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Review of stress corrosion cracking of pipeline steels in "low" and "high" pH solutions

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This paper reviews the current understanding of the mechanisms of stress corrosion cracking of pipeline steels. The similarities, the differences and the influencing factors are considered for the "high pH" stress corrosion cracking caused by a concentrated bicarbonate-carbonate solution, and for the "low pH" stress corrosion cracking due to a diluter solution. For high pH stress corrosion cracking, it is well accepted that the mechanism involves anodic dissolution for crack initiation and propagation. In contrast, it has been suggested that the low pH stress corrosion cracking is associated with the dissolution of the crack tip and sides, accompanied by the ingress of hydrogen into the pipeline steel. But the precise influence of hydrogen on the mechanism needs to be further studied. © *2003 Kluwer Academic Publishers*

1. Introduction

Stress corrosion cracking (SCC) from the external surface of a buried pipeline is a serious matter. There is always the chance that a pipeline could leak or rupture. A pipeline failure can have serious consequences for people living or working close by and also for the environment. A pipeline failure may be catastrophic and may cause significant economic and environmental loss. For Canada, there are typically 30 to 40 failures each year on pipelines regulated by the National Energy Board (NEB) [1], some of which have caused fatalities. For example a death occurred in 1985 when a drainage tile plow ruptured a gas pipeline [1]. In 1965, the first case of SCC was reported [2]. Since that time, SCC has been recognized as a cause of pipeline failures in countries throughout the world. Pipelines in Australia [3], Canada [4], Iran, Iraq, Italy, Pakistan, Saudi Arabia, the former Soviet Union and the United States [5] have all suffered SCC. The Trans Canada pipeline had three SCC failures in the Northern Ontario portion between March 1985 and March 1986, experienced its fourth SCC-related pipeline rupture in December 1991 near Cardinal, Ontario, and its fifth in July 1992 near Tunis, Ontario. In July 1995, SCC caused a rupture on the Trans Canada pipeline near Rapid City, resulting in a major explosion. These SCC related failures resulted in two public inquiries by the NEB of Canada. The second inquiry resulted in a substantial publication [1].

It is now recognized that there are two forms of SCC penetrating from the external surface of buried pipelines. One is intergranular SCC (IGSCC) and is usually called the "high pH SCC" or "classical SCC". The other is transgranular SCC (TGSCC), and is designated "near-neutral pH SCC" or "low pH SCC" or "non-classical SCC".

At the present time in China, there are plans to build a 4,200-kilometre natural gas pipeline, the total cost of which is expected to be more than 140 billion yuan (US$ 16.86 billion). It will be China's largest infrastructure project. Accordingly, it is essential and imperative to design the pipeline soundly and ensure that there are no SCC incidents. The present work is part of a research effort to understand pipeline SCC in order to minimize the possibility of its occurrence.

2. High pH SCC and near-neutral pH SCC of pipelines

There are many similarities between the two forms of pipeline SCC. Cracks of both forms usually occur on the outside surface in colonies, mostly oriented longitudinally along the pipe, primarily at the bottom of the pipeline [6]. These cracks coalescence to form long shallow flaws, that can lead to ruptures. The fracture surfaces are usually covered with black magnetite film [7] or an iron carbonate film [7]. However, there are many differences between the two forms of pipeline SCC. High pH SCC, engendered by concentrated bicarbonate or carbonate-bicarbonate solutions associated with pH of 9, has usually an intergranular morphology, and the cracks are sharp, with little lateral corrosion [1]. Near-neutral pH SCC, engendered by dilute ground water with a relatively low pH of around 6.5, has a trangranular, quasi-cleavage crack morphology with very little branching [1]. The transgranular cracks are generally wide with appreciable lateral corrosion of the crack sides. Moreover, the near-neutral pH SCC occurs over a wider potential range [8] than high pH SCC which has only narrow width of no more than 100 mV(SCE) [8].

SCC requires the simultaneous action of the following three factors: a potent environment at the pipe surface, a susceptible pipe material and a stress. All three factors must be present together for SCC to occur. If any of them can be eliminated or reduced, then SCC can be prevented. The following sections discuss these three factors in more detail.

