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SODIUM
COMPONENTS
DEVELOPMENT
PROGRAM
Information Meeting

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Preface

The Sodium Components Development Program (SCDP) is a Commission-wide effort to develop within the industrial complex of the United States competence for manufacturing safe, reliable, and economical components for advanced sodium-cooled nuclear power plants and the supporting technology to achieve this objective.

One of the purposes of the SCDP is to coordinate and foster the exchange of information in the area of components for sodium-cooled reactors. This information meeting is one of the ways in which the exchange of information is accomplished.

In accordance with the theme of the meeting, "Materials for Sodium-Cooled Reactors," the first day was devoted to development work in the area of materials and material experience from operating reactors. The second day included papers on current SCDP-sponsored work which had been completed since the last SCDP meeting in Hollywood, California on April 14-15, 1964. Also included in the second day were papers from the United Kingdom, Euratom, and Interatom.

Grateful acknowledgement is made of the services of the session chairmen, C. A. Pursel, R. J. Gariboldi, and R. H. Jones; and of the panel chairmen, D. Gurinsky and A. Amorosi. A special thanks to S. F. Burton, G. C. Gower, A. G. Hosler, D. I. Marshall, and C. L. Povalish for their assistance.

JOHN J. PURCELL*
Program Chairman

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Welcome

M. E. Jackson
U. S. Atomic Energy Commission

It is a pleasure for me to welcome you on behalf of the Chicago Operations Office to this information meeting of the Sodium Components Development Program. The fine turnout for this meeting is evidence of the strong interest in sodium component development work. In this meeting emphasis will be placed on material considerations in sodium-cooled reactor design. I have always taken a personal interest in material development, so I intend to use this meeting to bring myself up to date on material development for sodium-cooled reactors.

There will be no formal lunch either day of the meeting. However, for those of you who wish to eat lunch here at the hotel, the Pompeian Room on the first floor will be open. There will be a formal dinner this evening in this room preceded by a half-hour cocktail session.

At this time I would like to introduce to you Ralph Jones of the Liquid Metal Cooled Reactors Branch in Washington.

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Introduction

R. H. Jones
U. S. Atomic Energy Commission

Gentlemen, it's nice to again see so many people interested in the Sodium Components Development Program. I have assurances from John Purcell, who has handled all the arrangements for the meeting, that we are going to have some fine presentations. In accordance with our greater emphasis on quality control, we should have a most enjoyable meeting, since we have now standardized the sizes of slides which are acceptable for these meetings - so let's look forward to everyone's slides being readable!

The theme of our meeting here in Chicago is "Materials for Sodium Service." Instead of commenting on the problems associated with materials development, I would like to mention a few of the things which should be ground rules for our R&D programs:

1. The experiments should be run in environments representative of large primary and secondary sodium systems. Eventually, the primary environment should include typical fission products concentrations.
2. The levels of contamination in the test environment should be based on accurate measurements.

Now, my reason for stressing the importance of the experimental sodium environment is that, no matter how much R&D work has been done, no matter how wonderful the results, this work is of little value until it can be translated into the language of the designer, fabricator, and operator. To assure that this translation can be accomplished, the environmental factors must be completely and accurately known.

Hence, the goal of your materials development program will ultimately take the form of usable engineering data that can be used as a foundation for:

Introduction (Continued)

1. Good design practice,
2. Good experience application,
3. Good fabrication procedures, and
4. Good operating practices.

These four requirements for successful sodium-cooled reactors must be fulfilled from our research and development activities. Each of our sodium projects has demonstrated our dependence on these four factors; as examples, I will cite several that you are well aware of:

1. The Hallam IHX tube failures from excessive vibration,
2. The EBR-II oscillator rod bearing failure,
3. The SCTI secondary pump motor burn-out, and
4. The Fermi steam generator tube failures.

In each case, one or more of the four requirements were overlooked. To avoid these oversights, you will soon become aware of greater emphasis on:

1. Design review,
2. Quality control and inspection,
3. Proof-testing, and
4. Operating procedures.

Now, just a few words on our fast reactor program and how materials will influence it. Our program objectives are to produce a fast breeder prototype reactor by 1973, having a bulk sodium outlet temperature of 1200 F and turbine throttle temperature of 1050 F. On the basis of present technology, it appears that fuel cladding hot spot conditions may require materials other than stainless steel. However, our optimization studies will determine the best overall plant economics for various temperatures and materials. These studies could result in our accepting a back-off temperature condition to avoid certain materials problems. If a lower temperature is selected, we want it to be for optimum economics - not lack of technology. Therefore, your work on materials in sodium during the next few years is of utmost importance to the fast reactor prototype design effort.

On behalf of Dr. Wensch, I wish to convey his thanks to you all for participating in this meeting and his hope that you will find it of benefit to you and to your respective companies.

MECHANICAL PROPERTIES OF MATERIALS IN SODIUM

By

R. C. Andrews and F. Tepper

MSA Research Corporation

Callery, Pennsylvania

The Sodium Components Group sponsored the Mechanical Properties Research Program at MSA Research Corporation for the purpose of determining the effect of sodium on the mechanical properties of austenitic and ferritic steels. The underlying basic philosophy of this program is to determine if the allowable stresses, as established by mechanical property tests in air and inert gases, are adequate for the design of long-time systems and equipment in sodium.

The tests being employed are conducted in flowing air, static helium and flowing sodium. All of the liquid metal studies are being conducted at MSAR in two isothermal dynamic sodium loops of 316 ss containment material. A major portion of the air and helium tests are conducted at the University of Michigan under the direction of Dr. James Freeman. The tests being employed to compare the effects of the different environments include creep, creep-rupture and fatigue, plus tensile and impact on pre-exposed and original specimens. Appropriate chemical and metallographic analyses are used in evaluation of the effect of the environments on the composition and microstructure of the structural alloys.

The test materials selected for this program are Type 316 stainless steel at 1200 F, as representative of the austenitic steels, and 2-1/4% chromium-1% molybdenum steel at 1100 F, as representative of the ferritic steels. Type 304 stainless steel is scheduled as the next material to be tested. In addition to "clean" sodium, the effects of contaminated sodium on the mechanical properties are also being compared. The contaminants currently being studied are oxygen and carbon.

The previous information meetings covered the mechanical property studies with 316 ss in air, helium and low oxygen sodium, and 2-1/4 Cr-1 Mo steel in air, helium and both high and low oxygen sodium. Since the last meeting, the program has been concerned with the mechanical properties of 316 ss at 1200 F in both carbon-rich and oxygen-rich sodium systems. To date the operation in the high carbon system alone has resulted in 27,000 hours of mechanical testing with the generation of over 600 test specimens and monitoring tabs of various types. During this same period approximately 470 sodium samples have been analyzed, of which 365 were analyzed for carbon with the remainder for oxygen and emission spectrograph. Therefore, this program has provided a wealth of information on the carburization phenomenon in a high carbon sodium system. During this same period the studies in the high oxygen sodium system have

provided similar data of equal interest.

As reported at the time of the previous meeting, the effects of low oxygen sodium on the mechanical properties of 316 ss under these conditions did not appear to be significantly detrimental. A very brief summation of results shows:

1. The creep-rupture strength of 316 ss at 1200 F to be equal in all three environments (air, helium and low oxygen sodium) over the rupture times (100 to 4000 hours) covered by this program.
2. The creep rates of 316 ss at 1200 F in helium and low oxygen sodium, as compared to air, show a 12% decrease in the stress necessary to produce a minimum creep rate of 1% in 10,000 hours.
3. In comparison to air, the low strain fatigue life in helium and sodium increased by a factor of 4.

In contrast, the current studies in carbon-rich and oxygen-rich sodium environments at 1200 F are showing significant effects on 316 ss mechanical test specimens as well as monitoring tabs of 304 ss and 2-1/4 Cr-1 Mo steel. Generally the effects are detrimental and would result in a downgrading of the usefulness of these materials under these conditions. Both of these studies are in the final stages of testing. The metallographic examination of the many test specimens generated is in progress. Therefore, final conclusions should not be made at this time. However, sufficient work has been completed to allow us to indicate what we believe to be the main conclusions at this time.

EFFECT OF HIGH CARBON SODIUM ON MECHANICAL PROPERTIES OF 316 SS AT 1200 F

Without going into details at this time, the operation of the high carbon sodium environment can be stated as being centered around an instream carbon bed. A portion of the sodium flow at 1200 F is circulated as necessary through this bed to maintain a plugging temperature of 750-800 F. At the same time a circulating cold trap is used to maintain the oxygen level at less than 30 ppm. The operating procedures were established during preliminary runs and were found to be effective in maintaining the carbon concentration in the sodium system at a sufficiently high level to assure the high carburization environment desired for this study. The mechanical tests (creep-rupture, creep and fatigue) took place in this system at 1200 F as well as the 4000 hour exposure of other specimens for further testing in air, helium and high carbon sodium environments. Tabs for metallurgical examinations were also exposed during each run.

The extensive carburization under these conditions of 316 ss, 304 ss and 2-1/4 Cr-1 Mo is clearly substantiated by the results of the carbon analyses of the specimens and tabs, as well as their weight changes, microhardness profiles and photomicrographs. For example, the average changes in carbon content of the monitoring tabs accompanying the 4000 hour creep tests are shown in Table 1.

TABLE 1 - TOTAL CARBON CHANGE IN 4000 HOURS
OF 65 MIL SHEET SPECIMENS

<u>Material</u>	<u>Original Carbon ppm</u>	<u>Final Carbon ppm</u>	<u>Carbon Case Thickness-mils</u>
304 ss	667	9555	6.8
316 ss	480	6115	5.6
2-1/4 Cr-1 Mo	981	3334	None

During the preliminary runs, prior to the mechanical tests, the carbon diffusion coefficient for carbon in 316 ss was calculated based on thin tabs. A comparison of the total carbon pickup of all the 316 ss specimens against the calculated diffusion rate based on the carbon diffusion is shown in Fig. 1. The calculated curve is based on Fick's Law in the form:

$$\frac{C_m - C_o}{C_s - C_o} = \frac{1.128}{L} \sqrt{Dt}$$

Where: C_m = mean concentration at t
 C_o = original concentration
 C_s = surface concentration
 L = 1/2 specimen thickness (cm)
 D = diffusion coefficient
 t = time - sec.

While there is considerable scatter, the overall agreement appears to be good. Some of the scatter can be attributed to the different positions of the specimens in the test units. The mechanical test specimens were in the main stream while the monitoring tabs were in an annular space between the inner and outer shell of the test units. The carbon sample from the specimen may also have contributed to the overall scatter due to difficulties involved in obtaining uniform samples without changing the ratio between the carbon case and the overall sample. The analyses of the specimens as well as statistical analyses of the data is continuing.

As will be pointed out in the metallurgy section, extensive carburization resulted in the formation of a surface carbide case upon the 316 ss mechanical test specimens. This hard and brittle case directly influenced the mechanical properties of the specimens generally in the manner of losses

in ductility and increases in creep and rupture strengths. Specific results of the mechanical tests follow.

Creep-Rupture of 316 ss

Results of the six creep-rupture tests conducted in the high carbon sodium system at different stress levels, over times of 100 to 5000 hours, are shown in Fig. 2. The creep-rupture curve from the air, helium and low oxygen sodium environments is shown for comparison. The comparison distinctly shows the increase in the creep-rupture strengths after about 300 hours in the carburizing environment. The results of the mechanical tests in air and helium on 4000 hours pre-exposed specimens are incomplete at this time. Specimens exposed for 4000 hours, unstressed, in the high carbon sodium have been mechanically tested in the same environment and the data is included in Fig. 2. These results continue the trend of increased rupture strength with increased exposure times (increased carburization). A corresponding decrease in ductility was experienced in the high carbon-sodium environment. This is illustrated in Table 2 by comparison of the elongation and reduction of area data from the low oxygen-sodium with the high carbon-sodium runs at equal stress levels.

TABLE 2 - COMPARISON OF DUCTILITY OF 316 SS DURING CREEP-RUPTURE TESTS IN LOW OXYGEN-SODIUM AND HIGH CARBON-SODIUM ENVIRONMENTS

Stress Psi	Elongation - %		Reduction in Area - %	
	Low Oxygen-Sodium	High Carbon-Sodium	Low Oxygen-Sodium	High Carbon-Sodium
27,500	74	~ 37	50	20
24,000	62	24	49	17
21,500	70	20	46	17
20,000	53	8	36	0

Creep Rate - 316 ss

The influence of the high carbon-sodium environment on the creep rate of 316 ss was both significant and distinctive as compared to the other environments. Three creep runs at different stresses were conducted for 4000 hours. The change in the creep rate versus time is shown graphically in Fig. 3. The curves clearly show the influence of the carbide case being formed during the creep runs. The creep rates at the low stresses appear to pass through stages of varying rates as indicated by the inflections of the creep curves during the second stage. The final minimum creep rates were maintained for only a brief time. There did not appear to be characteristic second stage creep periods under these conditions. Without attempting to evaluate their meaningfulness at this

time the minimum creep rates obtained during these runs are compared in Fig. 4 to those from the air, helium and low oxygen-sodium environments.

The minimum creep rate reached under the highest stress shows substantial agreement with the minimum creep data from the creep-rupture runs. However, the two creep runs at the lower stresses do not appear to fit the other data. Because of the markedly changing character of the material during test, interpretation of the creep data is complex and should not be regarded in the same light as creep properties of homogeneous material.

Fatigue Tests

The high temperature, cyclic strain fatigue tests consisted of triplicate specimens run at three different strain levels in the high carbon-sodium system. As previously reported, the fatigue life of 316 ss is increased in the low oxygen-sodium system as compared to air under these test conditions. This continued to be true in the high carbon-sodium system but to a lesser degree. The change in fatigue life of 316 ss due to the high carbon-sodium system can be seen in Fig. 5 where the fatigue curve for the high carbon-sodium runs is compared with the low oxygen-sodium system. A consistently shorter fatigue life was experienced in the high carbon-sodium system as compared to the low oxygen-sodium system. Considering the brittle nature of the carbide case, and knowing that fatigue failures are sensitive to surface effects, it might be reasoned that even greater differences in the fatigue life of carburized specimens might be observed at lower temperatures where materials are less ductile. The fatigue of specimens exposed only 270 hours showed scattered results ranging from 100 to 300 cycles when tested in air, helium and high carbon-sodium.

EFFECT OF HIGH OXYGEN SODIUM ON MECHANICAL PROPERTIES OF 316 SS AT 1200 F

The mechanical testing in the high oxygen-sodium system was identical to the low oxygen program with the exception that the cold trap was operated at a temperature of 700 F. The difference in the oxygen concentrations therefore was 300 ppm rather than 30 ppm. The results of the testing completed to date will be discussed, but the final conclusions must await the completion of the mechanical tests on exposed specimens and complete metallurgical examination of the numerous specimens. While the influence of the high oxygen-sodium system on the mechanical properties may not appear as spectacular as the high carbon-sodium environment, the results may be more difficult to interpret. Some of the effects observed are: scattered weight changes of the test material; an increase in scatter of the mechanical test results; the formation of scale on the specimens; scattered carburization (although relatively low as compared to the high carbon environment); and, general mass transfer of metals. These

phenomena occurred in varying degrees during supposedly similar test runs. The data, therefore, offers a challenge in evaluating the significance of observed differences in test results.

Creep Rupture

The five completed runs and one run still in progress, each at different stress levels, are shown in Fig. 6. The creep-rupture of 316 ss in air, helium and low oxygen-sodium are also shown for comparison. Little difference is observed for short-time tests up to 1000 hours. However, the picture is not as clear for longer times. The 17,750 psi stressed specimen showed a rupture life in agreement with the former environments but with an unusually high loss in ductility. The succeeding two specimens revealed significant increases in rupture strength. This apparent scatter has not been resolved at this time. In general, the specimens exposed to this environment showed a small, but consistent, reduction in ductility.

Creep Tests

The primary and secondary creep rate curves appeared to be of normal character. However, no third stage creep rates were reached during the 4000 hour runs. The minimum creep rates, as shown in Fig. 7, show a good correlation with the values obtained from the creep-rupture tests. In the range of interest of this program (0.1% in 1000 hours) the minimum creep rate in high oxygen-sodium appears to be less than in either the low oxygen-sodium or helium environments, but higher than in air.

Fatigue Tests

Of all the mechanical tests completed to date, the most difficult to interpret at this time are the fatigue results for the high oxygen-sodium environment. As compared to the low oxygen-sodium environment a decrease in fatigue life was found at the low strain but an increase was apparent at the intermediate strain. Equal strengths were observed at the highest strains. When these two environments are therefore compared with regard to fatigue life two entirely different shaped curves are obtained, as shown in Fig. 8. Final conclusions and analyses may be clearer upon completion of the metallurgical examinations.

* * *

These then are the highlights of the mechanical tests in the two current environments under study. Time does not permit a discussion of the many other observations and studies associated with this program. The analytical program alone is worthy of considerable attention. All of these will be included in the topical reports covering these programs. However, in order to clarify the results as much as possible at this time, the overall broad results observed to date during the metallurgical studies are included in the following section.

EFFECT OF CARBON ON MICROSTRUCTURE AND COMPOSITION OF STRUCTURAL MATERIALS

Post test evaluation of samples exposed to a carbon-rich sodium environment included metallographic analysis, macro and microhardness measurements, weight change measurement, surface X-ray diffraction and microprobe analysis. The following generalizations describe the more important effects that were noted.

A number of tabular materials were exposed to the carbon-rich sodium environment. Gains in carbon content by a number of alloys of unit thickness and exposed for a single exposure time (521 hours) are of the approximate order: 321 > 304 > 309 > 316 > Incoloy 800 and 310 > 330 > 2-1/4 Cr-1 Mo > Inconel 600 > Nickel and Armco iron. The nickel and Armco iron did not absorb carbon in this test environment. The above relationship is likely to be altered at longer durations of exposure due to differences in carbon diffusion rates and differences in rate at which super-saturated carbides are precipitated from solid solution.

The effects of carbon-rich sodium on 316, 304 and 2-1/4 Cr-1 Mo were studied extensively. The study of 316 and 304 corrosion tabs resulted in the following conclusions:

1. For the first few days of exposure 316 and 304 carburize at approximately the same rate (0.23 mgms/cm²/day). After this initial period, the carburization rate of 316 declines more rapidly than that of 304. Fig. 9 compares the case thicknesses of carburized 304 and 316. The more rapid carburization of the 304 is accompanied by a greater transformation rate from austenite to ferrite as evidenced by metallographic observations. Greater ferromagnetic behavior is also seen in exposed 304, even when the 304 has absorbed less carbon than the 316.
2. Analysis of carbide residues from each alloy indicated that the more carbon-rich M₇C₃ phase is more predominant in 304, while the M₂₃C₆ phase exists in carburized 316 even at equivalent carbon levels in the two alloys.
3. Weight gain data tends to be less scattered than carbon analysis in indicating carbon absorption levels. This is due to difficulty in sectioning heavily carburized specimens, where cutting can occasionally result in loss of the brittle carbon-rich case.
4. Microhardness profiles are instructive in characterizing the carbon diffusion wave.

5. Little effect of stain-inducing carbon diffusion was noted in this carbon-rich sodium environment. Previous examination suggested that in a carbon-poor environment, carburization may have been enhanced by strain.
6. Microprobe analyses showed depletion of Cr and Ni from the surfaces of carburized 316 and 304 after having been exposed 2000 hours, but depletion only occurred to shallow depths (< 6 microns). Longer term exposure (5380 hours) to the carbon-rich sodium environment showed a distinct difference between 304 and 316. Fig. 10 shows a microprobe profile of 304 that had been carburized in the carbon-rich sodium environment. The 304 had formed a chromium-rich outer carbide layer, while the 316 still showed slight surface chromium depletion. The mass transfer of iron to nickel and Inconel was also noted (Fig. 11). An iron rich ($\sim 50\%$) layer was found on nickel to a depth of approximately one mil.
7. Differences in carburization rates were noted in various portions of the loop. Carburization appears to be less rapid farthest from the carbon bed, or in a stagnant flow environment. Scatter in carburization rates were also noted where the specimens were masked by a massive metal sink, such as in fatigue testing where the mandrel tended to shield fatigue specimens.
8. Thin austenitic structural components such as valve bellows were fully penetrated by carbon. With such carbon levels, these members were as friable as potato chips.
9. The room temperature impact properties of 316 and 304 were reduced almost 50% when aged in helium for 4000 hours. This reduction was a result of sensitization (carbide precipitation in the grain boundaries). When immersed in carbon-rich sodium, impact strength declined more sharply and reached a value only 20% of the original material. A transition from ductile to a brittle mode of fracture occurred when 304 and 316 were carburized.
10. No sigma phase was noted in any of the 316 specimens exposed to any of the environments. This may have been partly due to the fact that the 316 heat was specifically chosen with a composition that would avoid or retard sigma formation.

11. The mass transfer of metallic constituents to the carbon bed was noted. Analyses of the carbon constituents transferred and the extent of such transfer is still under investigation.
12. Specimens of 316 that were initially 0.060" thick swelled up to 1-2 mils in 5300 hours as a result of carburization. This swelling was approximately proportional to carbon pickup and is attributed to lattice expansion accompanying carburization.

Tabular specimens of 2-1/4 Cr-1 Mo were also exposed to the 1200 F carbon-rich sodium environment. This alloy rapidly absorbed carbon to an equilibrium value of approximately 0.5%. Chrome-moly specimens absorbed less carbon when in close proximity to stainless steel samples.

Carburization did not reduce the room temperature impact properties of 2-1/4 Cr-1 Mo, since the carbon that was absorbed formed spheroidized carbide precipitates rather than a brittle carbide case. In fact, a slight increase in impact properties was noted, but was shown to be the result of long term thermal effects rather than environmental effects.

The metallurgical observations described above, as well as other effects that have not been mentioned, provide a number of explanations for effects observed in mechanical property tests.

The formation of the carbide case upon the 316 mechanical test specimen is no doubt responsible for loss in ductility and an increase in creep and rupture strength. The carbide layer with its higher modulus and strength must yield before elongation of the uncarburized stainless substrate occurs. With increasing time and resulting increase in thickness of the carburized layer, the character of the structural material changes to greater strengths.

A similar explanation is evident for the unusual character of the creep curves. The initial creep properties are that of the virgin stainless but after sufficient time (greater than 2000 hours), the carburized material with its duplex character reduces creep rate during the second stage. Thus it would appear that the lowest creep rate just prior to tertiary creep is characteristic of the carburized material.

The effect of carbon on cyclic tests is more complex and difficult to interpret, primarily since exposure times are short and carbide cases constitute but a small portion of the cross-sectional area. The reduction of life is likely the result of the low ductility of the case. Cracks initiated in the case are able to proceed into the unaffected stainless. In essence, the high activation energy necessary for crack initiation is circumvented by the presence of the non-ductile surface.

EFFECT OF OXYGEN IN SODIUM ON MICROSTRUCTURE AND COMPOSITION OF STRUCTURAL MATERIALS

The effect of oxygen in sodium on the mechanical properties of 2-1/4 Cr-1 Mo were described at the Hollywood meeting. Since that time metallurgical evaluation of this study has been completed.

Surface oxidation of 2-1/4 Cr-1 Mo steel in 1100 F air appears to be responsible for the lower fatigue life in an air environment as compared to those of helium, sodium or sodium containing 200-300 ppm oxygen.

The results of carbon analysis show that decarburization is occurring during exposure of 2-1/4 Cr-1 Mo steel to 1100 F sodium, with higher decarburization rates resulting in the presence of added oxygen. Decarburization of Cr-Mo by sodium is thought to be partially responsible for a number of effects which were more prevalent in oxygen containing sodium than in pure sodium. These include:

1. Reduction of stress-rupture life below that of air and helium environments.
2. Increase of ductility.
3. Reduction in hardness (both macrohardness and micro-profile).
4. Further reduction of stress-rupture life of specimens pre-exposed for 4000 hours.

No compositional changes other than carbon depletion were noted after exposure of Cr-Mo to low oxygen-sodium. An oxide phase is formed on the surface of Cr-Mo exposed to high oxygen. Penetration of this corrosive phase into the matrix is increased by application of a stress. However, the observed corrosion has not measurably affected the mechanical properties within the limits of the tests. Such intergranular effects should be expected to ultimately affect mechanical properties, with measurable changes evidenced in longer times.

Metallurgical evaluation of the study on the effects of oxygen in sodium on 316 is not yet complete. However, preliminary analysis of this test series indicates that the effects of oxygen in sodium on microstructural and compositional changes are not as clear-cut as in the case of carbon contamination. This is a result of a number of factors including:

1. Corrosion of stainless steels by oxidation process resulting in scale-like corrosion products. Weight gain data is highly scattered due to the loss of corrosion deposits either by mechanical or thermal effects.

2. Carbon transfer can occur and superimpose effects over that of oxidative corrosion.
3. Mass transfer of metallic species occurs more readily with oxygen contamination.

Preliminary observations on this test series are as follows:

1. Stainless steels (304 and 316) are oxidized more readily in 1200 F sodium containing 300 ppm oxygen than in 1200 F air. X-ray examination of surfaces reveals the presence of oxides of the form $\text{Na}_x\text{M}_y\text{O}_z$. Microprobe results (Fig. 12) confirmed this layer to be rich in chromium, indicating that the oxide layer is sodium chromite. Extensive surface porosity was also indicated. Intergranular penetration of oxygen is not as rapid as with carbon.
2. A number of stainless steel samples showed slight carbon increases to approximately 0.1%. The source of this carbon is thought to be ingestion of trace quantities of CO_2 during opening and closing of test pots.
3. The impact properties of 316 and 304 were reduced below that resulting from thermal effects alone. However, the reduction in impact strength was not as great as that caused by carburization.

Tabular samples of 2-1/4 Cr-1 Mo were exposed to 1200 F sodium containing oxygen. Decarburization was found to occur much more rapidly than at 1100 F. Weight gain data, although scattered, shows that chrome-moly oxidizes considerably, although concurrent weight loss due to mass transfer is suspected. Slight increases in impact properties were noted, and are probably due to decarburization.

CARBURIZATION OF 316 SS AT 1200°F
IN A HIGH CARBON - SODIUM SYSTEM
FIG. 1

CREEP RUPTURE OF 316 SS AT 1200°F
COMPARISON OF HIGH CARBON SODIUM WITH AIR,
He, AND LOW OXYGEN SODIUM ENVIRONMENTS
FIG. 2

CREEP RATES OF 316 SS IN HIGH CARBON-SODIUM ENVIRONMENT AT 1200°F
FIG. 3

MINIMUM CREEP RATE OF 316 SS AT 1200°F
COMPARISON OF HIGH CARBON SODIUM WITH AIR, He, AND LOW OXYGEN SODIUM ENVIRONMENTS
FIG. 4

316 SS FATIGUE AT 1200°F IN LOW OXYGEN-SODIUM
AND HIGH CARBON-SODIUM ENVIRONMENTS
FIG. 5

CREEP RUPTURE OF 316 SS AT 1200°F
COMPARISON OF HIGH OXYGEN SODIUM WITH AIR,
He AND LOW OXYGEN SODIUM ENVIRONMENTS
FIG. 6

MINIMUM CREEP RATES OF 316 SS IN AIR, He AND Na AT 1200°F
FIG. 7

316 SS FATIGUE AT 1200°F IN LOW OXYGEN-SODIUM
AND HIGH OXYGEN-SODIUM ENVIRONMENTS
FIG. 8

304 ss 316 ss
 Exposure: Carbon-Rich Sodium
 at 1200 F For 21 Days
 Mag 133X Ferric Chloride Etch
 COMPARISON OF THE CARBURIZATION
 OF 304 AND 316 SS IN LIQUID SODIUM
 FIG. 9

MICROPROBE PROFILE OF 304 STAINLESS
 EXPOSED TO A HIGH CARBON SODIUM EN-
 VIRONMENT (5380 hrs, 1200°F)

Note Cr Enrichment At The Edge
 FIG. 10

MICROPROBE PROFILE OF NICKEL EXPOSED
TO A HIGH CARBON SODIUM ENVIRONMENT
(3800 hrs, 1200°F)

Note Fe Enrichment At The Edge

FIG. 11

MICROPROBE PROFILE OF 304 STAINLESS
EXPOSED TO A HIGH OXYGEN SODIUM EN-
VIRONMENT (5688 hrs, 1200°F)

Note Cr Enrichment At The Edge

FIG. 12

GENERAL ELECTRIC SODIUM MASS TRANSFER PROGRAM

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PROJECT STATUS

A study of mass transfer and corrosion of materials in flowing sodium has been in progress at General Electric since late 1959 as part of the AEC Sodium Components Development Program. The objective of this work has been to evaluate the use of candidate materials in sodium cooled reactors in non-nuclear loop tests and to develop a means of predicting corrosion or mass transfer rates in full sized systems. The initial phase of testing 316 stainless, $2\frac{1}{4}$ Cr-1 Mo ($2\frac{1}{4}$ Cr) and 5 Cr- $\frac{1}{2}$ Mo- $\frac{1}{2}$ Ti (5 Cr) steels in six pumped test loops is complete. This involved the determination of the effects on mass transfer of three primary variables: loop maximum temperature, overall loop ΔT and oxide impurity level as measured by plugging temperature. The materials from which hot and cold legs of test loops were constructed and in which specimens of like materials were tested are shown in Table I.

TABLE I

MATERIAL COMBINATIONS IN MASS TRANSFER TEST LOOPS

<u>Loop #</u>	<u>Hot Leg</u>	<u>Cold Leg</u>
1*	316 SS	316 SS
2	316 SS	$2\frac{1}{4}$ Cr
3*	316 SS	$2\frac{1}{4}$ Cr
4*	316 SS	5 Cr
5*	$2\frac{1}{4}$ Cr	$2\frac{1}{4}$ Cr
6*	5 Cr	5 Cr

Details of the design and construction of these loops have been reported in GEAP-3725. (1)

Five run types varying the primary variables were made in each of five loops (*Table I) comprising the various material combinations. These tests represented the minimum combination for two levels of the variables with one interaction run. The run conditions for this five run fractional replicate are listed in Table II (run Types I through V). Two additional interaction runs, Types VI and VIII, were also made in individual loops to strengthen the mass transfer data. The remaining run Type VII of the eight possible combinations was not made because of cold leg flow fouling due to higher corrosion and deposition rates at high temperature and high oxygen conditions. Plugging of small cold leg passages was experienced in run Types I and V.

TABLE II
TEST LOOP CONDITIONS

<u>Run Type</u>		<u>T max</u>	<u>Degrees F</u>	
			<u>ΔT</u>	<u>t_s^*</u>
	I	1200	500	450
	II	1200	500	305
	III	1100	500	305
	IV	1200	250	305
	V	1100	250	450
interaction runs	VI	1100	500	450
	VII	1200	250	450
	VIII	1100	250	305

* t_s plugging temperature 305 $\cong$ 12 ppm oxygen
450 $\cong$ 50 ppm oxygen

Loop #2, a duplicate of Loop #3, was operated at constant run Type II conditions to test the validity of extrapolating mass transfer data from 2800-hour runs (used in the fractional replicate program) to longer times. The longest run data obtained in Loop #2 was about 20,000 hours and yielded linear mass transfer rates compatible with short-time data.

Within the past year work has been concentrated in data analysis, complete metallurgical, electron microscope, and microprobe examinations of test loops, and the design and construction of two new corrosion mock-up loops. A list of project reports available and forthcoming is attached.

During the operation of the mass transfer loops, sodium has been analyzed regularly for carbon and metallic impurities. It has not been possible to establish any direct correlation between the level of impurities (other than oxygen) and the rates of mass transfer. Similarly, from the analysis of these elements and rates for calcium and hydrogen, it has not been possible to confirm any of several hypotheses which attempt to explain the appearance of multiple breaks in plugging meter flow curves. It is tentatively concluded that these phenomena are related to complex equilibria which involve compounds of the elements undergoing mass transfer. New techniques for obtaining and analyzing large sodium samples are needed to determine these effects, and such a program in sodium impurity separations and identification was initiated in late 1964. The methods selected for evaluation include liquid ammonia dissolution, distillation, and amalgamation. The processes are outlined in Figure 1. The chemical form of some impurity compounds present in the sodium may change during removal of sodium so methods that vary in temperature of operation and in chemistry are being used. Capacity for kilogram samples is being provided to insure high sensitivity and reliable identifications.

Emphasis is being placed initially on non-destructive analytical methods such as X-ray and electron diffraction for identification of compounds. Gases evolved during separations will be collected and analyzed to determine whether decomposition products of unstable compounds are present. Separated impurities will be encapsulated to prevent atmospheric contamination during identification procedures.

The amalgamation system hardware is complete and initial sodium samples have been successfully manipulated. Vacuum tweezers have worked well for removing residual materials from mercury surfaces. Assembly of the liquid ammonia system was completed in June. Filter materials for this system have been tested by X-ray analysis of residues. Duralon and polyethylene have been found sufficiently pure for use. Fabrication of the distillation system was completed in July.

MASS TRANSFER RESULTS

Statistical analysis of weight change data from the 25 loop runs in the five-run-fractional replicate plan, and from supporting runs yielded the following general empirical equation for metal loss in hot-leg regions:

$$R = V^{0.884} C^{1.156} \exp \left(12.845 - \frac{23,827}{T + 460} - 0.00676 L/D_1 + \frac{2.26}{t + 1} \right)$$

where

R = metal loss rate, $\text{mg}/\text{dm}^2\text{-Mo}$

V = sodium velocity, ft/sec

C = oxide impurity concentration, ppm

T = local sodium temperature, $^{\circ}\text{F}$

L/D_1 = number of diameters downstream in an isothermal region
(L = length and D_1 = hydraulic diameter)

t = time, months

There is a similar equation for the cold leg. The statistical procedures and details of data analysis are contained in GEAP-4183 covering preliminary methods and results. GEAP-4830 covers final correlation of the data.

The empirical equation is most readily used to estimate steady-state loss rates at the beginning of an isothermal region at temperature T ($^{\circ}\text{F}$)

by omitting the two last terms in the exponential containing diameters downstream and time in months. Effects of operating variables and the primary variables on mass transfer are separately listed below for clarity.

Oxide - Rates varied directly in proportion to the oxide impurity level in the sodium.

Velocity - Measured rates varied linearly with sodium velocity.

Time - After an initial transient period of several hundred hours, the observed rate approached linearity. The reciprocal time term in the exponential of the empirical equation was inserted to account for an initial non-linear transient. This transient appeared to correspond to the establishment of a modified Fe-Ni-Cr composition layer on 316 SS.

Sample temperature - An increase of 100°F produced approximately a three-fold increase in rate.

Loop maximum temperature - Loop T_{max} was found to have no effect on observed rates at locations other than the T_{max} position. Local sample temperature remained the controlling factor.

Loop ΔT - Varying ΔT between 250°F and 500°F produced no significant change in measured rates.

Mass transfer data analysis showed no statistically significant difference in rates between 316 SS, 2 $\frac{1}{4}$ Cr or 5 Cr steels. All were observed to undergo mass transfer to an equivalent extent within the accuracy of the weight change measurements obtained. An example of metal loss rates applicable to the three steels tested obtained from the derived empirical equation is shown in Table III.

TABLE III

MASS TRANSFER RATES VERSUS OXYGEN AND TEMPERATURE

Average Metal Loss in mils/yr at 20 ft/sec Sodium Velocity

<u>Temperature °F</u>	<u>Oxide Impurity Content in Na</u>	
	<u>10-15 ppm</u>	<u>45-50 ppm</u>
1000	0.05	0.25
1100	0.15	0.75
1200	0.39	1.9
1300	0.95	4.7

The material loss rates given in Table III are average values. A factor of ~5 variation in the data is observed. Taking into account the statistical variations, the maximum values expected for 1200°F operation and at the low oxide level shows loss rates in the vicinity of 2 mils per year. This is prohibitive for some in-core components such as thin claddings. This implies that for bulk temperatures above 1100°F, very low oxygen contents must be maintained, for example ≤ 5 ppm at 1200°F.

Of the initial primary variables the most significant is the oxide impurity level. A model of the downstream or saturation effect has been developed, which correlates most of the existing data, but this remains to be fully confirmed. The present data do not permit unequivocal distinction between local velocity and mass flow rate as a primary variable. Because of the complexity of the possible interactions in a loop system with a large ΔT , it has been concluded that reactor mock-up loops with appropriate scaling of geometry and flow are necessary to improve the range of applicability of loop testing results.

LOCALIZED CORROSION EFFECTS

The occurrence of specific transport effects have been reported previously. These effects involve the selective but not spatially localized transport of chromium and nickel in low oxide (~10 ppm) impurity runs, the selective transport of iron in high oxide (~50 ppm) impurity runs, and the release of carbon from $2\frac{1}{4}$ Cr-1 Mo and subsequent uptake by 316 SS in the same loop.

Metallurgical examination of the four test loops has revealed localized corrosion effects. Loops that contained 316 SS piping in the entrance to the cold leg or "hot-to-cold-leg cross-over region" have invariably revealed an intergranular corrosion in this location. The attack rate is approximately 1.7 mils/yr. Unless anticipated by design, such a rate might adversely affect the integrity of the primary system components corresponding to this region.

An example of the metallographic appearance of 316 SS showing intergranular attack is given in Figure 2. The observed distribution of this attack around the periphery of $\frac{1}{2}$ -inch pipe sections is uniform. It has generally been found to begin in pipe walls several inches downstream of loop hot-leg exits where make-up heat is no longer being supplied to the system. Occurrence of the attack ceases near the entrance to the first cooler. No significant correlation has been observed between attack depth and sodium oxide impurity level (12 versus 50 ppm Ox) or temperature (1200 versus 1100°F Tmax). Measurements made of 316 SS intergranular attack in different loops with different program run conditions have remained consistent with a rate of ~1.7 mils/yr.

Electron microscope fractographic examinations of pipe specimens from a corroded area have been made. Pipe specimens fractured at liquid nitrogen temperature (to cause intergranular fracture) revealed the morphology of grain surfaces and carbide precipitates. Figure 3 shows a carbon extraction replica illustrating the differences between attacked and non-attacked grain surfaces. The upper right hand corner of the photograph

shows the high density of particles identified primarily as Cr_{23}C_6 that is characteristic of heavily sensitized 316 SS and is observed on grain surfaces where no intergranular attack had occurred. The remaining areas show chemical attack on crystal faces resulting in a stepped pattern on grain surfaces in areas of intergranular attack. Relatively few larger carbide particles remain in these areas and they maintain sharp angular configurations that indicate no attack on the particles themselves. Chemical attack of grain surfaces apparently loosened the smaller carbide particles such that they were lost in the fracture process and only the larger particles remained pinned in place.

Results of the examination can be summarized as follows:

1. The stainless steel piping was heavily sensitized in the cross-over region.
2. There is no attack of the grain boundary precipitates themselves.
3. Chemical attack proceeded by solution of metal at the interface with the grain boundary precipitate.

A preliminary interpretation of this effect is that attack results from a high (effective) oxide impurity level created at the entrance to the cold leg. It is postulated that this region experiences the beginning of supersaturation and subsequent nucleation of metal precipitates, with the accompanying liberation of oxygen to sodium oxide. This causes attack in grain boundary areas which are suspected to be depleted in chromium by the precipitation of Cr_{23}C_6 at the grain boundaries.

Further tests to confirm the mechanism of this attack, and to determine differences in sensitivity of different alloys are in progress or scheduled.

CORROSION MOCK UPS FOR LARGE SODIUM SYSTEMS

As described earlier in this paper and in previous project presentations, the exposure effect of flowing sodium on steel is dependent on location, geometry and upstream conditions in the system as well as on the temperature, flow rate and impurity level. The system designer has some options on the temperature distribution, location and velocity parts of the exposure effect. The experimental data to date are regarded as sufficient to establish the corrosion model and calculational methods, but insufficient to permit the designer to make reliable quantitative trade-offs between these effects. This uncertainty would be reflected by the requirement of considerable safety margins with respect to temperatures, velocities and oxygen levels.

