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SUMMARY STATUS REPORT
INTERNAL CORROSION OF
RIBBED ALUMINUM PROCESS TUBES

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SUMMARY STATUS REPORT

INTERNAL CORROSION OF
RIBBED ALUMINUM PROCESS TUBES

by

P. A. Carlson
D. H. Curtiss
N. R. Miller
F. W. Van Wormer

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I. INTRODUCTION

The increasing incidence of leaking process tubes, and the approaching end of the useful life of process tubes in the C and K Reactors, have focused attention upon the various sources of aluminum process tube leaks. Further, the replacement of large numbers of process tubes with new ones, also of aluminum, will require continued attention to these sources to make efficient use of the new tubes.

One of the sources of process tube leaks is the corrosion that attacks the interior surface of the process tube. The factors influencing the extent of this corrosion attack are varied and complex, and in recent years, the corrosion service conditions have become increasingly more severe. Each of the factors involved in determining the rate of corrosion attack has thus become individually more important, and the need to understand the interrelationships among them has increased. It is the purpose of this report to discuss the technical factors contributing to the internal corrosion of the process tubes, to review the way some of these factors have varied in the past, to examine the means available for evaluating the extent to which corrosion has damaged the tube walls, to comment upon the ways in which knowledge of ~~the various~~ ^{these} factors can be utilized to obtain desirable service lives from the process tubes, and to evaluate the current status of C and K Reactor process tubes and their probable remaining lives.

There are, of course, other important sources of tube leaks and factors to be considered in the over-all determination of process tube life. Tube failures arising from stress conditions in the tube wall or the tube flange, operational considerations such as the ability to replace process tubes before failure versus the consequences of encountering some leaking tubes, and the effects of tube replacement and tube leaks upon over-all production continuity, while vital considerations in obtaining the most efficient and productive service from the process tubes, are not within the scope of this report.

II. SUMMARY AND CONCLUSIONS

The technical factors known or considered to have importance in determining the rate and extent of corrosion damage occurring at the internal surface of the aluminum process tube have been examined. Of these, water quality or corrosivity and the tube wall temperature have the greatest impact upon the amount of corrosion attack. Since water quality tends to remain relatively constant, the general picture is one in which temperature effects assume prime importance in determining the degree of corrosion damage occurring at the process tube walls. This being the case, great importance must be attached to those considerations tending to create undesirable high tube wall temperatures. The eccentric position of the fuel charge within the tube (initially toward the top of the tube and finally, after accumulated rib damage has occurred, toward the bottom of the tube) is a most important feature governing local tube wall temperatures.

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A review has been made of past efforts to maintain an adequate knowledge of the conditions of reactor process tubes. It appears that "loss of control" has occurred on some occasions. On most of these occasions, the results were not serious; unpredicted, large-scale retubing did not result, nor was the tube damage significantly greater than would have otherwise occurred. On a few occasions, the lapses have been serious. In particular, the failure to note the shift of the point of maximum corrosion to the bottoms of second-generation tubes resulted in the initiation of major replacement of these tubes being required about six months earlier than had been predicted. Also, the lapses of control have prevented the initiation of corrective action which might have extended the service lives of some tubes.

There are presently available three useful tools for ^{determining} ~~monitoring~~ process tube conditions: out-of-reactor destructive examination, the eddy current probolog, and the thermocouple probe that replaces the rear dummy pattern to measure local temperatures about the coolant annulus of individual fuel charges. Out-of-reactor destructive examinations provide detailed profiles of tubes removed from the reactors, but must be quite restricted in number. With proper allowance for its accuracy, the probolog is a useful instrument with which to ~~monitor~~ ^{determine} trends in the accumulated corrosion damage occurring in the process tube, and to guide tube replacement selections. The thermocouple probe can be helpful in measuring the eccentricity temperature effects that are important both in monitoring tube condition and in evaluating corrective actions such as the use of water-mixing elements in the fuel charge. The need for two more ~~monitoring~~ devices, a sector probolog measuring wall thickness at both the top and bottom of the tube and an instrument to measure process tube rib height in-reactor, is clearly established. Technical feasibility of the sector probolog has been demonstrated. When operational, the sector probolog will greatly facilitate maintaining an adequate current knowledge of tube conditions.

Over all, this study of the technical factors leads to the conclusion that future tube lives as limited by internal corrosion will depend largely on monitoring and corrective efforts. Specifically, with techniques and technology currently available, average central zone tube life of four years is considered attainable with 95°C reactor bulk effluent temperature operation, if appropriate monitoring and corrective measures are taken. It is beyond the scope of this report to conclude whether or not it is economical to take these steps.

Recognition of the effects of the technical items discussed, together with operational considerations such as tube replacement capability and production incentive, can be combined to obtain the greatest economic service life from the process tubes. In other words, internal tube corrosion may determine when process tubes will leak, but should not be considered by itself to establish tube replacement schedules.

For information, current evaluations of the condition of process tubes in the C and K Reactors have been included as Appendices A and B.

New process tube designs have been formulated. These designs provide for a concentric, uniform wall process tube. Greater wall thickness at the bottom

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of the tube should result in a measurable increase in tube life. Specific dimensions of these tubes are included as Appendix C.

III. RECOMMENDATIONS

The material reviewed in the course of this study and the conclusions summarized above give rise to the following general recommendations:

- A. The efficient use of ribbed aluminum process tubes in the existing reactors is ~~quite~~ dependent upon the existence of an adequate tube condition monitoring program. It is therefore recommended that a routine program of monitoring be established at all reactors which adequately provides for the characterization of the majority of the process tubes. This program should include routine and repetitive probologging of selected process tubes, the expanded use of thermocouple probes, provision for the efficient collection, reduction, and dissemination of monitoring data, and the early development and use of the sector probolog and of a device for the in-reactor measurement of process tube rib height.
- B. An integrated study should be conducted of all factors bearing upon the optimum use of aluminum tubes. In addition to the factors outlined in this report, many important operational and economic considerations should be included in this study. The study should be initiated concurrently with the establishment of the monitoring program recommended above, from which it would draw current data and upon which the study would depend for periodic reassessment.

It is recognized that these general recommendations have been previously advanced by others. Specific recommendations for action expected to prolong the usefulness of process tubes in the K Reactors have been included in Appendix B.

IV. DISCUSSION

A. Technical Factors Affecting Tube Corrosion

The factors that determine the useful life of the process tubes, and when and how they will fail, are varied and complex. Of these, those that are involved in the internal corrosion of the process tubes are especially complex and are such that the interrelationships of the various factors are not always capable of clear definition. In this section, the relative contributions of water quality, temperature, radiation, coolant flow, mechanical effects, and materials on the internal corrosion of process tubes are discussed. This is followed by a discussion of how these factors influence fuel element and process tube design.

1. Water Quality

Water quality variables have important effects on both the uniform and non-uniform corrosion rates of aluminum.

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a reducing the diffusion rate
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Uniform corrosion rates are apparently controlled by the rate of diffusion of an oxidizing agent (say, hydroxyl ion) across the aluminum oxide ~~corrosion~~ layer formed on the metallic surface. (1,2) To reduce corrosion rates, therefore, requires increasing the diffusional resistance of the oxide layer. This in turn requires an improvement in the quality of the oxide layer, an increase in the oxide layer thickness, ~~or a lower temperature~~ *or a combination of these steps*.

Tests have consistently shown that steps which minimize the rate of dissolution of the oxide layer in turn minimize corrosion rates. (2) Such steps can result in thicker films (with lower dissolution rates, more of the oxide formed will stay in place) or films of better quality (with low dissolution rates, the films formed have minimum porosity and give maximum protection). (3) For example, aluminum coupons exposed in static autoclaves are found to corrode at quite low rates, even at high temperatures. (3) Apparently, the water becomes saturated with an aluminum oxide specie, further dissolution of the oxide ceases, and the corrosion rate follows the parabolic rate loss (the rate is inversely proportional to the amount of corrosion that has previously occurred).

The only practical method known for minimizing the rate at which Hanford process water dissolves the oxide layer is to regulate the pH. Aluminum oxide is amphoteric; that is, its hydrolysis reactions may be represented by:

Hence, the solubility of aluminum to oxide is high at both high and low pH, and pH control is required to minimize dissolution rates and corrosion rates.

effect of pH on corrosion rate
(2,4) The pH which yields minimum aluminum corrosion rates varies with the water temperature. (2,4) At current tube outlet temperatures, this pH of minimum corrosion is probably between 5.0 and 6.0. Since the startup of the Hanford plant, the pH of process water has been decreased from 7.6 to 7.0. This apparently reduced corrosion rates by about a factor of four. A further pH reduction to 6.6 has been made at the K Reactors, but the ~~benefits~~ *benefits* have not yet been determined. A further pH reduction below 6.6 probably would further reduce corrosion rates. However, incremental acid costs, effects of the lower pH on carbon steel piping corrosion, and effluent activity effects would have to be carefully weighed before a further pH change is seriously considered.

In some experiments, aluminum corrosion rates have been reduced by maintaining a high dissolved aluminum ion content in the coolant. Since in the alum water treatment process, the coolant is already saturated with aluminum ion at the reactor inlet, it does not appear possible to further reduce rates by a "presaturation" technique.

phosphate ion
The addition of phosphate ion to water also appears to have the effect of reducing corrosion rates. (2,5) The phosphate ion apparently enters into

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the oxide layer, increasing its diffusion resistance. Because of the need to limit the amount of P^{32} released to the river, this approach to corrosion control has never been seriously considered.

The alum feed rate was increased in mid-1961. The small changes of trace impurities resulting from the increased feed would not be expected to affect the corrosivity of the water. The change in feed may have reduced the amount of colloidal film deposited on fuel and tube surfaces. However, since this colloidal film is not considered to be protective in nature, such a change would not be expected to change corrosion rates.

Certainly no effect of the high alum feed is apparent on C-Reactor or K-Reactor process tubes which have been studied in detail. Further, an ex-reactor test when purge material was continuously added to process water, corrosion rates remain the same as previously determined in the absence of purging. This purging was probably at least as effective as high alum feed rates in removing colloidal films. (6)

Non-uniform corrosion of aluminum can be caused by either chemical or mechanical factors. The basic cause of non-uniform attack is a local disruption in the protective oxide layer allowing a rapid corrosion attack. For example, when aluminum is exposed to cold water containing a few ppm of chloride ion, it will pit rapidly. Aluminum exposed to cold, pure water is not attacked. Apparently, the chloride ion enters the aluminum oxide lattice, perhaps introducing additional vacancies, but in any case destroying the protective nature of the thin film. Corrosion rates at these pits are thousands of times more rapid than the attack at adjacent, non-pitted surfaces.

Erosion-corrosion is another example of non-uniform aluminum corrosion. In this instance, the oxide layer is mechanically damaged, no longer offering an effective diffusion barrier, and corrosion rates can increase catastrophically.

Two general approaches are available for preventing or at least minimizing this non-uniform attack: elimination of conditions or factors leading to non-uniform attack, and utilization of inhibitors.

