

Probability Based Corrosion Control for Waste Tanks-Part II

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1 Executive Summary

As part of an ongoing study to evaluate the discontinuity in the corrosion controls at the SRS tank farm, a study was conducted this year to assess the minimum concentrations below 1 molar nitrate, see Figure 1. Current controls on the tank farm solution chemistry are in place to prevent the initiation and propagation of pitting and stress corrosion cracking in the primary steel waste tanks. The controls are based upon a series of experiments performed with simulated solutions on materials used for construction of the tanks, namely ASTM A537 carbon steel (A537). During FY'09, an experimental program was undertaken to investigate the risk associated with reducing the minimum molar nitrite concentration required to confidently inhibit pitting in dilute solutions (i.e., less than 1 molar nitrate).

The experimental results and conclusions herein provide a statistical basis to quantify the probability of pitting for the tank wall exposed to various solutions with dilute concentrations of nitrate and nitrite. Understanding the probability for pitting will allow the facility to make tank-specific risk-based decisions for chemistry control.

Based on previous electrochemical testing [1], a statistical test matrix was developed to refine and solidify the application of the statistical mixture/amount model to corrosion of A537 steel. A mixture/amount model was identified based on statistical analysis of recent and historically collected electrochemical data [2,3]. This model provides a more complex relationship between the nitrate and nitrite concentrations and the probability of pitting than is represented by the model underlying the current chemistry control program, and its use may provide a technical basis for the utilization of less nitrite to inhibit pitting at concentrations below 1 molar nitrate. FY'09 results fit within the mixture/amount model, and further refine the nitrate regime in which the model is applicable. The combination of visual observations and cyclic potentiodynamic polarization scans indicates a potential for significant inhibitor reductions at nitrate concentrations near 1.0 M without a significant increase in corrosion risk. The complete data sets from FY08 and FY09 testing have determined the statistical basis to confidently inhibit against pitting using nitrite inhibition with the current pH controls. Future testing will complete the spectrum of nitrate concentrations around 1 molar. These results will be combined to provide a complete spectrum for corrosion controls with a risk based component.

2 Introduction

Twenty seven compliant underground carbon steel tanks provide storage for radioactive waste at the DOE Savannah River Site (SRS). Over time, slight changes occurred in the grade of carbon steel used to build the tanks. The initial type III and IIIA primary tanks were constructed of ASTM A516 while the remaining type IIIA tanks were constructed from ASTM A537 Class 1 (A537). No known metallurgical difference exists between A516 and A537 that would impact corrosion behavior. An assessment of the potential degradation modes of the waste tanks determined that nitrate-induced pitting corrosion and stress corrosion cracking were the two most significant degradation modes. [4] Controls on the solution chemistry, see Figure 1, requiring minimum nitrite and hydroxide concentrations are in place to prevent the initiation and

propagation of pitting and stress corrosion cracking in the tanks [5]. These controls are based upon a series of experiments performed using simulated solutions on carbon steels used in construction of the tanks. [6] A detailed description of Figure 1 can be found in Part I of this study [1].