In order to obtain more complete exposure-effect data we are assembling two corrosion mock-up test loops, #8 and #9, which are designed to simulate the sodium exposure effect conditions in a large reactor system. These conditions include impurity levels (plugging temperatures), operating temperatures and temperature gradients, velocities and materials.

The pertinent thermal-hydraulic and geometric characteristics of the sodium systems of four recent fast reactor 1000 MWe design studies* were determined and used as a basis for estimating the corrosion characteristics of each plant.

The thermal, geometrical and flow characteristics of the corrosion mock-up test loops were designed so as to provide the same corrosion potential in the test loops as one of the reactor plants at the upstream end of each selected region of the complete system. This was accomplished by using the formula

$$\Delta S = \overline{CR} (A/W)$$

where ΔS is the change in impurity level of the sodium due to the corrosion

* COO 279 - "An Evaluation of Four Design Studies of a 1000 MWe Ceramic Fueled Fast Breeder Reactor"

process, CR is the average corrosion rate of the region, A is the surface area of the component, and W is the flow rate of sodium through the component.

The reactor plant regional impurity change, ΔS_R in ppt, was determined by utilizing the programs corrosion rate prediction equation* and the design study plant dimensions and operating conditions. Figure 4 shows these predicted impurities changes in the primary system regions, compared to maximum impurities changes from existing test loop data. The prediction equation for this particular comparison was normalized so that the maximum impurity change calculated for the mock-up loops would agree with existing loop data. In the normalization the assumption was made that the same saturation in sodium impurities would result at core and blanket outlet conditions as indicated by test loop hot leg outlet data. The normalized equation was not used for the new loop designs; however, the validity of this comparison can be checked by subsequent new loop data analysis. The basic driving force for corrosion in the hot leg of the primary system is the difference between the two curves in Figure 4.

The design of the mock-up test loop components (corrosion samples) was determined by equating the reactor and test loop regional impurity change

$$\Delta S_{i, \text{Test loop}} = \Delta S_{i, \text{Reactor}}$$

and calculating the dimensional characteristics using the reactor temperature and cold leg impurity level (plugging temperature). For a given flow rate this procedure results in many combinations of cross-section and lengths that will satisfy the above requirements. Fortunately relatively small size test loops can be designed that will satisfy the ΔS equivalence. Our loop scale down factor is 1/30,000. This size (~100 kW-6 gpm) was chosen as a compromise between cost of the test facility and corrosion

* See GEAP-4830 Sodim Mass Transfer XIV Statistical Analysis of 1961-1964 Sample Weight Change Data

sample sensitivity. An example of the variations in pipe and sample sizes satisfying specific geometry and impurity conditions is given in Figure 5. This figure illustrates the dimensional choices available from which 1300°F isothermal loop regions of various lengths (shown on the ordinate) can be designed to result in a change in sodium impurity level of 11 ppt for the conditions given. Note that these given conditions state that a 1300°F region exists immediately upstream of this section with a L/D of 300.

Since the core region of the plant contains the most corrosive region in the system and because it was most desirable to obtain realistic core conditions, Loop #9 was designed with the capability of accepting a three rod high heat flux heater to mock up, on a 1:1 basis a full scale unit cell section of the General Electric design study core. The test section is also designed to be able to accommodate any of the four "1000 MW Design Studies" issued in 1964. The characteristics of the General Electric core were most amenable to making an electrical heater mock-up due to the short length. The remainder of the primary system was designed by using the ΔS and the predicted corrosion rate equations. Loop #8 is identical to Loop #9 except that a low heat flux heater replaces the core "unit cell" heater. It was designed to have the same ΔS as the reactor core and the unit cell in Loop #9.

Predicted corrosion rates and ΔS values in the target 1000 MWe primary system and the mock-up test loops are presented for comparison in Table IV. The similarity of conditions and magnitude of anticipated corrosion rates reflect the extent to which the mock-up can be made to simulate a large system.

At present the new loops are being installed and the first developmental high heat flux heater unit for Loop #9 is being prepared for test.

The anticipated results from the corrosion mock-up loops include

1. More realistic exposure conditions
2. Covers the range of conditions required for localized intergranular attack
3. Verification of ΔS concept and corrosion rate formulas
4. Improved sodium impurity monitoring
5. Data to predict hot-channel flow-fouling.

TABLE IV
ΔS VALUES AND PREDICTED CORROSION RATES

<u>Reactor Areas</u>	<u>Test Loop Areas</u>	<u>T 100°F</u>	<u>Na Velocity v fps</u>	<u>Residence Time θ sec</u>	<u>ΔS ppt</u>	<u>Corrosion Rates</u>	
						<u>CR_{in} mg/dm²mo</u>	<u>CR_{out}</u>
Core		8-13	12	0.18	226	1.9	110
	Heater, HHF	8-13	12	0.16	254	1.7	100
	LHF	8-13	0.1-9	21.0	300	1.2	70
Blanket		13	12	0.39	452	87.0	2.1
	Zone A	13	12	0.36	442	76.0	1.5
P + Pipe		11	~17	2.7	1.8	1.0	0.9
	Zone B-C	13	~1	5.9	3.8	0.3	0.2
IHE		11-8	6	8.6	-398	-36.0	-17.0
	Cooler	13-8	3	1.8	-650	-21.0	-7.0
Return		8	19	3.2 ⁺	-5 ⁺	-30.0	-29.0
	Zone E-F	8	1-15	19.0	-306	-10.0	-0.1

⁺ These values do not include the tank flow region.

FIGURE 1 IMPURITY SEPARATIONS AND IDENTIFICATION

6.8 mils

29,311 HOURS

250X

FIGURE 2 316 STAINLESS STEEL INTERGRANULAR ATTACK

CARBON EXTRACTION

3000X

FIGURE 3 316 STAINLESS STEEL PRECIPITATE AND GRAIN FACE MORPHOLOGY

FIGURE 4 CORROSION IMPURITIES IN PRIMARY SYSTEM

FIGURE 5 TYPICAL DIMENSION MAP OF OPTIONAL COMPONENT GEOMETRIES

LIST OF SODIUM MASS TRANSFER PROJECT REPORTS

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IRRADIATION EFFECTS IN STAINLESS STEELS AT HIGH TEMPERATURES

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ABSTRACT

The effect of irradiation on the elevated-temperature mechanical properties of structural materials is described using type 304 stainless steel as an example. The general result is that the grain boundary fracture process, but not the deformation process, is affected. All data suggest the primary cause to be helium generated from (n, α) reactions. Several metallurgical techniques for improving the ductilities of irradiated alloys are suggested, and experimental data are given for type 304 stainless steel for which the degree of improvement is demonstrated.

INTRODUCTION

The effect of irradiation on the stress-strain behavior of structural austenitic stainless steels¹ tested above 500°C is different from that observed for the alloys tested below 500°C, as illustrated in Fig. 1. Irradiating an alloy below 500°C produces a structure with a higher yield stress, a lower strain-hardening coefficient (and hence a lower uniform strain), and lower ductility in terms of total elongation than the unirradiated alloy when tested below 500°C. In many instances, no change in true fracture strain or stress is observed for stainless steels irradiated to less than 1×10^{21} neutrons/cm².

Above 500°C the effect of irradiation on these materials is different; first, there are reductions in ductility in terms of uniform and fracture strains as well as total elongation, and the magnitude of these reductions is highly strain-rate sensitive. Generally, changes in yield stress and in the stress-strain relationship are not observed. Since the ductility is reduced, the ultimate tensile strengths and fracture strengths are also reduced, and the magnitude of this reduction is very sensitive to the work hardening capabilities of the alloy. The effects are schematically illustrated in Fig. 2. They are first noted at an homologous temperature of about 0.5, which represents the approximate temperature at which self diffusion becomes important and high-temperature creep becomes a consideration for reactor design.

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ELEVATED-TEMPERATURE EMBRITTLEMENT BY HELIUM

There are two very significant differences between the irradiation effects below and above 500°C. First, the radiation damage affecting the low-temperature properties can be eliminated by postirradiation heat treatment. Much of the damage is removed in situ if the irradiation temperature is above approximately 350°C. However, postirradiation annealing of iron- and nickel-base alloys at temperatures between 1050 and 1300°C does not remove the damage effecting the low ductility at elevated temperatures. The thermal stability of the defect causing the low ductility has been illustrated previously.^{2,3} A second significant difference is the dependence upon the neutron energy or the energy spectrum of the neutron flux. The low-temperature damage is due to the production of vacancies and interstitials by fast neutrons. The thermal neutrons contribute only a small fraction of the damage. However, the magnitude of the elevated-temperature embrittlement correlates best with thermal neutrons⁴ for normal iron- and nickel-base alloys. Table 1 illustrates this for two stainless steels.

Table 1. Dose Dependence of High-Temperature Ductility in Irradiated Stainless Steels

Experiment	Dose, neutrons/cm ²		Elongation (%)
	Thermal Neutrons	Fission Neutrons	
a	0	0	55
	2.2×10^{19}	3.8×10^{18}	48
	2.2×10^{19}	1.9×10^{16}	29
	1.5×10^{20}	7.4×10^{17}	18
b	0	0	47
	1.6×10^{19}	3×10^{19}	20
	1.6×10^{20}	3×10^{19}	14
	7×10^{20}	5×10^{20}	9

^aA 20-25-Nb stainless steel tested at 750°C by Roberts and Harries at Harwell.

^bType 304 stainless steel tested at 840°C by Martin at ORNL.

Of all the thermal neutron reactions possible, the (n,α) reaction with ¹⁰B appears to be the one capable of producing a defect structure with thermal stability previously noted. This reaction also produces a lithium isotope, but the work of Higgins and Roberts⁵ suggests that the helium produced is more embrittling than the lithium. It was not until recently^{6,7} that the (n,α) reactions with other elements and fast neutrons were considered important (see Table 2). However, we emphasize that helium (alpha particles) can be produced from the reaction of fast neutrons with Fe, Ni, N, and other elements and that the quantities

produced will be significant for normal engineering alloys irradiated to a dose level above 1×10^{21} neutrons/cm². We believe then that the embrittlement at elevated temperatures is due to the production of helium gas. The total amount of helium in the alloy can be calculated for a given alloy composition and neutron dose. A correlation of helium content with short-time tensile ductilities is given in Fig. 3. This curve illustrates that as the deformation temperature is increased the amount of helium needed to initiate the embrittling process decreases. Curves such as the one shown in Fig. 3 are typical for most structural materials. The ¹⁰B from which the majority of helium is generated in these commercial heats is segregated in grain boundaries. Thus the concentration of helium at the grain boundary can be several orders of magnitude greater than the average value calculated and presented in Fig. 3. The actual amount is dependent upon grain size, preirradiation heat treatment, etc.

Table 2. Effect of Boron Content on High-Temperature Irradiation Damage to Type 304 Stainless Steel

Irradiation Exposure	Boron Content (ppm)	Elongation (%)		Helium Concentration, ppm		Total
		at 704°C	at 842°C	¹⁰ B (n,α)	Fast (n,α)	
At high levels of irradiation exposure (approx 5×10^{20} neutrons/cm ²)	0.015	19	6	0.015	0.3	0.315
	0.023	23	6	0.023	0.3	0.323
	0.11	22	6	0.091	0.3	0.391
	0.13	20	6	0.110	0.3	0.410
	3.9	13	3	3.3	0.3	3.6
At low levels of irradiation exposure (approx 5×10^{16} neutrons/cm ²)	0.11	39	41	0.00004	0.00006	0.0001
	3.9	40	29	0.00150	0.00006	0.0015

Enhancement of the Intergranular Fracture Process

The nature of the irradiation embrittlement is such that the nucleation and rate of propagation of wedge-type grain boundary cracks are affected. This is illustrated in Fig. 4, which shows the variation in number and length of grain boundary cracks at fracture. For example, the unirradiated material strained to 14% would not have any grain boundary cracks visible at this magnification. The mechanism by which helium affects the process of intergranular fracture is not known, although some have been proposed.⁸

The irradiation effects noted thus far for stainless steels are applicable for a number of structural materials. The variation in

ductilities of irradiated alloys at elevated temperatures is given in Table 3, illustrating that irradiation embrittlement at elevated temperatures is a general phenomenon and not one specific to one given class of alloys. Also, the effects described for short-time tensile tests are in general applicable to high-temperature creep.

Table 3. Postirradiation^a Ductility of Materials

Material	Tensile Ductility		Creep-Rupture Ductility		Reference
	at 700°C	at 800°C	at 700°C	at 800°C	
Inconel 600				0.7-10.0	9
Hastelloy N	6-20	5-11	0.2-0.5		10,11
Hastelloy X	2-5	1	4-20		2,11
Rene' 41			1-11		2
Nimonic PE16	1-8				12
A 286			1-3		2
304 stainless steel	11-30	3-40	1-2	1-3	1,9
20 Cr-25 Ni-Nb	4-30		0.4-10.0	14-22	13,12
316 stainless steel	15-20	0-10			12,14

^aDose = 10^{20} to 10^{21} neutrons/cm².

METHODS FOR IMPROVING THE DUCTILITY

Irradiation studies at the Oak Ridge National Laboratory with type 304 stainless steel indicate that the magnitude of irradiation embrittlement can be reduced by several metallurgical techniques. These methods are believed to be successful because the quantity of helium per unit length of the grain boundary is reduced and, in most cases, the stress necessary to nucleate and propagate a grain boundary crack is increased. A detailed discussion of the mechanism can be found elsewhere.^{15,16}

The influence of grain size on the ductility of irradiated stainless steel is illustrated in Table 4. Grain boundary precipitates¹⁶ are important also. The effect of increased carbon content is shown in Table 5. Note that at 900°C where all the carbon is in solid solution for both the low- and high-carbon steels the ductilities are equal. The effect of carbon precipitate is illustrated for the higher carbon steel by the data in Table 6. These data show that aging at elevated temperatures to induce precipitation and spheroidization of carbides also improves the postirradiation ductility. We believe that the spheroidization of grain boundary carbides may retard the propagation of grain boundary cracks in much the same way as proposed by Weaver.^{17,18}

Table 4. Influence of Grain Size on
Irradiation Embrittlement

ASTM Grain Size	Total Elongation, %			
	at 704°C		at 842°C	
	Unirradiated	Irradiated	Unirradiated	Irradiated
3	30	19	23	8
5	39	23	36	11
8-10	58	42	47	16

To improve the ductility of irradiated materials, one must devise ways to reduce the helium concentration at the grain boundaries. It is only the helium bubbles at the grain boundary that are believed to be deleterious; that is, helium generated within the grains would not be as harmful if these atoms were unable to move to the grain boundaries. For many reactor applications, the preponderance of helium generated is due to the transmutation of ^{10}B . Boron normally segregates to the grain boundaries in the solid state and therefore a large quantity of helium is generated at these boundaries. If one could form a stable boron compound, insoluble either in the melt or at a very high temperature after solidification, it would be possible to get a homogeneous distribution of this compound. Therefore, the helium generated would stay at the precipitate-matrix interface, and the quantity at the grain boundaries would be greatly reduced. The precipitate-matrix interface would also serve as a depository for helium generated from other elements and fast neutrons. Thus in principle this system should result in material with a lower susceptibility to elevated-temperature embrittlement in thermal and fast neutron environments.

This approach for improving the ductility of irradiated alloys is currently being investigated using the 18-8 stainless steel. Among the most stable borides in this stainless steel are those of titanium. We have now accumulated data from two different irradiations, and typical data are given in Table 7. It is observed that additions of approximately 0.3% Ti in type 304 stainless steel are beneficial. These recent experiments illustrate that much can be done to improve the ductilities of irradiated materials.

Table 5. Elevated-Temperature Ductility of Unirradiated and Irradiated
AISI Types 304L and 304 Stainless Steels

Deformation Temperature (°C)	Irradiation ^a Temperature (°C)	Carbon Content (wt %)	True Uniform Strain ^b , %		Total Elongation, %	
			Unirradi- ated	Irradi- ated	Unirradi- ated	Irradi- ated
650	700	0.02	22.5	15.1	42.2	16.9
	700	0.06	20.9	21.8	36.8	28.5
	800	0.02	23.6	13.1	39.8	17.6
	800	0.06	22.2	20.5	41.7	29.9
	900	0.02		10.5		11.3
	900	0.06	23.7	11.4	37.4	12.4
704	700	0.02	17.7	11.2	38.9	13.1
	700	0.06	17.8	17.1	41.5	23.0
	800	0.02	18.0	11.2	36.2	13.3
	800	0.06	14.0	14.1	36.6	20.3
	900	0.02	17.8	6.1	31.4	6.4
	900	0.06	17.4	14.6	28.0	
775	700	0.02	10.2	7.1	33.0	9.1
	700	0.06	10.4	9.6	40.7	15.7
	800	0.02	10.0	5.0	30.0	7.2
	800	0.06	9.3	7.6	40.0	12.6
	900	0.02		2.5		2.9
	900	0.06	14.1	3.4	31.7	4.0
842	700	0.02	9.8	4.8	38.4	6.7
	700	0.06	7.0	6.3	31.9	10.6
	800	0.02	10.4	2.8	36.8	4.0
	800	0.06	10.4	5.8	39.0	8.8
	900	0.02	10.2	1.8	24.5	2.2
	900	0.06	10.8	2.4	25.6	3.0

^aAlloys irradiated to a neutron dose of 3.5×10^{20} neutrons/cm² ($E > 1$ Mev) and 4.5×10^{20} neutrons/cm² (thermal).

^bSpecimens strained at a rate of 0.2% per min.

Table 6. The Effect of Aging on the Postirradiation Ductility of Type 304 Stainless Steel^a

Preirradiation Heat Treatment	Deformation Temperature (°C)	Elongation (%)
1 hr at 1036°C	704	20
1 hr at 1036°C; 100 hr at 800°C	704	30
1 hr at 1036°C	842	8
1 hr at 1036°C; 100 hr at 800°C	842	14

^aDose = 10^{21} neutrons/cm² (thermal).

Table 7. Effect of Titanium Content on the Postirradiation Ductility^a of Types 304 and 304L Stainless Steel

Element (wt %)		Ductility	
Titanium	Carbon	Irradiated ^b at 50°C	Irradiated ^c at 700°C
	0.02	13	7
0.3	0.02	38	29
1.2	0.02	14	14
	0.06	17	11
0.3	0.06	29	19
1.2	0.06	30	15

^aTensile tests conducted at 842°C.

^bDose = 1×10^{20} neutrons/cm².

^cDose = 5×10^{20} neutrons/cm².

SUMMARY

In summary, irradiation damage at elevated temperatures is an effect on the process of intergranular fracture, and thus the degree of embrittlement is highly sensitive to strain rate and temperature. The embrittlement is apparently due to helium, generated from thermal (n,α) and fast (n,α) reactions. We have illustrated several techniques for improving the ductility in irradiated stainless steel alloys and suggest that similar techniques should be applicable to all structural alloys.

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Fig. 1. Effect of Irradiation on the Stress-Strain Curves.

Influence of Irradiation at Temperatures Above $\frac{1}{2}T_m$ on the Post Irradiation Tensile Properties of Stainless Steel.

Fig. 2. Influence of Irradiation at Temperatures Above $\frac{1}{2}T_m$ on the Postirradiation Tensile Properties of Stainless Steel.

Fig. 3. Ductility of Irradiated Type 304 Stainless Steel.

Fig. 4. Comparison of Intergranular Cracking for (a) Unirradiated and (b) Irradiated Type 304 Stainless Steel Strained at 0.002 in./min at 704°C.

EVALUATION OF NON FUEL MATERIAL BEHAVIOR IN HNPF*

R.K. WAGNER and J.D. STEARNS

ATOMICS INTERNATIONAL

INTRODUCTION

The Hallam Nuclear Power Facility (HNPF) consists of a 240-Mwt sodium graphite reactor and the associated equipment for generating electrical power. This facility, designed and built by Atomics International, is located near Lincoln, Nebraska.

The reactor is sodium cooled, graphite moderated and initially used uranium-molybdenum alloy fuel. The graphite moderator logs are clad with 0.016-in-thick, Type 304 stainless steel jackets. The sodium, which becomes radioactive as it is heated in the reactor core, is pumped through three separate primary loops. Each loop contains a heat exchanger where the heat is transferred from the radioactive primary loop to a non-radioactive secondary sodium loop. Each of the three secondary loops has a steam generator in which feedwater is evaporated and the resulting steam is superheated. The steam from the three generators is fed into a common header which serves the turbine generator.

In the operation of HNPF, a few instances have developed which required evaluation of some of the non-fuel materials. This evaluation included: (A) the examination of a failed sodium expansion tank and, (B) the determination of the swelling characteristics of graphite when impregnated by sodium.

CORROSION ON SECONDARY EXPANSION TANK

The secondary sodium system at the Hallam Nuclear Power Facility contains three stainless steel pressure vessels. These vessels, one located in each of the three secondary hot legs, function as expansion tanks to accommodate the thermal expansion of the secondary sodium. In addition, the expansion tanks collect and recirculate the sodium overflow from the pumps through an 8-in. line. These vessels, 7 ft. in diam. and 10 ft. high, were fabricated from $\frac{1}{2}$ -in-thick, Type 304 stainless steel plate. The top and bottom forged heads are $\frac{7}{8}$ -in-thick stainless steel. Type 405 stainless steel heater shields, heater brackets, and Incoloy sheathed heaters were located in a 1-in-thick gap between the tank shell and the insulation of hydrated calcium silicate. Aluminum sheet is wrapped directly over the insulation. Inside the vessel, a $\frac{1}{2}$ -in-thick, Type 304 stainless steel baffle plate is positioned parallel to the tank axis. The baffle, slightly off the tank axis, extended across the vessel and half way up the tank wall. The baffle was joined to the shell by a series of continuous fillet welds between the baffle and either the tank wall or bottom head. Because this was not a sodium containment weld joint, special weld specifications were not applied.

* Work done under contract AT(11-1)-GEN-8 for the U.S. Atomic Energy Commission.

On October 18, 1964, a puddle of sodium was discovered on the floor under the expansion tank located in the secondary sodium loop No. 3. Following the normal precautionary measures to contain and absorb the leaking sodium, the sodium flow and the pressure in the tank were reduced and draining operations initiated. At the time the leak was discovered, the loop was isothermal at 415°F. The pressure in the expansion tank was 18.5 psig. During power generation at HNPF, the normal secondary hot leg temperature was 915°F with the pressure in the tank at 19 psig. It is estimated that the system had operated under the combined conditions for a period of 15,000 hours prior to the discovery of the leak.

A model of an expansion tank connected to a model pump illustrated in Figure 1 shows the general piping configuration around the tank. The principal pipe lines attached to the tank include: an 8-in. overflow line connecting the pump and the left side of the tank; a 14-in. outlet line from the bottom of the tank and a 14-in. inlet line. The area where the leak had developed would correspond to the lower left portion of the tank in the region around the 8-in. overflow line.

In Figure 2, the region of an expansion tank near the 8-in. overflow line and nozzle is shown. In addition the aluminum skin, the insulation blocks, the stainless steel heat shielding and brackets, and the tubular electrical heaters can be identified.

TANK APPEARANCE AFTER FAILURE

After the affected expansion tank had been drained, the aluminum skin covering the insulation was removed for examination of the vessel. The outside surfaces of the aluminum skin did not show any evidence of sodium attack and could provide no clue as to location or the leak size. After the aluminum skin was removed, however, it was quite apparent from the amount of reaction products present that a significant sodium leak had developed. Figure 3 shows the 8-in. overflow line region of the tank No. 3 after removal of the aluminum skin and some of the insulation. The 8-in. overflow line is shown near the top of the photograph. The reaction products and the reacted insulation are evident.

In the area of the leak, near the vessel to pump overflow nozzle, the insulation had been severely attacked. The stainless steel heat reflector shield, positioned between the insulation and the heater elements also showed evidence of severe attack. Figure 4 shows two sections of Type 405 stainless steel where dissolution was complete. The section on the left is a corner of the heat reflector showing the complete penetration through the 0.065-in. thick material. The section on the right is a portion of the heater bracket material.

Before the vessel in the region of the overflow line could be examined for identification and location of the leak, the massive reaction products adhering to the tank wall required removal. Reaction products were found down the side of the vessel, on the bottom head and on the surfaces of the outlet pipe at the bottom of the vessel, Figures 5 and 6. The reaction products appeared as a massive glacier-like deposit adhering to the walls of the vessel and the piping. The color of the products covered a wide range from white to gray and black. In some isolated areas, there appeared a green to yellow-green deposit superimposed on the main concrete-like mass. It was apparent that the deposits were hygroscopic in nature since a slow reaction, presumably with the moisture in the air, continued throughout the cleanup effort.

The clean-up operation was conducted in three phases; the removal of soft putty-like deposits by scraping with suitable spatulas, the mechanical chipping of the hard and tenacious mass with portable jack hammers and finally, the rinsing, and removal of the small isolated deposits remaining with butyl alcohol and distilled water.

During the final clean-up, using a fine spray of water from an atomizer, it was noticed that the dissolving residue would occasionally produce sparks suggesting the presence of unreacted sodium embedded in the deposits. The cleaned and rinsed section of the vessel and overflow nozzle is shown in Figure 7. Location of the deposit and the general shape and flow of the reaction products is evident. Much of the heater bracket material missing in the photograph had dissolved.

FAILURE ANALYSIS

To determine the cause of failure, a section of the vessel wall, the 8" diameter nozzle and a section of the 8" diameter overflow line were removed from the tank. Figures 8 and 9 show views of the failed expansion tank after the suspect areas had been cut out. The orientation of the baffle section, joined by fillet welds to the vessel wall and bottom head, is clearly evident.

The sections removed from the expansion tank are shown in Figure 10 and 11. On these sections, the severity of the corrosive attack, at the deposit-tank wall interface, is very apparent. In general, the gross attack was localized and took the form of grooves where in some areas the depth was half the vessel wall thickness, $\frac{1}{4}$ of an inch deep. Figures 12 and 13 show the groove type of attack extending from the vessel wall on the left, through a very heavy fillet weld joining the vessel to the 8" diameter nozzle and on across the nozzle surface. The internal surfaces, vessel, nozzle and overflow pipe, were clean. There was no evidence of either corrosive attack or built-up deposits. Figures 14 and 15 show the internal surfaces of the nozzle and overflow line. These are typical of all the surfaces examined.

TESTS PERFORMED AND RESULTS

A variety of non-destructive testing methods were employed to locate the sodium leaks. Before sectioning the tank, pressure testing quickly isolated the pinhole leak which had developed in the nozzle portion of the overflow line. During this effort, an internal helium pressure of $3\frac{1}{2}$ psig was maintained in the vessel. The helium leak detection equipment was useful in defining the general area of the leak. A soap solution sprayed over the surfaces in the suspect area readily showed the exact location of the pinhole. Figures 16 and 17 show the general location of the secondary pinhole leak. This leak was found in the overflow nozzle at the 8 o'clock position. It was located $\frac{1}{2}$ inch away from the circumferential butt weld joining the nozzle to the overflow pipe. This was an area where the corrosion of the external surfaces appeared to be severe. A close-up of the pinhole leak, Figure 18, shows the internal nozzle surface.

Dye penetrant and radiographic examinations were performed on the section removed from the tank. The existence of the pinhole, previously defined, was confirmed. In addition, a series of cracks across the vessel wall, in the region of the baffle were revealed by dye penetrant, Figures 19 through 24. Subsequent examination showed that these cracks radiated from the primary leak which shows clearly in Figures 20 and 21.

METALLOGRAPHIC EXAMINATION

Sections for metallographic examination were taken from two major areas: the baffle to vessel weld joint (the suspected primary leak), and the pipe to nozzle joint (the suspected secondary leak). These areas were selected to determine the cause of the primary and secondary failures.

During the sectioning and preparation of the metallurgical samples, the following observations were made:

1. The visible cracks in the section containing the baffle, fillet welds, and the vessel wall increased in width suggesting high residual stresses.
2. The cavity between the baffle and vessel wall contained a hard residue black in color. This residue when contacted with water or alcohol produced reactions typical of a sodium bearing deposit. Similar behavior was noted in those areas containing cracks. The residue filled cavity is shown in Figure 25, where a section through the fillet welded tee joints is represented.

Examination of the section containing the baffle, vessel wall, and fillet welds, Figure 26, revealed a massive network of cracks, principally transgranular in nature. The type of cracks discovered are typical of stress-corrosion cracking found in austenitic stainless steels. Figures 27, 28, and 29 show typical cracked areas. Further sectioning and examinations indicated that the entire area was riddled with this branch type cracking. All of the cracks originated at the surface of the previously described cavity and propagated from this cavity through the baffle, fillet welds and of greatest importance through the vessel wall. It was noted that sodium continued to leak from many of these cracks after metallurgical preparation and examination.

Nothing unusual in the microstructure was found. The baffle and vessel wall exhibited a typical austenitic microstructure with grain coarsening in the heat affected zones. As expected, the structure showed grain boundary precipitation typical of a sensitized austenitic stainless steel exposed to temperatures over 800°F for extended periods of time. A large quantity of non-metallic inclusions were found in the baffle and vessel material. However, their presence did not contribute to the failure, primarily because their orientation was normal to the direction of cracking.

Sections taken in the nozzle to pipe weld clearly showed that gross external attack had occurred, (see Figures 30, 31, and 32). The initial wall thickness of 0.310 in. was reduced to 0.005 in. thick. In the thinned area, the pinhole shown in Figures 17 and 18 was clearly evident.

CHEMICAL ANALYSES

A variety of materials were submitted for chemical analyses, where the objectives were to establish material identity and the composition of deposits and residue.

It was established that the vessel wall material was Type 304 stainless steel, the heat reflector shield and the heater bracket material was Type 405 stainless steel and that the heater sheath material was Incoloy. All of these materials were consistent with the design requirements. Analyses performed on the insulation material defined its composition principally

as a hydrated calcium silicate with iron, aluminum, and magnesium also present.

Samples taken from the mass of reaction products adhering to the vessel wall were analyzed. As expected, the principal constituent was sodium. Other major and minor constituents include: iron, chromium, nickel, manganese, silicon, aluminum, calcium, magnesium and slight quantities of molybdenum. All of the metallic elements existed as oxides and complex oxides.

The most revealing information was developed on that deposit taken from the cavity between the baffle and vessel wall. These results are shown in Table I along with the results of analyses performed on coatings taken from two welding electrodes. It will be noted that the composition of the coatings show a striking similarity.

TABLE I

CHEMICAL COMPOSITIONS

Deposit from Tank

1. Qualitative Analysis

Na (major), Al, Ca, Cr, Fe, Mn, Ni, Si, and Zr.

2. Quantitative Analysis

Chlorine: 500 ppm.

Fluorine: 5 wt %.

Coatings from Two Welding Electrodes

1. Electrode #1 (Mfg A).

a. Qualitative Analysis

Major Constituents: Ca, Mn, Na, Si.

Minor Constituents: Al, Fe.

Trace: Cr, Ti.

b. Quantitative Analysis.

Chlorine: 524 and 560 ppm.

Fluorine: 17.4 wt%.

2. Electrode #2 (Mfg M).

a. Qualitative Analysis

Major Constituents: Ca, Mn, Si, Ti.

Minor Constituents: Al, Cr, Fe, Na.

b. Quantitative Analysis

Chlorine: 208 and 300 ppm.

Fluorine: 5.8 wt%.

The most important elements found in the samples analyzed include chlorine and fluorine. Both of these elements are regarded as significant contributors to the stress corrosion of austenitic stainless steels.

With the exception of zirconium and titanium, the compositions of the materials examined are comparable. Since it is expected that zirconium and titanium would behave similarly in a weld rod coating, the observed difference (deposit vs electrode coatings) may be attributed to manufacturers preference.

STRESS ANALYSIS, HNPF EXPANSION TANK

The shell stresses were calculated for that region of the shell near the 8-in-diam. overflow nozzle. Conditions considered included the basic design, the normal operating conditions, the stiffening effect of the baffle and the overflow pipe support condition existing during operation. A stress of 135,000 psi was calculated, assuming no shell flexibility due to the close proximity of the baffle to the nozzle. The magnitude of the calculated stress is sufficient to warrant consideration as a factor contributing to the stress-corrosion failure discovered in the vessel wall.

CONCLUSIONS

It is concluded that :

1. The primary leak that developed in the HNPF Expansion Tank #3 was caused by stress-corrosion cracking. The cracking occurred in an area of high stress where a cavity, containing a highly corrosive deposit, existed. The deposit has been identified as residual flux from coated welding electrodes.
2. A secondary leak developed in the wall of the overflow nozzle as a result of gross attack by the sodium reaction products produced from the sodium released through the primary leak.

Figure 1: Model of expansion tank connected to the pump with the tank on the right. Principal pipe lines attached to tank include: 8" overflow line attaching pump to left side of tank; 14" outlet line from bottom of tank and the 14" inlet line. The failed and corroded area would correspond to the lower left portion of the tank around the 8" overflow line.

Figure 2: Region of expansion tank, near 8" overflow line showing heaters, brackets, heat reflector, insulation and aluminum skin.

Figure 3:

Tank No. 3 after removal of the aluminum skin and some of the insulation, away from the leak area. The 8" overflow line is shown near the top of the photograph. The reaction products and reacted insulation are evident.

Figure 4:

Sections of Type 405 stainless steel, heat reflector and bracket material, taken from the leak area. The highly corrosive attack resulted in the complete dissolution of the 0.065" thick material.

Figure 5: Reaction products built-up on the top portion of the overflow line. The tank heaters and the heater brackets form the grid work on the vessel wall.

Figure 6:

A bottom view of tank No. 3 showing the 14" outlet line adhering reaction products, heaters, brackets, heat reflector shielding and the tank supporting structure.

Figure 7:

Same view as shown in Figure 3 after removal of the reaction products. The overflow line is shown with a portion of the insulation, heaters and aluminum skin removed. Much of the heater bracket material in this area had dissolved.

Figure 8: Tank No. 3 removed from HNPf secondary system. Failed area containing 8" overflow line has been removed, exposing end view of baffle. The 8" nozzle and overflow line were contained in that portion to the right of the baffle. Tank supports, 14" inlet and outlet lines, are shown.

Figure 9:

Baffle section exposed after removal of failed vessel wall area, reference Figure 8.

Figure 10:

Section taken from vessel wall, left of the baffle as shown in Figures 8 and 9.

Figure 11. View of section removed from the center and right of baffle shown in Figures 8 and 9. This section shows the vessel wall, a portion of the baffle, and the 8" nozzle and pipe.

Figure 12: Section of the vessel wall, 8" nozzle and pipe showing the deep grooves produced by the sodium reaction products. Note the localized reduction in the width of the shell to nozzle fillet weld, left center.

Figure 13: View of section removed from the center and right of baffle shown in Figures 8 and 9. This section shows the vessel wall, a portion of the baffle, the 8" nozzle and pipe.

Figure 14: View of section removed from the center and right of baffle shown in Figures 8 and 9. This section shows the vessel wall, the 8" nozzle and pipe.

Figure 15: Pipe and nozzle interior surfaces showing inside surface of pipe, pipe to nozzle weld joint, tapered portion of nozzle containing circumferential tool marks and nozzle surfaces. These surfaces were uniform and clean.

Figure 16: Side view of vessel wall, 8" nozzle and pipe in failed area. Note square outlined on nozzle which locates the secondary pinhole leak.

Figure 17: Closeup of area in Figure 16 where area in square shows slight pinhole and apparent crack. Area below shows deformation produced by the mechanical removal of reaction products. Nozzle to pipe weld is apparent.

Figure 18: Closeup of pinhole and crack which developed in the 8" nozzle.

Figure 19:

Trimmed edge of vessel wall containing cracks; after addition of dye penetrant. Also note crack in baffle and fillet weld.

Figure 20:

Closeup of vessel wall containing primary leak. Shows outside surface and the cut edge of the vessel wall. Note cracks on cut edge.

Figure 21:

View of primary leak in vessel wall. Note cracks radiating to left edge. Mag: 2 X.

Figure 22:

Side view showing cracks through vessel wall, in fillet weld and in baffle. This is same section as that shown in Figure 21, rotated 90° to examine left edge. Mag: 2 X.

Figure 23:

View of section shown in Figure 21 after rotating 180°, to examine baffle. Mag: 2 X.

Figure 24:

View of section shown in Figure 21 after rotating 270°. Shows crack in baffle, (polished edge on right), through baffle into fillet weld. Note crack has not progressed to vessel wall, polished edge on left (this edge corresponds to the right edge of Figure 21). Mag: 2 X.

Figure 25: Cross section through vessel wall and baffle, approximately 2" below primary leak. Shows section of vessel wall (curved section), baffle, fillet welds, and cavity (porous material). Mag: 2 X.

Figure 26: Cross section through vessel wall and baffle, approximately 1" below primary leak. Note severe cracking, particularly the large crack in baffle section. Mag: $1\frac{1}{2}$ X.

Figure 27: Cracks in typical section of fillet weld. Note cracks originate from edge of cavity, dark zone. Mag: 50 X.

Figure 28: Typical cracks appearing at fillet weld/vessel wall joint. Cavity is black area in lower left. Mag: 50 X.

Figure 29: Typical stress-corrosion cracking pattern. Cracks are progressing through the vessel wall. The bottom edge corresponds with the edge of the cavity from where all cracks seem to originate. Mag: 50 X.

Figure 30:

Metallurgical mount containing transverse section through pinhole, Figure 18; support clip to position sample is in top portion of photograph. The shorter section shows the radii at the root of the tapered nozzle. The longer section shows deformation produced during mounting. In area of maximum thinning, the remaining wall thickness was 0.005". The right edge represents the outside surface. Mag: 4 X.

Figure 31:

Photomicrograph of the thinned wall, 0.005" thick, shown in Figure 30. External wall surface is the right edge, shows smooth corrosion of surface grains. The slight deformation, hook, shown in the lower left was produced during specimen preparation. Note grains do not show evidence of distortion. Mag: 150 X.

Figure 32: This section was taken from the nozzle/pipe weld joint area; 180° around the joint from the pinhole. The outside surface is the edge showing the maximum width of weld bead. The opposite edge shows the multi-step radii at the root of the taper in the nozzle. The degree of attack which occurred on the external surfaces can be obtained by comparing this section, where the wall thickness on the nozzle side of the weld is 0.310", with the sections shown in Figure 30, where a minimum wall thickness of 0.005" exists. Mag: 4 X.

GRAPHITE DILATION

INTRODUCTION

The Speer mold grade graphite moderator in the Hallam Nuclear Power Facility (HNPF) is clad with .016 inch thick Type 304 stainless steel to protect the graphite from interaction with the liquid sodium coolant (Figure 1). It has been established that graphite swells when contacted by liquid sodium, and the structural integrity of the graphite is lost due to intercalation and cracking of the graphite. From a neutron economy standpoint, a loss in reactivity would be experienced since the moderating efficiency of the graphite would be diluted by the presence of large quantities of sodium.

Previous sodium-graphite interaction studies employing unirradiated graphite indicated that swelling of the order of 1% might be expected (1). Measurements performed on a Sodium Reactor Experiment (SRE) moderator element in which the cladding had ruptured revealed, however, that the graphite had swelled by more than 5% in the radial direction (normal to the direction of extrusion). The SRE is a sodium cooled, graphite moderated (clad with zirconium base alloys) reactor similar in design to the HNPF. A dimensional profile developed for the failed SRE element (Figure 2) suggested that the expansion of the graphite was dependent, at least in part, on previous neutron irradiation. If the HNPF elements swelled 5% or more after a cladding failure, a failed element might rupture or score neighboring elements as well as introducing moderator element and fuel element removal problems.

