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SMALL-LEAK BEHAVIOR SUMMARY REPORT ON SCOPING TESTS

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Small Leak Behavior
Summary Report on Scoping Tests

D. W. Sandusky

ABSTRACT

A scoping phase has been completed on a study of small steam leaks in Liquid Metal Fast Breeder Reactor steam generators. Methods were developed to fabricate leaks in the size range of 10^{-10} to 10^{-5} lb/sec H_2O in $2\frac{1}{4}Cr-1Mo$ steam generator tubing of prototypic wall thickness (~ 0.110 inch). A test system was modified to supply flowing superheated steam at 1525 psig to as many as three specimens immersed in flowing sodium.

An initial test in static sodium and static steam showed that a 2×10^{-5} lb/sec steam leak self-plugged on startup. After 100 hours exposure, the specimen showed no self-damage on the sodium side of the tube but considerable self-damage around the leak site on the steam side.

The first test in the flowing sodium/steam system included a specimen large enough ($\sim 4 \times 10^{-5}$ lb/sec) to stay open during exposure. After 1.75 hours, the leak rate suddenly increased about three orders of magnitude. The sudden increase was found to be the result of rapid oxidation of the steel surrounding the leak caused by the sodium/water reaction products. Similar observations have been made in France.

Small leaks that self-plug have been observed to reopen and grow during extended exposure to sodium and steam. Even the smallest leak tested ($\sim 10^{-10}$ lb/sec) showed self-damage. It was concluded that any through-wall defect that allows interaction of sodium and steam will eventually open and grow. The self-damage process appears to enlarge the leak path eventually causing the leak to unplug.

Preliminary evaluation of nondestructive examination methods to locate small leaks indicate that ultrasonic and eddy current techniques are promising. The reason these methods may prove successful is that self-damage of a plugged leak appears to enlarge the defect near the steam side of the tube wall. Further development of these techniques is required.

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Small Leak Behavior
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D. W. Sandusky

1. Introduction

Steam generators often have small leaks in the metal boundary between steam and the heat source. These leaks arise from fabrication defects, accelerated corrosion, stress corrosion cracking, fatigue, etc. In fossil-fired power stations and light water reactors, these leaks represent a problem of low to moderate concern from the standpoint of cross contamination between the two different systems (steam and heat source). However, for sodium-heated steam generators, cross contamination is one of the major design/materials/operation problems for Liquid Metal Fast Breeder Reactors (LMFBR's).

Through-wall defects in the steam generator tubing or tube-to-tubesheet welds will allow the injection of high pressure steam into sodium. If the leak is very large ($> 10^{-4}$ lb/sec H_2O), it has been observed that it can form a jet which may rapidly penetrate adjacent tubes.^(1,2) For leaks $< 10^{-4}$ lb/sec damage to adjacent tubes does not occur, but there will be self-wastage of the leaking tube. There have been reports that this self-wastage will continue for a period of time until the tube wall is penetrated resulting in a rapid increase in leak size.^(3,4) This phenomenon is made more serious by the fact that small leaks ($< 10^{-5}$ lb/sec H_2O) are not likely to be detected after steam generator fabrication. Neither are they likely to be detected during operation of an LMFBR plant until the tube fails.

Since there was little information on the tendency of leaking $2\frac{1}{2}Cr-1Mo$ steam tubes to self-damage in liquid sodium, a study of the phenomenon was initiated. The objective of the first phase of the study was to identify methods to fabricate and test steam tubes with small leaks in sodium and make initial observations of small leak behavior. This report summarizes the results of the scoping phase of small leak testing.

2. Conclusions

The following conclusions have been drawn from the observations of small leak behavior during the scoping tests:

- 2.1 Small leaks that are open during operation of the steam generator will suffer self-damage and grow resulting in a sudden increase in leak rate. The increase will be at least three orders of magnitude. The damage mechanism is a rapid attack of the $2\frac{1}{2}\text{Cr-1Mo}$ by NaOH formed by reaction of sodium and steam/water. Failure time is dependent on initial leak size and leak geometry.
- 2.2 Leaks smaller than about 2×10^{-5} lb/sec H_2O will generally self-plug with sodium/water reaction products. After an indefinite period of exposure, plugged leaks may reopen to become larger leaks. There appears to be a self-damage process which enlarges the leak path and dislodges the plug.
- 2.3 Even the smallest leak tested (10^{-10} lb/sec) appeared to suffer some self-damage within 500 hours. It is concluded that essentially no leak is too small to be of concern.
- 2.4 Thermal cycling and pressure cycling were not found to reopen self-plugged leaks.
- 2.5 Some nondestructive examination techniques, particularly ultrasonic and eddy current, show promise in locating self-plugged leaks. There is potential in these techniques because of enlargement of the leak path near the steam side of the tube wall caused by rapid oxidation of the steel.

3. Experimental

3.1 Test System

The principal system used for the scoping tests was Loop 9 located at GE, San Jose, California. Loop 9 was originally used to study high temperature sodium corrosion of LMFBR fuel cladding materials. For the small leak scoping tests, the loop was modified considerably to allow insertion of steam generator tubing specimens which would contain flowing steam or water. The modifications included installation of a test pot in a bypass line, construction of a high pressure water loop, and addition of a high capacity cold trap to remove sodium/water reaction products. A simplified schematic of the test system is shown in Figure 1. The capacity of the main loop and

cold trap combined is approximately 65 gallons of sodium. During testing, the full sodium flow (5 gal/min.) is routed through the cold trap. The leaks are monitored by two diffusion tube hydrogen meters. Since all the sodium is purified on each circuit of the loop, leak rate may be determined directly from the deviation of the meters from the background hydrogen level. The test pot is capable of exposing three tubular specimens simultaneously to flowing sodium and steam. The steam pressure is generated by a mechanical pump and controlled by back-pressure regulators. The capabilities of the test system are summarized in Table 1.

