Effect of Hydrazine on Flow Accelerated Corrosion

Technical Report

Effect of Hydrazine on Flow Accelerated Corrosion

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Final Report, March 2005

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REPORT SUMMARY

This final report presents results from a study jointly sponsored by EPRI and EDF to examine the effects of hydrazine on Flow Accelerated Corrosion (FAC) of carbon steel in single phase water. It documents work performed since publication of the interim report in November 2002 (1002768). The project team performed laboratory tests using the CIROCO loop with controlled water chemistry and mass transfer conditions. This study examined the effects of hydrazine concentration on the FAC rate of carbon steels at different temperatures and pH conditions.

Background

Researchers worldwide have studied Flow Accelerated Corrosion (FAC) of carbon steels for more than twenty years and the process is now fairly well understood. They have demonstrated, both theoretically and experimentally, the influence of several parameters like water chemistry (i.e., pH and oxygen content), temperature, hydrodynamic or mass transfer conditions (i.e., flow velocity, geometry, steam quality) and steel composition on the corrosion kinetics. However, research has yet to fully explore the effects of a reducing environment and variable redox conditions.

The establishment of a reducing environment is effective in increasing the resistance of steam generator tubing to intergranular attack / stress corrosion cracking (IGA/SCC) and pitting. Therefore, secondary water chemistry specifications recommend consideration of high hydrazine chemistry in the steam-water circuit to ensure reducing conditions. However, some previous work indicated that increased hydrazine levels, e.g., approximately 150 µg/kg, could have a detrimental effect on carbon steel feedwater piping by potentially enhancing the FAC process. The EPRI interim report published in November 2002 (1002768) demonstrated that hydrazine has no negative impact on FAC of carbon steels at 180°C. This work is primarily aimed at better quantifying the effect of hydrazine on FAC rates of carbon steel at 235°C and at an intermediate temperature of 210°C.

Objective

To study the effect of hydrazine on FAC of carbon steel at two different temperatures and at different pH conditions.

Approach

Tests were performed on carbon steel specimens with a chrome content equal to 0.009% (weight percent), close to that found in various carbon steel components of nuclear power plants. The project team carried out the FAC tests using the CIROCO loop under the following conditions:

- Temperature: 210°C or 235°C
- Pressure: 35 to 40 bar

- Oxygen maintained at < 0.5 µg/kg
- Single-phase flow with water velocity for tubular specimens: 5 m/s or 10 m/s
- Single specimen (8 mm internal diameter) or two specimens in parallel (5.5 mm internal diameter each)

The project team used flowrate to control the water velocity in the tubular test specimens, and used Thin Layer Activation (TLA) to provide a measure of the FAC rate on-line.

Results

The results show that temperature is an important parameter leading to a different effect of hydrazine on the FAC rate of carbon steels:

- At 180°C, hydrazine had no measurable effect on FAC rates other than it's effect on high temperature pH.
- At 210°C, hydrazine had no effect to a minimal effect (maybe 20% increase) in the observed FAC rate. However, it appears that any effect is well within the scatter of the measured rates.
- At 235°C, a hydrazine increase from zero to approximately 150 µg/kg may result in an increased FAC rate (best estimate as much as 60% larger). However, the data scatter and observation of increasing FAC rate, even when hydrazine is decreasing to zero, makes any quantification highly uncertain.

At 235°C, there is no discernable difference in the hydrazine effect on FAC at $pH_{25°C}$ values of 9.0, 9.3 and 9.5.

EPRI Perspective

Flow accelerated corrosion of carbon steel tubes has long been a significant concern in the nuclear industry. The recent experience at Mihama in 2004 underscores the importance of maintaining a vigilant mitigation and examination program of FAC susceptible components. Researchers have performed numerous studies to characterize the water chemistry and hydrodynamic conditions that affect FAC. In 1998, EPRI published TR-106611-R1, which was a collaborative effort among EPRI, EDF and Siemens Power Generation to provide a comprehensive assessment of FAC in the nuclear industry. Other EPRI sponsored work has included research to correlate FAC of SG internals to plant water chemistry (TR-111113), an investigation of the effect of oxygen on corrosion product transport (1000742), a review of alternate hydrazine injection locations on final feedwater corrosion product transport and ECP at South Texas Project (1001472), and a multivariable assessment of FAC and SG fouling (1003619). This study contributes to our understanding of how we can mitigate FAC of carbon steel components.

Keywords

Flow Accelerate Corrosion (FAC)
Carbon steel corrosion
Hydrazine

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1

INTRODUCTION

Flow Accelerated Corrosion (FAC) of carbon steel is a well known phenomenon, which has been studied for many years and is fairly well understood. It is a chemical type of degradation that affects carbon steel components in deaerated and alkalinized flowing water, mainly in the temperature range of 100°C to 300°C. Wall thinning rates are normally less than 0.03 µm/h (0.25 mm/year) but rates as high as approximately 0.34 µm/h (3 mm/year) have already been observed in operating plants [1]. Moreover, this type of degradation can lead to sudden piping failures, thus representing an important personnel safety concern for Nuclear Power Plants, such as the one that occurred very recently (August 2004) in the Japanese Power Plant Mihama 2.

The FAC mechanism consists of iron dissolution in flowing water and can be described as the action of four simultaneous reactions (Figure 1-1):

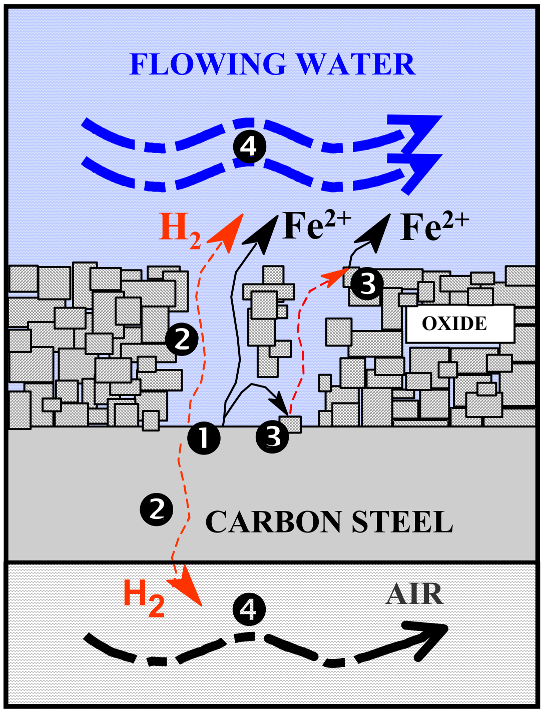

Figure 1-1
Schematic Description of the Flow Accelerated Corrosion Mechanism

1. oxidation of iron to soluble ferrous ions and magnetite at the internal interface between the steel and the oxide,

$Fe + 2\ H_2O \Rightarrow Fe^{2+} + 2\ OH^- + H_2$ **Equation 1-1**

$3\ Fe + 4\ H_2O \Rightarrow Fe_3O_4 + 4\ H_2$ **Equation 1-2**

2. diffusion of soluble species (iron and hydrogen) across the porous oxide and diffusion of hydrogen through the carbon steel,
3. dissolution and reduction of magnetite at the external interface between the oxide and the water,

$2\ Fe_3O_4 + N_2H_4 + 12\ H^+ \Leftrightarrow 6\ Fe^{2+} + 8\ H_2O + N_2$ **Equation 1-3**

4. transfer of soluble iron species towards the flowing water and transfer of hydrogen towards the air after diffusion through the steel.

A number of parameter influences have been studied, such as water chemistry, temperature, hydrodynamic and mass transfer conditions as well as steel composition [1]. As a matter of fact, most of these parameters are well described by models [1]. Nevertheless, the effect of some parameters still require investigation to try to better understand the mechanism of the degradation and to help update guidelines and improve FAC models. A more quantitative understanding of the effect of hydrazine on FAC is the purpose of this work.

Hydrazine is used in plant units to guarantee a reducing environment in the secondary circuit as it is an oxygen scavenger:

$N_2H_4 + O_2 \Leftrightarrow N_2 + 2H_2O$ **Equation 1-4**

and thus mitigates secondary side stress corrosion cracking of steam generator tubes. But recent EDF/EPRI results [2] have shown FAC could be increased in such reducing environments under specific chemistry, temperature and hydraulic conditions.

This report presents the recent results of FAC testing in the presence of hydrazine conducted on the CIROCO loop. The purpose of these latest tests was to reduce the scatter in data previously reported and to help model the influence of hydrazine on FAC.

2

PREVIOUS RESULTS

In fairly recent tests [2], two different effects of hydrazine on the FAC rate of carbon steel tubular specimens were exhibited on the CIROCO loop in strictly controlled water chemistry and mass transfer conditions, depending on the test temperature. The data at 180°C were first reported in [2], while the data at 235°C were reported in [2, 3]. Figure 2-1 shows how the FAC rate for a given hydrazine concentration relative to the average FAC rate obtained without hydrazine ("relative FAC rate") changed with the hydrazine concentration.

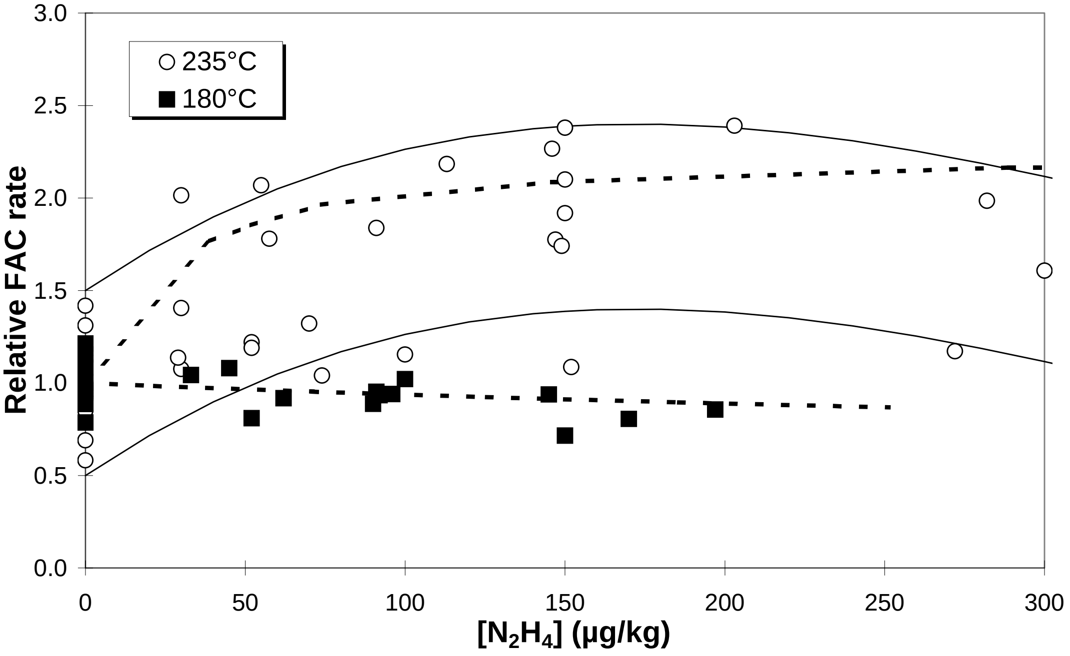

Figure 2-1
Relative FAC Rate Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow at 180°C and 235°C at $pH_{25°C}$ = 9.0 [2, 3]

These results show two different trends with respect to temperature:

- At 180°C, hydrazine had no measurable effect on FAC rates other than the effect it has on high temperature pH_T (6.2 to 6.3).
- At 235°C, an increase of the relative FAC rate for hydrazine concentrations from 0 µg/kg to 150-200 µg/kg was observed. In this range, the maximum FAC rate increased by a factor of 2.5 with an average increase of a factor of 2. Then, a decrease of the FAC rate was observed for concentrations above 150-200 µg/kg.

3
EXPERIMENTAL

3.1 Material and Specimen

Tests were performed on carbon steel specimens with a chrome content equal to 0.009 % (weight %) close to that found in various carbon steel components of nuclear power plants. Table 3-1 shows the reference and the chemical composition of this material. This material was also used in previous tests [2].

Table 3-1
Chemical Composition of the Studied Material (wt. %)

EDF Reference		Origin	Product	C	S	P	Si	Mn	Ni	Cr	Mo	Cu
T 86	E 24.2	Usinor	Plate	0.109	0.015	0.013	0.24	0.72	0.06	**0.009**	< 0.01	0.02

Specimens were straight tubes of 130 mm length, an external diameter of 16 mm and an internal diameter of either 8 mm (as were all the specimens tested during the previous tests) or 5.5 mm. The specimens with an internal diameter of 8 mm were tested individually while those with an internal diameter of 5.5 mm were studied by pairs set in parallel.

3.2 Test Facility: CIROCO Loop

3.2.1 Physical Parameters of the Loop

The FAC tests were carried out on the CIROCO loop, described previously [2], using the following conditions:

- temperature : 210°C or 235°C,
- pressure : 35 to 40 bar,
- single-phase flow with water velocity for tubular specimens : 5 m/s or 10 m/s,

The water velocity in the tubular test specimens (Table 3-2) was controlled by the flow rate (see equation (3-1)). The total flow rate of water circulating in the specimens was measured by means of mass flow meters which acted on the system regulating the circulating pumps.

$$v = \frac{Q}{\frac{\pi D^2}{4} \times \rho}$$

Equation 3-1

with v(m/s) = mean water velocity in the specimen,
Q (kg/s) = flow rate in the specimen,
D (m) = internal diameter of the specimen,
ρ (kg/m^3) = water density at temperature.

Table 3-2
Mass Flow Rate as a Function of the Water Velocity and the Temperature

Water Velocity (m/s)	Internal Diameter (m)	Test Temperature (°C)	Water Density* (kg/m^3)	Flow Rate (kg/s)
10	0.0055	210	852.8	0.202
5				0.101
10		235	819.7	0.195
5				0.097
10	0.008			0.410
5				0.205

*according to the database of the BRT-CICERO code [4].

The flow rates were continuously measured and recorded at the exit of both specimens, while the water velocities were continuously calculated and recorded only based on the flow rate of specimen 1. Nonetheless, the mean water velocities at the exit of specimen 2 during each step were calculated based on equation (3-1) as a function of the mean value of the corresponding flow rate during this step. These calculations showed the water velocity at the exit of specimen 2 was always either equal to the water velocity at the exit of specimen 1 or slightly lower (0.2 m/s lower maximum) during all four tests with two specimens tested in parallel (Test 1, Test 4, Test 5 and Test 6 – see Table 3-3).

Table 3-3
Test Conditions

Test	Specimens	Water velocity (m/s)	T (°C)	$pH_{25°C}$	[N_2H_4] range (µg/kg)
1	A and B	10	235	9.0	0-250
2	C	5	235	9.0	0-250
3		10	235	9.0	0-400
4	D and E	10	235	9.3	0-150
5		10	235	9.5	0-150
6	F and G	10 and 5	210	9.0	0-150

3.2.2 Water Chemistry

pH

The water in the loop was conditioned at $pH_{25°C}$ = 9.0, 9.3 or 9.5 by addition of ammonia and hydrazine. The total conductivity is a more sensitive parameter than the pH value to the variation in ammonia. So, the water chemistry pH was controlled and adjusted by the conductivity value inside the feedwater tank (make-up tank) of the loop. Appendix A presents the room temperature pH ($pH_{25°C}$), at temperature pH (pH_T) and the conductivity values versus the ammonia and hydrazine concentrations of the environments used for the FAC tests presented in this report.

Hydrazine

The hydrazine content was measured with a hydrazine-meter (ABB KENT 7835) in the make-up tank or immediately downstream of the test specimen. The expected hydrazine concentration was between 0 and 400 µg/kg for the tests at 235°C and between 0 and 150 µg/kg at 210°C. Unfortunately, the limited possible duration of each test based on the Thin Layer Activation (TLA) method (see section 3.3) together with the number of different hydrazine concentration steps for each test did not allow specimen testing for long periods of time at each hydrazine concentration (41h to 191h). Additionally, several difficulties encountered during the operation of t he CIROCO loop (replacement of components, leaks,...) during most of the testing complicated the testing in some cases. Consequently, the decision was taken to focus on the 0-150 µg/kg range, most representative of hydrazine concentrations used during operating conditions in the plants, for the final testing.