Sodium dichromate has proved to be an extremely effective inhibitor for the prevention of non-uniform corrosion of aluminum in Hanford process water. The amount of dichromate added to process water was reduced from 2 to 1 ppm in a half-plant test at C Reactor. Later, the same reduction was made at B, D, DR, F, and H Reactors. Data obtained from the C-Reactor test do not show that the change has had any effect on the uniform or non-uniform corrosion of process tubes. There is tentative, non-conclusive evidence that the non-uniform corrosion of fuel cladding may have been increased slightly. The C-Reactor test will be continued until the effects on fuel and tube corrosion have been rigorously defined.

of reduced dichromate

Just not due to
Na₂Cr₂O₇ - probably
Aluminum

Now have data
that indicate high
alum cause corrosion
in non-uniform
corrosion

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If the dichromate inhibitor were not added to process water, reactor purges could cause erosion-corrosion damage to process tubes. Experiments have shown that water turbidity can cause a very rapid erosive attack, particularly at points of fuel misalignment. While studies have not been made to determine specifically safe purging limits with dichromate inhibited water, the past practice used in limiting purging to 300 ppm - hours maximum in any 30-day period, would appear safe. Available information on this subject has been recently summarized elsewhere⁽⁷⁾ and will not be repeated here.

2. Temperature

a. Water Temperature

In the absence of heat transfer, the aluminum tube assumes the temperature of the water in contact with it. Temperatures, of course, have a large effect on corrosion rates. Figure 1 is a plot of corrosion rates as a function of temperature. In this figure, rates are calculated from the equation:

$$CR = 0.0008 (2)^{t/12}$$

Where CR = Corrosion rate, mils/month

t = Temperature, °C

Most such plots of corrosion rates versus temperature are made on semi-log graph paper, producing a straight line and tending to conceal the exponential relationship of temperature and corrosion rates. Figure 1 shows the effects on corrosion rates of the steady increases in temperature made over the years.

b. Fuel-Tube Eccentricity

It is a well-known fact that eccentric placement of the fuel element in the process tube results in a temperature unbalance in the annulus. Laboratory data which demonstrate this effect are shown in Figure 2, a plot of temperature versus eccentricity.⁽⁸⁾ Eccentricity is defined as the displacement of the center of the fuel from the point of perfect centering divided by the average annulus thickness. The data were obtained by carefully positioning an electrically-heated rod simulating the fuel element at various distances from the side of a K process tube. Water temperatures were measured at small intervals around the annulus. The plot shows that the temperature at the restricted annulus doubles when the annulus thickness is decreased by 57 percent from a centered position, and increases by more than a factor of four with a 90 percent displacement. The temperature in the open annulus is shown to decrease as it becomes larger.

All fuel types used with new ribbed process tubes produce an annulus which is hydraulically restricted at the tube top. This results in the top-of-annulus (TOA) coolant temperature being elevated above tube bulk temperature, and TOA corrosion rates are higher than rates elsewhere on the tube circumference. Alternately, the bottom of tube or between-the-ribs (BTR) annulus is relatively unrestricted; the percent of flow in the BTR annulus is greater than the percent of heat transferred to this part of the annulus and BTR temperatures are below tube bulk temperature. BTR corrosion rates are therefore relatively low in new process tubes.

As the tube ages, the initial conditions described above will automatically change. TOA tube corrosion and rib attrition combine to enlarge the top of tube annulus. TOA coolant flow rates are then increased at the expense of BTR and hole flow rates. Hence, TOA temperatures and corrosion rates decrease with time. BTR temperatures and corrosion rates correspondingly increase with time. In time, the extent of internal tube corrosion, either BTR or TOA, will reach a degree where the probability of a leak occurring becomes unacceptably high. At this point, the usefulness of the tube is ended and the tube should be replaced.

hole
probably
not

The temperature variations around the annulus are often expressed mathematically as "R Values." Where R is defined as:

$$R_x = \frac{\Delta t_x}{\Delta t_b}$$

Where R = R Value, unitless

Δt = Temperature increase, any ^{temperature} units

Subscripts:

x = A particular point on the tube circumference

b = Bulk coolant condition at x

Figure 3 shows the effect of R Values between 0.8 and 1.4 on tube corrosion rates at a constant tube outlet temperature of 110°C and inlet temperature of 10°C. Since both BTR and TOA R Values may range between 0.8 and 1.4 during the life of a tube, Figure 3 tends to show the inadequacy of tube outlet temperature measurements alone in describing corrosion rates. To restate this point, Figure 3 shows that at an outlet temperature of 110°C, a particular point on the tube at the rear of the fuel charge may be corroding at any rate between 0.2 and 4.3 mils/month.

The effect of fuel element diameter on the TOA and BTR R Values is shown in Figure 4, a plot of R Value versus Corrosion Index. The

Corrosion Index is a number which is related to the age of the tube and its outlet water temperature history; and, as such, is an indication of the accumulated corrosion of the process tube. These data were obtained in C-Reactor process tubes which had an initial rib height of 0.102 inches. Each plotted point is the average of the number of measurements shown in parenthesis on the chart. The curves are remarkably similar to those obtained in the laboratory. The TOA temperatures decrease as the tubes corrode and the BTR temperatures increase. Increasing fuel element diameter increases TOA temperature and decreases BTR temperature. The initial effect on BTR temperatures is not significant. However, the benefit of a larger fuel element as the rib height decreases is evident.

The data shown in Figure 4 were obtained from temperature measurements made at the end-of-the-active-section of the process tube and thus represent the accumulated effects along the length of the fuel element charge. The relationship between the bulk, TOA, BTR, and hole coolant temperatures for new process tubes is shown in Figure 5. In this case, the TOA and BTR thickness is constant along the length of the active section of the tube. This plot was computed by assuming (1) an initial R_{TOA} of 1.25, (2) a chopped cosine power distribution, and (3) no water mixing. The R Value at a particular point on the circumference is the same at each fuel element position. TC

Figure 6 is a similar plot for a corroded tube which shows how these relationships change after tube wall corrosion and rib attrition have occurred. This plot was obtained by geometrically determining top and bottom annulus thicknesses from ex-reactor measurements of wall thickness and rib height of C-Reactor irradiated process tubes.⁽⁹⁾ A local R Value was obtained for each position by referring to Figure 1. A chopped cosine power distribution, flow redistribution but no mixing were assumed. The local R Value varies in this case because the accumulated corrosion and rib attrition increase with water temperature. The temperatures are not significantly different from bulk water temperature up to about the center of the charge, where they diverge as the top annulus opens up and the BTR annulus closes. The hole coolant temperature is higher than in new tubes because the enlarged annulus demands a larger fraction of the total flow.

c. Tube Surface Temperatures

About six percent of the heat transferred to the cooling water in a Hanford process tube is generated in the graphite and is therefore transferred through the tube wall. This will raise the tube wall temperature above local water temperature, depending on the local heat flux and heat transfer coefficient.

The temperature drop across the water film on the tube surface was computed for an old-reactor tube operating at 1200 kw. Calculations were made for two cases. The first assumed a uniform gap between the tube

and graphite, and the second assuming a non-uniform gap with the tube and graphite in contact on one side. The results of the calculations are shown in Figure 7. For the uniform gap assumption, the maximum film drop is 2°C. Assuming the tube and graphite are in contact, the maximum film drop is 5°C. At positions in the tube where the maximum corrosion occurs, the film drop is 2°C. This difference is not large but in the direction of increasing corrosion rates.

d. Rib Temperature

Analyses of out-of-reactor tube examination data indicate that the process tube ribs corrode at a faster rate than the tube wall. This result is obtained after making an allowance for charge-discharge wear. This is most probably due to higher rib surface temperatures which result from heat transfer from the fuel element. Calculated rib temperatures are shown in Figure 7, assuming a contact coefficient of 800 Btu/hr-ft²-°F. Rib temperatures are shown to be about 8°C greater than the local water temperature. This increase in temperature could result in 50 to 70 percent higher rib corrosion rates when compared to tube wall corrosion rates. The rib surface temperature would not have a significant effect during the early stages of tube life because of the low water temperatures between the ribs; however, it would have a significant effect when between-the-rib water temperatures have increased due to rib attrition and top-of-annulus corrosion.

Another possible factor leading to high rib corrosion rates is that a stagnant flow condition may exist in pockets between the ribs and fuel element where high water temperatures or even local boiling may exist. Such a phenomenon has been observed in laboratory experiments using glass tubes containing electrically-heated rods with supports similar to those used on bumper and self-supported fuel elements.⁽¹⁰⁾ In these tests, boiling was observed to take place beneath the supports at the following conditions:

<u>Heat Flux</u> <u>B/hr-ft²</u>	<u>Sub-</u> <u>Cooling</u> <u>°C</u>	<u>Bulk Water</u> <u>Temp. °C</u>	<u>Pressure</u> <u>psig</u>
266,000	65	69	30
180,000	46	69	10.5

While the experimental conditions may not be exactly applicable to reactor conditions, the significant point is that temperatures at the fuel element - rib interface may be considerably higher than those shown by Figure 7.

e. Variations in Heat Generation Rates

With eccentric placement of the fuel element in the process tube, the variation in the thickness of the water annulus would be expected to

result in a variation in heat generation rate in an overmoderated system. This effect would tend to increase corrosion rates as the annulus became thinner because the poison effect of the water would be diminished. The effect of a completely dry annulus on the heat generation rate has been computed by R. J. Shields. These calculations show that the heat generation rate in a dry tube would be 12 percent greater than the heat generation rate in surrounding wet tubes. Out-of-reactor tube examination data reveal that the annulus between-the-ribs is reduced to about 0.050 inch at the time the tubes are replaced. Based on a maximum change of 12 percent, it is believed conservative to estimate a maximum increase of 2-5 percent for low ribbed tubes. Again, the effect on local water, wall, and rib temperatures would be small but in the direction of increasing temperatures.

3. Radiation Effects

It is of some practical importance to know the effects, if any, of reactor radiation fluxes on aluminum process tube corrosion rates. If radiation fluxes greatly increased corrosion rates, there would be added incentives for controlling the front-to-rear flux distribution. Optimum tube powers would be somewhat reduced and mixer element placement would be affected. This section presents available information on the effect of radiation fluxes on aluminum tube corrosion rates.

Radiation fluxes could affect process tube corrosion rates by: changing the properties of the aluminum metal, changing the properties of the protective oxide layer, producing short-lived changes in the process water, and producing long-lived changes in the process water.

If one of the first three effects were important, the "change" in corrosion rates would be proportional to the local flux intensity or, possibly, the local flux integrated over some period of time.

If the last effect were important, the "change" in corrosion rates would be proportional to the integrated radiation dose to the water at the point in question.

If either of these two types of changes of corrosion rates were present to an important degree, it should be possible to detect their effects by a careful examination of process tube corrosion patterns. Such an examination ~~of the data~~ has been performed and no important effect has been detected. The basis for this statement is amplified in the following paragraphs.

a. Evidence that Corrosion Rates are not Proportional to Local Flux Intensities or Dosages

If the rate of corrosion were increased by the local flux intensity or local accumulated dosage, the relative amount of corrosion from the front to rear of the tube would vary from that explainable by temperature

changes alone. Indeed, such a shift in pattern appears to occur in old, severely corroded tubes where the maximum amount of penetration is found as far as six feet upstream of the rear of the fuel charge. In these instances, one is tempted to invoke a flux effect to explain why the maximum amount of corrosion does not occur at the point of the tube coincident with the rear of the fuel train. While a convincing argument complete with proofs cannot be advanced to explain why the point of maximum corrosion is sometimes found to have shifted upstream, it probably is not the reflection of a flux effect.