TABLE 1

Loop 9 Small Leak Test Rig

Maximum Sodium Temperature	1000°F
Sodium Flow	5 gpm
Sodium Inventory	
a. Loop	15 gal
b. Cold Trap	50 gal
Maximum Steam Pressure	1800 psig
Maximum Number of Specimens	3
Leak Detection	H ₂ Meter (2)

A static sodium pot was used to expose one specimen while the system described above was under construction. This pot contained sodium stirred by natural convection. Steam was supplied to the specimen by static pressurization of a water reservoir by helium.

3.2 Leak Detection

The control of leak behavior studies is enhanced by being able to determine whether an individual leak is open or plugged. For a flowing steam system, the leak cannot be monitored on the steam side. An attempt was made to detect an individual leak by placing small diameter thermocouples in close proximity to the defect. In all cases the thermocouples were unable to detect a temperature rise near an open leak unless the leak was greater than $\sim 10^{-4}$ lb/sec H₂O. This observation agrees with calculations using heat of

reaction and sodium thermal conductivity, heat capacity and flow rate. The calculated temperature rise for a 10^{-5} lb/sec leak is at best a few degrees Fahrenheit. This is within the temperature fluctuations of the flowing sodium system.

Diffusion tube hydrogen meters were installed in the system as a back-up to the thermocouple technique. The meters were of the usual nickel membrane-ion pump variety with the addition of an ionization gauge. The presence of the ionization gauge allowed the meter to be calibrated using the saturation solubility of hydrogen in the cold trap. The vacuum pump was shut off, thereby allowing the hydrogen in the tube to equilibrate with the hydrogen level in the sodium. By performing this sequence for several cold trap temperatures, the hydrogen level in the tube (while the ion pump is operating) may be related to the hydrogen concentration in the sodium. With the calibrated hydrogen meter, a deviation from background hydrogen can be related to a specific leak rate. For Loop 9 with 5 gpm sodium flow at 920°F , a leak rate of 10^{-7} lb/sec H_2O is easily detectable. Unfortunately there is no way to distinguish individual specimens when more than one is exposed. Additional information on the hydrogen meter calibration is given in Appendix A.

3.3 Leak Specimens

Specimens for the scoping tests were tubing made of $2\frac{1}{2}\text{Cr}-1\text{Mo}$ according to ASME SA 213-T22. Several methods were attempted to drill small holes in 0.100 inch thick tubing. This included mechanical drilling, electrodischarge machining (EDM), and laser. The EDM technique presently appears to be limited to a minimum of 0.005 to 0.006 inch diameter. The laser technique produced a conical hole as small as 0.002 inch at the inner tube wall, but 0.020 inch on the O.D. Mechanical drilling consistently produced holes of constant cross section, 0.003 to 0.004 inch in diameter. For the scoping tests it was decided that mechanical drilling produced the most useful specimens.

The flow of superheated steam through a 0.003 to 0.004 inch diameter hole was approximately one order of magnitude greater than required for the small leak tests ($\sim 10^{-4}$ lb/sec H_2O). To reduce the flow rate to size range of interest (10^{-10} to 10^{-5} lb/sec), tubes containing drilled holes were swaged. By swaging through successively smaller dies, the leak rate could be reduced to a selected size range.

It was found that the leak rate of specimens produced by drilling and swaging was unstable when exposed to 1500 psi argon at 900⁰F. That is, increases in leak rate with time were observed. For this reason the specimens were heat treated at about 1700⁰F. This not only served to stabilize the leak rate, but also removed the cold worked structure caused by swaging.

The end product of the drilling/swaging process was a through-wall defect of slit-like cross section. A typical leak exposed to steam only is shown in Figure 2a and a cross section of the leak path in Figure 2b.

In addition to the drilling/swaging technique described above, an attempt was made to fabricate small leaks by making defective welds. Although this technique was marginally successful for leak paths less than 0.050 inch long, no leaks were produced in tubes of prototypic wall thickness (~0.110 inch). The methods employed in this effort are described in Appendix B.

3.4 Leak Calibration

Since most of the leak specimens were tested in the flowing steam environment of Loop 9, it was not possible to measure the leak rate through an individual leak while the test was in progress. This required that the leak rate of the specimens be measured before the test. The most convenient method to measure leak rate was to measure the flow rate of room temperature argon through the leak. This required that a relation be developed between the flow rate of argon at room temperature and superheated steam at 900⁰F (both at 1525 psig).

To establish a calibration curve specimens were inserted in a small furnace. A tube was first pressurized (1525 psig) with room temperature argon to measure the flow rate downstream of the leak. Then the specimen was heated to 900⁰F and the argon flow rate again measured. The rate was lower because of increased gas viscosity at 900⁰F. Steam flow was then initiated to the specimen. Flow through the leak was determined by condensing the steam and measuring the amount of effluent water with time.

Comparison of the argon flow rate to steam for a number of specimens showed that a linear relation existed (Figure 3). This fortunate occurrence allowed simple room temperature argon calibration of subsequent test specimens by reference to the curve.

