Modeling Corrosion and Corrosion Protection

By **ED FONTES** and **BERTIL NISTAD**

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INTRODUCTION

Almost every man-made structure involves the use of metals; often, nonnoble metals that are susceptive to corrosion. The estimated worldwide costs for corrosion-derived damages are about 2.5 trillion dollars. Proper corrosion protection could save 15–55% of the cost of these damages (Ref. 1). We can therefore safely claim that understanding corrosion and corrosion protection is financially well motivated.

Mathematical modeling and simulations are very efficient tools for understanding corrosion and corrosion protection. A validated high-fidelity model, based on the thermodynamics and kinetic properties of a system, can not only be used for understanding but also for making predictions, strengthening intuition, and fostering innovations.

This paper presents the basic theory behind the models that describe corrosion and corrosion protection. It also exemplifies how these models can be used for understanding, innovating, and optimizing devices and processes in order to mitigate corrosion.

THERMODYNAMICS, KINETICS, AND TRANSPORT PHENOMENA

The basis of modeling corrosion processes originates from the theory of heterogenous chemical reactions. Surface reactions where corrosion occurs involve a reduction and an oxidation reaction, where a metallic structure is in contact with an electrolyte. Oxidation and reduction occur on two different positions at the surface, called sites, and the transport of electrons from the oxidation site to the reduction site takes place through electronic conduction in the metallic structure. The circuit is closed through the electrochemical reactions and the transport of current through ionic conduction in the electrolyte.
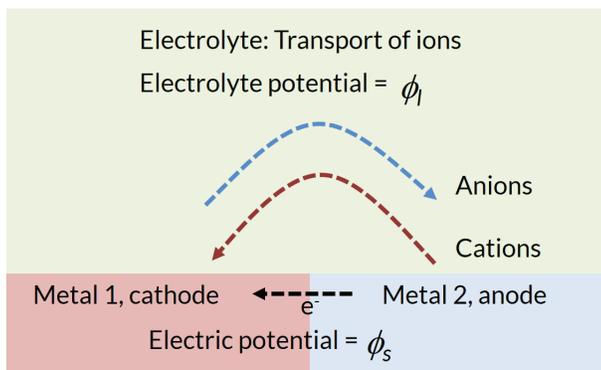
FIGURE 1. A galvanic cell where metal 1 acts as the cathode and metal 2 as the anode. The circuit is closed by the presence of an electrolyte, which covers both metal surfaces.

The oxidation reactions take place at the anodic site, which is the site to where the an-ions in the electrolyte migrate. The reduction reactions take place at the cathodic site, which is the site to where the cat-ions in the electrolyte migrate.

THERMODYNAMICS AND KINETICS

What determines whether the site acts as the anode or the cathode is the affinity to electrons of the metallic surfaces. The metal with a high affinity for electrons will act as a cathode and attract electrons, while the metal with a lower affinity will give away its electron and thus corrode. The affinity for electrons is determined by the Gibbs free energy of reaction (ΔG). Since electrons are involved, the electric potential affects the free energy. This is expressed through Faraday's law, which, in combination with Gibbs equations, yields the Nernst equation for electrochemical reactions (Ref. 2).

A common oxidation reaction (anodic) in corrosion processes is metal dissolution, while a common reduction reaction (cathodic) is hydrogen evolution.

$$Fe = Fe^{2+} + 2e^-$$

$$2H_2O + 2e^- = H_2 + 2OH^-$$
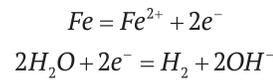
The rate of the anodic and cathodic reactions is determined by Arrhenius law, which states that the rate of a reaction depends exponentially on the activation energy. Again, since electrons are involved, the activation energy (ΔG$^{\neq}$) is affected by the electric potential. Combining Faraday's law and Arrhenius equations yields the Butler–Volmer equation for electrochemical reactions.
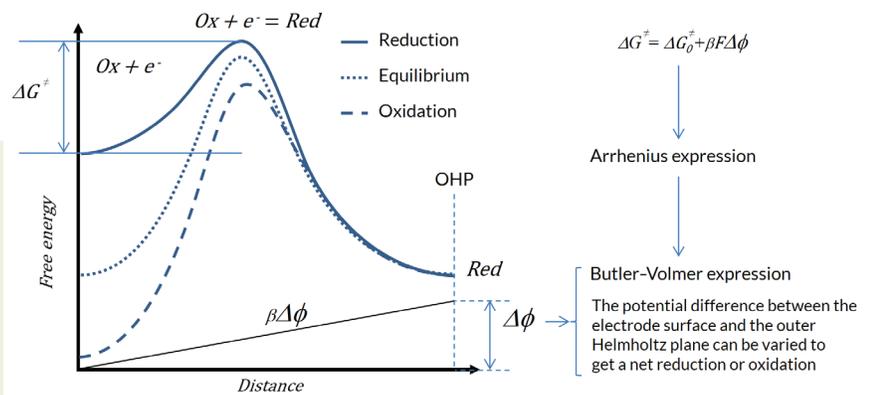
FIGURE 2. Depending on the electric potential at the metal surface, the surface can act as a cathode or as an anode. Connected to a more noble metal, a less noble metal surface acts as the anode. OHP denotes the outer Helmholtz plane of the charged double layer. Here, β denotes the symmetry factor, and F denotes the Faraday constant.

