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Predicted growth of through-thickness stress corrosion cracks in anhydrous ammonia nurse tanks

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ABSTRACT

Anhydrous ammonia is a toxic material that is transported from distribution centers to farm fields in steel pressure vessels called nurse tanks. Numerous accidents have occurred in which nurse tanks failed and ammonia was released, often with explosive force. The majority of such accidents are caused by stress corrosion cracking of the tank steel. Stress corrosion cracking is caused by the combination of stress in the tank's steel and the corrosive effect of ammonia. Neutron diffraction analysis was used to map the residual stress state in and near circumferential welds from two used anhydrous ammonia nurse tanks, one manufactured in 1966 and the other manufactured in 1986. Notched SA455 steel test specimens were held under tensile load (stress concentration factors of 40 to 80 MPa·√m) while immersed in NH_3 for seven months to generate crack propagation rate data. The results from these measurements were then used to predict stress corrosion crack growth rates for various pre-existing crack sizes at various temperatures. These data may be useful for estimating safe service lifetimes of nurse tanks that contain cracks. Copyright © 2015 VBRI Press.

Keywords: Hazardous material; stress corrosion cracking; welds; pressure-vessel failures; crack growth.

Introduction

Anhydrous ammonia (NH_3) is a widely used agricultural fertilizer that is transported to farm fields via nurse tanks (**Fig. 1**), which hold liquid NH_3 under pressure. Typical steels used to construct nurse tanks include ASTM A285, ASTM A455, and ASTM A516 grade 70; these are all low-carbon, low-alloy steels with mixed ferrite-pearlite microstructures. The Fertilizer Institute estimates that about 200,000 nurse tanks are in use across the United States; many are between 20 and 50 years old.

An international survey conducted in 1982 found that over half of all inspected spherical ammonia tanks contained cracks [**1**]. Anhydrous ammonia was reported in 1997 to be the most frequently released hazardous substance [**2**]. Liquefied ammonia flash vaporizes upon depressurization and causes severe injuries if it contacts human tissue [**3**]. The dangers posed by unintended NH_3 releases make the safe storage and transport of anhydrous ammonia an important concern for both agricultural workers and the general public.

Nurse tank failures can occur either by leaking or by explosion. Explosions have caused severe injury, death, and extensive property damage. Such failures are often attributed to stress corrosion cracking (SCC) [**4-8**]. SCC is crack formation and propagation in metal caused by the combined effects of corrosion and straining of the metal from residual or applied tensile stresses. Since nurse tanks do not contain manways, magnetic particle and fluorescent dye penetrant inspection methods can be used to find incipient cracks on interior tank surfaces only by cutting the tank open to expose its interior. Hydrostatic pressure testing, external visual examination and ultrasound wall thickness measurements are the only inspection methods in wide use today. Prior studies have shown that SCC can occur by three mechanisms: active path dissolution, hydrogen embrittlement, and film-induced cleavage [**9-14**]. Numerous reports have been published on the effects of water, oxygen, nitrogen, and carbon dioxide on SCC in ammonia tanks [**15-21**].

Fig. 1. Anhydrous ammonia nurse tank on running gear. (Photo courtesy of the U.S. Federal Motor Carrier Safety Administration).

Crack growth rates have been modeled by Lunde and Nyborg [**22, 23**]. Their model predicts crack growth rates on the order of 1 mm/yr for a nurse tank in routine service with a typical stress intensity factor (50 MPa√m) for the steels most commonly used to fabricate nurse tanks. For higher values of steel yield strengths and stress intensity factors the Lunde-Nyborg model predicts crack growth rates as high as 2 to 6 mm/yr.

Nurse tanks are fabricated by forming steel plate into cylindrical and hemispherical shapes, then welding those components into a completed tank. Steel in the fusion and heat-affected zones (HAZ) of welds is particularly susceptible to SCC because the metal retains high residual stresses from welding, which remain in the tank throughout its service life. Some regions near a weld retain a residual tensile stress; others retain a residual compressive stress. Tensile stresses are essential for SCC initiation and propagation, so only those regions with residual tensile stresses are vulnerable to SCC.

Some nurse tanks have been given stress relief anneals after welding to reduce residual stresses, but the great majority of the U.S. nurse tank fleet has not received stress relief annealing after welding. Accident analyses performed on failed nurse tanks often report that the crack leading to failure started near a weld [**7**]. Observations of cracks in and near welds typically show transgranular crack propagation in the fusion zone and intergranular propagation in the heat-affected zone. Some investigators have measured the residual stress state in and near nurse tank welds with x-ray diffraction analysis and/or strain gauge analysis [**14**]. However, such measurements are of limited value because the x-ray wavelengths most often used in diffraction analysis penetrate only a few tens of micrometers into the steel. This leaves most of the metal inaccessible to direct measurement of the lattice parameter shifts caused by residual stress. This problem can be overcome by performing diffraction analysis with a neutron beam, which can easily penetrate the wall thickness of a steel nurse tank.