During some test steps at 235°C, the location of the hydrazine analysis was changed from the make-up tank to the exit of the test specimen at some point during the test step. In these cases, the hydrazine consumption from the make-up tank to the test specimen was determined. The results indicated that the difference between these two hydrazine concentrations was equivalent for all test steps (Figure 3-1) and could be expressed as follows:

$$[N_2H_4]_{\text{cell}} = 1.004*[N_2H_4]_{\text{make-up tank}} - 4.622 \qquad \textbf{Equation 3-2}$$

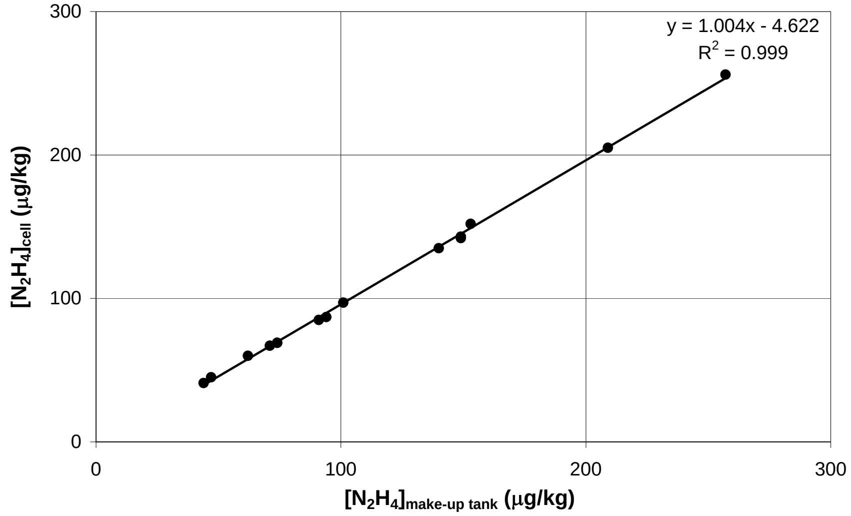

Figure 3-1
Hydrazine Concentration from the Exit of the Test Specimen as a Function of the Hydrazine Concentration from the Make-Up Tank for Testing at 235°C

Therefore, if the hydrazine concentration was only measured at one of the two points (make-up tank or specimens), the hydrazine concentration at the other point could be deduced from equation (3-2).

Oxygen

The dissolved oxygen was measured by an “Orbisphere” oxygen-meter in the make-up tank and in the test specimen and was always lower than 0.5 µg/kg. The oxygen concentration values in the make-up tank were most often close to 0.3 µg/kg and close to 0.1 µg/kg in the test specimen.

3.3 FAC Rate Measurement

The tubular specimen thickness loss rates were measured using the Thin Layer Activation (TLA) method with ^{56}Co as a radionuclide surface marker. The principle of the method has already been described elsewhere [2]. The carbon steel specimen was activated on a surface of 12.5 mm² ($\phi \approx 4$ mm) and a thickness of 150 µm. The linear activated depth over which a proper FAC rate could be determined was 100 µm.

The loss of material due to FAC could be related to the decrease of the material residual volumetric γ activity (accounting for the natural decay of ^{56}Co activity) which was measured on-line. The FAC rate was calculated with the slope of the thickness loss when the steady-state FAC process was reached as well as when the different measured chemical parameters (conductivity, hydrazine concentration) were stable. The FAC rates were calculated in µm/h and then converted to mm/year. FAC rates lower than 10^{-2} µm/h are not really satisfactory as the slope of the thickness loss measurement, over the step duration (approximately 100 hours), becomes highly uncertain relative to the measurement noise at these lower FAC rates (some examples are presented in Appendix B).

In order to compare different test steps, some of the results were expressed in terms of relative FAC rates (ratio between the FAC rate in the investigated environment and the FAC rate measured during the first step of the same test in the reference environment, e.g., without hydrazine[1].

3.4 Performed Tests

In order to determine the influence of hydrazine on the FAC rate of carbon steel, tests in deaerated water ($O_2 < 0.5$ ppb in the make-up tank) were performed at 210°C and 235°C. Each test included several test steps (with durations ranging from 41 h to 191h), with each step at a constant hydrazine concentration (detailed conditions and results in Appendix C). Table 3-3 presents the expected values of the principle parameters of the six tests presented in this report.

The first three tests were aimed to confirm the hydrazine influence evidenced previously [2] at 235°C. Using two specimens in parallel and extending the step durations were to help reduce the data scatter (see Figure 2-1).

Tests 4 and 5 were aimed to examine the potential modification of the hydrazine influence at other tested pHs.

Test 6 was conducted to evaluate the hydrazine influence at a temperature between the two previously tested temperatures (180°C and 235°C). The temperature of 210°C was chosen as it is slightly higher than the threshold temperature (205°C) above which the thermal decomposition of hydrazine was evidenced [5].

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[1] Previous tests defined the relative FAC rate as the ratio between the FAC rate in the investigated environment and the mean value of all FAC rates measured in the reference environment (without hydrazine) in the same test.

4
RESULTS

The results of all tests are presented below and detailed in Appendix C. On Figure 4-1 to Figure 4-6 are reported both the overall absolute FAC rate measured during each step of the tests (step rate), and the absolute FAC rates measured during 24h periods (24h rate), for tests 1 through 6, respectively. The latter FAC rates are generally considered less reliable than the ones measured over the whole period of the step, but they give an indication of the uncertainty of the measurement and the variability of the FAC rate during a step.

Figure 4-1 shows the absolute FAC rate at $pH_{25°C}$ 9.0 and 235°C under a 10 m/s water velocity. Both specimens showed an increasing FAC rate with increasing hydrazine concentration from 0 µg/kg to 113 µg/kg. However, as hydrazine was increased further to 256 µg/kg, the FAC rate for both specimens appeared relatively stable. Please note the scatter in the data between the "step rate", representative of the average FAC rate over the entire hydrazine step, and the "24h rates", representative of FAC rates over 24 hour portions of the hydrazine step. The largest percent deviation between the "step rate" and "24h rate" at any particular run varied from a maximum of 31% for specimen 2 at 0 µg/kg hydrazine to a minimum of 6% for specimen 1 at 113 µg/kg hydrazine.

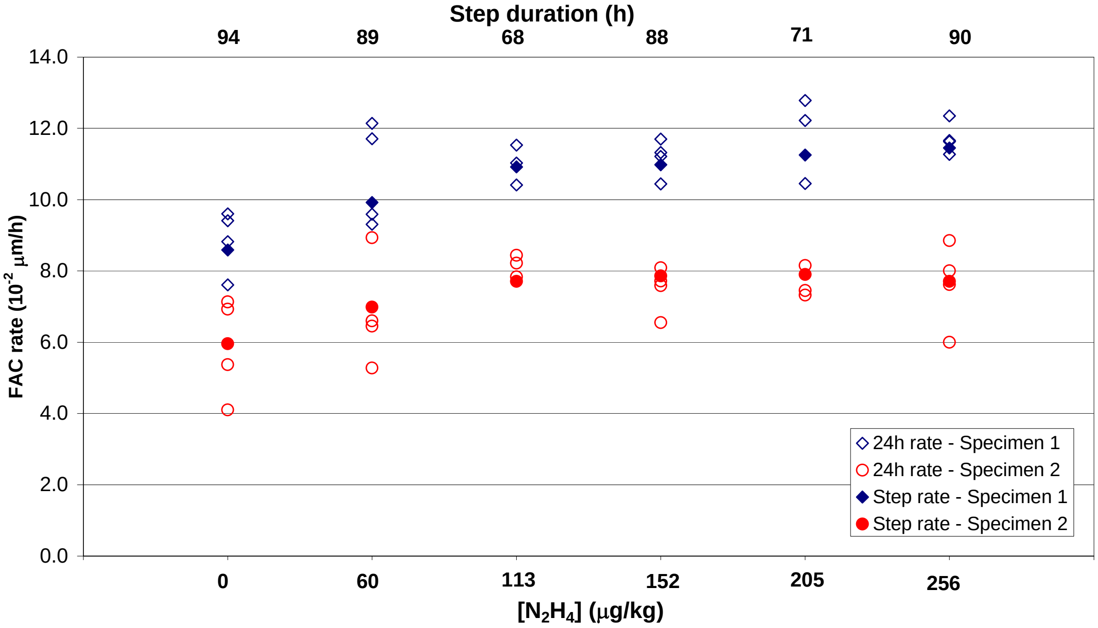

Figure 4-1
FAC Rate Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow with a Water Velocity of 10 m/s at 235°C at $pH_{25°C}$ = 9.0 During Test 1

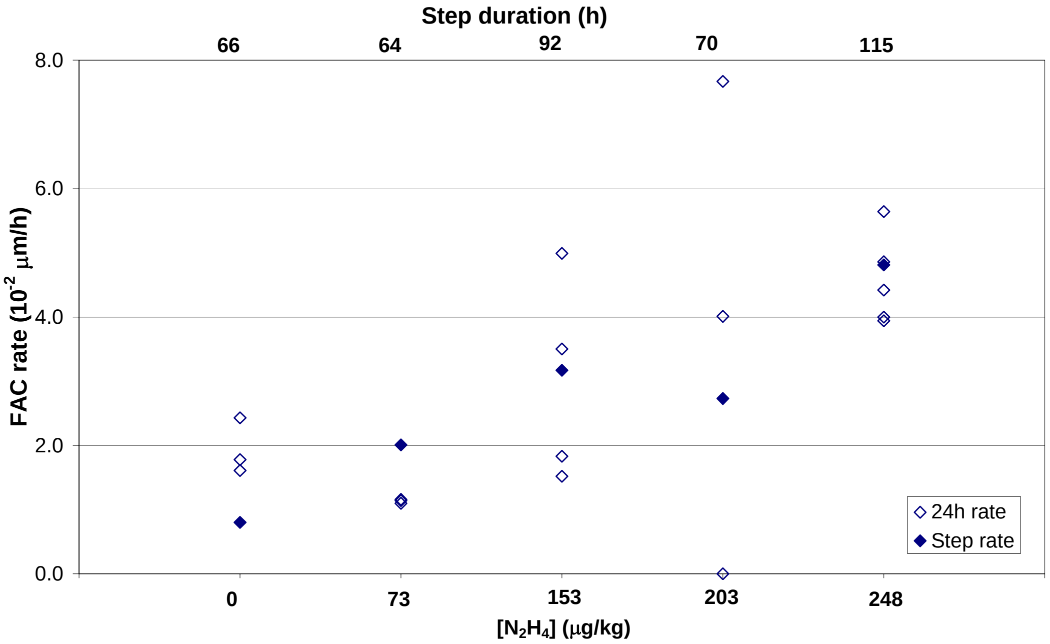

Figure 4-2
FAC Rate Versus Hydrazine Concentration for a Tubular Carbon Steel Specimen Exposed to a Single-Phase Flow with a Water Velocity of 5 m/s at 235°C at $pH_{25°C}$ = 9.0 During Test 2

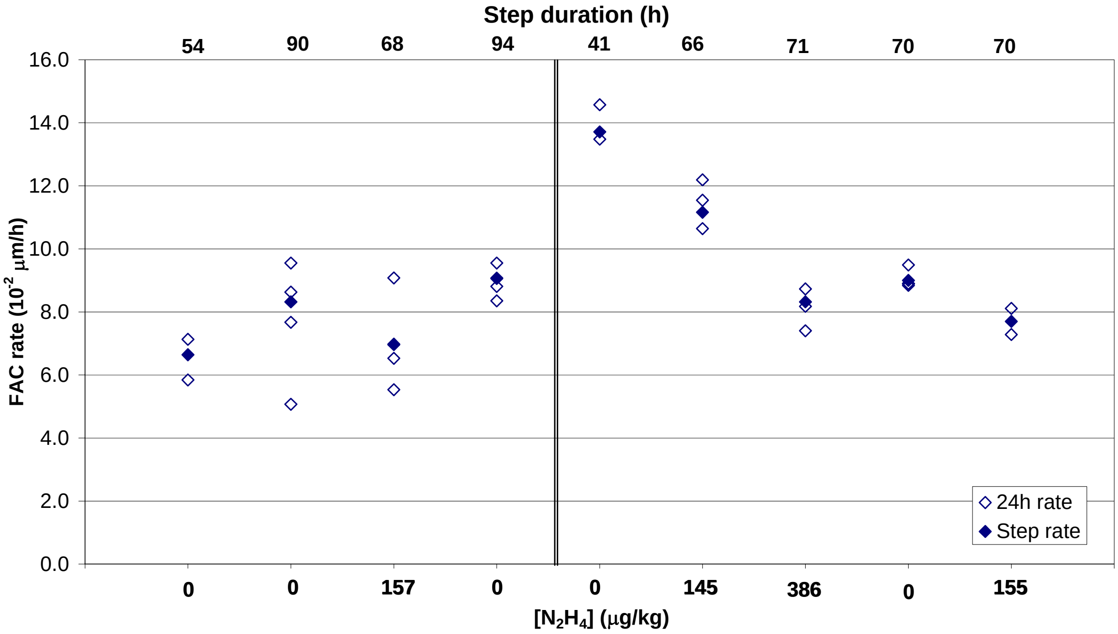

Figure 4-3
FAC Rate Versus Hydrazine Concentration for a Tubular Carbon Steel Specimen Exposed to a Single-Phase Flow with a Water Velocity of 10 m/s at 235°C at $pH_{25°C}$ = 9.0 During Test 3 Conducted Over Two Periods of Time (Steps 1 to 4 for Period 1 – Steps 5 to 9 for Period 2)

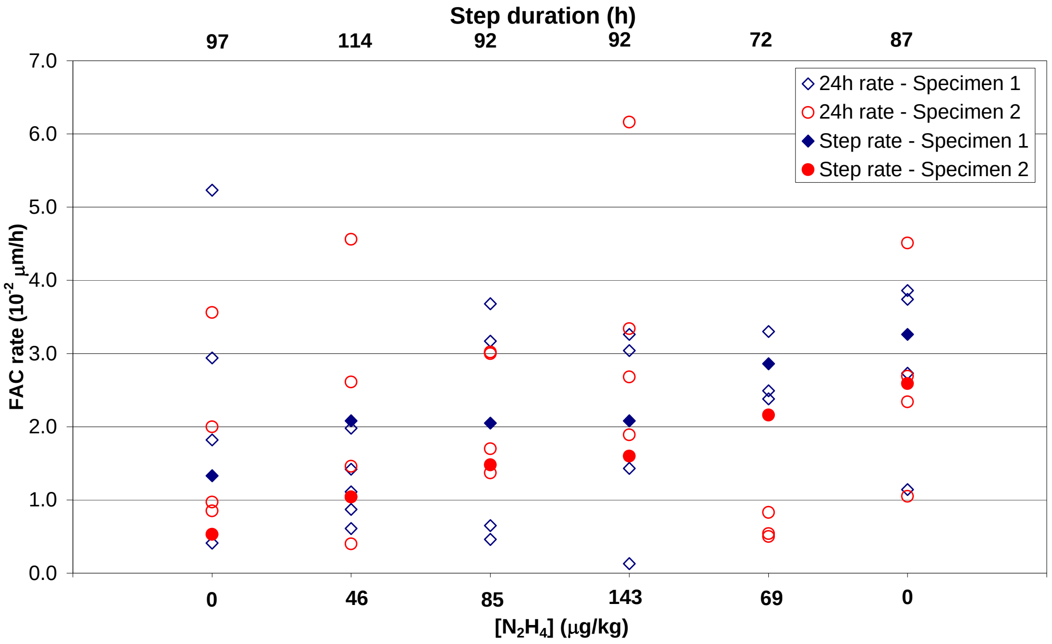

Figure 4-4
FAC Rate Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow with a Water Velocity of 10 m/s at 235°C at $pH_{25°C}$ = 9.3 During Test 4

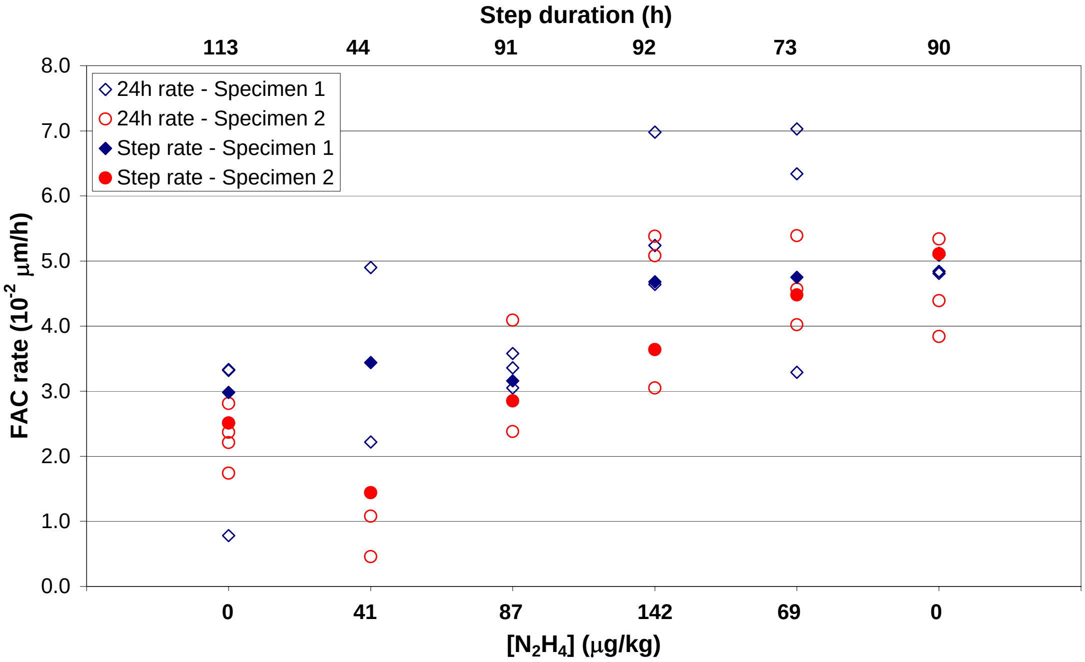

Figure 4-5
FAC Rate Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow with a Water Velocity of 10 m/s at 235°C at $pH_{25°C}$ = 9.5 During Test 5