The Butler–Volmer equation gives the rate of an electrochemical reaction as a function of the electrochemical potential at the surface site. Using Faraday's law, this gives the current density, since electrons are involved. (Ref. 2). The Butler–Volmer equation may describe, for example, the hydrogen evolution reaction:

$$i_{H_2} = i_{0,H_2} \left\{ \left(C_{OH^-} \right)^2 P_{H_2} \exp\left(\frac{3F}{2RT} \eta \right) - \exp\left(-\frac{F}{2RT} \eta \right) \right\}$$

In this equation, i_{H_2} denotes the current density perpendicular to the metal surface due to the hydrogen evolution reaction, assuming a reversible reaction. The exchange current density, i_{0,H_2}, represents the size of the anodic and cathodic current densities at equilibrium and reflects the rate constant of the reaction. A high exchange current density corresponds to a reaction with fast kinetics. The dimensionless concentration of the hydroxide ion is expressed as follows:

$$C_{OH^-} = \frac{c_{OH^-}}{c_{OH_{eq}^-}}$$

and represents the hydroxide ion concentration at the metal surface divided by that concentration at equilibrium. The corresponding dimensionless hydrogen partial pressure is denoted:

$$P_{H_2} = \frac{p_{H_2}}{p_{H_2,eq}}$$

Faraday's constant is denoted F, R denotes the gas constant, T temperature, and η the reaction overpotential. The reaction overpotential is defined as follows:

$$\eta = \phi_s - \phi_l - \left(\phi_{s,eq} - \phi_{l,eq} \right)$$

where ϕ_s denotes the electric potential at the metal surface and ϕ_l the electrolyte potential just outside of the charged double layer (OHP above, with the exception of extremely diluted electrolytes). The overpotential is thus the difference between the electric potential of the metal surface and the electrolyte potential close to that surface compared to that same potential difference at equilibrium. It is virtually impossible to measure the potential difference ϕ_s-ϕ_l (disregarding equilibrium or not), since this implies sticking another metal surface (for example, a microelectrode) in the electrolyte, which in turn has another difference between the metal and the electrolyte. Instead, a reference electrode connected to a very high impedance can be used. Then, this potential difference is at least always measured to the same potential difference. In a model, if the same reference is used, the overpotential can be expressed as follows:

$$\eta = \phi_s - \phi_l - E_{eq}$$

where E_{eq} denotes the equilibrium electrode potential, measured with the same reference and at the same

positions at equilibrium and nonequilibrium. In a model, this also means that the electric potential ϕ_s must be set at one point; for example, at one position on one of the metals and in theory kept at the same value and position at equilibrium and nonequilibrium. The electric potential is usually set to zero at the position where the reference electrode is connected (in theory) to the more negative electrode so that $\phi_s = 0$ V. If we do not set the potential in one point, then the problem becomes underdetermined.

If we look at a metal surface consisting of two different metals in electronic contact and in contact with the same electrolyte, then the surface sites with a higher electron affinity yield a higher electrochemical potential at equilibrium, E_{eq}, than the surface sites with a lower electron affinity. Since the surface sites are in electronic contact and in contact with the same electrolyte, we get a galvanic cell with a mixed potential at a zero net current (also called corrosion potential, E_{corr}) according to the plot below for anodic and cathodic sites that are very close to each other. We can see here that the surface with the lower energy affinity acts as the anode, which by definition is a positive charge-transfer current density at the surface. The surface with higher electron affinity acts as the cathode, which by definition is a negative current density. The absolute value of the current density for the anodic and cathodic reactions at a net zero current density is called the corrosion current, i_{corr}.
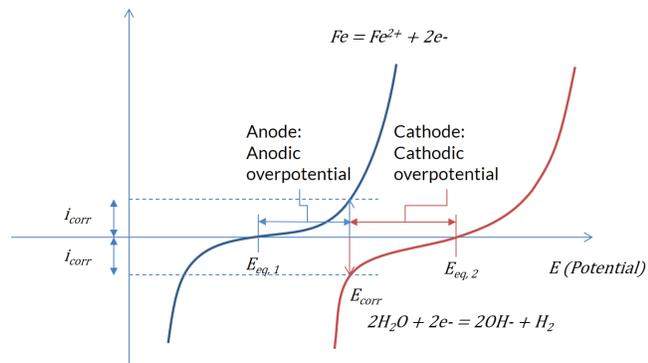
FIGURE 3. The Butler–Volmer expression for the reaction at the less noble metal surface (blue) and the more noble metal surface (red).

The electrode reactions can also be visualized in a so-called Evans diagram, where the electrochemical potential is plotted as a function of the logarithm of the absolute value of the current. Here, we can see the corrosion potential and corrosion current, again in the absence of mass transport resistance and neglecting ohmic losses.