Stress corrosion crack formation and propagation is a stochastic process, and the current understanding of SCC is inadequate to allow reliable prediction of whether a given piece of steel will develop a stress corrosion crack when loaded in tension and immersed in NH_3. Once a crack is present, models are available to make predictions of how rapidly the crack will propagate, but these models are approximate at best because the interplay of factors affecting crack growth rates is complex and incompletely understood. This study was performed in an effort to (1) provide residual stress measurements throughout the HAZ and fusion zones of welds, and (2) use these residual stress data to provide SCC failure predictions for anhydrous ammonia nurse tanks for various temperatures and crack sizes.

Experimental

Material and methods for residual stress measurement

The Spectrometer for Materials Research at Temperature and Stress (SMARTS) facility of the Los Alamos Neutron Scattering Center (LANSCE) has been described in detail elsewhere [**24, 25**]. It has the ability to mount a complete circumferential welded section (30cm wide and 110 cm in diameter) from a 1000-gallon nurse tank in the neutron beam. By measuring the neutron diffraction pattern of the ferrite phase in the steel, shifts from the equilibrium interatomic spacing are used to determine the strain on the steel in small cube-shaped volumes in the metal that are 2 mm on a side. Interatomic spacings smaller than the equilibrium value indicates compressive strain and interatomic spacings larger than the equilibrium value indicate tensile strain. The residual stress necessary to produce such strains can then be calculated by Hooke's Law. By sequentially measuring many 2 x 2 x 2mm regions, maps of residual stresses in the axial, hoop, and longitudinal directions were developed for extensive regions of the specimen.

Two 3800-liter (1000-gallon) nurse tanks were purchased for use in this experiment; one tank was manufactured in 1966, the other in 1986. Both tanks had been in continuous service from their manufacture date until they were purchased for this research project in 2009. A hoop section (**Fig. 2**) was cut with an oxyacetylene torch from each of the two nurse tanks. The cuts were made parallel to the weld used to attach the hemispherical head to the cylindrical body of each tank with the cuts made 15 cm from the weld fusion zone. Temperature measurements made on the hoop section during cutting indicated that the temperature remained below 100˚C in all but a 3 cm wide region beside the cuts. Both tanks were fitted with strain gauges prior to the oxyacetylene cutting to measure strain changes resulting from the cutting. In all locations, the strain changes measured by the strain gauges were small, indicating that the largest residual stress change caused by the cutting was less than 25 MPa.

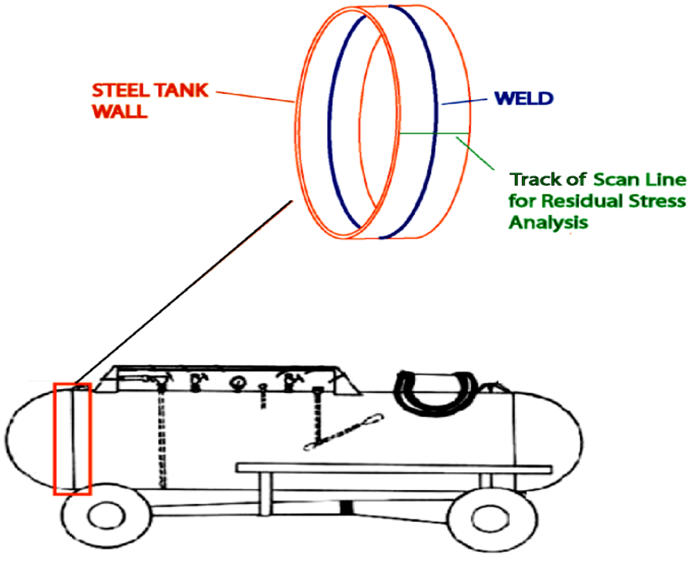

Fig. 2. Location of sections cut from nurse tanks for neutron diffraction analysis of residual stress.

Coupons were cut from other areas on the tank to use in fabricating tensile test specimens. These specimens were machined and tested in accordance with ASTM Standard E8 to determine the steel's yield strength, ultimate tensile strength, and ductility as percent elongation at failure.