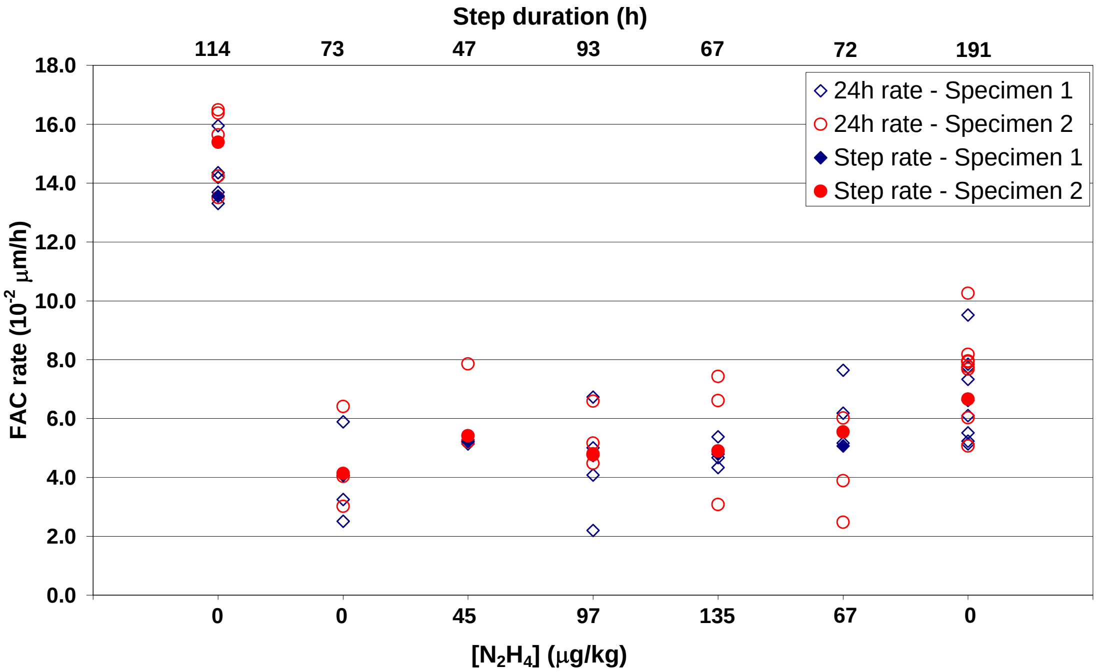

Figure 4-6
FAC Rate Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow with a Water Velocity of 10 m/s (First Step) or 5 m/s at 210°C at $pH_{25°C}$ = 9.0 During Test 6

Figure 4-2 shows the absolute FAC rate of the tested specimen at $pH_{25°C}$ 9.0 and 235°C under a 5 m/s water velocity. The FAC rate tended to increase with increasing hydrazine concentration from 0 µg/kg to 248 µg/kg. However, the absolute FAC rate ("step rate") at 203 µg/kg was lower than the ones measured at 153 µg/kg and at 248 µg/kg. Additionally, the variability between the "step rate" and "24h rates" was very large in a number of the hydrazine steps. The largest percent deviation between the "step rate" and "24h rate" at any particular run varied from a maximum of 205% at 0 µg/kg hydrazine to a minimum of 18% at 248 µg/kg hydrazine.

Figure 4-3 shows the absolute FAC rate at $pH_{25°C}$ 9.0 and 235°C under a 10 m/s water velocity. This test was conducted over two separate periods of time, as delineated by the vertical line in the middle of the figure[2]. The ordering of the hydrazine concentrations tested was different in this test than the previous testing shown in Figures 4-1 and 4-2. No systematic increase or decrease in the hydrazine concentration was done in this particular test. As shown in Figure 4-3, the results are different from previous testing as follows: (1) The FAC rate at 157 µg/kg hydrazine was lower than or equivalent to the three runs with no hydrazine during the first testing period; (2) Increasing the hydrazine concentration from zero to 145 µg/kg and then to 386 µg/kg hydrazine resulted in reduced FAC rates during the second testing period. In addition, the FAC rate was relatively stable even during subsequent hydrazine concentration changes

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[2] The loop was shutdown for maintenance over a period of 8 weeks between these two periods and the specimen was not replaced. No major event occurred during the two phases and the oxide that may have formed at the surface of the specimen between the two phases was taken away by staying several hours at neutral pH at 180°C (pH and temperature for which the FAC rate is very high) before starting the second phase.

from 386 µg/kg to zero then to 155 µg/kg hydrazine. The variability between the “step rate” and “24h rates” is also apparent in Figure 4-3. The largest percent deviation between the “step rate” and “24h rate” at any particular run varied from a maximum of 39% at 0 µg/kg hydrazine during step 2 to a minimum of 5% at 0 µg/kg hydrazine during step 8.

Figure 4-4 shows the absolute FAC rate at $pH_{25°C}$ 9.3 and 235°C under a 10 m/s water velocity for two specimens in parallel. The FAC rate for specimen 1 (“step rate”) increased from the first step (0 µg/kg) to the second step (46 µg/kg), was then stable during the next three steps (46 µg/kg - 85 µg/kg - 143 µg/kg) and finally increased during the last two steps (69 µg/kg - 0 µg/kg). Specimen 2 followed the same trend, with a stable absolute FAC rate during step three (85 µg/kg) and step four (143 µg/kg). However, there is a large amount of scatter in the data between the “step rate” and “24h rates” at each hydrazine concentration. The largest percent deviation between the “step rate” and “24h rate” at any particular run varied from a maximum of 572% for specimen 2 at 0 µg/kg hydrazine to a minimum of 17% for specimen 1 at 69 µg/kg hydrazine.

Figure 4-5 shows the absolute FAC rate at $pH_{25°C}$ 9.5 and 235°C under a 10 m/s water velocity for two specimens in parallel. The FAC rate (“step rate”) was relatively stable for both specimens between zero and 87 µg/kg hydrazine, except for a decline in the FAC rate for specimen 2 from zero to 41 µg/kg hydrazine. Both specimens showed an increase in the FAC rate from 87 µg/kg to 142 µg/kg hydrazine. However, the FAC rate for specimen 1 remained relatively stable during subsequent hydrazine concentration decreases to 69 µg/kg and then to zero hydrazine, while the FAC rate for specimen 2 increased slightly. This data clearly demonstrates that there is inherent uncertainty even in the “step rate” data as the two parallel specimens showed different behaviors under the same chemistry and hydraulic conditions. In addition, the variability between the “step rate” and “24h rates” is again apparent at each hydrazine concentration. The largest percent deviation between the “step rate” and “24h rate” at any particular run varied from a maximum of 74% for specimen 1 at 0 µg/kg hydrazine during the first step to a minimum of 6% for specimen 1 at 0 µg/kg hydrazine during the last step.

Figure 4-6 shows the absolute FAC rates measured on two specimens tested in parallel at $pH_{25°C}$ 9.0 and 210°C under a 10 m/s water velocity (step 1) and a 5 m/s water velocity (following steps). The water velocity was decreased from step 1 to step 2 as the measured absolute FAC rates were much too high to allow the test to be continued long enough. Under the 5 m/s water velocity, the trend for both specimens was the same : the absolute FAC rates (“step rates”) slightly increased while the hydrazine concentration was increased from 0 µg/kg to 45 µg/kg[3] and was then quite stable during the four subsequent steps (45 µg/kg – 97 µg/kg – 135 µg/kg – 67 µg/kg) and finally increased slightly during the last step (0 µg/kg). The scatter in the data between the “step rate” and the “24h rates” at each hydrazine concentration is generally as large as the slight changes observed in the identified “step rates” from one hydrazine concentration to the next. For example, the largest and smallest percent deviation between “step rates” is 31% from zero µg/kg hydrazine to 45 µg/kg hydrazine for specimen 2 and 1% from 97 µg/kg hydrazine to 135 µg/kg hydrazine for specimen 1, respectively. Similarly, the largest percent deviation between the “step rate” and “24h rate” at any particular run varied

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[3] This is not due to a slight decrease of the oxygen concentration in the test cell, as this concentration was very low (≤0.2 µg/kg) through the entire test.

from a maximum of 55% for specimen 2 at 67 µg/kg hydrazine to a minimum of 2% for specimen 1 at 45 µg/kg hydrazine.

Another interesting observation in a number of the tests, although not true in all cases, is that when the hydrazine concentration was step increased regularly, the FAC rate generally increased and when the hydrazine concentration was step decreased during the second part of the test, the FAC rate still continued to increase (see Figure 4-4 and Figure 4-5). These results are quite surprising. As a matter of fact, tests in this laboratory had never been conducted previously by systematically increasing the hydrazine concentration from one step to the other and then decreasing it back down to zero.

Some differences were observed between the FAC rates measured on specimen 1 and on specimen 2 (Table 4-1) during the tests where two specimens were tested in parallel (Test 1, Test 4, Test 5 and Test 6). The average ratio between the FAC rates ("step rates") measured on specimen 1 and specimen 2 was 1.32. These differences could be do to slight differences in material properties as different specimens were used when tested in parallel, even though all test specimens were made from the same carbon steel plate,. In addition, as noted previously, the flow rate of specimen 2 was always equal to or slightly less than (0.2 m/s lower maximum) than the flow rate of specimen 1. This is consistent with a slightly lower average FAC rate for specimen 2 compared to specimen 1 observed during Test 1, Test 4 and Test 5. Nonetheless, during three of the four tests, the two specimens showed similar trends (Figure 4-1, Figure 4-4, and Figure 4-6). The only significant differences between behavior of the two test specimens was noted in Test 5 (Figure 4-5). Material differences can generally be neglected when analyzing the relative FAC rates, as the same specimens were used throughout one test. As a matter of fact, the differences between relative FAC rates were generally small (Table 4-1), with an average ratio of specimen 1 to specimen 2 of 0.97. This demonstrates a posteriori the interest in testing two specimens in parallel to reduce the scatter in data, and the use of a relative scale to analyze FAC results.

Table 4-1
Range of Ratios of FAC Rates Measured on Specimen 1 and Specimen 2 for Test 1, Test 4, Test 5 and Test 6

Test	FAC Rate (Specimen 1)/ FAC Rate (Specimen 2)			Relative FAC Rate (Specimen 1)/ Relative FAC Rate (Specimen 2)		
	minimum	mean	maximum	minimum	mean	maximum
1	1.40	1.43	1.49	0.97	0.99	1.03
4	1.26	1.63	2.51	0.50	0.65	1.00
5	1.00	1.34	2.39	0.84	1.13	2.01
6	0.88	0.95	0.99	1.00	1.08	1.13

The relative uncertainty of a FAC rate measurement tended to be higher when the absolute FAC rate was low (Test 2 for example). Conversely, this relative uncertainty was rather low when the absolute FAC rate was high (Test 3 for example).

In most cases, the step rate value ranged between the minimum and the maximum 24h rates measured during that step. Nonetheless, in a few cases, this was not true. For example, during test 1 – step 3, the step rate measured for specimen 2 was lower than the three 24h rates measured during that step. This is due to the high sensitivity of the FAC rate measurement (Figure 4-7).

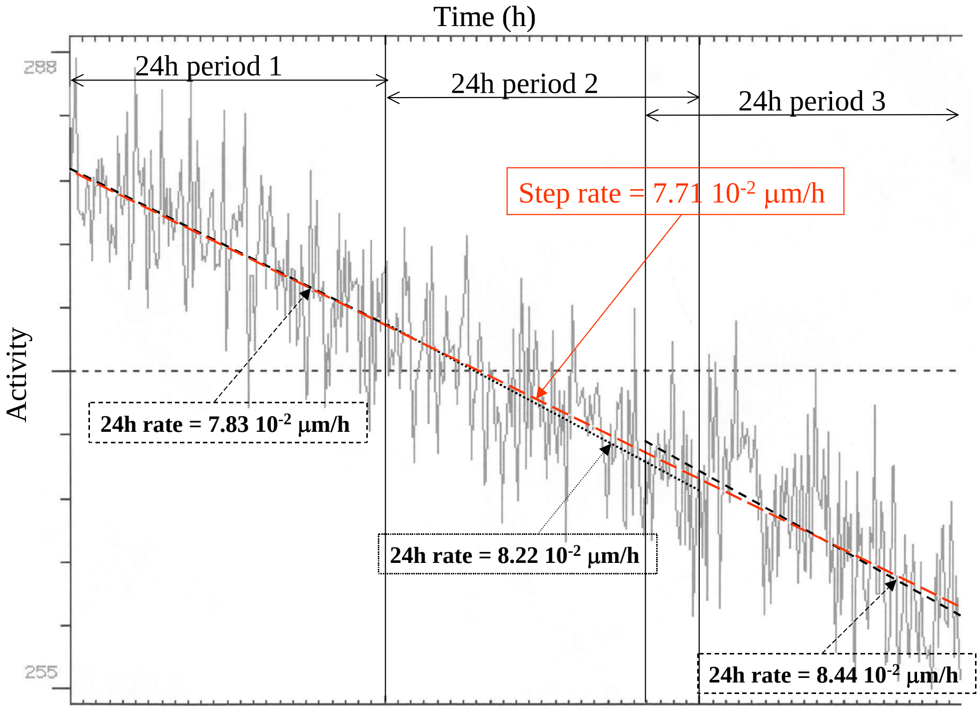

Figure 4-7
Activity of Specimen 2 Versus Time Measured During Step 3 of Test 1

The high variability in absolute FAC rate results could be due to several parameters:

- material differences from one specimen to the other,
- different initial surface states obtained after having suppressed the oxide at neutral pH and 180°C before starting the tests,
- slight differences in residual oxygen content (but always very low : < 0.5 ppb),

- differences between intended values and measured values (maximum temperature difference = 3.7°C (2%) - maximum water velocity difference = 0.2 m/s (4%) – maximum conductivity difference = 0.6 µS/cm (20%)). For example, a conductivity difference of 0.6 µS/cm at $pH_{235°C}$ = 9.0 can result[4] in a $pH_{235°C}$ difference of approximately 0.05 pH units if it's due to an inaccurate ammonia concentration and approximately 0.15 pH units if it's due to sodium contamination (approximately 76 ppb) [6]. If equation (1-3) is correct and the FAC rate is proportional to equilibrium iron solubility, then a pH change of 0.15 pH units could result in a FAC rate change of 50-100%.

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[4] pH calculations done thanks to MULTEQ using the database version 4:

T = 235°C - $[NH_3]$ = 265 µg/kg – $[N_2H_4]$ = 100 µg/kg -> conductivity = 2.74 µS/cm – pH_T = 5.90

T = 235°C - $[NH_3]$ = 351 µg/kg – $[N_2H_4]$ = 100 µg/kg -> conductivity = 3.33 µS/cm – pH_T = 5.95

T = 235°C - $[NH_3]$ = 265 µg/kg – $[N_2H_4]$ = 100 µg/kg – [Na] = 76 µg/kg -> conductivity = 3.33 µS/cm – pH_T = 6.05

5

DISCUSSION

5.1 Comparison between Previous and Recent Tests

Unlike previous tests, recent tests were conducted by step increasing the hydrazine concentration and then step decreasing it back to zero, when possible. As observed previously (see section 4), the absolute FAC rates measured without hydrazine at the beginning of the tests were always very different from the absolute FAC rates measured without hydrazine at the end of the tests. Therefore, the comparison between previous (which results are reported in Appendix D) and recent relative FAC rates could be done in two ways:

1. old definition of the relative FAC rate = absolute FAC rate/average value of all absolute FAC rates measured without hydrazine during the test,
2. new definition of the relative FAC rate = absolute FAC rate/absolute FAC rate measured during the first step of the test without hydrazine.