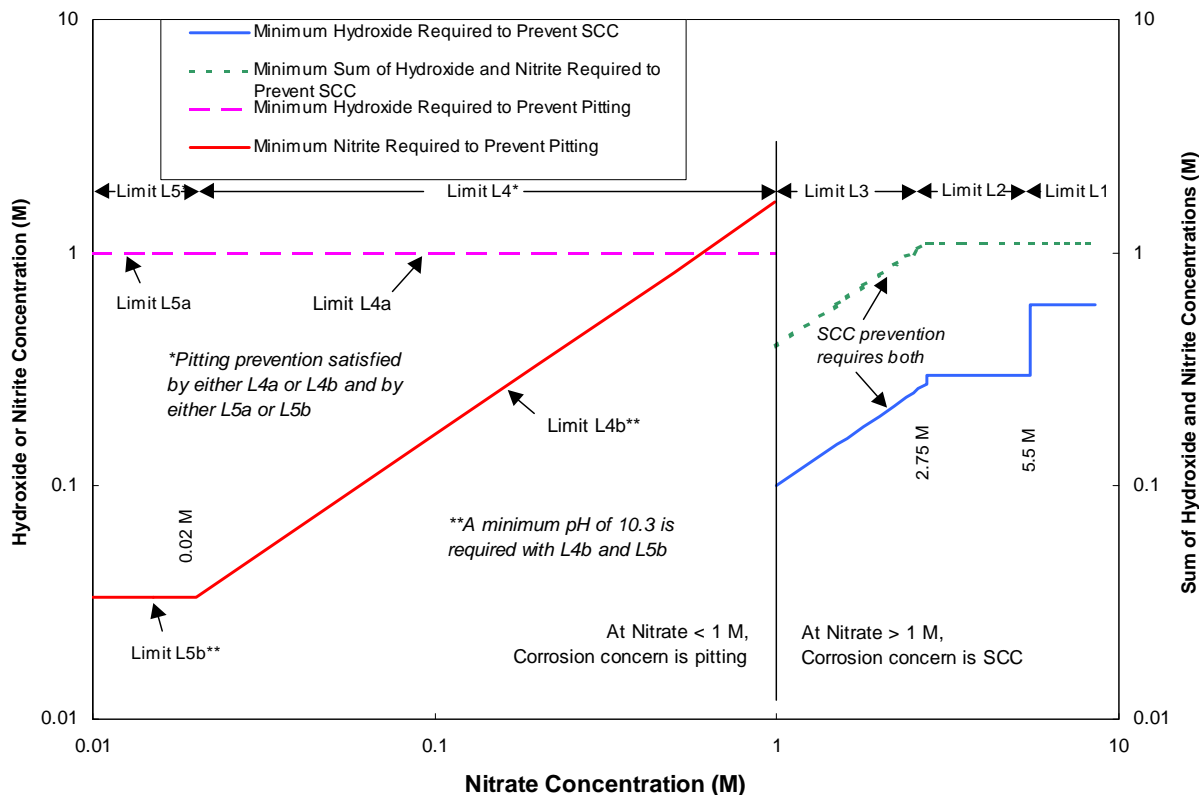


Figure 1 Inhibitor concentration limits under current chemistry control program.

In dilute waste solutions, nitrate-induced pitting, chloride-induced pitting, and sulfate-induced pitting are the corrosion modes of concern. [6] Based on the current chemistry control program, the minimum molar concentrations of nitrite required to prevent pitting in the 0.02 to 1.0 M nitrate concentration range at $T \leq 40^\circ\text{C}$ is:

$$[\text{NO}_2^-] = 1.66 \times [\text{NO}_3^-] \quad (1)$$

The limit is based on the results of cyclic potentiodynamic polarization (CPP) scans and coupon immersion tests. [7] In the event that the nitrate anion is in low concentration and is not the principal corrosive anion, minimum nitrite limits have been established based upon either the chloride or sulfate concentrations.

The chemistry control program has thus far been implemented by applying a safety factor on the data. The ideal chemistry control program would add the lowest volume of inhibitors to maintain the solution chemistry at a desired concentration to minimize corrosion vulnerabilities.

An experimental disconnect however exists between the chemistry control above and below 1.0 M nitrate due to a presumed change in dominant corrosion modes (i.e. pitting in dilute solutions and stress corrosion cracking in more concentrated solutions), as seen in Figure 1. The control limit below 1.0 M nitrate is based on a linear regression derived from experimental data primarily in the region of 0.5 M nitrate or less. It will be possible, and of particular benefit, to adjust the control program to minimize the disconnect in this region. It was proposed that a probability based approach be used to quantify the risk associated with the chemistry control envelope. [2] This can lead to an application-specific chemistry control program such as broadening the envelope for closure goals, while maintaining current levels for long-term storage.

Two options to revise the chemistry control program for wastes at dilute concentrations were considered: (1) Reduce the hydroxide limits, or (2) Reduce the required nitrite concentration. An experimental program has been undertaken to investigate the latter, which is the minimum molar nitrite concentration required to confidently inhibit pitting. A driver for focusing on the reduction of nitrite concentration is that nitrite does not deplete at the liquid-air interface, unlike hydroxide. An experimental matrix determined using logistic regression, provided the strategy for experimentation [2].

Statistically driven testing was initiated in FY08 with 104 test solutions. Based on visual observations of samples that had undergone CPP testing, a model based on the mixture/amount theory [8] was developed, see Figure 2. This model, while creating a good fit with the higher (0.5-1.0 M) concentrated nitrate, did not provide credible guidance at the lower concentration of nitrate (0.0-0.5 M). A second set of statistically determined solution sets was developed and analyzed in FY09. The intent of the second set is to provide increased confidence in and validity of the mixture/amount model by evaluating corrosion response in the solution space around the proposed limit.