(Potential)
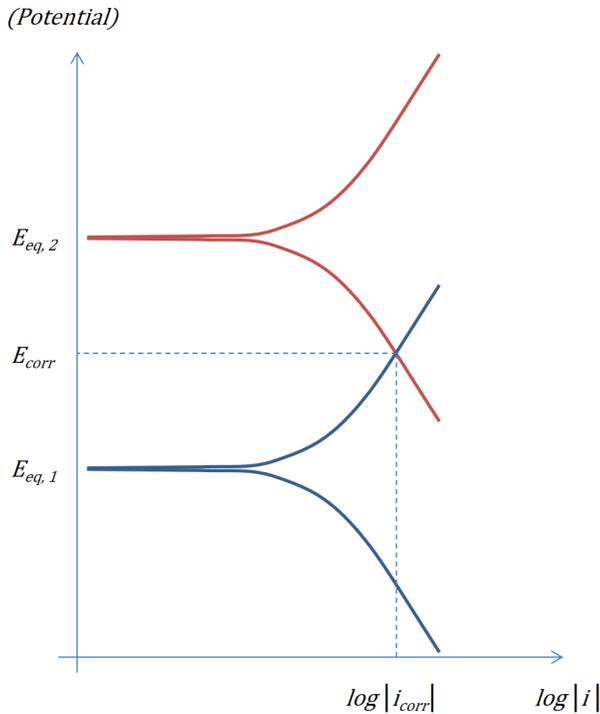
FIGURE 4. Evans diagram for two electrode reactions in the absence of ohmic and mass transport losses.

In the presence of mass transport limitations, the mass transport losses damp the exponential increase with overpotential. The current density at high values ceases to increase exponentially and may asymptotically approach a constant value referred to as the limiting current density. This is seen in the cathodic branch (negative) for oxygen reduction (red) in the figure below.

i (Current density)
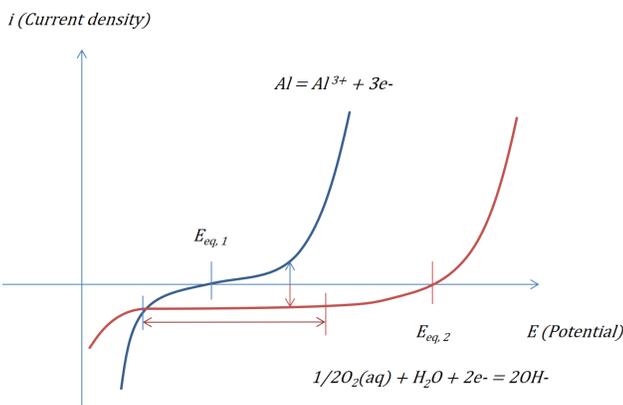
FIGURE 5. Oxygen reduction (red) is mass transport limited over a wide range of potential. The absolute negative current increases only at negative potential when hydrogen evolution starts.

The corresponding Evans diagram for transport-limited oxygen reduction and anodic metal dissolution is shown in Figure 6 (Ref. 3).

(Potential)

FIGURE 6. The Evans diagram for limiting current in the oxygen reduction reaction corresponding to the plot in Figure 5.

TRANSPORT PHENOMENA AND MODEL EQUATIONS

The electronic potential and the electrolyte potential may vary in space and time. In addition, the composition in the electrolyte may vary in space and time. A high-fidelity model of corrosion and corrosion protection must be able to describe the composition of the electrolyte as well as the potential distribution in the metal and the electrolyte. Such models introduce the flux of charged ions in the electrolyte, the material balance of all species in the electrolyte, and the electroneutrality condition in the electrolyte as model equations. The expressions for the electrochemical reactions discussed above are then introduced as boundary conditions at the metal surfaces.

The equations that describe the transport of charged ions in an electrolyte are the so-called Nernst–Planck equations. The flux of a charged ion, $\mathbf{N}_i$, consists of contributions from diffusion, migration, and advection. In a material balance, the flux variations in different directions are balanced by accumulation, which is the time-dependent term in the equations for below, where c_i denotes the concentration of species i, or by homogeneous reactions in the electrolyte, denoted R_i in the equations for the material balances.

The model equations in the electrolyte domain are as follows:

1. Material balances for 1 to n-1 species:

$$\frac{\partial c_i}{\partial t} + \nabla \cdot \mathbf{N}_i + R_i = 0$$

The flux is given by the Nernst–Planck equation:

$$\mathbf{N}_i = -D_i \nabla c_i - z_i u_{m,i} F c_i \nabla \phi_i + c_i \mathbf{u}$$

where D_i denotes the diffusion coefficient of species i, z_i the charge of an ion i, $u_{m,i}$ the mobility, and $\mathbf{u}$ the velocity vector that describes the flow of the electrolyte.

2. The equation for the balance of current density in the electrolyte, $\mathbf{i}_l$:

$$\nabla \cdot \mathbf{i}_l = 0$$

The current density in the electrolyte is given by the sum of the flux of all charges in the electrolyte, which, from the flux of ions and in combination with Faraday's law, gives:

$$\mathbf{i}_l = \sum_{i=1}^{n} z_i F \mathbf{N}_i$$

3. Poisson's equation:

$$\nabla \cdot \left(-\varepsilon \nabla \phi_l \right) = F \sum_{i=1}^{n} z_i c_i$$

where ε denotes permittivity. For most electrolytes (except for highly diluted electrolytes), this equation is approximated with the electroneutrality condition:

$$\sum_{i=1}^{n} z_i c_i = 0$$

In the electrolyte domain, we have n+1 unknowns: n species concentrations and the electrolyte potential ϕ_l. The material balance equations are n-1 in number. The equation for the balance of current density is the nth equation, which is a linear combination of the mass balances of all charged species. The electroneutrality condition gives the last equation (n+1) in the electrolyte. This gives the same number of equations as unknown variables (n+1).

For the electronic potential in the metal structure, we can use the equation for the conservation of current in combination with Ohm's law.

4. Balance of current density in the metal:

$$\nabla \cdot \mathbf{i}_s = 0$$

where

$$\mathbf{i}_s = -\kappa_s \nabla \phi_s$$

Here, κ_s denotes the electric conductivity of the metal.