3. Environment conditions

When new pipelines are built, they are coated for corrosion protection and cathodic protection is applied once the pipeline is in the ground. After a period of pipeline operation, the coating may deteriorate, leading to the formation of holidays and the diffusion of water, carbon dioxide and other species through the coating. Alternatively the coating may disbond. Different types of coating have different properties, and they have different propensities for deterioration or disbondment. The correlation of coating type to the incidence of SCC is most marked in relation to the near-neutral pH SCC where, with the exception of the involvement of asphalt in very high soil resistivity locations, SCC failures have been mostly associated with high electrical resistivity tape coatings. The explanation may be that a high electrical resistivity tape coating can shield the cathodic protection current from flowing to the pipeline surface, and if the coating is disbonded, then the near-neutral pH environment forms and TGSCC can become possible. If the coating has a high resistivity, then the near-neutral pH natural ground water is not changed by the cathodic protection current. If the good coating is disbonded or deteriorated, it can shield the cathodic protection current flowing at the pipeline surface, the natural ground water can come into contact with the pipe material and then TGSCC can become possible. This interpretation is supported by the NEB report [1].

For high pH SCC, SCC has been associated with pipelines with coal tar, asphalt and tape coatings, and no failures have been reported with modern thin film coatings. Most of the failures with coal tar coatings may be because this type of coating has been used on the older pipelines, these pipelines have had longer periods of service, and hence had more opportunity to crack. This is supported by NEB [1]. The consequence of coating defects is that they allow ground water access to the pipeline surface. The area of the surface exposed to the ground water solution depends on the disbondment of the coating. Furthermore, the composition of the groundwater solution depends on the amount of cathodic protection current reaching the pipe surface [1]. The ground water is not be changed if the coating does not allow the cathodic protection current to pass through, or if there is high electrical resistances within the soil or the solution in the crevice between the pipeline surface and the coating, or if there is no significant cathodic protection current reaching the exposed surfaces. The natural ground water solution has a pH from 6 to 7 resulting from the equilibrium between HCO_3^- and CO_3^{2-}. This solution can cause TGSCC. Laboratory tests often use NS1, NS2, NS3, NS4 solutions [9], the Nova solution [10] or 0.01 N $NaHCO_3$ [11] or N1 Trapped Water [11] to produce TGSCC.

However, a substantial cathodic current at the pipeline surface causes hydroxyl ions to be generated and accumulated, and the pH increases according to reaction (1) or (2):

$$2H_2O + 2e = H_2 + 2OH^- \qquad (1)$$

$$O_2 + 2H_2O + 4e = 4OH^- \qquad (2)$$

The solution chemistry also relates to the conversion of bicarbonate to carbonate ions. With time, the solution becomes concentrated and the concentration of carbonate is high, which leads to the tendency for the solution to passivate the steel surface, and IGSCC can occur. The extent that the pH increases depends on the partial pressure of CO_2. In the absence of CO_2, the pH increases to 11–12. The solution consists of nearly all carbonate ion, and the passive film characteristics and electrochemistry are not conductive to SCC. However, if there is an appropriate partial pressure of CO_2 the solution becomes buffered because of the equilibrium between bicarbonate and carbonate ions, and the pH remains in the region of 9. IGSCC can occur when the potential is in the appropriate range as defined by Fig. 1 [12, 13].

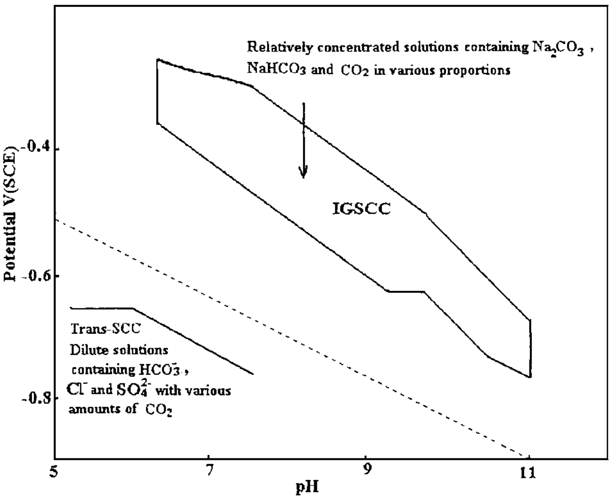

Figure 1 Potential-pH diagram showing the regimes for IGSCC and TGSCC at 24°C in solutions containing different amounts of CO_3^{2-}, HCO_3^- and CO_2 to achieve different pH values [12, 13].

The great majority of pipelines use cathodic protection so that the high pH solution is more likely to be generated than the low pH environment. This is in agreement with field experience that the intergranular form of SCC is much more common than the transgranular form.