Measurements obtained from process tubes removed from C Reactor are shown in Figures 17a through 17f in Appendix A. A study of the profiles obtained from tubes removed during late 1955 and throughout 1956 which had experienced little corrosion fails to show the effects of a radiation flux. While some scatter in the data is apparent, it is plain that the maximum amount of corrosion occurred at the rear end of the fuel charge. This indicates that corrosion rates do not vary with the local flux intensity; otherwise, the point of maximum corrosion would be shifted upstream. Also, the amount of BTR corrosion in the perf zone is about equal to that just upstream of the rear of the fuel charge. If there were a pronounced flux effect, one would ^{have} expected a "recovery" in the wall thickness downstream of the fuel-perf train junction. ~~Such~~ Such a recovery in the TOA wall thickness is expected because of reduction of TOA water temperatures and is observed.

The recovery in wall thickness upstream of the rear of the charge follows the pattern one would expect if corrosion rates were controlled by temperature alone. This latter fact is illustrated in Figures 20a through 20i in Appendix A. In these figures, the measured amount of corrosion in relatively new tubes is plotted against the "integrated cosine value." The integrated cosine value is the accumulated fraction of the total tube power from the inlet to the point in the tube where the measurement was made. The amounts of corrosion were obtained by subtracting observed wall thicknesses from those measured in the front, cold part of the tube.

These plots are equivalent to plots of corrosion penetration plotted against temperature. The data produce straight lines when plotted on semi-log paper, as would be expected if water temperature alone was the major factor affecting corrosion rates. Since the data from these tubes do not show the existence of a flux effect and since flux intensities and accumulated dosages were appreciable by 1956, it is concluded that shifts in the point of maximum corrosion upstream of the end of the fuel charge in other tubes must reflect the interplay of other factors.

b. Evidence that Corrosion Rates Are Not Proportional to the Integrated Flux Dosage to the Water

If corrosion rates were proportional to the integrated dose to the water (implying that the radiation flux produced a relatively long-lived

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change in the water chemistry), the effect would be expected to persist into the perf zone. The sections of C-Reactor tubes in the perf zone have corroded only about 10 mils throughout their entire 10-year life. Hence, long-lived water chemistry changes induced by the radiation fluxes apparently have not been important factors in accelerating corrosion rates.

Actually, the amount of corrosion measured in the perf zone is in good agreement with that predicted from the equation developed from laboratory testing. This has been previously reported⁽¹¹⁾ from measurements made in old-reactor tubes, and is illustrated again in Figure 21 in Appendix A, with data from C Reactor.

c. General Comments on Effects of Reactor Fluxes on Aluminum Corrosion

While from the foregoing it is concluded that reactor radiation fluxes do not have an important effect on process tube corrosion, it is of interest to discuss other data obtained from tests particularly designed to study such irradiation effects. Both Goldsmith and Richman have made such studies. In both cases, they performed studies in which process water was preheated (to 80°C by Goldsmith and to 90°C and 110°C by Richman) and then circulated past a number of aluminum samples. The aluminum samples were positioned in the reactor core so that they were exposed to flux intensities from virtually zero to near the maximum obtainable. Significant flux effects were reported in both cases.^(12,13) At a given temperature, corrosion rates of samples exposed to the maximum flux intensity were five or six times those observed at the zero flux point. In both cases, it appeared that the major effect was the local flux intensity or dosage, although Richman's test also indicates that a long-lived radiation effect of the water may also have increased the corrosion rate slightly. It is not known why a discrepancy exists between data obtained from the test and the data obtained from tube measurements. However, two potentially pertinent factors are known.

1) Exposure Time ^{data}

Tests by both Richman and Goldsmith were relatively short-term - of the order of three or four months. The data from process tubes reflect exposures of several years. It is possible that radiation could affect rates during the early period of exposure before corrosion films were well established and not affect rates later in the process. If this should be the case, initial radiation effects would soon be masked out.

2) Surface Preparation

Both Richman and Goldsmith used aluminum samples which had been pre-autoclaved to simulate fuel cladding surfaces. Such treatment reduces initial corrosion rates. The maximum rate reported by Richman for the autoclaved samples both at 90°C and 110°C - about 0.4 mil/month - is

about what is observed at 110°C on non-autoclaved tube surfaces in the perf zone. Hence, the radiation effect noted might merely be the destruction of the superior corrosion resistance of the autoclave film.

exposure + time of surface prep -
It appears that there is some theoretical support to concluding that the two preceding factors are important. This can be seen by discussing each possible effect of irradiation on aluminum corrosion.

Effect of Flux on the Metal

in explaining the description in
It is expected that effects radiation has on the metal itself would affect corrosion rates, if at all, only while the protective oxide layer was quite thin. With an oxide layer of steady-state thickness, the limit to the corrosion rate is thought to be diffusion of an oxidizing agent through the oxide, not the rate of reaction of an oxidizing agent with metal. Hence, if radiation damage to aluminum affected the rate with which it reacts with an oxidizing agent, one would expect to observe the effect on corrosion rates only during the early stages of the process.

Effects on the Oxide Film

It has earlier been postulated that the rate-limiting step is diffusion through the oxide layer. With the thick oxide layers obtained in process tubes, the rates observed at the water temperature attained cannot be explained by lattice diffusion.⁽¹⁴⁾ At these low temperatures, lattice diffusion rates are quite low. Hence, the process must be diffusion along very small cracks in the oxide. The formation and characteristics of the cracks would be expected to be determined by the gross chemical, physical, and metallurgical characteristics of the film, i.e., the film strength, thickness, and composition. None of these factors is likely to be affected by radiation fluxes.

~~On the other hand,~~ ^P lattice diffusion may be important in the early corrosion of autoclaved aluminum samples. Radiation damage to the autoclave film - at least during the early stages of the process - could affect the atomic structure of the film and hence, corrosion rates.

Further testing is obviously required if all phenomena observed are to be fully understood.

4. Coolant Flow Effects

a. Velocity

Tube corrosion rates should be coolant velocity dependent, to some degree. Increased coolant velocity may mechanically erode away the protective oxide layer. Even if this effect were not present, increases

in velocity would increase the potential for diffusion of aluminum ion from the oxide layer to the bulk coolant stream. In part, this diffusion rate increases with velocity in much the same manner as do heat transfer coefficients. While this effect probably is present, it is too small to be measured.

Insight on the effect of velocity on aluminum corrosion rates in treated Columbia River water has been obtained from both laboratory studies and examination of reactor process tubes. In 1952, (15) 1100 aluminum alloy corrosion rates at 95°C were measured at water velocities of 4.7, 10.9, 13.7 and 18.3 feet/second. No velocity effect was observed. These tests were conducted with pH 7.6 water, instead of the now standard pH 7.0 water. However, the more corrosive pH 7.6 water would be expected to exhibit as large a velocity effect as the lower pH water.

The coolant velocity in a process tube drops at the juncture between the fuel train and perf column. For example, the velocity in a BDF-type tube loaded with OIIN elements drops from about 37 feet/second to 10 feet/second when the tube flow is 45 gpm. In cases where an abrupt temperature change does not also occur at the juncture between fuel elements and perfs, the amount of corrosion is not measurably different in the two velocity regions. This lack of effect can be seen in the C-Reactor tube profiles taken of tubes removed before severe corrosion had occurred.

b. Bulk Boiling

Localized bulk boiling in a process tube can result from fuel cocking, column bowing, or extreme annulus eccentricity. The presence of bulk boiling implies a high temperature which in itself leads to high rates. However, boiling in itself apparently can cause a catastrophic further increase in corrosion rates.

Tube leaks have been found to occur in a number of new process tubes - tubes in service less than a year. These tubes leaked at the top, and the tube area associated with the leak appeared severely pitted. In most cases, the severe corrosion was found to initiate at some point a few feet upstream from the rear of the fuel charge. The width of the severely corroded area generally increases from the upstream point to the rear of the charge. At the juncture between the fuel column and the perf train, the severe corrosion disappears.

Radiometallurgical examination of sections taken from a typical leaker of this nature, Tube 2481-DR, showed that the metal in the vicinity of the severe corrosion was mechanically worked to a depth of 0.5 mils. (16) The very high corrosion rates observed, probably reflect the extremely erosive conditions brought about by boiling.

Sections

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To date, only TOA tube leaks have been attributed to be the result of boiling. It is not improbable, however, that bulk boiling also occurs between the tube ribs when tube corrosion and rib loss continue to an advanced state. However, since the ribs provide positive positioning of the fuel, BTR damage resulting from boiling can occur only after the tubes have been severely corroded. TOA boiling resulting from fuel misalignment may occur, and has occurred, in new tubes.

The desirability of avoiding conditions which lead to bulk boiling has been recognized for many years. Indeed, many changes in the process - development of self-supported fuel, bumper fuel, tru-line fuel elements, mixer elements, and the redesign of fuel and process tubes - have been made to minimize local, high temperatures in the tube.

5. Mechanical Effects

Evaluation of the important factors which directly influence internal corrosion (fuel and tube geometry, temperature, and water quality) of necessity assumes that the process tube dimensions as specified by the purchase specifications remain unchanged except as influenced by these same factors. Specifically, it is assumed that the tube remains round, and that the wall thickness is not impaired by such things as handling prior to installation; scratches, galls, or other damage from installation; external corrosion, or ruptured fuel removal efforts.

Recent tube installation experience at B and DR Reactors serves to illustrate problems of a mechanical nature, which of themselves are troublesome, but which can seriously impair the expected service life of the tubes. At DR Reactor, galling and scratching of the tube caused considerable difficulty in the recent tube outage. It is not known, but it can be speculated that some number of tubes which were installed with reluctance have scratches and galls of such a depth as to have only 30-35 mils of remaining wall.⁽¹⁷⁾ If this is true, these tubes may leak prematurely and unpredictably. A similar problem would exist if, as a result of installation, the bottom or top of the tube is flattened, thus reducing the annulus. Under these conditions, higher water temperatures would occur than were predicted. Thus, accelerated internal corrosion would occur, possibly resulting in premature failure. Dimensional probing at B Reactor has shown the potential of this problem.

External corrosion, a well known phenomenon at F Reactor, can similarly reduce the available wall for internal corrosion. Those factors, such as pinhole leakers or Van Stone leaks, that lead to external corrosion can thus have an important bearing on tube life predictions.

The deterioration of the process tube ribs results from a combination of mechanical and chemical effects. Rib profiles for a series of D-Reactor tubes are shown in Figure 8. It is evident that rather little reduction of rib height occurs upstream of the center line of the fuel

charge. For central zone tubes, rib attrition caused by mechanical wear amounts to about one mil/year.

The rib loss near the rear of the charge is quite high, amounting to two or three times the corrosion measured on the adjacent tube walls.

One factor causing the rapid rate of rib loss is the high rib temperatures resulting from contact with the fuel element. However, this is not the sole cause since high rib losses are also seen in the perf zone.

Another factor causing the rapid loss of rib height is the periodic removal of the protective aluminum oxide (and a very small amount of aluminum metal) by the charge-discharge operation. The bare aluminum surface then corrodes rapidly until the oxide film is re-established. This cycle is then repeated every time the fuel is discharged. This "extra" aluminum corrosion resulting from exposing the aluminum surfaces may amount to as much as two mils/cycle.

