

BNL-17901

CONF-730801--4

EXPERIENCE WITH STRESS CORROSION CRACKING AND MATERIALS

COMPATIBILITY AT THE HIGH FLUX BEAM REACTOR

R. W. Powell
J. G. Y. Chow
W. J. Brynda
M. H. Brooks
J. R. Weeks

Brookhaven National Laboratory
Upton, L. I., New York

The primary system components of the High Flux Beam Reactor are fabricated from two types of metals, aluminum 6061 and stainless steel type 304. The aluminum reactor vessel and fuel element cladding establish the basis for the primary coolant system water and cover gas chemistry.

An 18 inch diameter stainless steel weld-neck flange, which was a component of the primary coolant system piping, failed due to intergranular stress corrosion cracking.

Stress analyses showed that hoop stresses due to high bolt tension were sufficient, when coupled with other circumstances, to be of significance.

Detailed metallurgical analyses of some of the cracks which were removed from the pipe identified the cracks as intergranular which originated from the inside surface. The cracks seemed to originate in the heat affected zone but none extended into the weld material.

The replacement flange, of the stub end type, was not subject to hoop stresses shown to have existed in the weld neck type.

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Introduction

Stress corrosion cracking, after six years of operation, was experienced in one 18" dia. weld-neck flange in the HFBR primary piping circuit. Although two other failures had occurred in stainless reactor components in the past, this failure was of greater interest from a metallurgical point of view, since more environmental information was available and more extensive post failure examination was possible. Earlier weld failures of parts of the anti-critical grid and a failure on the baffle plates in the heat exchangers involved circumstances such as stress levels, time of failure, etc., which could not be clearly determined.

The Laboratory was fortunate that the failure occurred in one of two parallel cooling loops which permitted operating the reactor at 75% of normal power while materials for the repair were procured and preparations for the repair were made. Considerable down time was incurred, prior to resuming operation on one loop, because of a thorough examination to assure that no similar cracking was evident in the remaining loop.

A review of the stresses indicated that replacement of the weld-neck type flange with the Van Stone lap joint type flange would eliminate the largest of the stress components present in the flange which failed. Extensive review of system chemistry and materials in use resulted in no changes.

The quality assurance plan was extensive and provided some upgrading of the plant in the area involved insofar as codes, qualifications and inspections are concerned.

Since the HFBR was designed to be closely coupled to reduce the heavy water inventory, all pipes between valves, pumps and heat exchangers were as short as possible. Such a design tends to result in somewhat higher stresses due to thermal expansion and vibration. Replacing a flange in this system afforded an opportunity to measure the deflections which resulted from uncoupling the loops and thus redetermine this component of stress.

The major stress, however, was found to be due to the high bolt loads required to produce sufficient face loading for the type of gasket chosen.

With minor exceptions the repair went as planned and the plant was returned to normal operation. The total downtime attributed to this difficulty was 40 days; the 75% of full power operating time was 80 days. Fourteen half-used fuel elements were sacrificed so that there would be no fuel to contend with during the period of repair.

System Description and Environmental Conditions

The High Flux Beam Reactor (HFBR) is a heavy water cooled and moderated research reactor that operates at 40 MW. (1) The reactor vessel is fabricated from aluminum 6061-T6. The fuel element, side plates and fuel plate cladding material are of 6061 aluminum. The element is similar in many respects to the well known MTR element. A transition from 20 inch aluminum reactor inlet and outlet piping to stainless steel piping is made at flange joints. The isometric diagram, Fig. 1, illustrates the location of all major components.

The 20 inch diameter reactor outlet and inlet piping is divided into two 18 inch diameter primary coolant loops, A and B. Each loop consists of an outlet and inlet valve, primary pump, check valve, and horizontal U-type heat exchanger. The low system volume and high system flow rates causes the entire primary system content to pass through the core twice each minute, thus the N^{16} activity is high throughout the system. All primary components are therefore heavily shielded and inaccessible during operation.

The stainless steel piping and components were fabricated from type 304 S.S. with maximum carbon content of 0.06%. Most of the components were fabricated in 1964 and have been in operation since then.