There is a difference of the polarization curve measured in the high pH solution and the near-neutral solution. In the high pH solution, the curve exhibits an active-passive transition over a certain potential range as illustrated in Fig. 2 from the work of Parkins [14]. This transition has been shown to be associated with IGSCC of ferritic steels in various environments [15]. In contrast, the near-neutral pH environments do not promote passivation and do not exhibit an active-passive transition (Fig. 3) [12]. The influence of potential and pH on IGSCC and TGSCC is demonstrated in Fig. 1 [12, 13]. This shows the results from slow strain rate tests on a low-carbon steel in the solutions which contain bicarbonate, carbonate and carbon dioxide. For TGSCC, the potential should be applied at or slightly cathodic (max. −50 mV) to the Ecorr [16] to reproduce TGSCC.

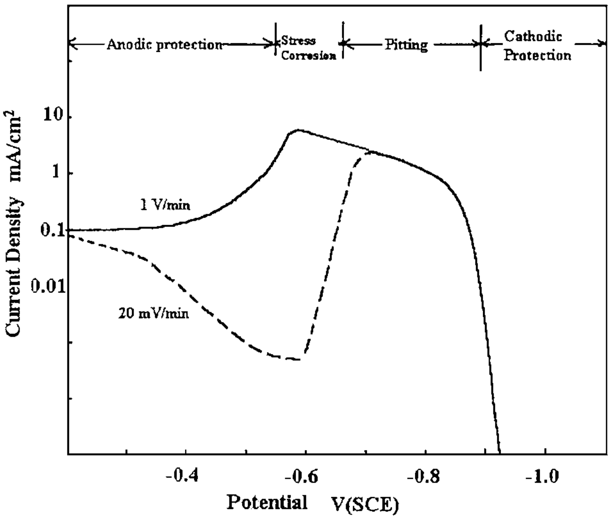

Figure 2 Potentiodynamic polarization curves showing the potential range for IGSCC in concentrated carbonate-bicarbonate solution at 90°C [14].

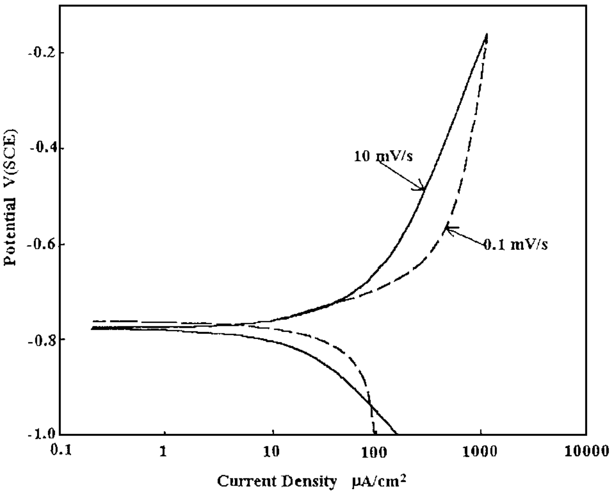

Figure 3 Fast and slow sweep rate polarization curves at 24°C for a line pipe steel in simulated ground water saturated with CO_2, pH = 5.8 [12].

Researchers have learned that a range of chemical species can promote near-neutral pH SCC. Near-neutral pH SCC has occurred in solutions with low concentration of carbonic acid, bicarbonate ions and other species including chloride, sulphate and nitrate ions where high pH SCC has not been observed [1]. Rebak *et al.* [17] showed that TGSCC can be reproduced in aqueous sulfate, bicarbonate and simulated soil solutions. So the environment producing near-neutral pH SCC may vary. In addition, there is another factor for operating pipelines that can make solution concentrated. If the surrounding ground is able to accept the evaporated water, which is influenced by the seasonal change, the water can slowly evaporate from the solution due to heat transfer through the pipe wall. This can concentrate the solution, as has been shown by Parkins [18].

IGSCC displays temperature sensitivity in the field and in laboratory tests. When the temperature increases, the intergranuar stress corrosion crack propagation rate increases according to the Arrhenius Law [19]. In the field, the temperature in the vicinity downstream of a compressor station is higher than in other places, and so the greatest risk of SCC is in this location. When the temperature is high, the coating deterioration also becomes worse. In contrast, there is no systematic temperature dependence in the range 5°C to 45°C for near-neutral pH SCC [20].

The effect of oxygen on SCC of buried pipeline steels needs to be clarified. Delanry [4] cited a Canadian study that showed SCC becomes more severe in the absence of oxygen. In that study, cracks in the field were observed mainly in areas where oxygen access was restricted and the propensity to SCC decreased with the presence of oxygen.

