The Roles of H$_2$S Gas in Behavior of Carbon Steel Corrosion in Oil and Gas Environment: A Review

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Abstract--Hydrogen sulfide (H$_2$S) is the most dangerous element which exists in oil and gas reservoir. H$_2$S acidifies water which causes pitting corrosion to carbon steel pipelines. Corrosion reaction will increase fast when it combines with oxygen and carbon dioxide (CO$_2$). Thus, they can significantly reduce service life of transportation pipelines and processing facilities in oil and gas industries. Understanding corrosion mechanism of H$_2$S is crucial to study since many severe deterioration of carbon steels pipelines found in oil and gas industries facilities. To investigate H$_2$S corrosion accurately, it requires studying physical, electrical and chemical properties of the environment. This paper concentrates, especially, on carbon steel corrosion caused by H$_2$S gas. How this gas reacts with carbon steel in oil and gas reservoir is also discussed. This paper also reviews the developments of corrosion prediction software of H$_2$S corrosion. The corrosion mechanism of H$_2$S combined with CO$_2$ gas is also in focused.

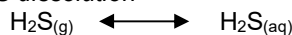*Keywords: H$_2$S Corrosion, carbon steel, oil and gas environment*

1. INTRODUCTION

The roles of H$_2$S gas in corrosion problems are important for the prediction of pipelines corrosion since many of the oil fields around the world contain this gas. The oil and gas environments which are dominated by CO$_2$ and H$_2$S gases contribute to the mostly deteriorations of pipelines. The corrosion mechanism will be complex if CO$_2$ and H$_2$S gases exist in the systems [1-3]. Intensive studies have been conducted to study the effect of H$_2$S gas in CO$_2$ system [4-10]. As discussed in many published papers, the complex chemistry and mechanism of corrosion process make it difficult to predict CO$_2$ and H$_2$S corrosion processes[10-13]. The corrosion process may involve combination of reactions between corrosion rate and film formation rate. Thus, further research is needed to investigate how H$_2$S gas affects corrosion rate in CO$_2$ system.

2. H$_2$S IN AQUEOUS SOLUTION

Hydrogen sulfide, which is weakly acidic when dissolved in water, is involved in a series of chemical reactions in the pipeline. The chemical reactions are [14]:
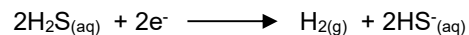
H$_2$S dissolution
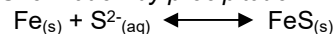$$H_2S_{(g)} \longleftrightarrow H_2S_{(aq)}$$

H$_2$S dissociation
$$H_2S_{(aq)} \longleftrightarrow HS^-_{(aq)} + H^+_{(aq)}$$

HS$^-$ dissociation
$$HS^-_{(aq)} \longleftrightarrow H^+_{(aq)} + S^{2-}_{(aq)}$$

H$_2$S Reduction

$$2H_2S_{(aq)} + 2e^- \longrightarrow H_{2(g)} + 2HS^-_{(aq)}$$

FeS formation by precipitation
$$Fe_{(s)} + S^{2-}_{(aq)} \longleftrightarrow FeS_{(s)}$$

At pressures less than 200 kPa, the solubility of molecular H$_2$S in water is given by Henry's law [15]:

$$M_{H2S} H = Y_{H2S} P$$

Where Y_{H2S} is the mole fraction of hydrogen sulfide in vapor, P is the total pressure, M_{H2S} is the molality of the molecular form of hydrogen sulfide in water (moles per kilogram of water), and H is Henry's constant.

The reactions of H$_2$S in aqueous vary with pH. At low pH (acidic solutions), the predominant form of the sulfide species is the molecular H$_2$S. This continues to be until a pH of about 6 when significant amounts of the bisulfide ion are present. Any increase in the pH will result in the formation of more bisulfide. At pH of slightly less than 7, there are equal amounts of the molecular and bisulfide forms. At a pH of 8, the concentration of the bisulfide ion is about ten times that of the molecular H$_2$S. The bisulfide ion is the dominant hydrogen sulfide species for pH greater than 8 [16, 17].