The hoop sections were transported to the LANSCE site and measured by neutron diffraction to determine residual stress distributions. After the diffraction experiments were completed on the hoop sections, coupons from the specimen were cut and annealed in inert gas to relieve all residual stresses. A 4mm thick slice containing the diffracted region was cut from the specimen by wire electrodischarge machining (EDM). Further EDM cuts made this slice into a comb-shaped piece with 4 x 4 x 6mm "teeth" to relieve essentially all residual stresses. These slices were then measured by diffraction in SMARTS to determine the ferrite lattice parameter "baseline value" for a stress-free state.

Material and methods for crack propagation rate measurements

To measure crack formation and initiation rates in the steel most commonly used to manufacture nurse tanks (SA455), notched specimens were held in tension stress and immersed in ammonia vapor or ammonia liquid for a period of seven months. These specimens were periodically removed, examined for cracks and measured, then returned to the ammonia environment. The goals of this work were to (1) determine how rapidly cracks began, and (2) to measure crack growth rate. Growth rate information, in particular, would be valuable in predicting likely failure times for tanks found to contain sub-critical-size cracks. Many nurse tanks contain cracks, but the cracks are usually too small and grow too slowly to pose any immediate safety threat. Such cracks can eventually grow to dangerous dimensions, and information on crack propagation rates can guide inspection procedures to assure that cracked tanks are repaired or removed from service before cracks approach critical size.

Stress corrosion crack propagation testing was performed in a custom-built 1800-liter steel test tank with a manway to permit specimens to be placed into the tank and subsequently removed. (This would be nearly impossible in normal nurse tanks, which lack manways). The rack held 56 pre-notched, direct tension samples preloaded to various stresses ranging from 40 to 80 MPa√m. These specimens were periodically removed from the tank, and each specimen was examined using a stereoscopic optical microscope to determine if SCC had occurred in that sample. At the end of the test, crack lengths were checked by opening (stressing to failure) specimens that contained cracks to check whether external evidence of crack length matched the actual crack dimensions, which can be determined accurately only by opening the specimens

Results and discussion

Results of residual stress measurements

The hoop residual stress distributions measured by neutron diffraction at SMARTS/LANSCE are shown in **Fig. 3**. The stress distributions in the 1966 and 1986 tanks were found to be quite similar. In each case the residual tensile stresses (denoted by the positive stress values on the plots) are nearly as large as the tensile yield strength of the steel. Once the tank is pressurized with its ammonia cargo, additional stresses are imposed on the metal such that the yield strength of the metal is exceeded, and small regions of the metal plastically deform. Thus, the metal is under nearly the highest possible tensile stress state, making it especially susceptible to SCC initiation in these regions. It is significant that the highest tensile stresses are found in the HAZ, not in the weld's fusion zone. **Table 1** shows the maximum residual stresses measured in all three directions on the 1966 tank: hoop, axial (parallel to the tank's long dimension), and radial. The table also shows the stresses imposed by pressurizing the tank to the maximum anticipated pressure in service, 1.7 MPa (250 p.s.i.) and the percentage of the steel's ultimate tensile strength when both stresses are summed. **Table 1** does not attempt to show a third source of stress imposed on nurse tanks in service, the transient stresses resulting from towing nurse tanks over roadways and farm fields.

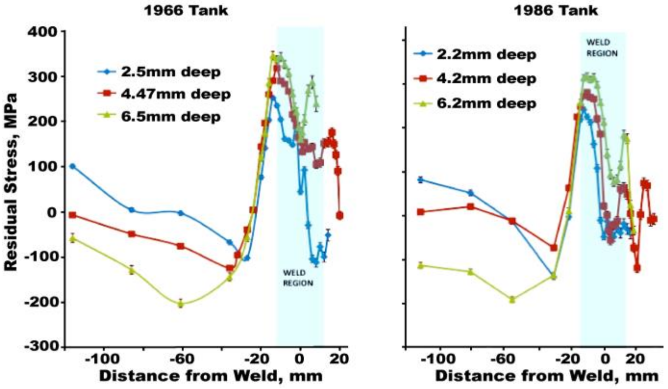

Fig. 3. Residual hoop stress distributions measured by neutron diffraction across a circumferential weld for a tank manufactured in 1966 (left) and for a tank manufactured in 1986 (right). The hemispherical tank head lies to the right of zero on the x-axis; the cylindrical tank body lies to the left of zero.

Table 1. Maximum hoop, axial, and radial stresses in a nurse tank resulting from internal pressure and from residual stress in the HAZ near welds.