6. Aluminum Alloys

Presently, aluminum tubes are fabricated from 1100 aluminum alloy (formerly designated 2S aluminum). This alloy is termed commercially pure aluminum, containing a minimum of 99 percent aluminum.

Investigation of other aluminum alloys for tubing materials has been performed in the past. Both powder metallurgy alloys and other wrought alloys have been considered. (18,19)

Of the alloys studied, 6063 alloy aluminum (formerly designated 63S) was specifically noted as an acceptable substitute for the 1100 alloy from the standpoint of uniform corrosion. Actually, any of the common wrought alloys acceptable from a nuclear standpoint would probably be satisfactory from a uniform corrosion standpoint; however, only 6063 alloy tubes were ever installed in a reactor. Of about twenty tubes installed, two of the 6063 alloy tubes leaked in service. One of the tubes leaked as a result of TOA corrosion; the second, installed in O2 row in H Reactor, leaked due to external corrosion.

While neither of these leaks should be considered strong evidence of inferiority of the alloy, the experience does not encourage the use of the 6063 alloy. Certainly, any incentive for adoption of the 6063 alloy must come from factors other than corrosion resistance. Should other strong incentives not arise, adoption of 6063 alloy as a tubing material is not recommended.

Within the last eight years, a new series of wrought alloys has been developed which has a superior resistance to corrosion at high temperatures. The present fuel cladding, Alloy X-8001, is one of these.

Process tubes made from these alloys have not been developed for two reasons:

- a. Because of the much lower heat fluxes, process tubes operate under much milder conditions than does fuel cladding. Hence, the superior high temperature corrosion resistance afforded by the new type of alloys is not required.

Under milder conditions, the "superior" alloys corrode at the same rate as does the 1100 alloy.

- b. There is evidence that these new alloys are more susceptible to non-uniform corrosion than is the 1100 alloy. (20)

However, if aluminum process tubes are to be used indefinitely, the testing of tubes made from the new type alloys should be considered.

7. Application of Technical Factors Affecting Fuel and Tube Design

From the preceding, it is concluded that there are three basic factors controlling tube corrosion rates and tube life as limited by internal corrosion.

- a. Water temperature.
- b. Tendency to non-uniform attack.
- c. Water pH.

Of these, water pH is independent of the fuel element and tube design. Of the factors producing non-uniform attack, local bulk boiling can generally be prevented by the use of bumper fuel elements which prevents fuel element misalignment. Assuming that non-uniform corrosion can be generally prevented, at a given water pH tube life becomes primarily a function of temperature. At a given bulk coolant inlet and outlet temperature, the temperature at a particular point in the process tube depends on the relationship between the fuel element and its process tube. This relationship is constantly changing because of tube wall corrosion and rib attrition. Maximum service life for given operating conditions is obtained from a ribbed process tube by taking full advantage of the wall thickness at both the top and bottom of the process tube. It has been clearly demonstrated that BTR temperatures and corrosion rates increase with time while TOA temperatures and corrosion rates decrease. Thus, the higher the initial rib height, the longer the delay in reaching these high terminal corrosion rates. However, the higher the initial rib height, the higher initial TOA corrosion rates. For a given fuel element, choice of the initial rib height for the process tube is, therefore, all important, for initial corrosion rates must not be so high as to cause premature failure through the top. Because of the wide variation in process conditions, it is impossible to arrive at dimensions which will assure that the top and the bottom will corrode an equal amount during the life of the tube. A compromise must be made in initial TOA corrosion rates to assure that an early failure will not

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occur. In practice, advantage is taken of water-mixing elements to control initial TOA corrosion rates. Their application is discussed in a later section of this report.

B. Past Experience and Monitoring

1. Past Experience with Process Tubes

The discussion of technical factors above is useful in giving insight into the past behavior of process tubes.

The original tubes in the central zones of the five oldest reactors were replaced starting in 1955 after up to 11 or 12 years of service. Coolant temperatures during the early years of operation were quite low - particularly at B, D, and F Reactors. Hence, even though the corrosivity of the cooling water then employed was roughly quadruple that of present process water because of the higher pH, corrosion rates were low. For example, the maximum tube outlet temperature achieved in D Reactor in 1951 was about 85°C, and these "high" temperatures were maintained only during the late summer when inlet temperatures were also high. Hence, during the years when the ribs in the tubes were high (R Values were about 1.25), temperatures were low and corrosion rates were moderate. By the time relatively high temperature operation was achieved, the ribs were worn and maximum temperatures occurred between the ribs. The original tubes in all five old reactors were therefore removed because of BTR corrosion. So far as is known, no tube leaks occurred in the original tubes as a result of TOA tube corrosion.

The second-generation tubes were exposed to an entirely different environment. By the last quarter of 1956 (when central zone retubing was largely completed), maximum tube outlet temperatures were over 100°C. High temperature operation continued, except for a short period following the completion of the water plant expansion project (CGI-558), when temperatures were reduced by increased flow rates. As reactor powers were again increased, maximum tube exit temperatures climbed as high as 115 to 120°C. Hence, when ribs were unworn and TOA R Values were high, tube exit temperatures were likewise high, TOA corrosion was therefore rapid. Concern was expressed in mid-1959⁽¹¹⁾ that the second-generation tubes would require replacement at an early date because of the TOA corrosion. While some tube leaks occurred because of TOA corrosion, the natural process of rib wear and corrosion plus the adoption of water-mixing elements apparently served to reduce TOA corrosion rates.^(11,21) Most of these second-generation tubes in the older reactors will be removed because of BTR corrosion.

C-Reactor tubes present a different history. Again, when the tubes were new, tube outlet temperatures were moderate (maximum tube outlet temperatures did not exceed 95°C during 1953). TOA corrosion rates were measurably greater than BTR rates, but the extent of TOA corrosion was not appreciable. By the time the tube ribs were becoming worn, the I&E fuel

element was adopted. The earliest I&E fuel elements irradiated had an OD of 1.474 inches, locating them eccentrically to the top of the tube, which produced elevated TOA temperatures. However, use of mixer elements, restricting outlet temperatures, and eventual reduction of fuel size to 1.460 inches prevented serious TOA corrosion.

In early 1960, it was concluded that BTR R Values were exceeding unity, even with the I&E fuel element, and that acceleration of BTR corrosion rates could be expected.⁽²²⁾ However, the use of mixer elements and displacement of fuel charge about two feet forward had the effect of minimizing corrosion rates and shifting the site of maximum corrosion rates to a thicker part of the tubes. In early 1960, few C-Reactor tubes were reported to have wall thickness of less than 41 mils.⁽²²⁾ Now it appears that relatively few tubes have wall thickness of less than 36 mils. Hence, in approximately two years of operation, the minimum wall thickness has decreased probably no more than 5 mils, although the thicker parts of the tubes have corroded more than this amount.

During the past two years, ribs have been further worn, BTR R Values have increased, and the tendency towards higher rates of attack has increased. Countermeasures have been recommended,⁽²³⁾ and significant additional tube life may be attainable. A more quantitative assessment of C-Reactor tubes may be found as Appendix A to this report.

The K-Reactor tubes are on the average thicker than C-Reactor tubes, and rib attrition has been less severe. Nevertheless, BTR R Values are greater than unity, and ever-increasing corrosion rates can be anticipated unless aggressive countermeasures are continued. The situation at the K Reactors is treated in detail as Appendix B.

2. Monitoring Efforts

Techniques for ~~monitoring~~ ^{measuring} process tube conditions have evolved over the past six or seven years. Prior to 1955, very little meaningful process tube monitoring was done. Initial ~~monitoring~~ efforts depended solely on the ex-reactor examination of process tubes, which consisted of visual examination of the surfaces and a laborious measuring of the residual thickness of the 72S cladding layer (original tubes were all clad with the 72S alloy). This technique of ex-reactor examination did not even produce enough meaningful data to give much insight into the status of the condition of tubes in the reactor, let alone their corrosion rates.

The development of the probolog was expedited during this period, and by the time tube replacement had become full scale at B and H Reactors, it had been perfected to the point that it was of major value in guiding tube replacement outages.

Meanwhile, the techniques of performing ex-reactor examinations were improved. More meaningful measurements were made as added experience

showed what measurements were important, and what measurements were of little value. Equipment and facilities were also developed which, with the better techniques, permitted the measurements to be made more rapidly. The ex-reactor examinations became primarily an adjunct to the probolog, indicating the meaning of the probolog traces.

Thermocouple probes were developed to measure the variation of the coolant temperature around the cooling annulus of the process tube. The first such probes produced data of poor quality for reasons discussed below. However, under the impetus of the conversion of the reactors to I&E fuel elements in late 1957, better techniques were developed for the fabrication and installation of the probes. This improvement of technique is continuing.

measurement
induct
At the present time, the three tube ~~monitoring~~ techniques are in use. The probolog instrument is used to gather enough data to ~~give a picture~~ of the general status of a large number of process tubes. Out-of-reactor tube examinations are performed to provide calibration for the probolog, to show better the pattern of corrosion, to measure rib height, to compare TOA and BTR corrosion, and to define the manner in which the TOA and BTR corrosion varies along the length of the tube. The thermocouple probes are used to measure the temperature variation around the coolant annulus and, hence, to show probable future trends of corrosion rates. The adequacy of this effort is discussed below.

3. Adequacy of the Monitoring Efforts

The two positive end results of the tube monitoring efforts are: (a) providing technical information to form a basis for positive action to extend tube life, and (b) providing information to form a basis for planning tube removals.

The adequacy and success of the monitoring efforts have varied from reactor to reactor and have varied with time when measured against the two end results.

removal of
It is pertinent to note that tube monitoring is not free. Removal of an isolated process tube for monitoring may require about four hours of outage time; probologging 200 process tubes requires about 10 hours of outage time; and the installation and the later removal of four thermocouple probes may require about an hour of outage time. Hence, at some point, it is probably better to accept less than maximum tube lives and less than perfect ability to schedule exactly upcoming tube outages rather than to expend more and more time on tube monitoring. Intuitively, it would appear that this point has never been reached. This question has never been rigorously studied. 20-0

a. Second-Generation Tubes - Old Reactors

Very little monitoring of second-generation process tubes was accomplished before 1959. During this period, the major attention at the old reactors

was focused upon original tubes which were still being replaced. The monitoring of second-generation tubes was accelerated after the TOA corrosion problem was identified and concern was expressed for the possibility that the service life of these second-generation tubes might soon be ended.

The natural processes of rib wear and tube corrosion plus mixer element usage apparently controlled TOA corrosion rates. Hence, while the monitoring was quite meager during the early service of the second-generation tubes, by-and-large, the results were not serious. However, such inattention to a large block of process tubes over an appreciable length of time seems unwise.

Probologging of the second-generation tubes continued; but the shift to high BTR corrosion rates could not be identified by the probolog. Neither were tube removals nor thermocouple probing efforts adequate to show the effect. Hence, the monitoring was not adequate on either of the two counts - prolonging the tube lives or providing a basis for scheduling tube removals. In early 1961, it was concluded from the results of an internal study performed to guide Research and Engineering Section planning that replacement of second-generation tubes would not be required at any reactor before April, 1962, at the very earliest, and would not begin before September, 1962, at most reactors. It is obvious that this conclusion was in error by about six months at a minimum. Hence, tube monitoring efforts were inadequate, providing neither a warning that action was needed to protect against BTR corrosion nor warning that major tube removal outages would be required at least six months earlier than expected. This difficulty was due in part to the wide range of tube conditions in the reactor. For example, the central zone of D Reactor contained tubes installed each year from 1955 through 1961. Each group of tubes was somewhat different from all others, and a completely satisfactory monitoring effort would have been difficult. In retrospect, the extra effort would certainly have been worthwhile.

