

H₂S Corrosion of Carbon Steel in Geothermal Fluids

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Keywords: *H₂S Corrosion, Geothermal, CO₂ Corrosion, Heavy Metal Deposition, Carbon Steel.*

ABSTRACT

Corrosion of carbon and low alloy steels in H₂S containing geothermal two phase fluids, separated steam and superheated steam has long been known to be controlled by the formation of adherent and continuous iron sulphide and iron oxide corrosion products that provide a barrier to the geothermal fluids. This corrosion control mechanism is effective within a narrow range of several factors including: H₂S concentration, pH range, CO₂ to H₂S ratio and chloride concentration. H₂S corrosion control can be lost for example by formation of acid condensate, precipitation of excess metal sulphides, deposition of heavy metals from two-phase or brine solutions and aeration. This paper will summarise the beneficial effects of H₂S in many geothermal fluids and contrast these with instances where H₂S corrosion control of carbon and low alloy steels is lost.

1. CORROSION CHEMISTRY OF GEOTHERMAL FLUIDS

The first step of evaluation of the corrosivity of geothermal fields is to compare the corrosion chemistry of such fields with that observed in characterized systems. Corrosion properties of fluids derived from production wells of geothermal fields in the Taupo Volcanic Zone (TVZ) of New Zealand have been extensively studied. The fluids produced from TVZ geothermal resources are primarily two-phase fluids at the wellhead and these are separated to provide the separated steam and separated water (brine) that are used for energy production. On separation of two phase fluid, a greater percentage of the corrosive gases CO₂ and H₂S tend to partition into the steam phase than gaseous NH₃ does. This means that after considerable steam separation, some ammonia is concentrated into the steam phase, but water retains an appreciable amount even after extensive steam formation. (Ellis and Mahon, 1977). Table 1 summarises separated steam gas chemistries for three New

Zealand geothermal fields, including Broadlands and Wairaki in the TVZ and Ngawha in Northland. Single phase dry, superheated steam wells are encountered in New Zealand and internationally. Single phase hot brine systems are also used for energy production internationally but are less common in New Zealand.

The TVZ separated steam condensate chemistries are in the near neutral pH of 6.3-6.6, with relatively low chloride concentration from separator carryover and have gas concentrations ranging from 0.1 wt% to 8 wt%. The Ngawha geothermal field in Northland, which is outside the TVZ of New Zealand, has a higher gas content, up to 10 wt% (Lichti and Julian, 2010, Wilson and Lichti, 1982). The corrosivity of the geothermal steam depends on the relative amounts of CO₂, H₂S, NH₃ and H₂. These species as well as dissolved solids present in the steam, as a consequence of carryover from steam/water separators and entrainment in discharge fluids from dry wells, together with small amounts of condensate arising from heat losses, determine the corrosion chemistry of air-free separated steam. Using the measured concentrations of gases in the steam phase and dissolved ionic species in the brine phase, high temperature and high pressure water phase chemistry of the corrosive water phases can be calculated (see for example, Glover 1980, Arnorsson et al, 1982, Arnorsson and Bjanmason, 1993).

Separated steam condensate chemistries were calculated for the three fields of interest for this paper using the data of Table 1 and the method of Glover, 1980. The results are given in Table 2 for the species required for thermodynamic modelling: Total CO₂, Total H₂S, Total NH₃, pH(T) and partial pressure of H₂. These calculated water phase chemistries characterise the corrosive electrolyte encountered by metals and alloys placed in these environments and this data can be used as input conditions for thermodynamic models that characterise the stability of iron based corrosion products for example.

Table 1. Separated steam gas chemistries of three New Zealand geothermal fields with different gas content (Julian and Lichti, 2010, Braithwaite and Lichti, 1981).