3. IRON SULFIDES FORMATION

There are three different possibilities of iron sulfide formation in aqueous. Firstly, the formation of visible black solid film due to anodic dissolution of iron occurs on the surface. The film grows through anodic process at the metal/sulfide film interface. The cathodic reaction takes place on the sulfide

film/solution interface and is limited by the diffusion of ferrous ions and electrons through the film (Figure 1.a). Second possibility is that ferrous ions dissolve into solution and react with sulfide ions in the solution, hence no film of corrosion product on the surface (Figure 1.b). Finally, is a mix of both where ferrous ions react both on the surface and in solution (Figure 1.c). The result is a porous film of iron sulfides. The porous surface facilitates the cathodic reaction and creates anodic dissolution of iron [18, 19].
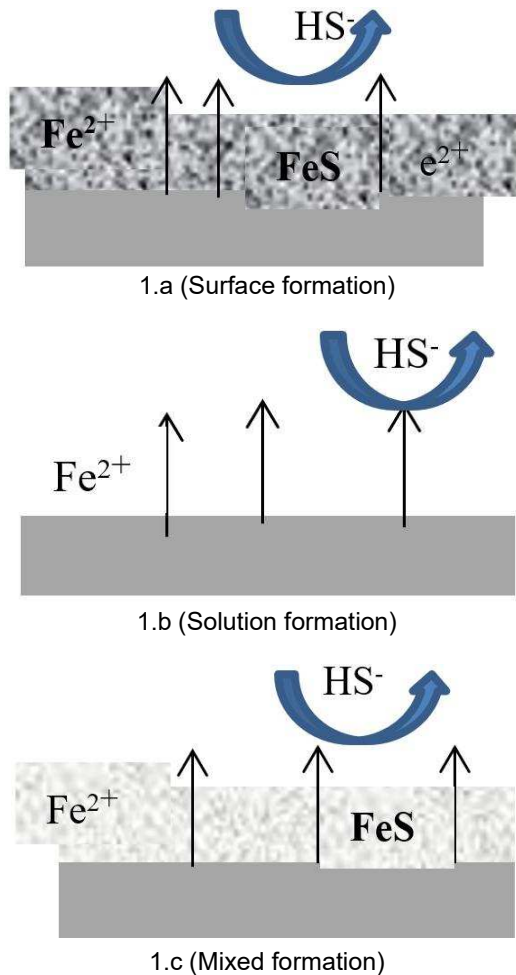

1.a (Surface formation)

1.b (Solution formation)

1.c (Mixed formation)

Figure 1. Possibilities of iron sulfide formation in H_2S saturated brine[18, 19]

Nesic et al. [18, 19], constructed a model to simulate film formation growth of CO_2/H_2S competition reactions as presented in Figure 2.3. From the simulated model, they identified that the growth of film formation containing H_2S/CO_2 gas, initially, was started by FeS film. The growth of FeS occur on the metal surface through solid state reaction. At that time, $FeCO_3$ is not been present. As the time goes on, the water chemistry changes (pH and Fe^{2+} increases). At that condition, the level of $FeCO_3$ solubility exceeds solubility permitting the dissolved $FeCO_3$ to precipitate by

diffusion reaction across the FeS film. Then, the FeCO3 film becomes thicker and denser at the metal/film interface due to an increase in pH and Fe^{2+} concentration.

4. EFFECTS OF H_2S GAS ON CORROSION

4.1 Experimental test of H2S corrosion

Kun Lin [15] set the experimental variable as; temperature 20^0C, pH 5, partial pressure 1 bar, flow rate 1000 rpm, concentration of H_2S in CO_2 in the range of 0 - 340 ppm. All of the experiments indicated that very small of amount of H_2S (10 ppm) in phase gas lead to rapid reduction of the corrosion rate. The research by Kun-Lin et al. also included microstructure analysis. Under SEM observation, they found that the scale formed on the surface that inhibited corrosion rate have a mackinawite structure. They stated that the mechanism of scale growth was not of mass-transport control, but rather a charge transfer controlled. Brown's and Kun Lin's arguments were quoted from Srinivasan who revealed that at 20^0C - 60^0C, a competition to form the protective film takes place between H_2S and $CO_{2.-}$ and the most protective film is formed at 60^0C.

A similar work was done by Hunnik et al., 1996 [31], who observed that the phenomena of accelerated corrosion in a CO_2 and H_2S environment occurs at low H_2S concentration. But, this effect diminished at H2S concentrations in excess of 15 ppm in the gas phase and did not appear to be dependent upon flow velocity. The conditions of their study were 60^0C and 0.79 MPa CO_2 partial pressure in a multi phase flow.

Bruce Brown, 2005 [32], working with temperature of 60°C, 0.77 MPa CO2, pH 6, 120 ± 10 ppm H_2S, found that three layers of film had developed on the surface as shown by an XRD of a cross-sectional view of the coupon. From the surface of the coupon, the first 60 micron of the layer was analyzed with EDS and found to have 0% Sulfur, 40% Iron, 13% Carbon, and 30% Oxygen. The next 60 micron layer was found to have 0% Sulfur, 45% Iron, 13% Carbon, and 16% Oxygen. The outermost 30 micron layer was found to have 11% Sulfur, 33% Iron, 22% Carbon, and 13% Oxygen.

