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RTR-146

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TO: A. A. JOHNSON

ALUMINUM CORROSION AND TURBIDITY

INTRODUCTION

It has previously been shown (Ref. 7, 9) that aluminum corrosion and turbidity formation in the reactors correlate with fuel sheath temperature. To further substantiate this correlation discharged fuel elements from R-3, P-2 and K-2 cycles were examined for extent of corrosion and evidences of breaking off of the oxide film.

SUMMARY

Two types of corrosion were observed, both of which showed a close correlation with fuel sheath temperature over a wide range. These were uniform corrosion (which has been studied extensively in the laboratory) and pitting corrosion which has not previously been observed for corrosion of 2 S aluminum in pure water at these temperatures. Pitting corrosion increases much more rapidly with temperature than does uniform corrosion. Moderator turbidity data can be explained on the bases that

- extensive pitting starts only after an oxide film has been built up to a critical thickness by uniform corrosion
- pitting releases portions of this oxide film into the moderator as the major source of turbidity.

Data are presented to show the correlations with sheath temperature and to permit prediction of the course of turbidity changes for any reactor cycle. These correlations and predictions are in part speculative because of the subjective method used to estimate extents of corrosion. They are presented to map directions for future quantitative studies.

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PROGRAM

1. Laboratory corrosion test should be conducted to duplicate uniform and pitting corrosion as seen in the reactor and determine means of controlling them.

2. Work should be pressed to determine whether moderator contains impurities which cause pitting attack (Ref. 8, 9).

3. Work under TA 1-725 will determine whether pitting corrosion and turbidity can be reduced and perhaps eliminated by controlling the moderator pH.

4. More precise methods of measuring extent of corrosion of discharged fuel should be developed for use in more firmly establishing the correlations presented in this report.

DISCUSSION

Background

Moderator turbidity leads to reduced visibility in the reactor tank, pluggage of instrument lines and accumulation of radioactive deposits throughout the reactor hydraulic and purification systems. A better understanding of the mechanism of turbidity formation is needed as a basis for selecting measures to alleviate these problems.

The turbidity is known to be hydrous aluminum oxide (Ref. 2,3,5) which is a product of corrosion of the aluminum reactor componetns. The amount of aluminum oxide formed by corrosion of these components can be estimated from the amount of deuterium released into the blanket gas unaccompanied by oxygen. Since only a small portion of this estimated amount of aluminum oxide appears in the moderator either dissolved or as turbidity, most of it presumably remains as an oxide coating on the aluminum surfaces where it was first formed.

The presence of this oxide film has been observed on the surface of discharged fuel slugs. The amount of oxide has been estimated from the Na^{24} activity in reactor moderator on the assumption that Na^{24} is formed by the $Al^{27}(n, \alpha)$ reaction in all of the oxide film and that all of it goes into the moderator (Ref 6). The amount is in agreement with that estimated from the deuterium evolution.

The deuterium evolution rate and formation of turbidity have both been shown to be dependent on the fuel sheath temperature (Ref. 7, 11). The deuterium evolution rate increases with sheath temperature in exact proportion to the increase in aluminum corrosion rate (as measured in laboratory tests) with temperature. Turbidity is not formed in large amounts when the highest sheath temperature is much below 135°C, and is usually found when this temperature is exceeded by a substantial amount. This temperature coincides roughly to the transition from one mechanism of aluminum corrosion to another (Ref. 4). While the correlation was made with sheath temperature, the correlation with neutron flux, power level, or central metal temperature would be nearly as good.

Deuterium evolution and buildup of Na^{24} activity has been substantial when there was no turbidity formation. Both occur continuously throughout the reactor cycle, indicating that the oxide film builds up continuously during the cycle. Why is it that under some conditions the oxide film will adhere to the aluminum surface, while under other conditions it breaks loose to form turbidity?

Perhaps an answer to this question can be obtained by examining discharged fuel elements for evidence of presence and adherence of the oxide film, which would give some understanding of the measures needed to prevent turbidity formation.

Examination of Discharged Fuel

Method Premeasured and other special slugs discharged from each R and P reactor cycle are inspected in the "Hot Lab" for changes in dimensions. A large number of these slugs were examined visually during the course of inspection of R-3 and P-4 slugs. Examination was done by the author while the Laboratory Section personnel were carrying out their normal routine of measurements.

With the exception of one slug, examination was by naked eye. Each slug was observed in two positions (a) while being held in front of the periscope to read its serial number and (b) while being rotated about its (horizontal) axis on the gaging stand. The ability to recognize evidences of corrosion improved with experience as more and more slugs were examined. Two types of corrosion were ultimately recognized. As soon as a type of corrosion was clearly recognized, slugs were graded subjectively on a rough quantitative scale of extent of this type of corrosion. Other observations were recorded so that if later some new type of corrosion was recognized, there would be some chance of interpreting these observations in terms of extent of the newly recognized type of corrosion.

In such subjective grading of any variable, it is difficult to guard against a wandering of the grading scale. It was felt during the observations that such a shift occurred sometime around January 28, probably in the course of first establishing the scale of uniform corrosion. This shift affected the grading of pitting but not of uniform corrosion. A correction was made for this shift as explained in Table XI and Figure 6. The correction consisted in multiplying the more recent values of pitting by 1/2, or the earlier values by 2. The uncorrected values are given in Figures 12 through 18 and Tables I through X. In Figure 1 through 6 the corrected values are used.

Because the grading was subjective, the quantitative results presented in this report are to some extent speculative. These results are presented to map out the directions which further study by more precise methods should take.

Types of Corrosion

Three extremes of slug appearance were observed which define the uncorroded state and the two types of corrosion. These were

(1) an "almost like new" appearance in which the slug had a medium high metallic lustre

(2) a bumpy slug with a heavy white coating resumbling flat white paint

(3) a mottled shiny appearance, with an egg-shell texture. Under magnification, the shiny patches were seen to be covered with many small pits or craters 1 to 10 mils in diameter. Areas between shiny patches or between more widely spaced pits looked like the white coating of case 2 above.

On the basis of all available facts and observations it appears that Case 1 represents the uncorroded state. Case 2 represents uniform corrosion to produce an oxide film in place, and Case 3 represents a pitting type corrosion in which pits penetrate through the oxide film.

Gradations were observed between Case 1 and Case 2, with the surface becoming whiter and showing less metallic lustre as Case 2 was approached. These cases were graded according to the amount of metallic lustre to give a scale of extent of uniform corrosion. On this scale the most lustrous (least corroded) slugs were grade 0 and the whitest, least lustrous (most corroded) were graded 3. Beyond grade 3 the film had become so thick that the metallic lustre was completely obscured and any further increase in thickness could not be detected visually.

Gradations between Case 2 and Case 3 were also seen, with the surface becoming more mottled and shiny and more completely covered with bright patches as Case 3 was approached. These cases were graded similarly, with 0 for Case 2 and 3 for brightest most mottled slug observed. Further increases in extent of pitting would be recognizable if they were to occur, so that this scale can be extended beyond 3 if necessary.

No gradations from Case 1 to Case 3 (that is to say, occurrence of pits in an otherwise like-new slug, without presence of the white coating) were observed. It would seem therefore that the oxide film must build up before pitting can occur.

<u>Mark VI-J</u> Two Mark VI-J fuel tubes discharged from K-2 cycle were examined underwater for evidences of corrosion, both by unaided eye and using the underwater periscope. One was a flat zone tube which had operated with a maximum sheath temperature of 154°C (twenty degrees higher than R-3 and P-4 flat zone slugs). The other was a buckled zone tube which operated at a maximum sheath temperature of 105°C (thirty degrees below the P-4 flat zone slugs).

Conditions of observation made quantitative grading of the extent of corrosion. Only rough limits could be placed on pitting corrosion. Grading of uniform corrosion did not seem possible, in part because of lack of time to acquire ability to recognize the gradation by underwater observation.

The buckled zone tube showed a degree of pitting which would have been rated practically zero under the conditions used in rating slugs. Some very small pits were seen under 15 X magnification, and there was a slight metallic reflection (seen by naked eye) over the portion of the tube that had experienced the highest sheath temperatures. The entire tube was coated with a fairly heavy oxide film which seemed to be thinner toward the cold ends of the tube.

The flat zone tube showed much more severe pitting than any slugs that have been examined. The pitted region covered a greater length of fuel column, while the pits were larger and the surface rougher. The difference was clearly visible to the naked eye at a distance of some fifteen feet, and was confirmed under magnification. A gradation in pitting was observed along the length of the column, as with slugs.

Under high magnification (X 15) pitting was seen to extend beyond the point at which no pitting would have been reported from unaided observation. This consisted of many minute pore-like openings in the oxide film, rather widely spaced. This "pitting" was the same as was seen in the buckled zone tube. It showed a gradation, going to smaller and smaller pore size and greater spacing toward the top end of the fuel tube.

<u>Results</u>

The results of these observations are presented in the accompanying tables and figures. Cumulatively they show a good correlation of extent of corrosion with fuel sheath temperature.

The observed extent of corrosion varied in the following ways, each of which can be explained by corresponding variations in sheath temperature:

- along the length of the fuel column
- along the length of each photographed slug (not enough care was taken in visual observation to detect this detail of distribution of corrosion)
- decreased corrosion at the end caps of slugs and on the two end sections of the Mark VI-J tube.
- between fuel columns in the same Mark VII-A quatrefoil
- between fuel columns which operated with different maximum sheath temperatures either in different reactor cycles or because of different location in the reactor.

Figures 12 through 18 and Tables 1 through 10 show that all fuel columns which operated at nearly the same maximum sheath temperature showed almost identical patterns of extent of corrosion, while those which operated at different maximum sheath temperature showed different corrosion.

Figures 1 and 2 show how the variations along the fuel column correlate with sheath temperature. The end cap effect is not shown in these figures. At the end cap the neutron flux is higher than elsewhere because of the Wilkins Effect, but the heat flux and sheath temperature are lower because of absence of fuel in this region. Similarly the sheath temperature is low in the end sections of Mark VI-J tube, which contains no fuel. Thus the extent of corrosion parallels the sheath temperature (and not the neutron flux) at the end cap.

Figures 3 through 6 show the correlation between extent of corrosion and fuel sheath temperature quantitatively. In Figure 3 the extent of corrosion is plotted against sheath temperature. Fuel elements are included from different reactor charges and which operated at widely different maximum sheath temperatures (P-4 flat zone at 135° and P-4 outer ring at 90° maximum in the same charge, and K-2 at 154° maximum in a different charge). Considering the variety of cases included, and the fact that the estimate of sheath temperatures is uncertain in some cases while the measure of extent of corrosion is crude, the correlation is surprizingly good.

In Figures 4 and 5 the same data (for R-3 and P-4 flat zone only) are plotted in the form of an Arrhenius plot. This plot (logarithn of rate against reciprocal of the absolute temperature) should give a straight line for any simple reaction mechanism. The slope of the line is characteristic of the mechanism and is proportional to the "activation energy". In the present case it is legitimate to plot the logarithm of extent of corrosion, since for a fixed cycle time (50 days for each of the two cycles considered) the <u>extent</u> is proportional to the <u>rate</u> of corrosion.

Departure from a straight line in Figure 4 is expected because of the limit beyond which differences in film thickness cannot be distinguished optically. The dashed curve represents the departure that would be expected if the optical density o film were 3.1 x 104 per cm. of film thickness (Lambert's law of light absorption). The slope of the Arrhenius line agrees closely with literature data on uniform corrosion of aluminum and with evidence from reactor D_2 evolution as to the effect of sheath temperature on aluminum corrosion (Ref. 4, 7, 11)

The slope of the Arrhenius line for pitting corrosion (Figure 5) is distinctly greater than that for uniform corrosion. This would imply that pitting is not the same process which gives rise to the major part of deuterium evolution, and that it is distinct from uniform corrosion.

The effect of differences in operating temperatures between fuel columns in the same Mark VII-A quatrefoil is shown in Figure 6. The sheath temperature is a nearly linear function of the channel effluent Δ T. The extent of pitting corrosion of an entire fuel column is measured by the area under the curve of extent of corrosion vs. slug position for that column. In Figure 6 this area is plotted against the channel Δ T for all four channels of each of five quatrefoils. The correlation between pitting and channel ΔT is clearly seen in this figure.

Preliminary Conclusions

From these results it is concluded that

1) Corrosion of the aluminum sheath of both Mark VII-A and Mark VI-J fuel elements occurs by two mechanisms (a) uniform corrosion to build up an oxide film and (b) pitting corrosion which penetrates through the film into the metal.

2) Both types occur at rates which increase with temperature.

3) The mechanism of pitting is distinct from that of uniform corrosion. The rate of pitting increases much more rapidly with temperatures than does the rate of uniform corrosion.

4) There is some evidence for two different types of pitting, one much more severe than the other. Severe pitting appears to start only after the oxide film reaches a critical thickness. This thickness is just reached at 121°C at the end of a 50 day reactor cycle (see Figures 1, 2, and 3).

Turbidity Generation

Pitting corrosion breaks through a previously formed oxide film and releases the oxide into the moderator. It must therefore produce turbidity. In Exhibit A a theoretical treatment is given to show how the buildup of turbidity during a reactor cycle could be explained quantitatively in terms of pitting corrosion.

The assumption is made that pitting starts on any minute area of aluminum surface only when the oxide film has built up to a critical thickness of approximately 1.3 mils. Figure 7 shows the number of slugs which have begun to pit at any time during the reactor cycle, and the resultant rate of turbidity generation, based on this assumption. The moderator turbidity rise in R and P reactors (which have no turbidity removal facility) will follow the dashed curve of Diagram B. In L, K and C (where turbidity is removed by the reboilers) the turbidity rise will follow the dashed curve of Figure 7.

Figure 7 represents the result of operation at 127° maximum sheath temperature. At other values of maximum sheath temperature the curves are shifted along the time axis such that the time of first appearance of turbidity is consistent with the theoretical curve of Figure 8. The time of first appearance of turbidity in several reactor cycles was in fairly good agreement with this theoretical curve, as shown in Figure 8. The course of turbidity buildup in these cycles was much as shown in Figure 7. There is thus at least qualitative agreement between the theory and the actual course of turbidity buildup in the reactors. The deviations could be acounted for by uncertainties in the laboratory data on uniform corrosion, on which the theoretical curve is based.