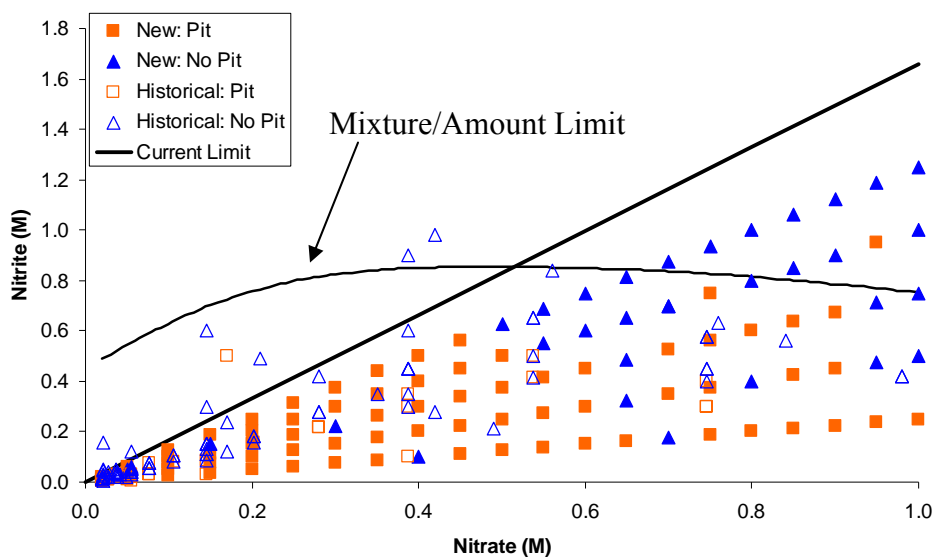


Figure 2 Results from Part I (FY 08) risk based corrosion results.

The mixture/amount model allowed for the investigation of a more complex relationship between the nitrate and nitrite concentrations and the probability of pitting than is represented by the model underlying the current chemistry control program as represented by equation (1). The model of equation (1) indicates that only controlling the ratio of nitrite to nitrate is sufficient to control the probability of pitting over the range of nitrate concentration of interest. For the mixture/amount, the probability of pitting is linked to a more complex function of the concentrations of nitrite and nitrate. The use of the mixture/amount model may provide a technical basis for the utilization of less nitrite to confidently inhibit pitting at concentrations below 1 molar nitrate as discussed below.

3 Experimental

Cyclic potentiodynamic polarization (CPP) testing was performed within the framework of the statistical test matrix to refine the current control limits. The CPP testing can be used to determine the electrochemical potential regimes in which low carbon steel is susceptible to pitting [9]. The material used for the CPP testing was semi-killed, hot-rolled A537. The nominal chemical composition for the alloy is 0.24 wt% C, 0.70-1.60 wt% Mn, 0.040 wt% S, 0.035 wt% P, and 0.15-0.5 wt% Si with small amounts of Cu, Cr, and Ni. The tests were conducted on disc samples of A537 that were nominally 5/8" diameter (Metal Samples, Munford, AL). Samples were ground using 800 grit grinding sheets to remove the native oxide layer and provide a flat surface. Grain size and shape were analyzed during previous testing [1].

3.1 Simulated Tank Solutions

The aqueous phase of radioactive waste is a complex solution containing numerous ionic species. These include the corrosive anion nitrate, in relatively high concentration and the corrosive anions chloride, sulfate, and fluoride, in relatively lower concentration. The protective anions are predominantly nitrite and hydroxide, but the waste also includes such protective anions as phosphate, chromate and molybdate in relatively low concentrations compared to the nitrite. Cost-effective, non-radioactive laboratory test solutions are simplifications of actual waste solutions. Corrosion testing experience in SRNL has shown that non-radioactive laboratory simulants of waste yield similar results to those of actual waste solutions [7,10]. It has also been shown that nitrate is the dominant corrosive anion. Chloride and sulfate have been shown not to require increased nitrite inhibitor concentration when they were individually increased over their typical waste concentrations. Sufficiently high concentrations of chloride and sulfate do eventually require higher nitrite concentrations to prevent pitting, and these higher nitrite concentrations have been quantified and incorporated into the waste tank corrosion control program.

A relatively simple non-radioactive simulant of waste was chosen for the testing reported herein. The major constituents were nitrate, nitrite, bicarbonate and carbonate. Chloride and sulfate were added for conservatism to ensure that the most potentially corrosive solution would be tested. Sodium nitrate and sodium nitrite were varied based on statistical modeling values. Sodium chloride and sodium sulfate were introduced to have a solution more representative of waste, which contains chloride and sulfate ions. Sodium chloride was added based on the

maximum amount allowable under current chemistry control limits. Sodium sulfate was added based on the 84 wt% of the maximum amount allowable under current chemistry control limits.