When the SRE underwent modification for the Power Expansion Program (PEP) sodium logged type TS-163 graphite from an SRE element in which the cladding had ruptured was made available. Samples of irradiated TSP graphite from an unfailed SRE moderator element were also exposed to sodium in capsule experiments to determine the amount of expansion that might be expected when the irradiated graphite is contacted by liquid sodium. In addition, small samples of unirradiated Speer mold grade and TSP graphite were exposed to sodium to confirm the earlier results. After exposure to sodium, irradiated TSP samples were submitted for X-ray diffraction analysis to determine if a mechanism for the greater expansion could be established. The sodium logged TS-163 graphite samples were also submitted to X-ray diffraction for purposes of comparison.

PROCEDURE

For the static capsule tests for the unirradiated and irradiated graphite sodium studies, a capsule, as illustrated in Figure 3, was employed. Zirconium tabs were added to the sodium bath to getter oxygen, reducing the oxygen content of the sodium to less than 25 ppm.

Each graphite sample (1 x 3/4 x 1/2 inch) was placed in a 304 SS capsule, sodium was added, and the capsule was then welded shut. Loading and unloading operations were performed in an argon filled glove box. The capsules were transferred to a furnace, exposed for the times and temperatures indicated in Tables I and II, returned to the glove box and opened (Figure 4). The capsules were then heated to melt the sodium, facilitating graphite removal. Adhering sodium was scraped off for post-exposure measurements. Dimensional measurements were made with a micrometer caliper (Figure 5).

RESULTS

A. Unirradiated Graphite Dilatometry.

The table below gives the changes in bulk dimensions of the unirradiated graphite after sodium exposure.

TABLE I
SPEER MOLD GRADE

<u>Exposure Time (hrs.)</u>	<u>Exposure Temperature (°F)</u>	<u>%Expansion</u>		
		<u>a</u>	<u>b</u>	<u>c*</u>
1	600	0.1	0.3	0.1
100	600	1.0	1.0	0.6
600	600	0.5	0.8	0.5
100	800	0.9	1.0	0.5
600	800	0.8	0.9	0.7
1	1000	0.4	0.8	0.0
65	1000	1.0	1.0	1.0
600	1000	0.4	0.8	0.8
100	1200	0.8	1.0	1.2
600	1200	0.3	0.8	0.8
<u>TSP GRADE</u>				
100	1000	1.1	0.9	0.8
100	1200	1.0	0.1	1.0

*Axis of Extrusion

B. Irradiated Graphite Dilatometry

The sodium logged irradiated TSP graphite data is given in Table II below and presented graphically in Figure 6.

TABLE II

Accumulated Fast Neutron Flux (nvt)	Exposure Time (hrs.)	Exposure Temperature (°F)	% Expansion		
			a	b	c*
1.7×10^{19}	100	1000	2.4	2.5	1.4
1.7×10^{19}	100	1000	1.2	2.6	1.6
1.7×10^{19}	100	1000	2.4	2.5	2.1
6.7×10^{19}	100	1000	1.8	1.9	0.9
6.7×10^{19}	100	1000	1.6	2.6	0.7
7.3×10^{19}	100	1000	2.2	2.3	1.3
5.5×10^{20}	100	1000	2.4	2.8	2.0
5.5×10^{20}	100	1000	3.0	4.4	2.1
5.6×10^{20}	100	1000	1.6	1.6	1.3
5.6×10^{20}	100	1000	3.2	3.1	2.2
9.1×10^{20}	100	1000	4.2	4.0	2.8
1.09×10^{21}	100	1000	3.0	3.1	2.0
1.09×10^{21}	100	600	2.2	2.0	1.2
1.11×10^{21}	100	800	2.8	4.0	2.0
1.11×10^{21}	100	1000	4.3	4.4	2.6
1.11×10^{21}	100	1200	3.9	3.9	2.0
1.12×10^{21}	500	1000	3.8	4.3	2.6

* Axis of extrusion

X-RAY DIFFRACTION

Sodium logged samples of irradiated TSP and TS-163 graphite were subjected to X-ray diffraction analysis to determine changes in lattice structure as a result of neutron irradiation and sodium exposure. The TSP samples were irradiated in the SRE and exposed to sodium in laboratory capsule tests after removal from the reactor. The TS-163 graphite was exposed to sodium in-reactor due to a cladding failure.

X-ray patterns from the sodium logged TS-163 graphite revealed a graphite cell that was expanded as much as 11.6% along the c-axis (Figure 7), whereas there was a slight contraction (0.4%) along the a-axis. Since lamellar compounds of sodium and graphite have been observed in the past⁽²⁾, the patterns obtained from the TS-163 graphite were compared with patterns of sodium-graphite lamellar compounds but no evidence of a lamellar compound could be found. When the sodium-logged TS-163 graphite was exposed to air, the c-spacing reverted to only 3.3% residual expansion. Sodium-oxygen compounds were found after exposure to air. Since it was found that the bulk expansion did not change significantly with exposure to air, it was postulated that the sodium-oxygen compounds formed in the pores of the graphite, therefore inhibiting bulk dimensional recovery of the graphite.

The expansion of the graphite lattice along the c-axis was attributed to intercalation of sodium between the basal layers. Since the X-ray diffraction patterns exhibited a loss of intensity, it was postulated that the intercalation is probably random rather than systematic as is the case in lamellar compounds of graphite.

Although the lattice expansion in this material is similar to that attributed to irradiation damage alone the subsequent (on exposure to air) reversion of c-axis expansion from 11.6% to 3.3% suggests that sodium intercalation contributed to the c-axis expansion.

In order to compare irradiated graphite exposed to sodium in-reactor and irradiated graphite exposed to sodium in capsule tests, several samples of TSP graphite irradiated in the SRE and exposed to sodium in the laboratory were subjected to X-ray diffraction before and after exposure to sodium. The diffraction pattern of these samples exhibited only about 6% expansion of the c-axis. Again, no lamellar compounds of sodium and graphite were observed.

DISCUSSION

In Table I, it is shown that expansion of the order of 1% may be expected when unirradiated Speer mold grade or TSP graphite is exposed to liquid sodium in the 600 to 1200°F temperature range. Although some anisotropic expansion was in evidence in the unirradiated graphite, it was not nearly so pronounced as with the irradiated graphite. Both irradiated and unirradiated samples were sectioned in air to insure that sodium had permeated through the entire volume of the sample. The extent of sodium permeation was readily discernible when the samples were sectioned in air, as sodium could be seen bubbling forth from the pores of the graphite from the hygroscopic reaction of sodium with moisture in the air. No cracking or chipping occurred when the unirradiated graphites were exposed to sodium.

The results reported in Table II show that much larger expansion of the graphite was realized when the irradiated TSP graphite was exposed to sodium. Some cracking and corner chipping occurred in the irradiated samples during exposure. The cracking and chipping was attributed to the larger internal stresses developed during the greater expansion. A marked increase in expansion was noted in the irradiated graphite with increasing neutron dose over the neutron dose range investigated (1.7×10^{19} nvt to 1.12×10^{21} nvt). It was concluded that the expansion of graphite in liquid sodium is highly dependent upon the amount of previous neutron irradiation to which the graphite has been exposed, but not highly sensitive to temperature over the 800-1200°F.

By referring to Table II or Figure 5, it may be seen that the expansion of the irradiated graphite is anisotropic, with expansion in the direction of extrusion less than expansion normal to the direction of extrusion. This is not unexpected since the greater expansion of the irradiated graphite is attributed to expansion of the c-axis, and since the extrusion process tends to align the c-axis normal to the direction of extrusion.

On the basis of both the bulk dimensional changes and the X-ray diffraction results, it appears that during exposure of unirradiated graphite to sodium, the sodium simply permeated the pores of the graphite resulting in relatively low expansion. In the case of the irradiated graphite, however, a greater expansion in bulk dimensions occurred due to random sodium intercalation of the basal layers.

CONCLUSIONS

1. Unirradiated extruded graphite expands about 1% when exposed to sodium in the 600 to 1200°F range.
2. The expansion of irradiated graphite in sodium increases with increasing accumulated neutron dose, but the expansion is not temperature dependent upon sodium exposure temperatures between 800 and 1200°F.
3. Bulk expansion up to $\sim 5\%$ was found when highly irradiated ($\sim 10^{21}$ nvt) TSP graphite was exposed to sodium.
4. The greater expansion of the irradiated graphite upon exposure to sodium was attributed to sodium intercalation of the basal layers expanding the c-axis.
5. The bulk dimensional expansion of the irradiated graphite in sodium is anisotropic, greater expansion being observed normal to the direction of extrusion.
6. The c-spacing lattice dimensions partially recover when sodium-logged graphite is exposed to air, but the bulk expansion remains essentially constant due to the formation of sodium-oxygen compounds.

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Figure 1. HNPf Moderator Element
 (from Left to Right, a Perspective, a Cutaway, and a Diagram
 Showing Relation of Moderator Element to Other Core Components).

Figure 2. Failed Moderator Element Dimensional Profile.

Figure 3. Test Capsule for Soaking Test to Determine Sodium Pick-Up.

Figure 4. Removing Sodium Soaked Graphite From Test Capsule in Glove Box.

Figure 5. Post-exposure Measurements After Removing Adhering Sodium.

Figure 6. Sodium Swelling of Irradiated Graphite as a Function of Integrated Dose and Axis.

Figure 7. Crystal Lattice of Graphite.

FERMI MATERIALS EXPERIENCE

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I. INTRODUCTION

A detailed description of the various Fermi components, together with the materials used, has been presented elsewhere.¹ Included in that paper was a discussion of the materials experience obtained as of that date. In this paper, an attempt has been made to cover the Fermi experience since that time. In particular, emphasis is placed on sodium compatibility experience in the Fermi plant, under-sodium bearing experience, and boronated graphite irradiation test work.

II. SODIUM COMPATIBILITY EXPERIENCE

A. SODIUM PURITY AND THERMAL HISTORY

During the Reactor Components Test program the primary system of the Fermi reactor was filled with sodium on November 30, 1960. The reactor vessel and primary system were maintained at an isothermal condition of 500 F through May 1, 1961. At that time, the system temperature was heated over a period of 10 days to 1000 F, maintained at that temperature for 7 days, and then decreased in steps to 550 F.

At the start of the 1000 F isothermal test, the sodium plugging temperature was 240 F. When the system temperature reached 950 F, the plugging temperature started rising and reached a peak of 560 F when the system temperature reached 1000 F. Continuous cold trapping subsequently reduced the plugging temperature to 240 F. The plugging temperature increase was attributed to outgassing of the graphite in the reactor plug and subsequent reaction of the gases with the sodium to form free carbon and sodium carbonate. This reaction resulted in the formation of carbon and oxygen impurities in the sodium, as well as a surface layer of solid reaction products. A chemical analysis of the solid reaction products indicated that it consisted primarily of carbon, sodium carbonate, and sodium hydroxide as shown in Table I.

TABLE I - ANALYSIS OF SOLID REACTION PRODUCTS

<u>Constituent</u>	<u>Per Cent, by Weight</u>
Carbon	20.6
Na ₂ CO ₃	47.6
NaOH	31.7

Analysis of the sodium cover gas, taken during the 1000 F run, showed high percentages of carbon dioxide and oxygen. Subsequent outgassing tests on graphite that was similar to that in the plug indicated that hydrogen in the form of methane and moisture was also present, indicating that the sodium also would have become contaminated with hydrogen.

In June 1962, the sodium was drained from the primary system in order to affect repairs to several reactor components. The system was refilled in November 1962, and has been maintained at temperatures of 400 F and higher since that time. The thermal history of the primary sodium system is shown in Figure 1.

The oxygen, carbon, and hydrogen impurities in the sodium for samples taken periodically since December 1963 are shown in Table II,

TABLE II - AVERAGE ANALYSES OF FERMI SODIUM
DECEMBER 1963-MARCH 1965

<u>Lab Sample Number</u>	<u>Fermi Coil No.</u>	<u>Date of Sample</u>	<u>Total^a Oxygen, ppm</u>	<u>Total Carbon, ppm</u>	<u>Carbonate Carbon, ppm</u>	<u>Total^b Hydrogen, ppm</u>
86	6	12-63	25	37	8	-
104	8	3-64	27	41	3	-
143	9	6-64	13	43	6	11
149	10	7-64	12	38	7 ^c	3
151	11	8-64	10	59	4	7
161	12	9-64	15	43	7	2
200	13	10-64	21	31	8	3
351	14	1-65	16	50	3	4
428	15	2-65	14	40	1	8
503	16	3-65	12	48	2	3

a. Oxygen calculated as sodium monoxide.

b. Results were obtained by alcohol dissolution method and are too high.

c. The accuracy of the method is not yet established, and total hydrogen is hydrogen exclusive of hydrogen in sodium hydroxide. Since the oxygen level is always low, the amount of hydrogen which could be present as hydroxide hydrogen is not more than 0.5 ppm.

B. GENERAL CORROSION

There has been no evidence of corrosion on any components examined to date that could be attributed to exposure to sodium. This is not surprising, however, due to the short times that the system has been at high temperatures and that the system has not been subjected to any temperature gradients.

However, there has been evidence of carburization of the stainless steel in the primary system and of hydriding of the zirconium cladding on the fuel pins.

C. CARBURIZATION

During the shut-down period in 1962, several components were removed from the primary system and examined to determine whether carburization had occurred. These components were in the system throughout the nonnuclear test program. The results of these examinations are listed in Table III.

TABLE III - DEGREE OF CARBURIZATION OF
FERMI PRIMARY SYSTEM COMPONENTS

<u>Component</u>	<u>Depth of Carburization, in.</u>	<u>Carbon Content, w/o</u>
Check Valve	0.005	Not analyzed
Support Plate Orifice Retaining Ring	0.001	" "
Dummy Subassembly		
1. Stainless steel filter screen, 0.004 in. wire	Complete	0.5
2. Backup screen	0.005	0.18
3. Wrapper tube	0.0005	0.3

Surface sample - 0.003 inch thick

The microstructure of the check valve is shown in Figure 2. This minimal degree of carburization is not considered detrimental to any of the reactor system components. However, continued carburization is undesirable, particularly with respect to thin, highly-stressed sections such as bellows. For this reason, a program for monitoring the carburization potential of the system is currently underway.

In this monitoring program, the sample exposure station is in the sodium inlet line to the hot trap, downstream of the economizer. The heating capability of the hot trap and the economizer is used to heat the sodium to the desired exposure temperature. Primary sodium is pumped from the overflow tank of the reactor through the exposure station at flow rates of 2 to 5 gpm.

The monitoring specimens are type 304, round stainless steel bars, 3/4 of an inch in diameter and 5 inches long, machined from completely annealed stock material. The bars are provided with nipples on the ends for machining after sodium exposure and for holding the sample bars in the test section. Before exposure, the specimens are electrochemically polished (4 mils removed on the diameter) to remove effects of cold working resulting from machining; this step was first utilized after Run 1. Six of the test bars are held in the 3-inch-diameter test section by spider supports which allow free flow of sodium around the bars. The test bars and sample station in various stages of assembly are shown in Figure 3.

The exposure time of the runs is determined as the number of equivalent hours at 1200 F, as follows:

$$t_{1200} = \frac{D_t}{D_{1200}} (t_T)$$

where:

t_{1200} = equivalent time at 1200 F, hours

t_T = actual time at operating temperature, hours

D_T = carbon diffusion coefficient at temperature T

D_{1200} = carbon diffusion coefficient at 1200 F

Carbon diffusion coefficients reported at Al^2 are used. Only exposure times above 1000 F are used in this calculation because of the relatively small carbon diffusion coefficient below 1000 F. Temperature data are recorded approximately each 4 hours during a run.

After exposure, the sodium is removed from the bars by alcohol immersion, and 1-mil samples are machined off the bars for chemical analysis. The first three samples are used to determine the surface carbon content. An additional 3 mils are turned off the diameter to remove exposure effects and are discarded. A fourth 1-mil sample is then obtained which provides information on the carbon content of the bars before exposure. Samples are also taken from each bar for metallurgical examination.

The bars are electrochemically polished and then reused in additional exposure tests.

The samples are analyzed chemically by a combustion technique using National Bureau of Standards samples of known carbon content for control purposes.

The surface carbon content of the specimens is estimated from the results of the chemical analysis by both graphical and analytical methods. The graphical method consists of extrapolating the carbon content to the surface on a plot of the log of the carbon content versus average sample depth. The equation for unidimensional diffusion³ is used as the analytical method. Mill reports of the stock material and results of chemical analysis of the unexposed bar material are used for the value of the initial carbon content in the diffusion equation.

Results from the three runs completed to date are shown in Table IV.

TABLE IV - SURFACE CARBON CONTENT OF TYPE 304
STAINLESS STEEL SPECIMENS EXPOSED TO
FERMI PRIMARY SODIUM
(INITIAL CARBON CONTENT: 0.06 PER CENT)

Run No.	Equivalent Time at 1200 F, hours	Exposure Period	Surface Carbon Content, Wt. Per Cent	
			Graphical Method	Diffusion Equation
1	77	June 1964	0.16 ^a	0.12 ^a
2	125	Feb. 1965	0.22 ^a	0.19 ^a
3	148	March 1965	0.07 ^b	-

a. Average of 4 bar specimens from Runs 1 and 2

b. Average of 2 bar specimens for Run 3

The surface carbon content in all cases has been well below 0.5 per cent. The results from the third run indicate that essentially no carburization potential currently exists in the sodium. Since the exposure tests require the use of the hot trap, carbon is removed during each test. Possibly, the carburization potential of the sodium has been lowered, although the carbon content in the sodium does not appear to have changed significantly.

The highest surface carbon content of 0.19 to 0.22 weight per cent was observed during the second run. However, the maximum depth of carburization was only about 2 mils.

Shown in Figure 4 are photomicrographs of specimens from the second and third runs. These figures show carburization to a depth of about 2 mils during the second run, and essentially no carburization occurring during the third run.

The results obtained to date with the 100-hour exposure tests indicate little carburization potential exists. However, a series of runs are now in progress for longer periods of exposure. Specimens from these runs will be removed after 300, 500, 800, and 1000 hours of exposure. Results from these longer tests should provide a positive assessment of the carburization potential of the primary sodium of the Fermi system.

D. ZIRCONIUM HYDRIDING

Prior to manufacture of the Fermi Core A fuel, U-10 w/o Mo alloy fuel pins clad with 0.005-inch zirconium, two demonstration runs were made with depleted uranium metal. The first of these runs consisted of 500 pins, the second of 1000 pins. These pins were subsequently used to build two proof-test (PT) subassemblies to be installed in the reactor core prior to loading the enriched fuel, for the purpose of monitoring the condition of the fuel after long time exposure to high sodium flow conditions. These subassemblies were installed in the reactor in November 1962.

PT-1 was removed in July 1963 after being exposed 5860 hours. Metallographic examination of the cladding showed little or no effect resulting from the sodium exposure. The microstructure of pins from each of the trial runs showed only faint traces of zirconium hydride as shown in Figure 5, the hydrogen content being estimated at less than 80 ppm. Vacuum fusion analysis of the zirconium end caps showed hydrogen concentrations of 32 and 75 ppm, again representing little or no increase.

PT-2 was removed from the system in November 1964, after 17,500 hours of continuous sodium exposure. Examination of one pin from each of the trial runs has shown varied and extensive hydrogen pick-up, as shown in Figures 6 and 7. On the pin from the first trial run, the hydrogen appears as zirconium hydride needles primarily orientated normal to the cladding (Figure 6). A vacuum fusion analysis showed a hydrogen content of 333 ppm. The pin from the second trial run had a hydrogen content of 2070 ppm; the precipitate appearing as a nearly continuous grain boundary network, Figure 7. Examination of other pins from PT-2 subassembly is in progress.

A review of the literature indicates that for hydrogen concentrations up to about 5000 ppm, the embrittling effect on Zircaloy-2 is not extensive at temperatures in excess of about 500 F.⁴ However, there is evidence of an increase in the nil-ductility temperature under impact conditions. For this reason, thermal shock tests were run in sodium. The fuel pin specimens used had hydrogen concentrations estimated to be 70, 250, 500, and 1000 ppm. The microstructure of the lower concentrations showed the needle-like precipitate, whereas that of the higher concentration showed the grain boundary precipitate. In these tests, the fuel-pin specimens were heated in argon to 1000 °F and quenched in 500 °F sodium. There were no adverse affects after 100 such thermal shocks.

E. OTHER ASPECTS OF PROOF-TEST SUBASSEMBLY INSPECTIONS

Visual examination of many pins from the PT-1 subassembly showed that the cladding had pitted at the region of contact with the stainless steel support grids. These pits or indentations were oval in shape with their long axis in the

direction of the fuel pin axes. They ranged in size to a maximum of a 0.035-inch-major axis and a 0.025-inch-minor axis. In some, there was evidence of zirconium metal pullout to a depth of 0.004 inch, as shown in Figure 8. Such pitting and metal pullout has not been observed on pins from the PT-2 sub-assembly.

It is now concluded that the pitting was primarily the result of a dimensional stability test that was run on the subassembly prior to insertion into the reactor system. In this test, the subassembly was heated in vacuum for 10 hours at 800 F followed by 10 hours at 1100 F and subsequently checked for dimensional stability. It is believed that during this heating, diffusion occurred between the steel grid and the cladding that lead to the observed pitting. Were this not the case, PT-2, with its considerably longer exposure period, would most certainly have shown comparable or greater pitting.

As a result of the long exposure to the hot sodium, some transformation of the U-10 w/o fuel alloy from the metastable gamma phase to the thermally stable alpha plus epsilon phases had occurred, as can be seen in Figures 6 and 7. The different degree of transformation reflects the faster cooling achieved following the heat-treatment of the second trial pin run relative to that of the first run.

III. PERFORMANCE OF OSCILLATOR ROD BEARING

A. LOW-POWER OSCILLATOR ROD

The oscillator rod assembly consists of two sections: the poison section located in the core, and the drive shaft section which extends from the top of the core up through the sodium, the cover gas, and through the plug.

1. Poison Section

The poison section has two bearings; an inverted pivot bearing at the lower end which supports the poison rotor (the pivot is stationary and the socket rotates) and an upper bearing (sleeve type) about 51 inches above the pivot bearing.

The pivot bearing consists of a spherical socket, 1 inch in radius, made of Kenna-metal 138-A (Titanium Carbide Cermet) provided with "oil" holes to permit the circulation of sodium. The mating pivot, which has a 1 inch radius spherical contour (lapped fit to the socket) is made of Kenna-metal K-95 (Tungsten Carbide Cermet) provided with radial "oil" grooves to permit continuous flooding of the bearing surface with sodium. The rotation of the socket develops a pumping action which draws sodium across the surfaces of the pivot bearing during operation.⁵

The poison section sleeve bearing is 2 inches in diameter and 1 inch long. The sleeve is a fused Colmonoy No. 70 overlay on a Type 304 stainless steel backing, and the journal is a fused Colmonoy No. 4 on the rotor shaft; the sleeve has a ground finish ~ 32-microinch RMS, and the journal

a turned finish ~ 64-microinch RMS. The journal has four grooves, 90° apart, for sodium flow. The diametral clearance between the sleeve and the journal is 0.005 to 0.007 inch.

2. Drive Shaft Section

The drive shaft section has four sleeve-type bearings supporting the shaft. The two lower bearings, like the poison section bearing, operate under sodium; and the two upper bearings operate in the argon sodium-vapor cover gas over the sodium in the reactor vessel.

The sleeves of the two lower bearings are made from Stellite 6B, and the journals are a Colmonoy No. 4 fused overlay on the drive shaft. Both the journal and the sleeve were machined to a finish finer than 125-microinch RMS, with a diametral clearance of 0.007 to 0.009 inch.

The sleeves of the two upper bearings are made of aluminum bronze, Ampco No. 22, and the journals are a Colmonoy No. 5 fused overlay on the drive shaft. The bearing surfaces were machined to a finish finer than 125-microinch RMS and the diametral clearance of the sleeves and journals was 0.004 to 0.005 inch and 0.009 to 0.010 inch.

3. Test Performance

After a total of 75 hours of test operation in sodium at rotational speeds of 300 to 600 rpm, with about equal times in 500 F and 750 F sodium, and an additional 10 hours at a reduced minimum speed of 0.06 rpm., the oscillator rod assembly was cleaned and disassembled for examination of the bearing.

All the bearings were found to be in excellent condition. The socket of the pivot bearing had developed a bright burnished finish on the bearing area (as did, presumably, the matting pivot which was not removed from the assembly). The sleeve bearings also had developed a slight burnished finish on the bearing contact areas. No evidence of galling or scoring was found on any of the five sleeve bearings. The two Colmonoy No. 5 journals on the drive shaft did exhibit some short hairline cracks, but no spalling of the hard facing material occurred.⁶

B. HIGH-POWER OSCILLATOR ROD

The drive shaft section used for the low-power oscillator rod will also be used for driving the poison section of the high-power oscillator rod.

1. Poison Section

The poison section of the high-power oscillator rod differs mechanically from the low-power section in that it has an additional sleeve bearing that gives lateral support, about 5 inches above the pivot bearing.

The sleeves of both the upper and lower sleeve bearings are fused Colmonoy No. 4 overlays on a Type 304 stainless steel backing, and the journals are fused overlays of Stellite 6 on the shaft. Both the sleeves and the journals were machined to a finish finer than 125-microinch RMS. The journals have three equally spaced grooves parallel to the axis to permit access of sodium to the bearing surfaces; the diametral clearance of the journals and sleeves are 0.004 to 0.005 inch.

The pivot bearing is identical in design and material to that of the poison section of the low-power oscillator rod.

2. Test Performance

The poison section was test operated in sodium at 600 F for a total of 96 hours in accordance with the following sequential rotational velocity cycles.

<u>Velocity, RPM</u>	<u>Cycle Time, Min.</u>
6	60
6	60
12	15
30	15
60	15
120	15
300	15
600	30
6	60
6	60
6	60
6	60

There was a 1-minute stop interval between each of above cycles of rotation. The above series of cycles was repeated without stopping until a total of 96 hours of test operation had been completed.

The two sleeve bearings and the pivot bearings were found to be in excellent condition at the completion of the 96-hour test as shown in Figure 9. The pivot bearing surfaces developed a smooth, polished finish with no measurable wear, and the sleeve bearings became slightly burnished and showed no evidence of galling or scoring.

IV. IRRADIATION STUDIES ON BORONATED GRAPHITE

The primary shield of Fermi was described previously¹ and the function of the boronated graphite portion of the shield discussed in considerable detail. To briefly review, there is a 6-inch inner layer of nominally 5 w/o boronated graphite (boron added as boron carbide (B_4C)) surrounding the reactor vessel. Its main function is to absorb low energy neutrons which escape from the reactor vessel. Considerable heat is generated in this inner boronated layer which is transferred back into the reactor by a layer of thermal insulation external to the 6-inch layer. An irregularly shaped region of unboronated graphite lies in areas outside of the thermal insulation. Piping, which passes through this region, is surrounded by rings of boronated graphite to decrease the neutron streaming. These graphite rings have a nominal boron concentration of 7 w/o. Surrounding the unboronated portion of graphite is another boronated graphite region which captures those neutrons which are thermalized in the plain graphite region.

The region of principal concern with regard to radiation stability is the inner, 6-inch boronated graphite region, although there is concern with the boronated rings which surround the reactor piping. The inner boronated layer was originally designed to accommodate an irradiation-induced nominal shrinkage of 2-1/4%, with a local maximum shrinkage of 6-3/4% and an expansion of 1/2%, by using corrugated straps which serve as springs to hold the graphite blocks in tight contact. The as-fabricated system has been reviewed and it has been concluded that expansion values of ~ 2% can also be accommodated without detrimental effects to the reactor, the coolant piping, or the primary shield tank. Although the dimensional stability was of prime concern, another important criterion was the compressive strength of the material in that it is required to support itself during irradiation.

Normally, the damage to graphite is primarily caused by fast neutron interactions with carbon nuclei; however, with boronated graphite the material is not only damaged in this manner, but also by recoiling helium and lithium nuclei. The nominal values for the helium generation rate in the inner boronated layer, at a reactor power of 200 Mw(t) has been calculated to vary from 0.3 to 1.0×10^{11} α 's/cm³-sec at the core axial centerline. The fast neutron flux ($E > 0.18$ Mev) decreases somewhat from a value of $\sim 2.0 \times 10^{12}$ nv at the inner boronated graphite surface as the neutrons pass through the blocks. The calculated temperatures are in the range of 600 F to 1000 F in the inner boronated graphite region at a reactor power of 200 Mw(t).

To study the effect of irradiation on boronated graphite used, APDA and PRDC initiated a program at the Hanford Atomic Power Laboratory (now Battelle-Northwest) under the auspices of the USAEC. In this phase of irradiation testing, samples were irradiated in three long term capsules. Each capsule was temperature monitored and had two sections which operated at ~ 930 F and 700 F, respectively. Within each section there were about 14 samples or 42 per capsule. The samples were irradiated in a helium atmosphere. The tests were designed in a manner that would allow some samples from the first capsule to be irradiated in the subsequent capsules. Some new specimens were added after each irradiation to replace specimens removed for destructive testing. In this part of the program, samples were to be irradiated in an extremely high thermal neutron flux to enhance damage in the boronated graphites, since it was felt that there was considerable solid solution of boron in the boronated graphites caused by high

graphitization temperatures. In fact, it was found that during production, material of a given boronation appeared to have two different appearances, a "gray" metallic-like appearance and a "black" appearance. J. M. Davidson and L. O. Gates,⁷ have postulated that the gray material reached the melting point of the $B_{11}C$ particles during graphitization, promoting recrystallization of the graphite and allowing more uniform dispersion of the boron within the graphite matrix. The black material, on the other hand, had the boron distributed as particles which retained much of the n, α damage produced within the particles, although it is suspected that there was some boron in solution even in the black material.

The black 5 w/o boronated graphite samples exhibited length contraction after irradiation at 700 F and 930 F in a direction parallel to the extrusion direction; the gray 5 w/o boronated samples, which had the same orientation, first contracted and then expanded. In the transverse direction the black samples expanded at 700 F; however, at 930 F some samples contracted while others expanded. The gray samples, on the other hand, initially contracted and then expanded at both irradiation temperatures, more at 700 F than at 930 F. At 930 F, the temperature closest to which the maximum neutron exposure will occur in the EFR, the maximum length change for the black material after a thermal neutron exposure of 2.5×10^{21} nvt_{th} (3.1×10^{20} nvt_f) was -0.75% in the parallel direction and + ~ 0.4% in the transverse direction. At the same neutron exposure, the gray, 5 w/o boronated material exhibited growth values of less than 0.5% in the parallel direction and less than 1.5% in the transverse direction. At the maximum exposure it is estimated that the B-10 burnout was greater than 50%.

J. M. Davidson and L. O. Gates⁷ also report changes in thermal resistivity, coefficient of thermal expansion, and compressive strength properties. Summarizing their results at the same irradiation conditions given above, it can be stated that both 5 w/o boronated materials increase in compressive strength by greater than 100% in both directions after irradiation. The thermal resistivity (measured at room temperature) increased by about a factor of 13 in the parallel direction and 17 in the transverse direction for both the gray and black material. The coefficient of thermal expansion between room temperature and 400 F also increases significantly in both orientations, although the scatter is quite large. J. M. Davidson and L. O. Gates⁷ describe an auxiliary program conducted on EFR materials which were irradiated in boron stainless steel thermal neutron shielded capsules in the ETR. In these tests, samples were exposed to a thermal neutron flux of up to 5.6×10^{19} nvt_{th} and 1.2×10^{21} nvt ($E > 0.18$ Mev) at temperatures similar to those described earlier. In these tests, the black and gray 5 w/o boronated graphites contracted in both directions. The black < 0.2% in the transverse and < 0.9% in the parallel direction, and the gray < 0.8% in the transverse and < 1% in the parallel direction.

If one assumes a 100% plant factor at 200 Mw(t) for 10 years, the maximum fast nvt ($E > 0.18$ Mev) is ~ 7×10^{20} nvt and the boron burnout is < 5% in the six-inch-inner boronated graphite region. It, therefore, can be seen that in the first set of irradiations cited the thermal neutron exposure, or n, α damage to the graphite far exceeded Fermi design conditions, while the fast neutron exposure was about one-half that anticipated for the EFR. In the shielded experiments described, the fast neutron exposure was about twice that anticipated in Fermi, while the thermal neutron exposure is assumed to be somewhat greater than expected in Fermi. Confirmation of this fact by chemical analysis is presently being done.

Since the dimensional changes observed in both irradiations were within design limitations and the compressive strength values increased, it has been tentatively concluded that the Fermi design is adequate from a dimensional stability standpoint. This conclusion is tentative since theoretical studies are to be done to correlate n , Δ damage and fast neutron damage obtained in these studies, and in the graphite for Fermi flux and spectrum conditions.

V. NEUTRON SOURCE

The neutron source for Fermi used to provide the necessary neutron flux for start up, is composed of a replaceable antimony rod that fits inside a beryllium assembly in the reactor. The antimony portion of the source must be irradiated in another reactor before use in Fermi. Thus far, considerable difficulty has been encountered with the fabrication and post-irradiation handling of the antimony section. It is the purpose of this section to describe the difficulties that have been encountered and in this way aid in future sodium-cooled fast reactor neutron source design and operation.

The antimony rod is about 0.7 inch in diameter and 25 inches long and is contained in a tantalum tube seal-welded in a helium atmosphere. Originally, the neutron source was designed for 300 Mw(t) operation and, conceivably, the antimony could have melted under abnormal reactor conditions. The choice of tantalum cladding for the antimony section was made on the basis of corrosion tests performed on readily available materials in a liquid-antimony environment. The tantalum also proved satisfactory for sodium operation.

The initial difficulty arose after the first antimony section of the source had been irradiated in another reactor. Upon receipt at the Fermi site, the source was immersed in a water leak detection can. When antimony activity was discovered in the water after allowing the section to remain in the water for several days, it was concluded that there might be a leak in the tantalum cladding. Originally, the tantalum specification required an arc-cast material to insure good weldability. The welds were suspected and, when checked, it was discovered that the vendor had used a powder metallurgy product that would be expected to have more inclusions and hence be more difficult to weld. Since it is extremely difficult to leak test the antimony section to the degree of accuracy needed prior to irradiation, all subsequent antimony enclosed tantalum sections were fabricated of either arc-cast or electron-beam melted stock. Since that time, difficulties of this nature have not been encountered with additional antimony sections.

Another source of difficulty has been the post-irradiation handling of the antimony rod. Because of the highly radioactive state of the rod, a large amount of heat is generated and considerable time must be allowed and/or adequate provisions must be made for cooling the rod. One of the antimony rods was removed from the vendor's pool, where it was being cooled after irradiation, and placed in a cask for about 2 days. Subsequent examination in the vendor's hot cell facility showed that about 0.010 inch of the wall thickness apparently had been oxidized and removed from the tantalum cladding. Since the tantalum was extremely radioactive, the vendor's cask and hot cell were considerably contaminated. If sufficient cooling time or adequate cooling had been provided, this problem would not have been encountered as it has since been established that the tantalum temperature was in the rapid oxidation range.

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Fig. 1 THERMAL HISTORY OF FERMI PRIMARY SYSTEM

Fig. 2 CARBURIZATION ON THE OUTSIDE SURFACE
OF THE PRIMARY SODIUM PUMP CHECK VALVE
MAG. 250 X

CARBURIZATION TEST SPECIMEN DISASSEMBLED

CARBURIZATION TEST SPECIMEN ASSEMBLED
BESIDE PIPE TEST SECTION

CARBURIZATION TEST SPECIMEN ASSEMBLED
IN PIPE TEST SECTION

Fig. 3 TEST SPECIMENS AND SAMPLE STATION USED
IN CARBURIZATION MONITORING PROGRAM

Run No. 2 - 125 hours at 1200 F.
Note slight degree of surface
Carburization

Run No. 3 - 148 hours at 1200 F.
Note complete absence of carburization

Fig. 4 MICROSTRUCTURES OF CARBURIZATION
TEST SPECIMENS
Mag. 250X

Fig. 5 CLADDING ON DEPLETED URANIUM
FUEL PIN FROM PT-1
Mag. 250X

Fig. 6 CLADDING ON DEPLETED URANIUM FUEL PIN
FROM PT-2 SUBASSEMBLY AFTER 17,500
HOURS EXPOSURE TO FERMI SODIUM
(PIN MADE IN FIRST TRIAL PIN RUN)
Mag. 250X

Fig. 7 CLADDING ON DEPLETED URANIUM FUEL PIN
FROM PT-2 SUBASSEMBLY AFTER 17,500
HOURS EXPOSURE TO FERMI SODIUM
(PIN MADE IN SECOND TRIAL PIN RUN)

Mag. 250X

Fig. 8 METAL PULLOUT OF THE ZIRCONIUM CLAD
ON A FUEL PIN FROM THE PT-1 SUBASSEMBLY

Mag. 100X

Fig. 9 PIVOT BEARING SHOWING POLISHED
WEAR PATTERN IN SOCKET AND BALL
(FULL SIZE)

EVALUATION OF THE PERFORMANCE OF MATERIALS IN SODIUM IN THE SRE*

By D.A. Mannas

ATOMICS INTERNATIONAL

The Sodium Reactor Experiment (SRE) is a sodium-cooled, graphite-moderated reactor power system constructed to provide technology for large sodium-cooled systems. Detailed descriptions of the reactor are available in the literature^{1,2,3}. Since the initial operation in March of 1957, the sodium system has accumulated 44,000 hours of sodium flow at temperatures from 300°F to 1030°F. In July of 1959, fouling occurred in the core when tetralin, an organic fluid used to cool the sodium pumps, leaked into the system and decomposed producing carbonaceous deposits. During a 12-month recovery period, several moderator cans were replaced, and the second SRE core (Core II) was installed. In February of 1964, modification of the system was initiated to permit operation at higher temperature and power levels. Since then, the entire primary system has been replaced. An examination of the material removed is being conducted to determine the effect of a sodium environment on a large heat-transfer-system.

The primary system is illustrated in Figure 1. Each line and component was examined visually as it was removed from the system. Most of the system was also visually examined after the residual sodium had been removed. A few typical items are shown in Figures 2,3, and 4. No evidence of erosion or wear has been observed.

Several items from the system have been examined in detail. They are categorized into hot-leg and cold-leg groups with the reactor core and the main intermediate heat exchanger (MIHX) separating them. Although there were 56 reactor scrams on Core I and 43 on Core II which produced temperature transients in the system, the hot leg and the cold leg sections normally were run under essentially isothermal conditions.

Hot Leg

The general integrated thermal and flow history of the hot leg is outlined in Table I. A section of the main primary line, type 304 stainless steel, 6-in.-diam., sched. 40 pipe, from the core outlet region is shown in Figure 5. The pipe wall in this area was heavily sensitized, exhibited fine grain size, and showed a light carburized case to a depth of 0.005 inch along the surface exposed to the sodium, Figure 6. Slight pitting along the surface may indicate some corrosion has occurred, but it is clearly negligible.

A saddle tee in the main primary line for the primary drain pit was subjected to thermal stresses because of the low temperature of the sodium in the connecting 2-in.-diam. line. Dye penetrant examination did not reveal any cracks on the surfaces of the tee that had been exposed to the sodium. Analyses of the slight deposit along the line, Figure 7, are listed in Table II. Typical cross sections of the main primary line and the weld joint in the tee are shown in

*Work performed as a part of Contract ESADA-1A with the ESADA group of investor-owned utilities companies in New York State.

SRE MAIN GALLERY

Figure 1. Diagram Showing Location of Samples in SRE Primary Sodium System

Figure 2. One inch line (arrow) from a thermal stress reducing tee which extended into the sodium stream in the 6-in.-diam., main primary hot leg. No erosion has occurred.