Figures 9, 10, and 11 show that there is an even more quantitative correlation. In K-2 cycle all of the oxide formed on 3/4 of the length of flat zone tubes was removed by pitting. The buckled zone tubes, constituting 40% of the reactor charge showed negligible oxide loss. Thus (3/4) (100-40%) = 0.450 of all the oxide formed on the fuel elements should have been released as turbidity. The amount of $Al(OD)_3$ released as turbidity up to any time during the cycle was calculated from a material balance of turbidity, using the daily turbidity data and assuming 100% removal by the reboilers at 6 gpm throughput. The total amount of $Al(OD)_3$ formed by corrosion was calculated from the deuterium evolution data. In Figure 1 the amount released as turbidity is compared with 0.5 times the total amount formed by corrosion. The agreement is good.

In Figures 10 and 11 the same thing was done for P-4 and R-3 cycles. In these cycles the equivalent of all the oxide was released from only about 3 out of 20 slugs in each column. This constitutes 15% of all the oxide generated on flat zone tubes or about 9% of the total oxide generated in the entire reactor. In Figures 10 and 11 the agreement with 10% of the total oxide is good.

This provides a means of predicting the turbidity generation during any reactor cycle. The rate of oxide formation can be predicted from the maximum sheath temperature. The fraction of total oxide which will be released by the end of the cycle can be estimated from the sheath temperature curve for flat zone tubes, using the correlation shown in Figures 1 and 2 and the limit of 121°C above which pitting occurs. The prediction is expected to be only roughly accurate, until more thoroughly tested.

CONCLUSIONS

1. Aluminum corrosion in the reactor depends primarily on the surface temperature of the aluminum.

2. Pitting corrosion becomes severe at and above 121°C.

3. Pitting corrosion has been a major contributor to turbidity formation in a number of reactor cycles.

4. Tentatively it should be possible to predict turbidity generation for any reactor cycle from fuel sheath temperatures, using the methods outlined in this report.

5. The phenomena of uniform and pitting corrosion as observed on discharged fuel elements is sufficiently reproducible that it should be possible to duplicate them in the laboratory.

PROGRAM

1. An RTA (Ref. 10) is being prepared which requests corrosion loop tests to duplicate uniform and pitting corrosion as seen in the reactor. These tests should do the following

   a) establish that these phenomena can be duplicated

   b) establish that pitting does cause turbidity formation

   c) determine the conditions which affect pitting corrosion, with the aim of controlling or eliminating it.

2. Pitting corrosion has not previously been observed on aluminum in pure water. The presence of contaminants such as mercury or chlorides has been necessary to cause pitting. Work proposed in references 8 and 9 should be carried out, to determine whether such contaminants are actually present.

3. Work under TA 1-725 (Ref. 1) to reduce aluminum corrosion by controlling moderator pH should reduce uniform corrosion by a factor of about 2 or more. There is a good chance that this may completely eliminate pitting corrosion, since uniform corrosion may be prevented from reaching the critical thickness.

4. More quantitative means of determining oxide film thickness and extent of pitting should be developed. These methods should be such that they can be used to obtain a large volume of data such as reported here. From these data the quantitative relationships presented speculatively in this report would be more firmly established.