To provide adequate net positive suction head for the primary pumps and eliminate possible cavitation at the outlet of the fuel element water channels, the surge volume at the top of the reactor vessel is pressurized to 200 psig with helium. The primary system is operated at a flow of 18,000 gpm (9,000 gpm/loop) with a maximum reactor inlet temperature of 55°C and a corresponding outlet temperature of 62.5°C. The quality of the primary D₂O is maintained by continuously passing a small flow (5 to 8 gpm) through a mixed bed resin column loop. The primary system pD is maintained between 5.0 and 5.2 by the periodic addition of nitric acid in an effort to control corrosion and oxide film buildup on the principal heat transfer surface of the aluminum fuel plates.

Conductivity of the purification loop influent and effluent are continuously measured and recorded. In-line measurement of pD at system pressure has proven to be difficult and has resulted in the practice of using the influent conductivity measurement as an indication of pD and as a basis for nitric acid additions. Frequent "grab" samples of the primary water are used to confirm the degree of control afforded by this method. The pD control range of 5.0 to 5.2 at 25°C represents a conductivity range of 3.0 to 1.9 $\mu\text{mho/cm}$ assuming essentially all conductivity is due to nitric acid ions. This method of control works well because the liquid system is completely closed for practical purposes. A resin column is considered to be exhausted when its effluent conductivity reaches 0.3 $\mu\text{mho/cm}$.

The pressure of the cover gas space at the top of the vessel is regulated by bleeding in high pressure helium or exhausting the cover gas from the reactor to the off gas system as necessary. In addition to its principal constituent (He) the cover gas also contains radiolytic decomposition products of D₂O, the gaseous corrosion products, some N₂ resulting from the inclusion of some air each time the vessel is opened, and other minor constituents of air. The cover gas is continuously circulated through a catalytic recombiner which provides a control for the D₂. Since corrosion provides an excess of D₂, control of this excess necessitates the addition of O₂ to the system.

By passing a small side stream of the circulated gas through an inline gas chromatograph, a continuous measurement and recording of D₂, O₂, N₂ and CO₂ is available and provides a basis for the addition of O₂. The O₂ is added periodically, usually in increments of about 2 liters at STP, once per shift, whenever the D₂ exceeds 0.075%. Although one would like to minimize the addition of O₂ to lessen its possible role in stress corrosion, tests run in 1968 indicated lowering the oxygen further would accelerate the decomposition of nitric acid and make pD control difficult. As soon as the initial excess O₂ resulting from opening the vessel is depleted, routine control by this method is practiced.

Control of chlorides and other halogens in the primary system are not difficult and normally chlorides are less than 10 ppb, which is the lower limit of the analytical method used.

Dissolved oxygen, a possible factor in stress corrosion cracking, is a result of the process and other essential controls.

Table I summarizes the principal constituents of primary system.

| | <u>Item</u> | <u>Abundance</u> |
|---------------|------------------|------------------------------|
| Primary Fluid | D ₂ O | 99.37 |
| | Conductivity | 4.6 - 3.0 μmho/cm at 45°C |
| | pD (neutral 7.4) | 5.0 - 5.2 |
| | Chlorides | < 10 ppb |
| | Nitrate | 330 - 550 ppb |
| | Turbidity | None visible through 20 feet |
| | Pressure | 200 - 250 psig |
| | Flow velocity | 37'/sec - 10'/sec |
| | Tritium Content | ≈ 3 Ci/l (March, 1972) |
| Cover Gas | D ₂ | 0.05 - 0.075% |
| | O ₂ | .0064% - 0.0088% |
| | N ₂ | .0800% - 0.020% |
| | CO ₂ | 0 |
| | | |

The Discovery of Cracks

In March 1972, during the last three weeks of an operating cycle, a continuous slow increase in tritium concentration in the exhaust air from the shielded primary heat exchanger rooms was detected. At the time of scheduled reactor shutdown the tritium level had increased approximately 18% above background. The tritium concentration in the primary system water at this time was 3.16 ci/liter. The piping and equipment in the heat exchanger room were inspected during the shutdown period. Two sources of D_2O were found: a leaky flange on a pump, which was immediately corrected by replacing the flange gasket, and five small longitudinal cracks in the body of an 18" weld-neck flange, Fig. 2.

