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POST OFFICE BOX P
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ORNL
CENTRAL FILES NUMBER

CF- 56-6-115

DATE: July 20, 1956

COPY NO. 3

SUBJECT: Examination of the North Carolina State College Reactor Core

TO: E. G. Bohlmann

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EXAMINATION OF THE NORTH CAROLINA STATE COLLEGE REACTOR CORE

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ABSTRACT

The postfailure examination of the North Carolina State College Reactor revealed a failure of the core wall in a very localized region just above the liquid level. The apparent cause of failure was a pitting type of corrosion. The failure of the cooling system was not located, but several possible sources of failure were observed metallographically.

INTRODUCTION

Early in May 1955, after a day of normal operation and three days of shutdown, the North Carolina State College reactor system was found to have developed a leak. Subsequent examination between then and early June revealed that there were apparently two leaks in the system. One of these had permitted the escape of a small volume of solution into the aluminum safety thimble, where it was retained. A second, very small leak had permitted some contamination of the cooling water.

The history immediately preceding the failure had been one of extended low-temperature operations at higher than normal power levels. There had been some difficulty, because of this schedule, with minor radiolytic-induced precipitation and erratic catalytic recombination. The latter phenomenon had resulted in some abrupt pressure surges in the system.

At the request of the College through the Oak Ridge AEC Office, Oak Ridge National Laboratory agreed to disassemble and examine the reactor core. This examination was assigned to the Corrosion Section of the Reactor Experimental Engineering Division.

The reactor core assembly in the safety thimble was transferred to ORNL in a carrier on July 30, 1955. At the same time a separate, shielded 50-gal drum was transferred, which contained the various rinse solutions used in cleaning the assembly.

DISASSEMBLY AND PHYSICAL EXAMINATION

The initial step in examining the reactor was of necessity the removal of the graphite rods, sheets, and pellets which were located inside the aluminum safety thimble on top of the core tank. This

removal was accomplished, while the reactor assembly remained in its shipping container, by the use of a vacuum cleaner with a disposable bag and a pair of standard laboratory tongs.

All material was monitored for radioactivity as it was removed; no activity above background was detected. The general activity above the core rose from 100 mr/hr to approximately 1 r/hr at 2 ft after the removal of the graphite.

At this point an unsuccessful attempt was made to lift the core vessel out of the safety thimble.

The entire core assembly was then transferred to the Corrosion Examination Facility Hot Cell for further operations. Because of the extremely thin walls of the core forcible removal of the core was avoided because it might result in mechanical damage which would mask the real failure. Consequently, the first step was to determine why the core was binding in the safety thimble. In order to do this, it was decided to remove the portion of the thimble above the top of the core vessel. Figures 1 and 2 show the method of holding and the location of the first cut. Figure 3 shows the core region after removal of the upper part of the thimble. Visually, the core seemed to be almost an interference fit in the thimble; so it was decided to slit almost through the aluminum thimble and try to peel it away from the core wall. In Fig. 3 one of the lines of cutting is indicated. Figure 4 shows the end result of the attempt to peel the aluminum envelope. Cables that were extended through holes in opposite walls of the cell were used in this operation. Dimples were punched in the aluminum thimble without relieving the bond between core and thimble. In an effort to break this bond a solution containing 800 ml of water, 200 ml of concentrated nitric acid, and 14 g of

Fig. 1. Holding Method for Cutting Top of Thimble.

Fig. 2. Location of First Cut for Removal of Thimble.

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Fig. 3. Reactor Core After Removal of Thimble Top. Lines of proposed slitting are shown on remaining portion of thimble.

Fig. 4. Reactor Core Assembly After Thimble Slitting and Attempted Mechanical Removal.

mercurous nitrate monohydrate was trickled into the crevice between the stainless steel core and the aluminum thimble. This solution readily attacks aluminum and aluminum corrosion products but does not significantly attack stainless steel. An attempt was made to recover all this solution for uranium analysis.

After this treatment it was possible to spread the aluminum thimble sufficiently to remove the core assembly from the thimble. Figure 5 is a typical view of the reactor core as removed from the thimble. It can be seen that not all the corrosion products were removed by this chemical treatment and that some deposits are still attached to the stainless steel walls.