Stress Direction	(a) Internal pressure	(b) Residual stress in HAZ	(a+b) as % of the steel's ultimate tensile strength
Hoop	110 MPa	350 MPa	82%
Axial	50 MPa	250 MPa	54%
Radial	2 MPa	23 MPa	4.5%

Results of the SCC crack formation and propagation measurements

Of the 56 pre-notched, direct-tension samples preloaded to various stresses, only seven specimens developed cracks during the seven-month period of NH_3 exposure. In every case, the crack lengths determined by external measurements were confirmed by crack opening results performed at the end of the test. The crack sizes measured in this experiment are displayed in **Fig. 4** as a function of the stress intensity factor for the specimen. (A higher stress intensity factor correlates to a higher stress on the specimen). This figure also indicates the location of the cracked specimen in the tank (i.e., whether it was above, at, or below the tank's vapor-liquid level). The location may be significant because there have been anecdotal reports from tank repair shops that most cracks occur in the vapor space above the liquid rather than on tank walls covered by liquid ammonia. Ammonia vapor may promote SCC more aggressively if it contains little or none of the 0.2% water content that is intentionally added to all anhydrous ammonia to suppress SCC.

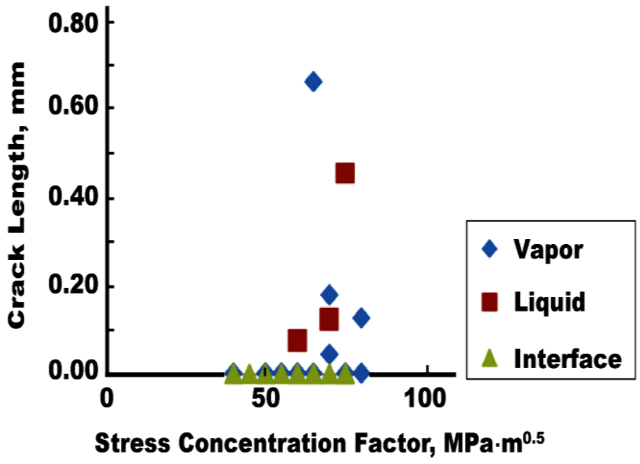

Fig. 4. Crack size as a function of stress intensity factor after four months exposure to NH_3. The symbols indicate whether the cracked specimen was tested in ammonia vapor only (vapor), in ammonia liquid only (liquid), or partially in vapor and partially in liquid (interface).

The cracks that formed were small (**Fig. 4, 5**), and their growth rates were modest, as would be expected during a trial that lasted just seven months. It was, however, possible to obtain growth rate values from this experiment, and these growth rates were used to formulate the failure predictions described in the discussion section. The data show only a loose correlation between stress intensity factor and crack size, which is typical of SCC data and reflects the stochastic nature of SCC crack growth rates.

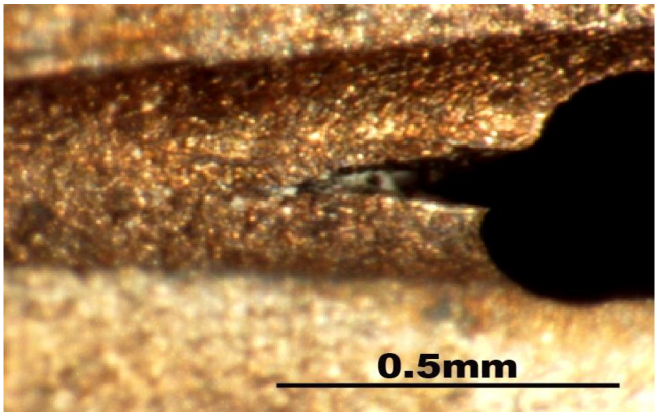

Fig. 5. Optical micrographs of a typical crack that initiated and grew during the SCC test. The specimen was exposed for two months in ammonia vapor on a specimen with a stress intensity factor of 65 MPa(m)$^{0.5}$

Service life prediction for tanks with cracks

The data obtained in these tests can be usefully applied to actual nurse tank crack propagation situations only if the stress intensity factors are known for steel at various locations in the tank. The following narrative describes how the SCC test data were combined with the residual stress analysis data from LANSCE to make predictions on failure time for tanks in various situations. The stress intensity factors for axial flaws in cylinders subject to internal pressure can be calculated by **Error! Reference source not found.** [**26**].

$$K_I = \sigma_h\sqrt{\pi a}\sqrt{1+0.52\chi+1.29\chi^2-0.074\chi^3} \qquad (1)$$

Where, σ_h is the hoop stress as given by **Error! Reference source not found.** and χ is given by **Error! Reference source not found.**.

$$\sigma_h = \frac{pR}{t} \qquad (2)$$

$$\chi = \frac{a}{\sqrt{Rt}} \qquad (3)$$

In these expressions *p* is the internal pressure, *R* is the radius of the tank, *t* is the thickness of the tank, and *a* is half the flaw length as shown in Error! Reference source not found.**6**. This holds true when *R* >> *t*. The hoop stress is twice the magnitude of the radial stress. For this reason, the stress is only calculated for axial flaws and not hoop flaws in this document.