Relative FAC rates obtained during the previous tests and the recent ones are compared for tests conducted under the same experimental conditions : at $pH_{25°C}$ 9.0 and 235°C (Figure 5-1) with the old definition of the relative FAC rate and Figure 5-2 with the new definition of the relative FAC rate). This comparison illustrates the fact that the aim of the recent tests to reduce the scatter in data by testing two specimens in parallel and by having longer step duration was not reached. As observed in both Figure 5-1 (old definition) and Figure 5-2 (new definition), the recent test results seemed to add to the data scatter when the latest 5 m/s results are included. However, the latest 10 m/s results are generally within the data scatter in Figure 5-2 (new definition), and are relatively independent of hydrazine concentration, i.e., remain at a relative FAC rate of 1 for all tested hydrazine concentrations, in Figure 5-1 (old definition).

To try to understand the scatter, the differences in flow rate conditions during the tests were examined. A first analysis showed that the hydrazine effect did seem different for tests conducted under a 5 m/s water velocity as compared to tests conducted under a 10 m/s water velocity (see Figure 5-1 and Figure 5-2). However, the latest 5 m/s results only include steps with an initial zero hydrazine step, but no subsequent zero hydrazine step. Alternately, all of the latest 10 m/s results, with the exception of Test 1, had at least two steps at zero hydrazine with most of those having them both at the beginning and the end of the test. This fact is important because these latest tests clearly show that the final step at zero hydrazine is often the one with the highest FAC rate. Including this step by using it to calculate an average FAC rate at zero hydrazine effectively reduces the calculated relative FAC rates as compared to only using the initial zero hydrazine step. The impact of including the additional zero hydrazine step can be seen by comparing the previous results at 5 m/s and approximately 150 µg/kg hydrazine in Figure 5-2 with the corresponding data in Figure 5-1.

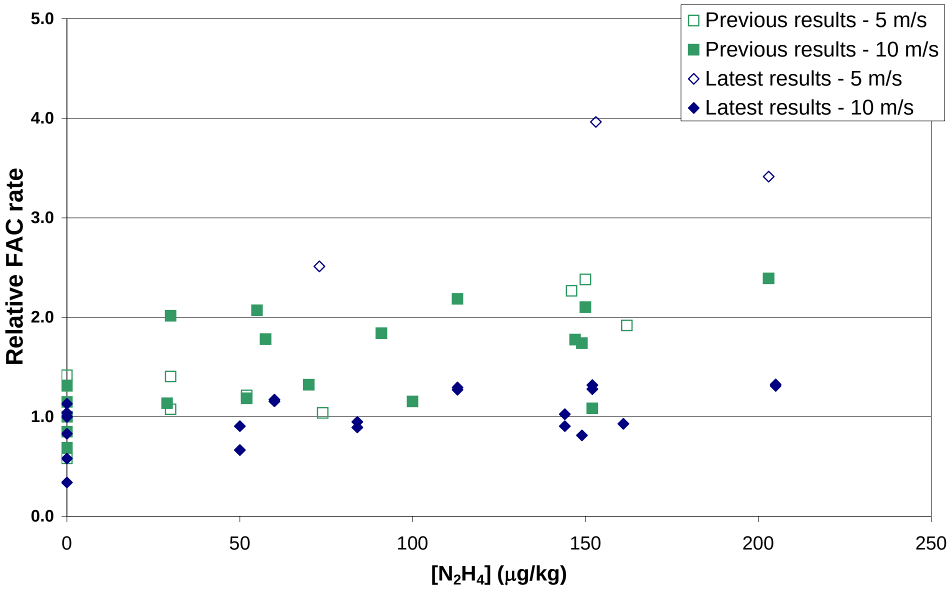

Figure 5-1
Relative FAC Rates (Old Definition) Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow at 235°C at $pH_{25°C}$ 9.0

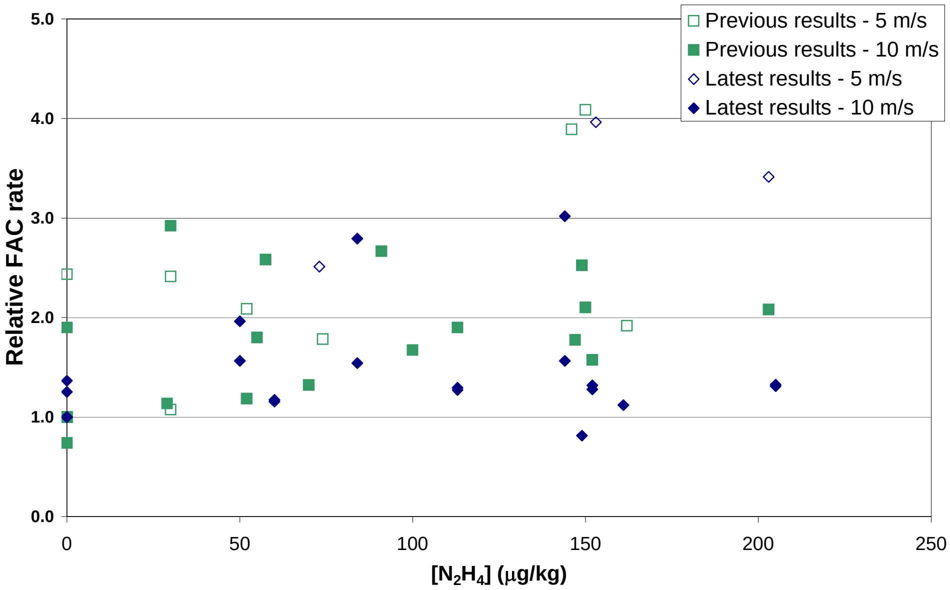

Figure 5-2
Relative FAC Rates (New Definition) Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow at 235°C at $pH_{25°C}$ 9.0

All the FAC results, both previous to this report (see section 2) and presented in this report (see section 4), showed that the hydrazine effect observed during different tests varied significantly from one test to the other and strongly depended on the experimental conditions. Test steps conducted in similar conditions (same water velocity, conductivity and temperature) gave very scattered absolute FAC rates (Figure 5-3).

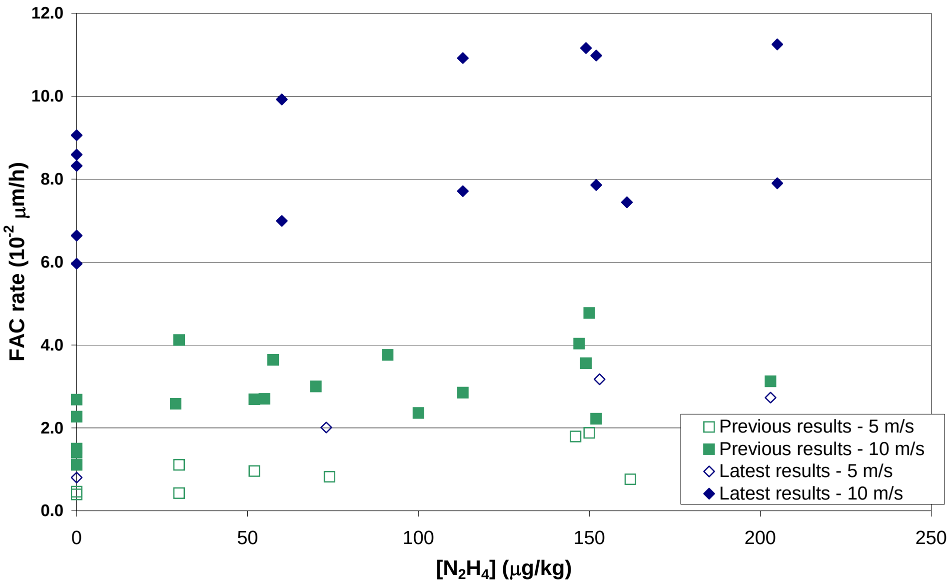

Figure 5-3
Absolute FAC Rates Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow at 235°C at $pH_{25°C}$ 9.0

5.2 Hydrazine Effect at Different pH Conditions

The comparison between the results obtained during tests on two specimens in parallel at 235°C with a water velocity of 10 m/s at three different $pH_{25°C}$ values (9.0, 9.3 and 9.5) during the hydrazine step increase is presented both on Figure 5-4 (absolute FAC rates) and on Figure 5-5 (relative FAC rates). These figures only show the FAC rates and relative FAC rates obtained during the first steps when the hydrazine concentration was step increased from 0 to approximately 150 µg/kg. As discussed in section 5.3, both the pH 9.3 and 9.5 tests showed continuously increasing absolute FAC rates with the final 70 µg/kg and 0 µg/kg hydrazine steps, which were higher than all previous FAC values – even those at higher hydrazine concentrations.

Figure 5-4 shows the hydrazine influence of the FAC rate followed a similar trend at all three studied pHs: when the hydrazine concentration step increased from 0 to 150 µg/kg, the FAC rate tended to slightly increase. It also shows the absolute FAC rates measured at $pH_{25°C}$ 9.0 were always higher than those measured at $pH_{25°C}$ 9.5, as one would expect. However, the measured FAC rates at $pH_{25°C}$ 9.5 were higher than those at $pH_{25°C}$ 9.3 in most cases. This is not totally coherent, as FAC rates should monotonously decrease with a pH increase. But, as it has previously been observed, the variability associated with the FAC rate measurements was quite high. Taking this variability into consideration (Figure 5-6), the effect of pH on the FAC results seem coherent as the FAC rates tended to decrease monotonously from $pH_{25°C}$ 9.0 to 9.5. Also, as noted previously, slight material differences between those specimens used at $pH_{25°C}$ 9.3 and 9.5 could have contributed to this observation.

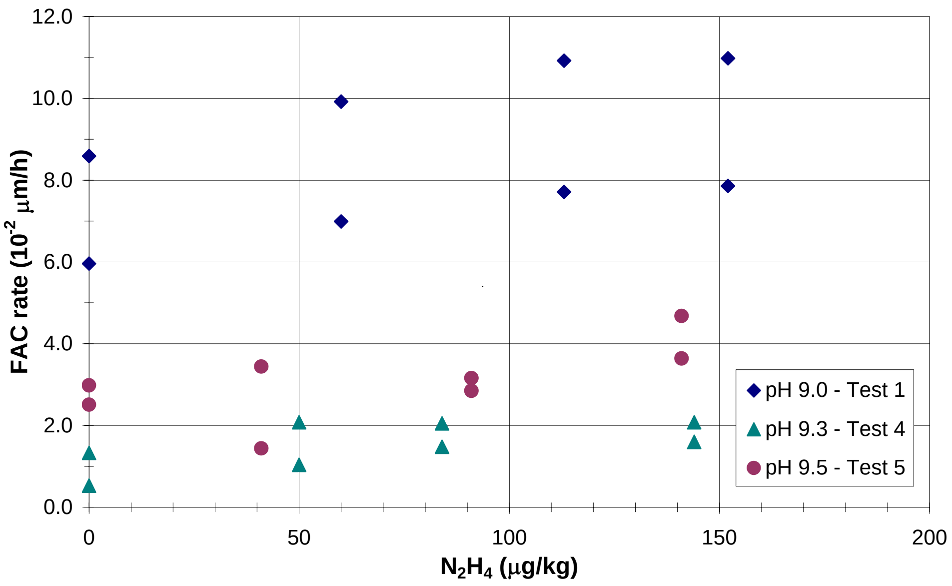

Figure 5-4
FAC Rates Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow with a Water Velocity of 10 m/s at 235°C at Different $pH_{25°C}$

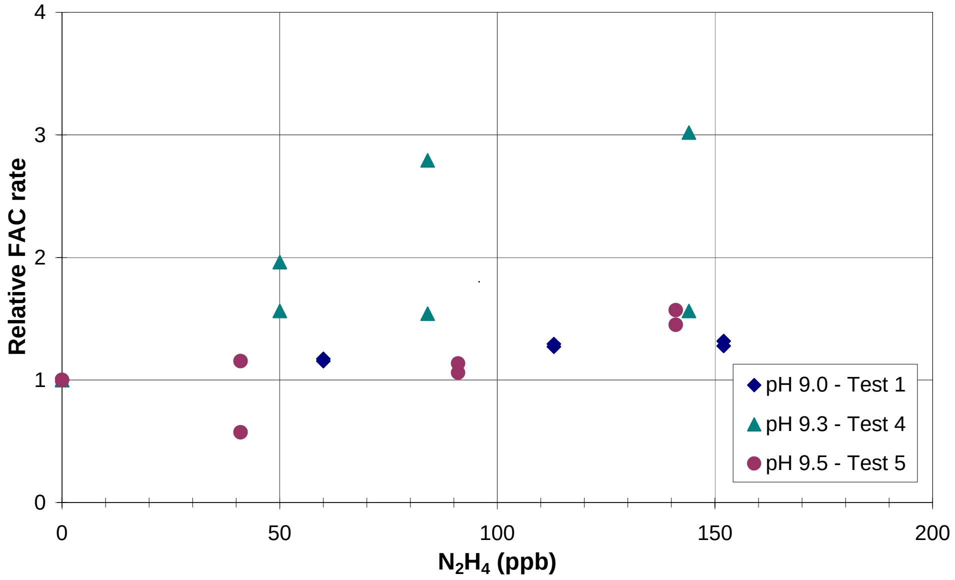

Figure 5-5
Relative FAC Rates Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow with a Water Velocity of 10 m/s at 235°C Using at Different $pH_{25°C}$

Figure 5-5 shows the increase of the relative FAC rate when the hydrazine concentration was increased. This increase was quite small at all tested $pH_{25°C}$ conditions of 9.0, 9.3 and 9.5 (maximum relative FAC rates close to 1.3, 1.6 and 1.6, respectively), except for the more pronounced effect of one of the parallel specimens at $pH_{25°C}$ 9.3 (maximum relative FAC rate of 3.0). This anomaly is not understood, but it is speculated that this could, in part, be due to the inherent uncertainty in the very low (and not satisfactory) absolute FAC rate (see section 3.3) measured during the first step (without hydrazine)[5]. The measured value was 0.53 10^{-2} µm/h, lower than the desired minimum of 10^{-2} µm/h. If the average zero hydrazine FAC rate had been used as the reference point in this plot, the relative rates would have been considerably lower.

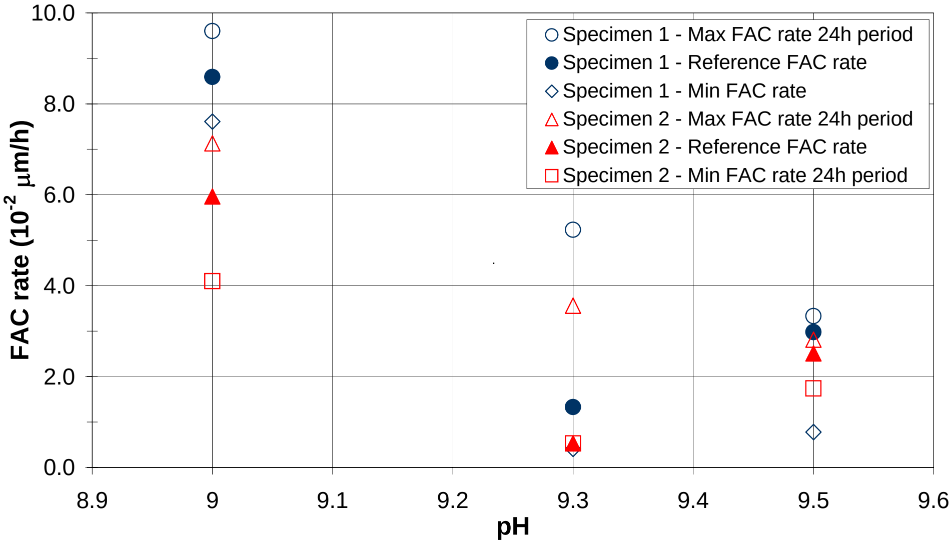

Figure 5-6
FAC Rates Measured Without Hydrazine Versus $pH_{25°C}$ for Carbon Steel Specimens Exposed to a Single-Phase Flow with a Water Velocity of 10 m/s at 235°C

5.3 Hydrazine Effect at Different Temperature Conditions

Figure 5-7 shows the hydrazine effect (during the hydrazine step increase only) on FAC of carbon steel using ammonia at $pH_{25°C}$ 9.0 under a 5 m/s water velocity at three different temperatures: 180°C, 210°C and 235°C:

- the results at 180°C, obtained during previous tests [2], show the relative FAC rate slightly decreased when the hydrazine concentration increased up to 150 µg/kg,
- the results at 210°C show a very slight increase of the relative FAC rate when the hydrazine concentration was increased up to 140 µg/kg,
- the results at 235°C, both including previous tests and recent tests, show a more pronounced increase of the relative FAC rate when the hydrazine concentration was increased up to 200 µg/kg.

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[5] Such a low absolute FAC rate could be due to a remaining residual oxide at the surface of the specimen.