The prompt detection and location of the leak was made possible because of the high tritium content of the system. The drip on the floor beneath the flange was about 2" in diameter and the estimated loss of heavy water from the system, based on stack analyses, was one liter. This is probably the only favorable statement we will ever make about the presence of tritium in this system.

These cracks were approximately 1/2" long and located at the transition from cylindrical neck to tapered body on the bottom quadrant of the flange. They were parallel to each other and also parallel to the direction of the pipe. The cracks did not appear to extend into the weld. Figure 3 illustrates five cracks are clearly shown as a result of the dye penetrant testing from inside the pipe.

The cracked flange was part of the "A" loop primary system piping. This flange was bolted to the outlet side of reactor outlet valve "A" and butt welded to 18" stainless steel piping, which directs primary flow to the suction side of primary pump "A", Fig. 1.

The specifications for the weld-neck flanges were: 18" - 150#, Bored to Sch. 10, A-182, F-304 stainless steel with a maximum carbon content of 0.06%. All primary piping was constructed to ASA Code for Pressure Piping, Petroleum Refinery Piping, ASA B31.3. (1)

Stresses in Cracked Flange

The hoop stresses in piping produced by normal fluid pressure at the point of difficulty are about 8000 psi.

Deflections observed upon unbolting (approximately 0.100") and the force required to reproduce this deflection (approximately 1000 lb.) indicated that there were very low "piped-in" stresses at this point and also that the movement caused by vibration and thermal expansion were insignificant additions. It was known, however, that during construction in 1965 the contractor had considerable

difficulty in making the two flanges (A and B loops) of this type tight at the 400 psig system test. The plant was, in fact, turned over to BNL with the gaskets in these flanges leaking.

It should be noted at this point, referring to Fig. 7, that there is a spacer between the weld-neck flange in question and the valve flange to which it is bolted. The thickness and taper of this spacer were determined as necessary to provide the final closure of each loop. In 1965 our analysis indicated that the leakage had occurred because the bolt loading required to develop the gasket face pressure needed to make the 300 lb. type Flexitallic gaskets tight had caused the weld-neck flange to deflect, thus creating an uneven gasket loading. Our solution to this situation, indicated in Fig. 8, was to provide a gage ring between the flanges to prevent deflection and to provide backup rings for the flanges to provide equal loading. Calculations and measurements were made in rebolting to assure no excessive stressing, but no calculations were done at that time to indicate the stress levels previously experienced in the repeated tightening attempts by the contractor.

Following the discovery of the cracks, extensive calculations of the stress in this weld-neck flange were made. Of particular interest were the hoop stresses produced in the crack area by a combination of the bolt load acting through the gasket, the hoop stress produced by the 400 psig tests, and the end loads.

During the initial system pressure testing stresses as high as 39,700 psi were calculated for the cracked areas. However, stresses continually present during normal operation are of greater importance. In order to estimate the highest possible of these, it was assumed that the gage ring shown in Fig. 8 was, for some unknown reason, ineffective. The 36,000 psi specified bolt stressing acting through the gasket would bring the total stress at the point of the cracks up to 16,800 psi.

Metallographic Examination of Specimens

Five cracks were readily observed visually; (Fig. 2); however, to determine the extent of cracking and deterioration of the metal, the fluid was drained from both loops and internal access was provided by removing the two valve operators and plugs for detailed nondestructive inspection of the flange. This facilitated extensive dye checking and radiographic inspection of both loops. Ultrasonic methods were also employed but with limited satisfaction due to interference from rough machine marks on the weld-neck flanges. Except for the area in question, no other indication of cracks were found.

In order to provide for laboratory examination of the cracks, three pieces of metal, 1-1/4" diameter, were trepanned in such a

way as to remove both ends of the cracks as well as a portion of the adjacent weld. Plugs were welded into the holes thus produced to provide a closure for A loop for subsequent operation of the reactor at 30 MW using B loop, even though A loop was valved off.