Field	Well	CO ₂	H ₂ S	NH ₃	H ₂	Residual	Gas in Steam	CO ₂ /H ₂ S	Produced fluid pH
		mmol/100mol steam					wt%		
Broadlands	BR22	841	17.6	5.4	0.79	16.2	2.1	47.7	6.5
Wairakei	HP steam	39.1	2.39	0.5	0.25	-	0.1	13.4	6.3
Ngawha	NG9	2825	35.2	34	7.6	79.3	7.3	80.3	6.6

Table 2. Calculated separated steam condensate chemistries of three New Zealand geothermal fields with different gas content using the method of Glover, 1980.

Variable	Wairakei	Broadland	Ngawha	Units
Temperature	160	160	160	°C
Pressure	618	618	618	kPa(a)
Partial Pressure				
pCO ₂	2.55E-01	5.44E+00	1.79E+01	kPa
pH ₂ S	1.56E-02	1.15E-01	2.30E-01	kPa
pSO ₂	0.00E+00	0.00E+00	0.00E+00	kPa
pNH ₃	3.26E-03	3.52E-02	2.22E-01	kPa
pH ₂	1.63E-03	5.16E-03	4.96E-02	kPa
pN ₂	0	1.06E-01	5.17E-01	kPa
Condensate Chemistry				
pH(160°C)	6.53	6.52	6.67	
CO ₂	2.16E-05	4.61E-04	1.52E-03	mol/kg
HCO ₃ ⁻	1.04E-05	2.20E-04	1.05E-03	mol/kg
Total C	3.20E-05	6.81E-04	2.57E-03	mol/kg
H ₂ S	4.44E-06	3.27E-05	6.54E-05	mol/kg
HS ⁻	2.03E-06	1.49E-05	4.31E-05	mol/kg
Total S	6.47E-06	4.76E-05	1.09E-04	mol/kg
NH ₃	3.23E-05	3.49E-04	2.19E-03	mol/kg
NH ₄ ⁺	2.19E-05	2.45E-04	1.11E-03	mol/kg
Total N	5.42E-05	5.93E-04	3.30E-03	mol/kg

These potential-pH diagram models called Pourbaix diagrams after their inventor (Pourbaix, 1974) can be prepared for example for the Fe-H₂O-H₂S system and for the Fe-H₂O-CO₂ system which are of primary interest for geothermal fluids. The potential-pH Pourbaix diagrams describe the equilibrium corrosion reactions of pure iron in contact with the water phase at the specified temperatures. Pourbaix diagrams are based on the calculated water phase composition and thermodynamic data which must often be extrapolated from the temperature at which they are measured to the temperature of interest.

However, there are some factors which should be considered regarding using these diagrams, they are as follows:

- They do not include the multiplicity of secondary alloying elements or all of the possible compounds that may form unless thermodynamic data is available for all the elements present in the alloy. However, for systems where stable corrosion products have been collected and are predominantly of a single alloying element, the predictions of these diagrams have been confirmed (Wilson and Lichti, 1982).
- The diagrams give no indication of corrosion rates and they cannot be directly used for prediction of corrosion rates for new geothermal fields under consideration. However, we can correlate known corrosion results for the well-known fields with their corrosion chemistry and compare this with that of the new field under consideration.

Pourbaix diagrams for separated steam at Broadlands, Wairakei and Ngawha fields originally prepared by Wilson and Lichti, 1982, have been reproduced using more recent thermodynamic data as given in Figure 1. The diagrams of

Figure 1 show that for Broadlands and Ngawha steam condensates, iron sulphides will form on carbon steel but they show greater stability at Ngawha than at Broadlands. The reason for this is that the condensate derived from Ngawha steam contains significantly higher concentrations of dissolved sulphide than that of Broadlands steam condensate. It is however difficult to predict the corrosion performance of carbon steel in these systems particularly in view of the known tendency to pitting corrosion (Lichti et. al, 1981) in chloride and sulphide containing solutions and the complexity of corrosion products.

In the case of Wairakei steam condensate the Pourbaix diagram shows stability of magnetite with smaller regions of stability for the iron sulphides. Corrosion investigations in Wairakei carbon steel geothermal steam pipelines showed magnetite (up to 3mm thick after 30 years of service) was the predominant corrosion product with particulates of pyrite (FeS₂) within the magnetite scale, which correlated with predictive thermodynamics models (Braithwaite, 1978, Lichti and Bacon, 1998).