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b. C Reactor

The quality of monitoring at C Reactor has by and large been satisfactory. Monitoring was not started in earnest before November, 1955; however, before then, efficient techniques had not been developed. C Reactor was the lead reactor in conversion to I&E fuel. This sparked a rather large-scale program of thermocouple probing and some probologging. This effort continued as the fuel elements were redesigned and as mixers were used to minimize first, TOA and, later, BTR corrosion.

It is concluded that loss of "corrosion control" at C Reactor occurred on only two occasions. The shift of maximum corrosion rates to the BTR position was noted in early 1959.⁽²⁴⁾ Had more rigorous monitoring been in practice, the shift might have been noted a few months earlier. The actual damage from this lapse was not significant.

The second loss of control occurred in September, 1961, when the charge was shifted to the rear, and a single mixer was placed at the fourteenth fuel position from the rear of the charge. The use of two mixers per column had been recommended when the shift in charge was made.⁽²⁵⁾ Moreover, the fourteenth fuel position was not the optimum location for a single mixer and may have resulted in the unnecessary loss of one or two mils of process tube wall.

Ample bases for scheduling tube removals at C Reactor have been provided. In early 1960, it was predicted that large-scale replacement at C Reactor would not be required before late 1961, although a major probologging effort prior to that time would be required to locate and remove a few exceptionally thin tubes. This prediction was modified in early 1961 by the results of the internal study⁽²²⁾ noted earlier, at which time it was concluded that major C-Reactor retubing would not be required before September, 1962. This later conclusion is still considered valid.

c. K Reactors

The tube monitoring effort at the K Reactors during the first six years of operation is considered to have been inadequate. Fortunately, this lack of effort did not result in an apparent significant reduction in tube life or an unheralded tube replacement program. In early 1961, when the shift of maximum corrosion rates was first noted, there was concern in some quarters that major retubing might be imminent. This led to the request for a monitoring program.⁽²⁶⁾ However, in the internal study done in January, 1961, it was concluded that major replacement at the K Reactors would not be required before April, 1963. In April, 1961, after concern had been expressed over the remaining life, it was predicted that major tube replacement would not be required until late 1963 or 1964, depending on adoption of corrective action, or until January, 1963, if no corrective actions were taken.⁽²⁷⁾ These estimates are still regarded as sound, and with a continued monitoring program, these estimates can be modified as required to give sufficient warning of impending tube removal requirements.

C. Technical Evaluation of Monitoring Tools

The monitoring of the condition of aluminum process tubes has been accomplished primarily by use of the probolog supplemented by out-of-reactor tube examination. The probolog is an eddy current device which can be used to measure the remaining wall thickness of the process tubes while in-reactor. The ex-reactor examination consists of obtaining small (~one inch) samples from various locations along a tube which has been removed from the reactor. The samples are either weighed or measured by micrometer calipers to determine actual wall thickness. Further, the rib height is obtained by direct measurement on similar samples. More recently, the use of thermocouple probes has been initiated to determine the temperature distribution of the coolant water around the annulus of the fuel elements and also in the hole of the I&E fuel elements.

Since the probolog and ex-reactor examination data are the basic means of establishing in-reactor corrosion rates and mechanisms and provide the basis for tube removal, it appears prudent to again examine the quality of these data.

The probolog is a device which uses eddy current technique to measure the residual metal volume in the process tube. The signal obtained from the probolog head is fed into appropriate circuitry to produce a trace on a recorder. Basically, the probolog head "sees" a circumferential ring (of the process tube) about 3/4-inch in length. The signal obtained is proportional to the volume of metal in this ring. In the initial development work, conducted by T. L. Deobald, (28) a number of tests were performed to determine what parameters would affect this signal and how these factors might be compensated for so that minimum wall thickness of process tubes might be routinely obtained. The following discussion is largely based on a review of this work.

To obtain wall thickness from the recorder trace, it is necessary to calibrate pen deflection (variation of the trace) against minimum wall thickness. A section of process tube was machined to various wall thickness by concentrically machining the OD. This section of tube, when probed, provided the basic calibration for future tests. A number of runs were made on flow lab process tubes to determine the reproducibility and stability of the instrument. These runs indicated the reproducibility and stability were excellent and that variations from trace to trace of the same tube were less than one mil. Additional detailed information is given in reference (29).

Following this basic testing, test runs were performed to evaluate: (1) the effect of the graphite tube block around the process tube; (2) the effect of water in the tube itself; (3) the effect of variation in temperature of the tube; (4) the effect of eccentric placement of the probe relative to ID surface; (5) effect of varying the speed of probe translation; (6) effect of water corrosion film buildup on ID surface; (7) effect of irregular surface as would be expected from actual corrosion. The following results were obtained:

1. The presence or absence of a graphite block around a process tube will alter the pen deflection equivalent to about one mil. Thus, in those process channels in which graphite tube blocks are separated by 3 to 4 inches, minimum wall thickness as indicated by the probolog trace can be in error by one mil. The indicated wall thickness is less than actual wall thickness.
2. The presence or absence of water has no significant effect on indicated wall thickness provided the temperature remains constant.
3. The probolog is quite sensitive to temperature variations. Increases in temperature give larger pen deflections for any given wall thickness. This can result in an error of 3 mils/°C in indicated wall thickness.

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To compensate for this sensitivity, a water flow is maintained in the process tube being probed, thus essentially eliminating any temperature variation over the length of the tube. In practice, the probolog trace is calibrated against wall thickness measurements obtained on samples taken from a process tube which was removed from the reactor immediately following the probologging. If it is assumed that the magnitude of the difference in deflection for changes in wall thickness is the same at 25°C as it is for 5°C, then summer-to-winter variations in temperature of the water flowing through the tube during probologging will not cause any significant error in the wall thicknesses indicated by the probolog.

4. No detectable difference in wall thickness was observed when the trace was obtained in the standard manner, and when the sample section of tube was turned "upside down" allowing the probe to rest directly on the wall rather than on the ribs. Thus, the eccentricity effect is negligible.
5. The indicated wall thickness is not affected by varying the speed of traverse of the probe through the tube, provided a speed of 24 feet/minute is not exceeded.
6. Runs were performed on tube sections with and without water film buildup. No effect was observed. Thus, variations in process water quality which lead to variable film buildup have no effect on probolog results.
7. The effect of pitted surface or scratches is to produce "hash" on the probolog trace. However, the character of the hash can sometimes be interpreted, e.g., external corrosion produces a characteristic hash. If the pitted area is sufficiently deep, the probolog will indicate a wall thickness equal to the weighted average of the pit depths and peaks.

From experience in probologging process tubes over the past 5-6 years, it has become evident that as the ratio of the maximum and minimum wall thickness to the average wall thickness varies, the indicated minimum may be in error. Laboratory tests on this effect were attempted but were not completed because a suitable sample could not be prepared. Reactor experience would indicate that this error is not more than 2-3 mils, except that in isolated cases this error might be 7-8 mils. (30)

In practice, the following procedure is used in probologging.

1. After setup on the front face, the response of the instrument is checked by inserting the probe in a standard machined tube sample and verifying laboratory calibration.
2. The probe is inserted in the process tube and pushed to the rear face. Water flow is maintained while the trace is obtained.
3. The probe is pulled forward to the front face at a rate of 12 feet/minute with a chart speed synchronized to give one inch of chart to one foot of tube. The traverse speed is doubled after the probe has passed the center line of the reactor.

4. A calibration chart which has been previously prepared giving wall thickness versus pen deflection is superimposed on the trace, and the minimum wall thickness is thus obtained. The base point is assumed to be 60 mils (original minimum wall thickness) and is taken one foot downstream of the end of the front gunbarrel.
5. To calibrate the probe, the process tube is removed following the obtaining of such a trace. Samples are taken at one-foot intervals from the rear half of the tube. Each sample is of standard area. The sample is carefully cleaned and weighed. Using the density of aluminum, weight and area of the sample, the average wall thickness is calculated. A calibration chart is then prepared using the indicated deflection and calculated wall thickness for a given sample.

Recently, a statistical analysis⁽³¹⁾ was made of probolog results obtained by three different technicians. It is apparent from this study that variations in reported results due to judgment, skill, and repeated tests are negligible, the principal error being the variation in the probolog results versus the results as measured from ex-reactor examination.

The over-all error, based on laboratory tests and several years of reactor experience, is ± 2 mils.

The thermocouple probe is an assembly designed to measure the water temperature around the annulus and in the hole of a column of I&E fuel elements. If one visualizes a column of I&E fuel elements, neatly aligned, and resting on the ribs of a brand new tube, as shown schematically in Figure 9a, potential for variations in coolant flow and thus coolant temperature in the annulus and in the hold are self-evident. As the top wall of the tube corrodes, the "between-the-rib" wall corrodes, and the ribs wear and corrode, cf Figure 9b, it is easy to see how these flow and temperature distributions can change with time and reactor operating conditions. The thermocouple probe is designed to monitor this temperature distribution with points of measurement as shown by numbers in Figure 9a. The probe head is shown in Figure 10. The probe assembly is shown in Figure 11. This assembly is installed in the rear of a process tube in lieu of a standard dummy pattern. A special nozzle-cap arrangement is used to bring the stainless steel sheathed thermocouples out of the reactor. The thermocouple leads are connected to a junction box located out of the tube pattern on the rear face of a reactor. The junction box is essentially a permanent installation with lead wires leading to the read-out instrument, which is accessible during reactor operation. The thermocouples are copper-constantan appropriately insulated and covered by a stainless steel sheath. The thermocouple beads are made in a standard manner. The thermocouple wires are soft soldered to extension leads which are in turn connected to a 19-prong amphenol connector and hence to the junction box. The soft solder joint is sealed with epoxy resin, as is the connector. Plastic tubing covers the extension wires. The junction box is similarly sealed such that the entire extension system is protected from water to prevent corrosion problems.

On a routine periodic basis, the probe temperatures are monitored by recording on a log sheet the temperatures indicated by the read-out instrument (in most cases a Wheelco 128-point indicating potentiometer).

Initial efforts to obtain temperature distribution data were plagued with faulty readings from poor thermocouples, corroded junctions, lack of positive orientation of the probe itself, and poor resolution in the temperature indicating instruments. Present procedure of fabrication has largely eliminated the corrosion problems at the junction box and lead extension joints. The use of bumpers on the probe head and the coupling of the first and second dummy pieces behind the probe head have largely eliminated the difficulties in obtaining positive alignment during installation. After probe assembly, thermocouples are given a heat response check. A calibration check of the thermocouple is not made. Normally, copper-constantan thermocouples are considered accurate to about $\pm 1^\circ\text{C}$, in the temperature region of interest. Further, the read-out system, including the leads to the junction box, has not been calibrated. The Wheelco instrument currently installed has a full scale range of $0\text{-}200^\circ\text{C}$. This amounts to about $25^\circ/\text{inch}$ of scale, or $1^\circ/\text{mm}$. Such poor scale resolution has in the past resulted in errors of recorded temperature of as much as 5°C . In the past, these combined errors could have amounted to as much as 5 to 8°C ; however, the nominal probably did not exceed 3°C . In analyzing fuel and tube design, and in establishing corrosion rates as function of tube outlet temperatures, an error of 5°C in probe data is significant. Considerable effort should be made to limit errors in probe data to the inherent accuracy of the thermocouple. Thus, the indicating instrument should be readily capable of being read to 1°C . The range of temperature which is of significance from a probe standpoint is from 70°C minimum to 160°C maximum.