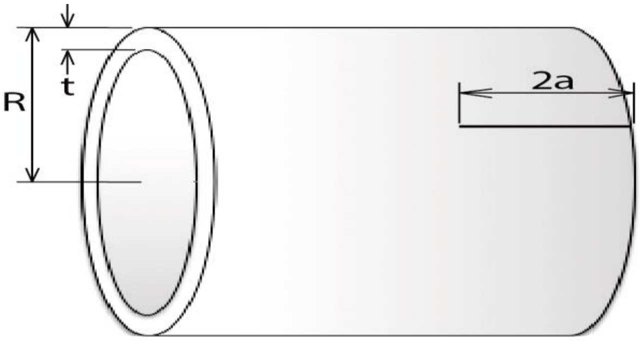

Fig. 6. Axial flaw in a cylinder subjected to internal pressure.

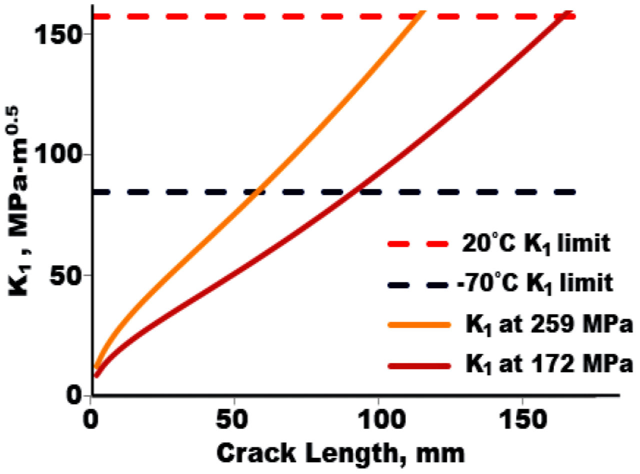

Fig. 7. K_1 versus crack length in a 3800-liter (1000-gallon) nurse tank.

The internal pressure in a nurse tank is regulated by a pressure-relief valve so the maximum internal pressure will not exceed 1.72 MPa. With a tank radius of 520 mm and a shell thickness of 7.94 mm:

$$\sigma_h = \frac{pR}{t} = \frac{(1.72MPa)(520mm)}{7.94\ mm} = 113MPa$$

The fracture toughness, J_{IC} value, of A516 Gr70 steel at 20°C is 114 kN/m [**27**]. This corresponds to K_{IC} of 158

MPa$\sqrt{m}$. However, when a breech occurs in the tank wall, rapid depressurization of the tank can cause the temperature in the local zone of the wall to fall to -67°C [**28, 29**]. Basko found that at temperatures of -70°C the critical stress intensity factor K_C is no less than 85 MPa$\sqrt{m}$ [**28, 29**]. In a nurse tank with an internal pressure of 1.72 MPa, diameter of 1040 mm, and a wall thickness of 7.94 mm, the stress intensity factor of an axial crack 94 mm long is 85 MPa$\sqrt{m}$ (**Fig. 7**).

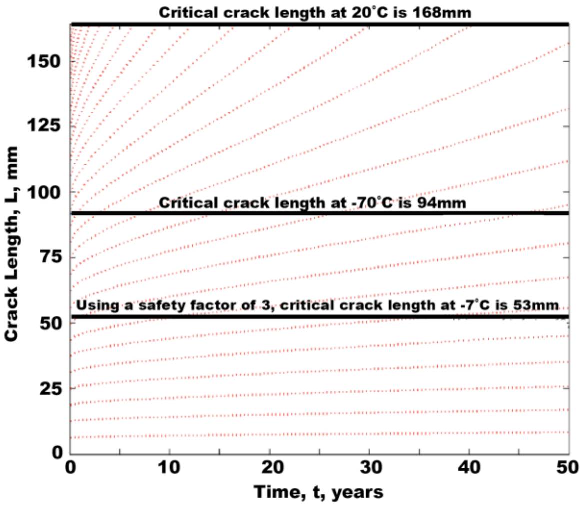

Fig. 8. Predicted growth of axial through-thickness cracks over a 50-year period, calculated for a 3800-liter (1000-gallon) nurse tank with an internal pressure of 1.72 MPa, 104.1cm outside diameter, a 6.05mm thick shell, and a crack growth rate of 3 x 10^{-7} m/yr. The middle black horizontal line represents the critical crack length at -70ºC. The upper border of the plot is the 20ºC critical crack length.