2. CORROSION PRODUCTS AND CORROSION RATES

Corrosion rates for carbon steel in transfer pipelines are mostly low and acceptable, and in conjunction with a suitable corrosion allowance, give a long life (Lichti et al., 1997). The successful use of mild and low alloy steels in geothermal steam depends on the formation and properties of corrosion product films which cover and protect the underlying metal. The corrosion products and corrosion rates depend on temperature and steam condensate chemistry.

Investigation of corrosion products and corrosion rate of mild steel coupons exposed at the Broadlands BR22 corrosion test rig (Borshevska et. al 1982) showed that the moderate hydrogen sulphide content of the Broadlands steam leads to the formation of a series of iron sulphides, including mackinawite, cubic iron sulphide, troilite, pyrrhotite and pyrite. Iron sulphide phase development in steam and bore fluid environments were toward sulphur rich iron sulphides with increasing time of exposure. The nucleation and growth of well-formed crystals depends on temperature and on the degree of supersaturating of sulphide ions. Transformation of the initial iron rich sulphides to sulphur rich sulphides depended on temperature.

Results for the Broadlands field showed that Pyrrhotite (iron deficient) was the major sulphide phase in bore fluid. Troilite (iron rich) was identified as major phase for separated steam (T=160°C). The presence of mackinawite, troilite and or pyrrhotite was confirmed for wet steam (T=105°C).

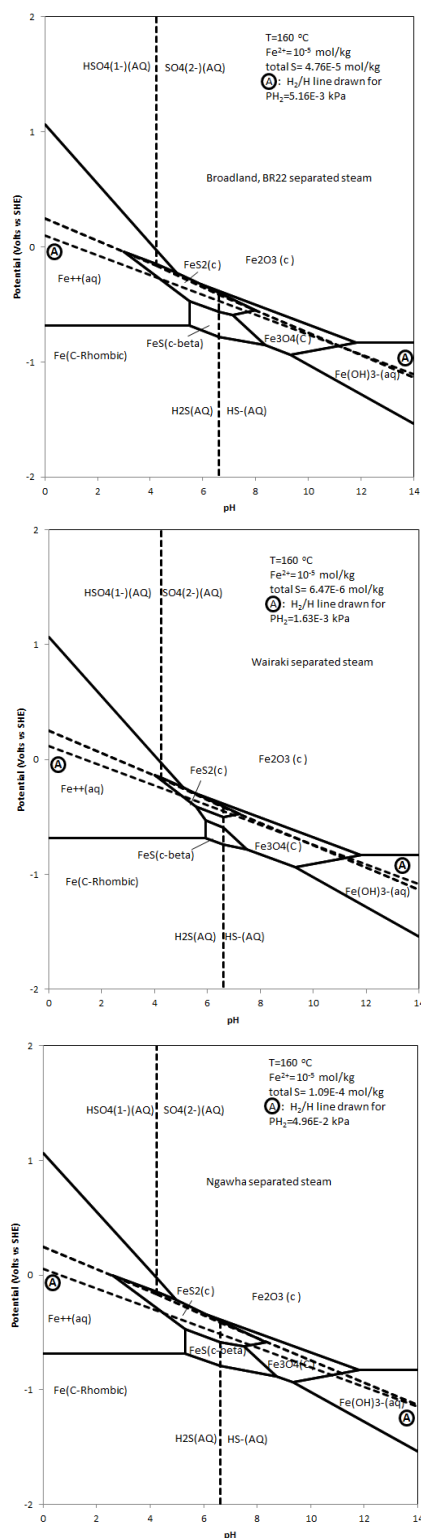


Figure 1: Potential-pH Pourbaix diagrams for separated steam at Broadlands, Wairakei and Ngawha, using the data of Table 2.