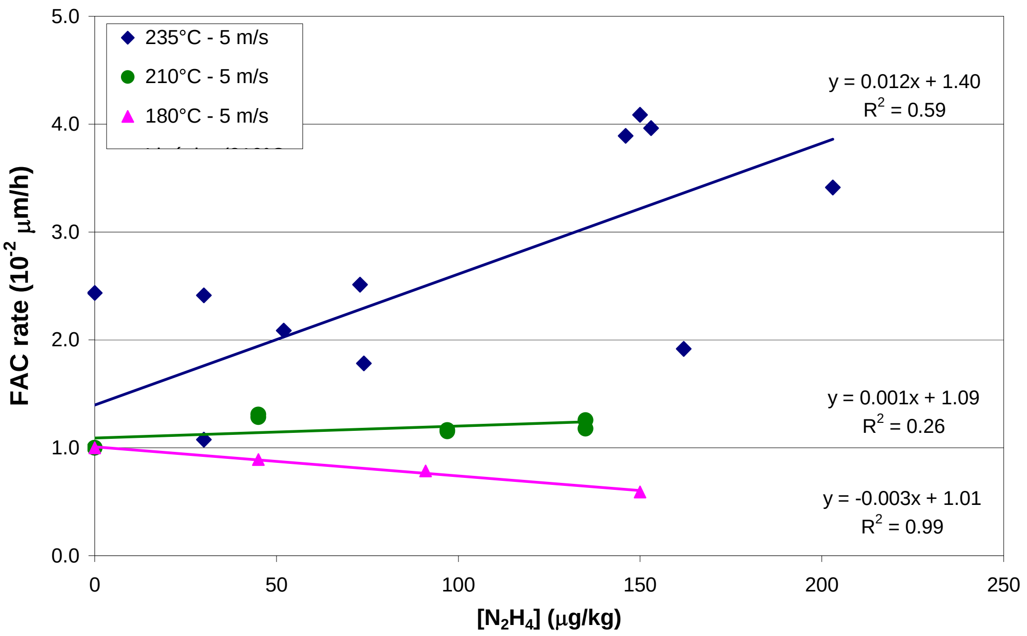

Figure 5-7
Relative FAC Rates Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow with a Water Velocity of 5 m/s at pH$_{25°C}$ 9.0 at Different Temperatures

Figure 5-8 shows the equivalent figure to Figure 5-7 for a water velocity of 10 m/s. At such water velocity, the only two studied temperatures were 180°C and 235°C. In both cases, the hydrazine effect was less pronounced than at a 5 m/s water velocity:

- the relative FAC rate slightly decreased (slightly less at 10 m/s than at 5 m/s) when the hydrazine concentration increased up to 197 µg/kg at 180°C,
- the relative FAC rate increased (much less pronounced at 10 m/s than at 5 m/s) when the hydrazine concentration increased up to 205 µg/kg at 235°C.

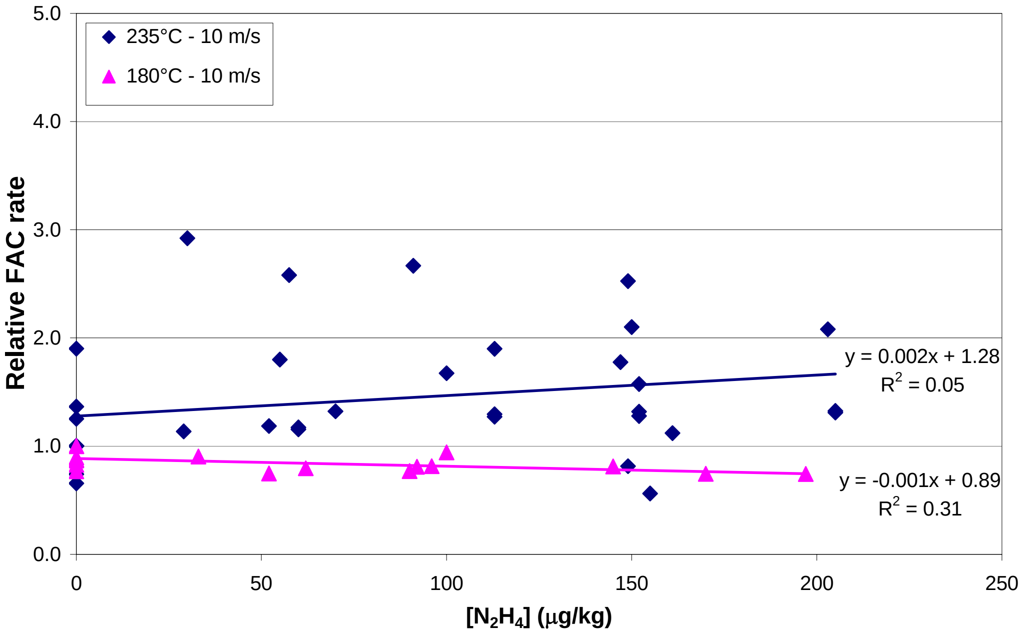

Figure 5-8
Relative FAC Rates Versus Hydrazine Concentration for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow with a Water Velocity of 10 m/s at pH$_{25°C}$ 9.0 at Different Temperatures

The slight decrease observed in the FAC rate with increasing hydrazine concentration at 180°C at both water velocities can be explained by the direct effect of hydrazine on pH. Increasing the hydrazine concentration increases pH, thus reducing the FAC rate. However, the very slight increase in FAC rate observed at 210°C and the relatively larger increase in FAC rate at 235°C cannot be explained by such an effect. The hydrazine effect at these two temperatures could be indirect, i.e. due to the hydrogen and/or the ammonia produced from the thermal decomposition of hydrazine, significant for temperatures above 205°C [5]:

$$2\,N_2H_4 \Leftrightarrow N_2 + H_2 + 2NH_3 \qquad \textbf{Equation 5-1}$$

This hypothesis is consistent with the above results showing a slight effect at 210°C and a more pronounced effect at 235°C. However, Figure 3-1 indicates that only a very small amount of hydrogen and ammonia (a few ppb or less) are produced on transport from the make-up tank to the test specimen as a result of hydrazine decomposition at higher temperatures.

Moreover, the trend observed at 235°C at pH$_{25°C}$ 9.3 and 9.5, showing the FAC rate did not decrease when the hydrazine concentration was step decreased towards the end of the tests (Figure 5-9 and Figure 5-10),was also observed during the test at 210°C (Figure 5-11). This trend, discussed elsewhere (see section 5.5), was less pronounced during Test 6 (pH$_{25°C}$ 9.0 – 210°C) than during Test 5 (pH$_{25°C}$9.5 – 235°C) and was most pronounced during Test 4 ((pH$_{25°C}$ 9.3 – 235°C).

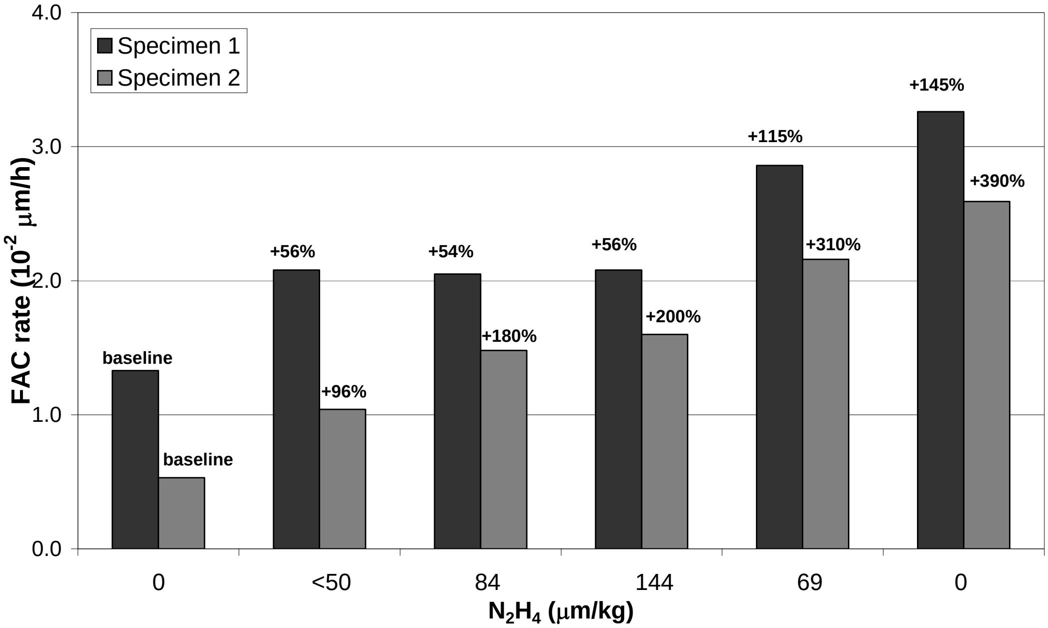

Figure 5-9
Evolution of the FAC Rates Measured During Test 4 (Water Velocity of 10 m/s – Temperature of 235°C - $pH_{25°C}$ = 9.3)

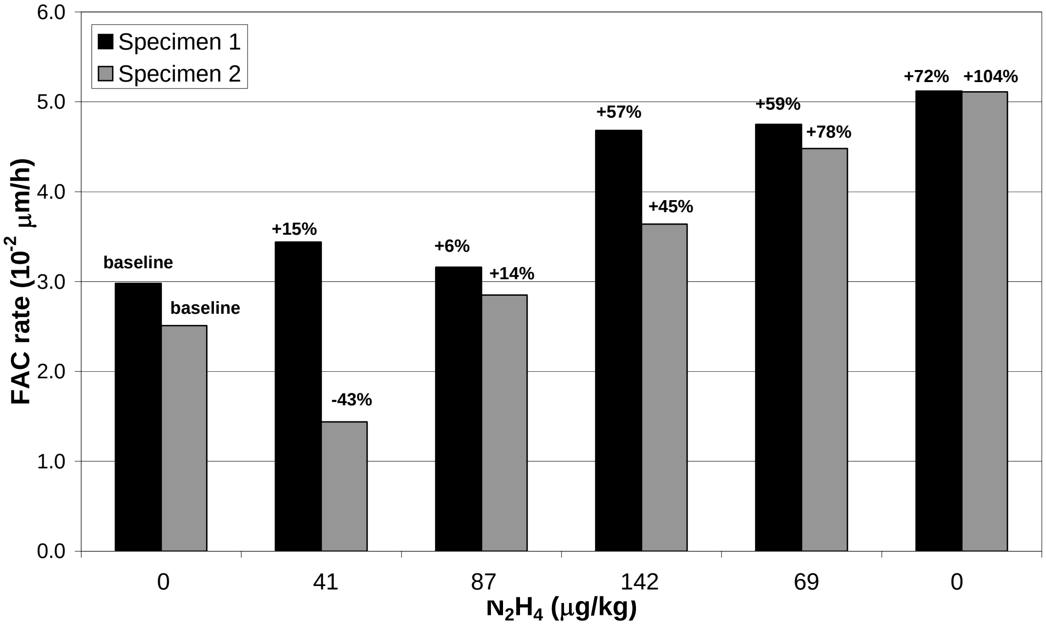

Figure 5-10
Evolution of the FAC Rates Measured During Test 5 (Water Velocity of 10 m/s – Temperature of 235°C - $pH_{25°C}$ = 9.5)

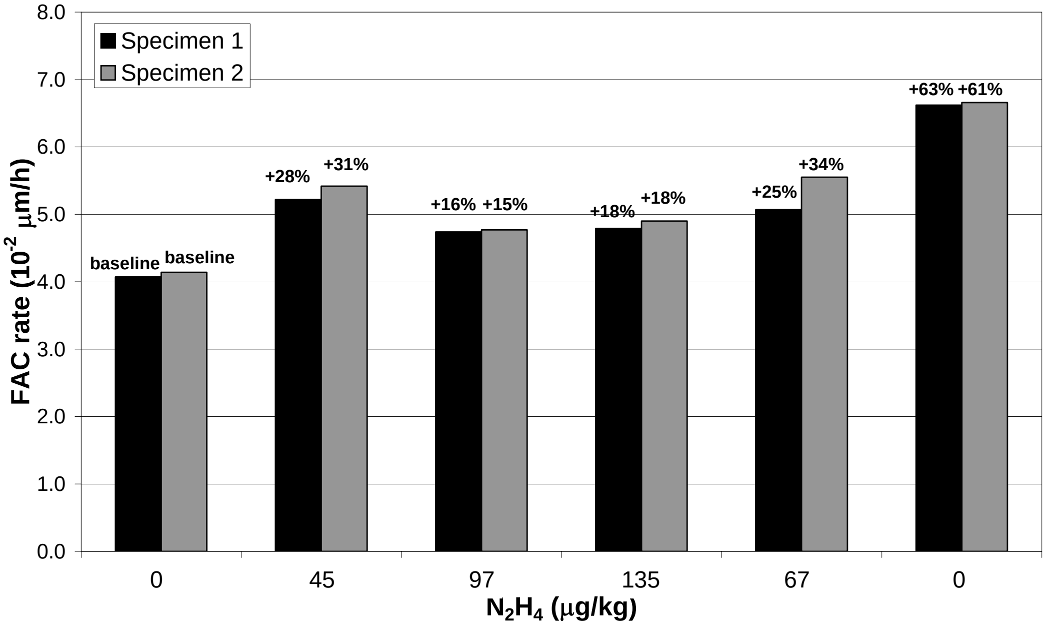

Figure 5-11
Evolution of the FAC Rates Measured During Test 6 (Water Velocity of 5 m/s – Temperature of 210°C - $pH_{25°C}$ = 9.0)

5.4 Impact of the FAC Rate without Hydrazine

It is interesting to note that the hydrazine effect observed at 235°C seemed to increase as the absolute FAC rates decreased. This was checked by comparing the relative FAC rates (based on the initial zero hydrazine step) at approximately 150 µg/kg hydrazine (± 15 µg/kg) versus the FAC rates without hydrazine during the initial runs obtained for the tests carried out on the CIROCO loop at 235°C (Figure 5-12). Data at 210°C and 180°C were also added for comparison. The 150 µg/kg hydrazine concentration was chosen because previous results showed the maximum effect of hydrazine on FAC rates was obtained for such a value (see Figure 2-1). This trend was confirmed at 235°C, at all tested $pH_{25°C}$ values (9.0, 9.3 and 9.5) and at both tested water velocities (5 m/s and 10 m/s). This tends to suggest that the slight differences in hydrazine effects at these three different pH values may be due to differences in the absolute FAC rates. The reason why hydrazine may have a more pronounced effect in conditions where the absolute FAC rate is lower is not yet understood.

As indicated previously, FAC rates observed in operating plant are normally less than 3 10^{-2} µm/h. In these conditions, the hydrazine effect can potentially be significant at 235°C (relative FAC rate increase from zero (no effect) to as high as a factor of 3 for a hydrazine concentration change from 0 µg/kg to 100 µg/kg). The purpose of taking this effect into account for the modeling of FAC would depend on the uncertainty of predicted FAC rates, which was also significant in this testing program. If this uncertainty is high, it might not be judicious or appropriate to model the hydrazine effect.

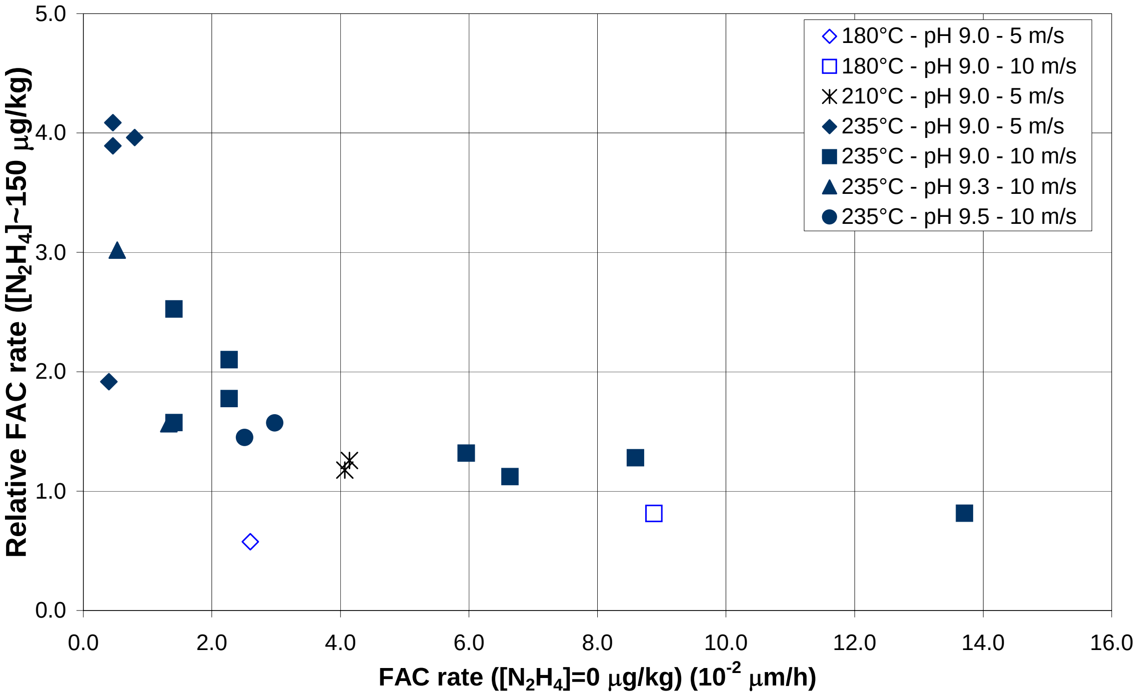

Figure 5-12
Relative FAC Rates (Based on the Initial Zero Hydrazine Step) Obtained at Hydrazine Concentrations of Approximately 150 µg/kg Versus FAC Rates Obtained Without Hydrazine for Tubular Carbon Steel Specimens Exposed to a Single-Phase Flow with Water Velocities of 5 m/s or 10 m/s at 180°C, 210°C and 235°C at Different $pH_{25°C}$

Hopefully, in the most worrying cases of FAC, e.g., where the wall thinning rate would be higher than 0.3 µm/h, the hydrazine effect should be negligible.