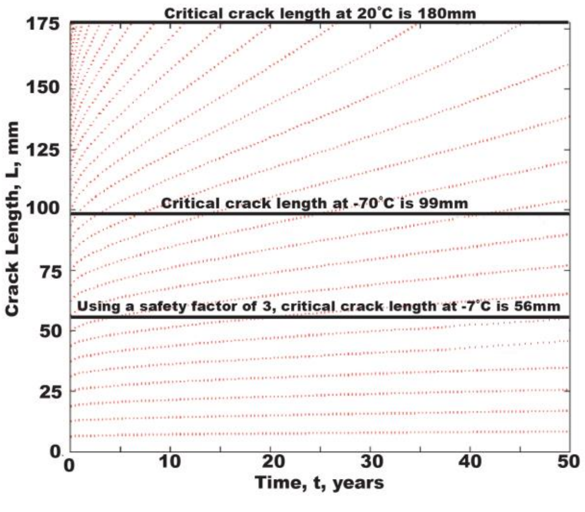

Fig. 9. Predicted growth of axial through-thickness cracks over a 50-year period, calculated for a 5500-liter (1450-gallon) nurse tank at 1.72 MPa (250 psig) internal pressure, 118.1cm outside diameter, a 6.88mm thick shell, and a crack growth rate of 3 x 10^{-7} m/yr. The center horizontal line represents the critical crack length at -70ºC. The upper border of the plot is the 20ºC critical crack length. Tanks with cracks less than 74mm long will not fail for at least 10 years.

SCC initiates on the interior wall of a nurse tank so visual inspection of the tank exterior cannot detect cracks unless they penetrate to the exterior surface (i.e., tank failure). Thus, dangerous tanks can appear safe by visual examination. Ultrasound thickness testing will also miss nearly every crack because angled ultrasonic inspection is necessary to find cracks. Hydrostatic tests subject tanks to a minimum internal pressure of 2.58 MPa. Tanks that pass this test may have cracks as long as 160 mm that can catastrophically fail when escaping liquid ammonia causes a drop in local temperature and a corresponding drop in fracture toughness.

The maximum crack depth for a given stress concentration for low carbon steel in ammonia, calculated by Lunde and Nyborg is shown in **Error! Reference source not found.**.

$$a_y = a_0 + 3.0 \times 10^{-7} K_{Ic}^2 \sqrt{y} \qquad (4)$$

where *a* is crack growth in meters with respect to time and *y* is time in years.

The crack growth rate for this is Equation 5.

$$\frac{da}{dy} = 1.5 \times 10^{-7} K_{Ic}^2 y^{-1/2} \qquad (5)$$

Combining **Error! Reference source not found.** with **Error! Reference source not found.** produces Equation 6:

$$a'(y) = \frac{1.5\times10^{-7}\pi p^2 R^2}{t^2}\left(a(y) + \frac{0.52}{\sqrt{Rt}}a(y)^2 + \frac{1.29}{Rt}a(y)^3 - \frac{0.074}{(Rt)^{3/2}}a(y)^4\right)\frac{1}{\sqrt{y}} \qquad (6)$$

Given the initial value of $a(0) = a_0$. This equation does not cover crack initiation, only crack growth. Solving the nonlinear first-order differential equation for several initial crack lengths generated **Fig. 8, 9**.

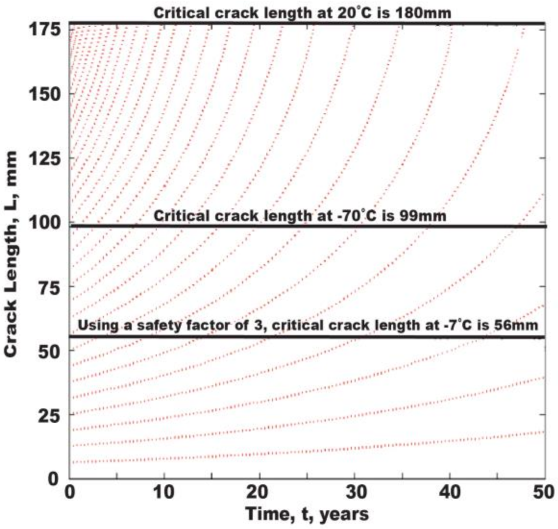

Fig. 10. Predicted growth of axial through-thickness cracks over a 50-year period assuming that crack growth rate remains constant and does not slow with time, calculated for a 3800-liter (1000-gallon) nurse tank at 1.72 MPa, 104.1cm outside diameter, a 6.05mm thick shell, and a crack growth rate of 3 x 10^{-7} m/yr. The center horizontal line represents the critical crack length at -70ºC. The upper border of the plot is the 20ºC critical crack length.