In all non-aerated test environments, i.e. bore fluid, separated steam and wet steam, a thin magnetite layer adjacent to the metal was observed which was covered by a layer of iron sulphide. The outer layer of sulphide acts to shield the magnetite layer from the sulphide containing solution. The results also showed that a greater degree of

shielding was obtained where dissolved SiO_2 was present or the condensate in contact with mild steel coupons had a higher pH. The beneficial effects of dissolved silica have been reported as being most important for low H_2S systems where magnetite stability dominates as in Wairakei steam pipelines (Braithwaite, 1978). Lichti and Bacon 1998 documented the level of silica required to stabilise magnetite. In the absence of silica the magnetite formed initially slowly dissolved leading to local breakdown under turbulent conditions giving accelerated localised erosion corrosion. The dependence of erosion rate on Reynolds Number was demonstrated by Soylemezoglu et. al, 1980 as illustrated in Figure 2.

Coupon exposed in test vessels with Broadlands near static steam environments and inspections of Broadlands steam pipelines, where iron sulfide stability dominates, showed no signs of silica influence. (Inman, 1994).

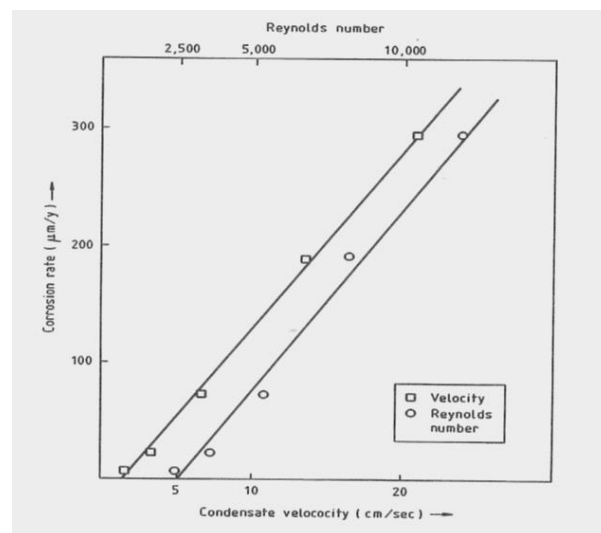


Figure 2: Effect of Wairakei steam condensate velocity and turbulence on corrosion rate of carbon steel, Soylemezoglu et. al, 1980.

3. EFFECT OF AERATION AND HEAVY METAL DEPOSITION

3.1 Effect of Aeration

H_2S corrosion control of carbon and low alloy steel in geothermal fluids is lost in the presence of aeration. Protective sulphide and oxide corrosion products in near neutral to alkaline pH fluids become unstable when oxygen is present even intermittently. Therefore, oxygen contamination of geothermal fluids must be avoided (Lichti, 2017). Braithwaite and Lichti, 1980, investigated the effect of continuous air ingress on surface corrosion of carbon and low alloy steel for Broadlands geothermal field. Their results showed that a thick massive corrosion product, which was blistered and porous, formed in aerated wet steam. In aerated condensate a thick deposit of non-adherent corrosion product formed, which easily washed off to reveal underlying metal. This was in contrast to adherent uniform films of corrosion product that were found on mild steel in bore fluid and non-aerated condensate. Corrosion rate results showed that carbon steel corrosion rates increased from 60.2 $\mu\text{m}/\text{year}$ in separated bore water to a high corrosion rate of

3.3 mm/year in aerated wet low-pressure steam. Material losses were also increased substantially on aeration of steam condensate. On aeration corrosion products in steam included iron oxides (hematite, goethite and magnetite), as well as iron sulphides and elemental sulphur. In the case of aerated cold condensate, iron carbonate, sulphur and goethite ($\alpha\text{-Fe}_2\text{O}_3\cdot\text{H}_2\text{O}$) were identified. The corrosion products on carbon steels were porous, poorly adherent and blistered. The effect is thermodynamically equivalent to changing the potential from reducing conditions to oxidising conditions on the potential-pH diagrams illustrated in Figure 1. The potential of the corrosion reactions moves into the region of stability of hematite, Fe_2O_3 and acidity increases through formation of sulphuric acid in the porous corrosion products. Soylemezoglu and Harper (1982) studied the rate of increase of corrosion in separated steam with increasing aeration using electrical resistance on-line corrosion monitors. Their work demonstrated that the corrosion was greatest downstream of the air injection point indicating some time was required for reaction of the oxygen with H_2S for formation of corrosive sulphur species.