D. Over-All Integration of Technical Factors

The over-all integration of the various factors affecting process tube corrosion hinges heavily on a sound and adequate monitoring program. The evaluation of such things as water-mixing elements, fuel-tube geometry, and water quality becomes exceedingly nebulous without adequate monitoring data. Further, unless the data from such a monitoring program are obtained and analyzed, the past fluctuations of concern over tube life and tube replacement will continue. While outside the scope of this document, it is evident from the above discussion that tube life from an operational point of view may not be determined solely by internal tube corrosion. The operational tube life may well be determined by such things as capability of replacement, product worth, and river contamination. To obtain the optimum tube life thus requires the integration of these factors along with those factors affecting internal corrosion. ~~These points are discussed in more detail in the following sections.~~

1. Tube Monitoring Program

The tube monitoring in the past has been plagued with a feast or famine approach, with the intensity of data collection directly proportional to

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the proximity of a major tube replacement. From the above discussion of the complexity of the corrosion problem, it is evident that such fluctuations in data collection are not conducive to the full and complete understanding of the problem. Basically, the tube monitoring program must have two primary objectives, namely:

- a. To enhance knowledge and understanding of:
 - 1) Tube corrosion with regard to mechanism, rate, and the influence of various technical factors.
 - 2) Methods of process improvement with regard to production or cost.
- b. To provide a basis for the planning, scheduling, and replacement of tubes with due regard for production, cost, and replacement capability.

To formulate a monitoring program which would attain the above objectives and which would be uniformly applicable to the old reactors is quite difficult. This arises largely because each reactor has experienced different conditions, e.g., different generation tubes - first, second, and even third; external corrosion at F Reactor; top-of-annulus corrosion at DR; installation difficulties at DR; cracked tube blocks at DR and H, among others. However, at C and K Reactors, the first major tube replacement is yet to come. Operation at these reactors has been relatively free of the difficulties experienced at the older reactors. If one assumes that the retubing of C Reactor will be accomplished essentially as a block, the following program is applicable. The data to be obtained are the in-reactor measurement of wall thickness of top and bottom of the tube and rib height. These measurements must be made on a repetitive basis on the same tubes. The frequency of the measurement should be consistent with the expected change and the sensitivity of measurement. These measurements must be periodically checked by ex-reactor tube examinations. The measurements should be initiated at the time of installation and should encompass a large enough number of tubes to characterize the "block." The number of tubes envisaged is 100-150 with repetitive measurements at every metal cycle. To provide these measurements, the development work on the sector probolog, the feasibility of which has been demonstrated, must be completed and the instrument made "operational." A device to measure rib heights must be developed. The instrument must not only measure the height of the rib above the wall but also measure the absolute reduction of the rib height and any variations in height between one rib and the other.

These measurements will characterize "what is happening," but an essential element in determining "how it happened" is temperature distribution. To monitor this, the thermocouple probe must be used in 5-10 of these 100-150 tubes on a continuous basis. It is not necessary that the same tubes be probed each metal cycle, but during the course of the tube life, the same tubes should be probed once every year. The use of thermocouple probe data in conjunction with probologging may well dictate possible

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methods for extending tube life to the maximum possible under any given temperatures and water quality conditions. Details of this are discussed in the next section.

The above program in general can be applied to the K and the older reactors. However, some variation in number of tubes will be required. For the K Reactors, a larger number of tubes would be needed because the central zone is twice the size of C Reactor. At the older reactors, more tubes might be required because of the variability in tube history.

As pointed out above, it is essential that the monitoring program be conducted in a manner so as to provide data points uniformly with time for the duration of the tube life. As an essential correlary, manpower and organizational responsibility must be in place to continually organize, integrate, and interpret the data obtained. An initial attempt at formulating such a program is documented in HW-72499.*

As the "technical monitoring program" indicates the need for mass probing or as the expected time for major tube replacement approaches, the central zone should be probologged to determine what specific tubes are to be replaced, and thus accomplish the second objective. As in the present case at C Reactor, the replacement of ~100 tubes in CY-1962 might well forestall major replacement (>1,000) for 6-12 months. Similarly, on the next replacement 3-5 years hence, the replacement of a small number of tubes preceding the major replacement may be desirable.

2. Extension of the Life of Residual Tubes

Measures that can be taken to increase the life of current process tubes include:

- a. Optimizing fuel size.
- b. Use of water-mixers.
- c. Shifting the charge position.
- d. Adjusting pH.

The first three of these involve decreasing the high local water temperatures resulting from rib attrition or shifting the point of maximum corrosion rate to a position in the tube where wall thicknesses are greater. The possible benefits of each of these measures are discussed below.

a. Optimizing Fuel Size

The tube life in reactors which contain tubes in the same general corrosion condition may be increased by optimizing the fuel size. The K Reactors are the only Hanford reactors in this state. The older

* Hough, C. G. Tube Corrosion Monitoring Program, HW-72499. 1/26/62.

reactors contain both new and corroded tubes, and at C Reactor, re-tubing is scheduled to begin in March, 1962.

Figure 4 demonstrates the effect of increasing fuel diameter on the R Values of corroded process tubes. Considering a tube condition represented by a Corrosion Index factor of 20, the BTR R Value is decreased by about 14 percent and the TOA R Value is increased by about 20 percent by changing the fuel diameter from 1.460 inches to 1.474 inches.

The penalties suffered by decreasing BTR corrosion rates by this method are:

- 1) A decreased tube flow rate which could result in a lower power level.
- 2) An increase in TOA corrosion rates. Thus, it must be determined that sufficient TOA wall thickness is available to withstand these increased corrosion rates.

b. Use of Mixer Elements

1) New Tubes

Experience at the Hanford reactors has shown that water-mixer elements can be used effectively to reduce TOA corrosion rates. The effect of water-mixer elements on R Values in new tubes is shown in Table 1. The water-mixers are shown to decrease TOA R Values by 10-20 percent and to increase BTR R Values by 0-5 percent. Referring to Figure 1 and assuming a 100°C tube coolant temperature, the mixer is indicated to reduce top-of-annulus corrosion rates in the K-Reactor case by 0.7 mil/month, and increase between-the-rib corrosion rates by less than 0.1 mil/month from the reduction in R Value alone. However, the ability of the mixer to achieve this reduction in TOA corrosion rates depends on its location. It is obvious that placing the mixer at the second fuel element from the rear of the charge would achieve a large reduction in R Value but may do no good in terms of tube corrosion. The location is chosen to provide approximately equal TOA water temperatures at the mixer location and end of the charge assuming a cosine power distribution and a mixer efficiency of 80 percent, as illustrated in Figure 12.

Mixing efficiency is defined by the equation:

$$E = \frac{R_T - R_M}{R_T F_x - F_x}$$

Where R_T = Top-of-annulus R Value

R_M = R Value with mixer

F_x = Fraction of bulk Δt from front of charge to position x assuming a cosine power distribution

E = Efficiency

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TABLE 1

Effect of Water-Mixer Elements on R Value - New Tubes

Reactor	Fuel Element	R _{TOP}		R _{BOTTOM}		Tube
		w/o Mixer	w/Mixer	w/o Mixer	w/Mixer	
D	OIIN	1.28	1.12 (7)*	0.9	0.9	Non-uniform
	OIINB	1.27	1.10 (7)	--	--	Non-uniform
	OIIINB	1.15	1.07 (7)	0.88	0.92	Uniform
C	CIN	1.70	1.34	0.85	0.85	Non-uniform
K	CIN	1.28	1.14	0.85	0.92	Non-uniform

* Numbers in parenthesis show mixer location in terms of fuel element position.

Current mixers have been experimentally determined to be 80 percent efficient according to the above equation. (11,21) The relationship between the TOA temperature with and without a mixer is shown in Figure 12, assuming an R_{TOA} without mixer of 1.25. Under the conditions shown, use of a mixer located at position 8 would reduce the maximum corrosion rate by 50 percent, while locating the mixer at position 5 would result in only a 27 percent reduction. Thus, use of mixers properly positioned can permit higher initial rib heights and longer tube life.

2) Corroded Process Tubes

As shown above, the water temperature between the ribs increases as the tubes corrode and wear in service. The question naturally arises as to whether water-mixers can effectively slow down the high terminal rates of corrosion between the ribs. Recent data are available from the C and K Reactors which show that the water-mixers do indeed reduce high BTR coolant temperatures. These data are shown in Figures 13 and 14, plots of R Value versus mixer position in the process tube. Position is shown as the number of fuel elements counting from the rear of the charge. BTR and hole water temperatures are reduced and TOA water temperatures are increased with the use of mixers. The magnitude of these changes depends on mixer location. With present K-Reactor tubes (Figure 13), a mixer upstream of the twelfth position has no effect on the R Values at current operating conditions. The reason for this may be seen by referring to Figure 6, a plot of Δt versus position in corroded process tubes. For the conditions plotted, the figure shows that the difference between the various water temperatures are not significantly different until after the water has passed about two-thirds of the length of the process tube.

This results from the fact that ribs are relatively high in the front section of the process tube. In these positions, the BTR temperature is actually lower than bulk and TOA temperature, though not significantly so. The temperatures begin to diverge downstream of the center-line due to decreasing rib height and tube wall thickness which opens up the top annulus and restricts the BTR annulus. Since the mixer combines and mixes water from the hole and annulus, a change in R Value would not be observed until the temperature differences were significant.

In the corroded tube case, the location of the mixer thus becomes more critical than in the new tube case, as can be seen by comparing Figures 5 and 6. If the mixer is placed too far upstream, it will have little effect on the BTR temperatures. Placed too far downstream, the measured temperatures are reduced, but the condition at the location of maximum corrosion rate has not changed. Thus, the mixer is located in corroded tubes by studying out-of-reactor tube examination data to establish the location of minimum BTR wall thickness. Because of the variations between tubes, a single location will not protect all tubes to the same degree, and it is further important that the mixer element does not increase corrosion rates as it would if placed in locations where the BTR temperatures were low in relation to hole and TOA coolant temperatures. This may be avoided by placing the water-mixer at a point in the tube where the BTR corrosion is equal to or greater than that experienced in the perf zone. Since the water temperature in the perf zone is uniform, and since corrosion rates are largely governed by water temperatures, the local BTR temperature at the water-mixer would be about equal to the bulk coolant outlet temperature. Therefore, the BTR temperature at the mixer must be considerably higher than local bulk temperature, and the mixer should be beneficial. Further, if conditions assumed in Figure 6 are typical, these calculations indicate that no matter where the mixer is located, the worst result would be no change in BTR temperature.

c. Shifting the Charge Position

Maximum corrosion rates and rib attrition occur in the downstream section of the tube where water temperatures are highest. It follows, therefore, that if the tubes may be short charged or the fuel elements moved upstream onto higher ribs, R_{BTR} will decrease because the overall BTR annulus becomes larger. Tests in K and C Reactors have indicated that this is so.