These calculations predict that a tank with a 76.2 mm (3”) crack will last more than 10 years. This assumes that the crack grows equally in both directions and that the internal pressure remains at 1.72 MPa throughout the life of the tank. It also assumes that crack growth slows over time as Lunde and Nyborg's research indicated. However, they mentioned that their tests maintained a constant K_{IC}. In a nurse tank, as the crack grows, the stress intensity would also increase which would lead to a higher crack growth rate.

If the crack growth rate remains constant and does not slow with time, then the equations change to:

$$da/dt = 3.0 \times 10^{-7} K^2_{IC} \tag{7}$$

$$a'(y) = \frac{3.0\times10^{-7}\pi p^2 R^2}{t^2}\left(a(y) + \frac{0.52}{\sqrt{Rt}}a(y)^2 + \frac{1.29}{Rt}a(y)^3 - \frac{0.074}{(Rt)^{3/2}}a(y)^4\right) \tag{8}$$

This causes the plots of crack growth to change significantly, as shown in **Fig. 10, 11**, which are given in the supporting information section.

The plots shown in **Fig. 10, 11** are not as forgiving as those in **Fig. 8** and **9** and indicate that a tank with a 76 mm crack can fail in about 7 years. Using the highest crack growth recorded in this project's tests (9.41 x 10^{-7} m/year) results in the following equations:

$$\frac{da}{dy} = 9.41 \times 10^{-7} K_{IC}^2 \tag{9}$$

$$a'(y) = \frac{9.41\times10^{-7}\pi p^2 R^2}{t^2}\left(a(y) + \frac{0.52}{\sqrt{Rt}}a(y)^2 + \frac{1.29}{Rt}a(y)^3 - \frac{0.074}{(Rt)^{3/2}}a(y)^4\right) \tag{10}$$

These equations yield values plotted in **Fig. 12, 13** which are given in the supporting information section. The plot's upper border lies at the 20˚C critical crack length. Tanks with cracks less than 32mm long will not fail for at least 10 years under these circumstances.

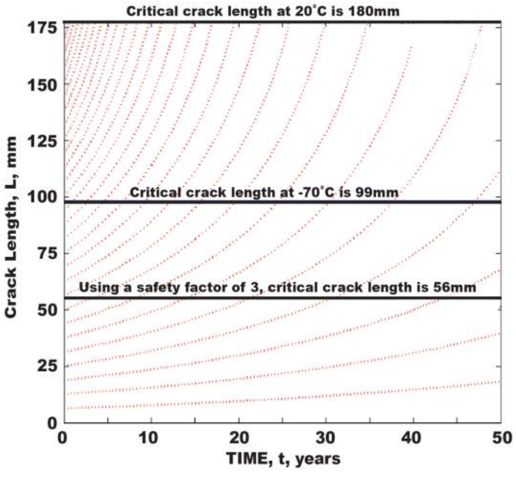

Fig. 11. Predicted growth of axial through-thickness cracks over a 50-year period, assuming that crack growth rate remains constant and does not slow with time calculated for a 5500-liter (1450-gallon) nurse tank at 1.72 MPa, 118.1cm outside diameter, a 6.88mm thick shell, and a crack growth rate of 3 x 10^{-7} m/yr. The center horizontal line represents the critical crack length at -70ºC. The upper border of the plot is the 20ºC critical crack length.

Fig. 12, 13 present calculation results for 3800- and 5500-liter tanks respectively when crack growth rates are 3.1 times greater than those used in **Fig. 8, 9**. Cracks with length over 38 mm can fail in around 10 years, and cracks with length of 64mm can fail in less than 5 years. The 3800-liter tanks fail slightly sooner than 5500-gallon tanks (with the listed thicknesses). In our side-angle ultrasound inspections, cracks only 6mm long gave clear ultrasound indications. Even the most extreme predictions shown in **Fig. 12, 13** indicate that if a tank has no crack over 25 mm long, its minimum lifespan is predicted to be 10 years. Thus, side-angle ultrasound inspection would be sufficiently sensitive to provide useful advance warning of tank failure with inspections spaced several years apart.