The effect of aeration can be seen in energy plant applications such as steam pipelines subjected to wet aerated shutdowns. These pipelines can show localised formation of such corrosion products over heavily corroded and pitted areas while other regions are unaffected, see Figure 3. (Lichti et al, 2003).

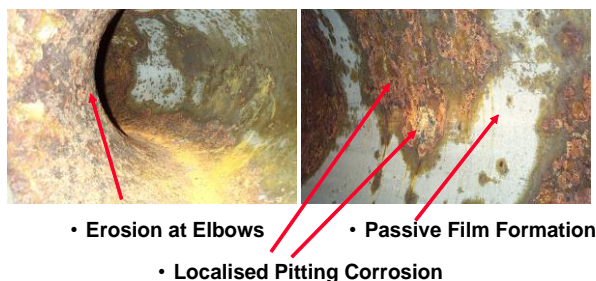


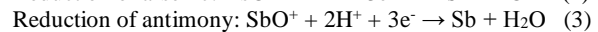
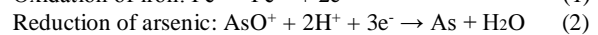
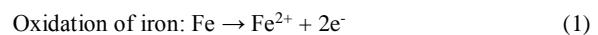
Figure 3: Illustration of steam pipeline under frequent wet aerated shutdown conditions showing erosion corrosion at an elbow (left) and a mix of protective film formation and localized corrosion (right).

3.2 Heavy Metal Deposition

The water phases of geothermal fluids in many cases may contain traces of heavy metals, such as gold, silver, thallium, arsenic, antimony and copper to mention a few. Under certain conditions, electrochemical reduction of these heavy metal species, particularly of antimony, antimony/silver and arsenic can result in deposited scales forming on freely corroding carbon steel. Precipitation of unwanted scales and corrosion in plant and piping causes major engineering challenges, leading to reduced efficiencies, plant shutdowns, costly scale removal and limits on downstream use of fluids. Apart from that, these scales, formed in conjunction with oxidation of iron have subsequently been associated with localised galvanic corrosion, under scale pitting and erosion corrosion of the underlying carbon steel (Lichti and Brown, 2013).

Under low pH conditions, both in two-phase fluids and in brines, arsenic and antimony have been observed to precipitate as metallic materials (see for example Osato et al 2016, Osato et al, 2017). Acid well fluids are not utilized in New Zealand geothermal production, however, acid process conditions can be encountered either through deliberate modification of the environment, for silica scale control for example or in power station circuits where the acidic geothermal gases (CO_2 and H_2S) are concentrated (Lichti et al, 2015).

Heavy metal deposition occurs through oxidation and reduction reactions that are illustrated below for arsenic and antimony as proposed by (Gallup, 1992):



Silver and lead have also been observed in the resulting heavy metal scales in some geothermal energy plants. The process of oxidation/reduction has been observed to show pH dependence as it appears that iron must be rapidly taken into solution and not precipitated as a protective corrosion product film for the elemental scales to form. Under alkali and near neutral to slightly acid conditions where iron oxides and iron sulphides are stable heavy metal precipitation has not been observed (Lichti and Brown, 2013). The conditions leading to corrosion are poorly understood.

These heavy metals can form uniform dense scales and stop corrosion but in most cases the scales have inherent flaws and defects that results in localised under deposit pitting corrosion. Laboratory testing with arsenic and antimony ion beam sputter deposited coatings on carbon steel coupons under deaerated pressurized high temperature conditions have shown that the primary damage mechanism observed on the underlying carbon steel was one of localized pitting corrosion (Lichti et al, 2016). The corrosion mechanism was similar for both arsenic and antimony coated carbon steel in the deaerated low CO_2 , low H_2S , chloride containing environment tested. Results also demonstrated enhanced corrosion damage accumulation by the galvanic effects of the coatings on pit propagation.