Metallographic examination of specimens removed from the cracked area of the flange yielded the following observations.

1. The cracks are intergranular.
2. They originated from the inside surface of the flange.
3. They originated in an area several mm away from the weld, possibly in the heat-affected zone or at the point of maximum stress - where the tapered section intersects the section of constant thickness.
4. The cracks do not extend into the weld metal.
5. The areas where cracks were observed were not heavily sensitized. However, there was some sensitization of the metal due to the welding operation.

From these observations it was concluded that the leak was due to stress-corrosion cracking of the intergranular type which normally requires low pH, dissolved oxygen, high temperature, high stresses and sensitization.

In the HFBR, water chemistry is based on compromises which, to the extent practical, minimize dissolved oxygen to the extent that its absence does not interfere with pH control. The temperature is relatively low. High stresses (80% of yield) were experienced at one time in the past. Normal operating stresses in the area of the cracks may be somewhat higher than normal pipe stresses because of the change in section at the point of difficulty. By making worst case assumptions, a stress of 16,800 psi may have been continually present. The sensitization observed is felt to be insignificant. It was recommended that replacement flanges be designed and installed in such a way as to minimize the locked in stresses and that they be fabricated of type 304-L stainless steel to minimize sensitization of the carbide segregation type in the heat-affected zone of the weld.

These recommendations were based on information obtained from two specimens which were initially examined metallographically to determine the nature of the cracking and whether it had been initiated on the inside or outside surface of the flange. Later, after the repair had been completed, the entire cut out section of pipe and flange was cleaned and made available for further examination.

Figure 2 indicates the locations of the initial cracks and the specimen designations as viewed from outside the pipe.

Figure 4 is 250X magnification of a specimen taken following

the repair and is from a location approximately 90° from sample A (Fig. 2) and in the same section. This and one other similar crack were found in the examination of two one inch diameter specimens. They were observed after the inside surface was polished to remove less than 0.010" of metal.

It should be noted that the crack is very localized in nature and this is believed to be the point of origin of a crack that could have, in time, penetrated the thickness of the metal.

Figure 5 is a 75x magnification of an etched cross section of sample A. (Fig. 2), with the inside edge of the metal in the top of the photograph. The cracking is shown to be entirely intergranular with no branching. The crack was situated in the heat affected zone but stopped at the weld and did not penetrate the area that had been molten. It penetrated the thickness of the metal and caused leakage of the reactor coolant. There was very slight sensitization of the metal and the amount of carbides in the grain boundaries is that expected from the short time that the material was heated by welding. In examining this sample, unlike the specimens obtained later and shown in Fig. 4, no evidence was available as to the origin of the crack.

Figure 6 is a 11x magnification of etched sample B. In this cross section two cracks were revealed; one penetrating the thickness and causing leaking, and the other extending from the inside surface part way through the thickness of the metal.

Examination of the cracks that penetrated the thickness gave no clues as to whether the cracks originated from the inside or outside surface. However, it was found that there were more cracks originating from the inside surface, which is the basis for the conclusion that the cracks originated from the inside. This can be seen in Fig. 4 and 6.

Problem Solution

The primary system is constructed with two types of flanges; one the weld-neck type (Fig. 7) which was subject to the aforementioned difficulties, and the other the Van Stone lap joint (Fig. 9), having a loose flange ring which, upon analysis, showed no hoop stresses due to bolting. In the primary system there are only two 150#, 18" weld-neck flanges. All other pipe flanges are of the 300# Van Stone lap joint type. Two types of gaskets were used in the plant; the Parker-O-Ring type which requires light face loading but exhibits more sensitivity to radiation damage; and the Flexitall type which requires very high bolt loading and exhibits little radiation damage.