The Butler–Volmer equation discussed above is used in the boundary conditions for the domain equations on both sides of the metal-electrolyte interface. For example, if an ion i participates in an electrochemical reaction at the surface, then its boundary condition is given by the fact that the flux at the boundary must match the reaction rate per unit area:

$$\mathbf{N}_i \cdot \mathbf{n} = -\frac{s_i}{nF} i_{BV}$$

where $\mathbf{n}$ denotes the normal vector to the metal surface, s_i the stoichiometric coefficient of species i in the charge transfer reaction, n the number of electrons, and i_{BV} is the Butler–Volmer expression for the electrochemical reaction that ion i is involved in. Note that this can be a sum of reactions. The corresponding boundary conditions for the balance of current density is the following:

$$\mathbf{i}_l \cdot \mathbf{n} = i_{BV}$$

Also, here, the Butler–Volmer expression, i_{BV}, can be a sum of expressions for several reactions. Analogously, the boundary condition for the balance of current density in the metal is obtained through the following expression:

$$\mathbf{i}_s \cdot \mathbf{n} = -i_{BV}$$

In a perfectly mixed electrolyte, we can neglect the concentration gradients everywhere except for a very thin microscopic boundary layer at the surface of the metal structure. The equation for the balance of current density becomes:

$$\nabla \cdot \mathbf{i}_l = 0$$

Here

$$\mathbf{i}_l = -\left(\sum_{i=1}^{n} z_i^2 u_{m,i} F^2 c_i \right) \nabla \phi_l + \left(F \sum_{i=1}^{n} z_i c_i \right) \mathbf{u}$$

However, the second term on the right-hand side of the equation above contains the electroneutrality condition as a factor, which is equal to zero. The factor within parentheses in the first term on the right-hand side of the equation above is equal to the electrolyte conductivity, κ_l. This yields the following expression for the current density in perfectly mixed electrolytes:

$$\mathbf{i}_l = -\kappa_l \nabla \phi_l$$

GALVANIC CORROSION

We have already introduced galvanic corrosion in our discussion above: Two metals with different electron affinities that are in electronic contact and in contact with the same electrolyte form a galvanic cell. This is a problem when welding different metals in the automotive industry and in shipbuilding. Close to the contact area between the two metals, the less noble metal is dissolved anodically in the galvanic process. Such structures have to be protected, for example, by using coatings that impede the direct contact of the metal surfaces with water containing ions (the electrolyte).

If we assume a perfectly mixed electrolyte, we can use the Butler–Volmer equations for hydrogen evolution and metal dissolution at the metal-electrolyte interface and set the electronic potential in one point in the metal. The figure below shows the resulting current density streamlines and the isopotential curves. In addition, the problem is solved on a moving mesh, where the dissolution of the anode is taken into account. The plot below shows how the initially flat surface of the less noble metal is consumed through anodic dissolution after being exposed to the electrolyte and in contact with the more noble metal for 72 h.
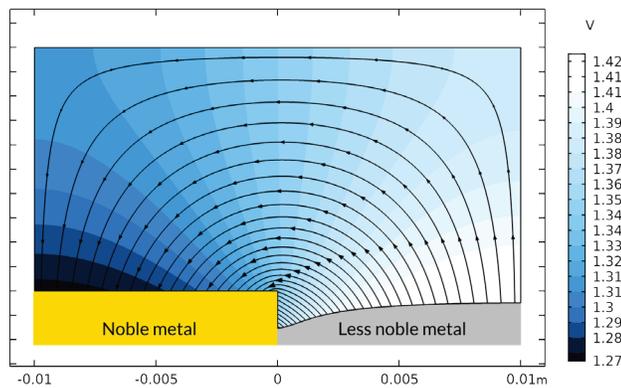
FIGURE 7. Streamlines of the current density and isopotential curves in the electrolyte in a galvanic cell. The initial surface of the noble metal and the less noble metal is horizontal at the initial state. After 72 h, the less noble metal is corroded through anodic dissolution.

The use of copper in the electronics industry together with other less noble metals (for example, soldering metals) is an example of galvanic corrosion in humid environments. When water condenses on unprotected surfaces, and ions are slowly dissolved to form an electrolyte, then galvanic cells are easily formed. The copper structure acts as a cathodic surface dissolving the less noble soldering metal, causing failure of the electronic device.

CREVICE CORROSION

In the discussion above, we have looked at galvanic corrosion due to differences in electron affinity in different metals. However, corrosion can also occur due to variations in electrolyte composition. For example, in crevice corrosion, the mouth of the crevice has a higher electrochemical potential, due to the higher oxygen activity, than the bottom of the crevice.

This process starts with small variations in composition at the metal surface that cause some parts to act as anodes and some parts to act as cathodes; i.e., general corrosion, discussed below. The cathodic oxygen reduction reaction consumes oxygen over the whole crevice, but eventually, this leads to oxygen depletion at the bottom of the crevice where the oxygen transport resistance is large.

At this stage, the corrosion process accelerates, since the bottom of the crevice gets a low electrochemical potential due to the low oxygen activity compared to the mouth. When this occurs, the bottom of the crevice is strongly anodically polarized, and anodic metal dissolution takes place, while the mouth of the crevice works as the cathode, with oxygen reduction as the cathodic reaction.