A significant observation was made during the development of the calibration curve. Small leaks less than 1×10^{-5} lb/sec H_2O tended to plug within one hour of initiation of steam exposure. Metallographic examination of the specimens showed the plugging to be caused by a buildup of Fe_3O_4 (normally formed in a steam atmosphere). This demonstrated that small leaks could easily be plugged during heat treating, unless an oxygen-free atmosphere is maintained.

4. Experimental Results

4.1 Static Sodium Test

While the flowing Na/Steam system was under construction, one specimen was tested in a static sodium pot with a high pressure steam supply system. (High temperature steam is generated as the water goes through small diameter tubing heated by the pot sodium. Pressure is set by inert gas pressure above the water supply, while the leak rate is monitored by following the water level in a sight glass.)

The specimen was placed in $950^{\circ}F$ sodium with an inert gas purge through the leak. Immediately before the test was initiated, it was confirmed that the leak was still open by shutting off the purge gas supply and observing a pressure decay. When water was injected into the leak, there was no indication of a temperature rise on the thermocouples adjacent to the leak. Neither was there any indication of flow through the leak from the water level in the sight glass. The leak was left in the sodium with high pressure steam in the tube for 100 hours. At the end of this time the pressure was raised to 1800 psig in an attempt to reopen the leak, but this was unsuccessful. The specimen was then removed and water cleaned for subsequent metallographic examination.

Visual examination of the outside of the tube showed only a buildup of material at the leak site. After visual examination, the tube was sectioned around the leak site to allow examination by scanning electron microscopy (SEM). Close examination of the outside surface of the specimen, shown in Figure 4a, did not give any evidence of accelerated corrosion around the leak. The buildup appears to be material extruded from the leak orifice. However, the inside of the tube (the steam side) was damaged for a considerable distance

from the leak center (Figure 4b). There appears to be a loss of material over an area 0.09 inch long by 0.06 inch wide. There also appears to be a mound of reaction products built up immediately around the leak which is higher than the surrounding base metal. No evidence of similar damage has been observed in leaks exposed to steam only.

A microprobe analysis of the leak site on the steam side showed only iron and chromium oxides. The only evidence of sodium was in the leak itself.

Optical metallography of the leak in a transverse section to the tube wall showed that the leak path was plugged with oxide reaction products. There was a loss of material in the shape of a shallow cone centered around the leak. The maximum loss was to a depth of approximately 0.010 inch in the base metal. The material appeared to have been removed by a general oxidation process.

4.2 Scoping Test Results

During the scoping phase of the leak behavior tests, a total of twelve specimens were exposed in the Loop 9 test rig. A summary of the leak sizes and exposure history is given in Table 2. A description of individual test runs follows:

4.2.1 First Scoping Test

Three specimens listed in Table 2 were loaded into the test rig and the system brought to test conditions (920⁰F sodium, 1525 psig steam). Argon purge flow indicated that the leaks were still open. When the steam in the economizer was at the test temperature, steam flow was initiated through the leaks. There was no initial indication of temperature increase at the leak sites from thermocouples attached to the specimens. However, within a minute of initiation of the test, the ion pump in the hydrogen meter went from 270 μ amp to 700 μ amp. This indicated that steam contamination was reaching the sodium. The hydrogen meter reading increased to 1000 μ amp and then began to decrease after about one hour. Venting of the test pot occurred twice during the first 20 minutes indicating a buildup of hydrogen pressure. The plugging indicator, which indicated clean sodium prior to test initiation, showed an increase in plugging temperature after about 5 minutes. In general all initial indications were that at least one leak was open with the hydrogen meter being the first and most positive indication.

TABLE 2

Summary of Small Leak Scoping Tests

Sodium Temperature: 920°F
Steam Pressure: 1525 psig

<u>Specimen Number</u>	<u>Run No.</u>	<u>Pretest * Leak Rate (lb/sec)</u>	<u>Exposure Time (hr)</u>	<u>Comments</u>
26	1	1.3×10^{-5}	1.75	Plugged at startup
27	1	1.0×10^{-5}	1.75	Plugged at startup
30	1	4.0×10^{-5}	1.75	Rapid increase in leak rate after 1.75 hours
21	2	1×10^{-6}	100	Plugged on startup; Na on steam side after 100 hrs.
23	2	8×10^{-7}	100	Plugged on startup
28	2	2.5×10^{-6}	100	Plugged before startup; Na on steam side after 100 hrs.
34	3	3×10^{-6}	81.3	Plugged on startup; opened at 75 hrs.; large leak at 81.3 hrs.
37	3	5×10^{-6}	81.3	Plugged on startup
38	3	7×10^{-6}	800	Plugged on startup; open < 1 hr. at 150 hrs; replugged to 800 hrs.
6	4	2×10^{-10}	535	Plugged on startup
22	4	2×10^{-7}	535	Plugged on startup; intermittently open during 500 hrs.
25	4	9×10^{-11}	535	Plugged on startup; inter- mittently open during 500 hrs.

* Calibrated with room temperature argon

After approximately 1.75 hours of steam flow, automatic venting of the test pot occurred indicating a pressure increase. It was observed that the temperature at the leak site of Specimen 30 was rising steadily toward the upper temperature set point (1100°F). Specimen 30 was isolated from the steam supply within 10-20 seconds after pressure venting was noted. This caused the temperature at the leak to start decreasing. Approximately 30 seconds after the first indication that the leak was opening, the sodium flow meter indicated a loss of main sodium flow causing both the sodium and water systems to be shut down in emergency mode.* The hydrogen meter went off scale on high level.