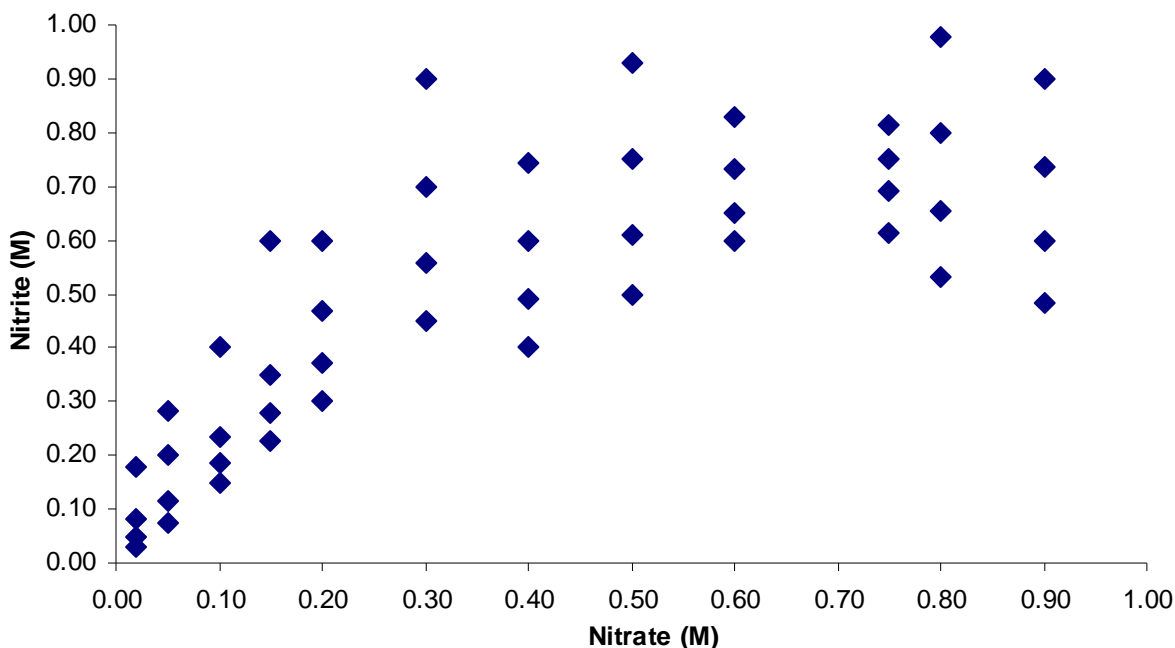


Figure 3 Testing matrix of Part II solution chemistries.

Simulated waste tank solutions were prepared using distilled water and reagent-grade chemicals: sodium chloride, sodium sulfate anhydrous, sodium carbonate, sodium bicarbonate, sodium nitrite, and sodium nitrate. The pH was maintained to 10.0 using a constant carbonate/bicarbonate molar ratio of 7 to 13. The gram amounts of carbonate and bicarbonate added were determined based on the nitrite amount. A total of 48 solutions were used for electrochemical testing, see Appendix A. Solutions were prepared based on a statistically determined experimental design [2]. The design space was defined by molar concentrations of nitrate between 0.02 and 1.0 M at a fixed temperature of 40 °C. The objective was to investigate the molar nitrite concentration required to confidently inhibit pitting and provide further confidence in the mixture/amount model fit, Figure 3.

3.2 Electrochemical Testing

The electrochemical cell was setup with A537 samples attached to a conductive wire and mounted in metallographic mount material as the working electrode and two graphite rods were used as counter electrodes (see Figure 4). The reference electrode was a saturated calomel electrode (SCE) connected to a Luggin bridge. The SCE in conjunction with a Luggin bridge was used to minimize noise/interference in the scans, as well as reduce the IR-drop which can form when large distances separate the working electrode and reference electrode. [11] Two scans were performed simultaneously on samples submerged in simulated solution and heated to 40 °C with heating tape and a hot plate. Aluminum foil was used to create Faraday cages around each electrochemical cell to minimize electrical interference. Samples were held at the open circuit potential (E_{OC}) for 2 hours prior to CPP testing.



Figure 4 Electrochemical experimental set-up.

Immediately following the open circuit potential hold, the CPP scan was performed on the sample. The cyclic polarization scans were performed by anodic polarization from -0.1 V vs. E_{OC} to 1.2 V vs. E_{OC} using a scan rate of 0.5 mV/sec. Immediately following the forward scan, a reverse scan was started at 1.2 V vs. E_{OC} and stopped once the potential returned to 0 V vs. E_{OC} .

After testing, the CPP scans were evaluated based on the shape of the CPP curve and the appearance of the sample after testing. The shape of the CPP curve was evaluated against a five category system to provide a method for assessing the degree of pitting, see Figure 5. The categories, in order of decreasing susceptibility, are:

- 1 Spontaneous pitting, significantly sloped forward scan, reverse scan never re-crosses;
- 2 Small hysteresis with E_{rp} returning at a lower potential compared to E_{corr} ;
- 3 $E_{rp} - E_{corr} < 300$ mV;
- 4 $E_{rp} - E_{corr} > 300$ mV;
- 5 Reverse scan retraces forward scan -OR- negative hysteresis (i.e., reverse scan occurs at lower current densities than forward scan)

E_{corr} represents the open-circuit potential, also known as the corrosion potential and E_{rp} is the repassivation potential, which is the potential at which the polarization re-crosses the passive portion of the cyclic polarization scan, indicating repassivation and the cessation of pitting.

