criteria must be answered before a method can be decided upon or definite conclusions drawn.

The first conclusion of this report is that a manual sampling and monitoring system should be installed. A sampling system has already been proposed for the 36 tube load, which could service the entire reactor. ⁽²⁾ Also, the 16 diversion headers should be tapped so that the limitation of five process tubes at a time can be diverted and sampled. After determining which set of five tubes the rupture is in, each of the five tubes can be sampled to determine which tube contains the rupture. Both of these sample systems could probably be installed for less than \$7500.

An automatic dual channel refrigerated liquid scintillation spectrometer system ⁽³⁾ such as the one referenced should be installed. This method is chosen for simplicity, sensitivity accuracy and would cost \$12,500. The analyzer could be used for the analyses of elements other than H^3 .

By sampling five tubes at a time, the reactor can be covered in 200 samples. Allowing ten minutes to trap and prepare each sample, it would require about 40 working hours to locate the rupture. Each sample will require at least a ten minute counting period, but samples can be prepared while others are being counted.

This analyzing equipment is available at the Hanford Laboratories in the 300 Area. If this equipment could be used when needed, the samples taken from the reactor could be sent directly to the 300 Area without further preparation.

The first conclusion is based on the apparent high reliability and integrity of the target elements, the low corrosion rate of aluminum and the resulting probable slow release rate of H^3 from a ruptured element, the apparent limits of H^3 release rates to the river and the availability of existing equipment on plant.

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The second conclusion of this report is based on the cost of sample analysis and possible reactor down time. Also, there is some uncertainty about the target elements as to how they may rupture, how fast the H^3 will be released and allowable H^3 concentrations in the primary loop.

At \$10 per sample analysis, it would cost \$2,050 plus possible reactor down time every time a failure had to be located. The estimate of \$10 per sample is what the Hanford Laboratories would charge to run several samples. However, this charge is primarily for technician's time. The estimated cost per sample for material is \$0.30. Two hundred samples would only cost about \$60 for analysis. A problem may arise in that the failure may not be located or sampled before the reactor must be shut down. Of course water will still be circulated while the reactor is down so a rupture may still be located.

It will be pointed out that to monitor one or two tubes at a time automatically will require about the same length of time as to do it manually with the same type of equipment since the counting time for H^3 detection is no less than ten minutes for sufficient accuracy. Thus, if a rupture can not be monitored manually, the monitor system should be able to detect a failure almost instantly in any tube. This implies that a tracer material should be developed so the present rupture monitor may be used. This would require much time and money. The alternate to this proposal would be to have enough on-line H^3 analyzers to cut the monitoring time of the entire load down to a reasonable figure.

It is important to point out that if the present rupture monitoring system is not used for instant (within one minute) detection and location, the most economical and practical method studied would be to take samples in groups of five process tubes manually and analyze the samples in a liquid scintillation analyzer. This would only have to be done when an element ruptured and the H^3 concentration in the loop exceeded a certain limit. An on-line H^3 monitor would not be significantly faster unless several analyzers were used. The cost of such a system could be prohibitive. Other problems would arise such as space and maintenance.

3.0 PROBLEM DESCRIPTION

3.1 Co-Product Reactor Loading

The initial co-producer load will be contained in 36 process tubes.

Later programs may involve the entire reactor. Needless to say, the monitoring problems for 36 tubes are much simpler than for 1004 tubes or some other large part of the reactor.

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3.2 Radioactive Waste Control

Some existing H^3 waste control limits are: ⁽⁴⁾

A. The Federal Radiation Council Guide (RCG)

The RCG has set a limit at 3×10^{-4} uc/cc. The total river burden at this concentration could be from 75,000 curies per day to 120,000 curies per day depending on the river flow rate.

B. HW-53612

This design criteria limits the H^3 release rate to the river from Hanford to 1% of the (RCG). The total H^3 in the river could vary from 750 curies per day to about 1200 curies per day depending on the river flow.