The plot below shows the concentration distribution of corrosion products and other ions in a crevice where the corrosion of iron takes place in an acetic acid/sodium acetate solution. The solution to the model equations reproduces Walton's findings (Ref. 4). The plot shows that the sodium concentration is significantly lower in the crevice, compared to the bulk (to the right in the plot), whereas iron ions and iron complexes have higher concentrations toward the tip (to the left in the plot).
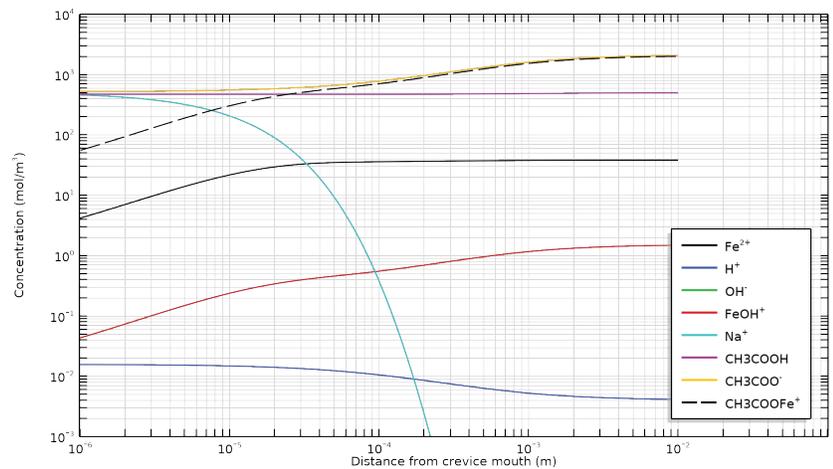
FIGURE 8. Concentration of ion and ion complexes along the depth of a crevice.

PITTING CORROSION

Droplets on the surface of a metal may cause a concentration cell similar to crevice corrosion. Uniform corrosion consumes oxygen at the surface of the metal covered by the liquid droplet. This causes oxygen depletion in the middle of the droplet, while the thinner film at the edges of the droplet have a lower oxygen transport resistance. The edges of the droplet then start acting as a cathode, while the surface underneath the middle of the droplet acts as an anode.

This may be the start of pitting corrosion. A small pit may form in the middle of the droplet. Once this pit is formed, humidity remains when the droplet on the surface has dried. The corrosion process then becomes very similar to crevice corrosion. The mouth of the pit acts as the cathode, due to the access to oxygen, while the bottom of the pit works as the anode, dissolving the metal and worsening the corrosion process.
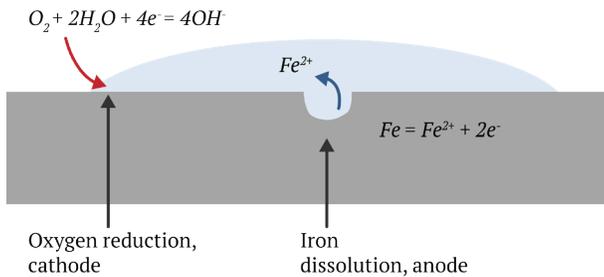
FIGURE 9. Corrosion may start due to uniform oxygen reduction and iron dissolution that eventually causes an oxygen gradient at the surface. This eventually gives a cathodic surface and an anodic surface; i.e., the reactions are no longer uniformly distributed.

STRESS CORROSION CRACKING

This type of corrosion is similar to pitting corrosion. However, stress corrosion cracking is initialized by a mechanical formation of cracks. These crack often repassivate; i.e., a protecting oxide film is quickly formed. If repassivation does not occur, then a similar process to pitting corrosion begins, with the mouth of the crack acting as the cathode and the tip of the crack acting as the cathode.
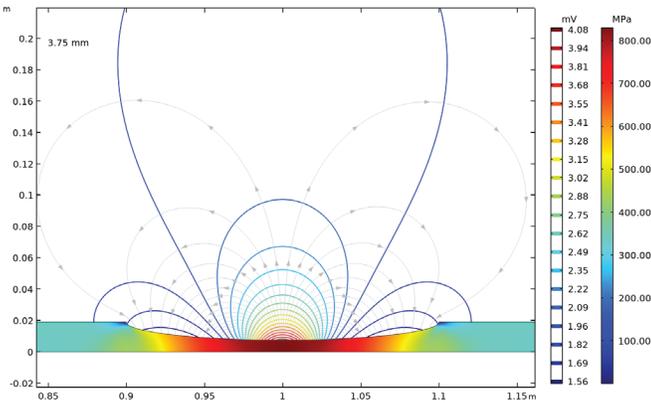
FIGURE 10. Stresses in a metal plate in contact with an electrolyte. The thinner region of the plate with the highest stresses acts as the anode.

The figure above shows stresses in a metal surface where the kinetic expression accounts for the formation of microcracks causing stress corrosion cracking. The model couples corrosion assuming a perfectly mixed electrolyte with a solid mechanics simulation of the metal. The middle of the metal plate, subjected to the highest stresses, acts as the anode and starts to corrode as the stresses increase.