The effect of charge position on process tube coolant temperatures is shown in Figure 15. For this plot, the conditions shown in Figure 6 are assumed as the base case, and the effect of removing three fuel elements from the rear and placing them at the front of the charge was determined. Thus, these fuel elements are relocated from an area of low ribs to an area of high ribs. R_{BTR} is shown to decrease about

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7 percent and R_{TOA} to increase about 8 percent. The area from which the fuel elements have been removed will now be exposed to water at the tube bulk outlet temperature. In this area, tube BTR corrosion rates are decreased. However, the corrosion environment at the rear of the relocated charge is more severe than before, at both the top and the bottom of the tube. Hence, unless the point of maximum corrosion is downstream of the relocated charge, the shift in charge has no apparent benefits.

d. Adjusting the pH

Reduction of the pH of plant process water below 7.0 almost certainly will reduce aluminum process tube corrosion rates. Such a reduction to pH 6.6 has been made at the two K Reactors on the basis of previous testing. However, a quantitative evaluation of the benefits at the K Reactors cannot be made because there is no "control" with which to compare the behavior at the lower pH.

After a controlled test is conducted, so that the benefits of the pH reduction can be balanced against costs, and so that any deleterious side effects can be identified, then the use of water of pH below 7.0 may be an available method for extending process tube life at all reactors.

3. New Tube Geometry

Very little freedom is available for revising ribbed process tube designs. The outside diameter is fixed at present values by the graphite channel diameter. Tube replacement will be accomplished on a piecemeal basis; therefore, it is necessary to design the tube to accommodate present fuel element designs. This places a limit on wall thickness to provide the present tube flow rates. The remaining variable, the rib height, is limited by the requirement that top-of-annulus failures be avoided. Available data indicate that an initial R_{TOA} of 1.25 is satisfactory, providing water-mixer elements can be used to control TOA corrosion rates. Appendix C lists uniform and non-uniform process tube designs which meet these criteria for all reactors.

From process considerations, the uniform tube designs are favored for the following reasons:

- a. These designs provide an extra 7 mils of tube wall in the area between the tube ribs which offer the prospect of additional months of service even at the high terminal corrosion rates that may prevail at that location near the end of the tube's life.
- b. The horizontal inside tube diameter is less, which will tend to provide a large fraction of the annulus flow to the top and bottom of the tube.

The only apparent disadvantage is that the top-of-annulus wall thickness has been reduced. Out-of-reactor tube examination data indicate that

the accumulated corrosion at the top-of-annulus can be controlled to less than 30 mils over the life of the tube with an initial R_{TOA} of the order of 1.25 when water-mixer elements are used. Since water-mixers are expected to be needed for both the non-uniform and uniform-wall designs, the latter designs are recommended.

4. Potential Life of Aluminum Tubes

With a given inlet and outlet temperature and at a given pH, tube life as limited by internal corrosion depends on the variation of temperature around the annulus and the change in this variation with time. Therefore, obtaining a given tube life requires a corresponding control of temperature variation about the annulus. The longer the tube life desired, the better the control required.

This scheme of events is graphically shown in Figure 16, where the required Effective R Value (ER) is plotted against tube life for operating conditions of 10°C inlet and 105°C outlet temperature. The Effective R or ER Value is defined as that R Value which, if held continuously, would produce the same amount of corrosion at a given point on the tube as actually observed. For instance, assume that the R Value at the top of a tube is 1.4 for one year and 1.0 for a second year. The value of ER for the top-of-annulus at the rear of the fuel charge for the two-year period is about 1.28.

The results shown in Figure 16 should be slightly conservative. It was assumed that the tubes would be replaced after experiencing about 25 mils of corrosion. The actual allowable corrosion in the new tube design is probably more nearly 30-35 mils. This "extra" 5 to 10 mils allows for variation from expected corrosion rates and errors in judgment of R Value trends.

The value of ER for the entire tube, both top and bottom, cannot drop below 1.0, which corresponds to a limiting tube life under the conditions outlined of slightly over seven years. This corresponds to perfect temperature distribution throughout the life of the tube - something unattainable.

It is of interest to discuss ~~probable~~ steps required to attain various tube lives. While the requirements to attain given lives are not exact, they do rest on a studied analysis of experience and data. The following discussion is based on: pH 7.0 cooling water, tube inlet temperature of 10°C, and tube outlet temperature of 105°C.

a. Two-Year Tube Life

As shown in Figure 16, a tube life of two years requires a value of ER of 1.23 or less. This should be relatively easy to attain. Adoption of a fuel-tube design which will produce an initial R_{TOA} of about 1.25 should produce the required life with no or very little added

effort. The use of mixers would probably not be required, tube monitoring requirements would be minimal. After about one year's service, a determination of wall thicknesses and R Values would be required to determine if satisfactory life as limited by internal corrosion would be obtained without further corrective steps.

b. Three-Year Tube Life

To attain a three-year tube life would require more effort than demanded by a two-year life. Again, a fuel-tube design would be selected which would produce initial R_{TOA} values of about 1.25. In this case, the initial use of mixers to reduce TOA R Values and corrosion rates would be recommended. Furthermore, tube monitoring plus thermocouple probing would be required to monitor wall thicknesses and R Values to assure internal corrosion did not shorten tube life to less than three years.

c. Four-Year Tube Life

Four-year tube lives are considered attainable with the expenditure of proper monitoring and corrective measures. Initial mixer usage would be required; and in fact, the use of two mixers per charge initially might be indicated. Close monitoring of process tube conditions and R Values would also be needed. During the final two years of service, the mixer or mixers would be located to minimize BTR corrosion rates. A fuel size change might also be needed.

d. Greater than Four-Year Tube Life

Tube lives greater than four years with pH 7.0 water are considered possible. However, it becomes increasingly difficult to obtain each additional increment of tube life. Attempts to attain tube lives appreciably greater than four years might result in diminishing returns.

The analysis of steps required to obtain given tube lives was made, assuming a coolant water pH of 7.0 and an inlet temperature of 10°C and an outlet temperature of 105°C . This is ~~believed to be~~ representative of most central zone tubes in operation at current bulk reactor outlet temperature limits and reactor orificing efficiencies. Changes in bulk outlet temperatures or changes in orificing efficiency would change the conclusions. For instance, a reduction in the reactor bulk outlet temperature of 5°C would ~~probably~~ increase tube lives by 40 percent.

5. Operational Analysis

This report has discussed the technical factors affecting process tube internal corrosion. It is evident from this study that this topic is only a portion of a much broader subject - the efficient use of ribbed aluminum process tubes in the existing reactors. There are many operational and economic considerations of great importance in their effect

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upon the practical life of process tubes, as well as upon the over-all operating plans of the reactors. Thus, there is a need for the integration of the technical considerations in extending tube life with the operational efforts to achieve the greatest practical service from the tubes, within the requirements of production considerations.

Operational analysis, of the type developed for the study of fuel element performance, should be applied to the utilization of process tubes, and the relationships developed should be considered in the development of reactor operating plans. The operational benefits resulting from reduced corrosion rates and longer process tube service lives that are obtained by adjusting water treatment, installing water-mixing fuel elements in the fuel charge, and tube monitoring efforts should be considered against the additional water treatment chemical costs, reactivity losses of mixing elements, and extra outage time for monitoring. The various costs, which may be considerable, incurred in extending the life of the process tube should then be considered with respect to tube replacement capabilities and the value of production. The resulting relationships should be integrated with other pertinent considerations now included in the "optimization" program to determine the best over-all operating plans for the reactors.

This approach, which is an attempt to formulate a quantitative relationship from an existing qualitative concept, should result in more clearly defined incentives for the various phases involved in the use of aluminum process tubes. The study might show that the cost of developing methods and equipment, and training of personnel to essentially double tube replacement rates may be less than the added acid cost of low pH, and the reactivity and production losses associated with mixers. An ability to replace the tubes in the entire central zone of a reactor in three to four weeks (even though this may not currently be considered possible) might more than compensate for the loss associated with a short tube life, and make it uneconomical to incur the incremental cost necessary to extend tube life.

To make the best ^{use} of process tubes now being, or soon to be, installed, the operational analysis should be initiated concurrently with the establishment of a tube monitoring program upon which it would depend for source data and for periodic reassessment.

The incentives for such an "optimization" study thus appear to be:

- a. A quantitative formulation which would result in the "best operating plan" for a given reactor.
- b. A better definition of the incentives for various development efforts associated with the use of aluminum process tubes.
- c. The minimizing of the cyclic concern and confusion associated with the periodic major tube replacement by the adoption of the optimization approach and its integration into the over-all reactor programming.

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These incentives appear sufficiently attractive to warrant the considerable effort required for the initiation and completion of an operational analysis, and to assure that it reflects current operational considerations.

The urgency for initiating this program of operational analysis stems from the need to collect and process in advance much data so that the necessary relationships can be developed before they are needed. Perhaps a year will be necessary for data collection and analysis, after which useful results will be obtained. By beginning the work now, effective use of the analysis can be well ahead of mid-life for process tubes now being installed.

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E. Bibliography

1. Dillon, R. L. A Proposed Mechanism for the Corrosion of Aluminum in Water, HW-59687. 3/10/59.
2. Lobsinger, R. J. Summary Report on the Corrosion of Aluminum in High Temperature Dynamic Systems, HW-59778 REV. 2/1/61.
3. Dillon, R. L. Dissolution of Aluminum Oxide as a Regulating Factor in Aqueous Aluminum Corrosion, HW-61089. 8/31/59.
4. Dillon, R. L. and R. J. Lobsinger. Progress Report; January 1957 - April 1957, Corrosion of Aluminum in Deionized Water, HW-49431. 3/20/57.
5. Lobsinger, R. J. and N. R. Miller. A Review of the Aluminum Development Program, HW-45054. 8/21/56. (Secret)
6. deHalas, D. R. and N. R. Miller. Slug and Tube Corrosion with Reduced Dichromate Concentration, HW-30952. 4/9/54. (Secret)
7. Hall, R. B., N. R. Miller and R. W. Pitman. Effects of Water Quality on Effluent Activity, Production Test IP-233-I, HW-59026. 1/27/59.
8. Kreiter, M. R. Unpublished Data, Thermal Hydraulics Operation.
9. Strege, D. E. Results of Actual Tube Wall Thickness Measurements, C Reactor, HW-72570. 1/30/62.
10. Batch, J. M. Monthly Report, Thermal Hydraulics Operation, January 1962. 1/26/62. (Confidential-Undocumented)
11. Miller, N. R. Status Report on the TOA Corrosion Problem, HW-62462. 11/8/59. (Secret)
12. Goldsmith, S. Interim Report, Production Test 105-510-E, Determination of Pile Irradiation Effects on Corrosion of 2S Aluminum, HW-27531. 6/1/53. (Secret)
13. Richman, R. B. Radiation Effects on Aluminum Corrosion in Hanford Process Water, HW-57854. 10/31/58. (Secret)
14. Gray, T. J., ed. "The Defect Solid State," Interscience Publication, Inc. New York: 1957.
15. Wilson, C. D. The Effects of Water Velocity on 2S Aluminum Slug Jacket Corrosion, HW-26033. 10/23/52. (Secret)