C. Savannah River Plant

The Savannah River plant uses a river concentration limit of 3×10^{-4} uc/cc. In the Columbia this would mean 75,000 curies to 120,000 curies per day depending on the river flow rate. This would correspond to the (RCG)_w.

D. Hanford 100-KE Reactor ⁽⁵⁾

The H^3 concentration limits in the loop while running target element tests were 400 uc/cc in a recirculating loop. With a maximum loop bleed rate of 1.5 gpm, the total release rate to the river could have been 3200 curies per day. The actual H^3 concentrations in the loop during the tests were less than 10^{-3} uc/cc.

E. Shippingport Reactor

The Shippingport Reactor in Pennsylvania is allowed a H^3 release rate of an average of 10 curies per day and a maximum of 50 curies per day.

It has been proposed that the maximum allowable H^3 release rate to the river from N-Reactor be 750 curies per day ⁽⁴⁾. This control limit would be consistent with the Hanford design criteria (HW-53612 Rev.) above. The H^3 concentration in the Columbia River ^{by here?} at present is about 10^{-6} uc/cc, or a background of 250 curies/day. The proposed 750 curies/day limit with the background of 250 curies/day would be only slightly over the design limits of HW-53612 with all eight of the older reactors operating at low river flow. Of course the proposal of 750 curies/day does not mean that 750 curies/day is expected as the normal release rate, but rather suggests a control point.

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It has been calculated that the H^3 concentration in a target element will be about 400 curies. With a bleed rate of about 600 gpm or 3.3×10^9 cc/day the loop concentration would be .22 uc/cc to release 750 curies/day into the river. With a loop volume of 8×10^8 ml, one element could create a loop concentration of .5 uc/cc if dissolved immediately. This means that the equivalent of one-half of a target element would have to be constantly dissolved in the loop to release 750 curies per day of H^3 into the river. This means that nearly two elements per day would have to corrode away to release 750 curies day to the river.

3.3 Personnel Radiological Hazard Control

The maximum permissible concentration of H^3 for 168 hours occupational work week is 0.03 uc/cc in water $(MPC)_w$ and 2×10^{-6} uc/cc of air as the oxide in air $(MPC)_A$. The $(MPC)_w$ is limited by the average amount of water consumed per week by an individual. However, it is not likely that anyone would be drinking primary loop water. If the H^3 in the loop were at the $(MPC)_w$ then 100 curies/day would be released to the river, and one-fourth of an element per day would be dissolved. The $(MPC)_A$ is based on the water vapor in the air a person breathes. The $(MPC)_A$ of 2×10^{-6} uc/cc of air as the oxide in air corresponds to 120° F saturated air with water containing the $(MPC)_w$ of .03 uc/cc. If it is possible that personnel might work in a room and breathe 120° F air saturated from primary coolant, then the loop should be restricted to an MPC of .03 uc/cc.

4.0 SURVEY OF TARGET ELEMENT FAILURE MONITORING TECHNIQUES

4.1 Tritium Detection Using Liquid Scintillators

4.1.1 Description⁽³⁾

A liquid scintillator contains only two essential ingredients, the solvent and the solute, however complicated the composition may be. The solvent absorbs and transfers the energy of a particle to the solute, which emits a burst of photons for each particle that transfers its energy to the solution. There are numerous solvents and solutes which may be used to detect H^3 , but is beyond the scope of this report to discuss the actual composition of the many liquid scintillators.

A reactor water sample is prepared for counting by double or triple distillation to remove beta emitting impurities, followed by the addition of a small sample of the condensate to the scintillator solution. This mixture is contained in a transparent vial which is counted in a well counter. The solutions are

Why don't H^3
leave ?
if T_2O it might stay
but T_2 stable here

prepared manually and the scintillator is not re-usable.

This method of H^3 counting does not lend itself to on-line monitoring since each sample is introduced to the analyzer by hand and the scintillator is not re-usable. The typical liquid scintillation counting system consists of two photomultiplier detectors monitoring the sample vial. The outputs of the two detectors are fed to an anti-coincidence circuit which provides discrimination between random detector noise and actual signals.