F. B. Longtin

F. B. Longtin

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APPENDIX

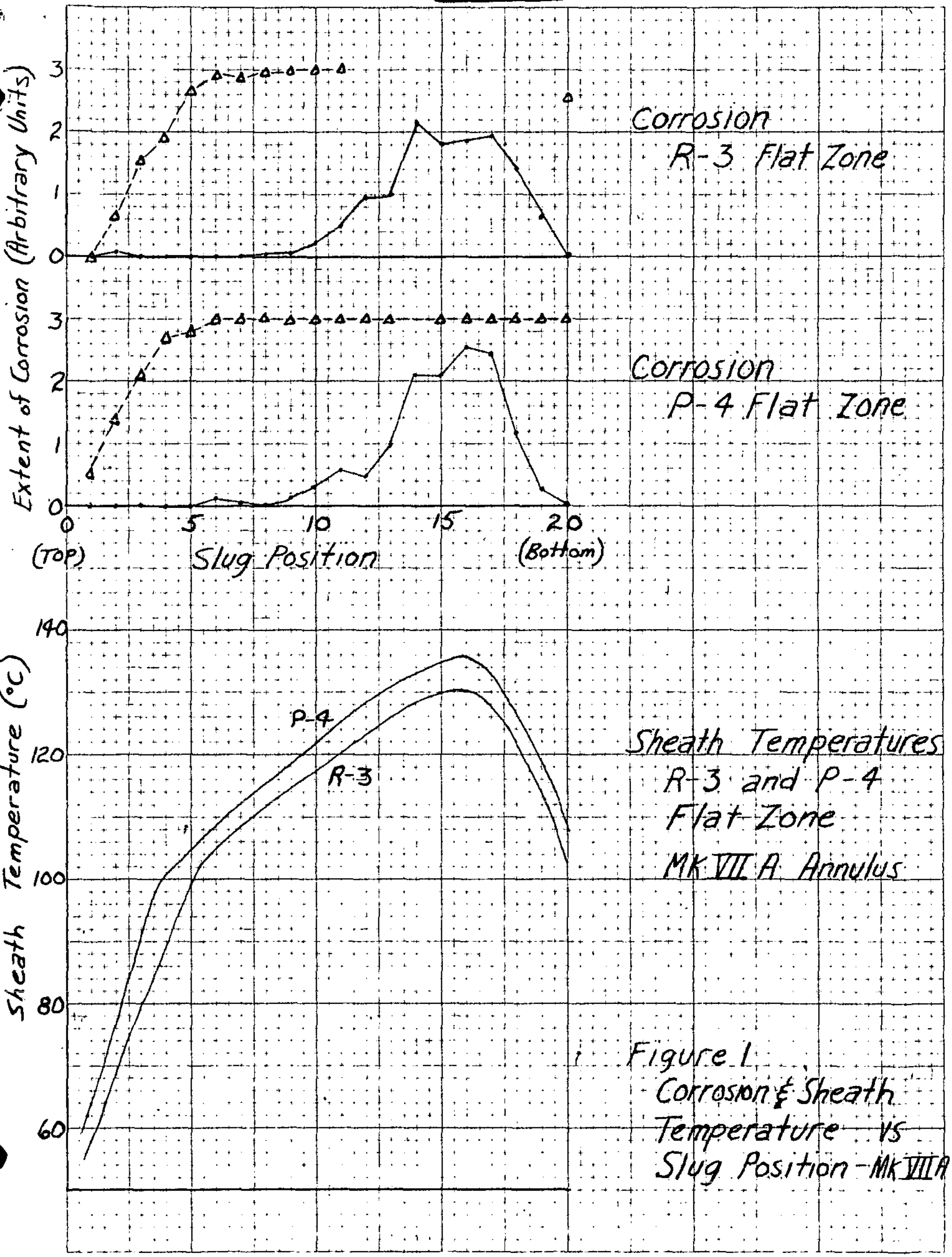

Figure 1
Corrosion & Sheath Temperature vs Slug Position - Mk VIIA

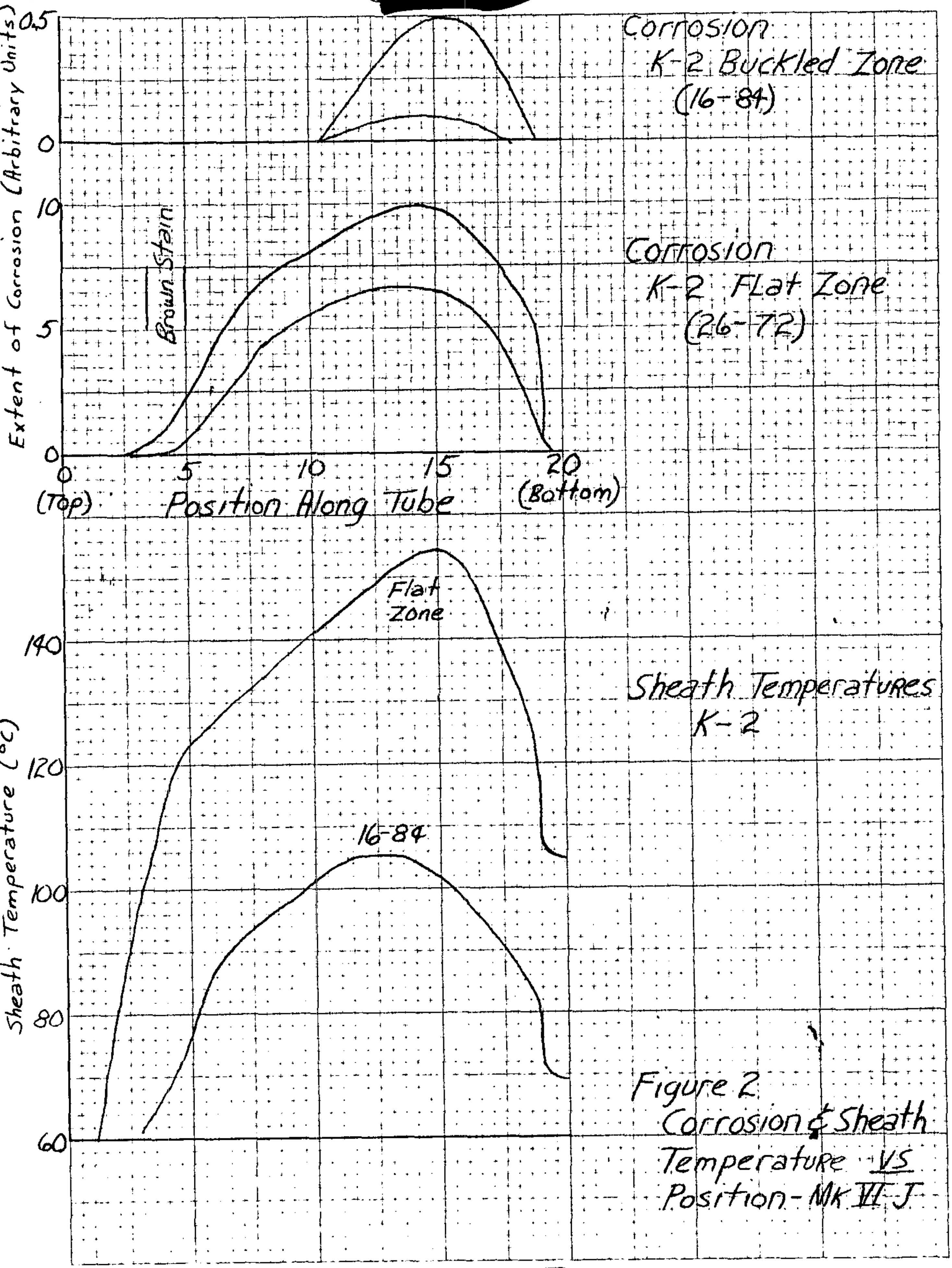

Figure 2
Corrosion & Sheath Temperature VS Position - Mk VI-J

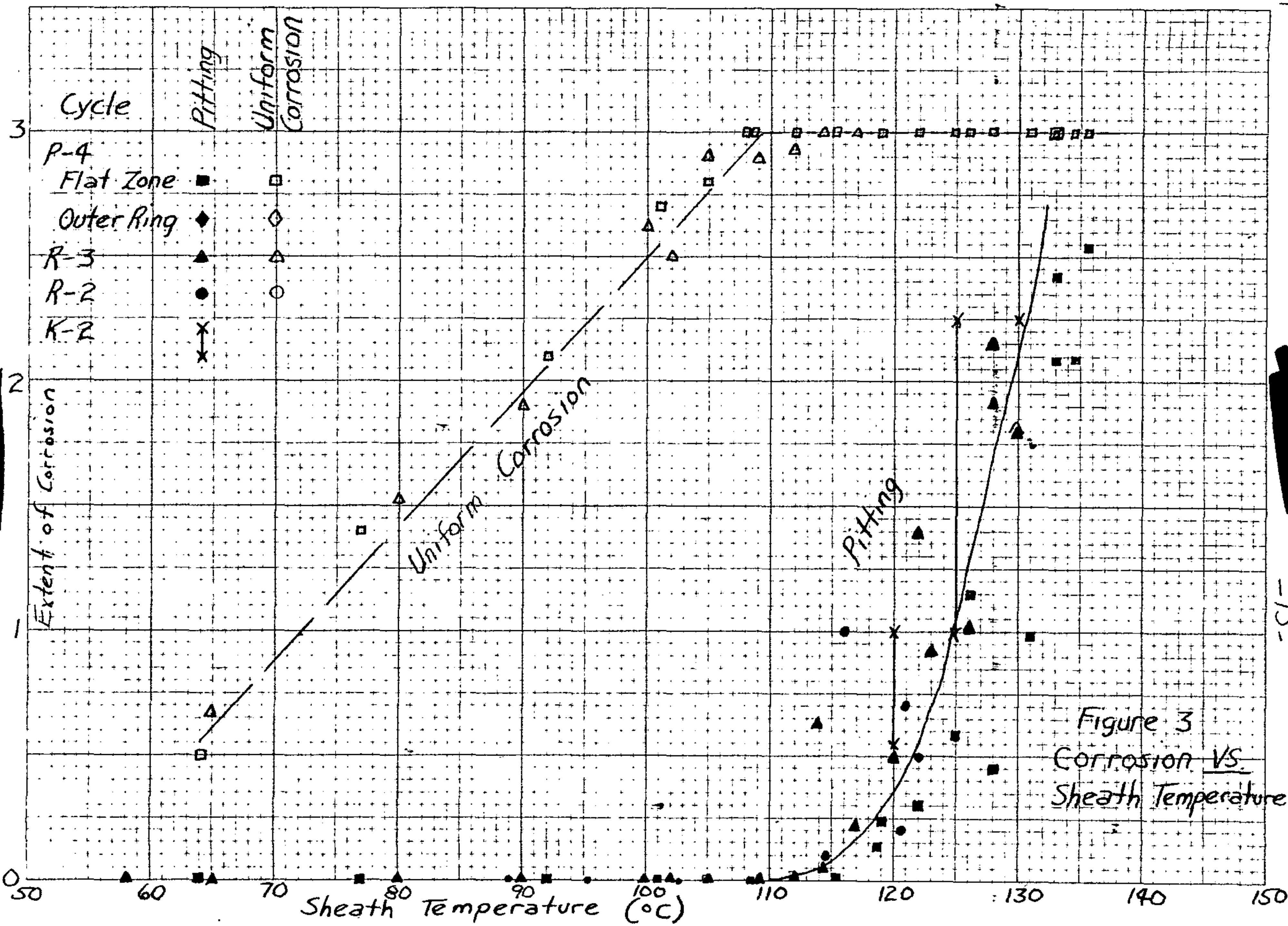

Figure 3
Corrosion VS. Sheath Temperature

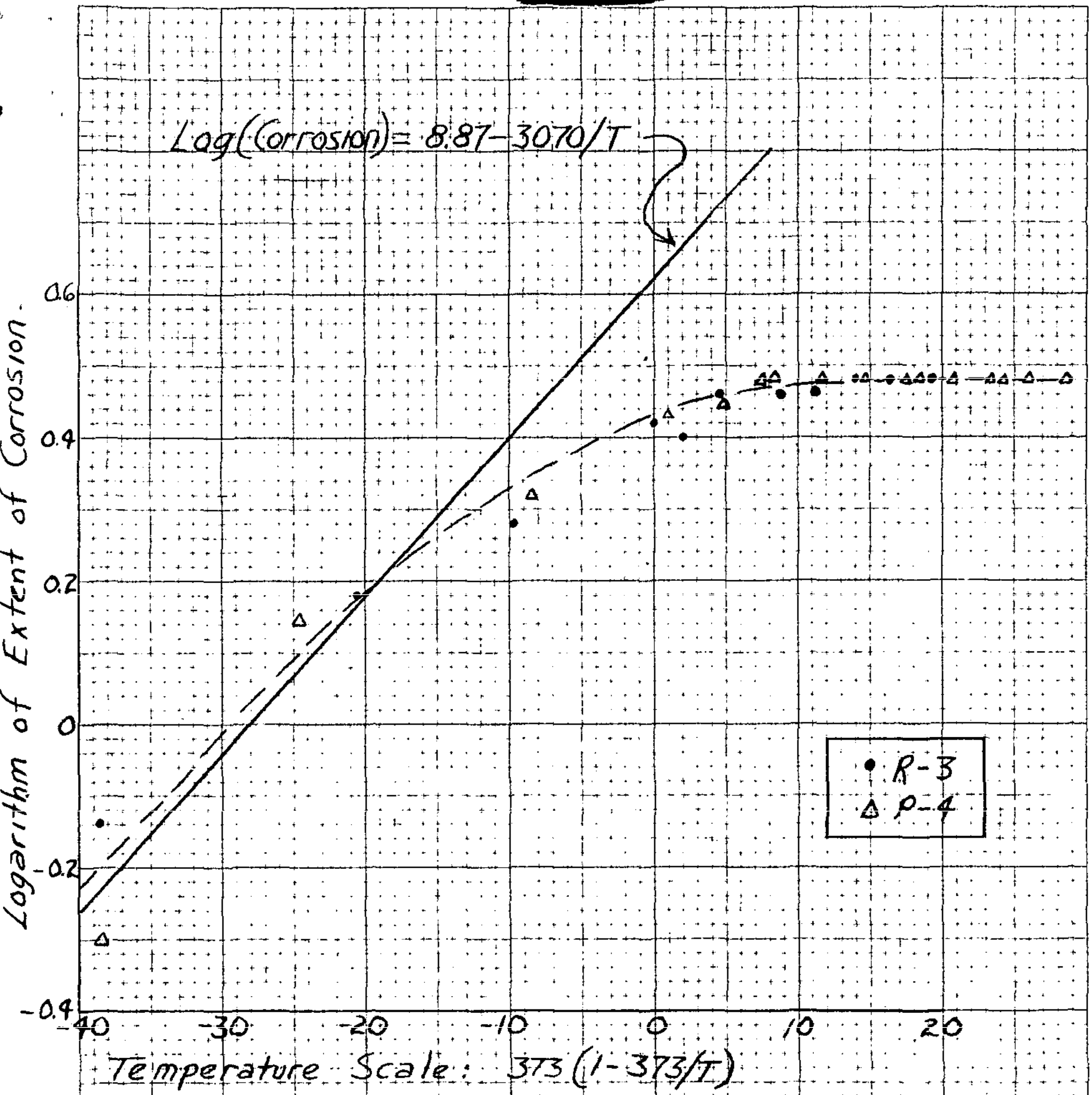

Figure 4. Uniform Corrosion Arrhenius Plot

[redacted]