Figure 3: Main primary pump impeller with original balancing, grinding marks, unaffected by service.

Figure 4. Cross Section of MIHX Tube Bundle where cold primary sodium was in tubes and cold secondary sodium entered around outside of tubes. All tubes were essentially free of deposits except those in upper left section which was in vicinity of secondary cold-leg line inlet.

TABLE I

Integrated Thermal and Flow History of SRE Hot Leg*

Temperature Range (°F)	Flow Time in Hours**	
	Core I	Core II
300	120	180
300 - 399	4,080	9,480
400 - 499	2,016	3,288
500 - 599	576	4,008
600 - 699	192	6,408
700 - 799	520	2,256
800 - 899	1,972	1,056
900 - 959	512	40
960 -1030	356	0
TOTAL:	10,344	26,716

* Interval: Core I - May 4, 1958 to November 10, 1959
Core II- July 22, 1960 to February 15, 1964

** Flow: Mean Flow 1100 ± 200 gpm (12.5 ± 2 ft/sec in 6-in. lines) with 6000 hr of Core II time at 1450 to 1560 gpm (16 to 17.5 ft/sec in 6-in. lines).

Figure 5. Section of 6-in.-diam. hot leg line removed from adjacent to the core outlet.

Figure 6.

A longitudinal section of the line above. The lightly carburized case at the surface averaged 280 Khn (100 gm. load) which dropped off to 180 Khn (100 gm. load) 0.005-in. below the sodium exposed surface as indicated by the microhardness indents. Etch: Ferric Chloride Mag: 150 X

Figure 7. Sodium exposed surface of 6-in.-diam. hot leg line with the thermally stressed tee connection (arrow) for the 2-in.-diam. line from the primary drain pit. The tee was at the top of the line in the system, and the film deposits were present after cleaning.

TABLE II

Analysis of Deposits from the Hot Leg at the Thermally Stressed Tee

Carbon Analysis (wt%)	X-Ray Diffraction Compound	Emission Spectrographic Analysis (wt%)									
		Ag	Al	Ca	Cb	Co	Cr	Cu	Fe	Mg	Mn
C	$\text{NaCO}_3 \cdot \text{H}_2\text{O}$	0.05	0.4	0.2	<0.01	0.3	>1.0	0.5	>1.0	0.1	1.0
6.03		Mo	Na	Ni	Pb	Si	Sn	Ti	V	Zn	Zr
		0.8	>1.0	>1.0	>1.0	>1.0	0.3	0.2	0.1	1.0	0.1

Figures 8 and 9. Carburization occurred to a slightly higher degree in the thermally stressed regions, i.e., the weld and the 2-inch diameter line, compared to the main primary line. Carburization has been shown to occur at a higher rate in stressed material⁴, and it is assumed that thermal stresses may have had an accelerating effect in this case. Except for the light carburized case, the tee was unaffected.

The largest deposit in the main primary line was found along the top of the main primary pump outlet line, Figure 10. The deposit was composed primarily of amorphous carbon, Table III. Since the tetralin entered the system through the pump seals and is known to decompose rapidly (to hydrogen, free carbon, and aliphatic hydrocarbons²) in sodium above 675°F, particulate carbon probably formed in the sodium at the pump outlet. Apparently, the high turbulence and frothing in the pump outlet region caused the carbon to impinge and cake along the top of the line. The deposit terminated about 3 feet downstream from the pump outlet -- a reasonable distance for high turbulence.

To determine the effect of direct contact of the carbonaceous material on the Type 304 stainless steel line, sections of the line directly under the deposit and from the bottom of the line which were essentially free of deposits were examined metallographically. Typical microstructures of both sections, Figure 11, do not indicate any significant difference caused by the direct contact of the high carbon deposit. The hardened case extends to a depth of 0.010 inch in both cases, and the grain boundaries are heavily sensitized to a depth of 0.030 inch, Figure 12. The high degree of sensitization is clearly indicated by the removal of whole grains from the structure, caused by the complete dissolution of the grain boundaries by the metallographic etchant. Similar attack by sodium has not been observed.

Cold Leg

The general integrated thermal and flow history of the cold leg is outlined in Table IV. The eductor, a jet pump used to provide coolant flow for the graphite moderators, was installed at the time of Core II. The velocity of the sodium changed from 13 ft/sec in the main line to 60 ft/sec in the nozzle, shown in Figure 13. When the eductor was removed from the system, sodium was found to be trapped by the nozzle restriction, Figure 14. The sodium was removed by hot oil (300°F) immersion followed by immersion in Dowanal, ethylene glycol butyl ether. After sectioning, the nozzle was easily removed from the suction shell. A film of powdery deposit was found on the surface of the nozzle, Figure 15. The standard procedure for decontamination (manually wiping the surface with a solution of Alconox detergent and water) restored a bright metallic finish to the nozzle except for some black staining on the inside of the reduced section of the nozzle, Figures 16 and 17. The concentration of the staining in the orifice of the nozzle--the highest velocity region--is believed to have been caused by impingement. Table V lists the analysis of both types of deposit. Dye penetrant examination of the nozzle did not indicate any surface defects. Despite the high velocity of the sodium in this region, evidence of erosion was not found.

Multiple sections of the eductor nozzle and the suction shell were examined metallographically. A hardened layer up to 0.001 inch thick was found along all of the surfaces that were exposed to sodium, Figure 18. The hardness of the layer ranged from 290 to 325 Khn, compared to 160 Khn for the 304 stainless steel substrate; the inclusions found in the layer, Figure 19, indicate that it is a morphological change of the base metal, rather than a plating. Attempts to etch the layer have failed to reveal any grain structure, Figure 20.

Figure 8.

Longitudinal section of the 6-in.-diam. hot leg line adjacent to the drain pit line tee. The carburized case along the surface exposed to the sodium (top) averaged 307 KHN (100 gm. load) and decreased to an average of 198 KHN (100 gm. load) 0.006 in. below the surface.

Etch: Ferric Chloride

Mag: 150 X

Figure 9.

Longitudinal section of the weld between the drain pit line and the hot leg line. The carburized case along the surface exposed to the sodium (top) averaged 350 KHN (100 gm. load) and decreased to 198 KHN (100 gm. load) 0.008 in. below the surface.

Etch: Ferric Chloride

Mag: 150 X

Figure 10. Carbonaceous deposit along the top of the 6-in-diam. hot leg line at the outlet of the main primary pump present after cleaning.

TABLE III

Analysis of Deposit from the Hot Leg Line at
the Main Primary Pump Outlet

-Spectro- graphic Analysis (mc/gm)	Carbon Analysis (wt%)	Emission Spectrographic Analysis (wt%)									
		Al	Bi	Ca	Cr	Cu	Fe	Mg	Mn		
Cs ¹³⁷ 1.6	C 73.8	0.5	0.05	0.1	2.0	0.3	10.1	0.1	0.3		
		Mo	Na	Ni	Pb	Si	Sn	Ti	Zr		
		0.05	5.0	2.0	0.3	4.0	0.1	0.05	0.03

Figure 11. Sections of Hot Leg Line, 304 Stainless Steel, at Main Primary Pump outlet. Surface Exposed to sodium is at top. Hardness averaged 307 Khn (100 gm. load) at surface, and decreased to an average of 142 Khn (100 gm. load) as indicated by increase in size of hardness indents.
 A - Surface below carbonaceous deposit, Figure 10.
 B - Surface along bottom of line that was free of deposits.
 Etch: Ferric Chloride Mag: 150 X

Figure 12. Typical transverse section of Hot Leg Line, 304 Stainless Steel, at the main Primary Pump outlet. Surface exposed to the sodium is at top. A highly sensitized structure near surface is indicated by absence of whole grains caused by complete dissolution of grain boundaries by etchant.
 Etch: Ferric Chloride
 Mag: 150 X

TABLE IV

Integrated Thermal and Flow History of the SRE Cold Leg*

Temperature Range (°F)	Flow Time in Hours **	
	Core I	Core II
300	120	180
300 - 399	4,080	9,480
400 - 499	3,024	7,848
500 - 599	3,048	7,216
600 - 699	72	1,968
TOTAL:	10,344	26,716

* Interval: Core I - May 4, 1958 to November 10, 1959
Core II- July 22, 1960 to February 15, 1964

** Flow: Mean Flow 1100 ± 200 gpm (12.5 ± 2 ft/sec in 6-in. lines) with 6,000 hr. of Core II time at 1450 to 1560 gpm (16 to 17.5 ft/sec in 6-in. lines).

Figure 13: Cross Section of Eductor Nozzle.

Figure 14. The inlet to the eductor nozzle after removal from the system. The sodium (white mass) was prevented from draining by the restriction of the nozzle.

Figure 15. The eductor nozzle section after cleaning and removal from the suction section.

Figure 16. The outside surface of the eductor nozzle after cleaning with detergent.

Figure 17. Inside surface of the eductor nozzle after cleaning with detergent. The black stain is a thin film deposit.

TABLE V

Analysis of Eductor Nozzle Deposits
(Spectrographic wt%)

Elements	Eductor Nozzle		
	<u>ID</u>	<u>OD</u>	<u>Stain</u>
Al	0.5	0.5	0.5
Ca	0.02	0.03	0.05
Cb	0.05	0.05	
Co	0.03	0.03	0.05
Cr	1.0	1.0	8.0
Cu	0.02	0.02	0.5
Fe	1.0	1.0	8.0
Mg	0.1	0.1	0.8
Mn	1.0	1.0	2.0
Mo	0.05	0.02	0.1
Na	1.0	1.0	
Ni	1.0	1.0	2.0
Pb	0.5	1.0	0.05
Si	1.0	1.0	3.0
Sn	0.05	0.05	
Ti	0.1	0.3	0.5
V	1.0	1.0	0.1
Zn	0.1	0.01	
Zr	0.1	0.2	0.05

Carbon Analysis in wt%

C	12.89	18.64
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Compounds by X-ray Diffraction

FeC
 Na_2CO_3

Etch: ferric chloride Mag. 50X 7633-1505

Figure 18. Section of the outlet of the Eductor Nozzle illustrating the layer formed on all surfaces exposed to the sodium. The ID surface is along the bottom, and the OD surface is along the top. Etch: Ferric chloride Mag: 50X

Figure 19. Section of Educator Nozzle with layer on right. Micro-hardness of the layer is 327 Khn (100 gm. load) compared with 160 Khn (100 gm. load) for base metal, as indicated by two hardness indents at lower right. Inclusions present in layer indicate it is part of original base metal. Etch: Ferric chloride Mag: 150X

Electron microprobe scans for Fe, Cr, Ni, Zn, Mn, Zr, Mo, and Si failed to indicate any change in composition between the layer and the Type 304 stainless steel base material. Analyses by X-ray fluorescence and diffraction have also yielded the typical Type 304 stainless steel patterns. Analysis of chips and thin sections of the layer with a minimum amount of retained base metal are listed in Table VI. Emission spectrographic analysis of a thin section with the layer compared with a section of the base metal with the layer removed indicated that molybdenum was the only element with any significant variation in content. The base metal averaged 0.1% Mo, and the thin section averaged 1.0% Mo. However, even a 3% Mo content would not account for the high hardness of the layer.

A section of the main primary line from the core inlet region is shown in Figure 21. Other than a hardened layer, Figure 22, on the surface exposed to the sodium similar to the one observed on the eductor, the material appeared to be unaffected.

Liquid-Vapor Region

A Type 304 stainless steel hanger rod from a temperature probe which had been in the core since the start of the SRE in 1957, was examined for effects at the sodium liquid-vapor interface. The surface of the rod had a rusty appearance which is attributed to the steam washing it received prior to being placed in storage. After the loose rust had been wiped off, a sharp change in coloration was observed in the central portion of a 20-inch long section taken from the liquid-vapor region, Figure 23. The line apparently marks the normal (at-temperature) sodium level.

Sections at several levels in the vapor and liquid regions were examined metallographically. The location, radiation levels, microstructures, and hardness measurements of the sections along the rod are shown in Figure 24. Analysis of thin sections (< 0.005 inch thick) of the surface of the rod in the liquid region averaged 0.64%C and in the vapor region, 0.69%C; confirming that the hardened case was caused by carburization. An indication of carburization in a sodium vapor has been reported⁴; however, gaseous hydrocarbons produced in the system by the decomposition of the tetralin probably contributed to the carburization.

The slight corrosive attack along the surface of the sections that were immersed in the sodium is attributed to dissolution by the sodium of corrosion products produced during steam washings. The degree of penetration is insignificant.

Summary and Conclusions

Although the SRE sodium-heat-transfer system has been subjected to a wide variety of operating conditions during the seven year period, no indication of an impending functional failure has been found. Evidence of cracking or erosion has not been observed. Detection of Fe, Cr, and Ni in thin film deposits and of slight surface irregularities in the hot leg imply corrosion may have occurred, but the degree is apparently insignificant.

The presence of appreciable carbonaceous deposits in the hot leg indicates that the sodium had been saturated with carbon since the introduction of tetralin into the system. Carburization in the main primary piping has not been significant either in terms of carbon content or of case thickness.

The layer found in the cold leg appears to be a morphological change along the surface of the stainless steel. The characteristics of brittleness have not been observed in the layer, and the high hardness may have been beneficial in

Etch: ferric chloride Mag. 500X 7633-1507

Figure 20. Section of Eductor Nozzle with layer on left. Grain boundaries are not detectable in layer. Etch: Ferric chloride Mag: 500X

TABLE VI

Chemical Analysis of Machined Chips from the ID of a Cold Leg
Section with the Layer

Elements	Fe	Cr	Ni	Mn	Mo	Si	C	N
wt%	69.2	18.1	10.2	1.59	0.44	0.40	0.07	0.05

Figure 21. Section of 6-in.-diam. cold leg line removed from adjacent to the core inlet.

Figure 22.

A longitudinal section of the line above. The layer along the sodium exposed surface is similar to the layer on the eductor. The hardness of the layer averaged 290 Khn (100 gm. load) with an average of 180 Khn for the base metal. Etch: Ferric chloride
Mag: 150 X

Figure 23. Section of temperature-probe, 304 stainless steel hanger rod from sodium liquid-vapor interface region of core. Change in coloration (arrow) indicates location of liquid-vapor interface. Taped end on left was in vapor region.

Figure 24. Carburized case observed along temperature probe, hanger rod, Type 304 stainless steel.
Etch: Ferric chloride Mag: 150X

preventing erosion in the nozzle of the eductor. Assuming linearity, a comparison of the thickness of the layer on the eductor with the one on the original lines gives a growth rate of less than 0.0003 inch a year.

Considering the severest case of any of the material changes observed in the SRE sodium-heat-transfer system, large sodium systems operated under SRE conditions can be expected to have reliable life expectancies in excess of 30 years.

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EBR-II Materials Experience

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During the 1963 Winter Meeting of the American Nuclear Society, A Symposium on materials for sodium-cooled reactors was held in conjunction with the Metallurgical Society of the AIME. The proceedings of this symposium have been published by the AIME in a book called "Nuclear Metallurgy, Volume IX." The Argonne National Laboratory presented a paper called "Materials for Argonne National Laboratory Sodium-Cooled Reactors." Since this paper described the materials and some of the components in the EBR-II Reactor, I will refer you to this book for background material. In this paper we shall attempt to bring our experience with these materials in the EBR-II Reactor up to date.

First, I would like to describe the secondary sodium system. The EBR-II secondary sodium system is the heat transfer system carrying heat from the radioactive primary sodium system to the steam generator. It is composed of the following major components:

1. Intermediate primary sodium to secondary sodium heat exchanger. This is manufactured of 304 stainless steel.
2. Secondary sodium pump. This is a linear electromagnetic pump. The pump duct, which is in contact with the sodium, is fabricated from type 347 stainless steel.
3. The surge tank. This is an 800 gallon tank fabricated from 304 stainless steel.
4. The storage tank. This is a 16,000 gallon tank fabricated from 304 stainless steel.
5. Cold trap and recirculating pumps. These are fabricated from 304 stainless steel.
6. Steam generator. On the sodium side the evaporators and superheaters are identical. There are ten units in all and they are fabricated from 2.25% Cr, 1% Mo, low alloy ferritic steel.
7. Interconnecting piping. The piping system is composed of 304 stainless steel and 2.25% Cr, 1% Mo, (P-22) ferritic steel. The stainless steel piping has an inside area of approximately 1900 sq. ft. and the P-22 piping has an inside area of approximately 10,300 sq. ft.

Unfortunately, the ferritic piping was installed without pickling to remove the mill scale. Based on chemical cleaning of the water side of the steam generator, it was estimated that the sodium piping contained approximately 400 lbs. of mill scale (Fe_3O_4). Since sodium will reduce the mill scale it was necessary to go through a special cleaning program after the system was filled with sodium.

This program took approximately four months and essentially consisted of filling the system, heating to 560°F, allowing the plugging temperature to rise to about 540°F, draining, heating the storage tank to 600°F, cold trapping until the plugging temperature was reduced to less than 250°F, cooling the sodium to 350°F (the filling temperature), and repeating the cycle. A total of five cycles was necessary before the plugging temperature stayed constant after fill and heatup.

The original cold trap was completely filled during this operation and a temporary cold trap was then installed. This temporary cold trap was used until the end of the cleaning cycle. It was then removed and a new, permanent cold trap was installed in the system. Because of the batch-type process used, it was rather easy to keep a close track of the amount of sodium oxide removed from the system. A total of 130.6 lbs. of sodium oxide were removed which only accounted for approximately one-third of the total oxygen we had initially through to be present. Subsequent operation since this cleanup has shown that the system is not stable but that oxygen is slowly being picked up by the sodium. We have operated the cold trap in a batch-type process allowing the plugging temperature to rise from approximately 250 to 300°F. Then we put the cold trap on the line and reduce the plugging temperature to 250°F. Currently, it takes about five days for the plugging temperature to rise from 250 to 300°F and two days of cold trapping will again reduce it to 250°F.

During the cleaning procedure large amounts of particulate material were present in the sodium. It is thought that these were small pieces of mill scale which had broken off the surface of the piping as the piping expanded with temperature. These gave us a considerable amount of trouble in our plugging run determinations, and it was necessary to install a micro-metallic filter in the line leading to the plugging meter so that meaningful plugging runs could be made.

While the low alloy ferritic material is technically satisfactory for the piping, a careful analysis should be made to see if it is the most economical material. Because the P-22 material in schedule 20 is a specialty, the mill price is actually higher than the comparable schedule 10S 304 stainless steel price. To this must be added the cost of pickling and the added welding expense brought on by preheat and stress relief requirements. These requirements are made more expensive by the necessity of providing an argon purge in the system to prevent formation of more scale. The use of stainless steel piping is complicated by the need for expensive transition welds where it joins ferritic material and cladding the piping with carbon steel if induction heating is to be used.

The pump duct of our secondary sodium EM pump failed early in 1964. The failure is believed to have been caused by a low npsh at the pump inlet which caused a vibration type of fatigue in the large flat duct. However, the choice of pump duct material is still believed to have been sound.

In the primary system, both our pumps failed but these failures were again not due to choice of material but rather were design and fabrication problems.

The EBR-II reactor oscillator functioned by moving a control rod up and down with a linear motion. Part of the assembly consisted of a ball bushing which had to operate submerged in sodium. This bushing completely disintegrated during operation, spilling in excess of 100 1/8-inch ball bearings into the reactor. By

analysis of those parts that we have been able to recover we have found that the bushing was made of carbon steel rather than the stainless steel which was specified. While a suitable stainless steel bushing may have lasted much longer, the consequences of a failure precludes the use of this type of mechanical arrangement.

The EBR-II reactor source is quite standard in that it consists of a hollow cylinder of beryllium metal canned in 304 stainless steel and an antimony metal slug canned in tantalum which fits inside. Since it has been determined that the sources in EBR-II are self-renewing we have had some second thoughts on the tantalum clad material. Tantalum will, of course, reduce sodium oxide and become embrittled in time and it is feared that this cladding material will fail. Antimony tetroxide (Sb_2O_4), which has sufficient density and chemical stability to be a suitable source material has been tested in stainless steel containers to 1900°F and found to be compatible with the stainless steel. We therefore have a long range program to replace our antimony tantalum-clad sources with sources containing antimony tetroxide contained in a 304 stainless steel container.

Aluminum bronze (Ampco No. 18-13) versus stellite No. 6 is used as a labyrinth seal and guide bushing where the control rod drive shafts penetrate the reactor cover as well as other places in the primary tank. The control rod labyrinth and bushings are approximately $2\frac{1}{2}$ inches in diameter and have about 0.020 inches diametral clearance. Four of these control rod drive shafts have had to be removed for various reasons giving us an opportunity to inspect the aluminum bronze surfaces. They were in excellent condition with practically no signs of wear.

Figure 1 is a sectional view of an extremely important component in the reactor. It is the so-called gripper mechanism which is used to grasp subassemblies that are removed and inserted in the reactor and also to grasp the fueled subassemblies that are the reactor control rods. There are twelve control rod grippers and one fuel handling gripper. The gripper jaws and gripper funnel are held to the jaw and funnel support piece. The jaw actuator which is a hollow tube rides up and down the support piece and cams the gripper jaws open and closed. The support piece is hollow and a sensing rod rides up and down through the center of it to sense whether the top of the subassembly is clear of the gripper jaws. The end of the sensing rod in the fuel handling gripper is shaped like a screw driver and fits into the slot on the top of a subassembly. This provides an orientation sense to the gripper. Since the control rod gripper does not need this sense, the end of the sensing rod is shaped like a hollow cone to fit over the end of the subassembly. We thus have an assembly which contains a number of rubbing surfaces and pivoted surfaces. The jaw actuator which is made of 304 stainless steel has a hard chrome-plated interior. This bears against four stellite inserts in the jaw and funnel support piece. The gripper jaws and gripper funnel are machined from a 420 series stainless steel and hard chrome plated. The jaw cams on the jaw actuator are half-moon shaped pieces of stellite. These are moved across the camming surfaces of the gripper jaw to open and close the jaws. The total travel of the jaw actuator is approximately $1\frac{1}{2}$ inches. The jaw pivots which have to be very strong as well as hard are made of 18-4-1 tool steel which is hardened and chrome plated. The sensing rod is made of 420 stainless steel with a hard chrome plate. The top of the subassemblies are made of 304 stainless steel having no protective coating on their surface. To engage a subassembly, the whole gripper assembly is lowered until the sensing rod seats in or on the slot at the top of the subassembly

and is forced upward. If the gripper assembly is at the right elevation showing the screw driver on the sensing rod went into the slot and is not crosswise to the slot, the jaws can be closed by driving down the jaw actuator. This motion cams the gripper jaws closed around the mushroom type head on the top of the subassembly. To disengage the grippers from the subassemblies in the case of the control rod, the jaw actuator was moved upward coming the gripper jaws open. At this point the subassemblies would fall free approximately 3 inches. The sensing rod would then fall downward $3/4$ inch showing that the control rods were indeed released. The full weight of the subassemblies were on the gripper jaws while this operation was being done. Since the mating jaw faces are flat and the bottom of the mushroom is rounded, there is only line contact resulting in very high unit pressure. It has proven difficult in some instances to perform this operation showing sticking between the gripper jaws and the mushroom head on the top of the subassembly. The procedure has recently been revised so that the subassemblies are sitting in the bottom position in the grid and the gripper jaws opened without the weight of the subassemblies on the jaws. After the jaws are opened, the entire gripper assembly is moved up approximately 3 inches allowing the sensing rod to move downward and showing that the grippers have indeed released the top of the fueled control rod.

In one instance the jaw actuator mechanism got out of adjustment allowing the jaw actuator to move upward too far. When it was then pushed downward to close the jaws the upper most lip of the gripper jaw engaged one of the jaw cams and sheared it off the jaw actuator. When this happened the particular jaw could not be opened preventing the release of the subassembly from the gripper assembly. By shaking and raising the sensing rod it was possible to disengage the subassembly from the jaw. The gripper was removed from the tank, the cam found in the gripper, and the gripper rebuilt. In another instance, interference developed between the jaw and funnel support piece and the jaw actuator making the movement of the jaw actuator very difficult. This assembly was also removed from the tank and it was found that presumably foreign material had deformed the cylindrical jaw actuator by $1/32$ inch; extremely high unit pressures must have been present to cause this deformation. However, where the stellite rubbing pieces mated with the jaw actuator, only moderate galling was evident. At present it is not known what this foreign material was which got between the support piece and jaw actuator. At present it is estimated that these jaws have been actuated approximately 500 times. In the case of the gripper which takes fuel in and out of the core the construction is almost identical. We have had one mishap of this mechanism. An offset holddown device is positioned on the subassemblies surrounding the desired subassembly when it is removed from the reactor. The gripper was improperly aligned with the holddown device and on lowering, the funnel interfered with the holddown and became bent. It was removed from the reactor and the funnel replaced.

Except for this one instance, the gripper has successfully made over 900 transfers of fuel into and out of the EBR-II reactor core. In view of the problems we have had on the EBR-II reactor, it is gratifying to be able to report on one mechanism which works so well under such severe conditions.

One item which is still causing some concern is the rotating seals for the reactor. These seals consist of a 304 stainless steel trough, 304 stainless steel blade, and are filled with a tin-bismuth eutectic alloy. During reactor operation, the top half of the alloy is frozen and the bottom is maintained in a molten condition. During fuel handling the alloy is completely melted to allow the plugs to rotate. It has become necessary to employ increasing higher temperatures to completely melt these seals and allow rotation of the plugs. Laboratory tests

have shown that even through this is a eutectic alloy, repeated freezing and melting cycles cause a segregation of the alloy with the bismuth concentrating at the bottom of the container and the tin concentrating at the top of the container. There is also evidence of weld cracking in some of the heater holes in the blade. However, it is not known at this time whether this was caused by faulty welds or if it is characteristic of the system.

Considering the complexity of the EBR-II reactor and associated equipment, we have had minimal troubles caused by materials selection.

FIGURE 1

THE UNITED KINGDOM LIQUID METAL DEVELOPMENT PROGRAMME

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Introduction

In his paper to the Third Geneva Conference⁽¹⁾, Sir William Penney gave the status at that time of nuclear power development in the United Kingdom. The first phase of this programme is the installation of 5,000 MW(e) of nuclear power by 1970, these stations being of the "magnox" type. All these stations have now been ordered.

A detailed economic study of the part which nuclear power could play in our electricity generating system with stations to be commissioned between 1970-75 had just been completed. The Government decision had been announced in a White Paper⁽²⁾ in April of that year and was that the Central Electricity Generating Board should plan to commission a further 5,000 MW of nuclear power in England and Wales in that period. This decision would be regularly reviewed and the possibility of another station in Scotland would also be examined. At that time, the choice of the type of station for the second phase was left open. The Central Electricity Generating Board had called for tenders for the first of these stations (Dungeness B) based not only on the advanced gas-cooled reactor system (A.G.R.), which is under development in the U.K., but also on water-moderated systems. It has now been announced that Dungeness B will in fact be an A.G.R. station.

It was stated in this paper that, even at that time, the largest single part of the Authority's development effort was devoted to fast reactor development. The immediate tasks seen were the development of fuel and cladding with an objective of a 5% burn-up; a detailed design study of a large fast reactor and development of the associated engineering techniques; and the provision of the basic reactor physics information for which the Zebra zero energy facility had been designed and commissioned. The paper foresaw that a really major expansion of low-cost nuclear power would depend on the successful development of fast reactors. Initially, the main benefit from the introduction of fast reactors should be the raising of plutonium values due to the more efficient use of plutonium possible in fast reactors than in thermal reactors, leading to a general lowering of nuclear power costs compared with those possible with only thermal reactors. Looking still further ahead, the contribution of the fast reactor could be in ensuring efficient utilisation of the uranium. The fast reactor has always been seen, therefore, as an essential part of the overall plan for nuclear energy development in the U.K.

Since that paper was given, considerable progress has been made in all the main areas. The Dounreay fast reactor (D.F.R.) played an essential role in the progress, especially in the area of fuel, cladding and sub-assembly development. The decision to build D.F.R. was taken in 1954: it first generated electricity in 1962 and reached full power in 1963⁽³⁾. It was designed as an experimental

metal-fuelled NaK-cooled fast reactor of 60 MW(t). It has demonstrated the feasibility of fast reactors running at a power of 60 MW(4). The reactor has now completed (mid 1965) its eighth and longest full power run, with a full loading of experiments relating to future fast reactor fuel and cladding development. It is steadily accumulating evidence on the irradiation behaviour of sub-assemblies of fuel pins in a fast flux environment at projected fuel ratings and temperatures.

Design studies of fast reactors have been made and, at the same time, engineering development of components for fast reactors and our knowledge in the field of sodium technology has advanced at a similar pace. The successful operation of D.F.R. at full power and to a planned programme has been a major contribution to the build-up of confidence in liquid-metal-cooled reactor engineering technology, as well as fuel element technology. The reactor physics work at Winfrith, using the Zebra facility, has also maintained its planned programme of work. The time, therefore, has now been reached where the next immediate objective can be set. This is the construction and operation of a fast reactor delivering some hundreds of megawatts of heat, which is sodium-cooled and fuelled with uranium-plutonium oxide. Such a reactor would comprise a core made up of sub-assemblies of uranium-plutonium oxide fuel pins grouped around control and shut-off rod tubes placed in a diagrid which provides the coolant inlet header to the sub-assemblies. This core, with primary sodium coolant circuits would be entirely contained in a stainless steel tank. Secondary sodium circuits would transfer the heat to the steam raising units.

The main components which therefore have to be developed are the fuel element sub-assemblies, the mechanical pumps, intermediate heat exchangers, steam generators, and reactor mechanisms such as re-fuelling machines and control gear systems. In addition, it is necessary to establish the operational sodium technology at temperatures up to 650°C. Instrumentation to operate under these conditions is also a vital area for development in this programme.

The Sodium Facilities

Although it is intended in this paper to outline the sodium facilities in order to indicate the programme, it is necessary to mention that they cannot be entirely considered in isolation. In the engineering development of a reactor system, as is common practice, the problems which can be solved by modelling out of sodium are done using air and water to avoid undue cost, complication and time. It is also a part of the general development philosophy that as much basic information is provided before design freeze to minimise the check out of actual reactor components. Finally, it has always been the policy, especially in fuel element and sub-assembly development, to obtain as much operational experience by irradiation and operational use in the D.F.R. Hence, for example, D.F.R. can irradiate sub-assemblies designed to principles likely to be used in larger reactors.

The facilities to be described are either in the Reactor Engineering Laboratory (R.E.L.) at Warrington (Fig. 1), or in the Experimental Reactor Establishment at Dounreay (D.E.R.E.). Materials problems are tackled at the Reactor Materials Laboratory, Culcheth, and there are eight sodium loops deployed on this work. The Reactor Materials Laboratory work will not be described in this paper.

Mechanical Pump Rig

It was decided at an early stage that mechanical pumps should be used in the projected fast reactors. It was also judged necessary to obtain first-hand experience of the design and operation of this vital fast reactor component. The pump chosen for study is of the centrifugal type with a vertical shaft supported by an upper, oil-lubricated roller bearing and lower sodium-lubricated journal bearing.² The size chosen for study was 6,000 gal/min with a pressure rise of 75 lb/in². This pump will give enough technological and operational information to extrapolate to larger sizes which will be of the order of 15,000 gal/min. The pump is being tested in a specially designed loop (Fig. 2). The loop consists of a stainless steel vessel housing the pump, and a re-entrant loop of 12 in. diameter containing suitable instrumentation for flow measurement. The loop is constructed entirely of 18/8 stainless steel. The loop impurity level is controlled by a small, subsidiary, pumped clean-up loop comprising a 1-in. electromagnetic pump and a cold trap. Impurity levels can be obtained by sampling, with subsequent laboratory analysis, and operational performance is checked by a plugging meter. The amount of sodium held up in the loop is about 12 tons and the dump tanks are on the floor level. The loop operates at a temperature of 400°C.

A careful programme was planned for commissioning this facility. This programme called for a check of impurity levels at all stages from initial sodium delivery to the final clean-up state of the pump. Attempts were made to assess the dirt burden which had to be dealt with when commissioning a facility of this type. Hence, as far as possible, an indication was obtained of the levels to be expected in a large reactor. In addition, the construction, inspection and initial cleaning of the facility were those which it was deemed could be used in future reactors. A newly-developed leak-rate measuring system was used based on a helium isotope dilution technique.

To study the detailed performance of the pump, special instrumentation had to be developed including transducers to monitor bearing clearances. The experimental programme is now under way and has reached the stage where the pump has been run at full power, and pump characteristics obtained. There is a further programme of experiments and tests necessary before a planned long endurance run as a final check out.

Further experiments, when the pump tests are complete, are now being planned. In particular, a study of gas entrainment problems and the special instrumentation necessary is at the moment being developed. It is expected that this will be the only very large sodium flow facility in the development programme.

Fuel Element Thermal Test Facility

Another vital component in the fast reactor is the fuel element sub-assembly. Again, at an early stage, the decision was taken that appropriate flow and temperature conditions should be available for sub-assembly and other test work. To this end a 4-in. loop facility was designed and constructed (Fig. 3).

The loop was designed around the electromagnetic pumps originally built for D.F.R. which can deliver 50 lb/sec of sodium at 35 lb/in² head and 220°C. By

fitting these pumps with a specially-developed high-temperature winding, the allowable metal temperature has been raised to 375°C, and two of these pumps are used in series to provide 350 gal/min at 70 lb/in².

To provide sodium at 650°C at the test section, the loop is fitted with a 10 MW heat exchanger, a 350 kW heater and a suitable cooler. An additional 4-in. pipe is provided which allows cold sodium to by-pass the heat exchanger and heater and feed directly into the test section. Thus, thermal shocks of an amplitude of 250°C, and at rates of 50°C/sec, can be obtained. In commissioning this loop, considerable difficulty was experienced with the immersion heaters and an alternative induction-type heater has had to be designed and at present is being fitted. The standard cold trap and impurity monitor and sampling system is provided. A carefully-planned programme of commissioning has yielded valuable information on impurity levels and clean-up in sodium circuits.

A subsidiary test section has now been fitted to this loop and contains an experiment to measure the eddy diffusivity of heat as a function of Reynolds number. The ratio of this value of the eddy diffusivity of mass in air is a necessary piece of information in the design and evaluation of hot spots in sub-assemblies. The experiment measures the decay of a spike of hot sodium along the axis of a large tube and, for its performance, the development of high-precision thermocouple scanning equipment has been carried out. First results from this test section are now available and show that at Reynolds numbers around 6×10^4 $\epsilon_H/\epsilon_{Mass}$ is of the order of 0.6.

This loop is also heavily subscribed for instrumentation work, particularly the development of flowmeters and temperature measuring systems. An experiment in which a model sub-assembly containing 70 heated pins, designed to study a.c. temperature measurement systems, has recently been successfully completed. The facility is heavily subscribed for the future for sub-assembly evaluation work and instrumentation development.

General-purpose Rig

The third facility in R.E.L. is a 1-in. loop which is an exact analogue in its circuit design to the 4-in. loop just described. It can supply 25 gal/min with pressures up to 10 lb/in² and temperatures of 650°C. It has been used on a wide range of mechanical development problems including sub-assembly engineering problems, valves and joints, and for the development of various pieces of instrumentation. We are planning at the moment to extend the 1-in. facilities available in the laboratory because of the heavy commitment.

Small Sodium Rigs

Generally supporting these major facilities are a number of minor rigs, some of them in fact in the laboratories rather than in the main experimental hall. These are listed below.

1. A multi-headed rubbing-pair rig which allows basic investigations into friction and wear of materials in sodium environments of known impurity, and temperatures up to 600°C. Both rotating and reciprocating experiments can be performed. The results of some of this work have already been described by Roberts⁽⁵⁾.

2. A facility for producing mixtures of sodium vapour and inert gas to reproduce reactor cover gas conditions for the study of friction and wear of reactor materials.
3. A laboratory $\frac{1}{4}$ -in. loop which was designed for basic investigations into sodium wetting of materials. It consists of a purified sodium supply system to a small, open rectangular pot enclosed in a glove box, in which the cover gas purity can be very carefully controlled. So far in this rig the field of sodium vapour mass transfer has been tackled, and the wetting work originally planned is yet to start.

The next few facilities to be described are situated at our Dounreay establishment.

D.E.R.E. 4-in. Loop

This is a loop similar in concept to the R.E.L. loop previously described but differing in certain details. It uses but one 4-in. D.F.R. pump and can supply 90 gal/min of NaK at 600°C, with a pressure drop of the order of 30 lb/in². This loop has been used for the check out on the sub-assemblies for operation in the D.F.R. It is also planned to do some cold trap development on this loop, and therefore it has glove box facilities. At the moment it is programmed for a study of boiling heat transfer in small rod clusters. As mentioned by Matthews⁽³⁾, acoustic techniques are being developed for the detection of the onset of boiling, and hence accident conditions, in fast reactors. These experiments are planned to give the necessary basic information for the design of such detection systems. A series of boiling experiments under various conditions will continue for some time.

1-in. Facility

This facility is based on the same pump as the R.E.L. 1-in., and at the moment it is arranged to produce sodium at temperatures in the range of 650°C to 700°C in the test section. The present programme of work is designed to study the problem of instrument design to detect hydrogen from the sodium/water reaction, especially in the cover gas space.

NOAH Rig

Study of the sodium/water reaction is obviously relevant to the use of liquid metal coolants, and work is in progress at Dounreay on the NOAH rig.

The rig is a 1/16th-scale model of a 250 MW shell and tube steam generator (Fig. 4). The shell is filled with sodium which is circulated by an electromagnetic pump giving a downward flow. The tube section is contained in a square-section ducting within the main vessel. Because of the complexity of the tube assembly in the model, this is represented mainly by a number of perforated metal plates packed loosely in the square duct to give approximately the same geometry. One water loop is provided and this is taken to the base of the duct assembly where it is looped in the design layout and surrounded by a group of wire dummy water tubes arranged in the same way. A leak into the vessel is simulated by using a special pipe fracturing device in this line. Termination of the leak is controlled by a quick-acting shut-off valve. The loop is fed by a pressuriser vessel in which a reserve of water is maintained at

the correct test conditions. Temperature and pressure changes within the test vessel due to reaction are measured and recorded on a high-speed recorder unit. Excess pressure in the test vessel can be relieved by a bursting disc which allows the gaseous reaction products to vent into a large surge vessel. The rig is housed in a concrete vessel, and is remotely controlled.

So far, a number of tube burst tests have been carried out and further work is planned.

Future R.E.L. Facilities

The future facilities will be those specifically designed to solve detailed design problems, especially those associated with reactor mechanisms. In general, the programme calls for an experimental study of the component parts of these mechanisms to give design information before the whole design is frozen. For example, any fuel handling machine must contain a number of bearings, screws and grab mechanisms which will be studied as separate problems. Therefore, a new sodium facility is planned which will contain a number of test vessels in which these components can be studied under the correct sodium environment and operating conditions. It is also planned that grab and sub-assembly raising and lowering mechanisms will be studied to full-scale in the same sodium test facility.

In a similar way the various problems on control and shut-off rod mechanisms have been defined, and separate, individual experiments, and hence sodium test facilities, specified. Further, it has been decided that as full as possible a check-out on a complete control rod mechanism will be necessary, and to this end a further large test section will be added on the mechanisms rig.

As mentioned previously, an extension of our 1-in. facilities for both mechanical and instrument development is deemed to be necessary.

Future Downreay Facilities

With a sodium pool operating in the region 600°C , the sodium fog, and hence mass transfer, problems can pose serious problems in certain design features. Downreay are therefore planning to convert one of their existing facilities to provide a sodium pool at appropriate conditions and to study certain specific sodium mass transfer problems. In addition, the question of whether scum formation on the sodium surface is a serious problem or not will be examined.