The detectors and counting well are enclosed in a refrigerated unit to lower the thermal noise of the photomultiplier tubes. There are two types of counting systems available, a semi-automatic counter where each sample vial is placed in the counting well manually, and an automatic counter which automatically counts up to 200 sample vials one at a time.

4.1.2 Sensitivity

This method of counting has a sensitivity in the order of 10^{-6} uc/cc of H^3 in the water sample with a normally prepared scintillator. The scintillator may be refined by electrolysis, yielding a counting sensitivity in the order of 10^{-8} uc/cc. With a 100 minute counting time the accuracy in both cases is $\pm 25\%$. If the concentration of the sample is in the order of 10^{-3} uc/cc, a 10 minute count would yield sufficient accuracy with the unrefined scintillator.

4.1.3 Time and Costs

The cost of the instrumentation ranges from \$5000 for a semi-automatic system to \$12,500 for an automatic system⁽³⁾. The cost of the scintillator is about \$0.30 per sample for the non-refined scintillator.

This monitoring method could be installed in a matter of a few weeks at N-Reactor along with a manual sampling system for \$10,000 to \$15,000 using shelf item equipment. The Hanford Laboratories have this same equipment available for a cost of \$10 per sample analysis for large numbers of samples.

4.2 Tritium Detection Using Solid Scintillation Crystals^(3,6,7)

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4.2.1 Description

This method uses solid anthracene crystals packed in a transparent tubing. Highly distilled tritiated reactor water is slowly and continuously passed through the tubing while two scintillation detectors monitor the activity in the same way as explained in method 4.1.

The advantage of this method over the liquid scintillator is that the crystals are re-usable and do not become contaminated from one sample to another. That is, one sample and then another may pass over the crystals without any cleaning or changing of crystals. The counting instrumentation and scintillator are shelf items.

4.2.2 Sensitivity

The sensitivity of this method is less than 10^{-3} uc/ml. A counting time of at least 10 minutes is required for sufficient accuracy.

4.2.3 Time and Cost

The cost of the counting instrumentation would range in the order of \$5000. The cost of the sample system would depend on the method of sampling and sample preparation.

4.3 Gas Proportional Counting (3,8,9)

4.3.1 Description

In this method tritiated water vapor is mixed with other gases such as methane, helium, butane, argon, etc., and counted in a proportional gas counter. This method may be used as a batch or flow counter. If the counter is not flushed with pure water vapor between samples, a slight contamination of the detector from sample to sample may arise.

Some of the difficulties with this method are control of the gas mixtures and pressures, the water vapor pressure, temperature of the counter, etc. These conditions must be controlled for accurate H^3 concentration measurements. However, such instruments are made and are shelf items.

As with the scintillator methods, the samples would have to be highly distilled to remove beta emitter impurities. However, the process of introducing the sample partially performs this distillation.

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4.3.2 Sensitivity

The sensitivity of this method can be greater than the liquid scintillation method as measured in 10^{-6} uc/cc. If a sensitivity in the order of 10^{-3} uc/cc is sufficient, good counting accuracies may be obtained from simpler and cheaper proportional counters in a reasonable counting period.

4.3.3 Time and Cost

Simple proportional gas counting instrumentation can be purchased for less than \$5000. However, the samples would have to be prepared and run through the system manually. The Beckman Instrument Company makes an on-line proportional counter for about \$20,000 which prepares and runs the sample through automatically. However, since the water samples would be from the reactor, the samples would have to be distilled before feeding them to the counter.

The time that would be required to install this method at N would, as in the case of the other methods, depend on the requirements and design of the sampling system.

4.4 Tracer Method

4.4.1 Description

The present rupture monitor could be used if a material could be placed between the aluminum and zirconium jackets of the target elements, which would become a gamma emitter after irradiation. This method would provide instant rupture detection and location, but there are several problem areas:

1. The aluminum and zirconium jackets have a close fit and heat conduction between the two must not be significantly altered.
2. A tracer material must not cause chemical reactions between the two jackets.
3. The irradiated tracer can not cause the element to be "hot" to handle without special handling facilities, since the zirconium jacket is removed manually in a glove box. In other words, the tracer must have a short half life.
4. The tracer must dissolve readily when a rupture occurs and create a continuous signal on the rupture monitor.
5. The tracer must be economical and feasible to apply to every target element.