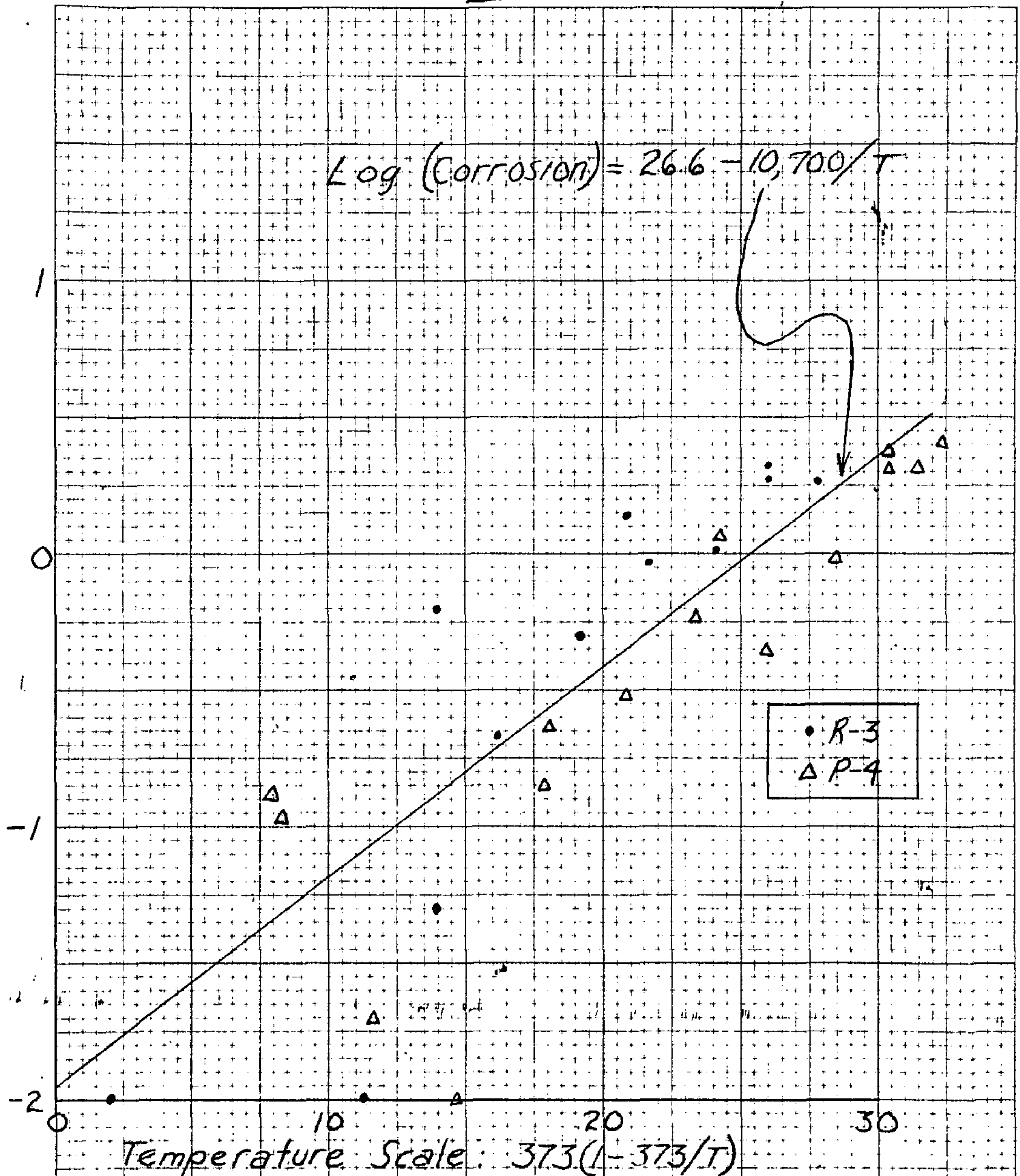

Figure 5 Pitting Corrosion-
Arrhenius Plot

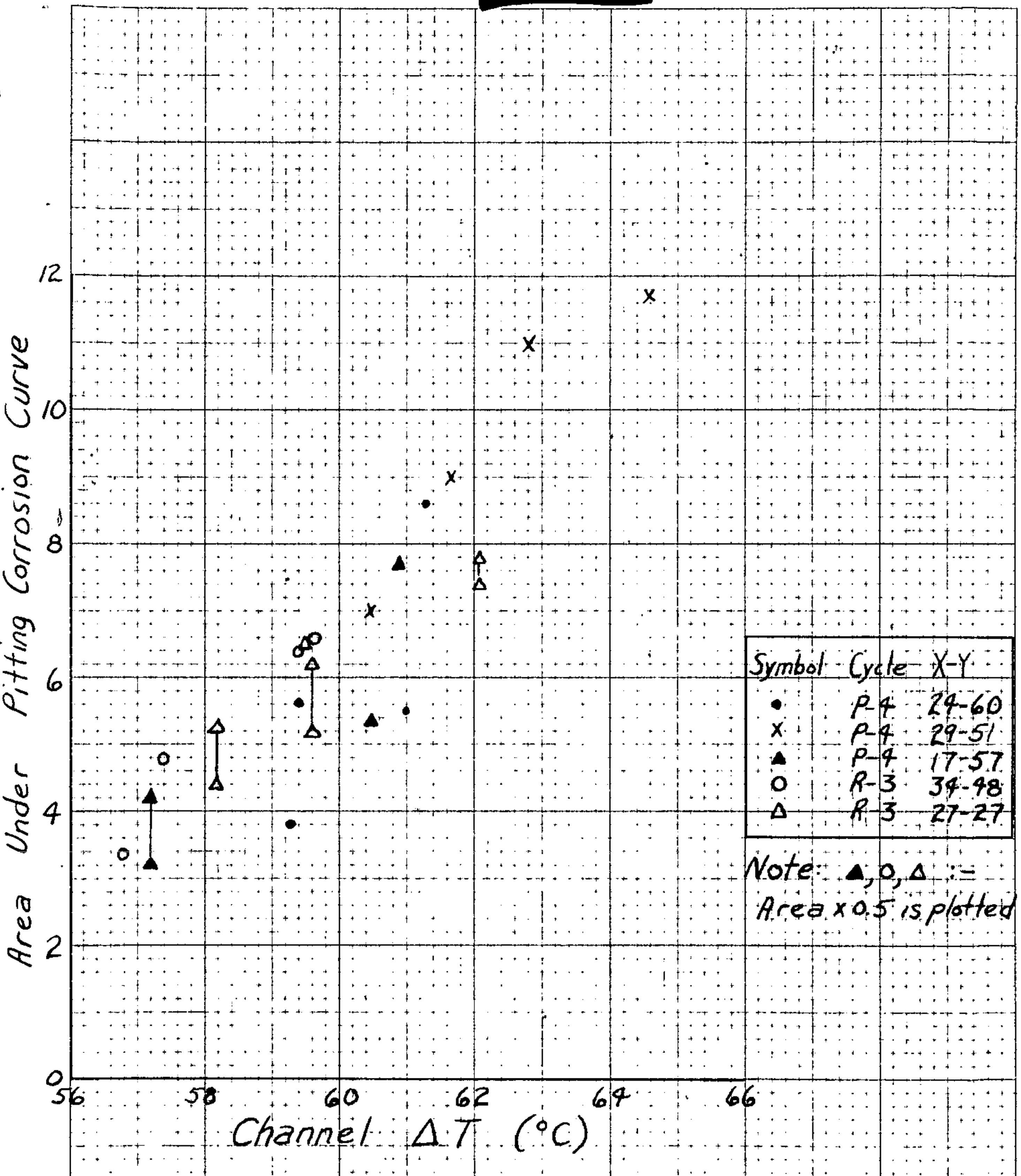

Figure 6. Corrosion in a Fuel Channel vs Channel ΔT

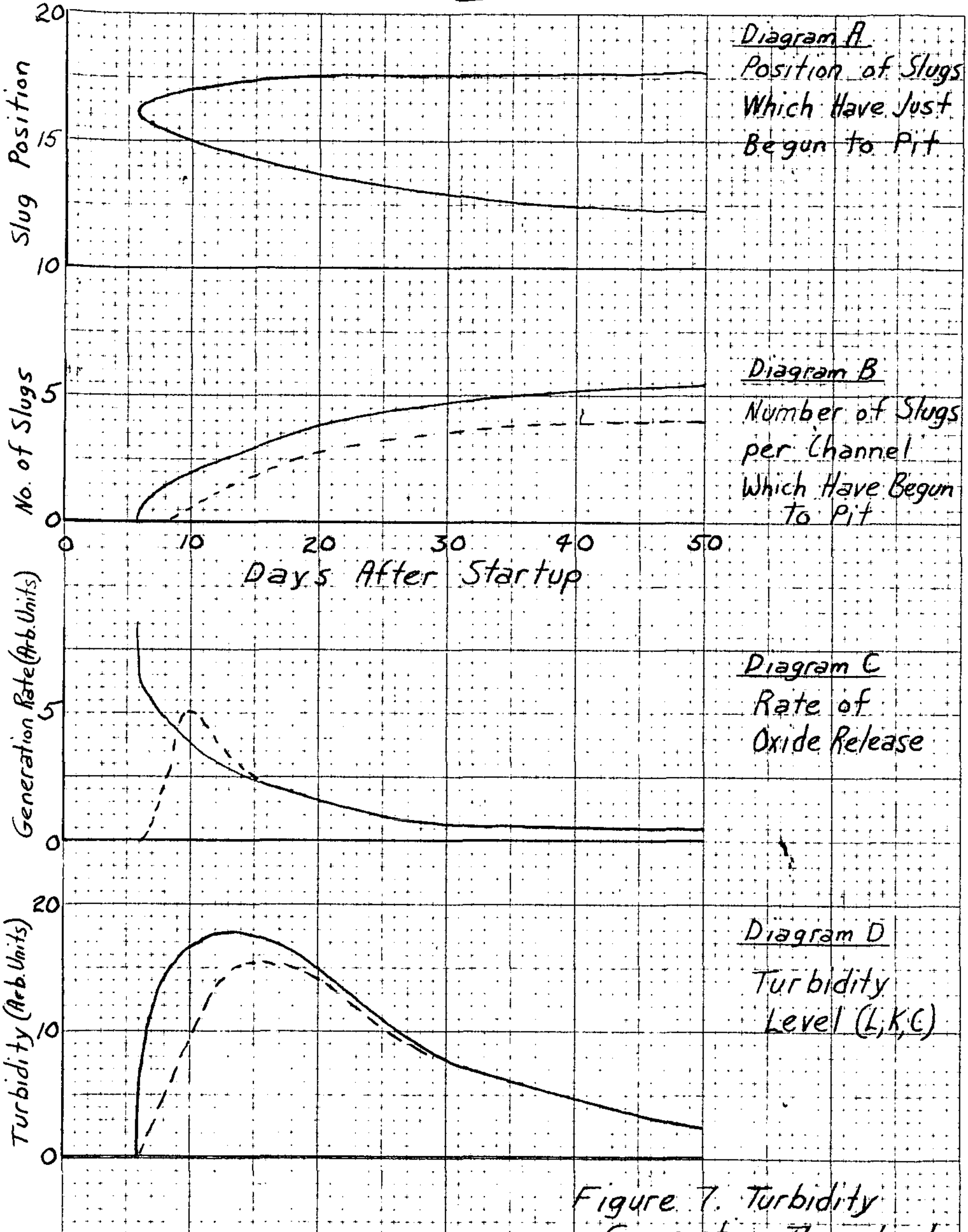

Figure 7. Turbidity Generation- Theoretical

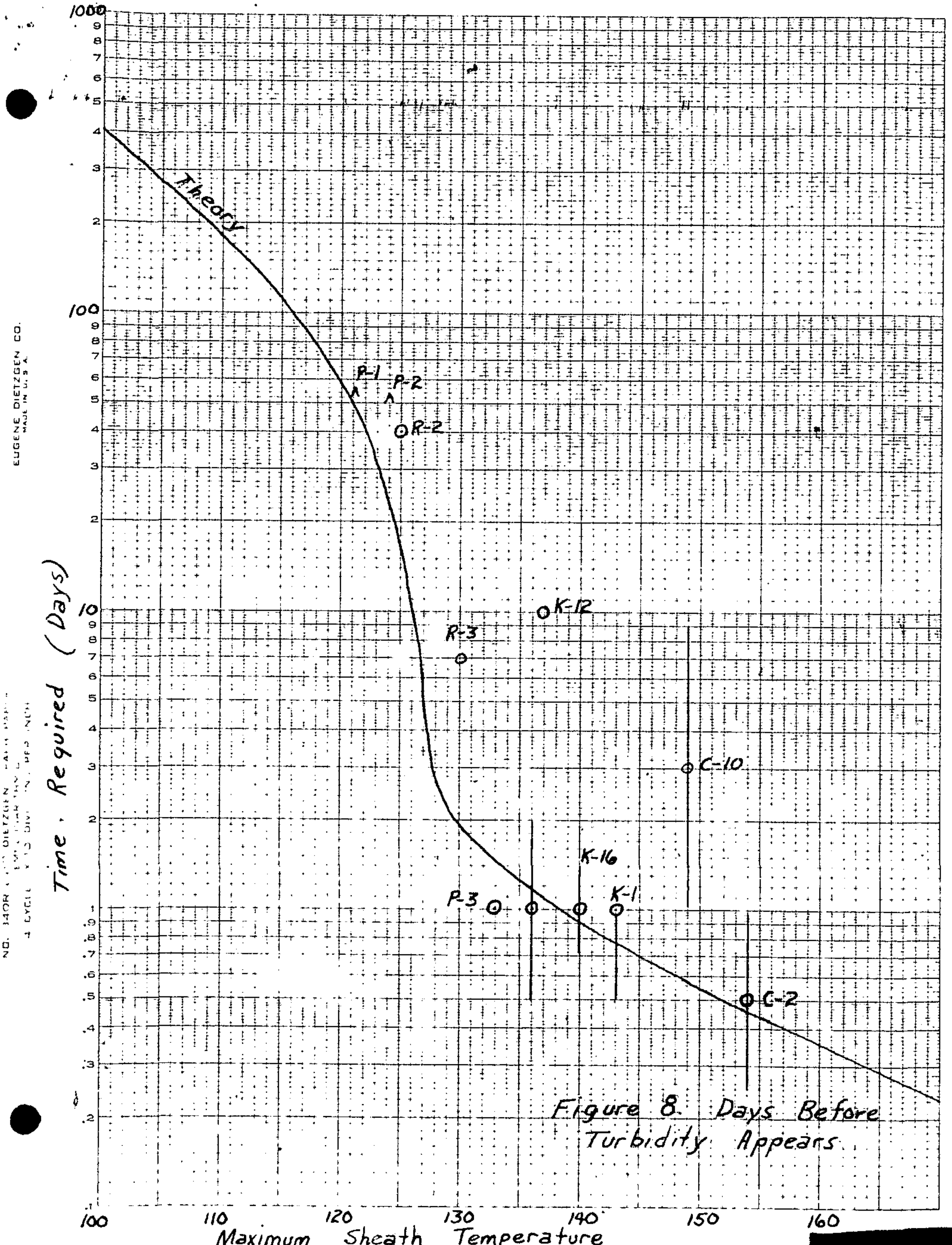

Figure 8. Days Before Turbidity Appears

[redacted]

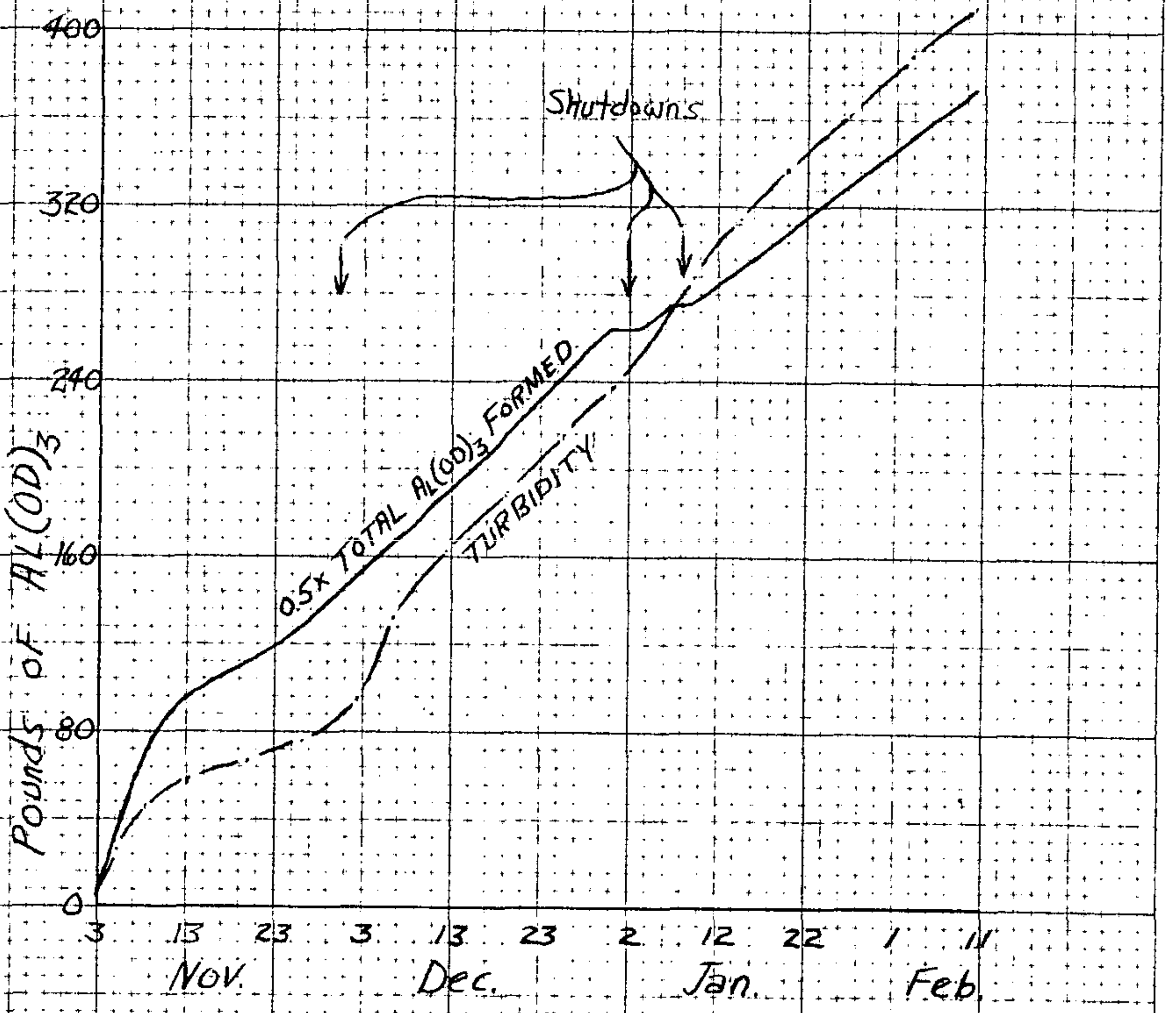

Figure 9. Turbidity Formation vs Total Corrosion- K-2

[redacted]

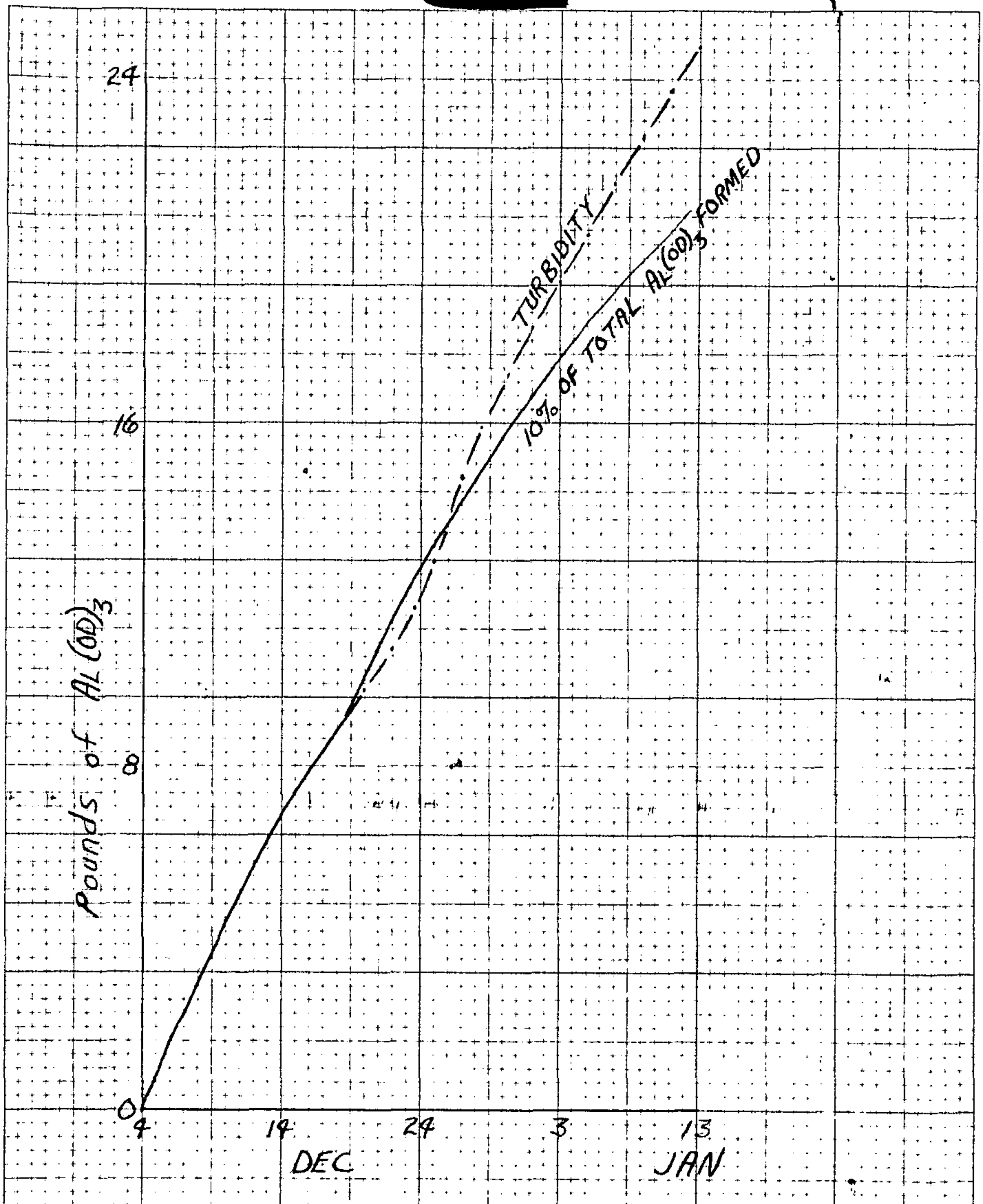

Figure 10 Turbidity Formation
VS Total Corrosion P-4

[redacted]