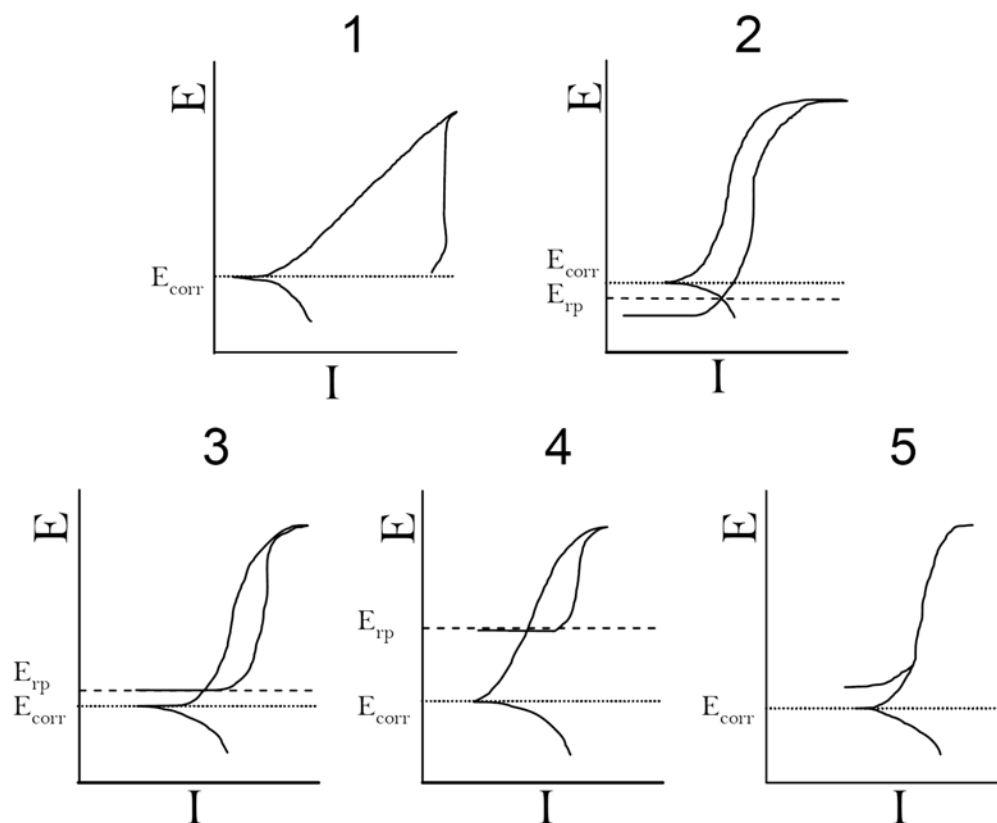


Figure 5 Cyclic polarization categories.

While the electrochemical scans were rated by a five category system, the visual results of the samples run in the electrochemical test were rated on a three category scale. Visually, the samples were rated on a three category scale: (1) heavy pitting, (2) moderate pitting, and (3) no pitting. To illustrate the three categories, see Figure 6.



Figure 6 Visual analysis of electrochemical samples.

4 Results and Discussion

4.1 CPP Testing

There were two criteria utilized to evaluate the results from the electrochemical tests. First, the potential-current plot was analyzed to determine critical potentials indicating breakdown (i.e., pitting potential) and/or repair (i.e., repassivation potential) of the passive film. Once these potentials were identified, they were then categorized on a scale from 1 to 5 as previously described. Secondly, at the completion of the test, each sample was evaluated visually for pitting and rated on a scale from 1 to 3 as previously described. The results are described below.

4.1.1 CPP Curves

A summary of the cyclic potentiodynamic polarization testing results based on the five category system of analyzing the cyclic potentiodynamic curves is shown in Figure 7. The results indicate that the ratio of nitrite to nitrate must be greater at low nitrate concentrations (< 0.5 M) than at high nitrate concentrations (~ 0.7 - 1.0 M). This result implies that the minimum nitrite required by the corrosion control program may be overly conservative in the vicinity of 1.0 M hydroxide.