At the onset of the test it was anticipated that one or more of the leaks might enlarge; however, it was considered that this would occur over a period of minutes. In fact, the leak opened up in about 30 seconds.

After flow was reestablished in the loop, the sodium system was shut down normally and the sodium pot isolated. The specimens were then removed from the test pot and steam cleaned. The specimens and holders showed no evidence of excessive buildup of sodium/water reaction products. The assembly appeared to be similar to an assembly removed from clean sodium.

Even before the specimens were cleaned of residual sodium it was obvious that Specimen 30 had a large hole in the tube wall at the leak site. After cleaning, visual examination of the three specimens showed that Specimens 26 and 27 had not opened up while Specimen 30 contained a hole in the tube wall greater than 1/8 inch diameter at the outside of the wall. The opening appeared to taper inward to a significantly smaller opening on the inside of the tube wall.

After visual examination of the specimens, the sections of tubing containing the leaks were cut out by a dry process to preserve the corrosion products on the inside of the tube. A detailed examination showed there was a dark circular stain around the leak site on the inside of Specimens 26 and 27. This stain was not observed in specimens exposed to steam only. A test with pH paper indicated a strong basic reaction at the leak sites. The indicated pH range was 12.0 to 12.5.

*Subsequent examination showed that in fact the loss-of-flow indication was a large hydrogen bubble passing the flow meter.

The dimensions of the hole in Specimen 30 were determined optically. They are given below:

Outside Surface: 0.180 by 0.120 inch

Inside Surface: 0.045 by 0.62 inch

Examination by scanning electron microscope (SEM) showed that specimens 26 and 27 were in fact plugged with reaction products on the outside surface. A slight depression at the leak site indicated that some erosion had taken place before the leaks plugged. The depressions at the leaks were filled in with the plugging material. There was some evidence of accelerated corrosion in the immediate vicinity of the leak sites on the inside of the tubes. The corrosion was in the form of oxide build-up and blisters which were not visible on the base metal more than 0.010 inch from the leak.

Examination of Specimen 30 was hampered by the fact that the hole in the tube wall was larger than the field-of-view in the SEM at lowest magnification ($\sim 35\times$). Figure 5a shows the hole from the outside of the tube wall. The walls are initially steep, but then begin to taper near the inside surface of the tube wall. The spongy oxide visible at the bottom of the hole was formed after the test by NaOH-accelerated air corrosion before the specimen was cut open to remove residual sodium. Viewed from the inside wall (Figure 5b), it is apparent that there was no erosion at the surface--as evidenced by the sharp edge of the hole.

Optical metallography of Specimen 30, Figure 6, shows a cross section of the leak in perspective with the tube wall thickness. As previously noted the hole is funnel-shaped expanding from the inside to the outside. Examination of one of the self-plugged leaks (Specimen 27) in full cross section showed that the leak had been open for some period of time, as evidenced by the eroded pit at the outside surface (Figure 7). The development of the pit was apparently arrested when the leak self-plugged. There was no evidence of loss of material on the inside wall around the leak site.

4.2.2 Second Scoping Test

The leaks used for the second scoping test were approximately one order of magnitude smaller ($\sim 10^{-6}$ lb/sec steam) than the first test. One of the specimens (No. 28) was inadvertently plugged during fabrication. It was

decided to test the specimen to see if exposure to sodium and steam would cause it to open.

The second scoping test was conducted using a low pressure argon purge in the steam system while the test was not in progress. The pressure drop from the steam side to the sodium side was approximately 70 psig before introduction of 1525 psig steam. This test was terminated after 100 hours exposure because of a leaking steam fitting external to the sodium system. After shutdown, it was noted that the steam supply lines to two of the leaks were plugged. The cause of the plugging was metallic sodium in the steam inlet lines.

The observations made during this test contained two significant features. The first was that the low pressure argon backpurge was insufficient to keep sodium from entering the steam side of the tubes by capillary action. The second significant observation was that one of the leak specimens with sodium in the steam lines was Specimen 28. This leak was plugged during fabrication such that the leak could not be detected with a standard helium leak check. Even so, after 100 hours sodium entered the steam side of the tube.

4.2.3 Third Scoping Test

The third scoping test was run with a 1000 psig inert gas purge in the steam system at all times during startup and shutdown, i.e. inert gas was present if steam pressure was less than 1000 psig. It was postulated that the high pressure gas would prevent sodium from reaching the steam lines in quantities sufficient to plug the lines. The method proved successful for this and all subsequent tests. Some traces of NaOH are still routinely observed on the steam side, but no intrusions of bulk sodium have occurred.

The leak rate of the specimens for this test were in the range of 3 to 7×10^{-6} lb/sec H_2O . When steam flow was initiated, all the leaks self-plugged. This was confirmed by shutting off the large cold trap so no hydrogen was being removed from the system. The hydrogen meters showed no appreciable build-up of impurities.

After 75 hours of exposure, one of the leaks reopened. The leak rate was at a relatively low level ($\sim 10^{-7}$ lb/sec) initially. The leak rate increased to about 5×10^{-7} lb/sec, decreased somewhat, then rapidly increased off the scale of the recorder. The resultant increase in hydrogen generation

caused the system to automatically shut down. This sequence of events is shown in Figure 8. The three specimens were removed from the system. Visual examination showed that Specimen 34 had opened. It was removed for metallographic examination along with Specimen 37 for comparison. Specimen 38 was reinserted for further exposure. After 150 total hours, Specimen 38 opened at a very low leak rate ($< 10^{-7}$ lb/sec H_2O). It remained open for about 45 minutes, then replugged. The exposure continued for a total of 800 hours with no further activity.