In line with recommendations made by the Metallurgy Division, the Van Stone lap joint type flange and the Parker-O-Ring gasket

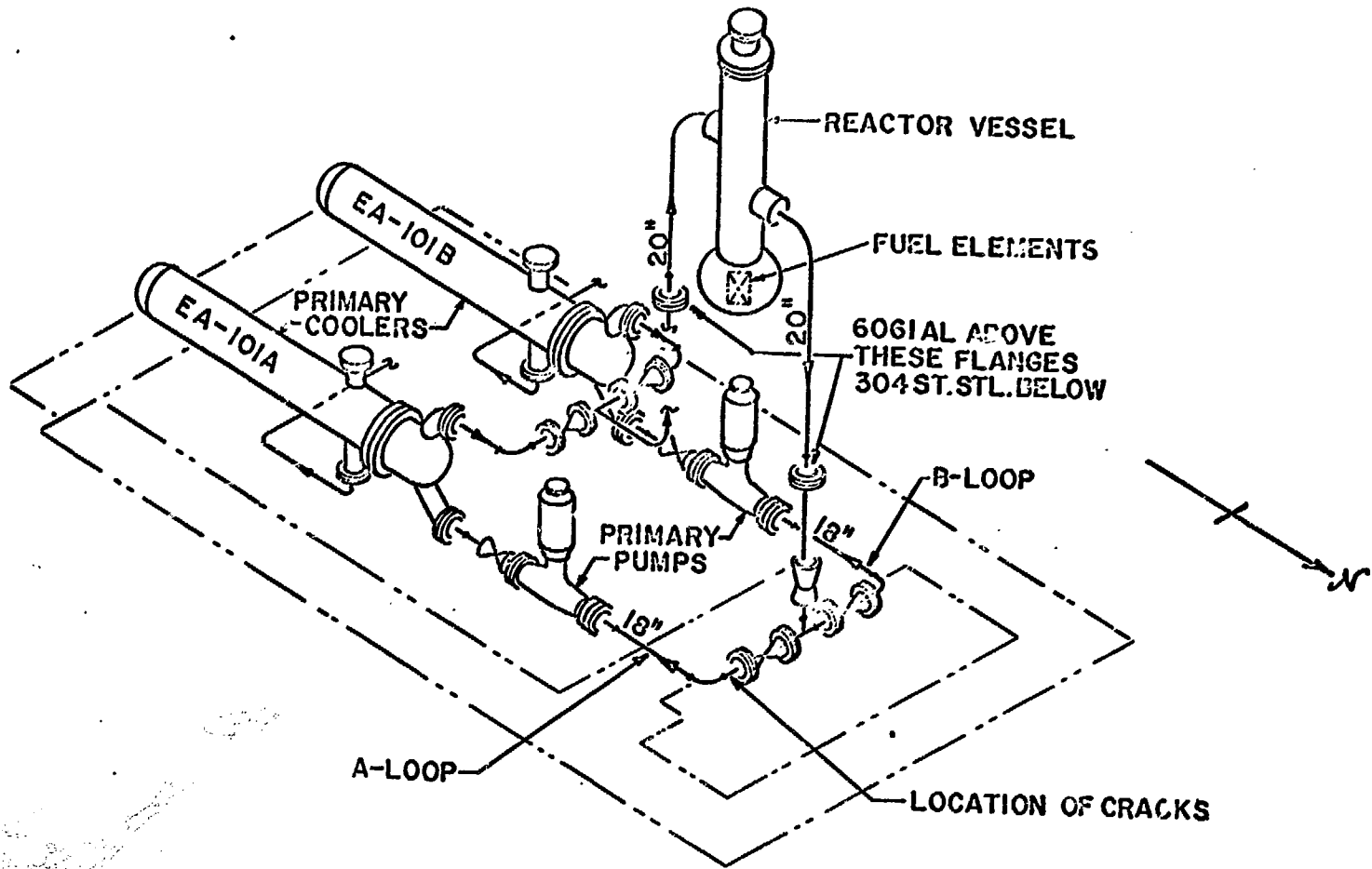
were chosen for the repair of the A loop. It was further decided to replace the Flexitallic gaskets with Parker O-rings at the similar weld-neck flange in the B loop, since actual radiation exposure had been less than anticipated in the design. The configuration for the A loop Repair is shown in Fig. 9 along with the original design which leaked in Fig. 7 and the BNL solution to the original leak problem in Fig. 8.