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6. The tracer must not degrade the integrity of the target element in any way.
7. The tracer isotopes must emit gammas within the energy range of the monitor.
8. The operating temperature of the elements will be from 250° C to 300° C.
9. No tracer yet developed meets the above requirements.

4.4.2 Sensitivity

The sensitivity of this method would be that of the rupture monitor.

4.4.3 Time and Cost

Since no such tracer has yet been developed, a program development and testing would have to be initiated. The time required to complete such a program might run from half a year to a year, assuming such a tracer could be developed.

The cost of the program would most probably be quite high, considering extensive reactor testing, materials, the development of tracer application to the elements and the time required.

5.0 PROPOSED MONITORING METHOD⁽²⁾

5.1 Description

A method for target element failure detection has been proposed for the initial 36 tube reactor load. This method consists simply of installing a manually operated sample system on the valve racks in both rupture monitor rooms (see Figure 1). The samples on both racks could be sampled one at a time. With a small reactor load, this method is simple, fast and practical for locating an element rupture. In a large reactor load this method may not be practical from the standpoint of sampling time.

5.2 Sensitivity

The samples taken may be sent to the Hanford Laboratories where the H^3 concentration can easily and inexpensively be measured to 10^{-6} uc/ml.

5.3 Modifications and Additions Required

The only modification required would be the addition of a tee connector on each sample flow calibration differential transducer (d/p) in both rupture monitor rooms as shown in Figure 1. The additions would only

*It is in sec. why
don't we sample lines
from 150th man so we
don't have them for all
programs? We need
them now.*

PROPOSED SAMPLE SYSTEM FOR THE 36 TUBE TEST**DECLASSIFIED**

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require 8 valves and several feet of tubing on both valve racks. These modifications could be completed within a week.

5.4 Installation and Monitoring Costs

The cost of installation plus material has been estimated at about \$2500. If one sample were taken periodically the cost of the analysis would be about \$15 per sample. If the entire load were sampled, the cost of analysis would be about \$10 per sample.

6.0 DISCUSSION

6.1 Monitoring Full Reactor Load

6.1.1 Sampling by Groups

The entire reactor could be sampled manually within 24 hours by diverting five process tubes at a time and sampling the diversion header. In this way there would only be 200 samples taken. All 200 samples could be analyzed within 48 hours by the Hanford Laboratories. Or, an analyzer could be installed at N-Reactor. With a H^3 concentration of 10^{-2} uc/cc in the loop a 10 minute count of each sample would yield an accuracy of $\pm 25\%$ and only 33 curies per day would be released to the river.

When the tubes are diverted in sets of five the bleed rate of the loop will increase by about 1000 gpm and the H^3 concentration in the loop will decrease rapidly. Since the H^3 concentration will change with the increased bleed rate, the bleed rate should be increased prior to sampling by the equivalent of five tubes until the H^3 concentration in the loop reaches equilibrium. The amount of time required to reach equilibrium is calculated as follows:

$$Q_L = \frac{Q_s f_B}{V}$$

1)

Q_L = concentration loss of the loop

Q_s = concentration in the loop

f_B = total bleed flow rate of the loop

V = loop volume

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$$Q_A = \frac{X}{V}$$

2)

where

Q_A = concentration gain of the loop

$X = H^3$ release rate from the rupture

$$\frac{dQ_s}{dt} = Q_A - Q_L = \text{loop concentration rate of change at any time } t$$

$$\frac{dQ_s}{dt} = \frac{X}{V} - Q_s f_B \quad 3)$$

$$\frac{dQ_s}{dt} + \frac{f_B}{V} Q_s = \frac{X}{V} \quad 4)$$

The general solution to equation 4) is

$$Q_s = A \text{Exp.} \left(\frac{f_B t}{V} \right) + B \quad 5)$$

from the general solution 5) the time constant (TC) can be seen to be

$$TC = \frac{V}{f_B} \quad 6)$$

where $V \approx 180,000$ gal.

$$f_B \approx 1600 \frac{\text{gal}}{\text{min}}$$

$$TC \approx 113 \text{ min.} \approx 1.9 \text{ hr.}$$

Allowing four time constants to reach equilibrium it would require that the loop be bled at 1600 gpm for about 8 hours before samples were taken by diverting five tubes at a time into the diversion header. As long as five tubes were constantly being diverted the H^3 concentration in the loop would remain constant.