Examination of Specimen 34 by SEM showed an enlarged leak path approximately 0.015 inch long by 0.005 inch wide. The estimated flow rate through a hole of those dimensions is about 10^{-3} lb/sec H_2O . Examination of Specimen 37 showed a deposit of crystalline NaOH around the leak site on the steam side. Optical metallography of the two specimens gave an interesting comparison of the leak paths (Figure 9). Specimen 34 shows a relatively constant cross section across the tube wall. The leak which remained plugged (Specimen 37) shows instead a relatively narrow leak path near the sodium side. However, near the steam side the leak path widens considerably. This tends to indicate a damage process which results in metal loss near the steam side of the tube. The cross section of the leak path is at least twice the original dimension.

Specimen 38, which was exposed 800 hours, was removed from the system and tested in the laboratory to determine if thermal cycling or pressure cycling would reopen the leak. For the thermal cycling, the specimen temperature was increased from room temperature to 900°F several times. The objective was to determine if differential thermal expansion between the plugging material and the base metal would crack the plug. During the thermal cycling, the specimen was connected to a helium mass spectrometer; if the plug cracked, it would be detected instantly. Neither slow nor rapid cycles produced any indication of a leak.

Similarly, pressure cycling was performed at 900°F in an attempt to dislodge the plug. Pressure cycling was not considered very likely to be successful because of the large shear stresses required to dislodge a firm plug. After cycling repeatedly from 0 to 1600 psig, there was no indication of a leak.

Following thermal and pressure cycling, Specimen 38 was examined by several nondestructive techniques. These were X-radiography, neutron radiography, ultrasonic and pulsed eddy current. The leak was readily visible in X-radiographs. The leak path appeared to be relatively wide near the steam side and sodium side of the tube wall and to neck down to a narrow path in the center. The leak was only slightly visible in a neutron radiograph. The presence of the defect was easily detectable by ultrasonic and pulsed eddy current techniques. The latter two observations were made using external probes. It remains to be determined if such defects are detectable from the internal bore of a tube.

Metallographic examination of Specimen 38 confirmed the observations of the leak path noted on the X-rays. There was a shallow depression caused by metal loss on the sodium side immediately surrounding the leak site (30 mils diameter, 3 mils deep). The leak path was about 2 mils wide near the sodium side narrowing to less than 1 mil near the midwall. The leak path then expanded in the form of an oxide plug to 4.5 mils near the steam side. At the I.D. surface the oxide plug spread out to about 40 mils diameter.

4.2.4 Fourth Scoping Test

The objectives of the Fourth Scoping Test were to (1) provide additional specimens for nondestructive examination development and (2) determine if very small leaks (10^{-7} to 10^{-10} lb/sec H_2O) were susceptible to unplugging and self-damage.

All of the leaks self-plugged when steam flow was initiated. After 29.5 hours exposure, there was a very brief, low level hydrogen excursion in the sodium. Again at 395 hours a hydrogen increase was detected in the sodium for several minutes. In both incidences the hydrogen level in the sodium returned to the background level indicating that the leak had replugged. The test was terminated after 535 hours total exposure.

A visual examination of the outside surfaces of the specimens showed that some metal was removed around the leak sites of Specimens 22 and 25. This indicated that each of these specimens had been open at some point in the exposure. Both specimens were thoroughly plugged after termination of the test. Examination of Specimens 22 and 25 has been deferred until several nondestructive examination techniques have been evaluated. Following the successful identification of the plugged leak in Specimen 38 by

ultrasonic and pulsed eddy current by examination from the outside (sodium side) of the tube wall, Specimens 22 and 25 will be examined from the inside wall. This requires some alteration in technique and probe design. In addition, the feasibility of leak location by Kryton-85 will be evaluated.

Specimen 6 was examined metallographically. A cross section of the leak showed that some oxidation and enlargement of the leak path occurred near the steam side (Figure 10). This shows that essentially any through-wall defect, no matter how small, is susceptible to self-damage. This is any small leak may eventually open.

5. Discussion

Observations made during the scoping tests of small leak self-damage fell into two categories: small leaks that are open and small leaks that self-plug. These two classes of leak self-damage are apparently related to somewhat different mechanisms and are therefore discussed separately.

5.1 Self-Damage of an Open Leak

The self-damage process of an open leak appears to be readily explainable in terms of accelerated corrosion. Several observations have been made in France of small leaks which grow to become large leaks.^(3,5) The general observation has been that an open leak will remain relatively constant for a period of time, then rapidly increase. These observations plus two open leaks observed in this work are shown in Figure 11 as a function of leak rate and time. As may be seen, the French results and GE results are comparable. The time to sudden enlargement appears to be related to the initial leak rate. A plot of log leak rate versus log time to sudden enlargement is in fact found to be linear (see Figure 12). There is some scatter in the data which may be attributed to variations between the different experiments in tube wall thickness, steam pressure, and leak geometry.

Although the data are still sparse, they suggest that geometry is a significant variable. In Figure 12 the French results along the bottom line were all leaks produced by fatigue cracking. The one French data point along the upper line was a leak resulting from a bad weld that would be likely to be of pin-hole geometry. The observations of Greene and this work resulted from leaks produced by drilling and swaging. This results in elliptic holes rather than linear cracks produced by fatigue. The available data then fall

into two categories: linear cracks and pin-holes/elliptic holes. The linear cracks appear to reach the stage of sudden enlargement sooner.