References

1. Hendrie, J. M., "Final Safety Analysis Report on the Brookhaven High Flux Beam Research Reactor", BNL 7661, April 1964.

List of Figures

1. Isometric of HFBR Primary Cooling Circuit.
2. Photograph of the lower side of the cracked flange, showing the location of the cracks and the samples examined.
3. Photograph of the inside of the cracked flange as indicated by dye penetrant testing.
4. 250X - Electrically etched in Oxalic Acid. Microcrack 0.010" below inside surface. Specimen extracted 90° from specimen A shown in Fig. 2.
5. Cross-section of Sample "A" showing intersection of crack with inner surface of flange; 75X, oxalic acid etched inside pipe - top.
6. General appearance of the cracks near both edges of the sample, showing both the major and lesser cracks. 11X, oxalic acid-etched. Inner surface is at upper side.
7. Original joint construction.
8. Startup solution for tight joint.
9. Proposed repair.



HFBR PRIMARY COOLING CIRCUIT

FIG. 1

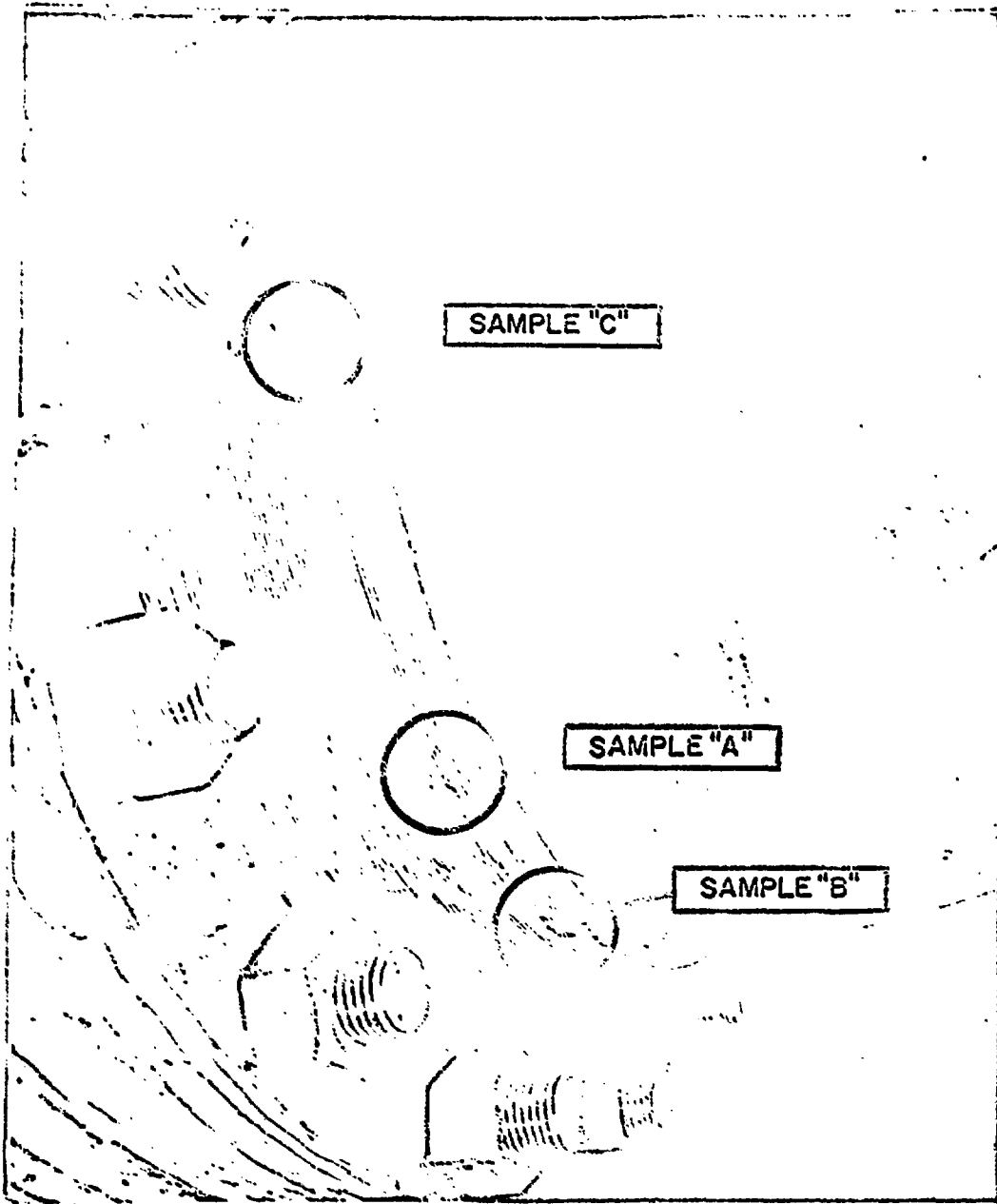


Figure 2

Photograph of the lower side of the cracked flange, showing the location of the cracks and the samples examined.