Alternative materials that do not readily corrode in the mildly acidic fluids can be specified but these are typically retrofitted (see for example Amends and Lee, 2013) as the risk of heavy metal induced corrosion cannot be predicted. Therefore, one of the challenges in materials selection for new geothermal fields is development of predictive geochemistry models for heavy metal reduction processes. New initiatives in scaling and corrosion control, alternatives to pH adjustment such as silica anti-scalants and the combination of pH adjustment plus corrosion inhibitors have been proposed as corrosion control options (Lichti, 2017).

4. LOW H_2S AND HIGH CO_2

Corrosion control by formation of protective sulphide and oxide corrosion products is only effective within a narrow range of factors including H_2S concentration, pH range, CO_2 to H_2S ratio and chloride concentration. In CO_2 dominated environments found in some liquid binary plant operations or in enhanced geothermal systems (EGS), the corrosion

tendency of steel has been shown to be dependant not only on the properties of magnetite but also on the properties of iron carbonate (siderite) corrosion product. The formation of a stable iron carbonate layer is highly dependent on factors such as partial pressure of CO₂, pH, temperature, flow velocity and iron concentration for example. It is well-known that the CO₂ corrosion rate increases with temperature up to a certain point (typically around 90°C) and then decreases at higher temperatures due to changing of the protective corrosion product on the steel surface from mainly iron carbonates to iron oxides, see for example Figure 4 reported by Humphreys et al, 2015.

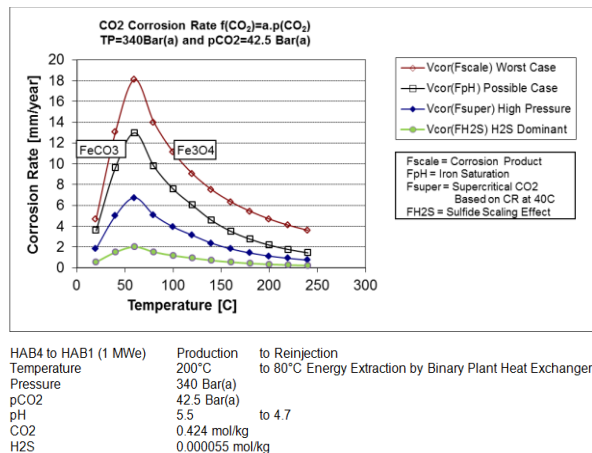
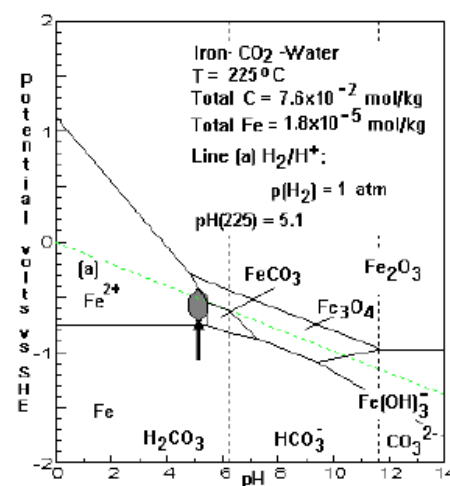


Figure 4. Example of corrosion model for CO₂ and H₂S corrosion at Habanero EGC system. (Humphreys et al, 2015)