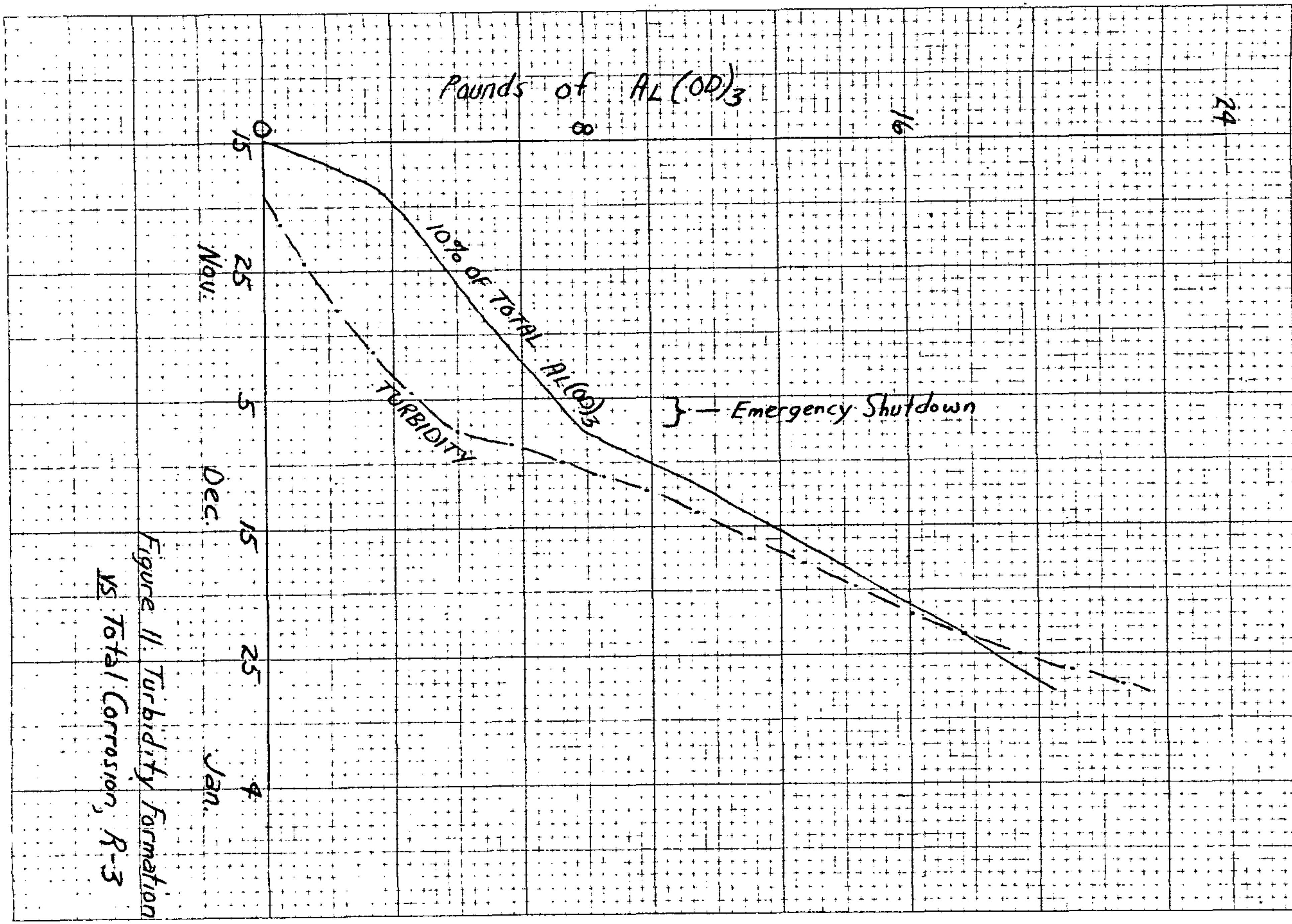

Figure 11. Turbidity Formation vs Total Corrosion, R-3

-21.

RTR 146

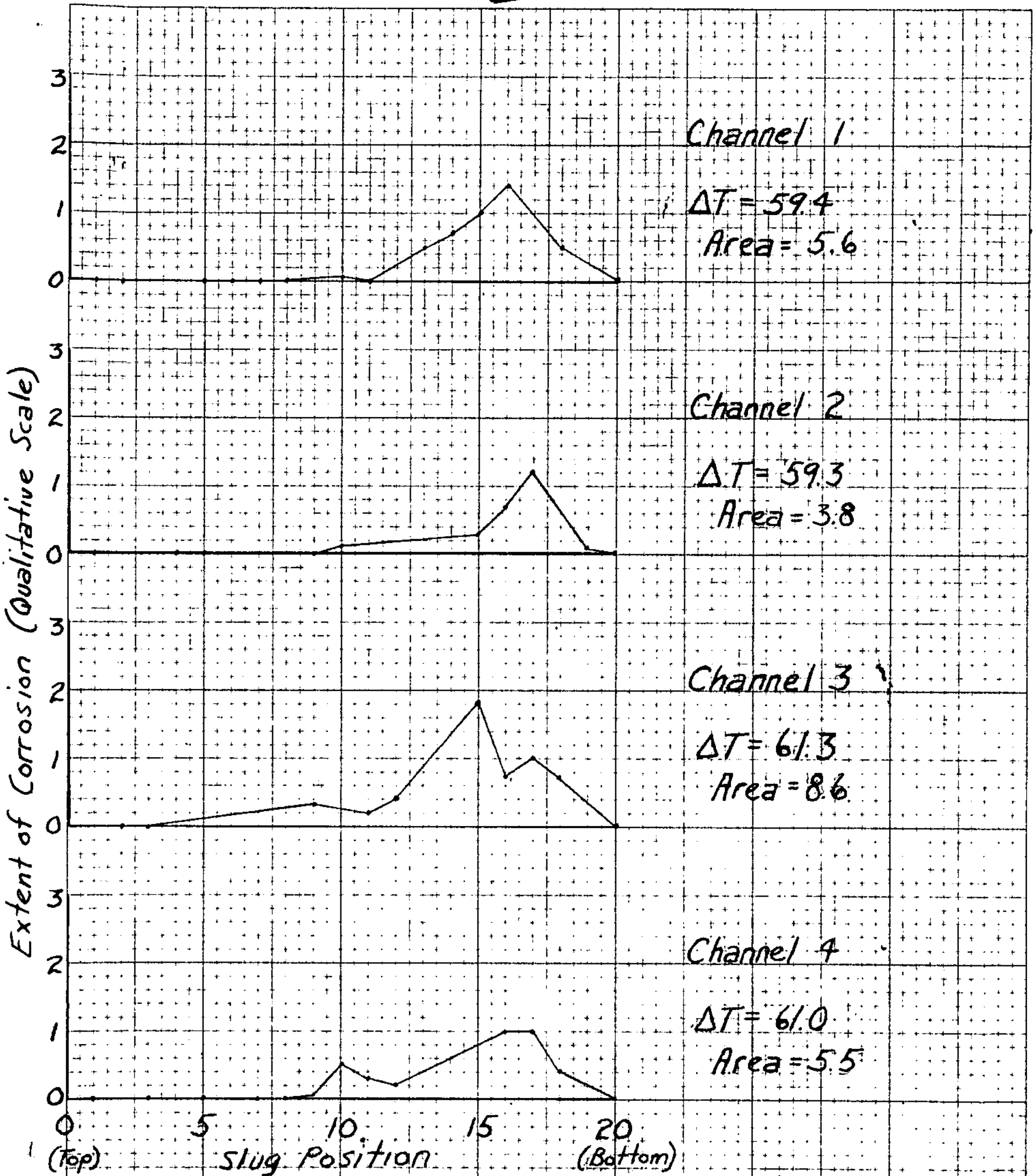

Figure 12. Pitting Corrosion
P-4 Fuel Element 24-60

[redacted]

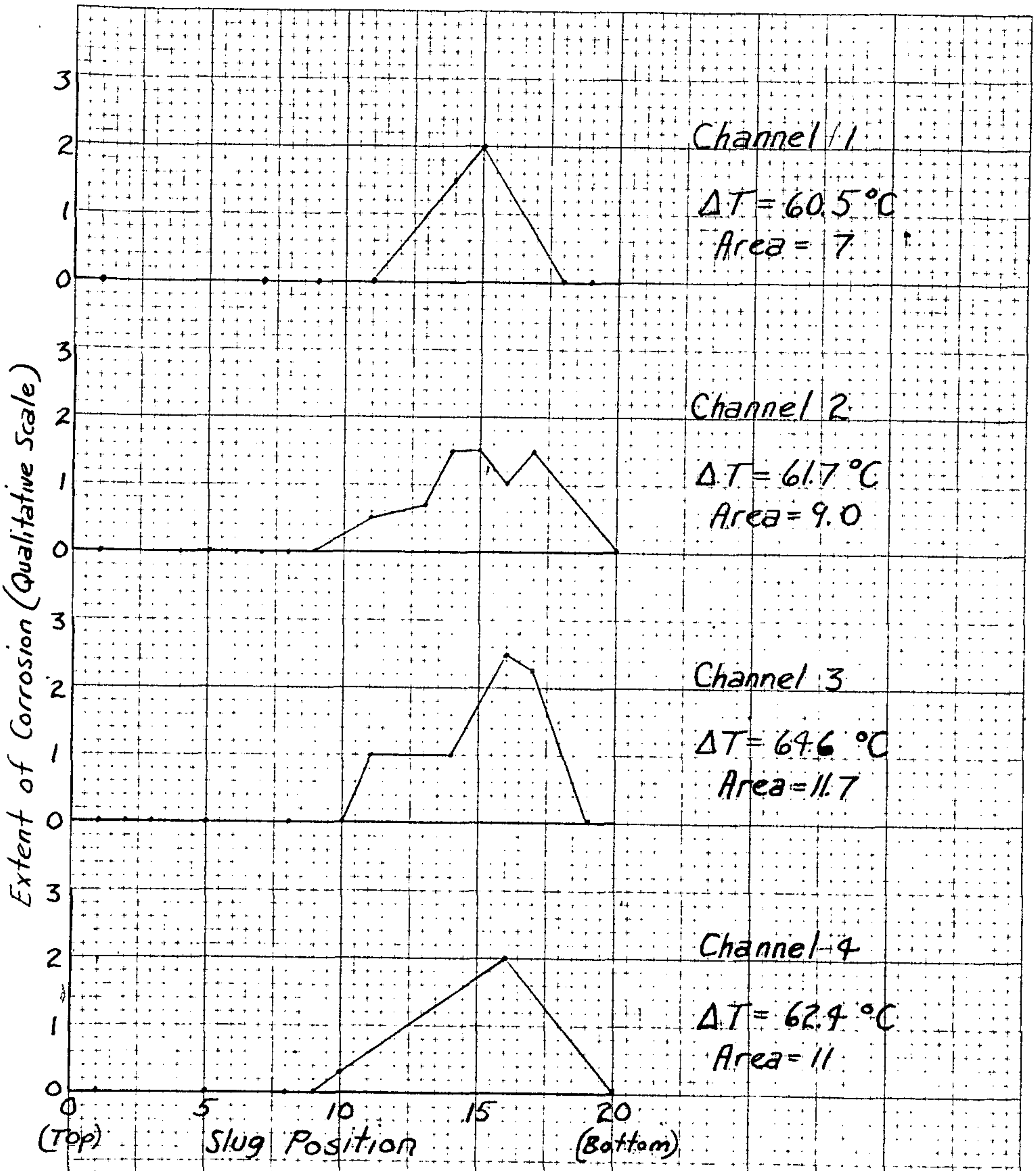

Figure 13. Pitting Corrosion
P-4 Fuel Element 29-51

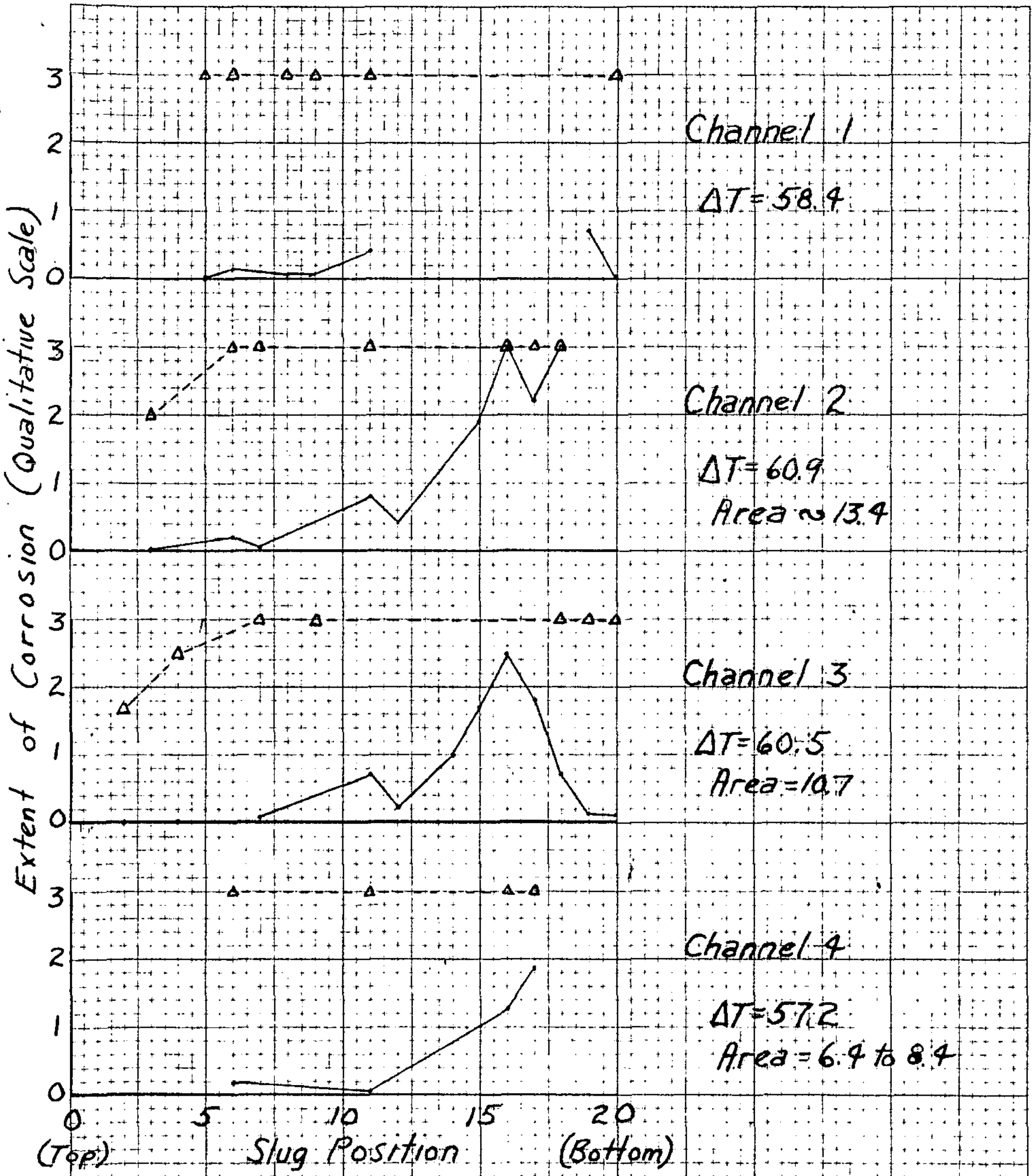

Figure 14 Extent of Corrosion
P-4 Fuel Element 17-57

[redacted]