4. Metallurgical conditions

So far there is no obvious evidence that provides a relationship between pipeline failure due to near-neutral pH SCC or high pH SCC and steel composition, grade, or microstructure. However, there is evidence from laboratory studies that shows certain batches of steel are more resistant to SCC. Asahi showed that, for a range of pipeline steels from X52 to X80 grades, thermo-mechanical controlled processing or quenched and tempered steels with fine-grained bainitic structures, or acicular ferrite, uniform microstructures, were more resistant to IGSCC than controlled rolled steels with ferrite-pearlite structures [21]. The work by Kimura *et al.* [22, 23] has shown that the low pH SCC susceptibility for the ferrite-pearlite structure is higher than that for uniform structure like a quench and temper steel. The homogeneous structure has a high resistance to SCC.

Grit blasting pipe surfaces before applying a coating can provide better coating bonding, and can be beneficial in avoiding the potential region for intergranular cracking. Moreover, if appropriately applied, grit blasting leaves the pipe surface in a state of compression that is beneficial in at least delaying, if not preventing, the incidence of SCC in a variety of systems [12].

It is well established [24, 25] that the mill scaled surfaces on pipeline steels are more susceptible to SCC than polished surfaces. The cause is not understood, but it has been accepted that the mill scale plays a detrimental role. Most cracks initiate at the base of pits where there are cracks in the mill scale [25]. The mill scale can hold potential within the cracking range [24, 25]. Furthermore, the potential for pitting on a pre-pitted surface is much below that for non-pitted surfaces, and this may facilitate changes in solution composition within new pit enclaves and also the discharge of hydrogen. Wang and Atrens [26] carried out observations of SCC initiation of X65 pipeline steel exposed to the high pH solution. They found for the pipeline steel tested with the original surface as was present in service, SCC initiation correlated with the fracture of the surface product film. This SCC initiation from the original surface occurred at a stress lower than for a polished surface of the same steel.

For near-neutral pH SCC, the following five factors have been suggested [11] to influence crack initiation: inclusions, aligned surface defects, persistent slip bands produced by mechanical pre-treatment of the steel, preexisting defects on the pipeline surface and coating disbondment. High pH SCC cracks are often found in the area containing pits in field failures [27] and in laboratory tests [25]. Pits cause a stress concentration, particularly when relatively deep. High pH crack nucleation appears to increase with increasing pit density.

Generally, it is common to use a resistant material in engineering structures for a particular environment to avoiding SCC. This may be feasible for new pipelines. However, the new steels must have high strength and fracture toughness, which is essential for the structures.

When a specimen is cold worked or notched, the initiation time is shortened and so cracking becomes easier than on a polished specimen. The reason is the stress concentration in the area of the notch or the cold work. Similarly the coarse grain heat affected zone is highly susceptible to SCC. Harle [28] showed that the coarse grain heat affected zone microstructure adjacent to the weld in an X65 steel, is significantly more susceptible to cracking than the base material in the near-neutral pH environment.

When pipeline steel is exposed to high pH solution, grain boundaries undergo preferential dissolution [29]. If a stress of an appropriate magnitude is not present, the dissolution does not penetrate far. The propensity for such intergranular attack is thought to be related to the presence of carbon, sulfur or phosphorus segregated to the grain boundaries in ferritic steels [30], as well as the properties of the solution that foster selective attack on the grain boundaries. Recently Wang *et al.* [31] measured the grain boundary compositions for X65 and X52 pipeline steels using analytical electron microscopy (AEM). The results showed that there was no segregation at proeutectoid ferrite grain boundaries, which indicated that species P and S are not responsible for preferential dissolution of grain boundaries during intergranular SCC [31–33]. Due to the ubiquitous hydro-carbon contamination of specimen surfaces, the distribution of carbon could not be measured using AEM [34]. For X42 steel, there was also no measurable S and P segregation at ferrite : ferrite grain boundaries [35].

5. Mechanical conditions

In order to propagate for SCC cracks, there must be an appropriate stress at the crack tip. The stress can be applied or residual in nature. SCC can initiate and propagate only if there is sufficient stress. Below some value of stress, the crack may not initiate. This stress is the threshold stress. Many cases of longitudinal SCC in the body of pipe were attributed to the hoop stress generated by the internal pressure. It was found that the threshold stress was markedly reduced for an actual pipe surface in the presence of mill scale or pits [36]. The number of cracks depends on the maximum stress [12]. For a higher stress, there are more cracks and they are closer spaced.