The French results do not report the leak rate after sudden enlargement. The tests performed for this work were terminated as soon as the leak rate rapidly increased to protect the test equipment. The final leak size then remains an open question. Examination of Specimens 30 and 34 after testing indicated that the leak rates at the termination of the test were approximately three orders of magnitude greater than the initial leak rates. The method of calculating the final leak rate for Specimen 30 is illustrated in Appendix C.

The cause of leak enlargement in $2\frac{1}{2}\text{Cr-1Mo}$ can be explained in terms of hot caustic corrosion of the metal surrounding the defect. The sodium-water reaction at the steam exit of the leak produces local high caustic concentrations which, at steam generator temperatures ($\sim 900^\circ\text{F}$), rapidly attack the steel.* Newman et. al.^(7,8) have done a considerable study of this phenomenon by impinging a sodium vapor jet on a steel surface in a water vapor atmosphere. They concluded that the wastage observed could be accounted for by hydroxide corrosion plus the action of the jet removing reaction products and loose grains of base metal. An analogous situation probably occurs during self-wastage of a leaking tube.

The phenomenon of a sudden rapid increase in leak rate is explained schematically in Figure 13. This representation is inferred from metallographic examination of specimens in this test program. The leak removes metal near the sodium side of the tube in the form of a roughly circular pit. Once formed, the pit advances through the tube wall at a relatively constant rate. Since the high-pressure steam is in choked flow, the leak rate does not change appreciably until the pit contacts the inner tube wall. At this point, only a thin edge remains. This is rapidly removed. In a short period of time then, the leak rate increases from that governed by the cross section of the original defect to that governed by the diameter of the pit.

5.2 Self-Damage of a Plugged Leak

The observation that a self-plugged leak reopens to become a large leak unfortunately is not as readily understandable as self-wastage of an open

*The rapid sodium/water reaction also probably produces local heating which further enhances the corrosion rate.

leak. At this point in the test program it is not possible to positively identify a leak opening process. However, some observations can be noted which give clues to possible processes.

The general observation both in this test program and that of Donati⁽⁵⁾ is that leaks smaller than about 10^{-5} lb/sec plug and reopen intermittently. While the leak is open, the self-damage mechanism described in Section 5.1 causes metal loss. The question which remains is, what causes a leak to reopen once it plugs.

There are two mechanisms which could explain the observation that small leaks plug and reopen. The plugging mechanism simply involves the buildup of solid reaction products produced by local high supersaturations in the leak path. Buildups of material ejected from the leak have been observed immediately around the leak site. The actual plugging material has not been completely identified. Of course the reaction of steam with high temperature sodium will produce Na_2O , NaOH , and NaH . Steam will react with the steel to form iron and chromium oxides. Newman⁽⁷⁾ suggests that the reaction on a $2\frac{1}{4}\text{Cr-1Mo}$ surface also produces NaFeO_2 and NaCrO_2 , (double oxides probably insoluble in sodium). It is expected that any or all of the above species may be present in the plug.

Once the leak is plugged, it is possible that the plug is removed by simple dissolution of the sodium oxide by the sodium. When the plug is dissolved, steam flow will restart thereby producing more reaction products. There are some reasons why this process acting alone may not be able to account for the observation of leaks opening in less than 100 hours. A plugged leak has a very long path length compared to the surface area of the plug exposed for dissolution. As dissolution progresses, the sodium near the plug will become saturated with oxide. The rate of dissolution of the plug will then be controlled by diffusion of the oxide from the sodium/plug interface to the bulk sodium. Experimentally, plugged leaks exposed to sodium with high pressure argon on the steam side failed to open during 100 hour exposures. Metallographic examination of the specimens after exposure showed very little penetration of the plug by the sodium.

Another possible self-damage mechanism would involve sodium diffusion through the plug. The sodium would then react with steam forming concentrated NaOH that would attack the steel. By this mechanism the reaction-product

plug would be undermined by the corrosion process and eventually dislodge. Rubright⁽⁹⁾ reports that high concentrations of aqueous caustic can result in a corrosion rate of 0.3 mil/hr at 900°F. Such accelerated corrosion may account for leaks opening within 100 hours.

There is some experimental evidence that sodium transport does occur through a plugged leak. Examination of self-plugged leaks has shown sodium hydroxide surrounding the leak site on the steam side in all cases. Sodium was identified both by electron microprobe and X-ray analysis in a scanning electron microscope. Measurements with pH paper in the vicinity of the leak has shown a pH greater than 12.0. Examination of self-plugged leaks by scanning electron microscope and optical metallography has shown in most cases enlargement of the leak path near the steam side of the tube wall. The area immediately surrounding the leak on the steam side is also subject to excessive corrosion. The most outstanding example of enlargement of the leak path was shown in Figure 10. The diameter of the defect was greater than the original diameter of the drilled hole.* Even a leak which was continuously plugged for 650 hours showed evidence of enlargement of the leak path by oxidation of the base metal.

One other piece of evidence suggests sodium transport through a plugged leak. Rubright⁽⁹⁾ reported that sodium hydroxide in excess sodium resulted in intergranular attack of 2½Cr-1Mo. Close examination of an oxide plug showed some intergranular attack of the base metal near the tip of the plug closest to the sodium side. This would be the most likely region to find NaOH in excess sodium. Closer to the steam side there would be excess H₂O and therefore excessive general corrosion.