5.5 Continuous Increase of the FAC Rate

During Tests 4, 5 and 6, FAC rates tended to increase as the hydrazine concentration was step increased, as well as during the hydrazine concentration step decrease down to zero (Figure 5-9, Figure 5-10, Figure 5-11). As a matter of fact, if the experiments had stopped after the fourth run (corresponding to the maximum hydrazine concentration tested), one could only have observed a slight and general increase of the FAC rate with the hydrazine concentration for both specimens, in coherence with previous results.

The reason why the absolute FAC rate did not decrease while the hydrazine concentration was step decreased during the second stage of these tests is not clear. One possible explanation is that it is due to an increase of the oxide porosity in presence of hydrazine. The sensitivity of the material to FAC would consequently have been aggravated. This would mean the effect of hydrazine may need to be considered in an integrated way, i.e., the hydrazine to be considered would not just be the immediate hydrazine concentration but the time-weighted integration of hydrazine concentrations the specimen would have been in contact with during the whole test. However, this is simply a theory that appears to be coherent with the test results.

Figure 5-13 shows the evolution of all FAC rates measured during 24 hour periods (24h rates) as well as the overall FAC rates measured during each step (Step rates) for Test 6 (210°C). It illustrates the high variability in 24h FAC rates even at a constant hydrazine concentration. The FAC rate was quite variable during each step and did not stabilize even after 7 days during the last step to zero hydrazine. Figure 5-13 also shows that linear extrapolations over the whole period of the test give similar trends both when calculated with the 24h FAC rates and with the step FAC rates, with a better correlation factor, R^2, in the latter case. Although the step rates provided a better correlation than the 24h rates (true for all tests - see Appendix E), the data clearly indicate that longer steps and tests would be necessary to get more reliable FAC rates.

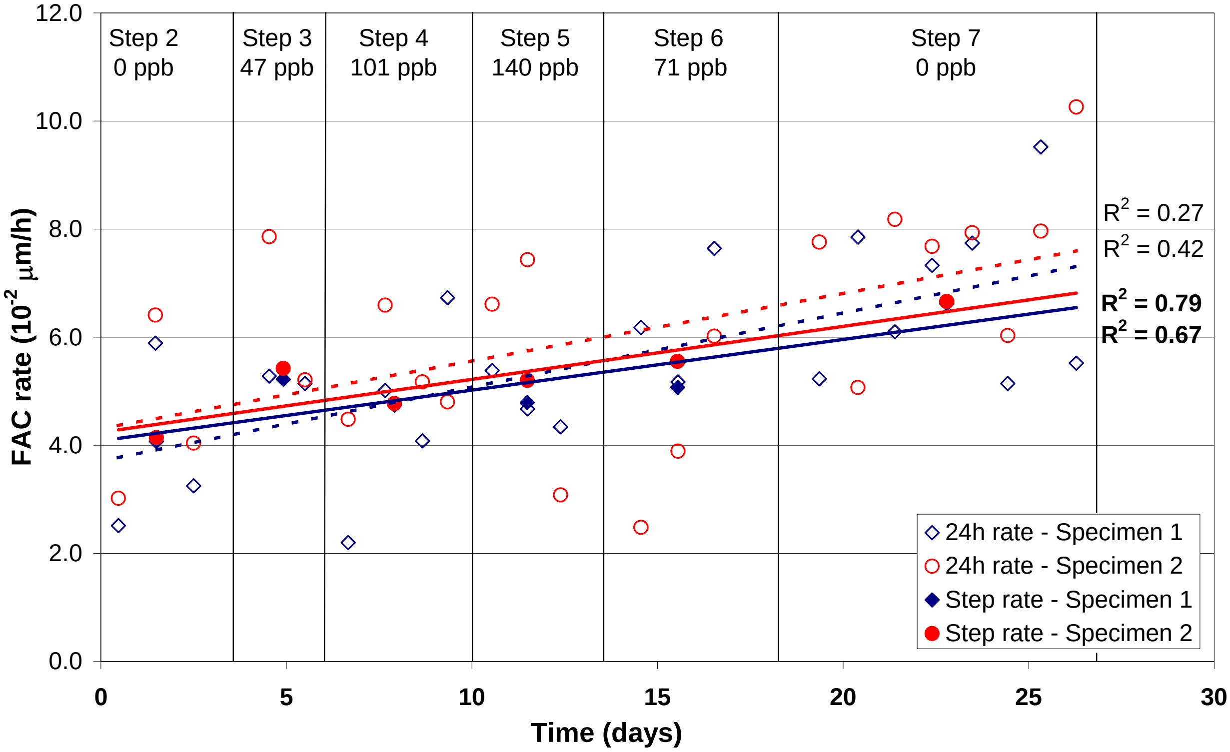

Figure 5-13
Evolution of FAC Rates Measured During 24 Hour Periods of Time and Over Step Periods on Carbon Steel Specimens Exposed to a Single-Phase Flow with a Water Velocity of 10 m/s at 210°C at $pH_{25°C}$ = 9.0 During Test 6

6

CONCLUSIONS

This report presents the latest tests conducted on the CIROCO loop where the hydrazine effect on FAC of carbon steel was studied. The purpose of the testing was to: (1) produce data with significantly less scatter than previously reported (see Figure 2-1); and (2) to help model the influence of hydrazine on FAC by better quantifying it.

The first objective was only partially met based on comparison of previous data to the three additional tests conducted as part of this latest program at $pH_{25°C}$ 9.0 and 235°C. The data scatter was significantly reduced when considering only the data at 10 m/s and using the same criteria for calculating relative FAC rates, i.e., based on the average of the zero hydrazine steps. In this case no significant effect of hydrazine on FAC was observed (see Figure 5-1). Alternately, when the relative FAC rate is based only on the initial zero hydrazine step, the scatter of the latest data and the previous data are similar (see Figure 5-2). However, two very important observations were identified in this latest testing that highlight the complexity of this issue. First, there is significant uncertainty in the measured FAC rates, i.e., large variability in the calculated FAC rates during each hydrazine concentration step. This is most likely a function of the relatively short duration at each condition relative to obtaining a stable oxide condition on the specimen and also to the variability in the chemistry conditions during the course of the tests (see Section 4). Second, FAC rates were observed to increase even when hydrazine was reduced from a high value back to zero. This observation is not understood and only hypothesized to potentially be caused by some long term effect (relative to the test duration) of hydrazine on the oxide porosity.

The second objective was also only partially met. Although it appears that there may be some increase of the FAC rate with increasing hydrazine concentration from 0 µg/kg to 150 µg/kg – 200 µg/kg at 235°C and $pH_{25°C}$ 9.0, the identification of significant uncertainty in the measured FAC rates and the observation of continued FAC increases even when hydrazine concentrations are decreasing creates significant doubt with regard to quantifying any hydrazine effect. The same observations and concerns apply to the results for $pH_{25°C}$ 9.3 and $pH_{25°C}$ 9.5 at 235°C. The results observed at 210°C and $pH_{25°C}$ 9.0 showed a similar effect of hydrazine on FAC rates as the data at 235°C, but to a much smaller degree.

The value of the relative FAC rate at approximately 150 µg/kg was measured in several pH, temperature and water velocity conditions:

- at 180°C, this value was consistently slightly lower than 1 at both 5 m/s and 10 m/s water velocities,
- at 210°C, this value was only slightly larger than 1 (about 20% larger) based on the first step of the test without hydrazine. However, it was slightly less than one based on the average of the zero hydrazine steps - at the start and end of the test. It appears that any effect is well within the scatter of the measure FAC rates (see Figure 4-6).

- at 235°C, this value may be larger than 1 (best estimate as much as 60% larger). However, the data scatter and observation of increasing FAC rate even when hydrazine is decreasing to zero makes any quantification highly uncertain.

The most surprising and perhaps important observation from this testing is that the FAC rates continued to increase even during the hydrazine concentration decrease in the latter part of the tests. This result is not yet understood, but is hypothesized to potentially be due to some effect of hydrazine on the oxide porosity of the carbon steel.

7

PERSPECTIVES

The study of the hydrazine effect should be completed by trying to better understand the observation of increasing FAC rates even in the presence of decreasing hydrazine concentrations. Longer tests would be necessary and should be followed-up with surface analysis. The hydrazine effect should also be studied with alternate amine chemistries, such as morpholine.

8
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A

PH AND CONDUCTIVITY OF THE ENVIRONMENTS USED FOR THE FAC TESTS

Table A-1
pH* and Conductivity of the Environments Used for the FAC Tests

T (°C)	NH_3 (µg/kg)	N_2H_4 (µg/kg)	$pH_{25°C}$	pH_T	Conductivity (µS/cm)	Corresponding Test-Steps
235	270	0	9.0	5.88	2.74	1-1 2-1 3-1 3-2 3-4 3-5 3-8
235	270	50	9.0	5.89	2.76	1-2
235	270	100	9.0	5.91	2.79	1-3
235	270	150	9.0	5.92	2.81	1-4
235	270	200	9.0	5.93	2.84	1-5
235	270	250	9.0	5.94	2.86	1-6
235	270	75	9.0	5.90	2.77	2-2
235	265	150	9.0	5.92	2.78	2-3 3-3 3-6 3-9
235	260	200	9.0	5.92	2.77	2-4
235	255	250	9.0	5.94	2.78	2-5
235	245	400	9.0	5.96	2.76	3-7
235	750	0	9.3	6.08	5.54	4-1 4-6
235	745	50	9.3	6.08	5.52	4.2
235	745	75	9.3	6.08	5.53	4-5
235	745	100	9.3	6.09	5.54	4-3
235	740	150	9.3	6.09	5.52	4-4
235	1560	0	9.5	6.23	8.77	5-1 5-6
235	1560	50	9.5	6.24	8.78	5-2
235	1560	75	9.5	6.24	8.78	5-5
235	1560	100	9.5	6.24	8.79	5-3
235	1560	150	9.5	6.24	8.79	5-4
210	270	0	9.0	5.88	2.74	6-1 6-2 6-7
210	270	50	9.0	6.02	2.76	6-3
210	270	75	9.0	6.03	2.77	6-6
210	265	100	9.0	5.90	2.75	6-4
210	265	150	9.0	5.92	2.78	6-5

* the pH values were calculated using the MulteQ code (V. 3.0 – database 12/00) [6].

B
EXAMPLES OF FAC RATE DETERMINATIONS BY THE TLA METHOD

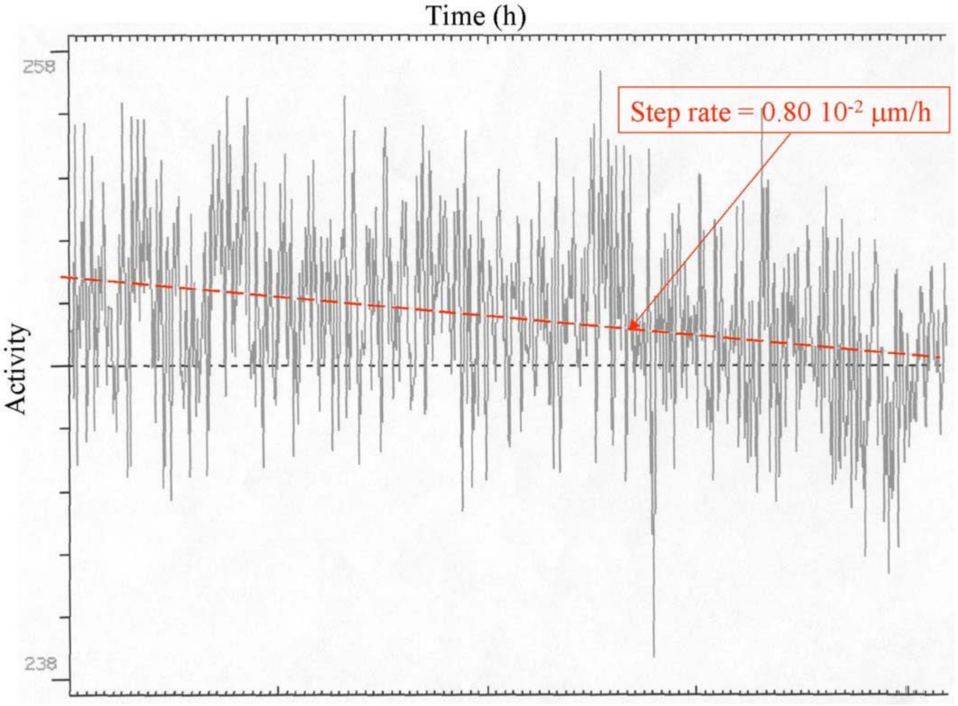

Figure B-1
Activity of Specimen 2 Versus Time Measured During Test 2 – Step 1 (66h Step)

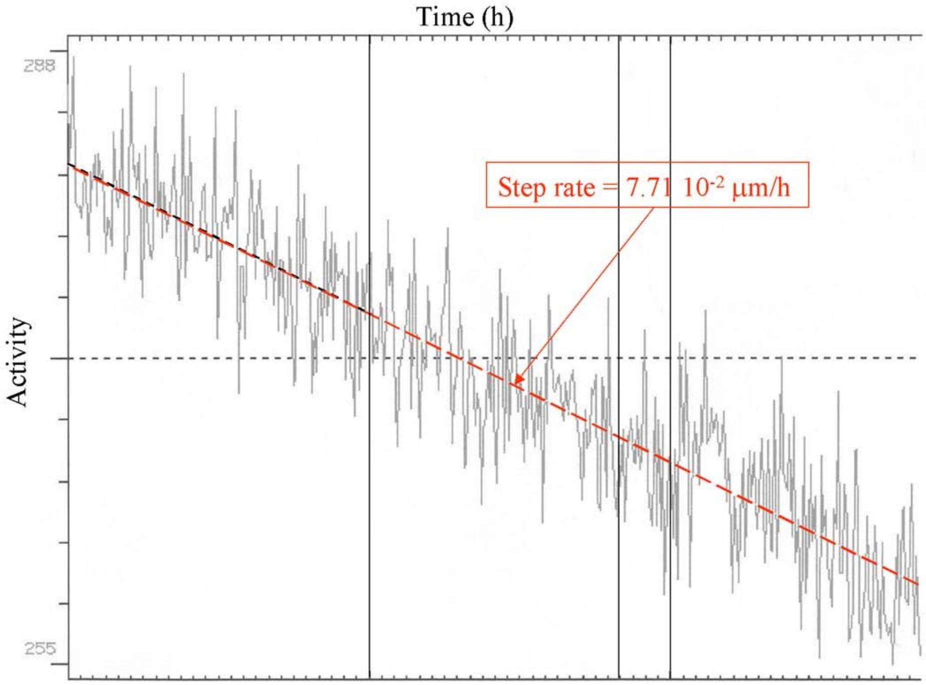

Figure B-2
Activity of Specimen 2 Versus Time Measured During Test 1 – Step 3 (68h Step)