Beavers [37] has indicated that no cracks have been found under constant displacement conditions. This points out the importance of pressure fluctuations. Pressure fluctuations may be necessary for cracks to occur not only with near-neutral pH SCC but also with high pH SCC.

Parkins [38] correlated the reduction in threshold stress with the strain-hardening behavior of the steel when subjected to cyclic stresses superimposed on a monotonically increasing stress. He found that the initiation becomes easy and the propensities for intergranular cracking of steels have a close relationship with cyclic stress-strain behavior as shown in Fig. 4. The superimposed cyclic stress (on the order of 15 to 80 M N/m^2) made plastic deformation start at much lower levels of mean stress, and the slope of the plastic portion of the curve changed abruptly at a certain stress, which corresponded to the threshold stress for SCC. Parkins showed that cyclic loading significantly decreased the threshold stress for IGSCC below that associated with a static load. While it has not been demonstrated by appropriate measurements, it seems likely that cyclic loading may also facilitate transgranular cracking in near-neutral pH solution and Parkins has remarked that cyclic loading of pipelines appears to play a major role on the propagation of both types of SCC [12].

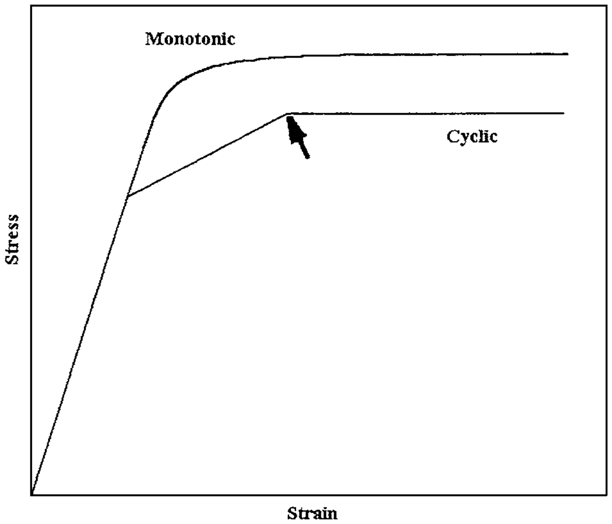

Figure 4 Comparison of a typical stress-strain curves produced with monotonic loading and with cyclic loads superimposed on the steady loads. Arrow indicates inflection point where the stress corresponds to the threshold stress for SCC [38].

There are also dormant cracks in pipelines. These are attributed to the decreasing micro-plastic strain rate with time. The phenomenon of dormant cracks may explain why relatively few failures occur in the field compared with the number of small cracks which are discovered in inspections. However, crack may grow in other cases, especially if adjacent cracks coalesce [39]. When a film is ruptured or an active crack is closer to the dormant one, the reactivation of a dormant crack may be possible and then coalescence may occur. This is consistant with Parkins [40]. Coalescence may possibly influence crack growth, but there is no predictive model which has been developed that successfully defines the effect of coalescence on crack propagation smoothly and the crack growth rate exactly. So it is incomplete from this point, and needs further to be studied.

To compare different steels, slow strain rate test (SSRT) has been used, which places emphasis on electrochemical effects (dissolution and passivation kinetics). In SSRT, the deformation of the specimen is imposed by the test at a given rate, which may override the creep rate and strain-hardening properties of the steels [41]. A better approach to comparing these steels may be using cyclic loading tests. Cyclic loading tests are more similar to the field conditions than SSRT.

6. Mechanistic understanding

Since the cases of high pH SCC have about 40 years of history, researchers have studied IGSCC extensively, and its mechanism, caused by preferential dissolution at the grain boundaries, is well accepted. For IGSCC, i.e., high pH SCC, Parkins showed that the crack rate for a variety of alloy in various environment can be evaluated from the dissolution current densities measured on bulk specimens under the same environmental conditions [40]. The crack propagation rate can be calculated according to Faraday's law:

$$\mathrm{CPR} = (i_{\mathrm{a}} \cdot M)/(z \cdot F \cdot d) \tag{3}$$

where CPR is the crack propagation rate, i_{a} is the anodic current density, M is the atomic weight, z is the valency of solvated species, F is the faraday and d is the density of metal. It has been found that the calculated propagation rate is in conformity with the estimated one [12, 29]. So the mechanism of high pH SCC is attributed to anodic dissolution resulting from selective dissolution at the grain boundaries and repeated rupture of passivating films that form over the crack tip.