One may conclude from the observations that any defect that allows sodium to come in contact with water can suffer self-damage and grow. Therefore susceptibility to self-damage is only a function of temperature and time, not leak size.

5.3 Small Leak Location and Shutdown

In the course of performing the scoping tests on small leak behavior, some qualitative observations were made on leak shutdown and location.

It was found that when a low pressure inert gas purge was used during shutdown of the system, metallic sodium intruded on the steam side of the system. Capillary flow by sodium at 900°F is very likely unless the hole is extremely small. This occurs because sodium is very effective at wetting

*The tube was subsequently swaged thereby reducing the cross section.

the steel surface. It is quite probable that a 70 psi pressure differential was insufficient to overcome the capillary action of the sodium. Although the capillary force cannot be interpreted directly in terms of an opposing pressure, it was postulated that a high purge pressure would keep the bulk of the sodium out of the steam side of the tube. High gas pressure on the steam side would result in gas flowing at sonic velocity through the leak. In practice it was found that use of 1000 psig purge pressure kept bulk sodium from entering the steam side. This observation has proved useful for the conduct of the succeeding tests, but further work is required before this method could be recommended for large systems.

Methods attempted during the scoping tests to reopen plugged leaks proved ineffective. It had been suggested that thermal cycling or pressure cycling may dislodge or crack the plug so that the leak could be located by helium leak checking. Neither of these methods was successful. Examination of the variables and materials involved show that this is the expected result. Differential thermal expansion between the base metal and the oxide for a small slit-like leak 0.2 mil thick, is only about 1 micro-inch (0.6%) for $1000^{\circ}\text{F } \Delta T$. If the wall of the leak was perfectly uniform, this may in fact dislodge the plug. Metallographic analysis of actual specimens have shown that the leaks generally have a convoluted path which varies much more than 0.6% of the thickness.

A simple calculation can also show that internal pressurization will not dislodge a plug. Assume that the maximum shear stress between the oxide and steel is 100 psi. Then for a 0.001 inch diameter plug the force required to remove the plug is 3×10^{-2} lb. This would require an internal pressure in the tube of about 40,000 psi. Clearly using pressurization to force out the plug will not work within the operating limits of the system.

From the preliminary observations made during the scoping tests, it appears that there are at least two nondestructive examination techniques which may be applicable to in-service inspection. It was shown that a self-plugged leak was undetectable by helium leak checking. X-ray examination gave a strong indication of the defect including the oxide plug. However, use of this technique for leak location in-service is doubtful because film would have to be placed on the sodium side of the tube. This region will not be accessible in the current design. Neutron radiography was not promising both from the standpoints of leak detection and applicability in service. By

far the most promising techniques for internal bore inspection of tubes are ultrasonic and eddy current. Both techniques easily detected a leak that had been exposed to sodium and steam. The inspection was made from the outside of the tube. Further development will be necessary to allow consistent results from internal bore inspection.

The reason there is considerable hope for development of ultrasonic and eddy current techniques is related to the self-damage phenomenon. It is presumed that only leaks too small to be detected will be present when the steam generator is put in service. It has been shown in the scoping tests that such small leaks suffer self-damage and grow. Therefore, even if a leak does not open (or remain open), it will be large enough to be detectable.

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Figure 1 LOOP 9 - SMALL LEAK TEST RIG

FIGURE 2. Slit-like Leak Produced by Swaging

a. Outside Wall

b. Cross Section - Midwall of Tube

FIGURE 3
RELATION OF ARGON FLOW RATE TO
STEAM LEAK RATE

FIGURE 4. Leak Exposed to Static Sodium (950⁰F) and Steam (1600⁰F)

a. Sodium Side

b. Steam Side - Note
Accelerated Corrosion

FIGURE 5. Leak Specimen Self-Wastage
(Initial Leak Rate: 4×10^{-5} lb/sec H_2O)

a. Sodium Side

Na Temp 920°F
Steam Pressure 1525 psig

b. Steam Side

Sodium Side

FIGURE 6. Cross Section of the Self-Damaged
Leak in Specimen 30

Sodium Side

FIGURE 7. Cross Section of Self-Plugged Leak No. 27

FIGURE 8 EXPOSURE HISTORY - SPECIMEN No. 34

FIGURE 9 Comparison of an open and a plugged leak
exposed 81.5 hours, 920°F Sodium, 1525 psig Steam

Open Leak

Self-Plugged Leak
(Note Enlargement
Near the Steam Side)

FIGURE 10. Small Leak Specimen No.6
Initial Leak Rate, 2×10^{-10} lb/sec H_2O
Exposed 535 Hours

FIGURE 11

FIGURE 12 RELATION OF SELF-ENLARGEMENT OF SMALL LEAKS TO EXPOSURE TIME

Figure 13

Development of a Large Leak From a Small Steam Leak in 2 1/4Cr-1Mo Tubing Exposed to Sodium

Appendix A

Hydrogen Meter Calibration

Sodium hydride dissociation pressures have been studied and discussed extensively in connection with the hydrogen meter for LMFBR's. A compilation of the data on this phenomenon is given by Rodgers and Dutina.⁽¹⁾ In the case of Loop 9, there is little question that the cold trap controls background hydrogen level because the volume of sodium in the cold trap is about five times that in the loop. Full loop flow is purified on each circuit. The cold trap temperature may then be related directly to hydrogen concentration (partial pressure) in the sodium. The equivalent hydrogen pressure is given by the NaH dissociation pressure for the temperature of the cold trap.