Andrzej Anderko and Robert D. Young [33] proposed a model involving thermophysical properties, electrochemical properties, and scale effects to predict corrosion rate. They reported significant drop in corrosion rate for partial pressures of H_2S ranging from 2.10 - 6 to 10 - 4 atm and the rate reached a plateau in a relatively wide range of H_2S partial pressures above 104 atm. Reduction in corrosion rates has been reported when the H_2S partial pressure exceeds 10 - 3atm in some systems. At substantial H_2S partial pressures (above ca. 10-2 atm), the aqueous H_2S, and HS- species become sufficient

to increase the corrosion rate. That observation is supported by Chengqiang Rena who found that corrosion rate in CO_2 system will decrease quickly as compared to sweet corrosion in low concentration of H_2S.

J. Kvarekval, R. Nyborg and M. Seiersten worked with 0.5 – 2 bar of H_2S [34]. Experiments with up to 2 bar CO_2 and temperatures up to 80°C resulted in slightly higher corrosion rates than in corresponding experiments without H_2S. The corrosion rates were in the range of 0.1-2 mm/y. In an experiment with 0.5 mbar of H_2S at a CO_2/H_2S partial pressure ratio of 4500, both iron sulfides (FeS) and iron carbonates ($FeCO_3$) were detected on the steel surface. The mixed sulfide/carbonate films were 30-80 μm thick. Experiments with CO_2/H_2S ratios of 1200-1500 resulted in formation of thin iron sulfide films (1- 10 μm) on the corroding surfaces. No iron carbonates were found in corrosion product films formed at CO_2/H_2S ratios below 1500.

Papavinasam and R.W.Revie [35, 36], in their experiment, concluded that corrosion rate increased both with H2S partial pressure and with rotation speed up to approximately 500 rpm. Beyond 500 rpm, the synergism was lost and the corrosion rate decreased (at 20 psi CO2). At 100-2000 rpm, corrosion rate increased due to H2S pressure until 75 psi, then decreased after that (at 20 psi CO2). At 25 – 100 psi H2S pressure, corrosion rate decreased with the rotation speed until 500 rpm, and increased beyond this value (at 20 psi CO_2).

Makarenko, S. P. Shatilo, Kh. Kh. Gumerskii and V. A. Belyaev [37] stated that corrosion process in CO2 environment is due to the combined effect of H_2S and CO_2. The process is accelerated by cathodic reaction of hydrogen ion reduction. It has been proven that CO_2 corrosion of carbon steel increases by 1.5–2 times with increase of H_2S content in the mixture (p H2S<0.5 MPa) in the temperature range 20–80°C. Further increase in H_2S content (p H2S≥0.5–1.5 MPa) weakens corrosion, especially in the temperature range 100–250°C, because of the influence of FeS and $FeCO_3$ on corrosion. It may relate to formation of protective film.

4.2 Effects of rotation speed on corrosion rate on H_2S corrosion

The effect of flow on corrosion rate has been studied by Silverman et al. [4-9, 27-29, 38]. They explained that flow increases corrosion due to a combination of mechanical effects due to water motion, and electrochemical effects of corrosion. Higher velocity is directly associated with higher turbulence that promotes mixing in the solution. This affects both the corrosion rate of the bare steel surface and the precipitation rate of iron carbonate. Prior to any film formation, high velocity will lead to increased corrosion rate. The

transport of cathodic species toward the steel surface is enhanced by turbulent transport. At the same time the transport of Fe $^{2+}$ ions away from the steel surface also increases, leading to a lower concentration of Fe $^{2+}$ ions at the steel surface. This results in surface supersaturation and thus precipitation rate becomes lower [38].