The core tank was next submerged in water contained in a battery jar, and approximately 5 psi of air was introduced into the core through the gas outlet pipe. Figure 6 is a photograph of the core vessel in water under air pressure. The location of the core leaks can be seen by the air bubbles rising from the holes in the tank wall. Figure 7 is a photograph of the same region without

the air flowing. Five distinct holes can be seen in this photograph. Careful inspection of the remainder of the core tank both immersed and with a saturated soap solution failed to locate any other leaks.

Thus it was found that the leaks occurred approximately $9\frac{1}{2}$ in. from the bottom of the core. Surface examination of the cadmium shim plates indicated that the solution which escaped from the core also reached a level in the safety envelope of approximately $9\frac{1}{2}$ in. The yellow-colored precipitate on the shim plates was removed with the same stock solution of nitric acid-mercurous nitrate used in freeing the core from the thimble. The two solutions were combined in one container for uranium analysis.

In order to facilitate sectioning of the core for interior examination, all external piping was cut off approximately 2 in. above the core cap. After careful examination of the construction drawings, sectioning lines were sketched on the core; these lines were to be followed during sectioning to prevent damage to the cooling coils inside the

Fig. 5. Stainless Steel Core After Chemical Removal of the Thimble.

Fig. 6. Core Tank Submerged in Water and Pressurized with Air. Note air bubbles escaping from holes in tank wall.

Fig. 7. Core Tank Submerged in Water. Five holes may be seen in tank wall.

tank. Figure 8 is a photograph of the top of the core tank after removal of the exterior piping and with the section lines sketched in.

After a considerable amount of manipulation, the core tank was quartered without damaging the cooling coils. Figure 9 is a view of the interiors of all four sections. The holes can be seen located just above what was the solution-vapor interface, as defined by a difference in corrosion products attached to the stainless steel surfaces. Figure 10 is a magnified view of this region. It is significant that the holes appear to be smaller at this inside surface than at the outside surface. This difference will be more apparent in later photographs.

Although the leak in the core wall had been located, there still remained the problem of finding the source of activity found in the cooling water. Consequently, one end of each coil was crimped and air pressure was applied to the other end while the whole unit was immersed in water or a thick soap solution. Although the end connections

were never made leakproof, sufficient air pressure was applied to reveal any large defect in the coils. None was found. Because of the high levels of radioactivity of the components and the necessity of direct manipulation in making tests for very small leaks, and because of the peculiar localization of the attack on the core walls, it was agreed with the North Carolina State representatives to remove sections of one coil in the region of the solution interface for metallographic examination. It was realized that the section containing the actual leak might thus be lost, but it was believed that metallography would yield a clue to the actual cause of the leak and greatly expedite the over-all examination. The coil chosen was the one adjacent to the holes in the core wall.

This quarter was then sectioned as follows:

1. The core wall and bottom were cut from the core cap just below the seal weld.
2. Three sections were then cut from the coil: a straight section at the outlet including the solution interface region, one U-bend from the same region

Fig. 8. Top of Core Tank After Removal of External Piping. Proposed sectioning lines are shown.

Fig. 9. Inside Surface of Sectioned Core Tank Showing Solution Level and Internal Piping.

Fig. 10. Close-up View of Internal Tank Wall in Area of Leaks.

and a second U-bend from the bottom, or totally immersed, region of the coil. Figure 11 is a photograph of the cooling coil after removal of the core wall. The location of samples taken from the interface region are shown.

3. Two sections of the core wall were taken: one which included the holes and the other from near the bottom of the core which included the weld between the spun bottom and the core wall.

All these sections were sent to the Remote Metallography Group of the Solid State Division for further sectioning and examination.

Since, as will be noted later, the metallographic examination failed to provide absolute evidence of a leak in the cooling coil sections examined, a helium leak test of the remaining three coils was made. This test also failed to locate any leaks.

Three additional metallographic samples were taken: a section of the core wall in the interface region diametrically opposite the section containing the holes and two additional pieces of the same cooling coil – a second U-bend from the bottom of the coil and a portion of the inlet including the tube to the core cap weld.