Instrumentation

It is necessary to mention the significant amount of effort being put into the development of instrumentation for liquid-metal-cooled reactors. The main urge behind this instrumentation arises from two main causes. First, the development of a comprehensive core monitoring safety system of instrumentation especially to guard against the sub-assembly blockage problem. Secondly, instrumentation which now has to work in sodium temperatures up to 650°C and, under fault conditions, higher than this. At the moment, we have under development burst can detection systems, boiling detectors, flowmeters, level gauges, pressure transducers and, in addition, a comprehensive study of the performance of thermocouples at these temperatures in sodium. Finally, I

would like to mention the work we have done in the field of impurity analysis. Here, the view has been adopted that, to understand operational performance of our rigs, specific impurity information is essential. Therefore, on all rigs there are extensive sampling facilities which are backed up by the appropriate analytical laboratory service. At the same time, the value has been recognised of continuously-indicating instruments of the rhometer and plugging meter type. We have just completed an extensive development on plugging meters and are now satisfied that we have an automatic, continuously-recording meter of sufficient sensitivity and stability for the reactor application. D.E.R.E. are developing the Blake rhometer for operation in sodium.

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Figure 1-R. E. L. General View of Sodium Building.

Figure 2-6, 1,000 gal/min Sodium Pump Test Facility.

Figure 3-Fuel Element Thermal Test Rig.

Figure 4-NOAH Test Vessel.

A CEA-EURATOM CONTRIBUTION TO

THE SODIUM TECHNOLOGY

- M.G. ROBIN -

COMMISSARIAT A L'ENERGIE ATOMIQUE

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I - INTRODUCTION

Sodium has been selected as a coolant for the reactors foreseen in our fast breeder program, and research in the sodium field was started, about eleven years ago, at the Fontenay Nuclear Research Center (figure 1 shows the location of the French Centers where sodium research is carried out).

Our first fast reactor, a plutonium fueled 20 MWt prototype called Rapsodie /1/, should be critical by the end of 1966. It will primarily be used as an irradiation facility and will not be devoted to electricity production (at least during the first years of operation). Consequently until 1963, our first efforts in sodium technology were chiefly devoted to such components as intermediate heat exchangers, pumps, valves, instruments, etc ... In 1961, a 1 MWt, and a 10 MWt loop were erected at the Cadarache Nuclear Research Center and then operated successfully /2/ ; the largest one is a full scale prototype of the two cooling loops of Rapsodie which is now under construction at Cadarache. Due to the limited time available, after giving a few indications on experimental sodium heat transfer coefficients, I will discuss our present experience on steam generators which we consider as a major problem in sodium technology.

II - SODIUM HEAT TRANSFER COEFFICIENTS

The 10 MW intermediate heat exchanger (figure 2) which is of the shell and tube unbaffled type (similar to the 10 MWt EFFBR heat exchangers as far as heat transfer is concerned) has been tested and the shell side Nusselt numbers, D_o being the outside tube diameter, are shown on figure 3. For comparison, experimental points obtained from two 200 kWt U-shaped heat exchangers, tested at Fontenay in 1961, have been included. In both cases the overall heat transfer coefficient was measured and the tube side Nusselt number was calculated according to the Lyon's equation.

The validity of this equation, within the Peclet number range of interest, has been checked using an electrically heated circular tube and 56 w/o K sodium-potassium alloy (figure 4).

The experimental results which will be published in one of the next issues of the International Journal of Heat and Mass Transfer show that the use of the classical Lubarsky-Kaufman equation should give conservative results and that the Lyon equation might be optimistic in the $10^2 - 10^3$ Peclet range.

Due to the necessity of evaluating the NaK heat transfer coefficient in our first steam generator prototype, research was conducted at Fontenay on a tube-in-tube economizer, whose annular cross section was similar to that of the steam generator (diameter ratio of 2). The useful Peclet range was explored and the experimental results are given on figure 5. The Dwyer (annulus) and Harrisson and Menke (parallel plates) curves are shown for comparison.

III - LIQUID METAL HEATED STEAM GENERATORS

The second French fast reactor will be a power plant prototype, called Phenix, whose construction could start at the beginning of 1969.

One of the most important new problems associated with this second reactor is the development of the steam generating equipment, and we have decided to devote a large effort to this problem.

So far, the following single reheat steam cycle has been selected as a reference design cycle :

- steam pressure	163	bars	(2400 psia)
- superheated steam temperature	500 to 510	°C	(930 to 950 F)
- feed water inlet temperature	280	°C	(536 F)
- reheated steam temperature	500 to 510	°C	(930 to 950 F)
- reheated steam pressure	33.5	bars	(490 psia)

In order to carry out a research and development program on sodium heated steam generators, a 5 MWt facility has been completed in 1964, on the Seine left bank, at Grand Quevilly, about 75 miles west from Paris. The schematic diagram of this facility is shown on figure 6.

It comprises an oil-fired sodium heater, a centrifugally pumped 4 in. ID sodium primary circuit for heating the shell side of a U bent intermediate heat exchanger, a centrifugally pumped 4 in. ID secondary NaK circuit for heating the shell side of a bank of six parallel tube-in-tube, double wall steam generating elements, and steam and water circuitry. Figure 7 shows the general arrangement of the double wall steam generator; static NaK is used as a third fluid and is argon pressurized at approximately 90 psig. The static NaK expansion tank is fitted with a rupture disc connected to a discharge pipe and separating tank vented to the atmosphere. Water flows upward in the innermost tube countercurrently to the secondary NaK and emerges as superheated steam in the upper header. This first steam generator designed in 1959 and constructed in 1962, is completely made of 321 stainless steel which is exposed to both water and NaK. After an initial start-up on June 1964, the design steam conditions were achieved on December 28, 1964 and so far the facility has undergone about 2 000 hours of operation despite mechanical troubles in the boiler feed pump and initial difficulties with water purification.

At the present time, typical tests conditions at 100 % load are :

- Thermal power	5	MW
- Steam pressure	1800	psig
- Steam temperature	1013	F
- Water inlet temperature	450	F
- Water flow rate	16,200	lb/hr
- Water steam pressure drop	60	psi
- NaK inlet temperature	1085	F
- NaK outlet temperature	656	F
- NaK flow rate	164,000	lb/hr

For the 100 % load, a typical temperature distribution along the steam generator length is shown on figure 8. The thermal balance is within 5 %.

The following overall heat transfer coefficients have been obtained along the heat exchange area (total length per tube 183 ft, inside diameter = 20 mm or 0.787 in).

- Water preheating section (28.7 ft)	472	Btu/hr-ft ² - F
- Nucleate boiling section (44.3 ft)	482	Btu/hr-ft ² - F
- Film boiling section (15.8 ft)	393	Btu/hr-ft ² - F
- Superheat section (94.2 ft)	470	Btu/hr-ft ² - F

Tests in steady state conditions have been carried out for 40, 60, 800, 100 and 110 % loads. During all these tests the steam temperature was steady within ± 1 F, and the steam pressure within ± 1 psi.

The water treatment system which is fed with city water consists of :

- a filter (crushed pebble bed)
- a cation exchange bed
- a CO₂ eliminator
- an anion exchange bed
- a mixed bed.

25 % of the total boiler water flow is retreated continuously in a mixed bed. Hydrazene and morpholine are added periodically to the feed water in order to maintain the pH value between 8.9 and 9.2 and to scavenge oxygen.

The following water purity is currently achieved :

- Resistivity	>	10^6	ohm-cm
- Silica content	<	30	microgram/liter
- Iron content	<	10	microgram/liter
- Cu content	<	20	microgram/liter
- Chloride content	<	50	microgram/liter

The NaK purification system is simply an air-cooled diffusion cold trap attached to the cold leg of the secondary system. A NaK plugging temperature of 200 F is achieved now after a few months of operation (375 F after the first start-up).

IV - SODIUM-WATER REACTION STUDIES

As part of our steam generator program , NaK-water reaction experiments are being conducted at Cadarache (figure 9). The main facility for this program is a circulating NaK-water system consisting of a 50 kW electrically heated loop including a once-through type , single-walled , single tube steam generator , a throttle valve , an attemperator , a condenser and a feed pump. The 0.079 in. thick 316 SS tube wall is perforated using an electric arc device which produces a 0.08 in-hole. Depending on the selected leak location , water or steam reacts with NaK ; the resulting pressure breaks the two rupture discs and the reaction products are discharged to the atmosphere. At the same time the failed steam generator is automatically isolated from the NaK loop and from the water loop by air operated valves (in about 50 milliseconds). During the first tests carried out with steam at 1800 psig and 1000 F , no NaK side peak pressure above 25 bars (350 psig) has been recorded (rupture pressure of the safety discs was 100 psig).

A second steam generator with a seven tube bundle has been designed : it will allow the study of the effect of a complete failure of the center tube.

We hope that the results of this several month experimental program will reinforce the current trend in France toward single-walled units , two of which are being contemplated for tests at Grand Quevilly about the end of next year. These two prototypes which are now under study are part of the steam generator program for Phenix and will be operated at the pressure and temperature indicated for the Phenix reference cycle.

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Fig 1 LOCATION OF FRENCH SODIUM FACILITIES (▲)

Fig.2 10 MW Na NaK INTERMEDIATE HEAT EXCHANGER

Fig.3 Shell Side Heat Transfer (Unbaffled Bundles)

Fig. 6 GRAND QUEVELLY FACILITY

Fig. 9 50 KW NaK - WATER REACTION TEST FACILITY

INTERATOM Activities in the Field of Sodium Technology

by R. Harde

INTERATOM has been actively engaged in the field of sodium technology since about 1960, mainly in connection with the design and development of a 20 MWe sodium cooled power reactor experiment (KNK-Project). Construction of this plant is expected to be started within the next few months. When completed the system will serve both as a compact high thermal performance sodium reactor experiment and as an irradiation facility for fast reactor fuel pins. The reactor plant has been described in a Geneva Paper (1). Some additional information on heat transfer components and developmental sodium test facilities were included in a paper presented by J.J. Morabito and H.W. Savage during a recent ANS meeting on sodium technology in Detroit (2).

This presentation will be limited to a discussion of some aspects of the use of carbon stabilized low alloy ferritic steel in high temperature sodium systems and to steam generator safety problems. In particular the experience gained at INTERATOM during the construction and start-up of a ferritic steel 5 MW Component Test Facility and an outline of the results of sodium-water interaction experiments will be reported.

1. The use of ferritic steel in sodium systems

The use of low alloy ferritic steel in high temperature sodium systems as a structural material has been contemplated at various places. In most cases a (2 1/4 Cr, 1 Mo)-type steel was considered which, however, had to be modified in order to overcome decarburization problems at higher sodium temperatures. The main incentives for the application of such a steel instead of an austenetic material are reduced component costs and the elimination of stress corrosion problems

in steam generators. Strength limitation at higher sodium temperatures may constitute a major disadvantage of such a material.

In cooperation with the steel manufacturer Phoenix-Rheinrohr AG, Düsseldorf, Interatom has investigated in the last five years the properties of a (2 1/2 Cr, 1 Mo)-steel which contained Nb in quantities about 10 times the carbon content (i.e. less than 1%). It could be shown that in thermal convection sodium loops essentially no carbon transfer to stainless steels of the types 18/8 and 16/13 took place at temperatures up to 600°C and over a period of 15.000 hours (3). Precautions must, however, be taken to avoid surface carburization of pipes and sheets during manufacture. As the yield strength and ductility for material produced in large scale heats and under standard mill conditions was not completely satisfactory, a small amount of nickel (0,5%) was added to the alloy in order to improve these properties. Table I provides a comparison of some of the more important properties of the standard (2 1/2 Cr, 1 Mo)-steel and its two carbon stabilized varieties. While it must be stressed that some of the data are still very preliminary the beneficial effect of the nickel addition seems unquestionable. It should be noted that the fatigue life for inelastic strain was measured for shock loading of the samples.

The very pronounced increase of the yield strength of the nickel contained steel is considered to be rather important because the design of at least the secondary sodium systems has also to take into account the potentially very high pressure transients that can occur after a steam tube failure in the steam generators for a short duration.

Large scale manufacturing and welding experience for the carbon stabilized steel has been obtained during the construction of a 5 MW-Sodium Components Test Facility (2,3) which was taken into service in January 1965 (Fig. 1). The quality rejection rate of the 100% X-rayed welds amounted to less than 3%. Also the postweld heat treatment could be shown to be no major cost item.

Table I: Properties of (2,25Cr; 1Mo)-type ferritic steels

Type A: standard (2,25Cr; 1Mo)-steel

Type B: C-stabilized (2,25Cr; 1Mo; 10xC Nb)-steel

Type C: C-stabilized (2,25Cr; 1Mo; 10xC Nb; 0,5Ni)-steel

(some data are preliminary)

property	unit	type	20°C	400°C	500°C	550°C	600°C
0,2% yield strength (1" sheet)	$\frac{kg}{mm^2}$	A B C	27 20 40	20 14 37	18 12 34	- 10 31	- - 27
10 ⁵ hrs creep strength to rupture	$\frac{kg}{mm^2}$	A B,C	- -	- -	15 12	7,5 7	4 (2,8)
fatigue strength >10 ⁷ elastic cycles to rupture	$\frac{kg}{mm^2}$	B,C	25	21	-	17	-
fatigue life for inelastic 0,2% strain of 0,4% 0,53% 1,0%	cycles	C	~25.000 ~2.000 ~600 ~100				
notch impact strength	$\frac{mkg}{cm^2}$	A B C	6 6 13				
ductile-brittle transition temp.	°C	B	~+ 40 ~- 40				

A matter of concern when using a ferritic material for construction of large sodium systems is the problem of system purification especially with regard to rust removal. During the construction of the 5 MW-Facility cleanliness and corrosion protection of pipes and components was maintained only to the degree that no major extra cost would be incurred. As a result after assembly all internal surfaces were covered with a thin layer of rust. The purification of the system was obtained in three phases. After filling and draining sodium at 250°C seven times, circulation of the sodium through mechanical filters was started at low rates gradually raising the sodium temperature to 550°C. Finally full flow was obtained with the 17 micron mesh size filter operated in a by-pass flow scheme. During the purification period one large cold trap (50 kg sodium oxide) was consumed. Details of sodium analyses carried out at the various purification steps are summarized in table II. The data are self explanatory and tend to indicate, that during the construction of a ferritic sodium system no expensive precautions for cleanliness are required. No pipe plugging by sodium oxide (smallest pipe diameter: 0,5" in auxiliary air cooler) was observed at any time.

2. Some problems and results related to steam generator safety

No representative operating experience of a highly rated single wall sodium heated steam generator is available or can be accumulated in the near future. Because of uncertainties with regard to long term corrosion and durability of protective magnetite layers on the water-steam side, the possibility of tube failures cannot be excluded in spite of close quality control of tubes and welds during manufacture. After a small or large water leakage into the sodium the reduction in plant availability due to cleanup and repair time can be very severe for large vessel type steam generators. In addition the amount of sodium involved in a leakage accident can be uncomfortably large. For these reasons and because the overall behavior of very large steam generator systems can be predicted by tests on rather small units, modular type steam generators are presently being preferred at INTERATOM. By proper design and application of mass production techniques the

Initial system purification

		Na as supplied	after fill & drain (7 times)	after full flow screen run	after bypass screen run
Operating time	hrs	0	16	51	508
Operating temper.	°C	-	250	250-550	550
Oxygen concentr.	ppm	245	330	350	< 10
Carbon concentr.	ppm	24	23	21	24
C in s.s.-sample	w/o	0,051	n.d.	n.d.	0,054
Particulate matter	ppm	28	720	65	36
part.comp.: Fe	w/o	56	95	95	95
SiO ₂	w/o	8,5	1,6	1,7	2,2
C	w/o	<0,1	0,46	1,10	1,93
part.size:>1 mm	w/o	28	21		5
0,1 - 1 mm	w/o	43	40		34
10 - 100 μ	w/o	28	28	n.d.	48
< 10 μ	w/o	1	11		13

cost disadvantage of modules as compared to large vessel units can be reduced to acceptable limits. Among the possible module concepts a concentric tube in tube design (2,3) was selected because of its simplicity and because it eliminates any danger of short term tube to tube interference after small water leakages. In addition such a system permits afterglow heat removal by passing air over the outer tubes. In this way the steam generators serve as a stable and effective emergency heat removal system even after a plant shut down as a result of a steam system failure.

In case of a water leak into the sodium the reaction pressure is limited by rupture discs which are connected to a pressure relief system. If the leakage is small causing however some pressure increase, the steam generator is separated from the rest of the sodium loop by fast acting shut off valves. A large water leak causes automatic isolation of the effected module or module group by non reversible shut off devices at the inlet and outlet end of each module or module group. In order to provide a rapid indication of small water leaks the sodium outlet of each module or module group can be equipped with a sensitive but simple (hydrogen) gas bubble indicator. The working principles of the non reversible shut off device and the gas bubble indicator are depicted in Fig. 2. Both devices are inexpensive and have been successfully tested under representative experimental conditions (shut off time 10 msec; sensitivity of bubble indicator 1 vol%).

The non reversible shut off devices are not fail safe; their proper response conveniently limits the amount of sodium involved in a water leak accident and permits - in principle - continued plant operation. If they fail to respond, protection of the loop until the pressure relief system becomes effective is achieved by a "single direction orifice" some distance upstream the sodium inlet, by an orificed sodium hold up vessel at the outlet and - by a high yield strength construction material. The term "single direction orifice" refers to a fixed orifice which is machined in such a way that it offers to a reversed flow a hydraulic resistance 3 to 4 times as large as under normal flow conditions. Since no flow reversal occurs at the sodium outlet a less effective orifice has to be combined with a sodium hold up tank. The latter prevents the reaction gas to enter the remainder of the sodium

system and eliminating thus the orifice effect while the pressure relief system is not yet fully effective. All protective devices have been designed according to results obtained in a series of sodium water interaction experiments.

Stable and controlled steam generation (510°C , 80 atm) has been achieved in a steam generator equipped with the aforementioned devices.

3. Sodium - water interaction experiments

A large number of sodium water interaction experiments in pipes have been carried out under various temperatures and pressure conditions. Fast quartz transmitters were used to record the pressure transients.

The principles of the arrangement of the experimental set up and the location of main instruments (p pressure transducers, k spark plugs, thermocouples) can be taken from Fig.5 Each Experiment involves the reaction of 5 to 10 kgs of sodium.

Typical recordings for saturated and subcooled water are given in Fig. 3 and Fig. 4.

Pressure readings at the various transmitter locations indicate a travelling shock wave which is reflected at the end of the first test section several times. Peak pressures up to 1200 at lasting less than half a millisecond were recorded. After about 100 msec the test section pressure remains for some time near the initial water pressure if the leakage was sufficiently large (e.g. complete tube rupture).

If the initial sodium temperature is much higher than the temperature of the saturated water a delayed secondary pressure surge superimposed on the primary reaction pressure can always be observed (Fig. 4). This effect can be interpreted as steam flashing due to a sudden release of heat stored in the hot steel pipe to the introduced saturated water. The high temperature difference between the pipe and the water causes initially steam blanketing of the pipe (film boiling), the

steam layer collapses some time later usually after a random pressure disturbance.

All results could be interpreted fairly well by a theoretical model which is briefly explained in Fig. 5.

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Fig. 1

FIG. 2

FIG. 3

FIG. 4

FIG. 5

SODIUM COMPONENT TEST INSTALLATION *

by

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In prior meetings, papers have been given which described the Sodium Component Test Installation--its design, construction, safety aspects, operating organization, and proposed testing schedules. This paper will review very briefly some of the salient features of the plant, report activities of the last year, indicate the current status of the plant, and describe the activities planned for the next year.

I. PLANT DESCRIPTION

The Sodium Component Test Installation (SCTI) is a large liquid metal system designed specifically for the developmental and proof testing of the prototype sodium-heated steam generators. The plant has a maximum capability of 35 Mwt with sodium temperatures up to 1300°F for short periods and 1200°F continuously. Flow capability is such that any prototype steam generator or intermediate heat exchanger with reasonable thermal temperature differences can be tested. Figure 1 is a schematic diagram of the SCTI process systems. The primary sodium loop includes a 35 Mwt gas-fired heater, an 8.5 Mw air blast cooler, a free surface pump, and the shell side of the intermediate heat exchanger. There are both hot and cold bypasses in the primary loop which were included to make possible the performance of rapid thermal transients. The secondary loop is a very simple circuit including another free surface pump, the tube side of the IHX, and the shell side of the steam generator. A single cold trap is provided for trapping both primary or secondary systems. Plugging meters and flow through sampling stations have been provided with each sodium loop.

On the right hand portion of Figure 1 is shown the steam side of the plant. No useful work is done by the steam produced in testing; instead, high pressure steam passes through three pressure reducing stations and two desuperheating stations before being discharged to a surface condenser. Ultimate heat rejection from the system is through a forced-draft cooling system which supplies cooling water to the condenser. There are three stages of feedwater heating including a deaerator. There is also a complete water treatment system included to provide control of boiler feed water standards comparable to those of a conventional power plant.

* Work performed under contract AT(11-1)-GEN-8 for the U. S. Atomic Energy Commission.

Adequate control and instrumentation has been incorporated to permit conducting transient temperature changes rapid as 35° per second for 14 seconds--up or down. There is also adequate instrumentation to record results of the testing program. Various design types of steam generators can be tested with only minor changes in plumbing, power, or instrumentation hookup.

Several figures included here afford a quick pictorial tour of the SCTI plant. Figure 2 shows the firing aisle side of the 35 Mw gas-fired heater, the forced air duct, heater-outlet sodium piping, and the primary fill/drain tank pit. An elevation view of most of the sodium piping, the IHX, cold trap, surge tanks, pump drives steam piping, and the inert gas control panel as shown in Figure 3. The top of the steam generator pit is in the center of the picture. Figure 4 is an isometric view of the steam generator pit with both ALCO and modular units shown installed. At the present, only the ALCO steam generator is in position. The two tanks mounted vertically are relief tanks connected to the rupture discs on the steam generator shell side. The horizontal tank is the secondary sodium fill and drain tank. Figure 5 is a general view of the feed and treated water piping. The main condenser is in the rear center of the picture, the larger pipe lines are circulating water for the condenser, the feedwater heaters and flash tanks are clustered at the right hand side of the photo. Next, Figure 6 shows the deaerating heater, the pre-heat boiler, and the sodium cooler.

II. ACTIVITY REVIEW

One of the principal accomplishments during the last year was the installation of the ALCO/BLH steam generator. This unit was received late in August, 1964, and all construction connected with its installation was completed at the end of January of this year. Figure 7 shows the steam generator just after it was taken off the truck, and Figure 8 shows it in a vertical position being lowered into the pit; these two pictures give an idea of the physical size of this unit--which is rated for 30 Mwt with outlet steam conditions of 2200 psig and 1050°F .

During the installation of the ALCO/BLH steam generator, it was found that certain thermocouples had been broken and had to be replaced, and also that some internal thermocouples had been omitted by the manufacturer. These situations have all been corrected. In connection with planning for the hydrostatic testing of the connecting piping, it was realized that both the stud bolts and the nuts for the head closures were made of stainless steel. This was, of course, undesirable because of the possibility of galling. Additionally, a calculation of bolting force required to maintain proper gasket loading indicated that the stud bolts were not strong enough. That is--the creep rate under initial loading would be high enough to unload the joint in less time than the length of the test program. As a result, the original studs were replaced with new ones made of high strength alloy, 19-9-DL.

A primary accomplishment of the staff during this last year has been the preparation of procedures and the performance of preoperational tests. Seventeen such procedures were outlined in our report of last year. Individual procedures were written for every major system and subsystem of the plant to provide a step-by-step process by which every piece of equipment, instrument, and control in the plant could be determined as properly installed and operable. Virtually all of these procedures have been completed. Performance of these detailed, step-by-step tests made manifest a number of problems, which if left uncorrected, would prevent

either proper operation or adequate control of the plant. Action required to rectify these problems has been completed.

Typical of some of the modifications and changes that were instituted as a result of the preoperational test program include the following:

1. Addition of several safety relief and check valves in the steam and feedwater system which were determined to be required.
2. Addition of a cold trap system, with connections to both primary and secondary sodium systems, complete with a plugging meter and an in-line sampling station for each loop.
3. Change in location of existing, and placement of additional steam and feedwater sampling lines.
4. Addition of sample holders for metallurgical determination of impurities in the sodium system.
5. Provisions of additional screen gates between the cooling tower and main circulating pump to permit cleaning the screens without shutting down the circulating water system.
6. Installation of a jib crane adjacent to the cooling tower to facilitate handling of screens, pump parts or other heavy equipment.

Construction work for all of these items has now been finished. Since last year, raw water conditioning equipment, a preheater for the sodium cooler in the primary system, and other equipment have been built into the plant, checked-out, and operated.

During the first quarter of fiscal year 1965, 88 barrels (36,600 lbs) of sodium were melted and put into the primary fill and drain tank. The primary system was purged and preheated following preoperational test procedures in preparation for system filling. At this time it was determined that additional sodium storage capacity was required. Several piping design changes had been made which increased system volume - after the original tanks were already installed. Therefore, a decision was made to purchase a new and larger tank for the primary system and to replace the existing secondary tank with the existing primary tank. The new tank has been installed, the old primary tank is in service as the secondary tank, and in addition, 100 drums (41,000 lbs) of sodium have been transferred into the new tank.

In terms of advancing the "state of the art" of sodium systems, there are several items that may be of interest. First, level gages operating on the principle of gamma attenuation have been installed for the steam generator and both sodium expansion tanks. After some initial position and alignment problems, these devices appear to be satisfactory. Another level indicator utilizes a standard float device (manufactured by Fisher Governor) in which change in float position is transmitted by a torque arm. So far, operation of this unit has been satisfactory. A recent addition to the instruments has been a gas chromatograph, set specifically to detect hydrogen in the cover gas over the sodium in the steam generator; this instrument samples and analyzes cover gas every six minutes. There is also a nickel membrane type hydrogen detector located at the sodium outlet from the steam generator--

which will give continuous indication. One other item of interest is an air preheating system which is installed on the primary sodium piping. Stainless steel tubing was used to "trace" the lines. Air compressed to 100 psi is heated in a gas-fired heater and distributed through several manifolds to the pipe tracing. Initially, a fair amount of time was spent in balancing air flows to obtain a good temperature distribution, but once set, the system has been very satisfactory, and needs very little attention. One desirable feature of this preheating method is that there is no heater burnout--as occurs in electrical heaters. Another item of hardware newly applied to sodium systems is butterfly-type valve for throttling service. There are six of these valves in our plant. These valves are very compact for their pipe size, are freeze stem sealed, have pneumatic operators, and the butterfly vane is supported by a stellite sleeve pivot bearing. On the basis of a very meager experience to date, these valves function well and give a good control over the full flow range.

III. STATUS AND FUTURE PROGRAM

At the time of preparation of this paper, almost all of the preoperational testing has been completed. This means that we have circulated sodium in both primary and secondary systems and determined hydraulic characteristics of those two loops. The steam and feedwater systems and their associated components have been chemically cleaned, circulation established, and operating capabilities of the pumps determined. The ALCO/BLH steam generator is preheated--prior to sodium filling--by circulating 350°F water through the tube side; this operation has been successfully performed several times. A considerable amount of work has been done in affirming proper installation and operation of the instrument and control systems. Although the preoperational phase of plant control check-out is finished, this work will continue as different tests are performed.

Immediately upon the completion of the preoperational test phase, plant performance tests will be started and continued for a month or two. The major objectives of this phase of the program are to: (1) demonstrate that the SCTI can be operated safely and efficiently over the full operating range, (2) verify that the plant has the performance capabilities for which it was designed, (3) assure continued dependable operation of the plant so that testing of steam generators will proceed expeditiously and, (4) familiarize operating personnel with plant responses under controlled conditions. During this phase, the plant will be taken to full power rating of 30 Mw. The ALCO/BLH steam generator will be in service during this period, and much operating data will be recorded. However, specific tests of the ALCO/BLH steam generator will be started after completion of most of the plant performance testing. The total program for the ALCO/BLH testing is described in detail in APAE-122, "Test Program for 30 Megawatt Prototype Sodium Intermediate Heat Exchanger and Steam Generator." Most of the tests detailed in that report will be performed. Assuming that all goes well, and that no major failures occur, nine months will complete the testing of the ALCO/BLH unit. After that, it is planned that the modular steam generators, designed and built by AI, will be installed and tested. These units will in turn be followed by the generator now being built by B & W.

SCTI SCHEMATIC FLOW DIAGRAM

Figure 1

Figure 2 SCTII - Gas-fired sodium furnace, forced-draft blower and primary sodium tank pit

Figure 3 SCTI, East view SCTI piping inside building

ALCO & MODULAR STEAM GENERATORS INSTALLATION IN SCTI

MSG SUPERHEATER MODULES (2)

SODIUM INLET

STEAM OUTLET

FEEDWATER INLET

BLOWDOWN GAS DISCHARGE

TEST PIT

SODIUM OUTLET

MSG BLOWDOWN

RUPTURE MEMBRANES

BLOWDOWN TANKS

MSG EVAPORATOR MODULES (2)

ALCO (B-L-H) STEAM GENERATOR

MSG BLOWDOWN

SODIUM DUMP TANK

Figure 4

Figure 5 SCTI water treatment area from cooling tower

Figure 6 SCTI deaerator, preheat boiler, and sodium cooler

Figure 7 Up-ending ALCO steam generator from horizontal position

Figure 8 ALCO steam generator being lowered into test pit

MODULAR STEAM GENERATOR FABRICATION*

L. T. Bogaert and D. F. Casey

With advancement of the sodium cooled reactor concept and its output of high temperature sodium, it becomes necessary to develop equipment with the capability of generating high temperature steam efficiently and economically (Reference 1). To provide such equipment, tooling and fabrication equipment must be developed. Fabrication and inspection techniques must also be developed to assure workmanship of very high quality.

Under the Sodium Component Development Program, Atomics International has developed fabrication techniques for and fabricated two evaporator and two superheater modular steam generators (Figure 1). The following report pertains to the fabrication of the steam generators. The evaporators were fabricated from 5%Cr-0.5%Mo steel and the superheaters from type 321 stainless steel with a 304 shell.

During the fabrication of the prototype steam generators, areas for change or improvement in the design, as pertinent to producibility, were demonstrated. Tooling requirements were defined, equipment needs established, and fabrication and inspection procedures determined. This initial fabrication effort was planned around the use of existing equipment and fabrication processes. All shell welding was done manually. Tube-to-tube-sheet welding was done by the process and equipment as developed in an earlier program for use in this effort (Reference 2).

All fabrication and assembly tooling was made rugged but as uncomplicated as possible. Tooling was made interchangeable for both the evaporator and superheater.

A detailed Manufacturing Plan was prepared for guidance in the procurement of materials, components, design and fabrication of tooling equipment requirements, assembly, inspection, test, and shipment of the completed modules.

All primary containment welding conformed to Section VIII of the ASME Boiler and Pressure Vessel Code. Tube-to-tube-sheet welding was done to a modified (more stringent) requirement of the ASME code.

During fabrication, tooling, techniques, and processes were improved wherever possible. Records were made for future reference and further improvement of design, fabrication, materials, and test procedures.

Materials

All materials and miscellaneous hardware items were purchased according to the specifications established by engineering. In some cases, such as the tube sheets where it was found that the specifications would not assure a satisfactory end product, the specification was supplemented to provide that assurance.

* Work performed under contract AT(11-1)-GEN-8 for the U. S. Atomic Energy Commission.

The tube sheets (Figure 2) called for material to ASTM specifications, vacuum melted, and also ultrasonic inspection. This requirement, while being satisfactory in intent, did not specifically define the quality of material required. Arrangements were made with the supplier for ultrasonic inspection of the ingot and also an inclusion count with established limits for acceptability. Forging blanks were cut from the billet in areas that were found acceptable by ultrasonic inspection. Since greater difficulty in welding was expected for the 5%Cr-0.5%Mo alloy, these forgings were subjected to a magnetic particle inspection prior to final machining operations. Nozzle sections, with less stringent specifications, were forged from the balance of the ingot, considered unacceptable for tube sheets. Since the highest impurity level is normally found at the center of the ingot, the center area was bored out of the forging in final machining operations to match the nozzle opening.

The pipe for the shell sections was purchased with a requirement for a partial ASME code stamp. This provided assurance that the pipe would meet all the test requirements once the modules were completed.

In cutting the 5%Cr-0.5%Mo pipe to length and making the weld joint preparations, small shallow checks and/or cracks were found in the inside surface of the pipe. These were probably caused in the drawing or sizing operations. Examination of samples showed that the depth of the defects were from 0.002 to 0.004 in. The pipe was pickled in a solution of 15% nitric and 3% hydrofluoric acid to remove the defects and then thoroughly rinsed and dried. The pipe was reinspected both visually and dimensionally. The pipe was cut to lengths required and weld preparations machined.

Examination of the type 304 stainless steel pipe showed that the pipe was not round, had not been checked using a "pass thru" gage, nor had partial ASME code stamped. The pipe was returned to the supplier for rework and completion.

Inspection of both the 5%Cr-0.5%Mo and type 321 SS pressure tubes revealed variation from wall thickness, ovality, and straightness requirements at the tube ends. Such variations would be detrimental to tube-to-tube-sheet welding operations. The tubes were then finished in a lathe on each end, matching the I.D. to the bore of the tube sheets and finishing the O.D. concentric to the bore with a minimum wall of 0.095 in. This preparation was made for one inch of length, blending to the original I.D. and O.D. Since many of the tube ends were bowed they were also straightened at this time.

Handling, machining of the ends, and the tendency of the 5%Cr-0.5%Mo tubes to show signs of rust and the accumulation of dirt and dust while unwrapped necessitated a preliminary cleaning. Just prior to installation in the tube bundle, the tubes were flushed with acetone and then cleaned with cotton wads blown through them.

Spacers (Figure 3) were investment cast, thereby avoiding costly machining operations. However, after a first article inspection at the source, it appeared that the pattern had been damaged. When the castings were positioned 180° to each other, the opposing holes did not match. The holes were reworked in pairs in a fixture and the hole locations corrected.

Tube Bundle

The tube bundle was assembled in a strong back fixture (Figure 4). It was designed around an I beam, quite simple but rugged and providing the assurance that the tube bundle would not be damaged in handling.

The spacers were first positioned in the fixture and the support rods installed and welded. The upper tee to be used for a thermocouple outlet, a short pipe section, a cone reducer and diffuser were added at this time. Once the welds were inspected the inlet tee was fitted and welded (Figure 5). The tubes were now inserted and the thermocouples (on the evaporators) threaded into the bundle and attached to the tubes by means of modified tube clamps. The bundle assembly completed, the upper tube sheet was fitted to the fixture in position, aligned with the tube bundle.

A mobile crane rig was used to pick up the tube bundle assembly and the fixture and position it in the pit (Figure 6). The pit was immediately covered and purged with dry argon gas. The dew point of the gas was monitored periodically. No corrosion or rust stains were observed while any of the Cr-Mo assemblies were in the pit. The tube bundle assembly was handled on the fixture at all times to prevent bending the tubes.

Tube-to-Tube-Sheet Welding

During the design phase of this program, development of the internal tube-to-tube-sheet welding was accomplished (Figure 7). Welding procedures for 2.25%Cr-1%Mo, 5%Cr-0.5%Mo, type 347 and type 304 stainless steel were developed. Weld parameters were also established. A complete program of testing and evaluation was carried out. Details of this effort are reported in NAA-SR-8541.*

Briefly, 1) the tube to be welded is brought to the tube stub on the tube sheet, 2) the copper chill and external gas manifold, not shown, is positioned aligning the tube to tube stub, 3) a ball of stainless steel wool, not shown, is inserted well below the weld to avoid intermixing of inert gas and air, 4) the weld head is positioned in the tube sheet bore to be welded, 5) the gas lines, power cables, and power supply are checked, 6) the weld cycle is initiated, 7) upon completion of the entire weld cycle the weld head and chill are removed, and 8) the ball of stainless steel wool is removed. The weld is now complete.

Tube-to-sheet welding was accomplished by use of a weld head similar to the one developed earlier for this program (Figure 8). This head was an improved model in that the drive motor was integrally mounted and the locating plate was simplified. Power was provided by the same power supply and programmer as used in the early development efforts. All tube-to-tube-sheet welding was performed with the units in a vertical position. This was accomplished by installing the units in a steel-lined pit approximately 50 ft. deep, in the assembly area floor.

The procedure for setup was the same for both the evaporators (5%Cr-0.5%Mo) and the superheaters (type 321 SS). The welding parameters were determined and the welding procedure specified for each alloy (Figures 9 and 10). The weld procedures specified the equipment to be used, the type of shielding gas, gas flow, electrode description, all of the automatic programming parameters, and detailed step-by-step setup instructions, plus a sketch of the weld joint setup.

During the initial tube bundle assembly fifteen tube-to-tube-sheet welds were made on a mockup of the tube-sheet configuration. One mockup was made from 5%Cr-0.5%Mo steel; the other from type 321 stainless steel. Welds were made using the same weld procedures, respectively, as those to be used on the modules. The samples made of Cr-Mo steel were post heat treated by the same procedure as intended for that tube sheet. These samples were x-rayed, dye penetrant and helium leak checked, and finally metallographically examined. When the procedures were satisfactorily checked out, the tube-to-tube-sheet welding operations were started.

Of the first 37 tube-to-tube-sheet welds made, two were considered questionable because of pores or inclusions upon evaluation of the x-ray film. Both welds were rerun, reinspected, and found to be within the limits of the specification. X-ray of the tube-to-tube-sheet welds (Figure 11) was done by the double wall exposure, shooting at a vertical angle of approximately 30°, imposing the weld image as an ellipse on the film. Two shots were made from opposite horizontal angles so that the weld would be fully exposed on film. Should an area be questionable, additional shots were made to clearly define that area. Porosity and inclusion allowances were established by using a specially prepared comparison plate allowing approximately one-third the inclusions and/or pores of that allowed by Sect. VIII of the ASME Boiler and Pressure Vessel Code, for 1/4 in. material. Thinning, undercut, offset, or tungsten inclusions were not permissible. The x-ray machine enclosures were fabricated of lead, thus providing shielding for personnel to within 13 ft. when the equipment was in operation.

By welding from the middle row of tubes outward, radiography was performed upon completion of every second row of welds. This permitted access to both rows of tubes. It also permitted dye penetrant inspection of the outside and inside of the welds.

The tube bundle was then removed from the pit and the entire tube sheet and welds placed in a furnace for post heat treatment of the evaporator units (Figure 12). Post heat treatment was accomplished by heating at a maximum rate of 400° per hour to 1325-1375°F, holding for one hour and cooling at a rate of 250-500°F per hour to below 600°F. Heating was monitored by clamping six thermocouples in the tubes at the weld joints and four additional thermocouples randomly placed around the tube sheet. Temperatures were recorded on tape for record. During heat treatment the entire heated area was purged with argon gas.

The welds were cleaned, helium leak tested, and dye penetrant inspected. The tube bundle was assembled to the shell by placing the shell in pit, then picking up the tube bundle and lowering it into the shell in a vertical position, a costly and time-consuming operation.

The tube ends were now faced to equal lengths by means of a spot facing tool in a drill press mounted over the end of the bundle. The tube sheet was mounted in position for welding of the lower end. Welding and inspection both proceeded much more rapidly and without difficulty. Since the tubes were now fixed at the opposite end, welding and inspection required lateral springing of the tubes (Figure 13) for operator access. This proved to be no obstacle and caused no damage to the tubes. Three sets of spacers, a 32-in. shell section, a cone reducer, and the lower cross were left off the assembly for this purpose, thus allowing over 60 in. of free tube length.

The first major discrepancy occurred on the second evaporator module. The 35th weld on the second end had been completed; a setup was made for the 36th weld. The weld head was erroneously replaced in the 35th position. Without the protection of the combination external chill and gas backup, the weld was burned through.