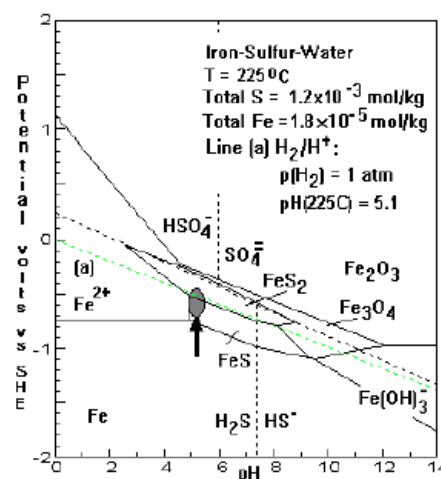
Where both CO₂ and H₂S species are present at high temperatures, both iron oxide and iron sulphide phases may form on the metal surface, see Figure 5. Although the formation of sulphide is favourable thermodynamically iron oxides may dominate at the high temperatures while iron carbonates dominate at the lower temperatures. Figure 5 illustrates that sulphide stability is predicted even at very low concentration of H₂S in the system at high temperatures, however, the kinetics of surface scale formation in the mixed CO₂/H₂S system are complicated, being dependent on the competitive kinetics of the two scale formation mechanisms and also on the chemistry of the brine (e.g. CO₂ to H₂S ratio) and the respective solubility of iron carbonate, iron oxide and iron sulphide phases.

CO₂ corrosion has been extensively studied in oil and gas applications where the combined CO₂/H₂S systems are generally divided into three regimes based on the concentration of H₂S (i.e. pCO₂ to pH₂S ratio) as defined by Pots et al., 2002 and illustrated in Figure 6. Nevertheless, the mixed CO₂/H₂S zone has been reported to begin at a much smaller ratio than the one displayed in Figure 6 (Brown et al., 2003). For example in glass test cells, corrosion testing completed by Sun and Nesic, 2006, iron sulfide scale, mackinawite, was detected even at CO₂/H₂S ratio of 1,000 with high Fe²⁺ concentration. However as discussed previously, the chemistry of iron sulfide scale formation is very complex and the scale characteristics and morphology can change with test conditions and time and can lead to very different scenarios of corrosivity. When H₂S is present in significant concentrations in a CO₂ containing system, the formation of sulphide and oxide phases can dominate

kinetically over the less protective carbonate phases and give a more stable protective layer.



Model Chemistry with pCO₂ = 6.9 bara



Model Chemistry with pH₂S = 0.02 bara

Figure 5. Potential-pH Pourbaix diagrams in pure CO₂ system (top) and with low levels of H₂S (bottom).

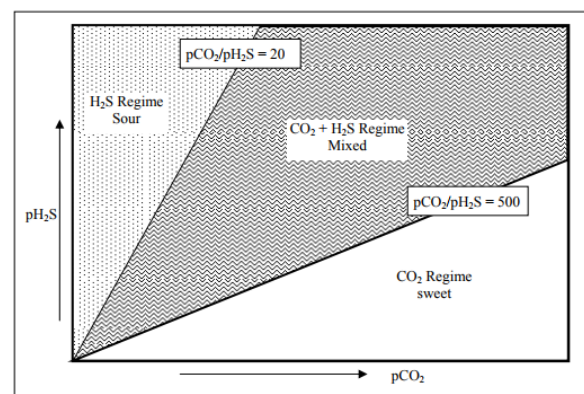


Figure 6. Corrosion regimes in CO₂/H₂S corrosion defined by Pots, et al. (2002).

5. PREDICTION OF STEEL PERFORMANCE IN NEW GEOTHERMAL FLUIDS

The prediction of materials performance in a geothermal fluid containing significant H_2S is possible even with relatively high levels of CO_2 present. Of particular importance in determining the performance in new fluids is the development of a material performance database. That is, understanding the chemical conditions that lead to corrosion or formation of protective corrosion products to predict the performance in new conditions. This may include knowledge gained from both field exposure tests and laboratory testing. Data may be used directly or extrapolated to help estimate the potential for corrosion product formation, deposit scaling and corrosion characteristics. Wilson and Lichti (1982) described how this methodology could be implemented utilizing data from Broadlands and Wairakei to estimate the performance at the Ngawha field and this was later proven in tests in a high gas geothermal fluid by Lichti and Julian, 2010.