Fig. 11. Section of Core After Removal of Core Wall; Set Up for Sampling of Cooling Coil for Metallography.

METALLOGRAPHIC EXAMINATION

The results of this examination are reported here almost verbatim from the original report.¹

Metallographic-sample preparation followed the conventional remote metallographic practice as employed in the Solid State Division at ORNL.

The appearance of the penetrations through the core vessel at the liquid-vapor interface level is shown in Figs. 12 and 13. Figure 12 is typical of the appearance of a penetration as it appears from the interior surface of the core vessel. Figure 13 shows the penetration as it appears from the exterior surface of the tank. A vertical cross section through the core wall in the area of this penetration is shown in Fig. 14. The extreme undercutting of the interior surface of the pit and the flaky appearance of the material remaining in the void may be noted in this photograph. Figures 15 and 16 are additional views of these penetrations. It is of interest to note that the pits are expanded below the surface and that the surrounding microstructure shows no abnormalities.

Corrosive attack by the fuel solution upon the core-vessel walls was slight except for the pitted areas. Figure 17 illustrates the appearance of the interior surface of the core vessel wall at a position slightly above the location of the pene-

trations. The area shown here was theoretically above the liquid level of the fuel solution and thus was exposed only to the vapor phase of the fuel; corrosive attack is less than $\frac{1}{4}$ mil and appears to consist of slight penetrations at the grain boundaries, resulting in what appears to be a strongly etched surface. Figure 18 shows an area from slightly below the penetrations. This area was below the fuel level and thus was exposed to the liquid phase of the fuel solution. Corrosive attack is identical in both type and degree to that shown from above the fuel level. Examination of the section of core wall from a position diametrically opposite from the penetrations showed similar results. The corrosive attack was similar in type and depth to that shown in Figs. 14 and 18. Again, there was no apparent difference between regions exposed to the liquid and those exposed to the vapor. In fact, it was impossible to determine the fuel solution level metallographically by using type or depth of corrosive attack as an indicator.

No evidence of knife-line attack was noted at the weld of the core bottom to the core wall (Fig. 19). Slightly accelerated corrosive attack was found in the heat-affected zone, where penetrations of 1 mil were noted (Fig. 20). Areas outside the heat-affected zone showed only slight corrosive attack, such as was previously shown in Fig. 18.

Corrosive attack by the vapor phase of the fuel solution upon the core header (Fig. 21) was again generally identical to that shown previously. Two areas exhibiting a pitting type of corrosive attack (Figs. 22 and 23) were noted in the heat-affected zone resulting from the header-to-tube welding operation. No accelerated corrosive attack was found in the crevice between the header and tube nipple (Fig. 24). The nipple itself showed no evidence of corrosive attack, and the header showed only slight attack, similar to that shown in Fig. 21. The crevice between the cooling tube and the nipple is shown in Figs. 25 and 26. No accelerated attack was evident, with the possible exception of the fusion zone of the tube-to-nipple weld (Fig. 26).

Figures 27 and 28 illustrate the appearance of the cooling coils. Figure 27 shows the inlet line in the area of the liquid-level interface. Figure 28 is typical of one of the U-bends, the area shown also being at the liquid-level interface.

¹A. E. Richt, *Metallographic Examination of Components from the North Carolina State College Reactor*, ORNL CF-56-3-158 (March 30, 1956).

Fig. 12. Interior Surface of Core Vessel Showing Pitted Area. $\approx 10X$.

Fig. 13. Exterior Surface of Core Vessel Showing Pitted Area. $\approx 10X$.

Fig. 14. Vertical Cross Section of Core Vessel Through Pitted Area. 20X.

Fig. 15. Interior Surface of Core Vessel at Pitted Area. 150X.

Fig. 16. Exterior Surface of Core Vessel at Pitted Area. 150X.

Fig. 17. Typical Corrosive Attack of Core Vessel by Vapor Phase of Fuel Solution. 250X.

Fig. 18. Typical Corrosive Attack of Core Vessel by Fuel Solution. 250X.