After evaluation of the damage, it was decided to attempt a salvage operation. The tube stub on the tube sheet was back faced to remove all cast metal, approximately 1/8 in. Two in. of the tube end was removed. The then existing end of the tube was prepared for welding and a 2-1/8 in. splice section also prepared for welding. The splice section was welded to the tube sheet using the original weld procedure. The tube was welded to the splice piece using a modified weld procedure (Figure 14).

Later in making the tube-to-tube-sheet welds on the superheaters, a short tube was accidentally installed in a tube bundle. This tube was not discovered until after it had been welded. Rather than make a splice on the opposite tube sheet, it appeared advisable to cut off the short tube and replace it with a tube of correct length. A new tube was inserted in the tube bundle without removing it from the pit. The tube stub on the tube sheet was refaced and prepared for welding. In welding the weld joint was "blown" into the gas annulus of the external chill. Examination revealed that a guide "bullet" used in inserting the tube had not been removed, causing excessive internal gas pressure during the welding operation.

Salvage required reshaping of the tube stub to nearly its original configuration using specially ground cutters and preparing a new splice section. The splice section was welded to the tube sheet using the original weld procedure. A modified procedure (Figure 15) was used to weld the tube to the splice piece. To assure control of the internal gas flow a small loosely wadded ball of stainless steel wool is inserted several inches below the weld joint. At this weld, a second ball was left in the tube and again caused excessive gas pressure causing a "blown" weld. This weld was also removed and a second splice section prepared and welded in place without incident (Figure 16).

In any future tube-to-tube-sheet welding operations of this nature where the weld configuration is dependent on the internal and external pressure of the shielding gas, a pressure control device will be utilized. Presently only flow control valves, actuated by the weld programmer, are being used. Pressure is established by orificing the gas exit ports on both sides of the weld. Various pressure control valves are being evaluated for application on this type of welding.

While a slight thinning in wall thickness was expected at the weld joints, most welds showed a slight reinforcement on the outside of the tube (Figure 17). This reinforcement was apparently due to a slight restraint exerted by the tooling in welding. Boroscopic examination and also checks with a plug gage showed the welds to be generally flush with the I.D. of the tubes.

Review of the assembly operations of the first evaporator and the cost of inserting the tube bundle in the shell showed that this operation should be changed. The spacers and support rods were assembled and inserted into the center shell section (Figure 18). The diffusers, cone reducers, and fittings were added, except for the lower cross and split shell section. The tubes were

now inserted. Assembly time was considerably reduced and radiography made easier with less possibility of damage to the tubes.

Shell Welding

Prior to any shell welding, several weldors were certified for welding both 5%Cr-0.5%Mo and type 321 stainless steel under Section IX of the ASME Boiler and Pressure Vessel Code. Weld samples were prepared for both the shell section and the tube sheet to nozzle section. Those samples of 5%Cr-0.5%Mo were post heat treated per the recommended heat cycles for the sample thicknesses. Additional samples were sectioned and examined by the Quality Control Laboratories and found satisfactory.

Initial welding was done on the upper shell sections without consumable inserts. Early welds required an unusual amount of rework. Procedures were changed to use a consumable insert in making the root pass which helped considerably in controlling the "drop thru" in the inside of the shell sections. Several different weld rods were used as filler and 508-18 rod produced the cleanest weld deposit. Later as the weldors gained experience the rework required became consistently less.

Fewer difficulties were encountered in welding of the shell assemblies for the superheaters. This material was type 304 stainless steel and all weldors had years of experience on this alloy. Due to the limited clearance between the inside diameter of the shell and the tube bundle, control of "drop thru" caused some problems. Weld shrinkage, particularly on the superheater shells, while within tolerance, caused interference in the insertion of the spacer and support rod assembly into the shell. However, once the spacers passed the girth welds they were free and without interference from the shell.

In both the evaporator and superheater the design of the cone reducer caused the most difficulty, the weld joint being located at the break point on both ends of the reducer. X-ray film showed a sharp line at the edge of the weld. Grinding to blend caused thinning in shell section. Alignment of the joint often proved difficult. Again interpretation of the film caused much disagreement. Often this line was interpreted to be lack of fusion or undercut. Visual examination of the weld was often impossible in final stages of assembly because the areas in question were hidden by the tube bundle. The only solution was to remove the area of weld in question and reweld.

In the design of any future modular steam generators, the cone reducers should be changed to standard concentric reducers. This would allow welding and radiography in straight sections of equal diametric with no change in section.

Post heat treatment of the evaporator shell welds was done by coiling the area with electrically insulated copper wire, and wrapping with insulation blanket. Each joint was monitored with calibrated thermocouples. Power was supplied with conventional welding power supplies. The heat cycle was manually controlled by varying the power output of the power supplies as required by the time-temperature cycle.

Closure Welding

With completion of the tube-to-tube-sheet welding, the units were prepared for closure of the shell (Figure 19). The clam shell section was fitted and tack

welded in position between the shell and cone reducer. A root pass weld was made consuming the insert and fusing the edges of the weld preparation. All welds were now carefully inspected, using a boroscope and dental mirrors. Any questionable areas were reworked. Filler passes were now made completing the welds. On the evaporators, x-ray of the welds revealed unacceptable porosity and inclusions and also that the cover passes had caused "drop thru" and concavity. These sections were removed, refitted, and rewelded. With further minor rework acceptable welds were obtained.

At this point, the remaining spacers were positioned using tubes as spacers on the support rod. The support bulk head was welded to the support rods. The lower cross was then added and positioned. The support bulkhead legs were welded to the cross. The cross was welded to the upper cone. This weld again caused difficulty because of the configuration. The lower cone was then fitted as a clam shell and welded into position, closing the entire shell side. The shells were then sealed and purged with dry argon gas.

An attempt was made to ultrasonically inspect the welds prior to post heat treatment because it was less costly than x-ray. Due to weld head roughness and variation of internal reinforcement, this method proved insufficiently accurate. Therefore, an information x-ray inspection was made of each evaporator shell weld prior to heat treat. After heat treat each weld was x-rayed again, dye penetrant, and visually inspected. Only one shell weld required additional rewelding and consequently a reheat treat after approval by preliminary inspection.

Closure of the shell indicated the need for elimination of the split shell section. It proved too costly and difficult to weld. Sufficient free length of the tubes could be gained by splitting only the reducers at the tube sheet ends of the assembly. Thus, the end cross and tee could be left off, plus two reducers. By sliding the shell down to the lower end of the bundle, the tubes could be bowed laterally as needed without damage.

Manual welding resulted in considerable rework and proved to be too slow. The quality of the welds, while satisfactory to Section VIII of the ASME Boiler and Pressure Vessel Code, would have been better if a semi-automatic submerged arc process had been used. The time required for welding would also be reduced significantly.

All tubes were carefully cleaned by flushing with acetone and cleaning by blowing cotton wads through them. The nozzle was fitted to the tube sheet with an insert and tacked in position. As on shell welding, the weld area of the evaporators was preheated to a minimum of 350°F. This preheat was maintained until completion of the weld. Following this, a 6 in. extension was welded to the nozzle to allow plugging for the final hydrostatic test. Information x-ray shots were made, the module head assembly was placed in the modified furnace for post heat treatment. Upon completion of the heat treat operation, it was submitted for final inspection.

At all times when the units were not in work they were sealed and purged with dry inert gas. Since all radiographic inspection on the module heads was performed on week ends, thus avoiding shutdowns on other work for protection of personnel, each unit was protected against corrosion internally while waiting for inspection.

Hydrostatic Test

Preliminary planning included a hydrostatic test immediately after completion of the tube-to-tubesheet weld. It was intended to hold a closure plate over the tubesheet end by clamping to a collar below the neck of the tubesheet. The extreme torque required to seal the closure plate caused the collar to indent the sheet surface in the neck area. Rather than risk damage to the tubesheet, this method of test was abandoned.

After completion of the entire unit, plugs were welded in the nozzle extensions and the shell sealed with closure plates. Hydrostatic testing, rinsing, and drying was accomplished as shown by Figure 20. Test of the tube-to-tubesheet welds was monitored by observing shell side pressure while tube side was under test pressure.

Upon completion of the hydrostatic test and acceptance by the State Inspector, the outside of the evaporator modules were wire brushed and given two coats of primer paint.

The evaporators were originally sealed by clamping closure plates over the shell nozzles and then coating them with a fiberglass epoxy. Movement and temperature changes caused these seals to leak. Therefore, a plate was fitted over the nozzles and fused to the weld preparation of the nozzle. The units were then again purged with dry gas.

All modules were then mounted on a shipping and storage fixture and shipped to the SCTI for storage until installation for test.

Conclusions

With the aforementioned changes in design and fabrication, it is felt that the modular steam generator concept lends itself very well to production techniques. The fact that production techniques can be applied, that all operations would be repetitive, the ease of handling and the simplicity of design, all indicate that a steam generator using the modular concept can be produced economically. This would become more apparent as the size of the installation becomes greater.

Admittedly, the tube sheet weld configuration is slightly more costly; this cost can be overcome by production machining practices. Actual welding cost appears to be about the same as for a tube configuration. Inspection is more costly because of radiography. However, this cost is certainly justified in the greatly increased confidence in knowing the quality of the weld. It also provides other advantages in a crevice free joint, improved tubesheet design, and reliability.

Fabrication of the prototype modules has also demonstrated that this concept can be fabricated in almost any shop qualified to ASME code welding. No special facilities or equipment other than the use of a crane is required.

REFERENCES

1. D. F. Casey et al, "Final Design of Sodium-Heated, Modular Steam Generators for the SCTI," NAA-SR-9826 (February 1, 1965)
2. G. Kazlauskas, "A Device for Internal T.I.G. Arc Welding of Tube-to-Tube-Sheet Joints," NAA-SR-8541 (March 15, 1965)

SCTI - HIGH TEMPERATURE MODULE

SCTI - LOW TEMPERATURE MODULE

Figure 1

Figure 2 Modular steam generator tube sheet

Figure 3 Spacer casting

Figure 4 Spacer, support rod assembly with thermocouple outlet fitting

Figure 5 Tube bundle assembly with tube sheet in position

FIGURE 6

Figure 6 Installation of tube bundle in pit

Figure 7 Weld head and tube sheet section

Figure 8 Weld head in position for welding

ATOMICS INTERNATIONAL MANUFACTURING ENGINEERING PROCESS PROCEDURE				NO. 1120 REV		OPERATIONAL SEQUENCE	
				UNG			
				APPROVALS		DATE	
				WELDING ENGR			
				DEV SUPERV			
				MFG DIRECTOR			
				QUALITY CONTROL			
PROJECT	MOD STEAM GEN	SPEC NO.	N7593 PE001				
PROCESS	T.I.G.	CLASS					
ALLOY	5 CR 5 MO	FACILITY					
POWER SOURCE	MFG VICKERS	PROGRAM DATA		MIN	MEAN	MAX	
	MODEL D C ARC WELDER	PREPURGE TIME			60		
	SER NO. 4712A1	START CURRENT			25		
TORCH	MFG A I	START DIAL REF			30		
	MODEL	PREHEAT TIME			40		
NOZZLE	TYPE NOT REQUIRED	UPSLOPE TIME			3 0		
	SIZE	FIXTURE DELAY			4 5		
SHIELDING GAS	TYPE AIRCO 75	WELD CURRENT			63		
	BACKUP C.F.N. 3	WELD DIAL REF			65		
	COVER C.F.N. 8	WELD TIME			50		
FLUX	TYPE NOT REQUIRED	1 REV TIME			40		
	SPEC NO.	DOWNSLOPE TIME			10		
ELECTRODE	TYPE 2% T	SPEC NO.	EWTH	DOWNSLOPE CURRENT		15	
	DIA 1/16	EXT	90° B BEND	DOWNSLOPE DIAL REF		2 0	
	SHAPE POINTED	190		SEQUENCE STOP TIME		19	
FILLER METAL	ALLOY			POSTHEAT TIME		40	
	SPEC NO.			START ARC CURRENT DIAL		0	
	DIA			START ARC TIME DIAL		0	
	FEED I.P.M.			POSTPURGE TIME		60	
	DIAL REF			CARRIAGE SPEED I P.M.			
	S. DELAY (SEC)			CARRIAGE DIAL REF		74	
	C. FILL (SEC)						
WELD SETTINGS	AMP	POLARITY	DCSP				
	RANGE	VOLTS					
WELD POS	2G						
HEAT	PREHEAT 500	TIME	PREHEAT PASS				
	POSTHEAT 1325	TIME	1 HOUR				
	METHOD OVEN						
TOOLING	NO. T 6001052						
	TYPE AUTOMATIC						
INSPECTION REQ	LAB NO.						
	VISUAL	X					
	DYE CHECK	X					
	X-RAY	X					
	LEAK TEST	X					
	ULTRASONIC						

OPERATIONAL SEQUENCE	
1. MACHINE PIPE ENDS TO SIZE	
2. CLEAN WELD SURFACES WITH ACETONE	
3. INSTALL AUTOMATIC HEAD TO TUBE I D	
4. LOCATE ELECTRODE FLUSH WITH WELD JOINT	
5. REMOVE AUTOMATIC WELDING HEAD	
6. INSERT GUIDE PIN FOR PROPER JOINT ALIGNMENT	
7. INSERT STEEL WOOL WADDING BELOW THE JOINT	
8. FINAL ACETONE CLEAN ALL MATING PARTS	
9. INSTALL O D CHILL AND GAS MANIFOLD	
10. INSERT WELD HEAD	
11. MAKE FINAL PROGRAM DATA CHECK	
12. WELD COMPLETE	
13. WIRE BRUSH I D AND O D	
14. REMOVE STEEL WOOL WADDING	
15. SUBMIT FOR INSPECTION	
16.	
17.	
18.	
19.	
20.	

SKETCHES (Edge preparation, passes, joints, etc)

Figure 9

ATOMICS INTERNATIONAL MANUFACTURING ENGINEERING PROCESS PROCEDURE				NO 1069 REV DWG 7593 449306 B			APPROVALS			DATE		
PROJECT	STEAM GEN.	SPEC NO.	SEC. VIII ASME	WELDING ENGR	DEV SUPVR							
PROCESS	AUTO TIG	CLASS		MFG DIRECTOR								
ALLOY	321 S S	FACILITY	789	QUALITY CONTROL								
POWER SOURCE				MFG VICKERS			PROGRAM DATA			MIN	MEAN	MAX
				MODEL D C PROGRAMMER			PREPULSE TIME				60	
				SER NO. 47121A1			START CURRENT				15	
TORCH				MFG A I			START DIAL				30	
				MODEL			PREHEAT TIME					
NOZZLE				TYPE NOT REQUIRED			UPSLOPE TIME				3 0	SEC
				SIZE			FIXTURE DELAY				4 5	
SHIELDING GAS				TYPE			WELD CURRENT				70	
				BACKUP C.F.W. ARGON 3 4			WELD DIAL				70	
				COVER C.F.W. AIRCO 75 8 10			WELD TIME				50	
FLUX				TYPE			1 REV TIME				40	
				SPEC NO.			DOWNSLOPE TIME				10 9	
ELECTRODE				TYPE 2%			DOWNSLOPE CURRENT				10	
				DIA 1/16 EXT			DOWNSLOPE DIAL				10	
				SHAPE POINTED 190			SEQUENCE STOP TIME				20	
FILLER METAL				ALLOY			POSTHEAT TIME					
				SPEC NO.			START ARC CURRENT DIAL				0	
				DIA			START ARC TIME DIAL				0	
				FEED I.P.M.			POSTPURGE TIME				60	
				DIAL REF			CARRIAGE SPEED I.P.M.					
				S. DELAY (SEC)			CARRIAGE DIAL				76	
				C. FILL (SEC)								
WELD SETTINGS				AMP			POLARITY DCSP					
				RANGE			VOLTS 18					
WELD POS				2G								
HEAT				PREHEAT			TIME					
				POSTHEAT			TIME					
				METHOD								
TOOLING				NO. T 6001052								
				TYPE AUTOMATIC								
INSPECTION REQ				LAB NO.								
				VISUAL			X					
				DYE CHECK			X					
				X-RAY			X					
				LEAK TEST			X					
				ULTRASONIC								

OPERATIONAL SEQUENCE

1. MACHINE PIPE ENDS TO SIZE
2. CLEAN WELD SURFACES WITH ACETONE
3. INSTALL AUTOMATIC HEAD INTO TUBE I D
4. LOCATE ELECTRODE RADIUS FLUSH WITH WELD JOINT SURFACE ON
5. MODULAR HEAD
6. REMOVE AUTOMATIC WELD HEAD
7. INSERT GUIDE PIN FOR PROPER TUBE WELD JOINT ALIGNMENT
8. FINAL ACETONE CLEAN ALL MATING PARTS (HANDLING PARTS AFTER
9. CLEANING WITH WHITE GLOVES)
10. INSERT WELD HEAD
11. MAKE FINAL PROGRAM DATA CHECK
12. WELD COMPLETE
13. WIRE BRUSH O D & I D
14. SUBMIT FOR INSPECTION
- 15.
- 16.
- 17.
- 18.
- 19.
- 20.

SKETCHES (Edge preparation, passes, joints, etc)

FIGURE 10

FORM 725-K NEW 1 64

Figure 10

Figure 11 Radiography of tube to tube sheet welds

Figure 12 Post heat treat of tube to tube sheet welds

Figure 13 Tubes spread for welding, lower end

ATOMICS INTERNATIONAL MANUFACTURING ENGINEERING PROCESS PROCEDURE				NO. 1120 R REV				OPERATIONAL SEQUENCE			
				DWG		APPROVALS		DATE			
PROJECT	MOD	STEAMGEN	SPEC NO.	N7593-PE001							
PROCESS	T.I.G.		CLASS								
ALLOY	5 CR 5 MO		FACILITY	QUALITY CONTROL							
POWER SOURCE	MFG VICKERS			PROGRAM DATA				MIN	MEAN	MAX	
	MODEL D.C ARC WELDER			PREPURGE TIME					60		
TORCH	SER NO. 47121A1			START CURRENT					25		
	MFG A.I.			START DIAL REF					30		
NOZZLE	MODEL			PREHEAT TIME					40		
	TYPE NOT REQUIRED			UPSLOPE TIME					3 0		
SHIELDING GAS	SIZE			FIXTURE DELAY					4 5		
	TYPE AIRCO 75			WELD CURRENT					58		
FLUX	BACKUP C.F.N. 2-3			WELD DIAL REF.					60		
	COVER C.F.N. 11-12			WELD TIME					50		
ELECTRODE	TYPE NOT REQUIRED			I REV TIME					40		
	SPEC NO.			DOWNSLOPE TIME					10		
FILLER METAL	TYPE 2% T			DOWNSLOPE CURRENT					15		
	DIA 1/16" EXT 90° BEND			DOWNSLOPE DIAL REF.					2 0		
WELD SETTINGS	SHAPE POINTED 190			SEQUENCE STOP TIME					19		
	ALLOY			POSTHEAT TIME					40		
WELD POS	SPEC NO.			START ARC CURRENT DIAL					-0		
	DIA			START ARC TIME DIAL					-0		
HEAT	FEED I.P.M.			POSTPURGE TIME					60		
	DIAL REF			CARRIAGE SPEED I.P.M.							
TOOLING	S. DELAY (SEC)			CARRIAGE DIAL REF					74		
	C. FILL (SEC)										
INSPECTION REQ	AMP			POLARITY DCSP							
	RANGE			VOLTS							
	PREHEAT 500 TIME PREHEAT PASS										
	POSTHEAT 1325 TIME 1 HOUR										
	METHOD OVEN										
	NO. T-6001052										
	TYPE AUTOMATIC										
	LAB NO.										
	VISUAL X										
	DYE CHECK X										
	X-RAY X										
	LEAK TEST X										
	ULTRASONIC										

REPAIR PROCEDURE			
16.			
17.			
18.			
19.			
20.			

SKETCHES (Edge preparation, passes, joints, etc)

Figure 14

ATOMICS INTERNATIONAL										NO. 1069		REV		OPERATIONAL SEQUENCE		
MANUFACTURING ENGINEERING PROCESS PROCEDURE										DWG 7593-449306 "B"						
										APPROVALS		DATE				
										WELDING ENGR						
										DEV SUPER						
										MFG DIRECTOR						
										QUALITY CONTROL						
PROJECT	STEAM GEN.			SPEC NO.	SEC VIII ASME											
PROCESS	AUTO T.I.G.			CLASS												
ALLOY	321 S.S.			FACILITY	789 SHOP											
POWER SOURCE	MFG VICKERS				PROGRAM DATA				MIN	MEAN	MAX					
	MODEL D. C. PROGRAMMER				PREPURGE TIME					60						
	SER NO.				START CURRENT					15						
TORCH	MFG A. I.				START DIAL REF					30						
	MODEL				PREHEAT TIME											
NOZZLE	TYPE NOT REQUIRED				UPSLOPE TIME					3 0	SEC					
	SIZE				FIXTURE DELAY					4 5						
SHIELDING GAS	TYPE				WELD CURRENT					65						
	BACKUP C.F.W. ARGON 2-3				WELD DIAL REF.					65						
	COVER C.F.W. AIRCO 75 11-12				WELD TIME					50						
FLUX	TYPE				I REV TIME					40						
	SPEC NO.				DOWNSLOPE TIME					10 9						
ELECTRODE	TYPE		2%	SPEC NO.	EWTH 2		DOWNSLOPE CURRENT			10						
	DIA		1/16	EXT	90° BEND		DOWNSLOPE DIAL REF.			10						
	SHAPE		POINTED		190		SEQUENCE STOP TIME			20						
FILLER METAL	ALLOY				POSTHEAT TIME											
	SPEC NO.				START ARC CURRENT DIAL					-0						
	DIA				START ARC TIME DIAL					-0						
	FEED I.P.M.				POSTPURGE TIME					60						
	DIAL REF				CARRIAGE SPEED I.P.M.											
	S. DELAY (SEC)				CARRIAGE DIAL REF.					76						
	C. FILL (SEC)															
WELD SETTINGS	AMP		POLARITY	DCSP												
	RANGE		VOLTS	18												
WELD POS	2G															
HEAT	PREHEAT		TIME													
	POSTHEAT		TIME													
	METHOD															
TOOLING	NO.		T 6001052													
	TYPE		AUTOMATIC													
INSPECTION REQ	LAB NO.															
	VISUAL		X													
	DYE CHECK		X													
	X-RAY		X													
	LEAK TEST		X													
ULTRASONIC																

1. MACHINE PIPE ENDS TO SIZE

2. CLEAN WELD SURFACES WITH ACETONE

3. INSTALL AUTOMATIC HEAD TO TUBE I.D.

4. LOCATE ELECTRODE FLUSH WITH WELD JOINT

5. REMOVE AUTOMATIC WELDING HEAD

6. INSERT GUIDE PIN FOR PROPER JOINT ALIGNMENT

7. INSERT STEEL WOOL WADDING BELOW THE JOINT

8. FINAL ACETONE CLEAN ALL MATING PARTS

9. INSTALL O.D. CHILL AND GAS MANIFOLD

10. INSERT WELD HEAD

11. MAKE FINAL PROGRAM DATA CHECK

12. WELD COMPLETE

13. WIRE BRUSH I.D. AND O.D.

14. REMOVE STEEL WOOL WADDING

15. SUBMIT FOR INSPECTION

16.

17. REPAIR PROCEDURE

18.

19.

20.

SKETCHES (Edge preparation, passes, joints, etc)

FIGURE 15

Figure 15

Figure 16 Spliced and repaired tubes

Figure 17 Completed tube to tube sheet welds

Figure 18 Center shell section of modular steam generator

Figure 19 Clam sheild weld

Figure 20 HYDROSTATIC TEST SET-UP & PROCEDURE MODULAR STEAM GENERATOR

LOW TEMPERATURE CONFIGURATION (5 Cr-1.2 Mo)

1. MIX 1.8 OZ. SODIUM NITRITE GALLON OF DISTILLED WATER
100 GALLONS REQUIRED. MIX 5.8 OZ. OF SODIUM NITRITE WITH
5 GALLONS WATER IN BOTTLES AND TRANSFER TO TWO 50-GALLON
DRUMS (LINED)

2. PERFORM COMBINED TESTS OF TUBE SIDE AND SHELL SIDE CON-
CURRENTLY AFTER MAKING CLOSURE AND NOZZLE WELDS. THE TESTS
ARE TO BE WITNESSED BY THE STATE CODE INSPECTOR

2.1 TEST PROCEDURE TUBE SIDE

- a. INSTALL COVERS TUBE SIDE AND ASSEMBLE PLUMBING
PER SKETCH
- b. EVACUATE UNIT SUFFICIENTLY TO BRING WATER LEVEL IN
FILL TUBE ABOVE VALVE. CLOSE VALVE.
EVACUATE TO 29.8 in. MERCURY (APPROX.)
- d. OPEN FILL VALVE. CONTINUE PUMPING UNTIL BUBBLE FREE
WATER FLOWS INTO VACUUM PUMP TRAP
- e. CLOSE VACUUM LINE VALVE
- f. START PRESSURE PUMP
- g. OPEN PRESSURE VALVE
- i. PRESSURIZE TO 7,550 psig
- FLUSH WITH DISTILLED WATER UNTIL THE CONDUCTIVITY OF
THE FINAL RINSE WATER IS 25 MICROMHOS OR LESS
- j. DRY BY EVACUATING

2.2 TEST PROCEDURE SHELL SIDE

- a. INSTALL COVERS AND ASSEMBLE PLUMBING PER SKETCH
- b. CONNECT PUMPS AND PLUMBING TO SHELL SIDE AND CON-
DUCT TEST AS IN 2.1 a THROUGH g
- c. PRESSURIZE TO 780 psig
- d. RELEASE PRESSURE AND DRAIN
- e. FLUSH WITH DISTILLED WATER UNTIL THE CONDUCTIVITY OF
THE FINAL RINSE WATER IS 25 MICROMHOS OR LESS
- i. DRY BY EVACUATING WHILE HEATING WATER ENTRAPMENT
AREAS TO 250°F. EXIT AIR DEW POINT TO BE 10°F OR LESS
- g. FILL BOTH SYSTEMS WITH DRY NITROGEN AND SEAL

HIGH TEMPERATURE CONFIGURATION (SS)

1. MIX 1.8 OZ. OF SODIUM NITRITE GALLON OF DISTILLED WATER
100 GALLONS REQUIRED. MIX 5.8 OZ. OF SODIUM NITRITE WITH
5 GALLONS WATER IN BOTTLES AND TRANSFER TO TWO 50 GALLON
DRUMS (LINED)

2. PERFORM COMBINED TESTS OF TUBE SIDE AND SHELL SIDE CON-
CURRENTLY AFTER MAKING CLOSURE OR NOZZLE WELDS. THE TESTS
ARE TO BE WITNESSED BY THE STATE CODE INSPECTOR

2.1 TEST PROCEDURE TUBE SIDE

- a. INSTALL COVERS TUBE SIDE AND ASSEMBLE PLUMBING
PER SKETCH
- b. EVACUATE UNIT SUFFICIENTLY TO BRING WATER LEVEL IN
FILL TUBE ABOVE VALVE. CLOSE VALVE
- c. EVACUATE 29.8 in. MERCURY (APPROX.)
- d. OPEN FILL VALVE. CONTINUE PUMPING UNTIL BUBBLE FREE
WATER FLOWS INTO VACUUM PUMP TRAY

e. CLOSE VACUUM LINE VALVE

- f. START PRESSURE PUMP
- g. OPEN PRESSURE VALVE
- h. PRESSURIZE TO 7,060 psig
- i. FLUSH WITH DISTILLED WATER UNTIL THE CONDUCTIVITY OF
THE FINAL RINSE WATER IS 25 MICROMHOS OR LESS
- h. DRY BY EVACUATING

2.2 TEST PROCEDURE SHELL SIDE

- a. INSTALL COVERS AND ASSEMBLE PLUMBING PER SKETCH
- b. CONNECT PUMPS AND PLUMBING TO SHELL SIDE AND CON-
DUCT TEST AS IN 2.1 a THROUGH g
- c. PRESSURIZE TO 1,210 psig
- d. RELEASE PRESSURE AND DRAIN
- e. FLUSH WITH DISTILLED WATER UNTIL THE CONDUCTIVITY OF
THE FINAL RINSE WATER IS 25 MICROMHOS OR LESS
- f. DRY BY EVACUATING WHILE HEATING WATER ENTRAPMENT
AREAS TO 250°F. EXIT AIR DEW POINT TO BE 10°F OR LESS
- g. FILL BOTH SYSTEMS WITH DRY NITROGEN AND SEAL

- ① INLET HIGH TEMPERATURE 3-1/2 X DOUBLE EXTRA HEAVY ASTM A 312 TYPE 321 H1/6 in. LONG
- OUTLET HIGH TEMPERATURE 3-1/2 X DOUBLE EXTRA HEAVY ASTM A 312 TYPE 321 H1/6 in. LONG
- INLET LOW TEMPERATURE 3-1/2 X DOUBLE EXTRA HEAVY ASTM A 106 GRADE B 6 in. LONG
- OUTLET LOW TEMPERATURE 4 X 0.671 WALL AST A379 P22 OR F22 6 in. LONG
- CONFIGURATION SHOWN IS FOR 5 CHROME 1/2 MOLYBDENUM ASSEMBLY (LOW TEMPERATURE)

Sodium Heated Steam Generator Design and Materials
Examination of a Model Steam Generator

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ABSTRACT

The completed Preliminary Designs of the Full-Size Steam Generator and the Prototype Steam Generator are described. A test program to assess the compatibility of Croloy 2-1/4 and 316 SS, particularly with reference to the design parameters of the full size steam generator is described. Results of a metallurgical examination of piping removed from a model sodium heated steam generator during intermediate shutdown after 1000, 2000, and 4000 operating hours are presented.

A gradual decarburization of the commercial Croloy 2-1/4 piping exposed at 1000F was detectable after 4000 hours to about 10 mils metallographically, and somewhat deeper by microhardness techniques. Tempering effects and gradients in the as-received material complicate assessment of the extent of the slight decarburization.

Electron microprobe examination disclosed composition gradients, within about 0.3 mils of the sodium-steel interface, in both the as-received Croloy 2-1/4 (attributed to oxidation during mill treatment) and the 4000 hour sample. Surface (at 1 micron) compositions indicate the following changes during exposure:

<u>Wt %</u>	<u>Original Surface</u>	<u>4000 Hour Surface</u>	<u>Interior</u>
Cr	0.80	1.98	2.30
Mo	1.00	1.00	1.00
Si	0.10	0.24	0.35
Ni	0.22	1.00	0.12
Cu	0.39	Not Detectable	0.09

No preferential mass transfer was detectable in the 316 SS samples, exposed at either 1000F or 1100F, although some uniform loss is probable. The material is characterized by persistent banding of chromium, nickel, and molybdenum.

Gradual sensitization, which is further advanced at the higher temperature, accounts for observed metallographic changes of the 316 SS piping. Accelerated sensitization near the surface correlates

with artifacts of heavy deformation of the surface region. Carbide formation is thus attributed to precipitation of carbon originally dissolved in the supersaturated austenite, not to carburization.

OBJECTIVE:

The objective of this Contract is to develop a large Sodium-Heated Steam Generator of improved design. This Steam Generator Design will be available for use in the Atomic Energy Commission Sodium Reactor Development Program which has the ultimate objective of developing reliable, economical, large central station nuclear power plants.

SCOPE:

The overall scope of work under this Contract is as follows:

Phase I:

- A. Preliminary Design of Full-Size Sodium-Heated Steam Generator
- B. Supporting Research and Development Work
- C. Preliminary Design of Prototype Steam Generator

Phase II:

- A. Detail Design of Prototype Steam Generator
- B. Fabrication of Prototype Steam Generator for Installation and Testing at SCTI
- C. Final Design Report Relating the Performance of the Prototype Steam Generator to the Design of the Full-Size Steam Generator.

Work was begun on this Contract on March 20, 1963, and the Present Status of the various projects is as follows:

1. Design Work:

The Preliminary Design of the Full-Size Steam Generator has been completed, and the Preliminary Design Report was issued July 1, 1964.

2. The Preliminary Design of the Prototype Steam Generator has been completed and the Preliminary Prototype Design Report will be issued soon.

3. Fabrication:

Fabrication has not yet begun on the Prototype Steam Generator, although some material has been ordered so that fabrication can start without delay. It is planned to begin fabrication on July 1, 1965.

Figure 1 is a simplified sectional side view of the Full-Size Steam Generator. Sodium enters the steam generator through two sodium inlet nozzles only one of which is shown on the figure, flows downward through a distributor system that distributes the sodium out over the top of the coiled superheater bundle. The sodium flows downward in crossflow over the coils of the superheater bundle and continues downward over the coils of the boiler bundle, and leaves through the sodium outlet nozzle at the bottom of the steam generator.

Feedwater enters the steam generator through four feedwater inlet nozzles, one of which is shown on the figure, flows downward through annular rows of downcomer tubes and enters the boiler bundle. As the water flows upward through the coils of the boiler bundle it is heated to saturation temperature, boils, and is slightly superheated. The steam continues upward through the boiler riser tubes and leaves through the four boiler outlet headers. External piping, not shown on the figure connect the boiler outlet headers and superheater inlet headers. In this piping is located the valves for the start-up system that is commonly used on once-through boilers to allow warming up and rolling the steam turbine early and allows matching of the steam temperature at the turbine with the turbine nozzle block temperature after a turbine trip-out. Steam from the

interconnecting piping enters the four superheater inlet headers, flows downward through annular rows of superheated downcomer tubes to the bottom of the superheater coil bundle. The steam is superheated as it flows upward through the coils of the superheater bundle and leaves through the superheater riser tubes and the superheater outlet headers. The steam pressure at the superheater outlet is 2450 psig and 1000F the feedwater temperature is 530F.

The outer shell and liner of the steam generator is made of Croloy 2-1/4 material, as are the tubes in the boiler bundle. The tubes of the superheater are made of Type 316 Stainless Steel because of its superior strength at high temperatures.

Figure 2 shows the Prototype Steam Generator. It was designed to model as closely as possible the performance and fabrication problems of the Full-Size Steam Generator. As one can see it has essentially the same general arrangement of heating surface. The tube diameters, thickness, material, and heated length are the same in the Prototype Steam Generator as the Full-Size Steam Generator. Since the Prototype Steam Generator is approximately 1/30th. the size of one of the Full-Size Steam Generators it has proportionally fewer tubes. The Prototype Steam Generator has twenty-five tubes in the boiler bundle and forty-five tubes in the superheater bundle. It has only one each of the various tubesheets and nozzles. Because of the similarity of the heating surface the Prototype Steam Generator should model the performance of the Full-Size Steam Generator quite closely.

It will be noted that the feedwater inlet header and tubesheet appeared to be larger than the other tubesheets. This tubesheet is an exact model of one of the four feedwater inlet tubesheets of the Full-Size Steam Generator. This tubesheet was modeled full-size because the feedwater inlet tubesheets undergo the most severe thermal stresses. Since the Prototype Steam Generator does not have enough tubes to completely fill this size of tubesheet the remaining tube holes in the full pattern are blind drilled to model the flexibility of the tubesheet. Thermocouples and strain gages will be installed in this tubesheet and thermal sleeve where it attaches to the vessel to obtain steady state and transient data to verify the stress analysis calculations that have been made.

Figure 3 is an outside view of the Full-Size and Prototype Steam Generators drawn to the same scale to show the relative size. It will be noted that the Full-Size Steam Generator is roughly twice as large in diameter and twice as high as the Prototype Steam Generator.

Supporting Research and Development Work

One of the projects being carried on under the R&D portion of this contract is the operation of a one-tube model steam generator simulating a Full Size plant including steam generator, intermediate heat exchanger, piping, and pump. The model is constructed of the same materials proposed for the full size unit, Croloy 2-1/4 (2-1/4 Cr - 1 Mo) and AISI Type 316 stainless steel (17Cr-12Ni-2.5Mo). In so far as possible, the surface to volume ratios, mass flow rate, temperatures, pressure, and other physical and operating parameters of the model have been selected to simulate the full size steam generator.

The circulation of liquid metals at elevated temperature usually results in observable dissolution of the constraining walls at some positions and subsequent deposition of material at other positions. These phenomena may appear in many guises, all of which may be called mass transfer, if temperature gradients, composition (activity) in-

homogeneities, or both are present. Dissolution may occur either as a rather uniform removal of metal or, more commonly, as a preferential leaching of certain constituents. Grain boundaries may be preferentially attacked. Deposition may occur in several ways, including physical plugging.

Mass transfer is fairly well understood, qualitatively. Diffusion rates, composition limits and activities within the various phases, possible interface controls or diffusion barriers, and especially the temperature dependence of the aforementioned are generally recognized as being of critical importance in determining the type of extent of mass transfer. At present, however, both lack of basic information and the complexities of geometry, temperature distribution, the large number of chemical species involved, and so forth, make predictions of the extent of mass transfer in a realistic steam generator uncertain.

Transfer of carbon from the ferritic Croloy 2-1/4 to the austenitic 316 SS is of particular concern because this decarburization-carburization reaction is expected, on the basis of previous work, to be the fastest and most extensive process to occur in the secondary sodium system. Tube dimensions are not seriously changed by this process; carbon transfer resembles a leaching rather than uniform dissolution. However, the properties of both materials may deteriorate, the decarburized Croloy 2-1/4 becoming weaker and the stainless steel becoming embrittled. If significant fractions of wall thickness experience this type of transfer, serious loss of structural integrity may result. Mass transfer involving wall thinning can lead to premature failure because of increasing stresses associated with reduced cross sectional areas.

Operation of the model sodium heated steam generator is a proof test for a proposed combination of materials and operating conditions. Following 8000 hours of operation, the model will be destructively examined to determine the extent of mass transfer.

Small sections of piping have been removed during intermediate shutdown of the model to check the progress of any mass transfer at two positions. This paper presents the information derived from these samples. Final interpretation of the model test program and the assessment of mass transfer must await the more extensive results of the final destructive examination.

EXPERIMENTAL PROCEDURES

Sample History

The exposed samples discussed in this report are sections of seamless commercial 1-1/4" schedule 40 pipe removed from two isothermal U-bend sections of a model sodium heated steam generator during intermediate shutdowns following 1000, 2000 and 4000 operating hours. The locations of the high temperature (1100F) U-bends are indicated in the schematic plan, Figure 4a. Figure 4b is a photograph of the model steam generator. The 1100F U-bend was constructed of a single section of 316 SS. The 1000F U-bend contained a welded juncture between 316 SS and Croloy 2-1/4. The Croloy 2-1/4 sample was at the maximum temperature for this material. Similarly the 1100F 316 SS samples were from the maximum temperature zone for this material. Samples of the as-received pipe used in these sections have also been examined.

During the scheduled shutdowns, the sodium was drained from the steam generator and replaced with an inert gas. The U-bends were removed and rinsed with alcohol. Sections of pipe, about 3" to 4" long, were sawed off each leg of the U-bends. The remaining portion of the U-bends were welded in place and reinsulated. The hot and cold traps

have maintained the oxygen content of the sodium at less than 30 ppm, as indicated by plugging temperature.

Thus, four samples of piping were removed during each shutdown: two samples of 316 SS (which were essentially duplicates) from the 1100F U-bend and one sample each of 316 SS and Croloy 2-1/4 from the 1000F U-bend.

Two special 316 SS samples were prepared by heating pieces of the as-received pipe in air at 1250F for 16 and 48 hours. These special samples demonstrated the susceptibility of the 316 SS to sensitization, thus accounting for observed carbide formation in the loop samples. Small samples of each of the four Croloy 2-1/4 conditions were given a special austenitizing heat treatment (1650F, 15 minutes, rapid cool) in a vacuum furnace. The special Croloy 2-1/4 samples were prepared to minimize the tempering effects found in the loop samples but to maintain any carbon gradients. Tabulated identification of the samples is given in Table I.