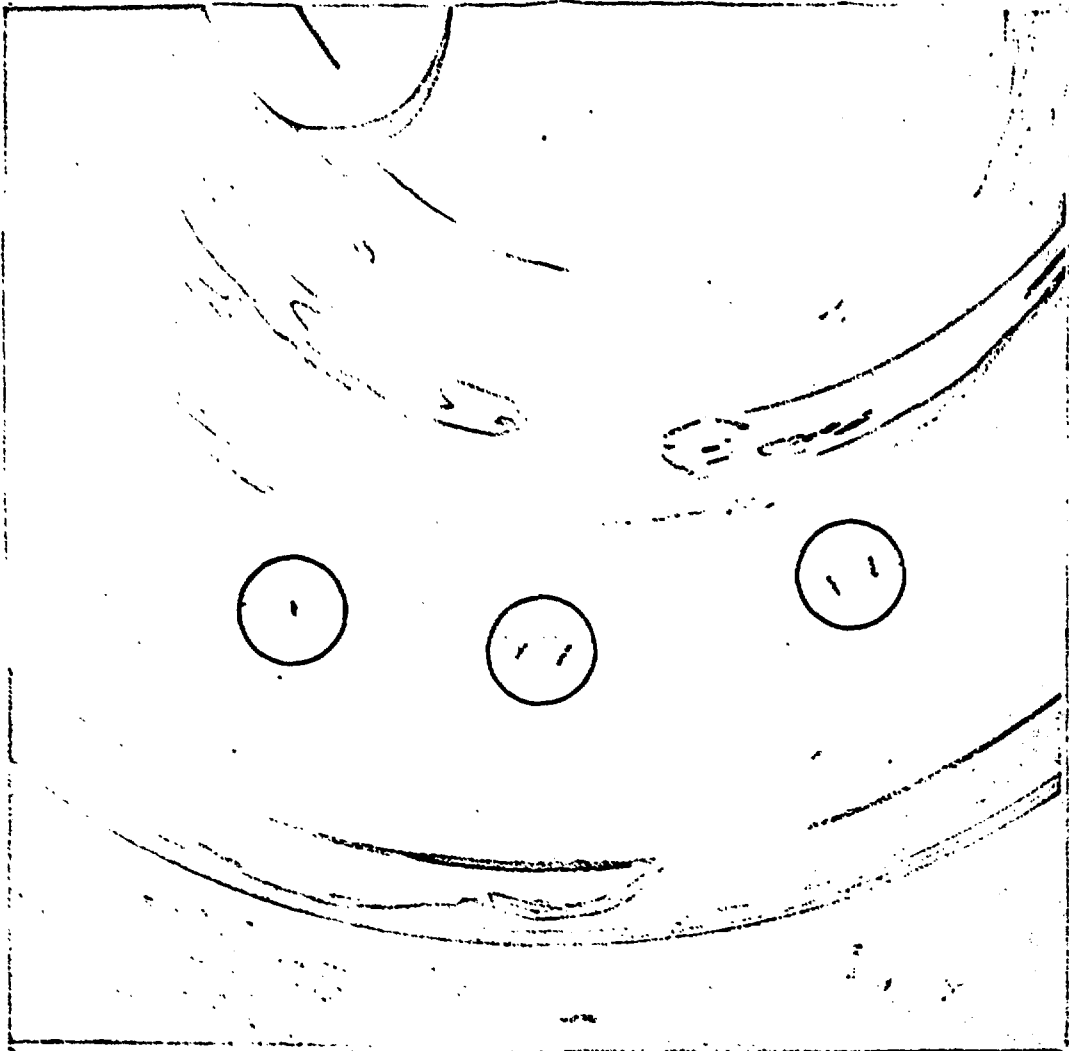


Figure 3

Photograph of the inside of the cracked flange as indicated
by dye penetrant testing.



Figure 4

250X - Electrically Etched in Oxalic Acid

Microcrack 0.010" below inside surface. Specimen extracted 90° from specimen A shown in Fig. 2.



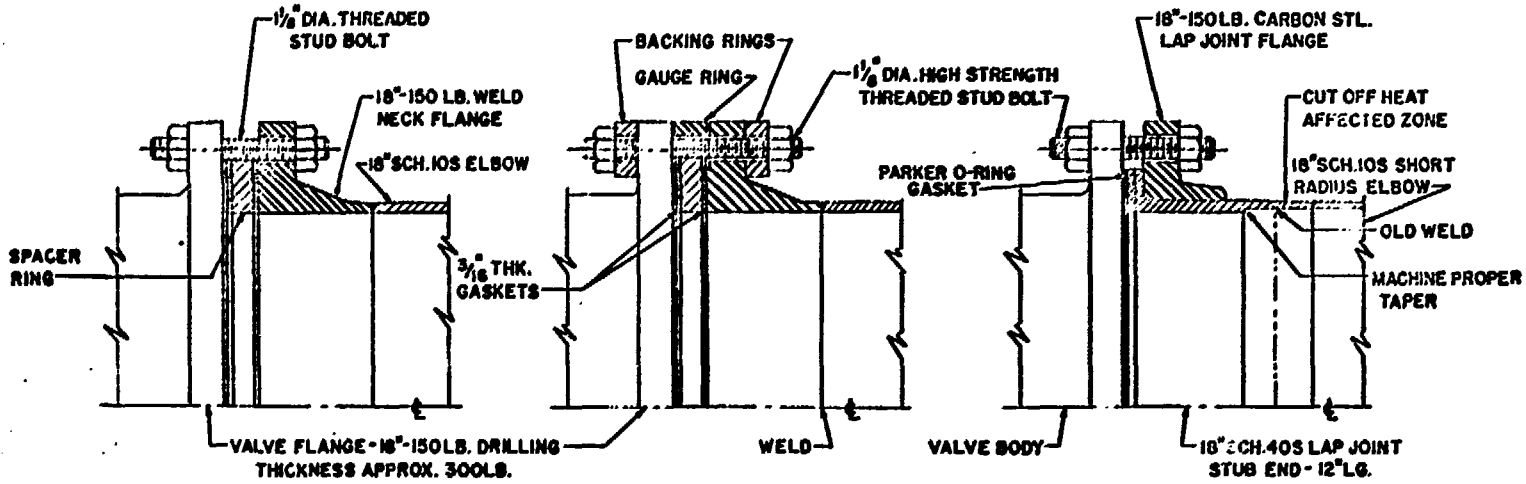
Figure 5

**Cross-section of Sample "A"
showing intersection of
crack with inner surface
of flange; 75X, oxalic
acid etched inside pipe - top**



Figure 6

General appearance of the cracks near both edges of the sample, showing both the major and lesser cracks. 11X, oxalic acid-etched. Inner surface is at upper side.



ORIGINAL JOINT CONSTRUCTION
FIG. 7

**START UP SOLUTION
FOR TIGHT JOINT**
FIG. 8

PROPOSED REPAIR
FIG. 9