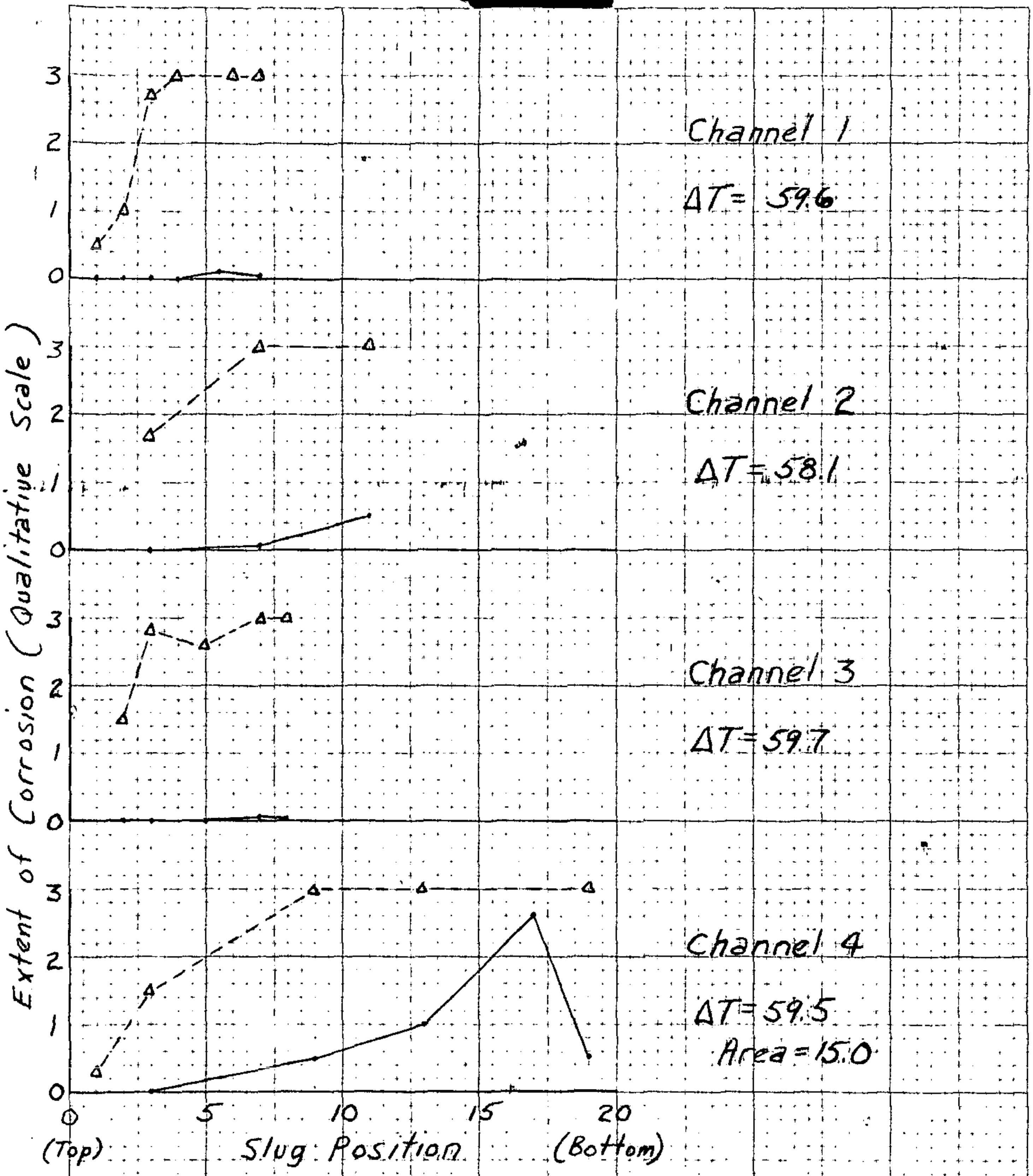

Figure 15. Extent of Corrosion P-4 Fuel Element 21-57

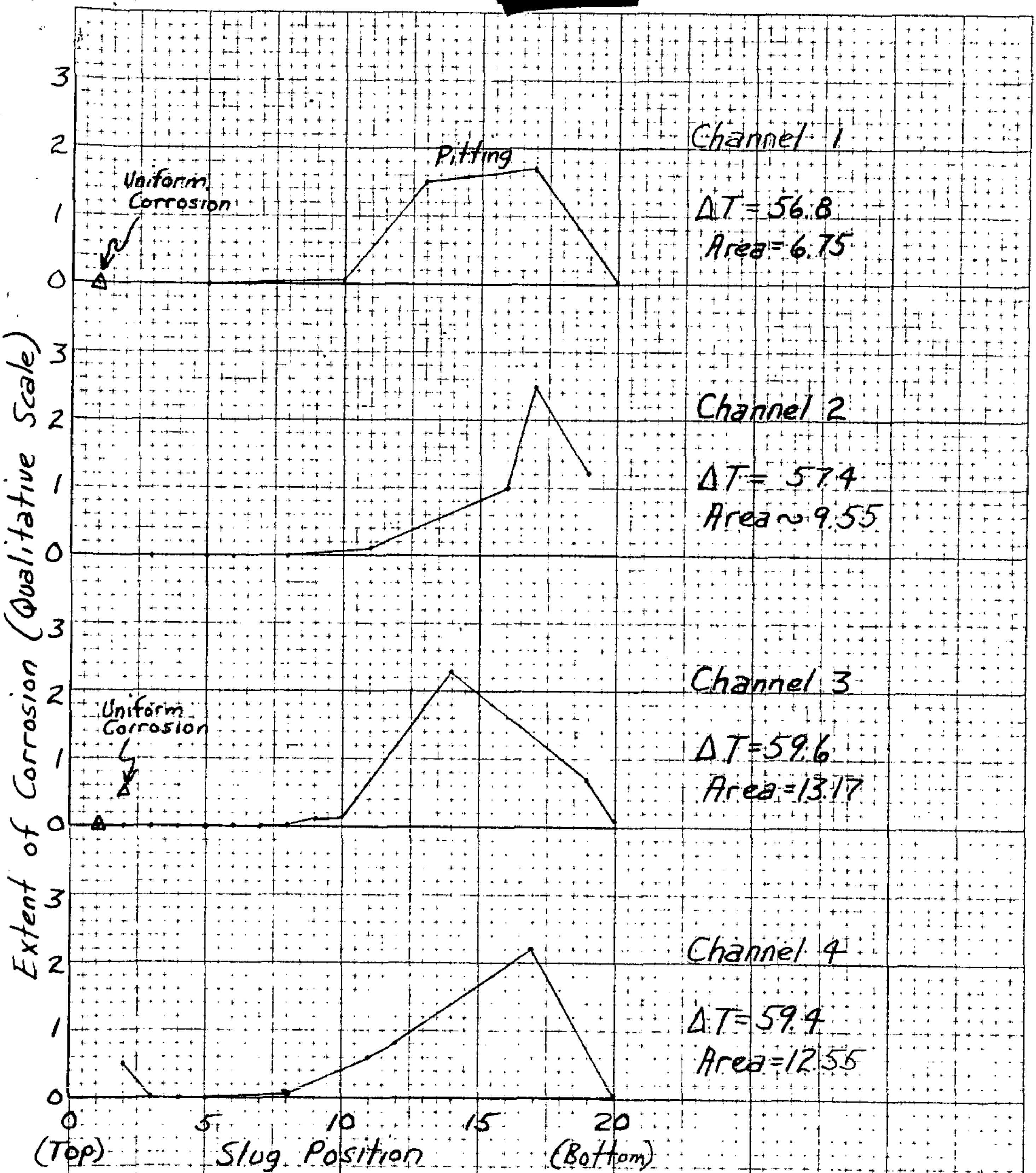

Figure 16. Pitting Corrosion
R-3 Fuel Element 34-48

[redacted]

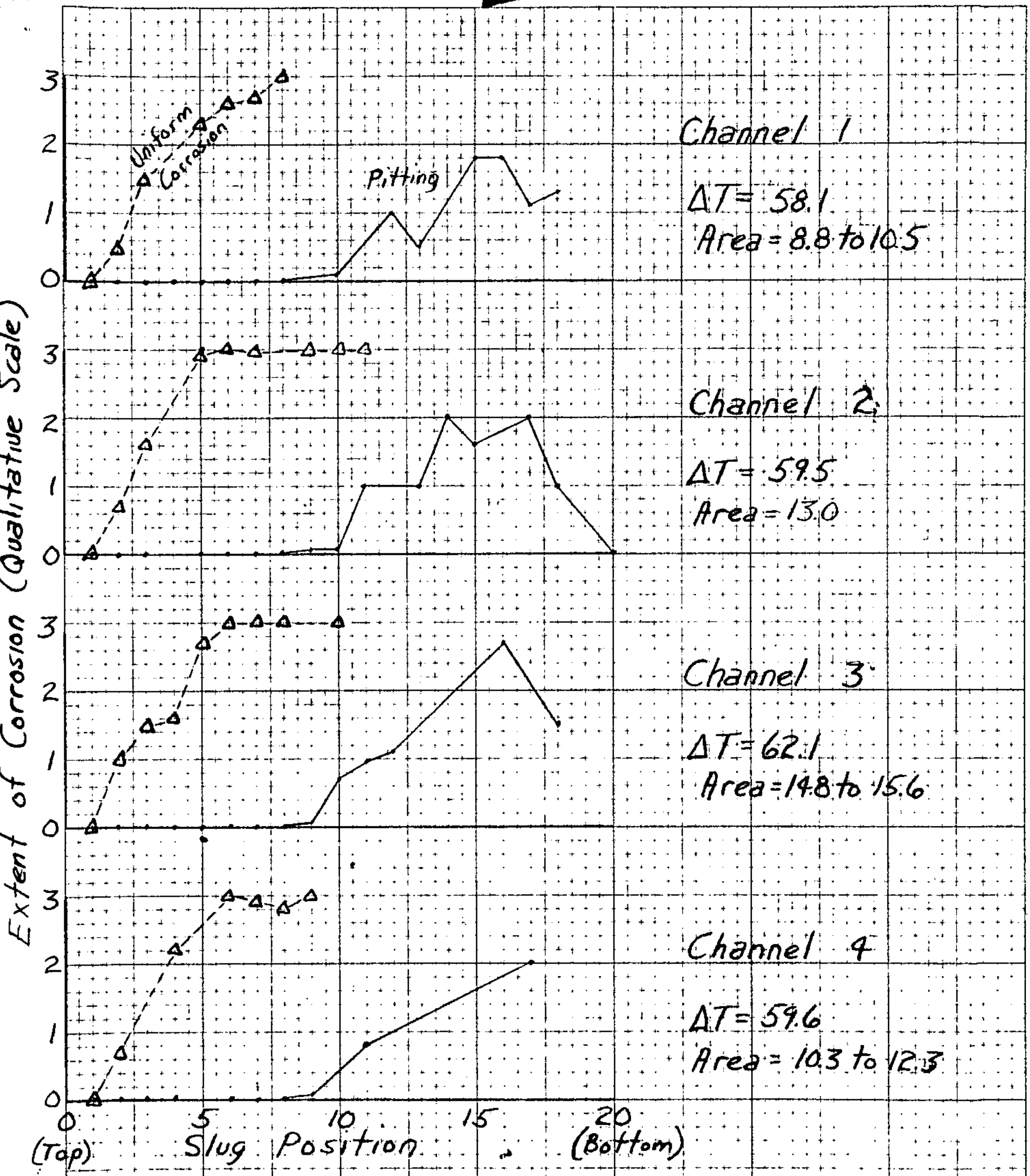

Figure 17 Extent of Corrosion
R-3 Fuel Element 27-27

[redacted]

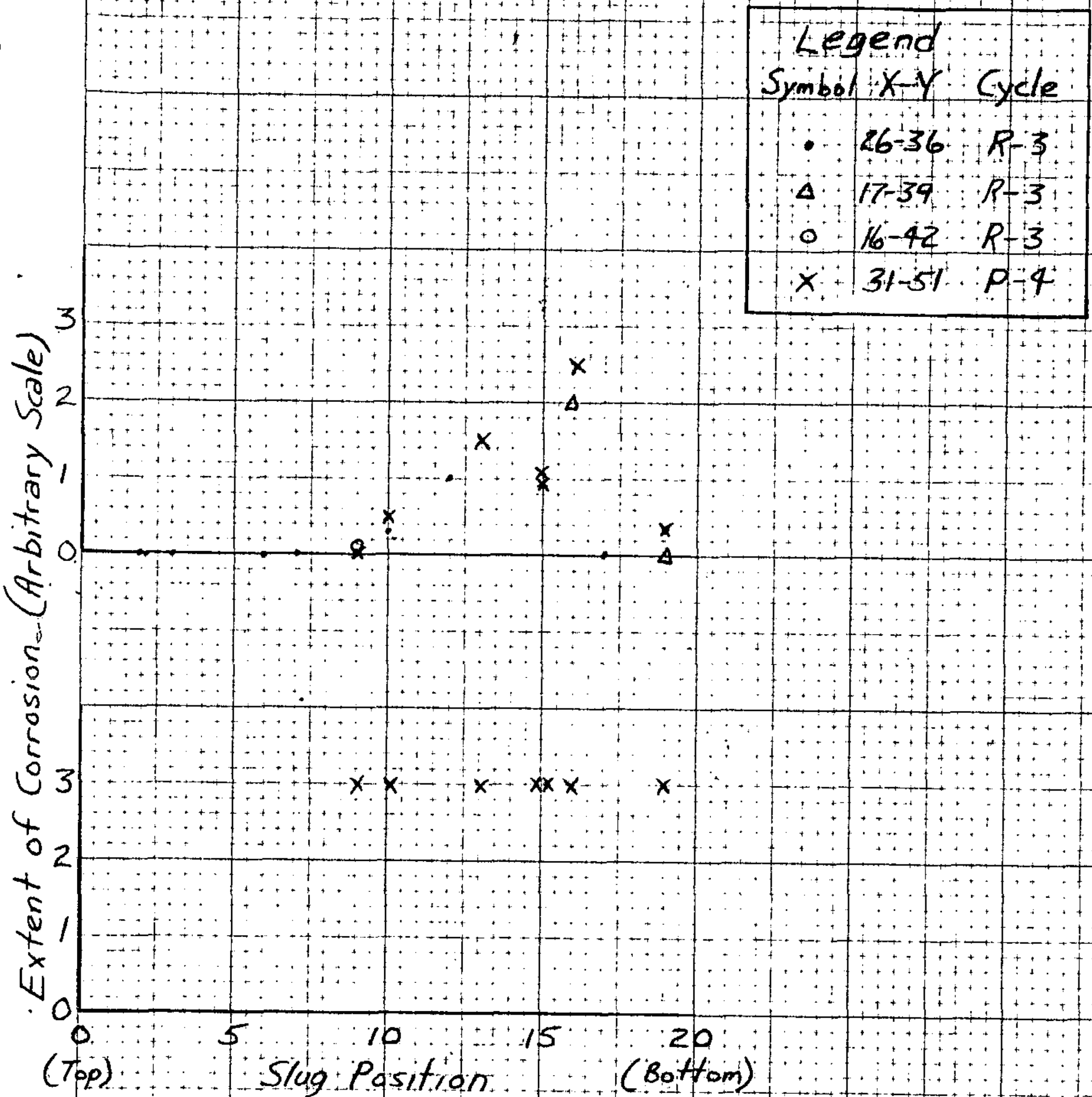

Figure 18. Miscellaneous Fuel Elements

[redacted]

TABLE I

P-4 FUEL ELEMENT 15-15
(Outer Ring - Mk I)

Serial No.	Channel & Position	Film Thickness †	Pitting *	Bumps
—	2-20	uniform light white		
—	-07	" " "		
—	3-20	" " "		
—	1-10	" heavier "		
—	1-11	" " "		slight
—	2-19	" light "		
—	2-18	" " "		faint
—	2-13	- -		medium
—	2-14	uniform medium white		slight
—	1-09	" " "		moderate
—	2-09	" " "		"

* Pitting was not recognized until later. It is fairly certain that none was present in any of these cases.