Specimen Preparation

The inner surfaces (the sodium side) of all pipe samples were visually examined. Longitudinal and transverse sections of each sample were separately wrapped in silver foil and mounted in Bake-lite. The mounted specimens were polished using standard procedures for metallographic preparation.

Photomicroscopy

In general, standard etching and photomicroscopy practices were followed. Except as noted, Croloy 2-1/4 specimens were etched in Nital, the 316 SS specimens in Marble's reagent. Although both transverse and longitudinal sections were examined, in most cases only the microstructures of the transverse sections were recorded at both 100X and 500X. Additional photomicrographs were made when other magnifications, etches, or the recording of special features seemed appropriate.

Hardness Measurements

Diamond Pyramid Hardness measurements, using a 100 gram load and standard techniques, were made on the four Croloy 2-1/4 samples and the four 316 SS (1100F) samples. Two traverses were made on each specimen from the inner surface to about 40 mils from the surface. About 30 indents were made on each Croloy 2-1/4 specimen, about 15 on each of the 316 SS specimens.

Several hardness transverses were also made across the special austenitized Croloy 2-1/4 specimens, using a Knoop indenter with a 100 gram load.

Microhardness measurements were made on polished and lightly etched surfaces so that microstructural features could be observed during testing.

Microprobe Examination

The as-received and 4000 hour metallographical specimens of Croloy 2-1/4 and 316 SS (1100F) were repolished and examined by electron microprobe X-ray analysis. Point counting was employed in one micron steps from the inner surface to roughly one mil from the surface. Several traces were made on each specimen, three elements being analyzed with each trace. Several continuous profile scans were made over much larger distances, in many cases extending from the inner surface about 25 mils. Analyses were made on both materials for iron, chromium, nickel, silicon, molybdenum, and copper. Backscattered electron traces were made with most point counting runs to pinpoint the surface position. Copper and nickel analyses of the Croloy 2-1/4 were also made near the outer surface. Copper and nickel contents of the Croloy 2-1/4 were determined from an

appropriate series of standards. Microprobe readings at the interior of the tube were normalized to the bulk chemical analysis to determine percentages of other elements.

OBSERVATIONS AND RESULTS

Croloy 2-1/4

a) Summary

A slight gradual decarburization of Croloy 2-1/4 exposed to 1000F sodium in this system has been detected. Tempering effects and original gradients complicate exact assessment of the rather small carbon loss detected. Partial decarburization is metallographically detectable to about 10 mils after 4000 hours. The greater affected depth indicated by microhardness measurements involves a very small carbon loss and may be considered of little significance.

Microprobe results indicate pick-up of nickel (to about 1% at the surface) and copper depletion during exposure. Leveling out of initial chromium and silicon gradients was also observed, part of which may be actual pick-up.

b) Macroscopic

Visual examination of the inner surfaces (sodium side) of the removed pipe samples did not reveal any signs of mass transfer. Neither deposits nor dendritic growths were apparent.

c) Microstructure

The microstructure of the ID surface region of the as-received Croloy 2-1/4 piping is shown in Figure 5a. This can be compared with Figures 5b, 5c, and 5d, which show the microstructure after 1000, 2000, and 4000 hours at 1000F respectively.

The carbides in the as-received mill processed piping are, for the most part, in colonies of poorly defined pearlite. The apparent area per cent carbides on the polished and etched surface slightly decreases between the interior and inner surface of the pipe, indicating a slight decarburization during manufacture of the pipe.

The most obvious structural changes during loop operation are attributed to the tempering effects of exposure at 1000F for long periods of time. Although these changes are visible in Figures 5a through 5d, Figures 6a and 6b of the center of the as-received and 4000 hour specimens illustrate the tempering effects more clearly. The carbide colonies have quite different appearances as carbides gradually spheroidize and presumably change composition as well as size and number with increasing time at temperature. The development of a dot-like carbide in what was initially clear ferrite grains is noteworthy. Tempering changes are unrelated to the environment and must not be attributed to the sodium in the system. However, tempering may influence the course of any decarburization and certainly complicates metallographic assessment of decarburization.

No completely carbide free rim has developed even after 4000 hours exposure to sodium at 1000F. The apparent progressive decrease in area per cent of carbides on the metallographically prepared surface between unaffected interior and surface, as illustrated in Figure 5 does indicate partial decarburization. The actual carbon loss to account for these changes, which are detectable to about 10 mils after 4000 hours (with questionable detection, in view of the complications, to perhaps 15 mils) and a lesser distance after 2000 hours, is small. Detection of carbon loss in the 1000 hour specimen is uncertain.

The microstructures of the briefly austenitized specimens consisted of ferrite in the surface regions and bainite in the in-

terior, the transition region consisting of a mixed structure. The small ferrite rim on the as-received material indicates that this material was initially decarburized. The exposed samples had, as a group, a larger ferrite rim although an increased depth with increasing exposure time is detectable.

d) Microhardness

The results of microhardness testing of the Croloy 2-1/4 series of loop specimens are summarized in Figure 7. The bar graphs were derived from envelopes of the original data, plotted as hardness versus distance. The large scatter for each specimen is for the most part real. The hardness depends on the structure, which may be viewed as a non-uniform dispersion of hard carbides in a soft ferrite matrix, in the immediate vicinity of the indent. At almost any distance from the surface an indent might fortuitously be made in a carbide free region or conversely might land on a local carbide colony. However, the probability of encountering carbides decreases as one approaches the decarburized surface.

The extrapolated surface hardness of all exposed specimens is about the same (145 DPH). This may be taken as the characteristic ferrite hardness. The tempering effects are shown by the decreasing hardness with increasing exposure time of the interior of the pipe which is not influenced by the presence of sodium. The decrease in width of the scatter band is another indication of tempering. The gradual drop in hardness between interior and surface of the specimens is an indication of the extent of decarburization although the affected depth is difficult to exactly specify because of the scattered nature of the data and because no natural demarkation exists between unaffected material and decarburized material.

The special austenitized specimens were prepared to minimize the hardness scatter resulting from the type of carbide distribution in the loop samples and to minimize the complicating tempering effects which are of only passing interest. Results of microhardness traverses across these special specimens are summarized in Figure 8. As in Figure 7, the results are shown in bar graph form, the envelopes of the original data being used to construct the bars. Some leveling out of carbon gradients may be expected during austenitizing. However, the data show that carbon redistribution has been local, that is, the carbide-ferrite aggregates of the untreated samples have been for the most part replaced by a more uniform structure, bainite, but gradients occurring over several mils are still detectable. The hardness decrease near the surface of all specimens is a measure of decarburization. The area between curves on a hardness-distance plot is a better measure of carbon loss than a distance of affected depth. Thus, although an almost constant hardness, for all samples, does not occur until some 25 mils from the surface, clearly a progressive decrease, with time, of the area under the hardness-distance curves does indicate slight gradual carbon loss. It should be noted that only differences between exposed and as-received material may be attributed to carbon loss during exposure. A significant part of the gradients must be attributed to the original decarburized condition. Hardness differences at depths greater than 20 mils are of little consequence, considering residual tempering differences and probable experimental data scatter. Small hardness gradients well away from the surface may be attributed to the small temperature gradient across the pipe wall during operation. This would cause a "tempering gradient" which would result in a slight hardness decrease between inner and outer pipe surfaces, independent of other effects.

e) Microprobe

Microprobe results of a point count examination of the sodium side of the as-received and 4000 hour samples are summarized in Figures 9a and 9b respectively. The many scanning traces gave no more real information than is contained in Figure 9; that is, composition throughout most of the material is quite uniform except within about 0.3 mils of the surface. Thus, the more accurate point counting results are given greater emphasis. The gradients in the surface region of the original material are attributed to diffusion which occurred during oxidation during some heat treatment in the manufacture of the pipe. The chromium and silicon would be expected to be preferentially oxidized depleting, as observed, the material next to the oxide. Nickel and copper would be rejected from the oxide, preferring to remain in the metallic constituent, and thus are piled up next to what was at one time an oxide-steel interface. Because of the vagaries of oxidation, exact reproduction of the data obtained on rather widely separated traces should not be expected. However, quite similar gradients were found near the outer surface of the as-received material.

When examining the composition gradients in the 4000 hour specimen one must, as with the decarburization information, compare the final gradients with the pre-existing gradients. Thus, although the surface of the 4000 hour sample is depleted in chromium, substantial chromium enrichment (compared with the original surface) is nonetheless indicated. A surface peak would normally be expected if chromium were being picked up by the steel. The absence of such a peak may be indicative of a small availability of chromium in the sodium, indicating a rather low chromium activity. However, the contribution of a simple diffusion leveling out of the chromium gradient is uncertain and may account for the bulk of the observed changes. Similar comments apply to the leveling out of the silicon gradient. The nickel peak and copper depletion are indications of mass transfer. However, the effects are certainly small, although in the direction anticipated.

No indication of composition changes of a magnitude needed to cause austenite formation has been found. As in the as-received material, only slight variations from trace to trace were found, and this must include any effects of the inherited differences in initial gradients. It is more appropriate to consider these differences between traces over different regions as real and thus to study individual traces rather than the average of several separated traces. The data presented were obtained from repeated traces over the same area; incomplete coverage of other regions was made to check the typical aspect of the information. The changes may be represented by comparing surface compositions (at a depth of 1 micron) with the average composition (at depths greater than 9 microns) as given in Table II.

316 Stainless Steel

a) Summary

No preferential mass transfer effects have been found in the 316 SS pipe, although some uniform removal of material is probable. The steel does sensitize readily and is further characterized by banding, both phenomena being unrelated to the sodium environment.

b) Microstructure

The microstructures of the as-received and 4000 hour specimens exposed at 1100F are illustrated in Figure 10 (100X) and Figure 11 (500X.) The different etches used on exposed and as-received material should be noted. The as-received material was difficult to etch,

grain boundaries being outlined only after a relatively long etching time. On the other hand, the grain boundaries of all exposed specimens were easily delineated, although apparent width generally increased with increasing time.

Note the banding effect persistently observed in all specimens and the change in surface profile shown in Figures 10a and 10b. The evidence of extensive deformation of the surface region in the as-received material, illustrated in Figure 11a, is especially noteworthy. The gradual precipitation of carbides during exposure is attributed to sensitization, not to carburization. This familiar process is the precipitation of carbon, as carbides, from the originally supersaturated austenite, preferentially at grain boundaries and twin boundaries but also, in advanced stages, within the grains in a Widmanstätten pattern. Figure 11b shows the typical appearance of the 316 surface regions after 4000 hours at 1100F when polished and lightly etched. Note that the thicker appearing grain boundaries occur to a depth similar to the depth of heavy distortion in the as-received material. Figure 11c, with a slightly heavier etch, shows a less typical but not rare area in which the Widmanstätten effect is quite apparent. To substantiate the attributing of carbide formation to carbon present in the as-received material, specimens of as-received material were heated in air at 1250F for 16 hours and 48 hours respectively. The structures of these two special specimens are shown in Figures 12a and 12b. Allowing for the effect of differences of temperature, the same microstructural features formed during exposure in the model steam generator are developed. Thus, carburization is not indicated, although the very first indication of carbon pick-up might be masked by the more obvious sensitization. That sensitization occurs throughout the pipe wall demonstration that this process can not be attributed merely to carburization.

Especially noteworthy is the advanced sensitization near the surface of the special specimens as well as the loop specimens. This is attributed to the known accelerating effects of the deformation observable near the inner surface of as-received material.

The 1000 and 2000 hour specimens and all specimens exposed at 1000F look rather similar. The grain boundaries etch quite readily. All specimens appear to be undergoing sensitization, the process being more advanced in specimens exposed for longer times or at the higher temperature.

No evidence of preferential mass transfer is metallographically detectable. However, changes in the surface profile during exposure indicate that some uniform attack has probably occurred. This may be a non-recurring attack of the initial pickled surface. Once susceptible grains are removed, the attack stops.

c) Microhardness

Microhardness transverse of the 316 SS (1100F) specimens are summarized in Figure 13. No data points were more than 5 microhardness units away from the curves. The initial high surface hardness substantiates the metallographic evidence of a cold worked surface. The gradual decrease in both surface hardness and interior hardness may be called a tempering effect. Carbon, originally dissolved in the austenite, contributes to grain hardness. Removal of carbon as rather ineffectual grain boundary carbides softens the steel. Recovery in strained regions also contributes to the softening. Removal of the initially harder surface grains by mass transfer could contribute to the lower surface hardness of the exposed piping.

d) Microprobe

No mass transfer is indicated by the microprobe examination of

the 316 specimens. Figure 14 summarizes the results for the 4000 hour specimen obtained from repeated traces over the same path. The banding or segregation of nickel, chromium, and to a lesser extent molybdenum is apparent. Averages of several traces would obviously obliterate the banding details, yielding only average percentages. The maximum and minimum chromium and nickel contents were plus and minus about 1/2% of the average concentration. Iron, of course, also shows segregation to make the total metal content remain constant. As observed metallographically, the banding occurs throughout the steel. However, small regions of a mil or so are apparent which do not have this characteristic.

No probe traces indicated mass transfer effects, although naturally, because of the nature of the banding, different probe traces yielded slightly different surface compositions. Because of the gradients (from banding) extending to the surface the very initial stages of preferential mass transfer (say within 0.1 mil) might be masked.

The total metallic content of the as-received surface dropped off slightly in a region within some 0.1 mils from the surface, due to the appearance of small gradients in all constituents. A tentative explanation is that a sub scale, formed during mill treatment and remaining after pickling, is present. This effect is not detectable in the exposed specimen, substantiating the probability of a uniform removal of surface grains.

DISCUSSION

I. Croloy 2-1/4

Detection and assessment of the small observed changes in carbon content, attributable to mass transfer, is complicated by the obvious tempering effects and the initial composition gradients. After 4000 hours, the affected depth is metallographically discernable to about 10 mils. The major hardness gradients also occur within this distance. Although distinct hardness differences are present to roughly 20 mils, slight differences at greater depths may be of little significance. Although the U-bends are described as "isothermal", a small radial temperature gradient may have resulted in lesser tempering with increased distance from the sodium side, thus contributing slightly to the apparent continued increase of microhardness with depth. This partial decarburization, which represents only a small carbon loss, is perhaps better described by a measure such as the area between curves on a hardness distance plot if, as for the special specimens, the hardness at the unaffected interior is similar for all specimens, thus minimizing the tempering effects. Using this measure, only a fraction of the total carbon within the affected region has been removed, as also shown metallographically.

Because of the scatter of the microhardness data expected in this type of microstructure, designation of a depth of decarburization when no natural demarkation exists between unaffected and affected material is tenuous. The changes in visual appearance of the microstructure due to tempering induced changes in actual carbide characteristics and etching behavior must be remembered when visually assessing the area per cent carbides, which is a measure of carbon content. Similarly, the usual analysis techniques for determining the percentages of various constituents are of little assistance.

The loss of copper from Croloy 2-1/4 has not been, to our knowledge, previously reported although the small amounts of this trace element are not apt to be of engineering significance. Nickel

pick-up is also a mass transfer process. The increase in chromium and silicon contents after 4000 hours must be interpreted with caution because the contribution of natural leveling out of the original gradients is difficult to determine. In any case, the amounts of transferred material are not nearly enough to cause austenite formation and the affected depth is small. Although mass transfer of these heavier elements is not apt to change bulk properties of piping, such transfer may influence the course of the more rapid carbon transfer and thus indirectly exert an influence of engineering concern. Pick-up of the heavy elements is apparently controlled by the behavior of the source, 316 SS. The absence of detectable preferential depletion in the 316 SS pipe examined correlates with the small pick-up by Croloy 2-1/4 of nickel and chromium. However, a more extensive 316-sodium interaction, caused by a rise in temperature for instance, might very well change the behavior of Croloy 2-1/4 at 1000F. These comments are meant as a reminder that the system must be viewed as a whole, that events at one portion of the loop can control behavior at another portion.

II. 316 SS

The interesting banding and sensitization effects are only incidental to the assessment of mass transfer in the model steam generator. The differences between commercial pipe and carefully prepared laboratory samples may influence observed mass transfer effects. Susceptibility of pickled surfaces to attack by sodium, influence of a possible subscale, and the effects of a deformed surface are not commonly studied with prepared samples and are indeed generally considered of only slight importance.

The absence of preferential mass transfer is significant because material examined was at the highest temperature in the system. Possible down-stream effects and the indication of some uniform attack need to be further evaluated.

III. General

The examination of pipe samples has been made to monitor the mass transfer effects throughout the system. Results are encouraging from the view of confirming the compatibility of materials and overall operating conditions, although larger changes would be easier to describe and numerically assess with confidence. The final destructive examination should provide more definitive information, not only because of the more extensive changes probable after longer times, but also because the entire system can be examined with an aim of evaluating possible down-stream effects and perhaps even attempting a form of mass balance, although the latter would be a quite lengthy and tenuous procedure.

The results must not be viewed simply as showing the behavior of Croloy 2-1/4 in 1000F sodium or the behavior of 316 SS in 1100F sodium. The type of mass transfer at a local region depends on the nature of the sodium in contact with the steel as well as the characteristics of the steel. Although processes within the steel, such as diffusion with the associated strong dependence on temperature, may well be local rate determining steps; the direction of preferential transfer and driving forces depend on the activity of the various constituents in the sodium as well as in the steel. It is this latter factor which accounts for the necessity of viewing the system as a whole because of the interaction of processes occurring at widely separated positions.

A continuing mass transfer obviously requires a source and a sink. Preferential transfer of carbon, for instance, requires many

steps. Carbide disintegration to feed carbon to a matrix, diffusion to the sodium-steel interface, transfer across the interface, transfer through the sodium and essentially the reverse processes of carbon pick-up most of all occur. Over-all rates are commonly identified with a slowest step. However, the relative surface and volumes of source and sink also are of critical importance; a small ferritic sample in an austenitic sodium loop will behave quite differently from the same ferritic material in the form of a similar loop, containing a small section of the austenitic material. Failure to identify copper and carbon sinks in the examined sections is not of great concern. The small amounts of material removed may either be residing in the sodium or may be discovered at other locations during the more complete destructive examination.

These comments are meant as indication of the considerations to be made in interpreting the results presented and as a general rationale of the approach to the assessment of a particular system.

TABLE I

Sample Identification

Original Composition*

	<u>C</u>	<u>Cr</u>	<u>Ni</u>	<u>Mo</u>	<u>Si</u>	<u>Cu</u>	<u>P</u>	<u>S</u>	<u>Mn</u>
AISI 316 SS	0.05	17.30	13.72	2.30	0.47	0.14	.019	.007	1.67
Croloy 2-1/4	0.11	2.30	0.12	1.00	0.35	0.09	0.013	0.015	0.45

	Thermal History		Special Examination	
	Temp. (°F)	Time (hours)	Microhardness	Microprobe
Croloy 2-1/4				
1	-	0	✓	✓
2	1000	1000	✓	
3	1000	2000	✓	
4	1000	4000	✓	✓
5	-	0**	✓	
6	1000	1000**	✓	
7	1000	2000**	✓	
8	1000	4000**	✓	
316				
9	-	0	✓	✓
10	1000	1000		
11	1000	2000		
12	1000	4000		
13	1100	1000	✓	
14	1100	2000	✓	
15	1100	4000	✓	✓
16	1250	16		
17	1250	48		

*Mill analyses (except Cu and Ni is Croloy 2-1/4 which were microprobe analyses).

**Plus 1650F, 15 min, rapid cool

TABLE II

Outline of Microprobe Results for Croloy 2-1/4

	1 Micron from Original Surface	1 Micron from Exposed Surface (4000 hours, 1000F)	Interior
Wt % Cr	0.08	1.98	2.30
Mo	1.00	1.00	1.00
Si	0.10	0.24	0.35
Ni	0.22	1.00	0.12
Cu	0.39	Not Detectable	0.09

Figure 1 - Full-Size Steam Generator

Figure 2 - Prototype Steam Generator

FULL SIZE STEAM GENERATOR

PROTOTYPE STEAM GENERATOR

Figure 3 - Outside View of Full-Size and Prototype Steam Generator Showing Relative Size

Figure 4a - Schematic of Model Sodium-Heated Steam Generator

Figure 4b - Photograph of Model Sodium Heated Steam Generator

a

b

c

d

Figure 5 - Microstructure of Surface Region of Croloy 2-1/4 Pipe
Exposed to 1000F Sodium. 250X Nital Etch

a) 0 hours (as-received)
b) 1000 hours

c) 2000 hours
d) 4000 hours

a

b

Figure 6 - Microstructure of Interior of Croloy 2-1/4 Pipe to Show Tempering Effects of Exposure at 1000F Unrelated to the Sodium Environment. 500X Nital Etch

- a) 0 hours (as-received)
- b) 4000 hours

MICROHARDNESS OF CROLOY $2\frac{1}{4}$. DIAMOND PYRAMID HARDNESS (100 GRAM LOAD) NEAR THE INNER SURFACE OF PIPE EXPOSED TO 1000F SODIUM

FIGURE 8

MICROHARDNESS OF SPECIAL CROLOY 2 $\frac{1}{4}$. KNOOP HARDNESS (100 GRAM LOAD) NEAR THE INNER SURFACE OF PIPE EXPOSED TO 1000F SODIUM AND SUBSEQUENTLY AUSTENITIZED

MICROPROBE ANALYSIS OF AS-RECEIVED CROLOY 2 $\frac{1}{4}$
FIGURE 9 a

MICROPROBE ANALYSIS OF CROLOY 2 $\frac{1}{4}$ EXPOSED TO 1000F SODIUM FOR 4000 HOURS
FIGURE 9 b

a

b

Figure 10 - Microstructure of 316 SS Pipe to Show Change in Surface Profile During Exposure to 1100F Sodium and Persistent Banding. 100X

a)	0 hours (as-received)	70% HNO_3 , Electrolytic
b)	4000 hours	Marble's Reagent

a

b

c

Figure 11 - Microstructure of Surface Region of 316 Pipe. 500X

- a) 0 hours (as-received) Note Deformation Markings
70% HNO_3 , Electrolytic
- b) 4000 hours Typical Sensitization Marble's Reagent
- c) 4000 hours Advanced Surface Sensitization Marble's Reagent

a

b

Figure 12 - Microstructure of 316 Pipe After Special Sensitizing Treatment in Air at 1250F. 500X Marble's Reagent

- a) 16 hours
- b) 48 hours

MICROHARDNESS OF 316 SS. DIAMOND PYRAMID
HARDNESS (100 GRAM LOAD) NEAR THE INNER
SURFACE OF PIPE EXPOSED TO 1100F SODIUM

FIGURE 13

MICROPROBE ANALYSIS OF 316SS EXPOSED TO 1100F SODIUM FOR 4000 HOURS

FIGURE 14

REVIEW OF ATOMIC POWER DEVELOPMENT ASSOCIATES' SODIUM-WATER REACTION PROGRAM

H. O. Muenchow

I. INTRODUCTION

The development of sodium-cooled reactors is highly dependent on the development of reliable system components. Of these components, the steam generator is recognized by the AEC as the weakest link in the heat transport chain. The possible occurrence of a sodium-water reaction and the problem of designing a steam generator to accommodate such a reaction has always been a major concern of the design engineer. In the early sodium-cooled reactors, the basic problem was avoided by attempting to minimize the possibility of a sodium-water reaction through the use of double-walled tubes to separate the reactants. EBR-I, EBR-II, SRE and Hallam all embody this principle. The lack of information available on the sodium-water reaction at the time these plants were designed certainly justified this action. However, it is clear that the double-walled tube does not answer the question for plants of the future which must operate over long periods of time since it is conceivable that a failure can occur in a double-walled tube sometime during the life of the plant. The use of a double wall also leads to economic penalties that may make sodium-cooled reactors uneconomical. As the AEC has recognized, it is important, that the problem of sodium-water reactions be faced squarely, and early, in any program to develop future sodium components.

In the past the designers of sodium-heated steam generators, as typified by the Fermi unit, have concentrated on the problem of a large reaction and the effects of high temperature and pressure for a relatively short period of time on the structural integrity of the unit. This led to the incorporation of reaction pressure relief devices, such as rupture disks, into the systems. In December 1962, a Fermi steam generator underwent a sodium-water reaction due to tube failures produced by vibration. This failure served as an excellent proof test of the relief system, which functioned properly to protect the shell of the steam generator and limit the reaction effects to the steam generator proper. However, subsequent examination of the unit indicated that a general corrosion had occurred in the area of the sodium-water reaction, producing failure of some of the adjacent tubes. Also, it appeared that corrosion was not the only mechanism associated with this destructive phenomenon and, therefore, the tube damage in a sodium-heated steam generator was called "wastage". Since the failure of the tubes progressed from small to large leaks, it is not clear when the tube wastage occurred. Investigation of the data available indicates that a small leak existed for a period of approximately 40 minutes, and the large leak existed for a period of approximately 1 minute.

This occurrence has re-emphasized the interest in sodium-water reactions throughout the international sodium-cooled reactor community. Many European and American organizations now have experimental programs under way to investigate the effects of these reactions.

A review of tests performed by The Detroit Edison Company (DECO) and a preliminary summary of some work performed at Dounreay lend supporting evidence to the hypothesis made after examination of the reaction in the Fermi generator; that a small leak can produce severe tube wastage in a few minutes.

It was clear that a comprehensive program should be conducted to investigate small leaks as well as large leaks, and that the program should have a scope sufficiently broad so that new tests would not have to be conducted every time a new steam generator was designed.

II. OBJECTIVES

The use of such generalized words and phrases as small leak, relative, wastage, evidence, and hypothesis point out the fact that the complete mechanism, the controlling parameters, or the limits of the parameters involved in sodium-water reactions occurring in steam generators are not known. Therefore, in support of the AEC's steam generator development program, APDA has been awarded a contract to conduct a series of sodium-water reaction tests which will:

1. Establish a means of estimating the extent of tube wastage during a small leak of water into the sodium of a sodium-heated steam generator.
2. Determine the dominant parameters of tube wastage during a small water leak so that they may be applied to the design of a unit.

III. ESTABLISHING THE TEST PROGRAM

A review of the factors that probably affect wastage, i.e., corrosion rate, erosion rate, reaction products concentration, exposure time, reaction temperature, and tube wall temperature, indicated that the following independent variables were probably controlling.

- A. Rate of water leak
- B. Duration of water leak
- C. Distance of leak from impingement surface
- D. Velocity of bulk sodium
- E. Temperature and pressure of reaction water
- F. Initial temperature of sodium

G. Tube material

H. Flow rate of tube-side coolant

To restrict the number of tests (the operating cost per test is expected to be quite high) and still obtain sufficient data, a statistical approach to the program was taken. However, limiting a statistical program to two levels of each of the eight variables still results in 2^8 or 256 tests. Therefore, the four variables that are probably most important were selected and set up as a statistical program based on two levels of each variable that results in 16 tests. The program was then reduced to a fractional factorial of $1/2$ for the initial series of tests.

Initially, variables A through D will be studied at two levels, E and F will be held constant, while G and H will be considered in-run variables and studied at two levels during every run. The test program is tabulated below:

Run No.	VARIABLE COMBINATIONS*			
	Water Leak Rate (A)	Leak Duration, Minutes (B)	Impingement Distance, Inches (C)	Sodium Velocity, ft/sec (D)
1	4 cc/min	10	1/4	1.5
2	16 cc/sec	10	1/4	1.5
3	75 cc/sec	10	1/4	1.5
4	A	b	1/4	1.5
5	A	b	1/4	1.5
6	A	B	1/4	3.0
7	A	b	1	3.0
8	A	B	1	1.5
9	a	B	1	3.0
10	a	b	1	1.5
11	a	B	1/4	1.5
12	a	b	1/4	3.0

* Upper case letters indicate high level of variable, and lower case the low level.

As shown in the table, during the first three runs only the leak rate will be varied in order to establish the level of leak rate that will give significant wastage. After the data from these runs are reviewed, the high and low levels of the leak rate and leak duration variables will be established for a statistically based program. Runs 4 and 5 should be the same as one of the first three runs. The data from these identical runs will be used to establish the confidence level or the degree of reproducibility of the test.

IV. DISCUSSION OF TEST LOOP

Figure 1 is a photograph of the support structure, which contains the test loop and components. The instrumentation and control room is located in the concrete building on top of the cliff to the right side of the support structure.

The test stand and flow directions are shown schematically in Figure 2. The test stand supports five main systems: (1) the sodium system, (2) the circulating water system, (3) the reaction water system, (4) the reaction products system, and (5) the nitrogen cover gas system.

During a small leak test, sodium will be circulated by a centrifugal pump from the bottom of the test unit through the filter tank and back to the test unit. Large particles will be removed from the sodium by a fine mesh filter screen, designed to be removed through the top of the filter tank for cleaning. Figure 3 shows the top flange of the filter tank. The sodium system also contains a cold trap, Figure 4, and dump tank, Figure 5.

The circulating water loop is made of Schedule 160 steel pipe and contains a pressurizer, air-cooled heat dump, and three quick-acting valves for emergency isolation. The system is pressurized from a 6000-psi nitrogen station. High-purity boiler feedwater at 2650 psia and 600 F flows by natural circulation from the pressurizer down through the test coil where it is heated. The hot water passes through the air-cooled heat dump and returns to the pressurizer.

The reaction water supply is contained in a storage tank which is separate from the circulating water. The same high-purity water will be used as that used in the circulating system, and may be heated to 600 F with electrical heaters. The system is pressurized from the same 6000-psi nitrogen supply used to pressurize the circulating water system. The reaction water flows through a Schedule 160 steel pipe, which is flanged to the reaction water trigger tube. The line contains a quick-closing valve for emergency operation.

The reaction products can be vented through two paths during a reaction. During the tests the hydrogen, with entrained sodium, will be vented through the hydrogen cooling tank to the atmosphere. In the event of an uncontrolled reaction, the products will be discharged through the 24-inch rupture disk, located in a 30-inch pipe, into a reaction products separator, Figure 6.

The test unit, Figures 7 and 8, consists of a pressure vessel, 3 feet in diameter and 16 feet long, containing a removable bundle of tubes, one of which is a natural circulation water coil. The remaining tubes have nitrogen sealed inside. The bundle is approximately 2 feet high and is positioned at the bottom of the vessel under 10 feet of sodium.

The trigger tube assembly is installed and removed through the top flange of the test unit. This device is used to inject a controlled amount of water into the sodium for the reaction tests. The trigger tube

is approximately 16 feet long with the flow control orifice for the reaction water located at the lower end, and a set of valves and flange connections at the upper end that connect to the water and nitrogen supply vessels.

The water flow orifice, located at the lower end, is isolated from the sodium prior to the reaction by a rupture disk set to burst at a maximum of 2500 psia (2300- to 2500-psia range). Nitrogen is used to pressurize the 600 F reaction water to 2650 psia in the storage vessel. The valve in the trigger tube supply line is opened and the pressure, transmitted through the water column in the tube, bursts the rupture disk.

V. DATA ANALYSIS

The following data will be taken during each test run:

1. Sodium flow rate will be sensed with an EM flowmeter and will be recorded.
2. Reaction zone temperatures will be sensed by 33 thermocouples and will be recorded. Twenty-one rapid response thermocouples, capable of sensing 90% of a temperature step change in less than 500 milliseconds, will be located in the test bundle on the tube walls and in the sodium between the tubes. These temperatures will be recorded on a rapid response recording oscillograph. The remaining 12 sensors will be standard grounded 1/8-inch OD sheath thermocouples placed on the tube bundle in various sectors and elevations away from the immediate reaction zone. These will indicate differences in temperature patterns among the various reactions.
3. The gas temperature and pressure in the cover gas space above the reaction will be recorded. The hydrogen in the cover gas will be monitored by a thermal conductivity cell analyzer and the concentration of the hydrogen will be recorded.
4. Reaction water pressure, temperature, and flow rate will be recorded.
5. The temperature and flow rate of the circulating water in a coil of the tube bundle will be recorded and the pressure will be indicated.
6. After each run, the tube bundle within the reaction vessel will be removed for inspection. The bundle is designed so that each ring and coil may be disassembled from the support members for inspection. Photographs will be taken. Each tube will be dimensionally checked, and diameters will be measured with a micrometer. When significant wastage occurs, the tubes will be sectioned, wall thickness measured, and photomicrographs will be made.

In analyzing the data, the prime objective is to define which variables have the greatest effect on wastage and to attempt to establish the relative degree to which they affect wastage.

The temperature versus time traces for the 33 thermocouples on the tube bundle will be analyzed. A correlation of the effects of leak rate, leak duration, and sodium velocity on the location of the maximum reaction temperature and the general reaction zone will be attempted. Also, a definition of the general reaction zone will be made through a comparison of reaction temperatures and wastage at specific locations.

Comparisons of wastage on the nitrogen filled tubes with that of the water coil will be made to establish if tube cooling from the inside has much effect on the degree of wastage. Also, wastage comparisons will be made among the 2-1/4 Cr - 1 Mo, 5 Cr - 1 Mo, type 316 SS, and type 304 SS tubes in each test.

Although the data will be limited, an attempt will be made to correlate the data from the hydrogen detector with the leak rates in these tests. However, hydrogen detection in the cover gas may prove to be too slow a method of leak detection to be useful with high wastage rates. Response rates of other leak detection methods may require investigation. In addition, the hydrogen detection data will be compared with the data obtained by DECO in the Fermi steam generators to point out the effects of the different system geometries and dynamics.

During the analysis of the test results from the initial series of tests, it is anticipated that new factors concerning wastage will be uncovered. In order to investigate these new factors, study the interaction of the dominant parameters, and expand the test data concerning the dominant parameters additional tests will be required. With the additional data, correlations will be attempted to establish the degree of wastage which will occur for a particular leak rate with respect to time. Should this correlation prove feasible, it might be combined with a leak detection versus leak rate plot to produce a correlation of leak detection, wastage, and time for a specific system. Then, at any leak detection level, a known time is established in which to effect a safe system shutdown. At a high leak detection level the shutdown might require scram action; however, at the lower levels normal shutdown procedures could be followed with varying degrees of urgency.

VI. CURRENT STATUS

All of the test loop components have been erected. The loop piping and valves have been installed and the systems are presently being leak tested.

Installation of the electrical components and instrumentation equipment started May 14. Vessel and piping heaters and the thermocouples that monitor the temperature of the vessels and piping will be installed first. The loop will be insulated while the electrical work such as erecting cable trays and pulling cable is completed.

According to the present schedule, loop check-out and shakedown should start the last week in July. The way the work is presently progressing the schedule should be met.

Fig. 1 Sodium-Water Reaction Test

Fig. 2 Loop Support Structure

Fig. 3 Top of Filter Vessel

Fig. 4 Cold Trap Vessel, Without Cooling Duct

Fig. 5 Top of Storage Tank

Fig. 6 Reaction Products Separator

Fig. 7 Top of Reactor Vessel, Showing 24-Inch Pipe Leading to the Separator

Fig. 8 Bottom of Reactor Vessel

ESADA
SODIUM-WATER REACTION
TESTS

By

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The Empire State Atomic Development Associates (ESADA), in continuing support of reactor power plant related research and development, have contracted with Atomics International to conduct a sodium-water test program. The primary purpose of this work is to study the effects of a sodium-water reaction resulting from a ruptured tube in a modular steam generator. Although a number of sodium-water experiments have been performed by others, the range of pressures and temperatures involved with the modular design have not been attempted in prior work. The study is expected to yield information that will permit the design of a modular steam generator system in which damage to the system is minimized in case of a sodium-water reaction.

The most important information needed from the test program is the shell, or sodium side, pressure-time relationship for a given mass flow rate of water and steam at operating conditions when reaction takes place. This information, correlated with performance characteristics of relief devices and the relief system, will provide the basis for the design of an integrated system in which pressure effects are minimized. In addition, data assuring adequate structural design of the steam generator module will be obtained.

A program of testing has been devised in which a simplified test system is used to obtain relative pressure-time data. A tube is forced to rupture within the system and resulting pressure transient measurements are made. Extrapolation to a full-sized system will be accomplished by mathematical model. It is expected that these experiments using the simplified test system will provide the basis for additional tests using the steam generator module.

Construction of the test facility has been completed and testing is in progress. The test system, Figure I (7633-1512A) consists of the test section in which reaction takes place, the relief system and

receiver tank for the products of the reaction, a high pressure steam supply system, a high pressure water supply, and a sodium storage and supply system.

The electrically-heated steam supply system is designed to provide steam at 2400 psi at 1000°F. Flow is limited to approximately 4 lb/sec. The water supply, also electrically heated, is designed to provide water at average conditions of 2800 psi and 470°F. The rate of flow is approximately 6 - 8 lbs/sec. Capacity for 10 - 15 seconds flow is provided by each system.

The relief system provides three locations for the installation of 6-inch diameter burst diaphragms. Normally, two will be used, one located at each end of the test section. They are rated at 125 psi. Upon rupture of the diaphragms, test section pressure and reaction products are discharged into the reaction products tank through 6-inch pipes. The tank is rated at 150 psi at 850°F. A 16-inch relief valve on top of the tank will pass up to 100 cubic feet of vapor per second at 5 psi. During each test, low pressure steam flows into the tank to complete reaction of sodium received from the relief system. Following the test, the relief system, test section, and reaction products tank are flushed with water and drained.

The sodium supply tank is used for intermediate storage of sodium at approximately 300°F. It has sufficient storage capacity for 4 barrels. The test section is filled through the transfer line by means of pressurizing this tank with inert gas. At completion of the filling operation, two valves in the transfer line are closed and the line is allowed to cool. The sodium remaining in the line solidifies, and thus aids in isolating the sodium supply tank from the test section during tests.

A photograph of the test facility, taken at completion of construction, is shown in Figure 2 (7633-10202). The longer pipe column in the center of the photo is the main test section. All auxiliary test sections have been assembled, as shown, during construction to ensure proper assembly and fit-up. Those sections, not required for the first tests, were subsequently removed.

A view of the reaction products relief tank with the relief valve in place on top of the tank is shown in Figure 3 (7633-10200). The valve cover is arranged to pivot upward to open, and the adjustable weights on the valve cover are such that 1-1/2 psi can be maintained in the tank. Unreacted sodium discharged from the test section and relief system is exposed to low pressure steam in the reaction products tank. This steam is supplied at 90 psi by a 30-h.p., gas-fired utility boiler. All sodium is thus reacted prior to flushing the system with water and draining.

The main test assembly in which initial reaction occurs is shown diagrammatically in Figure 4 (7633-15092). It consists of a tube bundle that surrounds a rupturable tube, all of which is enclosed in a

large heavy-walled pipe. The tube bundle is made of 36 stainless-steel tubes approximately 4-ft long that are sealed at both ends. Spacers of the same design and material as those used for the modular steam generator, space the tubes radially on approximately 1-in. pitch. This establishes three rows of tubes radially around the center. The center is occupied by the rupturable tube.

The shell is made of two sections of 8-in. sch. 80 stainless steel pipe. When assembled, the two sections have a total length of 16 ft. The lower section has four lateral nozzles, two of which are shown closed with blind flanges. These two are provided for connection to auxiliary test sections when required. The remaining two nozzles are connections to the relief system. The two flanged joints contain the 125 psi burst diaphragm assemblies.

The upper section of the test assembly is an 8-in., sch. 80 stainless steel tee. The rupturable tube is secured in the center of the side outlet of the tee by means of a blind flange with a hole machined to accept the tube. The upper and lower ends of the test assembly, shown closed with blind flanges, are for connection to additional test sections that will increase length to 44 ft.