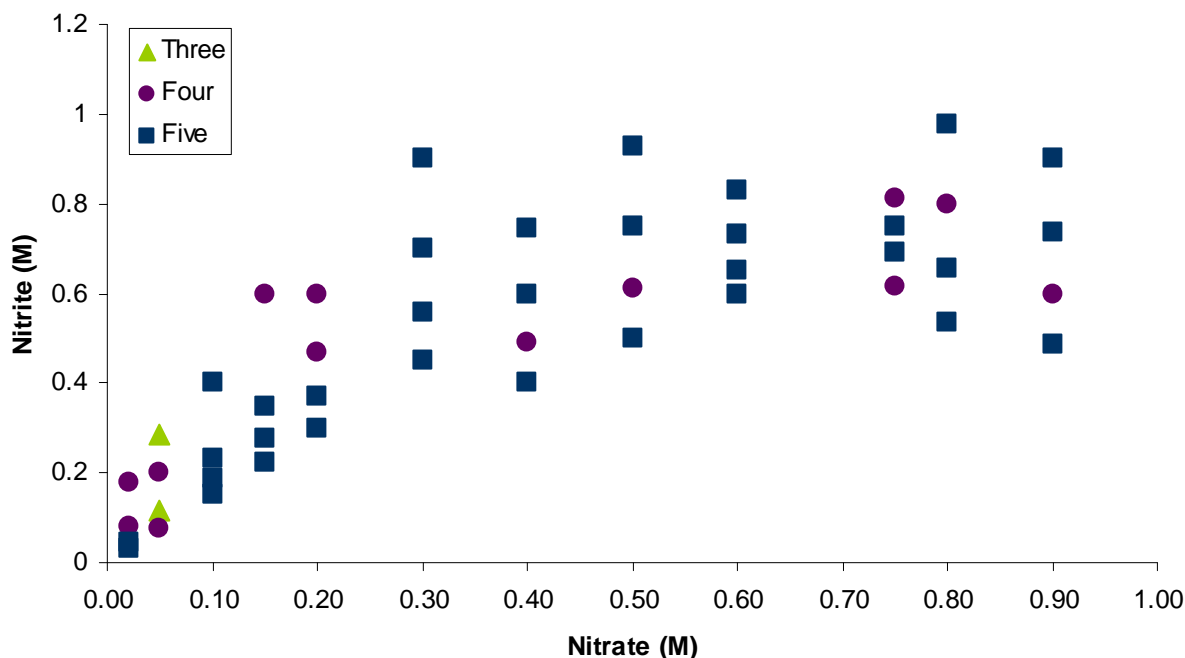


Figure 7 Summary of electrochemical results referring to the 5 category system where a rating of 1 suggests significant corrosion and 5 suggests no corrosion. Note, no curves resulted in a rating of 1 or 2.

The open-circuit potential for each solution was analyzed to determine the effect of the solution chemistry on the E_{corr} . No significant correlation was identified.

4.1.2 Visual Observations of CPP Samples

Electrochemical testing resulted in a range of corrosion responses from heavily corroded (♦) to unaffected metal surfaces (■). Overall, samples exhibiting significant amounts of corrosion were from solutions with low levels of nitrite, see Figure 8. Note several samples showed evidence of crevice corrosion on the interface between the mounting material and the metal sample. Crevice corrosion was discounted during visual examination of samples.