5. PREDICTION OF CORROSION RATE OF CARBON STEEL IN H_2S GAS

5.1 Mechanistics model [39-46]

A mechanistic model of uniform hydrogen corrosion can be modeled using fundamental physicochemical laws [47]. It consists of convective diffusion equation, molecular diffusion equation and diffusion via solid film equation. Convective diffusion reactions (flux) through boundary layer are caused by mass transfer processes (Km), bulk concentration (Cbi) and outer concentration (Coi). (6) (ii) Molecular diffusion (flux) through the liquid in the porous outer scale: From the this parameters, expressed a mathematical corrosion theory (CR) as [14,31]:

$$CR_i = A_i e^{\frac{B}{RT_K}} \ln \frac{c_{b,i} - CR_i \left(\frac{\delta_{oc}}{D_i \varepsilon \Psi} + \frac{1}{k_{m,i}} \right)}{c_{s,i}} \qquad (1)$$

Where:

K_m is mass transfer coefficient of species I (m/s)

$c_{b,}$ is bulk concentration of species i (mol/m^3)

c_o is the interfacial concentration of species i at outer scale/solution interface (mol/m^3)

D_i is diffusion coefficient for dissolved species i (m^2/s)

ε is outer scale porosity

ψ is tortuosity factor

c_i is interfacial concentration of species i

δ_{os} is the thickness of outer film sacle

δ_{hbl} is turbulence boundary layer thickness

δ_{mbl} is mass transfer boundary layer thickness

δ_f is film thickness

A is Arhrhenius constants

T_k is temperature (Kelvin)

c_s is surface concentration.

5.2 Electrochemistry Processes in CO_2/ HAC/ H_2S System

The electrochemical is chemical reactions where electrons are transferred between molecules which called oxidation/ reduction reactions. To investigate the corrosion mechanisms, electrochemical processes take account into the metal surface and transport process for species that participate in the reactions. The model focuses in cathodic and anodic reactions which happen in the systems involved.

5.3 Corrosion prediction software

There are many equations that predicts the corrosion rate in CO_2 environments. These include the de Waard [de Waard [48] and its many subsequent derivatives, Yuhu, Vera and Nesic et al. [49]. All of these were developed based on different systems and assumptions. Some corrosion predictions softwares that have been developed based on semi-empirical approach are discussed herewith.

- **NORSOK [50]**

The NORSOK standard is owned by Norwegian Oil Industry Association and Federation of Norwegian Manufacturing Industries. The program covers only calculation of corrosion rates where CO_2 is the corrosive agent. It does not include the corrosivity, e.g. contamination of O_2, H_2S etc. The model is an empirical model for CO_2 at different temperatures, pH, CO_2 fugacities, wall shear stresses, and temperatures from 20 to160°C.

The model calculates pH and wall shear stress. The effect of acetic acid is not account for in this model, but it is still valid to predict corrosion rate if the concentration of acetic acid is less than 100 ppm. The corrosion rate is calculated as in the following equation:

$$CR_t = K_t fCO_2^{0.62} x (S/19)^{0.146+0.0324\log(fCO_2)} f(pH)t \quad (2)$$

Where CR is the corrosion rate (mm/yr), K_t is the constant for temperature t, f CO_2 is the fugacity of CO_2 (bar), S is Wall shear stress (Pa), $f(pH)t$ is the pH factor at temperature t.

- **ECE (Electronic Chemical Engineering) [51]**

ECE program software calculates corrosion rate based on the modified de Waard and Milliams method. ECE model includes oil wetting correlation based on field correlation. For low horizontal flow velocities < 1 m/s, the F_{oil} =1. ECE proposes a corrosion prediction expression as follows:

$$V_{cor} = \frac{1}{\frac{1}{V_r}+\frac{1}{V_m}} \quad (3)$$

Where, V_r is corrosion reaction and V_m is mass transfer effect.

Where mass transfer effect is determined by temperature, pCO_2, fugacity CO_2, pH of pure water saturated with CO_2 at prevailing temperature and pressure. The fugacity of CO_2 is similar to its partial pressure, but corrected for non-ideality of CO_2 at high pressure and temperature. The mass transfer represents the main part of the dependence on flow velocity U and pipe diameter.

- **Cassandra (DWM 93) [52]**

Cassandra is a model developed based on the experiences of de Waard and BP [53]. The input includes pH, CO_2 concentration, temperature, and water contaminant. This model does not consider scaling temperature. The user must set an assumption of the scaling temperature. The basic formula to calculate corrosion rate is expressed as in temperature dependency. This model has important aspects which influence rate of corrosion, namely corrosion inhibitor availability and corrosion risk categories. The model also accounts for the presence of acetate in water as acetic acid. The major input to the model are: CO_2 mole %, temperature, total pressure, liquid velocity and water chemistry. Besides that, the model has secondary input, such as hydraulic diameter and glycol concentration, oil type (crude or condensate) and water type (condensed water or formation water). The effect of oil wetting in this model is not included.