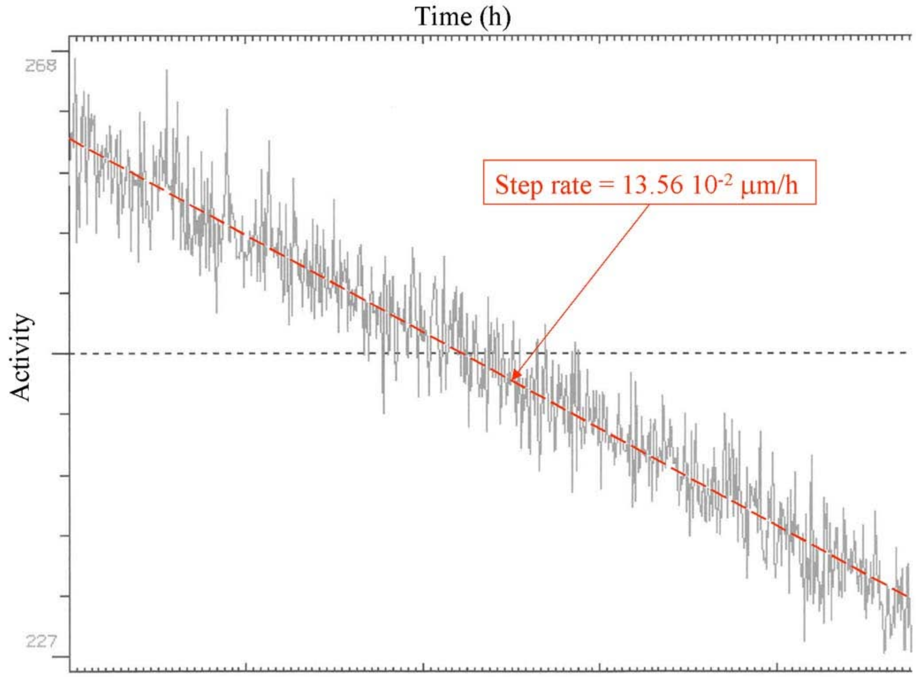

Figure B-3
Activity of Specimen 1 Versus Time Measured During Test 6 – Step 1 (114h Step)

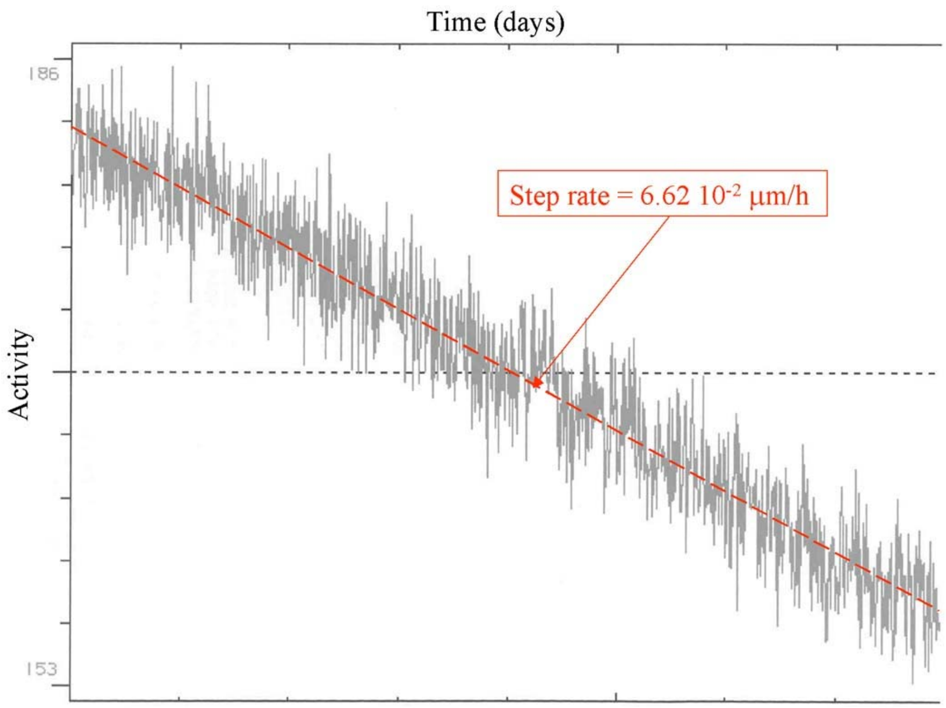

Figure B-4
Activity of Specimen 1 Versus Time Measured During Test 6 – Step 7 (191h Step)

C

EXPERIMENTAL CONDITIONS AND RESULTS OF THE TESTS

Legend

The calculated water velocities in the cells correspond to the one calculated at the blow down of specimen 1.
Values of Hydrazine concentration in italic: calculated thanks to equation (3-1)
Values not available: -

Table C-1
Experimental Conditions and Results of Test 1 (T = 235°C – $\mathrm{pH}_{25°C}$ = 9.0 – v = 10 m/s)

		Make-up Tank			Cells			Specimen 1				Specimen 2			
Test - Step #	Step duration	Conductivity*	$[N_2H_4]$	$[O_2]$	Water Velocity	$[N_2H_4]$	$[O_2]$	Temperature	FAC rate		Relative FAC rate	Temperature	FAC rate		Relative FAC rate
	h	µS/cm	ppb	ppb	m/s	ppb	ppb	°C	µm/h	mm/year		(°C)	µm/h	mm/year	
1 - 1	94	2.73	0	0.2	10.0	0	0.2	232.6	0.0859	0.752	1.00	234.8	0.0596	0.522	1.00
1 - 2	89	2.80	62	0.2	10.0	60	0.2	232.6	0.0992	0.869	1.15	234.8	0.0699	0.612	1.17
1 - 3	68	2.84	*117*	0.2	9.9	113	0.3	232.6	0.1092	0.957	1.27	234.8	0.0771	0.675	1.29
1 - 4	88	2.82	153	0.2	9.8	152	<0.5	232.6	0.1098	0.962	1.28	234.8	0.0786	0.689	1.32
1 - 5	71	2.83	209	0.2	9.8	205	0.2	232.6	0.1125	0.986	1.31	234.8	0.0790	0.692	1.33
1 - 6	90	2.84	257	0.2	9.8	256	0.2	232.6	0.1145	1.003	1.33	234.8	0.0771	0.675	1.29

* In this test, the ammonia concentration was not reduced when the hydrazine concentration was increased. Therefore, the conductivities were slightly higher than was initially planned. According to MULTEQ calculations, the difference in terms of temperature pH is only of 0.06 pH unit, which should be negligible in terms of FAC rates.

Table C-2
Experimental Conditions and Results of Test 2 (T = 235°C – $\mathrm{pH}_{25°C}$ = 9.0 – v = 5 m/s)

		Make-up Tank			Cells			Specimen 1 not exposed				Specimen 2			
Test - Step #	Step duration	Conductivity	$[N_2H_4]$	$[O_2]$	Water Velocity	$[N_2H_4]$	$[O_2]$	Temperature	FAC rate		Relative FAC rate	Temperature	FAC rate		Relative FAC rate
	(h)	µS/cm	(ppb)	ppb	(m/s)	(ppb)	ppb	(°C)	µm/h	mm/year		(°C)	µm/h	mm/year	
2 - 1	66	2.74	0	0.1	5.0	0	0.4					233.0	0.0080	0.070	1.00
2 - 2	64	2.73	73	<0.5	5.0	*69*	<0.5					233.0	0.0201	0.176	2.51
2 - 3	92	2.73	153	<0.5	5.0	*149*	<0.5					233.0	0.0317	0.278	3.96
2 - 4	70	2.74	203	0.1	5.0	*199*	0.3					233.1	0.0273	0.239	3.41
2 - 5	115	2.75	248	0.1	4.9	*244*	0.3					233.2	0.0481	0.421	6.01

Table C-3
Experimental Conditions and Results of Test 3* (T = 235°C – $pH_{25°C}$ = 9.0 – v = 10 m/s)

Test - Step #		Make-up Tank			Cells			Specimen 1 not exposed				Specimen 2			
	Step duration	Conductivity	$[N_2H_4]$	$[O_2]$	Water Velocity	$[N_2H_4]$	$[O_2]$	Temperature	FAC rate		Relative FAC rate	Temperature	FAC rate		Relative FAC rate
	(h)	µS/cm	(ppb)	ppb	(m/s)	(ppb)	ppb	(°C)	µm/h	mm/year		(°C)	µm/h	mm/year	
3 - 1	54	2.75	0	<0.5	9.9	0	0.1					233.4	0.0664	0.582	1.00
3 - 2	90	2.76	0	<0.5	10.0	0	0.1					233.5	0.0832	0.729	1.25
3 - 3	68	2.76	161	<0.5	10.0	*157*	0.1					233.4	0.0698	0.611	1.05
3 - 4	94	2.75	0	<0.5	9.9	0	0.1					233.4	0.0906	0.794	1.36
3 - 5	41	2.75	0	<0.5	9.9	0	0.1					233.6	0.1371	1.201	2.06
3 - 6	66	2.75	149	<0.5	10.0	*145*	0.1					233.6	0.1116	0.978	1.68
3 - 7	71	2.76	*389*	<0.5	10.0	386	<0.5					233.6	0.0832	0.729	1.25
3 - 8	70	2.73	0	<0.5	10.0	0	0.1					233.6	0.0900	0.788	1.36
3 - 9	70	2.74	*159*	<0.5	10.0	155	<0.5					233.6	0.0770	0.675	1.16

*Test 3 conducted in two periods: steps 1 to 4 during the first period and steps 6 to 9 during the second period.

Table C-4
Experimental Conditions and Results of Test 4 (T = 235°C – $pH_{25°C}$ = 9.3 – v = 10 m/s)

Test - Step #		Make-up Tank			Cells			Specimen 1				Specimen 2			
	Step duration	Conductivity	$[N_2H_4]$	$[O_2]$	Water Velocity	$[N_2H_4]$	$[O_2]$	Temperature	FAC rate		Relative FAC rate	Temperature	FAC rate		Relative FAC rate
	(h)	µS/cm	(ppb)	ppb	(m/s)	(ppb)	ppb	(°C)	µm/h	mm/year		(°C)	µm/h	mm/year	
4 - 1	97	5.54	0	0.6	10.0	0	0.1	232.7	0.0133	0.117	1.00	234.8	0.0053	0.046	1.00
4 - 2	114	5.53	50	0.5	10.1	*46*	0.1	232.7	0.0208	0.182	1.56	234.8	0.0104	0.091	1.96
4 - 3	92	5.52	91	0.5	10.1	85	0.1	232.7	0.0205	0.180	1.54	234.8	0.0148	0.130	2.79
4 - 4	92	5.54	149	0.5	9.9	143	0.1	232.7	0.0208	0.182	1.56	234.8	0.0160	0.140	3.02
4 - 5	72	5.53	74	0.5	9.9	69	0.1	232.7	0.0286	0.251	2.15	234.8	0.0216	0.189	4.08
4 - 6	87	5.51	0	0.5	9.8	0	0.1	232.7	0.0326	0.286	2.45	234.8	0.0259	0.227	4.89

Table C-5
Experimental Conditions and Results of Test 5 (T = 235°C – $pH_{25^{\circ}C}$ = 9.5 – v = 10 m/s)

		Make-up Tank			Cells			Specimen 1				Specimen 2			
Test - Step #	Step duration	Conductivity	$[N_2H_4]$	$[O_2]$	Water Velocity	$[N_2H_4]$	$[O_2]$	Temperature	FAC rate		Relative FAC rate	Temperature	FAC rate		Relative FAC rate
	(h)	µS/cm	(ppb)	ppb	(m/s)	(ppb)	ppb	(°C)	µm/h	mm/year		(°C)	µm/h	mm/year	
5 - 1	113	8.77	0	0.3	10.0	0	0.1	232.7	0.0298	0.261	1.00	234.8	0.0251	0.220	1.00
5 - 2	44	8.82	44	0.3	9.9	41	0.1	232.7	0.0344	0.301	1.15	234.8	0.0144	0.126	0.57
5 - 3	91	8.8	94	0.3	9.9	87	0.1	232.7	0.0316	0.277	1.06	234.9	0.0285	0.250	1.14
5 - 4	92	8.83	149	0.3	9.9	142	0.1	232.7	0.0468	0.410	1.57	234.9	0.0364	0.319	1.45
5 - 5	73	8.74	74	0.3	9.9	69	0.1	232.7	0.0475	0.416	1.59	234.9	0.0448	0.392	1.78
5 - 6	90	8.76	0	0.3	9.9	0	0.1	232.7	0.0512	0.449	1.72	234.9	0.0511	0.448	2.04

Table C-6
Experimental Conditions and Results of Test 6 (T = 210°C – $pH_{25^{\circ}C}$ = 9.0 – v = 10 m/s and 5 m/s)

		Make-up Tank			Cells			Specimen 1				Specimen 2			
Test - Step #	Step duration	Conductivity	$[N_2H_4]$	$[O_2]$	Water Velocity	$[N_2H_4]$	$[O_2]$	Temperature	FAC rate		Relative FAC rate	Temperature	FAC rate		Relative FAC rate
	(h)	µS/cm	(ppb)	ppb	(m/s)	(ppb)	ppb	(°C)	µm/h	mm/year		(°C)	µm/h	mm/year	
6 - 1	114	2.76	0	0.2	10.0	0	0.2	207.6	0.1356	1.188	1.00	209.8	0.1539	1.348	1.00
6 - 2	73	2.76	0	0.3	5.1	0	0.2	206.4	0.0407	0.357	0.30	208.5	0.0414	0.363	0.27
6 - 3	47	2.73	47	0.3	5.1	45	0.2	206.4	0.0522	0.457	0.38	208.5	0.0542	0.475	0.35
6 - 4	93	2.78	101	0.3	5.1	97	0.2	206.4	0.0474	0.415	0.35	208.5	0.0477	0.418	0.31
6 - 5	67	2.77	140	0.3	5.1	135	0.2	206.4	0.0479	0.420	0.35	208.5	0.0520	0.456	0.34
6 - 6	72	2.78	71	0.1	5.1	67	0.1	206.3	0.0507	0.444	0.37	208.5	0.0550	0.482	0.36
6 - 7	191	2.77	0	0.2	5.2	0	0.2	206.3	0.0662	0.580	0.49	208.5	0.0666	0.583	0.43

D

EXPERIMENTAL CONDITIONS AND RESULTS OF THE PREVIOUS TESTS [2] USED IN THIS REPORT

Test - Step #	Step duration	Conductivity	[N_2H_4]	[O_2]	Water Velocity	Temperature	FAC rate		Relative FAC rate
	h	µS/cm	ppb	ppb	m/s	°C	µm/h	mm/year	
1 - 1	61	3.42	0	0.1	5.1	236.0	0.0040	0.035	1.00
1 - 2	13	3.42	30	0.7	5.1	235.9	0.0043	0.038	1.08
1 - 3	18	3.42	162	0.4	5.1	236.0	0.0076	0.067	1.92
2 - 1	41	-	0	-	5.0	-	0.0046	0.040	1.00
2 - 4	69	3.49	146	0.1	5.2	235.7	0.0179	0.157	3.89
2 - 5b	50	-	74	0.1	5.2	235.7	0.0082	0.072	1.78
2 - 6	75	3.54	52	0.1	5.2	235.7	0.0096	0.084	2.09
2 - 7	56	3.54	30	0.1	5.2	235.7	0.0111	0.097	2.41
2 - 8	68	3.52	0	0.1	5.2	235.7	0.0112	0.098	2.43
2 - 12	46	2.84	150	0.1	5.2	235.7	0.0188	0.165	4.09
2 - 4b	11	3.49	150	-	10.0	-	0.0477	0.418	2.10
2 - 5a	19	-	70	0.2	10.5	-	0.0300	0.263	1.32
2 - 6b	18	3.54	52	0.1	10.5	236.0	0.0269	0.236	1.19
2 - 7b	19	3.52	29	0.1	10.5	236.0	0.0258	0.226	1.14
2 - 8b	20	3.40	0	0.1	10.5	236.0	0.0227	0.199	1.00
2 - 12b	23	2.95	147	0.1	10.6	236.0	0.0403	0.353	1.78
3 - 1	65	2.74	0	0.1	10.2	234.3	0.0141	0.124	1.00
3 - 3	90	2.69	0	0.1	10.2	234.3	0.0268	0.235	1.90
3 - 6	79	2.72	152	0.1	10.2	234.1	0.0222	0.194	1.57
3 - 7	72	2.68	100	0.1	10.2	234.2	0.0236	0.207	1.67
3 - 8	72	2.76	58	0.1	10.2	234.2	0.0364	0.319	2.58
3 - 9	86	2.69	149	0.1	10.2	234.4	0.0356	0.312	2.52
3 - 10	72	2.72	91	0.1	10.1	234.2	0.0376	0.329	2.67
3 - 11	71	2.69	30	0.1	10.0	232.8	0.0412	0.361	2.92
4 - 1	93	2.75	0	0.3	10.2	234.1	0.0150	0.131	1.00
4 - 2	69	2.77	55	0.2	10.2	234.1	0.0270	0.237	1.80
4 - 3	91	2.73	113	0.1	10.2	234.2	0.0285	0.250	1.90
4 - 4	54	2.72	203	0.1	10.2	234.2	0.0312	0.273	2.08
4 - 7	37	2.65	0	0.1	10.1	234.2	0.0111	0.097	0.74
6 - 1	117	2.72	0	0.5	10.1	180.3	0.0746	0.653	1.00
6 - 3b	87	2.73	100	0.3	10.1	180.3	0.0703	0.616	0.94
6 - 4	48	2.69	0	0.4	10.0	180.3	0.0668	0.585	0.90
6 -6b	69	2.70	0	0.5	10.0	180.3	0.0653	0.572	0.88
6 - 7	70	2.72	52	0.4	10.0	180.3	0.0558	0.489	0.75
6 - 8	74	2.74	170	0.4	9.8	180.3	0.0555	0.486	0.74
12 - 1	124	2.78	0	0.1	9.8	179.4	0.0888	0.778	1.00
12 - 2	46	2.72	33	0.2	9.5	179.4	0.0803	0.703	0.90
12 - 3	44	2.70	62	0.2	9.5	179.4	0.0706	0.618	0.80
12 - 4	62	2.72	92	0.3	9.5	179.4	0.0719	0.630	0.81
12 - 5	42	2.71	145	0.3	9.5	179.4	0.0722	0.632	0.81
12 - 6	47	2.74	197	0.3	9.5	179.4	0.0659	0.577	0.74
12 - 7	51	2.74	96	0.3	9.5	179.4	0.0723	0.633	0.81
12 - 8	42	2.73	0	0.3	9.4	179.4	0.0736	0.645	0.83
12 - 9	69	2.74	0	0.3	9.7	179.9	0.0772	0.676	0.87
12 - 10	65	-	90	<0.5	9.7	-	0.0683	0.598	0.77
12 - 11	26	-	0	<0.5	9.7	-	0.0682	0.597	0.77
13 - 2	64	2.74	0	0.1	5.0	178.3	0.0256	0.224	1.00
13 - 3	66	2.76	45	0.2	5.0	178.3	0.0228	0.200	0.89
13 - 4	92	2.76	91	0.2	5.0	178.3	0.0201	0.176	0.79
13 - 5	70	2.76	150	0.2	4.9	178.3	0.0151	0.132	0.59