† What was being observed here was the same thing later recognized as thickness of the film resulting from uniform corrosion. It was being recorded with the purpose of estimating the film thickness, and is probably a fairly good measure of the quantity which was subsequently graded numerically. All slugs would have been rated 7 to 9 on the numerical scale, as based on remark recorded January 27, 1959 when this scale was first set up.

[redacted]

A. A. [redacted]INSON   TABLE II   P-4 FUEL ELEMENT 29-51
SECRET Page 30   (Flat Zone Mark VII-A)   RTR-146

Serial No.	Channel and Position	Film Thickness A	Film Thickness B	Pitting On Stand	Pitting At Scope	Bumps	Remarks
9N03090	4-05	10				✓	
9N03024	3-08	11				Less	
	1-01	8				0	Film Uniform Medium
	4-08	9				Some	
	2-05	8.5				✓	
	1-09	9				✓	
	1-11	10				✓	
	3-14	9		✓		✓	(Pitting First Seen)
	3-11	9		✓		✓	
	2-17	8.5		Big Patch			Film Flaking Off?
	2-11	9.5		Few Streaks		✓	
	4-20	11		0		Less	
	1-14	8		✓	✓	✓	Small Pock Marks
	1-15	9		✓	Many	✓	Small Pock Marks
	2-20	9.5				Not Much	Clean { Indicates no
	2-07	9.5				Not Much	" Stain but also
	2-13	9-		Some			
	1-19	9		0			" by Inference,
	4-09	10					" Absence of Pits
	4-20	10.5					
	2-15	9-		Medium	✓		Some "Speckling" ✓
	2-01	9					Very Uniform Film
	3-19	9.5				Some Low	
	4-10	9+		Faint			
	2-08	9					
	3-05	11	Heavy			Some	
	3-03	10.5		0	0	Some	Med. Heavy White Film
	2-04	10+		0	0	"	" " " "
	2-09	11		0	0	"	" " "
	2-14	10-		1	2	✓	" " "
	3-16	9		2	3	✓	
	3-10	11		0	0	✓	Heavy White Film
	1-18	11		0	0	✓	" " "
	3-02	11		0	0		" " "
	3-01	11		0	0	Few Faint	" " "
	1-07	11		0	0	Some	Heavy White Film
	2-16	10		1	1	✓	
	4-16	10		2	2	✓	
	4-01	10+		0	0	Faint	Med. Heavy White
	2-06	11		0	0	✓	" "

Note: Remark "heavy white film" toward end of table probably meaningless. At this time attention was concentrated on pitting, so that the estimate of film thickness was distorted.

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TABLE III

P-4 FUEL ELEMENT 24-60
(Flat Zone Mark VII-A)

1-28-59

Serial No.	Channel and Position	Film Thickness A	Film Thickness B	Pitting On Stand	Pitting At Scope	Bumps	Remarks
	3-15	10		2	1 to 2	✓	Film Thickness 11 before pitting
	1-01	10		0	0	0	Medium White Film
	1-02	10		0	0	Slight	Medium White Film
	2-10	10		?	0	✓	Not Much Pitting
---4356	3-17	9.5		1	1	Some	Light Pitting Allover
---0117	2-19	9.5		?	0	Slight	Perhaps Faint Pitting
---0153	2-20	10		0	0	"	
---0124	1-16	9.5		1	1 to 2		Medium Pitting All over
	1-15	10		1	1		Light Pitting All Over
?4333	1-05	10		0	0	Faint	Clean White
0125	2-17	10		1	Heavy?	✓	Pitting on One Side
0112	2-15	10		0.5	0	Some	Light Pitting on One Side
4366	4-20	10		0	0	0	Clean White
0126	1-18	10		1	0	Some	Faint Overall Pitting
4333	1-05	10		?	0	Faint	Little or No Pitting
1231	3-18	9.5		1✓	Faint	Some	Pitting - One Side Only
0146	2-18	10		1	0.5	Some	Pitting Somewhat on One Side
0147	1-20	9✓		0	0	0	Clean White
--04331	4-18	10✓		0.7	0	Some	One Side Only
4305	3-12	10		0.7	0	✓	LSW*
0144	2-05	9.5		0	0	✓	Clean White
0156	2-09	10		0	0	Low	Clean White
1222	3-03	10		0	0	Faint	Clean White
1217	3-16	10		1	?	✓ Low	LSW
0113	2-14	10		0.5	1	--	LSW and General Pitting
0122	2-16	10		1	0	✓	LSW
4322	3-20	10		0	0	Faint	Clean White
1274	1-08	10		0	0	Some	Clean White
0123	1-13	10		0.5✓	0.5	✓	Overall-More LSW
4365	1-11	10		0	0	Slight	Hint of Faint Pitting
4325	4-03	10		0	0	Faint	Clean White
0111	2-04	10		0	0	Faint	Clean White
2755	3-02	10		0	0	Very Faint	Clean White
4351	4-07	10		0	0	Some	Clean White
0157	2-01	10		0	0	0	Clean White
1215	4-01	9.5		0	0	0	Clean White
4314	3-11	10		0.3	0	0	Faint Pitting LSW

[redacted]

TABLE III (Continued)

Serial No.	Channel and Position	Film Thickness A	Film Thickness B	Pitting On Stand	Pitting At Scope	Bumps	Remarks
---4324	1-06	10		0	0	Low	Clean White
4350	3-09	10		0.3	0.3	✓	LSW
4348	4-10	10		0.3	0.7	✓	Faint to Medium Light Pitting
4372	4-09	10		0?	0	Low	Clean White (Faint Pitting?)
1284	4-11	10		0.3	0.3	✓	LSW
1296	1-10	10		0✓	0✓	Low	Some Hint of Pitting
4310	1-04	10		0	0	Slight	Clean White
1268	1-07	10		0	0	✓	Possible LSW**
1249	4-05	10		0	0	Some	Clean White
4319	4-16	10		1	1	✓	Overall Large Pits
4347	4-17	10		1	?	✓	LSW (Some noted at scope)
4369	4-12	10		0.3	0	✓	Faint LSW
4329	4-08	10		0	0	✓	Clean White

------

* LSW = Pitting concentration on "low" (concave) side of warp of the slug.

** Probably just the greater reflection from concave side of warp on unpitted slugs which was later noted in R Area.

Note: At this time it had been decided that apparent film thickness was of little consequence and recording of observations pertaining to film condition became perfunctory. Any variations noted must have been extreme.

[redacted]

TABLE IV
R-3 FUEL ELEMENT 26-36

1-28-59 (2:00 PM)

Serial No.	Channel and Position	Film Thickness A	Film Thickness B	Pitting On Stand	Pitting At Scope	Bumps	Remarks
---6659	2-10	9		0.5	0	0	LSW (Poor Lighting)
---6656	1-07	9		0	0	0	
---6684	3-12	9		1	1	-	LSW
---6649	3-17	9		0	-		Clean White (poor Light)
6664	2-06	9		0	0	0	Clean White
6611	3-03	9		0	0	0	Clean White
6607	1-02	9		0	0	0	Fairly Clean White
6625	2-02	9		0	0	0	*
----							

* Lighting appears different on opposite sides of slug warp.

TABLE V
OBSERVATION OF R-3 PHOTOGRAPHED SLUGS

1-29-59 (11:00 PM)

Serial No.	Channel and Position	Coordinate X - Y	Pitting On Stand	Pitting At Scope	Remarks
03N33567	4-09	16-42	---	0	Eggshell Texture around bumps; seen as pits through periscope.
03N02912	3-19	7-39		0	Faint texture (as from pitting)
03N01664	3-16	17-39	2/	1.5	Under periscope, a uniform covering of closely spaced pits or pock marks (about 5-10 mil diameter) was seen.
----2908	---	----		0	Some texture (as from pitting).

[redacted]

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TABLE VI

R-3 FUEL ELEMENT 34-48

1-29-59(9:00 AM)

Serial No.	Channel and Position	Film Thickness A	B	Pitting On Stand	At Scope	Bumps	Remarks
	3-03	9		0	-		
---1962	3-06	9.5		0	0		
---1900	3-07	-		0	0		
1943	3-08	10		0	0	Low	
1973	3-19	10		1	0.2		LSW
4863	4-11	10		0.3	1	Low	
1915	1-05	10		0	0	Faint	Clean White
3661	4-02	?		0.5	0.5		
1967	1-13	9.5		1	2		LSW
3668	4-12	9.5		0.5	1		Large Pock Marks
1949	3-14	9.5		2.5	2		LSW
4885	4-08	10		0	0	✓	Faint texture on clean white
1982	1-20	10		0	0	0✓	Clean White
4875	4-17	9		1.5	3	✓	
4858	2-06	10		0	0	✓	Clean White
4801	4-05	10		0	0	✓	Clean White
1994	2-17	9		2	3	✓ Low	Heavy Overall Pitting
1922	3-04	10		0	0	✓ Low	
1975	2-19	10-		1	1.5	✓	LSW
4874	3-05	10		0	0	Low	Clean White
4813	4-20	10		0	0	Faint	
1995	2-08	10		0	0	✓	Clean White *
4862	4-03	9.7		0	0	Faint	Clean White
4826	3-20	10		0.?	0.?	0	Faint texture
1954	2-16	-		1	1		Pitting heavier on LSW
1955	3-10	10		0.2	0.1	Low	
3517	3-02	10.?		0	0		(Film very thin and patchy)
4869	2-11	10		0.1	0.1	✓	Faint texture
1987	3-09	10		0.1?	0.1?	---	Faintly brighter patches
1916	1-17	9.5		2	1	✓ Low	Mottled, more on LSW
1964	3-01	8		0	0	0	Mottled Sheen **
4800	1-01	8		0	0	0	"
4823	4-04	10		0	0	-	Clean white
----	2-05	-		0	-		
0335	1-10	10		0.1	0		
1934	2-03	10		0	0	0	

----

* Reflected light much brighter on concave side of warp.

** Technicians remarked that this slug looked " as good as when it went in the reactor." It was at this point that gradation of film thickness was first vaguely recognized.

[redacted]

TABLE VII
R-3 FUEL ELEMENT 27-27

1-29-59 (1:30 PM)

Serial No.	Channel and Position	Film Thickness A	Film Thickness B	Pitting On Stand	Pitting At Scope	Bumps	Remarks
01K45318	2-17	9	-	2	2	Low	
01K50191	2-10	10	3	0.1	0	Some	Some Visible Pits
6479	4-08	10	2.8	0	0	0?	
6310	1-08	10	-	0	0	Faint	
6372	2-13	9	-	1	1	√	More on LSW (pits visible)
9420	2-18	9	-	1	1	Low	LSW (pits visible)
01K46404	3-08	10√	3	0	0	Some	
0988	1-18	9	-	1	1.5	Low	Many Visible pits
2219	4-06	10	3?	0	0	Faint	
6300	3-18	9	-	2	1	Low	Mottled
9429	2-20	9.5	0	0	0	0	
2564	1-03	9.5	1	0	0	0	Almost Like New
9415	1-05	10	2	0	0	Some	
9136	3-03	9.5	1	0	0	0	
6453	3-10	10	3	0.5	1	Some	Some Visible Pits
9123	4-04	9.7	2	0	0	Faint	Many Visible Pits
6380	1-06	9.9	2.5	-	0	Some	
2675	2-11	9.5	3	1	1	Low	
6455	3-01	8	0	0	0	0	Like New
6813	2-01	8	0	0	0	0	Like New
6463	4-06	9.5	2.5	0	0	0	Like New Except Heavy Film
6338	4-07	9.7	2.9	0	0	Some	Like New Except heavy Film
5482	2-09	10	3	0.1	0	Little	
6384	1-07	9.7	2.7	0	0	Some	Like New Except Heavy Film
6343	2-14	9	-	2	2		
6835	2-02	8.5	0.5	0	0	0	Almost Like New
6381	2-15	9	-	2	1.5	Low	
6309	3-12	9.5	-	1	1-	√	
2641	3-13	9	-	1.5	0.7	√	
5488	2-08	10	2.95	0	0	Some	
1668	4-09	10	3	0.1	0	Some	
2832	4-11	9.7	-	0.5	1	Some	
2612	4-17	8	-	2	2	√	Mottled
6373	2-06	10	3	0	0	√	
6431	3-16	8	-	2.5	3	√	
6450	3-09	10	-	0.1	0	Low	

[redacted]