6. ACCURACY OF CORROSION PREDICTION MODELS OF H₂S CORROSION

6.1 Comparison model of H₂S corrosion with experiments data

The comparison of the model with Hedge's [54] and George's [55] experimental data are shown in references [25]. A good agreement between experimental and calculated data based on this corrosion prediction model. Comparing to Hedge's experiments at 60°C, the model has R^2 of 94%, correlation of 97% and standard error estimation deviation of 0.28. In comparison to George's experimental data, the model shows a relationship with R^2 of 93%, correlation of 97%, and standard deviation of 0.5. George's experiment showed a good relationship in correlation and regression relationship; but it provided less precision (0.5) in standard error estimation.

6.2 Comparison corrosion rate (mm/y) at Various Conditions in pH 4 and CO₂/H₂S system

Table 1 shows comparison between corrosion software FreeCorp [46] and model experiments [25]. Below the table is presented performance of the model compared to software. It can be seen that root sum square (RSQ) of the model are, 0.5 temperature 25°C, 0.7 for 30, 40 50 and 70°C. Correlation from 0.7 – 0.9 for various temperature tested. And standard error estimation from 0.1 – 0,3.

Table 1. Corrosion data calculated using FreeCorp [46] software and model from experiments

Temp. (ºC) Rot. (rpm)	25		30		40		50		60		70	
	Frc	Model	Frc	Model	Frc	Model	Frc	Model	Frc	Model	Frc	Model
100	1.1	1.5	1.3	1.6	1.6	1.7	1.9	1.9	2.4	2.2	2.9	2.6
1000	1.5	1.6	1.6	1.7	2.1	1.8	2.5	2.0	3.3	2.3	4	2.7
2000	1.5	1.7	1.7	1.8	2.1	2.0	2.6	2.2	3.4	2.4	4.1	2.8
3000	1.5	1.8	1.7	1.9	2.1	2.0	2.6	2.2	3.4	2.5	4.2	2.8
4000	1.5	1.9	1.7	1.9	2.1	2.0	2.6	2.3	3.4	2.6	4.2	2.9
5000	1.5	1.9	1.7	2.0	2.2	2.1	2.6	2.3	3.4	2.6	4.2	2.9
RSQ :		0.5	0.7		0.7		0.7		0.6		0.7	
Correlation :		0.7	0.8		0.8		0.8		0.8		0.9	
St. error :		0.1	0.1		0.1		0.3		0.3		0.1	

7. EFFECTS OF FILM FORMATION OF H2S GAS ON CARBON STEEL CORROSION

Figure 2 presents the simulated corrosion rate of carbon steel due to the presence of H_2S in CO_2 environment, calculated based on Formula 1. The individual effects of H_2S on corrosion rate under two different conditions were simulated i.e. free of film formation and with film formation. In free film formation, the corrosion is found to be controlled by activation process. On the other hand, when film formation exists, corrosion is controlled by diffusion limiting current. As can be seen in the graph, in free film condition (red line) the corrosion rate will increase if H_2S concentration increases. In contrast to film formation condition, an increase in H_2S concentration will reduce the corrosion rate. These results meet argument that carbon steel release atomic weight in the CO_2/H_2S environments during corrosion process.
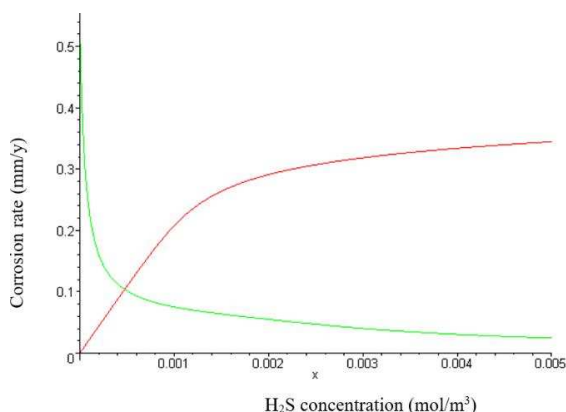

Figure 2. Comparison carbon steel corrosion with film formation (green line) and without film formation (red line)

8. CONCLUSION

H_2S corrosion has major impacts on safety, reliability, and integrity of oil and gas industries. It damages pipeline transportation and process equipment. Combining this gas with other elements such as CO_2 and O_2 will accelerate corrosion rate. Early detection is crucial issue for controlling corrosion process. Successful model prediction is needed to predict corrosion rate. However, a data experiments are still required for verification. The current level of effectiveness of corrosion software prediction is not sufficient to address the safety, reliability, and reduce risk of corrosion. To overcome this situation, studying corrosion process should involve understanding of electrochemical process, physical properties, and chemical behavior.

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