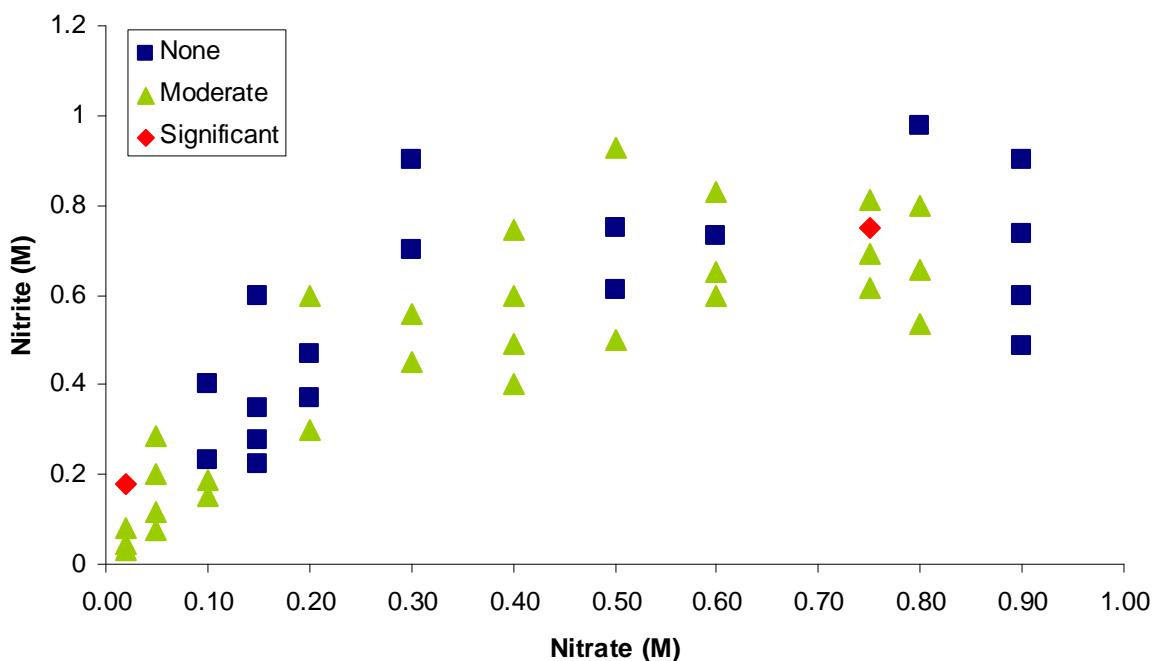


Figure 8 Summary of visual observations of samples after an anodic polarization cycle.

These results also indicated that the ratio of nitrite to nitrate must be greater at low nitrate concentrations (< 0.5 M) than at high nitrate concentrations (~ 0.7 - 1.0 M). However, some discrepancy occurred between the electrochemical and visual observations in terms of degree of corrosion for a given experimental condition. Since visual observation is a more direct corrosion evaluation method as compared to evaluation of electrochemical curves, the electrochemical statistical analysis was based on the visual results summary. Visual evaluation provided a better fit for the overall electrochemical data and allowed for discrimination between pitting and crevice corrosion.

4.1.3 Statistical Analysis of Results

Both CPP and visual observation results were evaluated. The corrosion results were treated as a binary function, leading to either a pit or no-pit outcome. CPP results with category values less than or equal to 4 were classified as corroded; visual observation results any amount of corrosion, either significant or moderate, were classified as corroded. Statistical models based on logistic regression were employed to analyze the results. [8]

The advantage of using the model is that the amount of nitrite needed to inhibit corrosion can be predicted based on percentage probability, allowing the facility to make quantifiable risk-based decisions.

When the optimal result from FY09 were combined with the even solution trials from FY08, the mixture/amount model still applied and indicated that a lower concentration of inhibitor is needed at 0.8-1.0 M nitrate compared to the 0.4-0.8 M nitrate regime (see Figure 9). While the additional experimental effort in the extremely dilute regime continued not to follow the mixture/amount model, when all the results of solutions containing less than 0.1 M nitrate are removed, the data support the mixture amount model, as seen in Figure 9. It is hypothesized that at the concentrations of nitrate less than 0.1 M, a separate chemical reaction involving the chlorides and sulfates is controlling the corrosion result [12], which creates difficulty when trying to incorporate the solution sets into the mixture/amount model.

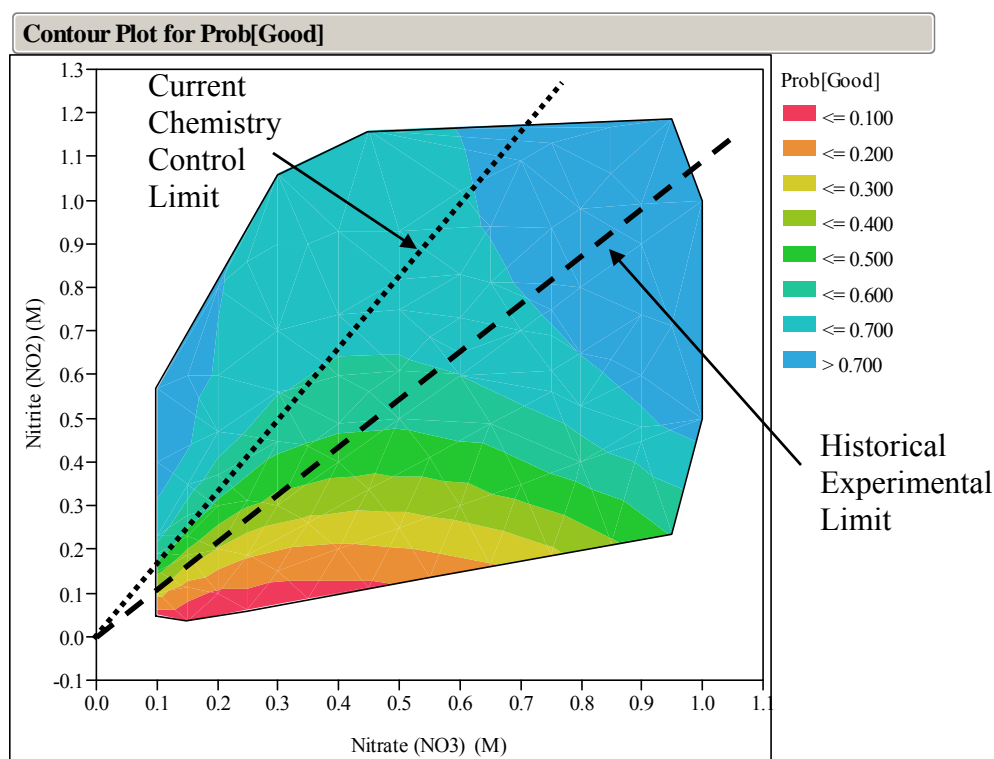


Figure 9 Contour plot for probability of a "no corrosion" outcome developed from the FY09 and even trials of FY08 developed from the mixture/amount model. The shaded regions represent probabilities of a good outcome, i.e. no pitting.