GENERAL UNIFORM CORROSION

A metal surface consists of grains and grain boundaries of varying composition. This implies that different grains and grain boundaries may have different affinities to electrons. This leads to the formation of microscopic galvanic cells when the metal surface is covered with an electrolyte. This type of corrosion is localized at the microscale, but to the naked eye, it looks like a uniform corrosion of the surface. The figure below shows how crystallites of a less noble metal are dissolved by crystallites of a more noble one. The original surface is completely flat while, after 60 hours of contact with an electrolyte, the corrosion process makes the surface more rugged.
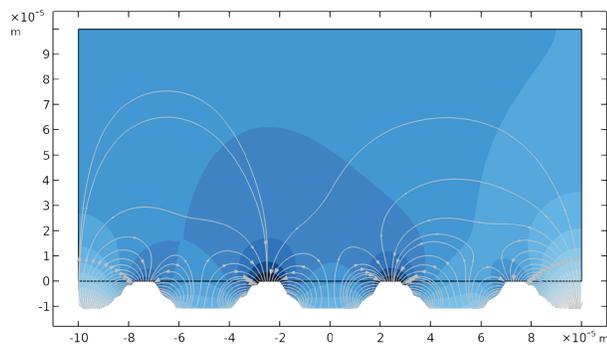
FIGURE 11. Results of a corrosion simulation using moving mesh. The initially flat surface starts to become slightly rugged after 60 h in contact with an electrolyte.
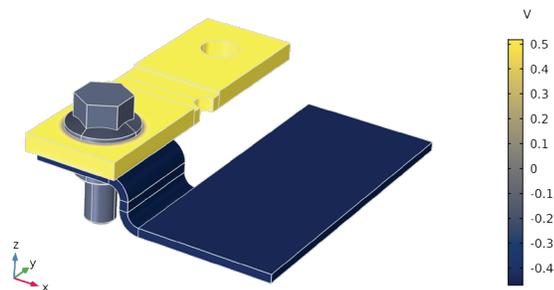
FIGURE 12. Electrode potential on the surface of a busbar assembly subjected to humid air. The structure is covered every now and then with a thin layer of condensed electrolyte. The simulation yields the corrosion rate of the different metals: copper (upper busbar), zinc (bolt), and aluminum (lower busbar).

Atmospheric corrosion is one example of uniform corrosion. It is a large problem in humid environments, since the metal surface is often covered by a liquid film. Metal structures in coastal areas are also subjected to mist containing chloride ions, which accelerate corrosion. In general, atmospheric pollution may also worsen the effects of atmospheric corrosion.

ANODIC PROTECTION

Oxide films formed by a highly controlled oxidation of a metal surface can protect the metal from further corrosion. Anodic protection sounds almost like a contradiction, since a metal surface working as an anode usually dissolves. In the case of anodic protection, however, the oxide film is stabilized by the anodic potential and only a very small corrosion current density is obtained. This very small current density actually contributes to the stabilization of the protecting oxide film. Anodic protection is not a very common method, but it is used for extremely acidic environments, such as stainless steel tanks containing phosphoric acid and titanium tanks containing sulphuric acid.

The modeling of anodic protection makes it possible to study the potential distribution on the surface of the metal that is to be protected. The current density at the anode can be kept constant at the passivation current (boundary condition). The potential must be high enough to be on the passivation region, but the variations should be small enough to avoid the reactivation that occurs at potentials above the passivation window. The model equations are very similar to those for cathodic protection, with passivation being treated analogous to oxygen reduction in cathodic protection.
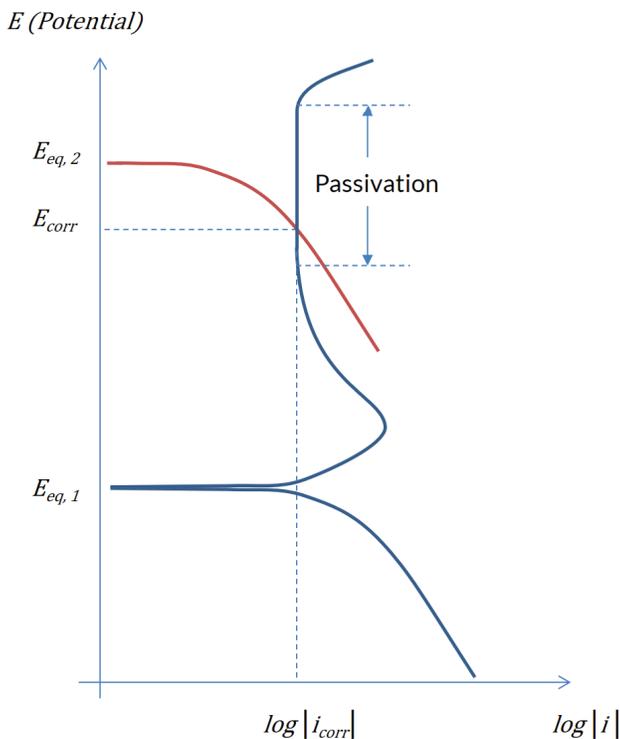
E (Potential)

FIGURE 13. In anodic protection, the potential must be kept within the passivation window.

CATHODIC PROTECTION

The most common method of actively protecting a metal surface is by polarizing it cathodically. The potential of the surface is lowered to such an extent that oxygen reduction is the only reaction (or, at least, the main reaction) that occurs on the cathodic surface. Oxygen reduction gives an alkaline environment at the protected surface and a calcareous layer may be formed. This layer additionally protects the surface and results in an even lower protecting current being required.

In principle, there are two different processes for cathodic protection: cathodic protection with sacrificial anodes and impressed cathodic current protection (ICCP).