Table VII (Continued

Serial No.	Channel and Position	Film Thickness A	Film Thickness B	Pitting On Stand	Pitting At Scope	Bumps	Remarks
01K4907	3-06	10+	3	0	0	Slight	
6678	3-05	9.7	2.7	0	0	0	Like New Except Heavy Film
3513	1-17	9		1+	1	0	Some Visible Pits?
6382	1-02	8.5	0.5	0	0	0	Like New, Less Lustre
5248	1-15	8		1.5	2	Some	Visible Pits
1940	1-01	7	0	0	0		Like New
6321	2-05	10	2.9	0	0	✓	
1586	2-07	9.7	3	0.1	0.1	✓	Faint Sheen (nature doubtful)
5253	3-04	9	1.5	0	0		Almost Like New
0967	4-01	7	0	0	0		Like New
2648	3-07	10-	3	0	0	Low	
6305	4-03	8	1	0	0		Almost Like New
2658	2-03	9	1.5	0	0		Almost Like New
5243	1-16	8-		2.5	1		Visible Pits
0911	4-02	9.7	0.5	0	0		Almost Like New
6423	1-10	10		0.2	0		
6454	3-02	9	1	0	0		Almost Like New
6367	1-12	8.5		1	1		Mottled
2647	1-13	9		0.5	?		
5752	1-04	9	1.5	0	0		Almost Like New

[redacted]

TABLE VIII

P-4 FUEL ELEMENT 17-57 2-2-59

Serial No.	Channel and Position	Film Thickness A	Film Thickness B	Pitting On Stand	Pitting At Scope	Bumps	Remarks
----2725	2-17	9(10)		2.5	1.5		LSW; visible pits
2723	2-15	8.5		3	1-		
2767	1-20	10	3	0	0	√	No visible pits
2734	2-97	10	3	0.1	0	√	
2762	1-06	10	3	0.2	0.1	√	Corrosion sheen around bumps
2747	1-11	10	3	0.3	0.5	Some	
2733	2-16	8(10)		3	3		LSW; heavy visible pits
2739	2-11	9.5(10)		1	0.3		
2716	1-08	10	3	0.05	0.05	Low	
2627	4-16	10		2	?	√	LSW
2707	2-06	10	3	0.3	?	√	Corrosion sheen around bumps
2759	3-19	10	3	0.1?	0	Low	Corrosion sheen around bumps
2740	2-18	8(10)	3	3	3		Corrosion sheen around bumps; LSW
2764	1-05	10	3	0.5	0.1		Some sheen-nature doubtful
2768	3-16	8.5		2.5	2		Visible Pits -"3"
1277	3-18	(10)		0.7	0.5		Visible Pits - "1"
2753	2-14	--		2.5	--		LSW
2650	3-07	10		0.1	0		Faint sheen around bumps
2743	3-17			1.5	2		Mottled
2669	3-12	10		0.3	0		Pitting around bumps
2663	3-02	9	1.5	0	0	0	Almost Like New
2719	1-09	10		0	0		Some sheen around bumps?
2756		10	2.9	0	0	Low	
2686	4-11	--		0.1√	0	Some	Sheen around bumps
2642	3-15	9.7		1.5 to 2	1.5 to 2		
2649	4-12	10		0.5√	?	√	Pitting Around bumps
2763	3-20	10		0.1	0.05		Pitting around bumps
2727	1-19	9.5		1	0.3		LSW
2742	3-11			0.5	1		LSW
2705	2-03	9	2	0	0	Slight	Almost Like New
2702	3-04	9.5	2.5	0	0	Some	*
2696	4-06	10		0.3	0	√	Pitting around bumps
2694	4-17	8(10)		2-	2-	√	Pitting around bumps; LSW

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* Brightness around bumps suggests that light reflection is brighter here, in absence of pitting.

[redacted]

TABLE IX
P-4 FUEL ELEMENT 21-57

2-11-59 (1:00 PM)

Serial No.	Channel and Position	Film Thickness A	Film Thickness B	Pitting On Stand	Pitting At Scope	Bumps	Remarks
----8769	3-05	-	2.6	0	0	Some	
8767	3-08	-	3	0.05	0	Slight	Sheen Around Bumps
3215	1-07	-	3	0/	0/	Few	
8711	3-07	-	3	0.05	0.0/	Few	
1878	4-17	-	-	2.5	2	Some	
1816	1-02	-	1	0	0	-	
	4-03	-	1.5	0	0	0	Almost Like New
8714	3-02	-	1.5	0	0	0	Almost Like New
8784	2-03	-	1.7	0	0	Faint	Almost Like New
8781	4-09	-	3	0.5	0.5	√	LSW
8703	2-07	-	3	0.05	0.05	Some	LSW (?)
3288	1-06	-	3	0.1	0.1	Slight	LSW
1886	1-04	-	3	0	0		
1882	4-13	-	3	1	1		LSW
1849	1-03	-	2.7	0	0		
8740	3-03	-	2.8	0	0		Clean Heavy White
1829	4-19	-	3	0.5	0.5	Some	
8730	2-11	-	3	0.5	-		
8756	3-01	-	0.5	0	0	0	Almost Like New
8792	4-01	-	0.3	0	0	0	Almost Like New

TABLE X
P-4 FUEL ELEMENT 31-51

2-11-59 (3:00 PM)

Serial No.	Channel and Position	Film Thickness A	Film Thickness B	Pitting On Stand	Pitting At Scope	Bumps	Remarks
----0433	1-10		3	0.5	0.5	Low	Sheen around bumps
2189	4-15		3	1	1 to 1.5	Low	Visible pits
0402	1-15		3	1-	~1	Low	LSW
4639	1-13		3	1	2		LSW*
2134	4-19		3	0.5	0		LSW
4656	3-09		3	0.05	0	0	Heavy Film
0621	1-16		3	3	2		**

----

* Heavy pitting in a narrow band on LSW. Rest of slug fairly free of pitting.

** Heavy white incrustation in patches. Consists of small white conical mounds ("anthills") about 20 mils in width and height.

[redacted]

TABLE XI
AVERAGE EXTENT OF CORROSION
(Arbitrary Units)

Slug Position	Pitting: Flat Zone R-2*	Pitting: Flat Zone R-3	Pitting: Flat Zone P-4 †	Pitting: Outer Ring P-4	Uniform: Flat Zone R-3	Uniform: Flat Zone P-4	Uniform: Outer Ring P-4
1		0	0	None	0	(1)	
2		0.06	0	None	0.68	1.7	
3		0	0	None	1.53	2	
4	0	0	0	None	1.9	2.5	
5	0	0	0	None	2.63	2.8	
6		0	0.11	None	2.9	3	0
7	0	0	0.02	None	2.89	3	
8		0.01	0.01	None	2.93	3	1.5
9		0.05	0.14	None	3	3-	0.5
10		0.22	0.3	None	3	2.9	0.5
11		0.5	039	None	3	3	
12	0.1	0.94	0.45	None			
13		1.02	0.98	None			
14		2.15	2.08	None			
15	0.2	1.8	2.09	None			1.5
16	0.5	1.82	2.54	None		3	
17	0.7	1.92	2.42	None		3	
18	1	1.4	1.15	None		3	0
19		0.63	0.24	None		3	0
20		0.01	0.013	None	2.5	3	0

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* Only one slug for each position; data from examination of photographs of discharged slugs.

† Data for reactor positions 29-51 (Table II) and 24-60 (Table III) multiplied by 2 to correct for shift in grading scale.

TABLE XII

ESTIMATED SHEATH TEMPERATURES, °C*

Position	Flat Zone R-2	Flat Zone R-3	Flat Zone P-4	Flat Zone K-2	Outer Ring P-2	$(T_t - T_b)$** R-2
1	57	58	64	-	51	11.5
2	68.5	65	77	78	57.5	11.0
3	79	80	92	88	64	10.5
4	89	90	101	106	71	9
5	95.5	100	105	123	75	5
6	99	105	108.5	127	76.5	3
7	102.5	108.8	112	130.5	78	3
8	105	111.8	115.2	134	79.7	3.3
9	107.5	114.5	118.8	137	81	2.4
10	109.8	117	122	141	83	2.3
11	112.0	120.2	125	144	84.2	2.1
12	114.5	123	128	148	85.3	2.5
13	117	126	131	151	86.8	2.5
14	119	128	133	153.2	88	1.9
15	120.8	130	134.5	154.2	89	1.4
16	122.0	130	135.5	151.2	90	0.6
17	121.0	128	133	142.5	89.5	-3.2
18	116.0	122	126	134	86.3	-5.4
19	110.7	114	119	120	83.5	-5.4
20	105.0	102	108	113	80.2	-6.0

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* Sheath temperatures calculated for graphs prepared for this purpose by J. M. Boswell. Values calculated for outer surface of fuel. Mark VII-A values take into account observed flow split and monitoring efficiency.

** Difference in sheath temperature, top of slug minus bottom of slug (measure of difference in extent of corrosion to be expected between two ends of slug).

EXHIBIT A. THEORY OF TURBIDITY GENERATION

Uniform Corrosion

Uniform corrosion of aluminum occurs in two stages (a) an initial rapid buildup of the film and (b) a subsequent slow constant rate stage. The second stage has been studied in some detail (Ref. 4). The constant rate increases with temperature according to the Arrhenius law. The first stage has not been studied very carefully. The extent of corrosion during this stage appears to increase with temperature, but the laboratory results are erratic.

We will assume, for lack of any better data, that the extent of corrosion during the first stage varies with temperature in the same way as does the subsequent constant rate. This assumption is to some extent justified by the fact that the total extent of uniform corrosion in the reactor at the end of 50 days varies with temperature in just this way (Figure 4; the slope of the straight line is that previously found for the constant rate period - Ref. 7, 11). Since the major part of the total corrosion occurs in the initial period, the variation with temperature shown in Figure 4 characterizes mainly this initial period.

The total extent of corrosion of 2 S aluminum up to any time (0 to 20 days) at 125°C as given in Reference 4 is taken as the basis for estimating the extent of uniform corrosion at any time and temperature. This basic extent of corrosion, C, is tabulated below:

Time (day)	0.5	1	2	5	10	20	30	40	50
C (mg/dm^2)	4	8	12	13.5	14	15	16	17	18

At temperatures other than 125°, the foregoing assumption gives the extent of uniform corrosion as proportional to $W = C \times 10^{-3300/T}$. The quantity W is a measure of the thickness of oxide film built up, expressed in arbitrary units.

Pitting Corrosion

Pitting corrosion is assumed to start when this oxide film reaches a critical thickness (hence when W reaches a critical value, W_c). Inspection of both slugs and fuel tubes as reported here, shows that aluminum exposed at 121°C has just begun to pit at the end of 50 days. This gives for W_c the value 3.76×10^{-6}, corresponding to a critical film thickness of 1.3 mil (266 mg of Al corroded per dm^2).

The time required to reach the point of incipient pitting may be estimated from the above assumptions, as described below. First the extent of corrosion C_c which would be attained after this same period of exposure at 125°C (instead of the actual temperature T) is calculated

$$C_c = W_c \times 10^{3300/T} \qquad (1)$$

Then the number of days required to attain this extent of corrosion at 125°C is estimated from a plot of C vs. time which represents the basic data tabulated above.

Diagram A in Figure 7 represents the results of such a calculation for a typical R-3 P-4 flat zone fuel column. For each slug position along the column the value of C_c was calculated from equation 1 corresponding to the sheath temperature (Figure 1 and Table X) at that slug position. The times to reach these values of C were then plotted against slug position as shown.

Diagram B is a plot of the number of slugs lying between the two branches of the curve in Diagram A. All of these slugs have either reached or passed the point of incipient pitting, and hence show varying degrees of pitting.

Turbidity Formation

The basic assumption is that when pitting occurs, the previously formed oxide film is broken at the site of each pit, and the aluminum oxide which covered the area of the pit is released into the moderator as turbidity.

Let M be the total amount of oxide released per unit length of fuel column from any particular differential element of length dn of a particular fuel column. The unit of length is here taken to be the length of one slug. The total amount of oxide released into the moderator by the entire reactor charge is then

$$\text{Total release} = \sum \int M\,dn \tag{2}$$

where integration is carried over the entire length of one fuel column and summation is over all fuel columns.

Let us define the fractional extent of pitting p by the expression $M = p\,M_o$, where M_o is the value of M for a "completely pitted" surface. The value M_o is so chosen that p is always less than, but approaches unity for severely pitted surfaces. The value of p differs significantly from zero only in the flat zone and within that portion of each fuel column for which pitting has started.

The total release is expressed in terms of p as

$$\text{Total release} = \sum M_o \int p\,dn$$

$$= \sum M_o \quad \bar{p}n$$

where n is that length of the particular fuel column on which pitting has started, and $\bar{p}$ is the average extent of pitting over this length.

Inspection of slugs shows that $\bar{p}$ and n are roughly constant throughout the flat zone. Hence $\sum M_o$ pn for the flat zone alone has the value $M_o\ Z\ \bar{p}\ n$, where Z is the total number of fuel columns in the flat zone. The contribution from fuel columns outside the flat zone is assumed to be negligible because of much lower sheath temperatures. Hence

$$\text{Total release} = (M_o\ Z)\ \bar{p}\ n \tag{4}$$

$$\text{Rate of release} = (M_o\ Z)\ d\ (\bar{p}\ n)/dt \tag{5}$$

Note that the coefficient $(M_o\ Z)$ is nearly constant at least throughout any one reactor cycle.

In Figure 7 the heavy curves represent the case in which $\bar{p}$ is assumed to be unity, while the dashed curves result from a crude estimate of the actual value of $\bar{p}$. Diagram B is a plot of $\bar{p}$n against t, while Diagram C is a plot of $d(\bar{p}n)/dt$ against t (obtained as the slope of the corresponding curve in Diagram B). These curves give respectively the total amount of turbidity formed and the rate of turbidity formation in arbitrary units such that one unit represents an amount $M_o\ Z$ of oxide released (per unit time in the case of Diagram C).

In R and P reactors, which have no means of turbidity removal, the total amount of turbidity released remains in the moderator. Thus Diagram B also represents (in arbitrary units) the way in which turbidity should increase during a reactor cycle.

In L, K and C reactors turbidity is removed by the distillation reboilers. A material balance of turbidity generation and removal gives

$$V\, dT/dt = (M_o\ Z)\ d(\beta n)/dt - FT \tag{6}$$

where V is the total moderator volume, T is the turbidity level (amount of turbidity solids per unit volume of moderator) and F is the rate of flow of moderator through the reboilers. Integration of this equation gives

$$T = (M_o Z/V) e^{-(F/V)t} \tag{7}$$

The results of this integration are shown in Diagram D, with the factor $(M_o Z/V)$ taken as the arbitrary unit. Buildup and decay of turbidity in this fashion has been observed repeatedly in K and C reactors and frequently in L reactor.

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