Unlike a simple linear fit, the more complex mixture/amount model requires significantly more experimental data to ensure a degree of high probability. Based on the current amount of data, the model is only able to determine probability limits up to >70%. However, when comparing the probability contours of the mixture/amount to the current chemistry control limit follows the slope of the model contours in the nitrate concentration region of 0.1 – 0.4 M.

5 Conclusions

Electrochemical CPP testing provided evidence to suggest that pitting susceptibility in A537 carbon steel depends on the ratio of aggressive to inhibitive anions (e.g. nitrate to nitrite), as well as the concentration of each species. The mixture amount model does not apply at concentrations below 0.1 M nitrate, however, it fits the solutions sets between 0.1 and 1.0 M nitrate. In the upper regime of nitrate concentration (0.8-1.0 M) the model suggests that less nitrite is needed compared to the intermediate regime of 0.4-0.8 M nitrate. This result further supports the reduction of the chemistry control nitrite inhibitor concentration in the regime of 0.8-1.0 nitrate.

6 Future Work

In FY10, need exists to explore the solution region above and below 1.0 M nitrate to further address the step change in the chemistry control program at 1.0 M nitrate. Historically, it has been determined that in dilute solutions, the primary corrosion mode is pitting. In concentrated solutions, the primary corrosion mode is stress corrosion cracking. In the development of the chemistry control program, 1.0 M nitrate was taken as the nitrate concentration by which the dominant corrosion mode transitions from pitting to stress corrosion cracking. No experimental evidence is present for this decision to switch chemistry control rationale at the 1.0 M concentration. It may well be that pitting continues to dominate for concentrations greater than 1.0 M. A series of experiments is necessary to determine where pitting truly stops being the dominant corrosion mode and stress corrosion cracking begins.

7 Acknowledgement

Thanks to Tracy Murphy for electrochemical testing and Philip Zapp for discussions.

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9 Appendix

Appendix A: Solution trial concentrations of nitrate and nitrite used for electrochemistry.

Trial ID	NO ₃ (M)	NO ₂ (M)	Cl (M)	SO ₄ (M)
1	0.02	0.03	0.001	0.008
2	0.02	0.05	0.002	0.013
3	0.02	0.08	0.002	0.025
4	0.02	0.18	0.004	0.067
5	0.05	0.08	0.002	0.024
6	0.05	0.12	0.003	0.040
7	0.05	0.20	0.005	0.076
8	0.05	0.28	0.006	0.115
9	0.10	0.15	0.004	0.054
10	0.10	0.19	0.004	0.069
11	0.10	0.23	0.005	0.091
12	0.10	0.40	0.008	0.173
13	0.15	0.23	0.005	0.087
14	0.15	0.28	0.006	0.112
15	0.15	0.35	0.007	0.148
16	0.15	0.60	0.011	0.280
17	0.20	0.30	0.006	0.123
18	0.20	0.37	0.007	0.158
19	0.20	0.47	0.009	0.208
20	0.20	0.60	0.011	0.280
21	0.30	0.45	0.009	0.199
22	0.30	0.56	0.010	0.257
23	0.30	0.70	0.012	0.337
24	0.30	0.90	0.014	0.454
25	0.40	0.40	0.008	0.173
26	0.40	0.49	0.009	0.220
27	0.40	0.60	0.011	0.280
28	0.40	0.74	0.012	0.362
29	0.50	0.50	0.009	0.226
30	0.50	0.61	0.011	0.287
31	0.50	0.75	0.012	0.366
32	0.50	0.93	0.015	0.472
33	0.60	0.60	0.011	0.280
34	0.60	0.65	0.011	0.308
35	0.60	0.73	0.012	0.356
36	0.60	0.83	0.013	0.412
37	0.75	0.61	0.011	0.288
38	0.75	0.69	0.012	0.332
39	0.75	0.75	0.012	0.366
40	0.75	0.81	0.013	0.402
41	0.80	0.53	0.010	0.244
42	0.80	0.65	0.011	0.311
43	0.80	0.80	0.013	0.395
44	0.80	0.98	0.015	0.501
45	0.90	0.48	0.009	0.217
46	0.90	0.60	0.011	0.280
47	0.90	0.74	0.012	0.358
48	0.90	0.90	0.014	0.454

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