The principle for cathodic protection is shown in Figure 5. A galvanic cell is formed on purpose, where the anodic reaction dissolves, for example, aluminum anodes in a controlled way. As long as there are aluminum anodes left, the cathodic polarization is sustained on the metal surface to be protected.
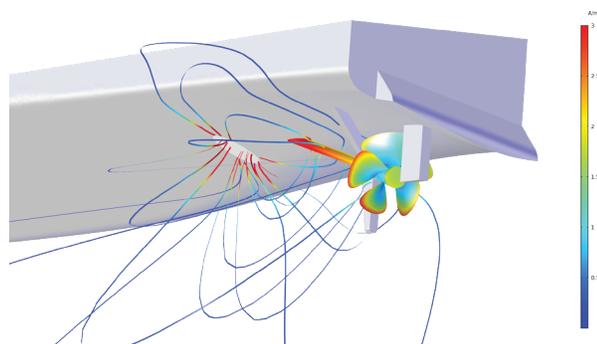
FIGURE 14. Streamlines in seawater around a sacrificial anode with impressed current that protects the propeller in a large cargo vessel. The inner parts of the impeller blades are slightly less protected than the rest of the structure.

The modeling of cathodic protection is done with the purpose of positioning sacrificial anodes in such a way that the whole metal surface to be protected is within the potential window for oxygen reduction. The boundary condition is a constant current density at the cathode surface. The resulting potential distribution should be well within the oxygen reduction window. If not, then the sacrificial anodes must be repositioned. The risk of large variations in the electrochemical potential is twofold: The surface can be excessively cathodically polarized with undesired hydrogen evolution as a result, or the surface may be unprotected with accelerated corrosion as a result. Hydrogen evolution may cause hydrogen embrittlement of, for example, steel structures.

When an object is submerged in seawater, a rather complex reaction mechanism causes calcareous deposits. These deposits act as a protective film, reducing the

oxygen transport toward the bare metal surface, and thus the current demand for the cathodic reactions. By accounting for this, a more accurate model for the lifetime of the structure can be obtained. A rigorous model of the full reaction mechanism would be too complex for a full-scale subsea structure, but by fitting measurements to a simpler model for the precipitation of calcareous deposits, one can obtain quick and realistic results.
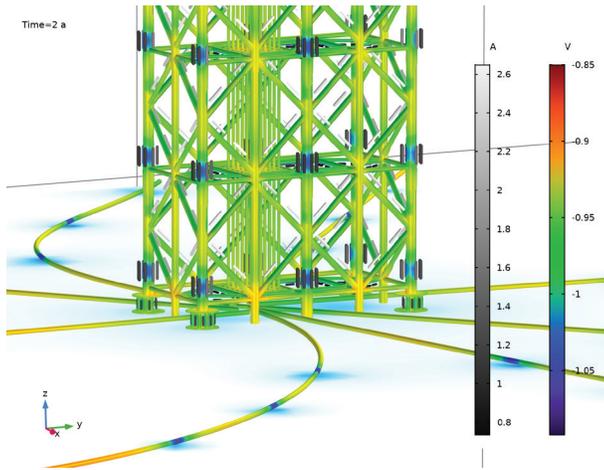
FIGURE 15. Potential distribution for an oil platform jacket structure and current output from the sacrificial anodes, two years after installation.
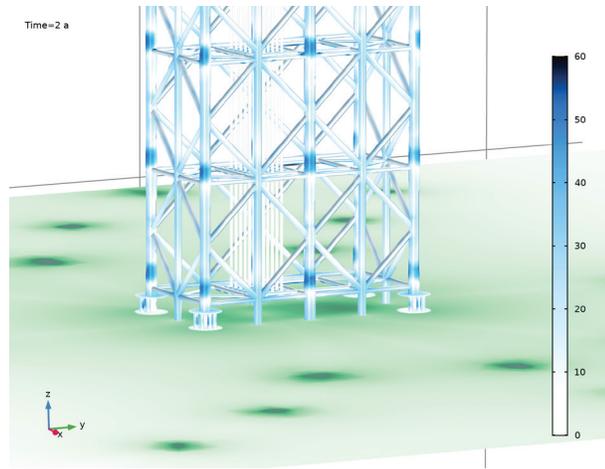
FIGURE 16. Deposited species in dimensionless units two years after installation of jacket structure. The deposited species affect the current distribution due to a reduced oxygen diffusion through the layer which reduces the amount of current passing through the surface