The prediction of geothermal well corrosion has been successfully completed using a combination of geochemistry models, wellbore simulators and potential-pH Pourbaix diagrams. TVZ systems have been mainly dominated by near neutral two-phase fluids that give limited corrosion due to the formation of protective iron sulfides and underlying iron oxides. (White et al 2000) Internationally, these conditions are not always replicated and in Japan and the Philippines for example lower pH fluids are often encountered. Sanada et al (2000) illustrated possible downhole corrosion processes that may be encountered, see Figure 7. These authors demonstrated how knowledge of the downhole corrosion processes are required before processes and materials can be applied to control the risk of corrosion.

Carbon steels are typically the materials of choice for downhole casing and above ground piping for geothermal energy plant. For carbon steel in a H_2S containing geothermal fluid the corrosion mechanisms are controlled by maintaining a stable iron-sulfide layer and a stable underlying magnetite layer. The modelling of thermodynamic conditions and historical corrosion testing has been used to help predict corrosion rates.

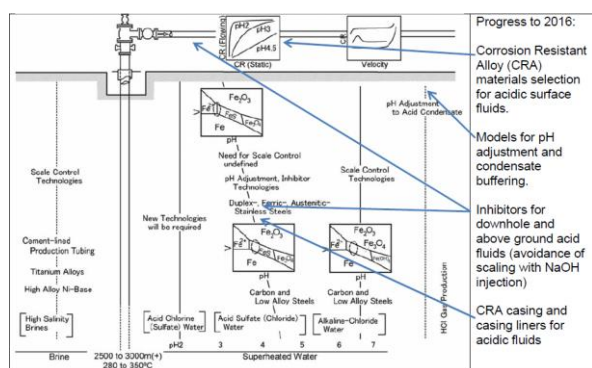


Figure 7: An illustration of the how to control corrosion and select material for deep and acidic geothermal wells. (base figure from Sanada et al, 2000, with updates from Lichti 2017)

pH adjustment has been demonstrated to successfully control downhole corrosion in some instances (Costa Rica). The use of Pourbaix diagrams or other thermodynamic

modelling allows for the target pH to be determined. (see White et al 2000, Osato et al, 2017)

While the materials selection for geothermal energy plant is predominantly based on the continuous operation of the plant, it is important to also understand how the plant will perform during upset and shutdown conditions. This may be related to ingress of air or loss of process control procedures. For example when considering the use of the pH adjustment for plant there is the risk of losing control of the pH adjustment. Rippon (2001) demonstrates this for an oil and gas scenario when, for high-risk pipelines, the choice of selecting a corrosion resistant alloy may be required to mitigate the risk during a failure of corrosion control.

6. CONCLUSION

Materials selection and prediction of materials performance in geothermal fluid has been a challenge in geothermal energy projects for many years. Corrosion of carbon and low alloy steels has been shown to be controlled by formation of protective sulphide and oxide corrosion products in near neutral pH fluids. Under acidic conditions these corrosion products are unstable and corrosion control processes are required. Thermodynamic models can be used to predict when such interventions are required in new fields.

The protective corrosion products are unstable when oxygen is present. Therefore, oxygen contamination must be avoided.

Other factors such as condensate velocity and turbulence, temperature, pH range, CO_2 to H_2S ratio and chloride concentration have an important role in the stability of corrosion products and corrosion mechanisms, and subsequently on service life of assets and maintenance.

The development of predictive geochemistry models for heavy metal reduction process under low pH conditions is also a challenge.

New initiatives in scaling and corrosion control, alternatives to pH adjustment such as silica anti-scalants and the combination of pH adjustment plus corrosion inhibitors should be considered.

While carbon steels are typically the materials of choice for most new geothermal plants and their potential performance can be predicted based on the new field geochemistry and available database of materials testing for well-known geothermal fields, the material selection for new geothermal fields does at times require further materials-testing.

ACKNOWLEDGEMENTS

The authors express their thanks to Quest Integrity NZL Ltd. for support and permission to publish this work.

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