Equilibration of the hydrogen meters with hydrogen in the sodium was carried out for cold trap temperatures of 225, 250, 275 and 300°F. In addition, dynamic (ion pumped mode) pressure readings were obtained for cold trap temperatures of 350 and 400°F for comparison to the literature data. The relation of dynamic pressure to equilibrium pressure is shown in Figure 1, (this of course applies only to the particular meter in question). The associated temperature may be obtained from:

$$1. \log_{10} P_{(\text{torr})} = 11.66 - 6160/T(^{\circ}\text{K}) \text{ (Ref. 1)}$$

The temperature inferred from the equilibrium pressure may in turn be related to hydrogen concentration in the sodium by:

$$2. \log_{10} C_{\text{ppm}} = 6.067 - 2800/T(^{\circ}\text{K}) \text{ (Ref. 1)}$$

Then if the dynamic mode hydrogen pressure is 2×10^{-7} torr, this relates to an equilibrium pressure of 3.5×10^{-4} torr. From Equation 1 this gives a saturation of 274°F which may then be used in Equation 2 to give a hydrogen concentration of 100 ppb.

The saturation pressures determined in this test were also compared to the literature compilation given in Reference 1. Figure 2 shows the selected literature data and the observations made in Loop 9. It may be seen that, especially at the lowest cold trap temperatures, the observations are somewhat higher than predicted by the correlation. This may be attributed to several difficulties associated with such low hydrogen concentrations. In particular

vacuum system purity and cold trap temperature control can significantly affect the observations. It is concluded absolute values of NaH dissociation pressures measured by this method below 300⁰F (150⁰C) must be viewed with caution.

Reference:

A-1 D. N. Rodgers and D. Dutina, NEDM-12473, July 1974.

FIGURE A1 HYDROGEN METER CALIBRATION

FIGURE A2 DISSOCIATION PRESSURE OF NaH IN EQUILIBRIUM WITH Na(l)

Appendix B

Small Leak - Defected Welds

G. R. Dodson

Methods were sought, without success, to produce defective welds with through-wall defects. Small leaks generated by such defects would simulate the type of leaks that might appear in the welded areas of the steam generator.

Holes were drilled in .165" wall 1.5" diameter 2½Cr-1Mo tubing and closed by TIG welding. Weld shrinkage and cooling stresses from self-quenching produced some small leak rate defects. However, the wall thickness near the leak was reduced to about 0.050 inch. This thickness did not adequately represent the steam generator weld thickness. The addition of weld metal during the TIG process increased the wall thickness, but no through wall defects were generated. Thermal cycling and thermal shocking failed to open the thickened TIG welds.

Arc welding with a moistened weld rod was also attempted, but the steam produced during welding did not generate a through-wall defect. Along the same lines, a tube was pressurized with argon and the hole filled with weld metal. This method produced a through-wall defect but it was too large for the small leak tests planned in this program. Contamination of the weld pool by addition of foreign substances such as carbon, lead, etc., was also attempted. However, the concentrations of contaminant necessary altered the weld chemistry well beyond any deviations expected in actual tube-to-tubsheet welds. Since corrosion appears to be a dominant phenomenon in small leak behavior, it was considered that use of defects caused by gross deviations in chemistry would be generally unproductive.

Appendix C

Flow Through Specimen 30-End of Life

The following calculation is patterned after a sample calculation for superheated steam in pipes given in a engineering handbook.⁽¹⁾

Parameters:	Temperature	900°F
	Pressure	1525 psig
	Tube wall (L)	0.110 inch
	Hole diameter *(D)	0.056 inch

$$K = f \frac{L}{D}$$

where: K = resistance coefficient
 f = friction factor

Assume the internal roughness ($\frac{\epsilon}{D}$) is $\frac{.0002}{.056}$

Then from a plot showing $\frac{\epsilon}{D}$ and f:

$$f = .030$$

also: $K_I = 0.5$ at the inlet
 $K_O = 1.0$ at the outlet

$$K_{\text{total}} = f \frac{L}{D} + K_I + K_O = 1.56$$

$$\text{Pressure drop} = \frac{\Delta p}{P} = \frac{1525 - 20}{1525} = .987$$

From a plot showing K and $\frac{\Delta p}{P}$, the maximum possible pressure drop for K - 1.56 is 55%. Therefore, the steam flow is limited by the sonic velocity.

$$\Delta p = .55 (1525) = 839 \text{ psig pressure drop}$$

$$P (\text{downstream}) = 1525 - 839 = 686 \text{ psig}$$

$$V_s = (k g 144 P_d \bar{V})^{\frac{1}{2}}$$

*From the equivalent cross sectional area.

Where: V_s = sonic velocity

k = 1.3 for superheated steam

g = acceleration of gravity (32.2 ft/sec^2)

P_d = downstream pressure

$\bar{V}$ = specific volume of steam @ 686 psig

$$V_s = (1.3 * 32.2 * 144 * 686 * 1.16)^{\frac{1}{2}}$$

$$V_s = 2190 \text{ ft/sec}$$

$$W = \frac{V_s d^2}{0.0509 \bar{V}} \quad \text{mass flow rate}$$

$$W = \frac{2190 (56 \times 10^{-3})^2}{0.0509 (1.16)} = \underline{\underline{3.25 \times 10^{-2} \text{ lb/sec}}}$$

References:

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Crane Co., Chicago, Tech. Paper #410, (1957)