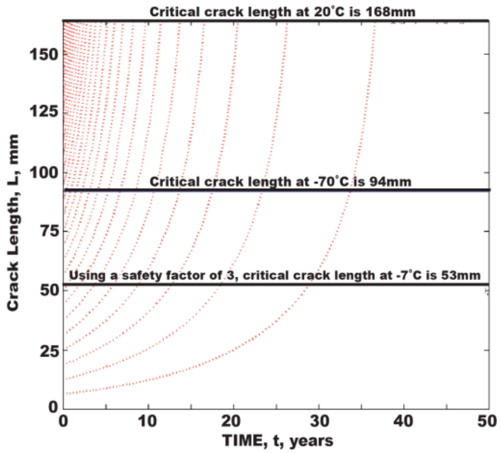

Fig. 12. Predicted growth of axial through-thickness cracks over a 50-year period, assuming that crack growth rate remains constant and does not slow with time calculated for a 3800-liter (1000-gallon) nurse tank at 1.72 MPa, 118.1cm outside diameter, a 6.88mm thick shell, and a crack growth rate of 9.41 x 10^{-7} m/yr. Tanks with cracks less than 32mm long will not fail for at least 10 years.

Because many of these cracks occur next to the circumferential weld, they can grow in only one direction. This would cut the growth rate in half, since growth is predicated on a crack propagating from both its ends. Nurse tanks are subjected to cyclical loading in service that can lead to faster crack growth as brittle corrosion products break open at the crack tip.

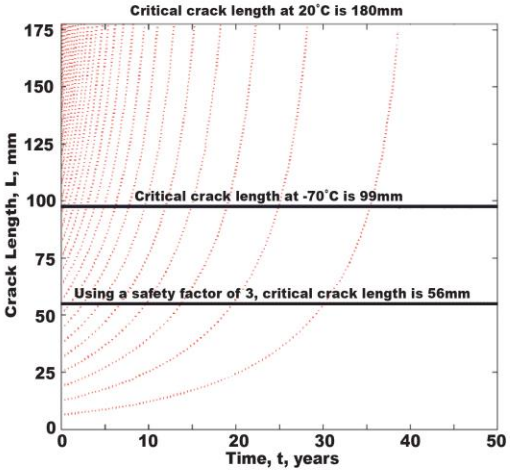

Fig. 13. Predicted growth of axial through-thickness cracks over a 50-year period, assuming that crack growth rate remains constant and does not slow with time calculated for a 5500-liter (1450-gallon) nurse tank at 1.72 MPa, 118.1cm outside diameter, a 6.88mm thick shell, and a crack growth rate of 9.41 x 10^{-7} m/yr. Tanks with cracks less than 32mm long will not fail for at least 10 years.

These calculations were done using a constant pressure of 1.72 MPa. This value represents the vapor pressure of ammonia at 50˚C and the setting of the pressure relief valve. In reality, tanks are seldom exposed to internal pressures this high. These calculations did not incorporate the residual stress due to welding because the region of high residual stress in the axial direction does not extend more than 25 mm from the weld (**Fig. 3**). The residual stress is a danger for the initiation but not the growth of axial cracks. These calculations are conservative because they were done for through cracks. Most cracks will not penetrate through the thickness of the steel and thus will have lower stress intensities. The residual stress is more dangerous in its effect on cracks that grow in the hoop direction. Axial stress acts directly on these cracks, and they can grow from both crack tips without leaving the region where tensile stress is highest (**Fig. 3**).

Conclusion

The high residual tensile stresses measured in this study are undesirable in nurse tank steel. These stresses make SCC in the welded nurse tanks more likely, because the tanks are stressed to nearly the highest possible elastic stress during most of their service life. This would be expected to accelerate SCC crack initiation and, to a lesser extent, propagation. There is some consolation to be found in the fact that the high residual tensile stresses present beside welds fall rapidly to much lower stress values as one moves to regions farther from the weld; thus, crack initiation conditions are severe near welds, but the stresses tending to propagate the cracks diminish with distance from the weld, which would be expected to diminish the speed of SCC crack expansion as the crack grows outward from the weld. Steel is a high toughness material that has large critical crack sizes (the critical size is the crack dimension that results in instantaneous failure), so these findings suggest that crack initiation conditions are severe, but that cracked tanks will still be slow to expand those cracks to the size that will cause failure.

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