Fig. 1 shows that there is not a gradual transition from IGSCC to TGSCC. There is a discontinuity between the two forms, and the potential range for TGSCC is lower than for IGSCC. This implies that they have different mechanisms. If the mechanism of near-neutral pH SCC were anodic dissolution, then SCC in the pH 5.8 solution extends to lower potential from about −0.67 V (SCE), the highest potential for transgranular cracking in Fig. 1. Fig. 3 shows that the current density is about 1×10^{-4} A/cm^2 at −0.67 V (SCE). Then the CPR is about 4×10^{-8} mm/s according to Equation 3, which corresponds to the observed stress corrosion crack velocity in field or in laboratory tests. However, when the potential is below −0.67 V (SCE), the current density is reduced according to Fig. 3, and so should the CPR. When the potential is lowered below the open circuit potential, the CPR would be very small, even not detected, cease. However, this is not a case. TGSCC can occur near the open circuit potential and especially below the free corrosion potential and the CPR calculated from Equation 3 is lower than the measured CPR [42]. So a dissolution mechanism is not supported. Nevertheless, dissolution does occur within the crack for near-neutral pH SCC as evidenced by the heavy lateral corrosion on the crack sides. It is difficult to imagine that such dissolution occurs only on the crack sides and not at the crack tip.

Nevertheless, it is concluded that there is some process other than dissolution involved in the crack propagation in the low pH solution. Fig. 1 includes the line for equilibrium discharge of hydrogen. The potential range for TGSCC is below this line. This indicates that in the low pH solution, hydrogen discharge is possible, so the mechanism for near-neutral pH SCC may be related to hydrogen. Gu [10, 43] deduced an equation for the synergistic effect of hydrogen and stress on the anodic dissolution rate. He suggested that the mechanism of SCC for pipeline steels in near-neutral pH solutions might be as follows. As the anodic potential comes close to $E_{\mathrm{corr.}}$, local dissolution or pitting occurs, generating H^+, which result in local acidification within individual pits. That acidification could facilitate the crack initiation and propagation process. So Gu suggested that near-neutral pH SCC is dominated by the mechanism of hydrogen-facilitated AD. At cathodic potentials, it is suggested that, when the hydrogen concentration reaches a critical value, hydrogen induced cracking controls the cracking process.

There is a considerable amount of evidence [20, 44, 45] that hydrogen plays a critical role in the near-neutral pH solution. However the precise mechanism has not been identified. It is well known that the hydrogen can reduce the ductility of the steel without producing multiple cracks, but the same amount of hydrogen in the steel held below the yield stress may not have any noticeable effect. Normally, when severe SCC occurs, the reduction area (RA) value is low, but the embrittling effect of hydrogen can confuse the issue. Parkins [20] showed that in some specimens held near Ecorr in the near-neutral pH environment, the RA decreased from about 70 percent to 30 percent even in the absence of secondary cracking. Apparently, enough hydrogen may be introduced into the steels during the test that the ductility is reduced to 30 percent elongation. However, Fessler pointed out that an operating pipeline is never expected to undergo anywhere near 30 percent elongation, so the results from laboratory slow strain rate tests may not necessarily be applicable to an operating pipeline, and the extrapolating the laboratory results to the field can be dangerous [46]. This is reasonable.

Some investigators think that the magnitude of the RA correlates with the rate of hydrogen absorption [10]. And some think that hydrogen can reduce the yield strength of the steel, then promote the onset of plastic deformation, or can lower the total deformation which can be tolerated by the steel before fracture [47].

Whilst the mechanism of the high pH SCC is almost universally accepted, there is less agreement on the mechanism of the near-neutral pH SCC. It needs to be further studied.

7. Summary

1. When the coating deteriorates and disbonds, the ground water can be in contact with the pipeline surface. In the absence of a cathodic protection current through the crevice between the coating and the surface, TGSCC can occur. If the current flows at the surface, then the environment is changed into the high pH concentrated solution and that give rise to IGSCC.

2. For high pH SCC, the mechanism for cracking is believed to be anodic dissolution caused by preferential dissolution of some elements or active phase at the grain boundary.

3. In case of near-neutral pH SCC, there is less agreement on the mechanism. It appears that both corrosion and hydrogen may be important to the mechanism, but the precise role of hydrogen needs to be further clarified.

Acknowledgments

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