The rupturable tube enters through the side outlet of the tee, passes centrally down through the tube bundle and emerges through the lower blind flange. It is made of type 304 stainless steel tube of 5/8-in. outside diameter and .090-in. wall thickness, the same material as that used for tubes in the modular steam generator. A flat, 4 in. long, is machined on the tube to reduce wall thickness to 0.005 - 0.007 in. at a point tangent to the inside diameter of the tube. The flat is positioned vertically midway between the two burst diaphragms as shown in Figure 4. The tube is ruptured when steam or water pressure inside the tube reaches approximately 1100 psi. Shown on the right in Figure 4 is a cross section of the machined tube.

Steam is admitted at the upper end of the rupturable tube; water at the lower end. Pressure is withheld from the tube by means of a blocking disc inserted at either end of the tube. When flow is required, the disc is opened with a spring-driven pointed valve stem. Prior to use, the spring is compressed and held by a latching mechanism which, in turn, is secured by an explosive bolt. When the bolt is electrically fired, the spring is released, driving the pointed valve stem into the blocking disc. In this way, the rupturable tube is abruptly pressurized. A tube rupture caused by steam at 1575 psi is shown in Figure 5. No sodium was present in the test section.

Pressure transients occurring in the test section are detected by means of differential transformer type transducers using a 10 kc primary supply with output demodulation and amplification sufficient to drive a 1000 cps recording galvanometer. The linear differential transformers are suitable, with internal temperature compensation, for operation at temperatures up to 1000°F. Natural frequency of the Inconel X, heat-treated diaphragm is in the 600 - 1000 cps range, varying with individual

units. The transducers are rated at 500 psi, and 1000 psi for short term overload. The pressure system is nominally response limited by the transducers to a minimum of 600 cps. Thus, pressure instrumentation delay for step-function input is about 0.6 ms up to 90% of final value. For an exponentially increasing function on a period of one ms, the delay is near 0.3 ms.

Pressure transducers are installed on the test section shell by means of flanged nozzles such that the transducer diaphragm is in direct contact with the sodium. One transducer is located on the shell at the point where tube rupture and initial reaction takes place. Two, the first 5 ft above the point of rupture, and the other, 5 ft below are provided as well as one on each of the two lateral nozzles leading to the relief system, for a total of five pressure transducers on the main test section. Up to 11 transducers may be installed on the 44-ft test assembly. By means of observing pressure occurrence at each location, pressure-time data can be obtained.

Strain gages also are installed at each pressure transducer location to obtain strain information occurring in the shell. This is primarily for determining feasibility of using strain gages for obtaining structural data from later tests in which the steam generator module is used. The strain gage monitoring system consists of a standard 20 kc bridge excitation with compatible demodulator-amplifier assembly. Recording of strain information is by means of an oscillograph with 5000 cps galvanometers.

Thermocouples are affixed mechanically to the rupture tube, and are thus immersed in the sodium at the point of tube rupture, and at points 5 ft above and below conjunctional to pressure transducer locations. The thermocouples are 1/8-in. sheathed chromel-alumel type with the junction grounded. The sheathed junctions have time constants of approximately 100 ms. Thermocouple recording is direct, with no amplification. Galvanometers have 60 cps natural frequency.

The different test assembly configurations possible to obtain with presently available components are shown in Figure 6 (7633-1511A). The asterisk denotes the location of tube rupture in each assembly. The sequence shown is tentative, it is expected that development of the mathematical model will dictate the order in which the different assemblies and conditions appear. The five different configurations shown are achieved with the use of interchangeable pipe spool sections. Flexibility is required to study the effects of distance from the point of tube rupture to the relief system on pressure-time data. Also, the effects of different column heights of sodium above and below the point of tube rupture can be observed.

Test conditions are tabulated below each configuration. Each series consists of three identical tests to check data repeatability. The first assembly, for test series A and B, is the simplest, requires 300 pounds of sodium, and is for the purpose of checking system performance, test procedures, and instrumentation. Series C represents a tube rupture

occurring near the lower end of the superheater module where sodium at 915°F will react with water at 470°F. Series D conditions are for the case where tube rupture occurs near the lower end of the evaporator module. Sodium temperature is lowered to 675°F to react with water at 470°F. Similarly, Series E through H represent tube failures at the center and at the top of the two modules, with test conditions representative of operating conditions that would be expected at these locations. Test Series C through H require approximately 600 pounds of sodium for each test.

Series J tests are for the purpose of observing pressure effects on a parallel system. The test assembly is identical to that used for Series C and D except for a parallel test section added to the main assembly. Approximately 700 lb of sodium will be required for each of these tests.

In all tests, the test assembly will be incompletely filled with sodium so that a space equal to 8 - 10% of the sodium volume will remain. This space will be pressurized with inert gas at 50 psi for the evaporator tests, and 75 psi for the superheater tests. These cover gas conditions are approximately those expected in the steam generator modules.

Test No. 1 of Series A was performed on April 29, 1965. Three-hundred lb of sodium at 750°F were used. The central tube was ruptured by steam at 2060 psi and 970°F. The steam control valve malfunctioned, interrupting the required sequence of events and delayed tube rupture and reaction approximately 30 min. Consequently, pressure transient measurements were not obtained, nor were photographic records secured. However, post test examination of the system verified structural adequacy. The relief system performed as desired, and the post test water flush was adequate to clean the system. Figure 7 is a photograph of the tube rupture shown for comparison with the tube that was ruptured previously in the absence of sodium.

Figure 8 is a photograph of the tube bundle after test. All tubes were deformed by bending in relation to the longitudinal axis. None were fractured or deformed circumferentially. A bend in one of the central tubes was about 1-in. from the normal axis. All deformation occurred adjacent to the central tube rupture location.

Although little quantitative data were obtained from the first ESADA test, there appears little doubt that the test plan will deliver optimum information acquisition commensurate with cost and schedule. If development of the mathematical model for steam generator design based on data from early testing shows promise of success, consideration will be given to revising the test plan for inclusion of a typical steam generator module for tests in the present facility. Additionally, the feasibility of revising certain parameters such as flowing sodium rather than static, closer simulation of pressure drop in parallel system sections, and revision of the relief system for collection and storage of reaction products will be studied.

TEST SYSTEM DIAGRAM

Figure 1

Figure 2

Figure 3

TEST SECTION

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

SODIUM TECHNOLOGY LOOP
EVALUATION OF IN-LINE INSTRUMENTS
AND PURIFICATION DEVICES

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I. INTRODUCTION

Under the sponsorship of the AEC's Reactor Division, the sodium technology loop has been operated by APDA since 1962 to evaluate in-line instruments and purification devices for sodium impurities. The program is designed to include the following activities:

1. Instrumentation for sodium systems, including the development and evaluation of in-line instruments for the detection and determination of impurities in sodium.
2. Sodium cleanup processes, including the development and evaluation of sodium purification techniques and devices.
3. Chemical and physical behavior of sodium impurities, including the investigation of the chemical and physical behavior of impurities in sodium loop environment.
4. Laboratory analysis, including complementary development and refinement of analytical techniques for the determination of sodium contaminants.

During the Sodium Components Meeting held at Hollywood during April 1964, the available results of evaluations of a Rhometer, hydrogen meter, and plugging meter were presented. The recent additions to the loop and supporting facilities will be described, and the status of the instruments and devices being evaluated in the loop and the supporting chemistry program will be discussed.

Since April 1964, the sodium technology loop has been modified to include two additional Rhometers, four oxygen meter cells, a filter cascade, a hydroclone, and improved sodium and cover gas sampling facilities. The addition of these tools has not only increased the scope of the investigations which can be undertaken, but also has allowed a comparison of the relative behavior of these devices under controlled conditions.

In order to undertake a meaningful program of development, a chemical laboratory program is required to provide an immediate and accurate evaluation of the performance of the instrument or clean-up device during its development. Chemical techniques which are used are described.

II. SODIUM TECHNOLOGY LOOP MODIFICATIONS

The loop, as now modified, is shown in Figure 1. The new circuits extending from the original 4-inch loop are all type 304 stainless steel, and are designed to operate at temperatures up to 1000 F at an average flow rate of 7.5 gallons per minute.

The two new Rhometers are installed in a differential arrangement, so that it is possible to monitor the inlet and outlet streams from any of the clean-up devices installed in the circuit. An economizer is provided upstream of both meters to equalize the temperature at the two monitoring points. Oxygen cells 1 and 2 are installed near RM-1 and cells 3 and 4 near RM-2. Thus it is possible to directly compare their readings with the Rhometer indications.

The effluent header returns either to the pump surge tank directly, or to the plugging meter and original Rhometer installed in the 4-inch loop. The supply header and effluent header may be operated in series (with no clean-up devices across the line) passing through the differential Rhometers and through the two sample stations.

The sample stations are U-bends of stainless steel tubing which can be isolated from the circuit when a sample is desired. Eventually they will be placed in a vacuum dry box to eliminate possible atmospheric contamination when sample tubes are removed or replaced.

The extended loop and components are serviced by a 200-gallon auxiliary storage tank (not shown) into which the system drains by gravity. The loop is vented at all high points with lines connecting to the original loop vent system, and the new tanks are connected to the original loop pressure equalizing systems. The thermal decomposition tank is equipped with manual heaters so that its temperature can be raised to 1200 F.

The filter cascade is an arrangement of three progressively finer micro-metallic filters in series with provision to drain and isolate the assembly for removal from the loop by means of grayloc fittings. When this assembly is transferred to a dry box for disassembly and examination, each filter chamber is made accessible for examination by breaking open the 3-flange separators in the cascade assembly. Filter elements may also be removed and disassembled.

The Doxie hydroclone installation consists of two Dorr-Oliver units mounted in parallel. This arrangement is designed to pass a 7.5 gal/min flow of sodium through the differential Rhometers with an anticipated 30 to 40 pound pressure, with a calculated ability to effect a 95% separation of 2-micron particles or larger. To provide operating flexibility, the hydroclone units are provided with plugging rods to deactivate any number of the individual cyclone separators. These units were adopted for sodium service by sealing the removable high-pressure heads with a stainless steel Flexitalic ring gasket. The overflow, or clean discharge stream, empties into the effluent header; the underflow stream, which contains the removed contaminants, passes through a throttle control and a flow-meter to a run-off pot for accumulation and sampling. Figure 2 is a photograph of the loop as now modified.

III. CHEMICAL PROCEDURES

The Chemical Laboratory is equipped to analyze sodium for total carbon,

Figure 1 Diagram of Sodium Technology Test Loop

Figure 2 Sodium Technology Test Loop

carbonate carbon, oxygen by mercury amalgamation, total hydrogen, hydroxide hydrogen and nonhydroxide hydrogen. The analytical determinations for a given sample depends upon the investigation being undertaken. The chemical basis of the procedures are as follows.

A. Determination of Total Carbon

The analytical method used by APDA for the determination of total carbon¹ in sodium metal is based on a stepwise conversion of the total carbon in the sodium sample to sodium carbonate followed by thermal decomposition of the sodium carbonate to carbon dioxide. The carbon dioxide is collected at liquid nitrogen temperature and subsequently measured by gas chromatography.

A representative series of replicate sodium samples analyzed by this method had a standard deviation of ± 3 ppm and a relative standard deviation of 8.1%. This method is applicable to the determination of carbon in sodium in the concentration range of 10 to 150 ppm.

The conversion of carbon (C) in sodium (Na) to sodium carbonate (Na_2CO_3) and subsequent evolution and collection of carbon dioxide (CO_2) is carried out in the apparatus shown in Figure 3. The analytical system consists of five sections: (1) gas metering and mixing, (2) gas purification, (3) sodium sample combustion, (4) CO_2 collection, and (5) measurement.

B. Determination of Carbonate Carbon

The analytical method developed for the determination of carbonate in sodium² is based on the carefully controlled dissolution of the sodium sample in water, the evolution and collection of carbon dioxide, and the measurement of the carbon dioxide by gas chromatography. An investigation of the relative merits of water-dissolution and alcohol-dissolution technique resulted in the selection of water as the solvent. Standard deviations of 0.2 to 0.26 ppm and relative standard deviations of 4% to 7% were obtained on standard sample using the water-dissolution technique.

A gas chromatograph and a sample dissolution carbon dioxide evolution and collection assembly are the two essential components of the apparatus used for the determination of carbonate carbon in sodium metal. Figure 4 is a schematic of the apparatus.

A three-necked distillation flask, is used for the dissolution and decomposition of the sample. The flask is fitted with connectors that facilitate the removal and assembly of the vessel in the apparatus. When assembled in the apparatus, the three standard taper, ground glass joints of the flask are used as the inlet for the helium purge gas, the reagent addition tube and the outlet for the evolved CO_2 . A thermocouple is located near the bottom-center of the flask and a Sim-Ply-Trol controller is used to regulate the reaction temperature. A magnetic stirrer is used to agitate the reaction. A water-cooled condenser and a dry ice-acetone trap are inserted between the reaction flask and the carbon dioxide collection loop to remove water vapor and acid fumes which may be carried over by the helium to the collection loop.

C. Determination of Oxygen

The Pepkowitz and Judd method for the determination of oxygen in sodium has been modified so that relatively low concentrations of oxygen³ in sodium can

Figure 3 Combustion Train for Determining Carbon in Sodium Metal

Figure 4 Gas Evolution Apparatus

be determined with improved reliability. The APDA procedure is applicable to the determination of 10 to 100 ppm of oxygen with a relative standard deviation of approximately 10 per cent. The separation of sodium carbonate and sodium hydroxide with the sodium monoxide in the amalgamation process has been demonstrated to occur.

All sampling procedures and the extraction of the sodium metal from the insoluble residues are done in a dry box containing an inert argon atmosphere. The argon is recirculated at the rate of 1500 cu ft/hr through a gas purifier which contains a bed of hot copper to remove oxygen and molecular sieves to remove moisture. The moisture level in the argon is maintained well below 1 ppm and is continuously monitored.

In the APDA method, a clean, dry tubing cutter is used to remove a 1/2 in. to 2 in. section from the 1/2 in. stainless tube containing the sodium sample. This sample size should be in the 1 to 5 gram range. It is weighed to the nearest milligram in the glove box. The sample is then transferred in the glove box to the amalgamation flask which contains 50 ml of triple-distilled mercury. The flask is then agitated to facilitate complete dissolution of the sample.

After the amalgam has cooled, it is agitated for 5 minutes and allowed to stand for 3 minutes. The amalgam is drained into a stoppered bottle retaining a residue of 1 to 2 ml which contains the insoluble residue. Twenty-five milliliters of new mercury is added to the 1 to 2 ml of amalgam, and the extractions repeated seven times. After the seventh extraction, a 1 to 3 ml sample of the effluent from the flask is collected in a 50 ml beaker and removed from the glove box. Five milliliters of deionized water and 1 to 2 drops of phenolphthalein solution are added and if a negative alkalinity test is obtained, as indicated by a colorless solution, the extraction of the sodium is complete. Additional extractions and tests are necessary if a pink color is obtained in the alkalinity test.

Fifty milliliters of deionized water are added to the extraction flask, and the resulting solution is collected in a 50 ml beaker. The flask is rinsed with two separate 25 ml portions of deionized water. Modified methyl red indicator and exactly 1.000 ml of a standard 0.002 N sodium hydroxide solution are added to the solution. It is titrated with 0.002 N sulfuric acid solution until the end point is detected.

The oxygen concentration is calculated using an expression which assumes the extracted oxide is in the form of sodium monoxide.

D. Determination of Hydrogen

The determination of hydrogen⁴ in sodium is based on the principle that boiling mercury will displace the contained hydrogen. Thus, the sodium is heated with mercury to reflux temperature, and the hydrogen evolved is analyzed by gas chromatography. In determining hydrogen, the most difficult problem is to be sure that the reflux flask, Figure 5, is not a source of hydrogen. This is assured in APDA's procedure by pretreating, or conditioning, the reaction vessels which involves refluxing mercury and sodium in the vessel at the highest temperature attained during the analysis to remove any residual water. After refluxing, the atmosphere in the vessel is analyzed by gas chromatography. If no hydrogen is present, the vessel can be considered ready for use; however, if traces of hydrogen are present, another cycle is needed.

After the hydrogen vessel is prepared it may be used for several analyses until the amalgam becomes solid at room temperature.

Figure 5 Hydrogen by Vacuum Reflux

The actual analytical procedure is similar to that used to clean the vessel. In determining nonhydroxide hydrogen the sodium sample is added to the mercury and the cover gas pressure adjusted so that refluxing occurs at 400 F. Three refluxing cycles are generally required to remove the nonhydroxide hydrogen.

To obtain hydrogen from NaOH the amalgam is heated to 680 F. It generally requires five cycles of refluxing to remove all hydroxide-hydrogen.

After refluxing, the vessel is returned to the dry box and the sample tube removed and weighed. The hydrogen concentration can now be calculated.

IV. LOOP INVESTIGATION

The program planned for the loop will eventually include an investigation of the effects of various individual impurities of concern in fast reactor operation on instruments and purification devices. Since the April 1964 report, a study of sodium carbonate and water reaction products have been completed.

The general procedure for adding impurities to the loop is to add an amount of impurity calculated to be equivalent to one-half the accepted solubility level at the temperature of the test, covering a temperature range of 450 to 900 F. After the addition of the impurity at constant temperature a temperature ramp is carried out to investigate the effect of temperature on the response of the instruments being evaluated. At the end of each series of additions a maximum amount of impurity is present in the loop and at the time a purification device is cut into the loop and its performance is evaluated.

A. Sodium Carbonate Additions

For this study⁵ a previously described 7-inch teabag was loaded with 11.17 grams of granular Na_2CO_3 (20-45 mesh), which was prepared by fusing primary standard Na_2CO_3 in a platinum crucible followed by crushing and screening. During this test, the teabag was immersed approximately 4 inches in the flowing sodium stream for 4 hours at a temperature of 450 F. Inspection of the teabag in the glove-box revealed that sodium had contacted the outside of the teabag but did not fully penetrate into the teabag through the 0.011-inch slots. A total of 11.05 grams of Na_2CO_3 was recovered from the teabag, indicating that no injection was obtained.

Because the Na_2CO_3 did not go into solution, the effectiveness of a teabag as a means of adding Na_2CO_3 was evaluated at 450 F loop temperature using 13-to 30-mil Na_2CO_3 particle size and 200-gal/min flow. Additional tests were run at 850 F, with the other conditions remaining the same. In this test the sodium did penetrate into the teabag, but an insignificant amount of Na_2CO_3 was dissolved out of the teabag. Results of the chemical analysis showed that 9.18 grams of a total 9.97 grams remained in the teabag.

It was concluded that this method of adding impurities is unsatisfactory under the conditions tested and a new procedure should be developed.

Several alternate methods for adding Na_2CO_3 to the loop were then considered:

- Building a side contamination loop

- A hot finger method

- The direct addition of Na_2CO_3 to the sodium in the surge tank

- Forced flow through Na_2CO_3 in a retainer basket

A side contamination loop is now being designed and will be used for future work; however, it could not be fabricated in time for this test.

The hot finger method consisted of a 1/2-inch pipe, which contained the Na_2CO_3 . The hot finger was coupled into the bottom of the flowing sodium line and was heated. The dissolved sodium carbonate was supposed to diffuse through the sodium in the finger into the flowing sodium stream. The results of the test indicated that very little Na_2CO_3 , if any, had dissolved.

To determine whether Na_2CO_3 will dissolve in the presence of large amounts of sodium, the direct addition of Na_2CO_3 into the sodium was tried. In this test the sodium analyses showed that there was no increase in the carbonate carbon content. However, this method is not suitable for standard operation because there is no direct method of accounting for the residual Na_2CO_3 .

The last method evaluated involved the forced flow of sodium through a specially designed container loaded with 10.82 grams of granular Na_2CO_3 . The flow rate through the unit was 7.5 gal/min for 28 hours. The temperature of the sodium was maintained at about 850 F. A chemical analysis of the material remaining after the container was removed from the loop showed that none of the sodium carbonate had dissolved.

It was then concluded that the sodium carbonate solubility data reported in the literature is greatly in error. To verify this, a solubility apparatus has been built which can be operated entirely in the laboratory dry box. This apparatus is shown in Figure 6. The equipment is constructed so that sodium can be distilled into the solubility vessel containing the sodium carbonate. The contents of the solubility vessel can be stirred until equilibrium is reached. When removing a sample, the solubility container is pressurized with argon purified with hot copper, molecular sieves, and a hot sodium trap. The effluent line is heated to the same temperature as the main vessel to avoid precipitation. The entire sample is added directly to the analytical vessel to avoid a segregation effect upon cooling.

B. Water Injection Program

The recent program of injecting water into the loop sodium has provided extremely valuable information concerning the effect of water reaction products on the response of both the Rhometers and oxygen meters, and the relation of this response to the chemical interactions taking place subsequent to the injection.

The program involved adding water in 5-ml increment to 320,000 grams of sodium contained in the loop, or the equivalent of about 14 ppm of oxygen and about one ppm of hydrogen for each increment of water. A total of 11 such injections were made over a period of two weeks making a total added contamination equivalent to 170 ppm of impurities. The starting oxygen concentration, by chemical analysis, was about 70 ppm making a final expected oxygen contamination of about 225 ppm. Chemical analysis showed that this amount of contamination was not present in the sodium, and the instrumentation did not indicate that the oxygen was dissolving in the sodium. Since the water was injected below the level of the sodium, there can be little doubt that all the water added reacted with the sodium. However, because the hydrogen in the cover gas was rapidly absorbed by the sodium, the concentration of hydrogen in the cover gas cannot be used as an independent check on the amount of water actually entering the sodium.

The conclusion was that some of the initial water reaction products are solids, and that most of these solids do not dissolve in the sodium under the

Figure 6 Sodium Carbonate Solubility Apparatus

conditions investigated. These solids probably precipitate in the loop and remain there until a much higher temperature is reached.

1. Rhometer Response to Water Additions

The response of the loop Rhometers to water injection⁶ is extremely interesting and the curves shown in the APDA Quarterly Report for March-June 1965 allow an interpretation of the effects of water injection, and hydrogen evolution and re-solution, which occur during actual water injection as well as during the subsequent temperature ramps. It is quite clear that the Rhometer responds immediately to an injection of water showing an increase of sodium resistivity with each injection. Further, a continued increase in resistivity seems to correspond to the subsequent absorption of hydrogen by the sodium. At 450 F, this latter effect on the Rhometer is greater than the effect of the water injection. However, when the injections are made at 675 F the effects are much smaller. When desorption of hydrogen occurs after a temperature ramp, the effect on the Rhometer is also apparent.

Although the response of the Rhometer to water injection is quite encouraging, it is important to note that as the programs just described were getting underway, some fundamental defects were discovered in the Rhometers which required attention before a meaningful evaluation could be started.

Shifts occurred in the zero reading of the Rhometer which were caused by movements of the loop during valve operation or other adjustments. It was found that a small movement of the winding assembly on the toroid transformer can produce a large upset in the meter reading, particularly if the coil is skewed relative to the core axis. As manufactured, the winding form is fitted loosely on the core leg, probably to allow air circulation, and about one-fourth inch of movement possible. It was demonstrated that movements which occurred during valve adjustment caused resistance shifts of 0.1 to 0.12%. This caused an error in the range of 100 ppm oxygen. In order to "fix" the winding in place, narrow wedges of Maranite were placed at the four corners inside the form window. This reduced the movement to a negligible volume; however, this approach is not a permanent solution to the problem.

The large effect of temperature on the reading of the Rhometer is illustrated by the equation which Blake⁷ derived to express the effect of temperature and impurities on the resistivity of sodium:

$$\rho = 5.17 [1 + 0.00263 (T-200) + 0.0001W]$$

ρ = resistivity in micro-ohms per inch

T = temperature of sodium in °C

W = concentration of oxygen in parts per million

It will be quite obvious that temperature compensation is extremely important since a change in temperature of one degree centigrade produces an effect on resistivity equivalent to 25 ppm of oxygen.

2. Rhometer Temperature Compensation and "Noise"

The three Rhometer units have been responsive, with apparently good correlation, to all eleven water injections made in the recent tests; however, plots of the Rhometer readings of all three instruments show a significant noisiness in the form of random excursions. The noise plus base-line shifts have complicated the task of extracting information from the plots. Except for differences in base-line readings and base-line shifts which occur when switching ranges, the three instruments are reasonably alike in performance.

A base-line shift could certainly be caused by a change of resistance of the compensator. The degree of base-line shift exhibited by RM-3 and RM-2 is insignificant insofar as affecting temperature compensation. In the case of the RM-1 Rhometer, however, it is felt that some degradation of temperature compensation has occurred as the result of the approximately 0.8% base-line shift of several months ago.

The noise present in the Rhometer reading plots consists of occasional spikes to a level as high as perhaps 0.2%, positive or negative, with relatively frequent smaller excursions below about 0.05%. The spikes and excursions have been studied for simultaneity of occurrence with possible errors in dial setting and abnormal flow; the latter would most likely occur during sample taking. Sensitivity of all three Rhometers was checked, with special attention given to the RM-2, by offsetting the decade dials and noting the correspondence (or lack of it) with the recorder reading. All three instruments were found satisfactory.

Base-line shifts when ranges are switched, base-line shifts were definitely found for all three Rhometers, the greatest being that of the RM-1. Its base-line shift is about +0.15% when going from range 1 to range 2. RM-2 and RM-3 Rhometers exhibited base-line shifts of about -0.05% and -0.06%, respectively, when switching from range 1 to range 2. Direction or polarity of the shift is reversed in going from range 2 back to range 1.

A possible explanation of the noise in the Rhometer readings is the effect of small, variable deposits of impurities, perhaps NaOH, lodged on rough areas inside the toroid pipe. Welds in the "V" areas where the toroid pipes separate appear to have been ground down but left rather rough. To account for an excursion of 0.2%, the amount of sodium displaced would have to be about 0.53 ml. To provide a basis of reasoning in this area, if 10 ml of water produce 8 ml of NaOH (20 grams with a density of 2.5), a reading of +3.3% would be obtained if all of this material were deposited in the toroid.

V. EVALUATION OF THE UNITED NUCLEAR OXYGEN METER

The oxygen meter⁸ has been previously described. Four electrolytic cells have been in service since early May at all times when the temperature of the loop sodium is below 650 F, a temperature limitation imposed by the objectives of the present evaluation program.

The four loop cells have yielded extremely interesting information during the water injection series.⁶ It will be noted that none of the cells showed any well-defined response during the injection of water and the chemical analysis showed that the oxygen content of the sodium was not changing which is in agreement with the lack of response. The voltage of all cells increased in sodium temperature indicating that a different temperature correction

factor might be needed. However, all the increase in voltage noted may not be caused by temperature because the chemical analysis shows that the oxygen concentration is decreasing. This effect may be related to hydrogen absorption and subsequent chemical interaction⁶

The most interesting data obtained to date is the decrease in voltage which occurs at about 655 F at exactly the same time the hydrogen in the cover gas shows a rapid increase.⁶ The expulsion of hydrogen may relate to the conversion of sodium hydroxide to sodium monoxide which goes into solution and thus causes the oxygen meter to indicate an increase in oxygen concentration. This effect has been observed at the same temperature in another run.

Additional studies are planned to investigate this effect since it may be important to the design of purification devices.

VI. IN-LINE HYDROGEN METER

The APDA in-line hydrogen meter is based on the principle that oxygen can be used to "pump" hydrogen through a metal wall by using it to catalytically oxidize the hydrogen to water as it emerges from the wall. Thus sodium containing hydrogen can be passed through a tube, for example, and the hydrogen diffusing through the tube wall is oxidized to water and the water concentration can be measured. The rate of hydrogen diffusing through the tube wall is related to the hydrogen concentration in the sodium.

The meter (Figure 7) has as its principle component a length of palladium tubing approximately 9 feet long wound as a 4-inch-diameter helix. The tubing has a 1/4-inch O.D. and a 10-mil wall thickness. The surfaces which will contact the sodium will be plated with electroless nickel (2 to 3 mils). The unit should be operable up to a temperature of 900 F; however, it must be maintained at isothermal conditions regardless of the loop temperature. Flow through the unit would be about 0.1 gal/min which would give a residence term of approximately 1.5 minutes in the detector. The oxygen throughput would be 200 cc/min which is the flow rate required by the present moisture monitor. The unit is calculated to have a diffusion rate high enough to detect 0.1-ppm hydrogen in sodium; however, its probable accuracy is not known.

Figure 7 In-Line Hydrogen Meter

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THE LIQUID METAL OXYGEN METER

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An on-line instrument for continuously monitoring the effective oxygen content in sodium has been developed at United Nuclear Corporation. The meter is rugged and suitable for use in sodium systems under field conditions. It is specific for oxygen, and useful at low oxygen levels in the range from 1 ppm to 100 ppm. It is extremely sensitive to small changes in oxygen concentration, particularly at low levels. Its response to changes in oxygen content is rapid, it is relatively insensitive to changes in sodium temperature, and it is independent of sodium velocity over reasonably large ranges.

Development of the meter was started approximately 3 years ago as part of the Atomic Energy Commissions's Sodium Component Development Program. The initial phase of the program demonstrated the feasibility of the electrochemical approach on which the meter operation is based. A subsequent research and laboratory test phase determined the most satisfactory cell composition and its characteristics. These tests were performed in thermal convection loops and utilized experimental models. Details of the operating principles and of the feasibility and laboratory tests are available in References 1a through 1e.

The objectives of the work during the past year were directed toward the development, production, and testing of meters of improved design which would be practical for use as on-line oxygen monitors under field conditions. These objectives have been met. The Liquid-Metal Oxygen Meter is now undergoing field tests at APDA. It is scheduled to be in operation at LASL and ANL shortly.

The instrument developed in this program consists of two components, a Sensor and an Indicator. These units are shown in Fig. 1. The Sensor is installed in a main or bypass line of the sodium system. The Indicator may be located either adjacent to the Sensor or in the system control center.

The Sensor consists of an electrode assembly and a housing as shown in Fig. 2. The housing provides an all-stainless steel containment envelope for the electrode assembly. Its principal parts are a 1/2-in. standard pipe tee, a finned cooling section, and a Conoseal union.

The key elements in the electrode assembly are a solid electrolyte, thoria yttria tube, and a copper-copper oxide reference electrode located inside the bottom of the tube. The electrode assembly is inserted into the housing and coupled by means of the Conoseal union. Sodium is allowed to freeze in the annular space between the electrolyte tube and the finned tube. The sodium in the annulus provides a primary seal and protects the electrolyte tube from thermal and mechanical shock. The electrode assembly is easily installed and replaceable in the field.

The lower end of the electrode assembly, which contains the reference electrode, projects into the sodium flowing through the tee. The electrode assembly in contact with flowing sodium forms a galvanic cell which develops a voltage that is a direct measure of the activity (or effective concentration) of oxygen in the sodium.^{2,3}

The Indicator measures the voltage developed by the Sensor. It is a specially designed differential voltmeter with a very high input impedance. The input circuitry is guarded and special precautions are taken with insulators, electrical connections, and cables to eliminate error-causing leakage currents. The Indicator is provided with a recorder output and alarms to indicate malfunctions and high or low limit oxygen contents.

The development and testing of this meter was performed in a forced circulation sodium loop built of type 304 stainless steel pipe. The loop contained an electromagnetic pump and flowmeters, main heater, surge tank, cold trap, plugging meter, four parallel test sections, and provisions for sampling and injecting impurities. The loop contained approximately 100 lb of sodium. The flow past the oxygen meters could be varied between 0.2 and 3 gpm. Cold trap temperatures could be varied from 250 to 500°F. The cold trap was loaded with sufficient excess oxide to saturate the entire sodium inventory at all operating temperatures. Oxide content was then controlled by adjusting the cold trap temperature. The loop was designed and operated to insure that no inadvertent diffusion cold traps existed in the circuit, and to insure that the lowest sodium temperature occurred in the cold trap.

The electrochemical oxygen meters were tested by measuring the cell voltage vs oxygen content of the sodium. The oxygen content was varied by means of the cold trap and by injections of sodium oxide using a tea bag technique. The oxide content was determined from cold trap or plugging temperature using published solubility data. In addition, tests were performed to determine the effects of cell temperature and flow rate on meter calibration.

The response of the meters to changes in oxygen content is reproducible and in excellent agreement with theory. Evaluation of data indicates a linear relationship between the logarithm of oxygen concentration and cell voltage. The experimentally determined slope is reproducible and within 10% of the theoretical value. The absolute voltages are also within 10% of theoretical values, but are subject to somewhat greater cell-to-cell variability.

Because of the logarithmic relationship between oxygen concentration and voltage the meters are extremely sensitive to changes in oxygen content. Based upon measured response, a change in oxygen content of as little as 20% at any level (e.g., 0.4 ppm change at 2 ppm) produces the same voltage change of 5 mv as a change of 2 ppm at a 10 ppm level. This is easily detectable provided meter temperature is reasonably constant (see below).

The response time of the meter appears to be as rapid as changes in oxygen content of the sodium can be effected. In oxide injection tests, initial response was observed within one minute after oxide arrived at the meter. Part of this delay may be due to the time required for the oxide to dissolve in the sodium.

Meter temperature coefficients have been determined in the range from 580 to 655°F. The coefficient is 0.52 mv/°F, linear and reproducible for all meters within this temperature range. A temperature variation of 10°F (causing a voltage change of ~5 mv) corresponds to an equivalent oxygen concentration change of approximately 18%. Temperatures can be controlled easily to within 10°F, and the coefficient appears sufficiently reproducible to apply a correction. Test at sodium flow rates within the range from 1/4 to 3 gpm showed no apparent effect upon meter calibration or response to changes in oxygen concentration.

The meter is presently limited to an operating temperature below 650°F, if stable calibration and cell life in excess of 3000 hr are to be assured. Meters have been operated at significantly higher temperature, but experience is limited and thus calibration and life are uncertain. The oxygen meter in its present form provides a means for monitoring oxygen content in full scale systems and laboratory loops. It can be used to follow cleanup operations as well as for continuously monitoring sodium purity during extended operation. The meter provides the only available means for measuring changes in dissolved oxygen activity in the sodium.

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3. Kolodney, M., Minushkin, B., and Steinmetz, H.: Electrolytic Method for the Continuous Measurement of Oxygen in Liquid Sodium, to be published in Electrochemical Technology, July-August 1965.

Figure 1 Liquid Metal Meter Sensor and Indicator

Figure 2 Liquid Metal Oxygen Meter - Sensor

EXAMINATION OF THE FERMI COLD TRAP

by

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Introduction

Under AEC contract, Westinghouse sampled, analyzed, and evaluated the performance of the non-radioactive primary sodium cold trap from the Fermi Atomic Power Plant. The final report, WCAP-4321, is being reviewed prior to distribution. The objectives of the program which follow, were met.

1. Determination of the chemical and physical composition of the trap contents.
2. Determination of the spatial distribution of the chemical species in the trap and their physical form.
3. Evaluation of the operational performance of the trap with particular regard to maldistribution of flow and flow bypassing.
4. Development of criteria for cold trap design.

The Fermi primary sodium cold trap (Figure 1) was a NaK cooled, 500-gallon capacity stainless steel vessel containing 11 mil stainless steel mesh. The maximum flow rate was 100 gpm and the usual operating rate was 50 gpm. The cold trap was used for 3700 hours to purify the half-million pounds of sodium in the primary system.

Results

The difficulty of obtaining suitable sample cores of sodium and mesh was underestimated. Tools that worked well on either sodium or mesh alone failed on the mixture of sodium and mesh. The investigation of a variety of tools and techniques led to the successful sledge hammer method wherein a hole saw was first used to cut through the steel shells. A sharp-tipped, stainless steel tube was then hammered manually into the cold trap to take representative sample cores an inch in diameter and up to twenty-seven inches long. Fifty-five samples were taken in this manner. Samples from the high-oxide region were dissolved in liquid ammonia, and microscopic examination revealed that the oxide coating on the wire mesh was about three mils thick. Metallurgical examination of the stainless steel mesh showed no evidence of surface corrosion or intergranular attack. An experiment employing gamma radiation survey techniques to determine density gradients in the trap was unsuccessful.

Based on the results of over 400 analyses, it was estimated that the trap contained fifty pounds of oxygen, one pound of carbon, and lesser amounts of nitrogen, hydrogen and metallic impurities. Figure 2 shows that the contaminants concentrated in the lower third of the mesh area. The horizontal

distribution of impurities was relatively flat. The carbonate carbon values were about half the total carbon values except in the upper mesh area, where the total carbon was unexpectedly high. The metallic impurities in the cold trap were examined by emission spectrography and are reported in Table I. The high copper content was surprising since there was no apparent explanation of its source.

The fifty-pound value for oxygen was calculated by integrating the oxygen concentrations measured in all zones of the trap. This amount is in harmony with the 32-plus pounds of oxygen removed from the primary system based on plugging temperatures. Since the designed oxygen removal capability was 290 pounds, less than 20% of the expected life of the cold trap was achieved, the trap being prematurely removed from service. The low impurity level of the Fermi primary sodium system and the accumulation of contaminants in the cold trap demonstrated the suitability of the cold trap for general sodium purification.

The inner wall of the cold trap was designed as an inverted cup with straight walls. During examination it was noted that the wall was severely collapsed around half its circumference for almost all of its height (Figure 1). The maximum deformation was about eight inches, and the lowest mesh area was displaced upwards from two to four inches. The most likely explanation of this condition is that a layer of sodium oxide accumulated in the lower mesh area and allowed more than 1 psi of pump pressure to be applied across the innermost wall of the cold trap, causing that wall to collapse. The pressure was then applied to the mesh and caked oxide, displacing them upwards until oxide-free areas opened up and allowed the sodium to flow through. Based on the chemical analyses in the cold trap, flow maldistribution was not evident.

Recommendations

It is recommended that a radial flow design be used to avoid the collapsed wall problem and the biased oxide buildup encountered in the Fermi cold trap design. The inlet sodium would flow in the top and down the sides as before, but would then travel radially into a vertical collection pipe at the center of the trap. The flow pattern is short-circuited at the top mesh area initially. Then, as the sodium oxide accumulates there, the flow pattern would move progressively downward. This would automatically result in a more uniformly loaded trap.

ENRICO FERMI PRIMARY SODIUM COLD TRAP

Figure 1

VERTICAL DISTRIBUTION OF CONTAMINANTS IN THE FERMI COLD TRAP

Figure 2

METALLIC IMPURITIES IN SODIUM

	<u>Min. Detect.</u>	<u>Fermi Cold Trap</u>							<u>Fermi Oct. 1964</u>	<u>U. S. (a) Avg.</u>
		<u>In</u>	<u>Below Mesh</u>	<u>Low Mesh</u>	<u>Mid Mesh</u>	<u>High Mesh</u>	<u>Above Mesh</u>	<u>Out</u>		
Al	8				9					10
Ca	3									190
Cr	3	8		4	25	7		13	27	5
Cu	3	5	5	230	320	63		3		2
Fe	3	48	8	40	160	27	5	30	11	3
K	30	140	150	240	200	140	250	75	75	100
Mg	3			3	3					10
Mn	2			12						2
Ni	3			6						2
Si	8									5
Ti	3			6						-

a. Results are in ppm $\pm$ 30% relative on the basis of sodium metal.

b. Blank spaces are less than minimum detectable limits.

c. The following elements were not detected in any of the samples. Numbers in parentheses indicate detection limits in ppm.

Ag(2), B(25), Ba(3), Be(8), Bi(3), Cd(50), Co(3), Cs(15), In(50), Mo(8), Li(1), Nb(250), Pb(8), Rb(8), Sb(250), Sn(8), Sr(3), Th(250), V(8), W(250), Zn(80), Zr(15).

d. From Liquid Metals Handbook, Sodium Na-K Supplement, TID-5277, July 1955.

Table I

List of Attendees

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A T T E N D E E S

Information Meeting
(Sodium Component Development Program)

Held at: Pick-Congress Hotel
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June 16 and 17, 1965

Sponsored by:
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