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16. Gruber, W. J. Examination of Severely Corroded Process Tube from 2481-DR, HW-64612. 4/11/60. (Secret)
17. Hovorka, D. L. Personal communication.
18. Taylor, A. T. Status Report, Aluminum Process Tube Development, HW-34229. 11/27/54. (Secret)
19. Wilson, C. D. Corrosion Testing of Aluminum Alloys, HW-37414. 6/22/55. (Secret)
20. Hope, R. S. Personal communication. 2/21/62.
21. Miller, N. R. Relocation of Water-Mixing Elements at the Five Oldest Reactors, HW-68424. 2/8/61. (Secret)
22. Miller, N. R. Status Report, C-Reactor Process Tubes, HW-64418. 3/21/60. (Secret)
23. Letter, P. A. Carlson and N. R. Miller to R. E. Dunn, "The Use of Water-Mixing Fuel at C Reactor." 2/2/62.
24. Graves, S. M. Use of Mixing Slugs for Tube Corrosion Considerations, HW-60356. 5/13/59. (Secret)
25. Letter, G. F. Owsley to W. J. Ferguson, "C-Reactor Charge Displacement." 9/19/61.
26. Letter, S. L. Nelson to N. R. Miller, "Program for Tube Corrosion Monitoring - K Reactors." 3/21/61.
27. Letter, D. H. Curtiss and N. R. Miller to W. J. Gartin and S. L. Nelson, "Process Tube Program and Procurement - K Reactors." 4/7/61.
28. Deobald, T. L. Unpublished data.
29. Warren, William J. "Probolog Application of Eddy Current Technique to Non-destructive Testing," Corrosion. October, 1954.
30. Strege, D. E. Personal communication.
31. Fouts, J. M. The Probolog's Role in the Measurement of Reactor Process Tubes, HW-72124. 12/28/61.
32. Letter, E. J. Nesselson to W. J. Ferguson, "Relocation of Mixing Element in Natural Metal Columns." 6/5/61.

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FIG. 1

EFFECT OF TEMPERATURE
ON CORROSION RATES

pH 7.0 PROCESS WATER

CORROSION RATE MILS / MONTH

TEMPERATURE, °C

ANNULUS TEMPERATURES
AS A FUNCTION OF
ECCENTRICITY

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FIG. 3

EFFECT OF R-VALUE
ON CORROSION RATES

INLET TEMP. = 10°C
OUTLET TEMP. = 110°C
PH 7.0 PROCESS WATER

FIG. 4

R-VALUE VS. CORROSION INDEX

C ~ NON-UNIFORM TUBE

RIB HEIGHT = 0.102" (NEW)

- ▲ 1.474 X .375
- 1.460 X .375
- 1.454 X .375

TEMPERATURE VS. POSITION
NEW TUBES - C REACTOR
GEOMETRY

TOP (R=1.25)
BULK
HOLE (R=0.94)
BOTTOM (R=0.9)

FIG. 5

FLOW

FUEL POSITION

$\Delta T, ^\circ C$

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FIG. 6

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FIG. 7

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FIG. 8

RIB HEIGHT ATTRITION

D-REACTOR

RIB HEIGHT ~ MILS

DISTANCE FROM RYSE ~ FT.

TUBE NO.	DATE CHARGED	DATE REMOVED
3463-D	8/56	9/61
2481-D	4/56	10/61
479-D	3/56	10/61
0963-D	12/44	9/61
1565-D	4/56	10/61

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FIG. 9A
NEW TUBE

THERMOCOUPLE
PROBE POINTS

ANNULUS

O.D. PROCESS TUBE

O.D. FUEL ELEMENT

FIG. 9B
BADLY WORN
TUBE

FIG. 12

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FIG. 13

R ~ VALUE VS. MIXER LOCATION
K ~ PROCESS TUBES

FIG. 14

R~VALUE VS. MIXER LOCATION
C~REACTOR

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FIG. 15

FIG. 16

TUBE LIFE AS A FUNCTION
OF THE EFFECTIVE R VALUE

BASIS:

TUBE INLET: 10°C
TUBE OUTLET: 105°C
PH: 7.0

DECLASSIFIEDAPPENDIX AEVALUATION OF C-REACTOR TUBES1. Past History

Operation of the C Reactor began in November, 1952. In November, 1955, after three years of service, a tube removal program was started to obtain information on the status of tubes at that time. As shown in Figures 17a through 17f, the extent of corrosion and rib wear exhibited by tubes removed during the end of 1955 and 1956 was relatively small. The amount of TOA corrosion in the perf zone was about 7 mils, and the amount of TOA corrosion just upstream of the rear of the fuel charge was 9-10 mils. This comparison shows that even with 1.44-inch diameter solid fuel, TOA temperatures were elevated over bulk temperatures. was

The amount of BTR corrosion in the perf zone averaged 6 mils, and the amount of BTR corrosion just upstream of the rear of the fuel charge was also about 6 mils. As expected, the BTR corrosion in the perf zone is very nearly equal to TOA corrosion. The near equality of BTR corrosion in the perf zone and just upstream of the rear of the charge indicates that BTR temperatures were not then excessive.

However, during the period 1956 to 1960, the amount of rib wear became significant. At the rear of the charge, the ribs had been reduced 20 to 30 mils from the nominal new tube values. At the middle and front of the tube, the amount of rib removal was much less, e.g., 10-20 mils of rib removal was observed at a location corresponding to the middle of the charge. +

While a detailed calculation was not made, it does appear that by the end of 1956, BTR temperatures were higher than bulk temperatures. Increased BTR corrosion rates leading to the end of use life of C-Reactor tubes were probably averted by the adoption of I&E fuel starting on a large scale at C Reactor at the end of 1957. The element first adopted had an OD of 1.474 inches, 34 mils greater than the solid element it replaced. This size was large enough, not only to center the fuel on the worn ribs, but actually to produce high TOA temperatures in most tubes. on

To avert high TOA fuel and tube corrosion rates and TOA boiling, the fuel size was reduced to a nominal value of 1.460 inches. This reduction of size, while large enough to ease TOA corrosion problems, did not give rise to excessive BTR temperatures.

However, by the end of 1959, it appeared that sufficient additional corrosion and rib wear had occurred to produce BTR R Values greater than 1.0. Typical profiles of process tubes in early 1960 are shown in Figure 16.

By comparing the data obtained in late 1955 and early 1956 with that obtained in 1959, it appears that the tube bottoms just upstream of the fuel charge had corroded about 9-10 mils during that three-to-four-year period. Use of

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mixer fuel elements was recommended and adopted in 1959 to avoid rapid corrosion rates. Also, for other reasons, the fuel charge was displaced two feet to the front of the tube. This had the effect of reducing corrosion rates at the maximum corroded point in the tubes and reducing BTR R Values.

During the two years following adoption of mixer elements and displacing the fuel charge, from 1959 to 1961, the value for minimum wall thicknesses changed very little. However, contrary to the implication of this statement, it was known that corrosion rates had not halted, but rather, the maximum rate was occurring upstream of the minimum point in the process tube. Based on this knowledge, in mid-1961, a change in mixer location was recommended, from the seventh to eleventh fuel position from the rear of the charge.⁽³²⁾ This step was taken to provide maximum protection to the section of the tube corroding most rapidly. However, before the effects of this move could be evaluated, the fuel charges were shifted two feet back to the rear of the tube. The mixer element was then placed at the fourteenth position from the rear of the fuel charge, leaving it at the same point in the tube. This combination of mixer position and fuel charge position apparently resulted in an increase in corrosion rates. While it would not be wise to put excessive reliance on the small amount of data obtained, it appears that since September, 1961, corrosion rates may have been as high as 1 mil/month. Certainly, BTR R Values were increased by the change.

Concern over an early end to tube lives led to a recommendation in January, 1962, that two mixers per charge be employed at the ninth and twelfth fuel positions from the rear of the charge.⁽²³⁾

2. Current Corrosion Profiles of Process Tubes

On the average, the original tubes have experienced about 20 mils of TOA corrosion, leaving a remaining wall thickness of about 55 mils. Hence, most of these tubes still have relatively thick tops, and TOA corrosion isn't expected to limit C-Reactor tube lives.

Almost all C-Reactor tubes have thinner bottoms than tops; however, the total BTR corrosion is about equal to TOA corrosion. The thin section of the tube bottoms is located in a band 10 and 16 feet from the rear flange. This is different than observed previously. For instance, in 1956, the thinnest part on the tube bottom was about 8 feet from the rear and there was little corrosion at the 16-foot point. The bottom of the tube 8-10 feet from the rear of the tube has corroded little during the past two years - certainly less than 5 mils. However, at the 16-foot point, the attack may have amounted, on the average, to as much as 10 mils. This change in pattern may be attributable in part to the protection given by the mixer to the part of the tube downstream of it. Figure 19, which shows repetitive probologs of the same tubes, illustrates the fact that the major corrosion attack during the past two months is upstream from the rear of the fuel charge. The process tube rib loss at the rear of the fuel charge now ranges between 40 and 60 mils. Together with wall corrosion, this certainly has resulted in a reduced annulus between the ribs, thus producing high values of R_{BTR} .

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3. Current Condition of Tubes

Extensive probologging done during the past few months at C Reactor allows characterizing the current ranges of wall thickness. Extrapolating the data to the entire central zone of 1100 first-generation aluminum tubes, the following distribution is obtained:

<u>Wall Thickness Mils</u>	<u>No. of Tubes w/Given Thickness</u>	<u>No. of Tubes w/Wall Equal to or Thinner Than Given Thickness</u>
35	70	70
35	30	100
36	100	200
37	100	300
38	150	450
39	250	700
40	200	900
41	100	1000
42	50	1050
42	50	1100

*Central zone Probologger
may affect their number
very slightly*

Hence, it appears there are a few tubes, perhaps 70, which are relatively thin and may be leak prone. On the other hand, the number of tubes less than 37 mils is relatively small, about 200.

4. Outlook

A tube removal outage is scheduled for C Reactor in March. Presumably, it will be possible to remove all tubes from the reactor with wall thickness of 35 mils or less, and all tubes with appreciable external attack or non-uniform internal attack, as determined by probologging. If it is assumed that the balance of the tubes can be left in service until walls are corroded to 30 mils, it appears that little danger of tube leaks will exist until, at the earliest, the last of this calendar year.

This conclusion is based on the following factors:

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- a. The water-mixing element appears to be reasonably effective in producing uniform temperatures around the annulus in corroded process tubes. With two mixers per tube, extremely high water temperatures should be avoidable even with the eccentric annulus in evidence. Corrosion rates were perhaps about 1 mil/month with a single mixer element poorly positioned. With the use of two mixers properly located, it should be possible to reduce rates well below this figure for the balance of the calendar year.
- b. A satisfactory tube monitoring program is in progress at C Reactor. It is assumed that this program will be continued. Hence, if the mixer locations selected are non-optimum now or become non-optimum in the future, it is believed that this will be detected quickly and rectified before catastrophic damage occurs. *it is believed that*

will be While it is difficult to predict tube lives with high accuracy, ~~it is believed~~ that with the completion of the March tubing, little likelihood of tube leaks will prevail during the balance of this calendar year. ~~On the other hand~~ it is also probable that most of the central zone will require retubing by the end of CY-1963 in spite of best efforts to prolong life.