Fig. 19. Knife-Line Area of Core-Vessel Weld. 250X.

Fig. 20. Heat-Affected Zone of Core-Vessel Weld. 250X.

Fig. 21. Typical Corrosive Attack of Header. 250X.

Fig. 22. Pitted Area in Core Header. 250X.

Fig. 23. Pitted Area in Core Header. 250X.

Fig. 24. Header-to-Nipple Crevice. 150X.

Fig. 25. Nipple-to-Tube Crevice. 250X.

Fig. 26. Fusion Zone of Nipple-to-Tube Weld. 250X.

Fig. 27. Cooling-Coil Inlet Line at Fuel Liquid Level. 10X.

Fig. 28. Coil Bend at Fuel Liquid Level. $\approx 10\times$.

Corrosive attack by the fuel solution upon the exterior surfaces of the cooling coils was generally similar to that shown to have occurred on the core-vessel walls. Figures 29 and 30 are typical of the type and depth of attack above and below the liquid level of the fuel solution. As in the case of the core-vessel walls, there was no apparent difference in type or degree of attack with respect to the fuel solution level. In addition, there was no indication that residual stresses resulting from the bending of the cooling coils in fabrication had any effect upon the corrosive resistance of the tubing. A number of transverse sections from the cooling coils showed evidence of two areas of localized attack (Figs. 31 and 32). The depth of attack was generally 2 to 3 mils and was noted only in sections which were below the liquid level. The area between the penetrations (which always occurred in pairs) appeared to be somewhat dendritic in nature when the specimen was examined microscopically and slightly out of focus. As a result, it seems possible that these penetrations are a result of a knife-line type of corrosive attack in this welded and drawn tubing.

Corrosive attack on the interior surface of the

cooling coils by the cooling water was sporadic. Generally there was little or no evidence of corrosive attack, as shown in Fig. 33. All sections, however, showed isolated areas of intergranular attack such as is shown in Fig. 34, in some instances to a depth of approximately 4 mils. These areas did not seem to be related to the residual-stress pattern or the location of the tube weld. A longitudinal section through one of the U-bends of the tubing showed evidence of what appeared to be transgranular cracking (Fig. 35). This occurred at the outer radius of the bend at what would be the position of highest residual tensile stress. This particular area of interest was lost when the sample was repolished after an unsuccessful etching attempt, and, as a result, only an unetched photograph is available. Localized corrosive attack was noted at the interior surface of the cooling coils (Figs. 31 and 36). The similarity of this attack to that shown in Fig. 32, which occurred on the fuel side of the tubing, may be noted. Again this attack occurred at what appears to be the weld deposit of a welded and drawn tubing.

Fig. 29. Typical Corrosive Attack of Cooling Coils by Vapor Phase of Fuel Solution. 250X.

Fig. 30. Typical Corrosive Attack of Cooling Coils by Fuel Solution. 250X.

SOLUTION ANALYSIS

It had been estimated by the staff at North Carolina State College² that between 14 and 23 g of uranium could have been lost from the reactor core.

A fair proportion of this uranium was probably contained in the rinse water which had been used

²C. K. Bect et al., *Further Data on Reactor Operations up to Removal of Fuel from the Fuel Solution*, NCSC-108 (Aug. 10, 1955).

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Fig. 31. Location of Areas of Localized Corrosive Attack of Cooling-Coil Tubing.

by them to flush the core and the annulus between the core and safety thimble after the reactor had been drained. All these rinse solutions, together with a sample of the fuel solution removed from the core, were brought to ORNL for analysis. As a further precaution, every effort possible was made at ORNL during the dismantling operation to recover any remaining uranium. It was for this reason that the various solutions used in dismantling and inspecting the core were saved and analyzed.

Table 1 is a listing of the weight of uranium recovered in the various rinses and examination solutions.

A sample of the fuel solution as shipped to Oak Ridge at a later date gave the following analysis:

pH	2.75
U, mg/ml	54.73
SO ₄ ²⁻ , mg/ml	22.48
Cu, µg/ml	23
Ni, µg/ml	28
Cl ⁻ , µg/ml	218
Cr, µg/ml	0
Fe, µg/ml	169
Pu, counts/min/ml	116

Fig. 32. Localized Attack of Cooling Coil by Fuel Solution. 250X.