This all assumes that the H^3 release rate (X) from a rupture remains constant.

If X is a constant and the loop concentration is at equilibrium then

$$X = Q_s f_B \quad 7)$$

or in other words, the H^3 gain equals the H^3 loss.

$$Q_h = Q_s + \frac{X}{f_D} \quad 8)$$

where Q_h = diversion header concentration

f_D = diversion header flow rate

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Substituting 7) into 8)

$$\frac{Q_h}{Q_s} = 1 + \frac{f_B}{f_D}$$

9)

with a bleed rate of 1600 gpm (normal bleed of the loop + five tube diversion bleed)

and a diversion flow of 1000 gpm equation 9) becomes

$$\frac{Q_h}{Q_s} = 1 + \frac{1600}{1000} = 2.6$$

This shows that as long as X remains constant Q_h will be at least 2.6 times as great as Q_s .

If X is not constant, then there may be trouble in determining in which set of five tubes the rupture is located. In this case equation 8) must be left as is, so Q_h exceeds Q_s by $\frac{X}{f_D}$, and then $\frac{X}{f_D}$ shall have to be more than 25% greater than Q_s

for detection since the counting accuracies anticipated are at best $\pm 25\%$. Also, if X is not constant, a reference sample of the loop will have to be taken at the same time the diversion header is sampled. If not, it must be assumed that the rate of change of X is small enough so that two succeeding diversion samples may act as reference for one another; these are all pretty shaky assumptions and serious problems could arise if the H^3 release ceased before that tube was sampled. It is a known fact that the corrosion rate of aluminum decreases because of an oxide build up. This fact also indicates that the H^3 release (X) would probably not be constant.

6.1.2 Individual Tube Sampling

If each process tube were sampled manually with the system described in section 5, it would require about 50 hours of sampling time to cover the reactor. This is based on taking two samples at a time, one in each monitor room, and requiring five minutes per sample.

With a counting time of ten minutes per sample it would require about 170 hours to analyze a full reactor load. This time could only be reduced by using more than one analyzer.

6.2 On-Line Continuous Monitor

The problems with an on-line monitor are as follows:

1. Unreasonableness of having a separate monitor for each process tube.
2. Automatic sample preparation for each tube.
3. Scanning from sample to sample.

4. Design and development of the sampling system.
5. Possible lack of need for analyzing the entire load until the loop H^3 concentration exceeds set limits.

The cost and space that would be required to sample and analyze each process tube for H^3 would be prohibitive and probably not justified on the basis of quick detection and location.

The samples must be double or triple distilled to eliminate beta emitting impurities. To have a separate distillation unit for each sample may not be practical. One solution would be to have one sample being prepared while another sample is in the process of being analyzed.

The problem with scanning from sample to sample is the pressure that the loop is under. The samples could be withdrawn from the low side of the present rupture monitor sample system heat exchangers where the water has been cooled to about 120° F and the pressure is about 1600 psi. To automatically connect and disconnect from a 1600 psi source would present some difficulty.

An on-line monitor should have at least two monitoring systems so that one sample analysis could be used as a reference for the other. Since it still requires at least ten minutes to count each sample, it would require about 83 hours to scan the entire reactor analyzing two samples at a time. At this rate only one representative sample of the loop should be analyzed periodically to determine whether or not a H^3 releasing ruptured element exists. Then the rupture can be located. Actually, the samples could be taken, prepared and analyzed manually about as fast as they could automatically since with a manual system, samples can be taken and prepared while others are being analyzed.

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