E

EVOLUTION OF FAC RATES MEASURED DURING 24 HOUR PERIODS OF TIME AND OVER STEP PERIODS

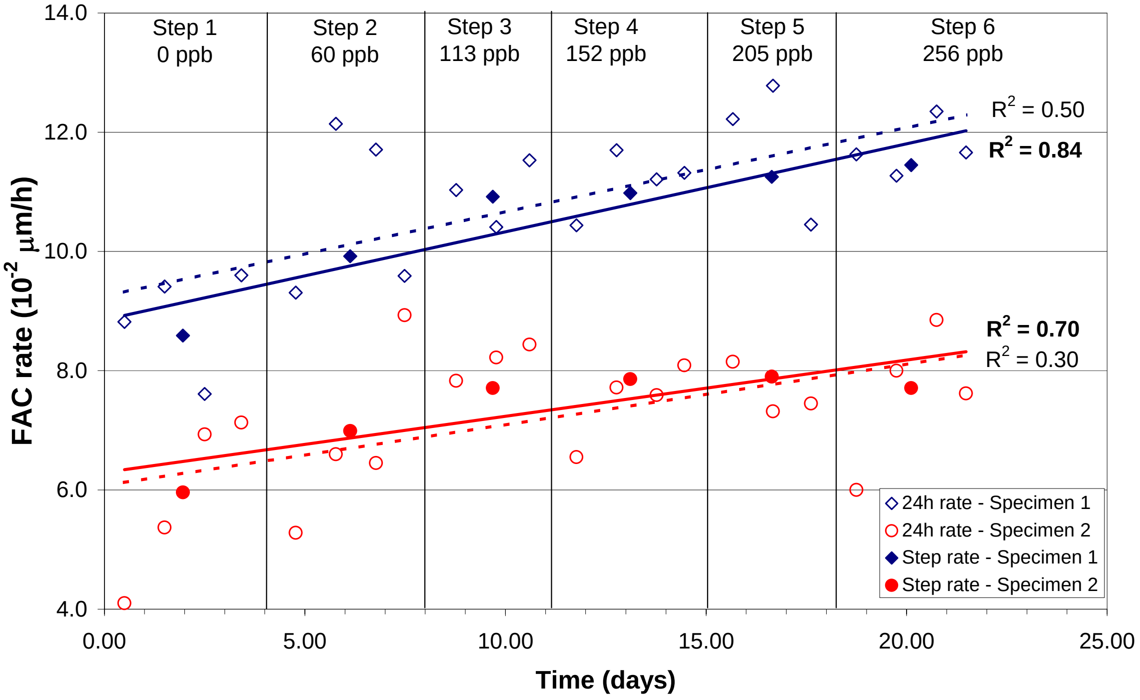

Figure E-1
Evolution of FAC Rates Measured During 24 Hour Periods of Time and Over Step Periods for Test 1

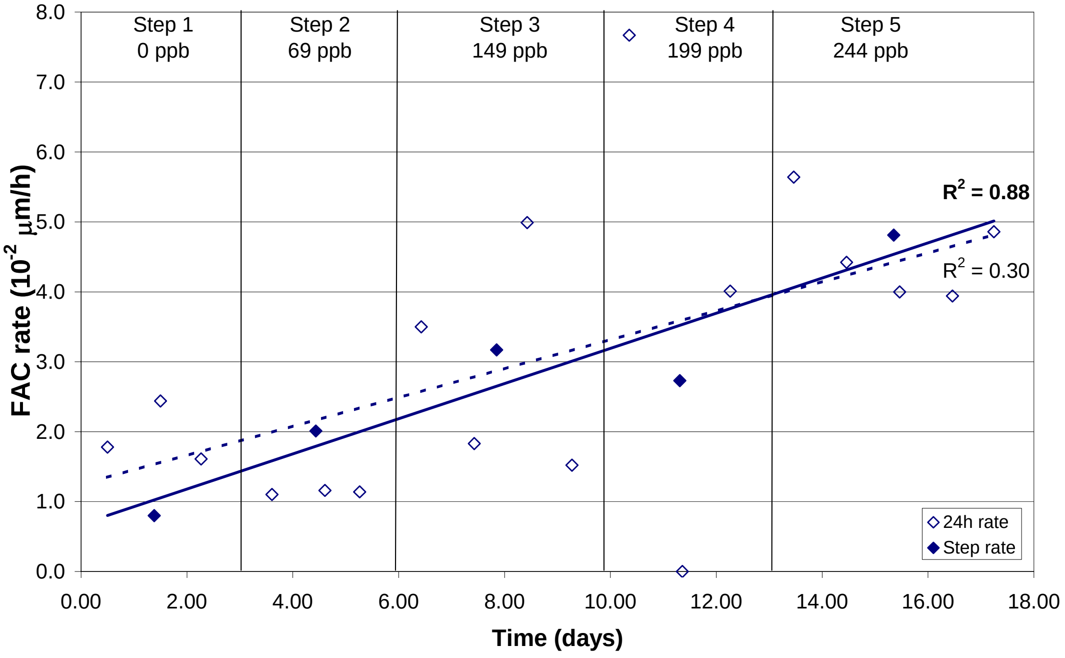

Figure E-2
Evolution of FAC Rates Measured During 24 Hour Periods of Time and Over Step Periods for Test 2

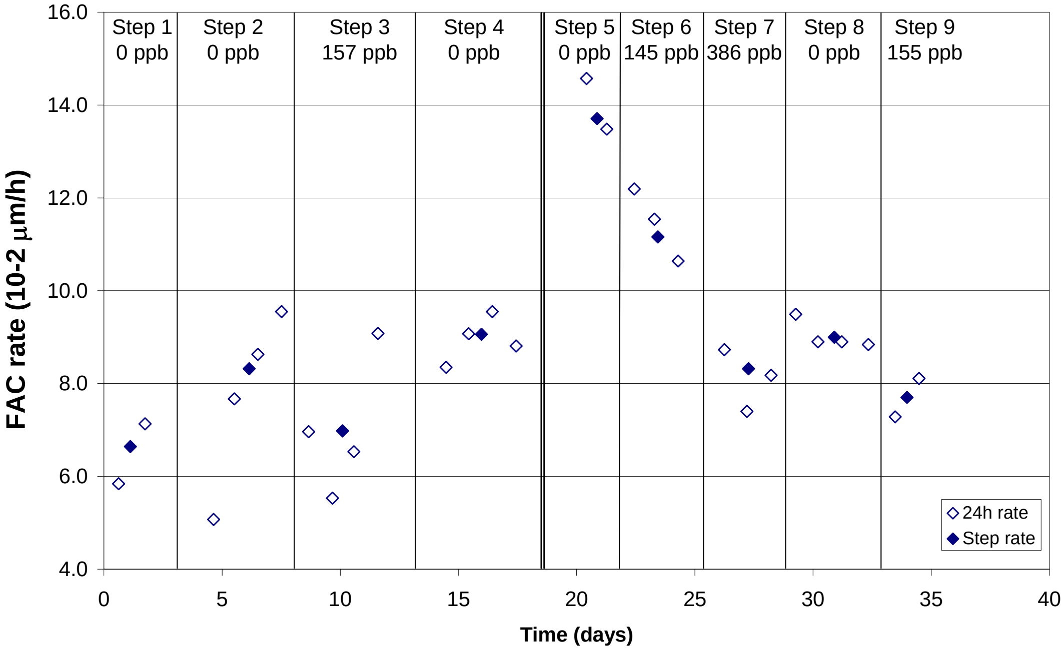

Figure E-3
Evolution of FAC Rates Measured During 24 Hour Periods of Time and Over Step Periods for Test 3

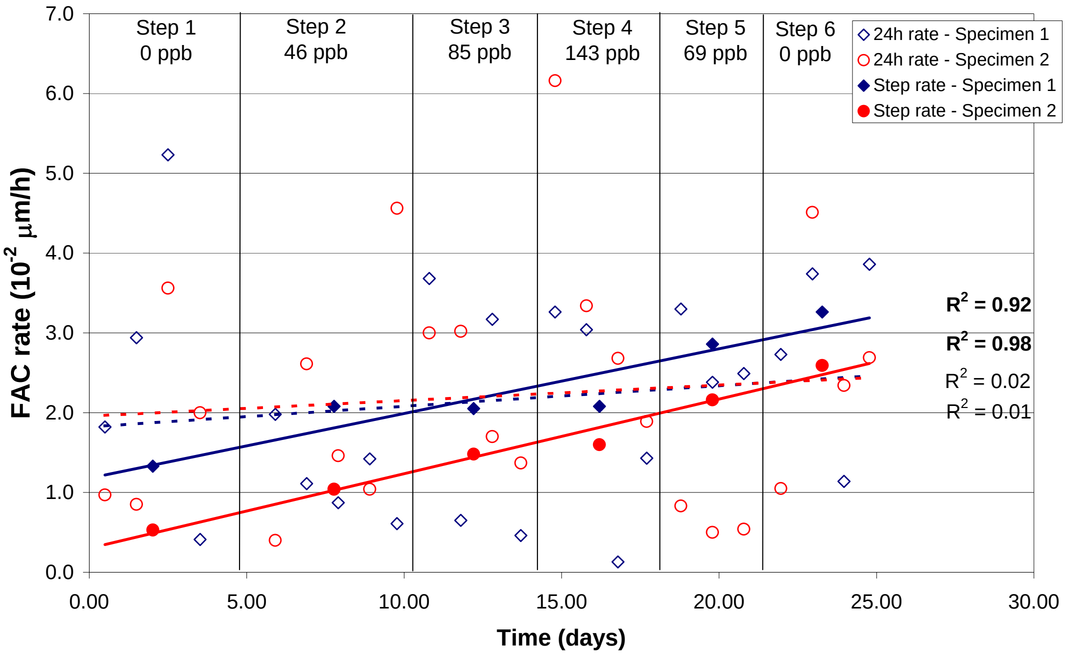

Figure E-4
Evolution of FAC Rates Measured During 24 Hour Periods of Time and Over Step Periods for Test 4

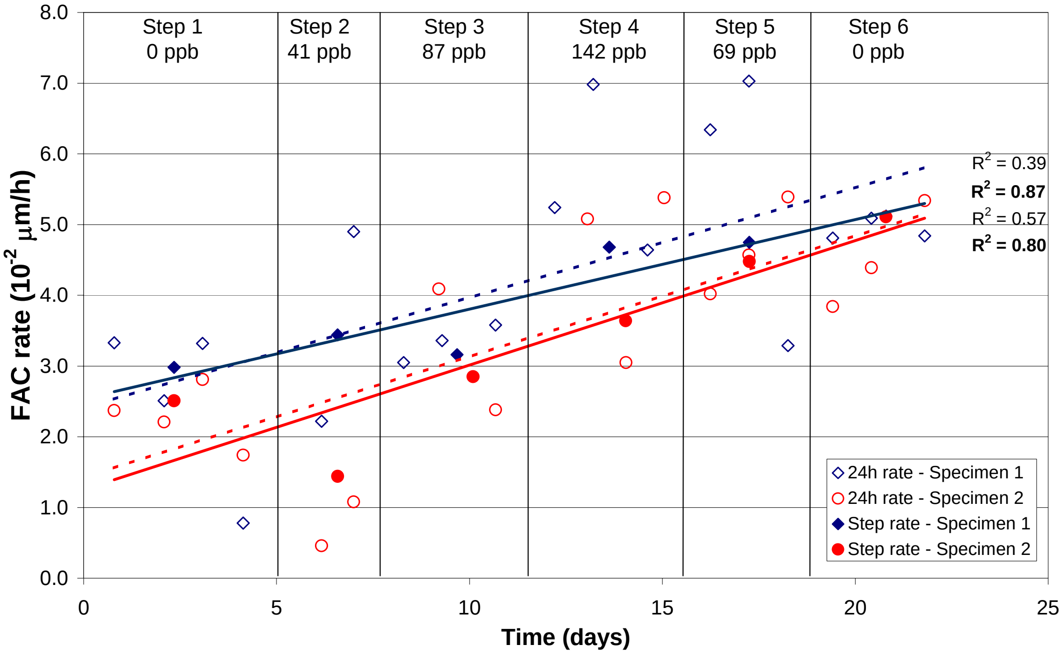

Figure E-5
Evolution of FAC Rates Measured During 24 Hour Periods of Time and Over Step Periods for Test 5

Evolution of FAC Rates Measured During 24 Hour Periods of Time and Over Step Periods

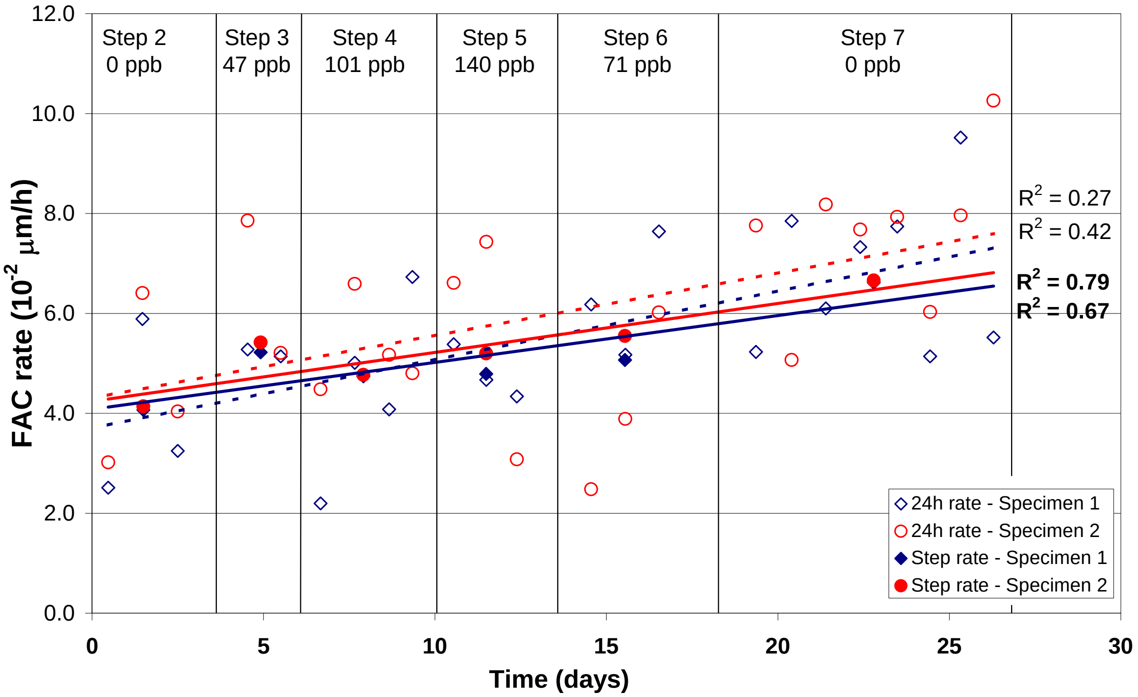

Figure E-6
Evolution of FAC Rates Measured During 24 Hour Periods of Time and Over Step Periods for Test 6

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Nuclear Power

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