Fig. 33. Typical Attack of Cooling Coils by Cooling Water. 250X.

Fig. 34. Intergranular Attack of Cooling Coils by Cooling Water. 250X.

Fig. 35. Possible Stress-Corrosion Cracking of Cooling Coils. 250X.

Fig. 36. Localized Attack of Cooling Coils by Cooling Water. 250X.

The most significant part of this analysis from our point of view is probably the high chloride content.

TABLE 1. URANIUM RECOVERED FROM VARIOUS RINSE, DISMANTLING, AND INSPECTION SOLUTIONS

Solution Identification	Uranium (g)
North Carolina State core rinse No. 1	3.25
2	1.48
3	3.14
4	0.52
North Carolina State annulus rinse No. 1	0.56
2	0.20
Core-removal solution	8.21
Core-immersion solution	2.06
Coil-immersion solution	0.85

DISCUSSION OF RESULTS

The examination of the core vessel itself failed to show any leaks other than the five holes located slightly above what appeared to be the solution-vapor interface. Metallographic examination of various sections from the core wall and core cap, or header, did not reveal any metallurgical defects which might account for the failure. Some evidence of a pitting type of attack was found in the core header, but this was only in the areas highly affected by the tube-to-header welds. No such pitting was found in the region of the core-wall-to-bottom weld, and there was no indication of heat-affected areas in the region of the actual failures. Qualitative evaluation of the surfaces indicates that there was very little generalized corrosion of the core vessel or the cooling tubes by the reactor solution.

Some experiments performed at ORNL by Moore³ in which various chloride additions had been made to uranyl sulfate solutions showed pitting attack of type 347 stainless steel in the vapor phase. These tests were made at 100 and 250°C with specimens in both the solution and the vapor phase. Even at the higher temperature the samples exposed to the solution appeared to be only

slightly affected by chloride up to 100 ppm. However, the presence of only 5 to 10 ppm of chloride at 100°C caused severe pitting of the vapor-phase samples (Fig. 37).

The similarity in appearance of the pits in the reactor core with those from the vapor-phase samples in the laboratory tests (Figs. 14, 15, 16, and 37) leads us to believe that the high chloride content of the fuel solution may have been the cause of the reactor core vessel failure. Unfortunately, such a comparative evaluation can only be indicative and not conclusive and does not explain why there were only five penetrations in the particular location and configuration found. Inclusions and arc strikes were also considered but no supporting evidence for such hypotheses was found. In any case, the chloride content of uranyl sulfate solutions in stainless steel equipment should be kept below 5 ppm and preferably below 1 ppm.

Unfortunately, the reported leak in the cooling coils could not be located. The one coil that was sectioned before helium leak testing may have contained the failure. The widespread but spotty intergranular attack of the coils on the water side and the possibility of stress-corrosion cracking in the area of the U-bends may explain such a leak. Another possibility is the localized attack on both the water and solution sides adjacent to the seam welds in the tubing. This possibility could be eliminated in the future by the use of seamless tubing.

Fig. 37. Pitting of Vapor-Phase Sample in Laboratory Test. 75X. Reduced 32%.

³G. E. Moore, *Summary of Laboratory Tests*, ORNL CF-55-12-70 (Dec. 12, 1955).

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It was suggested by Foley⁴ of the Babcock & Wilcox Co. that carburization may have been responsible for the intergranular attack on the water side of the tubes. Subsequent metallographic examination failed, however, to show any indi-

⁴W. R. Foley, letter dated March 27, 1956 to M. A. Cordovi of the Babcock & Wilcox Co. covering the discussion concerning the North Carolina State Reactor at ORNL on January 12, 1956.

cations of such carburization. Thus the possibility remains that the chloride content of the tap water used in cooling the reactor may be responsible for the attack found on the cooling coils.

ACKNOWLEDGMENTS

The authors would like to thank W. H. Wagner, B. W. McCollum, and R. L. Plemons, who performed the remote operations in dismantling and examining the reactor.