Impressed cathodic current protection is similar to the use of sacrificial anodes for cathodic protection. However, as an alternative to dissolving anodes, dimensionally stable anodes (DSA) may also be used. In addition, an external current is applied, and the anodes can be placed further away from the metal surface than they can without impressed current. This also implies that the electronic connection between the anodes and the cathode is
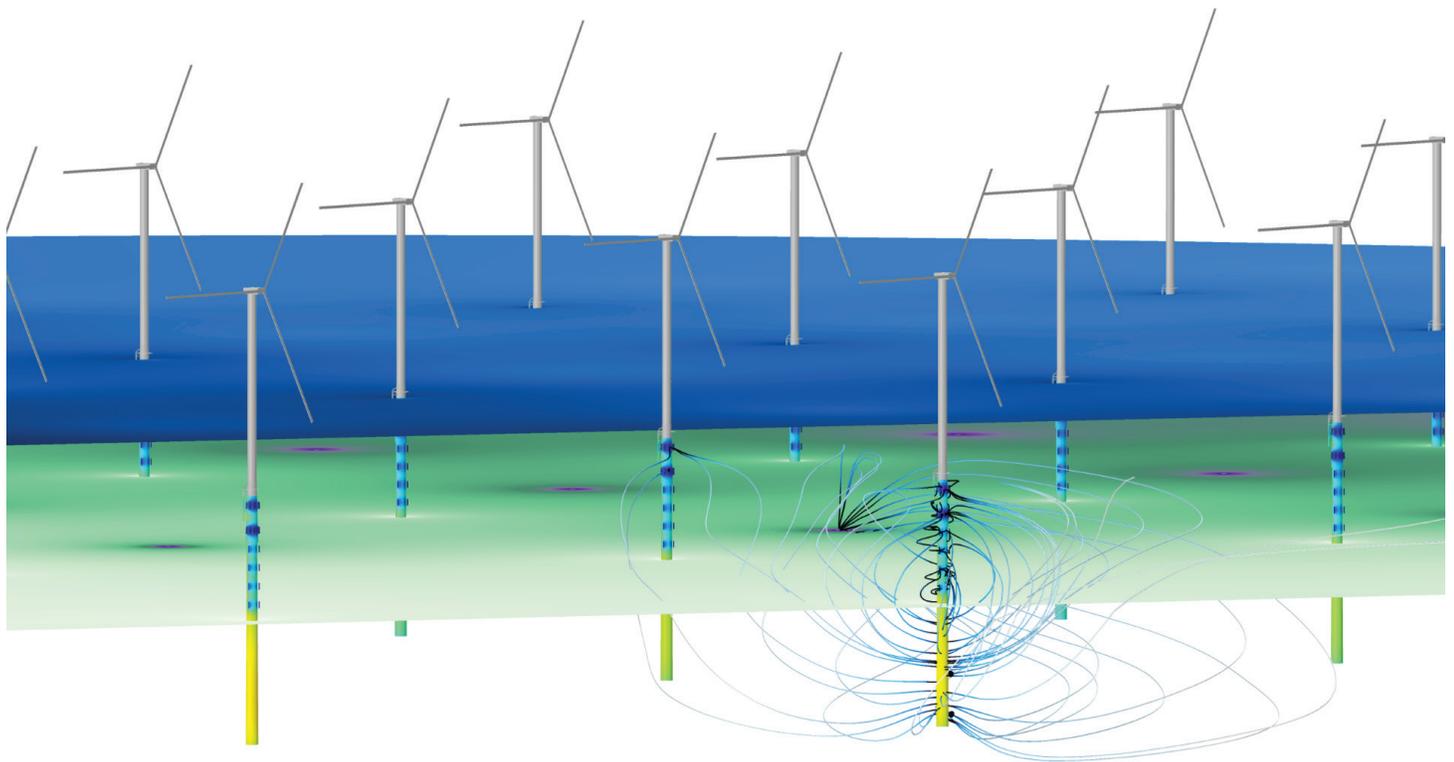
FIGURE 17. An offshore wind farm with parts of the structure below the sea surface is protected from corrosion using ICCP. The simulation is done with the purpose of positioning the anodes in order to protect the structure.

obtained through external cables that are connected to a power supply for delivering the desired direct current.

The modeling of ICCP is very similar to the modeling of systems without external currents. The only difference is that the boundary conditions for the electric potential are different. A system without external currents is short-circuited, which means that the potential at the contact point between the two metals is identical. In ICCP, a difference in potential is supplied by the external power unit. Also, in this case, the positioning of the anodes has to be done in such a way that the cathodic potential is within the oxygen reduction window. However, the freedom in the positioning of the anodes is larger. In addition, other effects, such as power consumption and overprotection, may become more relevant.

ICCP also introduces problems with bipolar effects. For example, a boat that is anchored close to a protected structure over a longer time may act as a bipolar electrode and thus corrode and interfere with the ICCP system.
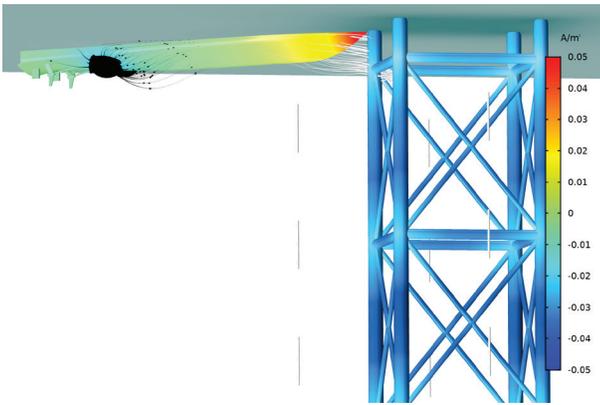
FIGURE 18. A boat anchored close to an oil rig is subjected to the currents from the cylindrical anodes that protect the rig structure from corrosion. The boat acts as a bipolar electrode, where parts of the hull close to the oil rig may corrode anodically. The black streamlines show the output from the ICCP anodes, and the gray streamlines show the direction of current toward the jacket structure.

The freedom of operation and the flexibility of ICCP also introduce some difficulties in that other metallic structures in the vicinity of the protected system may take up some of the current, causing so-called stray current effects.

CONCLUDING REMARKS

It is clear that modeling and simulation using high-fidelity 1D, 2D, and 3D models can contribute to the understanding of corrosion and corrosion protection processes. In addition, this understanding can be used to design devices and protection processes to mitigate corrosion. The benefit of modeling and simulation is that understanding, design, and optimization can be done at a lower cost, which also lowers the risk of expensive redesigns and reparations, compared to purely empirical designs or designs based on simplified lumped models.

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