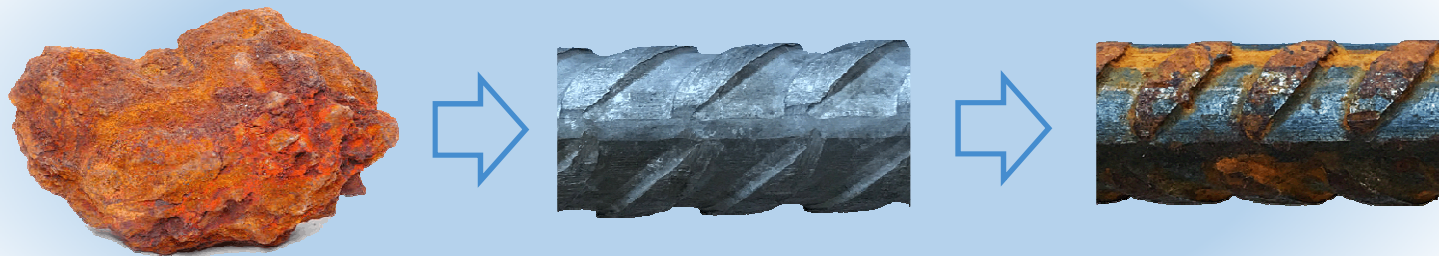


WORKSHOP ON CORROSION OF STEEL IN CONCRETE

17-18 September 2018, University of Aveiro, Portugal

CORROSION OF STEEL



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ciceco
aveiro institute of materials



Long Lasting Reinforced Concrete for Energy Infrastructure under Severe Operating Conditions

Coordinator: SINTEF

Objectives

Introduce (in 40 minutes) aspects of metallic corrosion that will be appearing recurrently through the Workshop.

Provide a refresh of concepts and bring the language to a common ground.

Overview of iron and steel corrosion but no focus on concrete, which will be highlighted in the next session.

Keep a phenomenological description. Experimental methods and results will be presented in a subsequent communication.

Contents

Definition of corrosion.

Why metals corrode?

Thermodynamics.

Corrosion reactions.

Mechanism and kinetics.

Passivity.

Critical environment factors (pH, O₂, humidity, ...)

Forms of corrosion.

Corrosion control.

Coatings and paints.

Corrosion inhibitors.

Cathodic protection.

Conclusion.

Definition of Corrosion

International standard definition of corrosion:

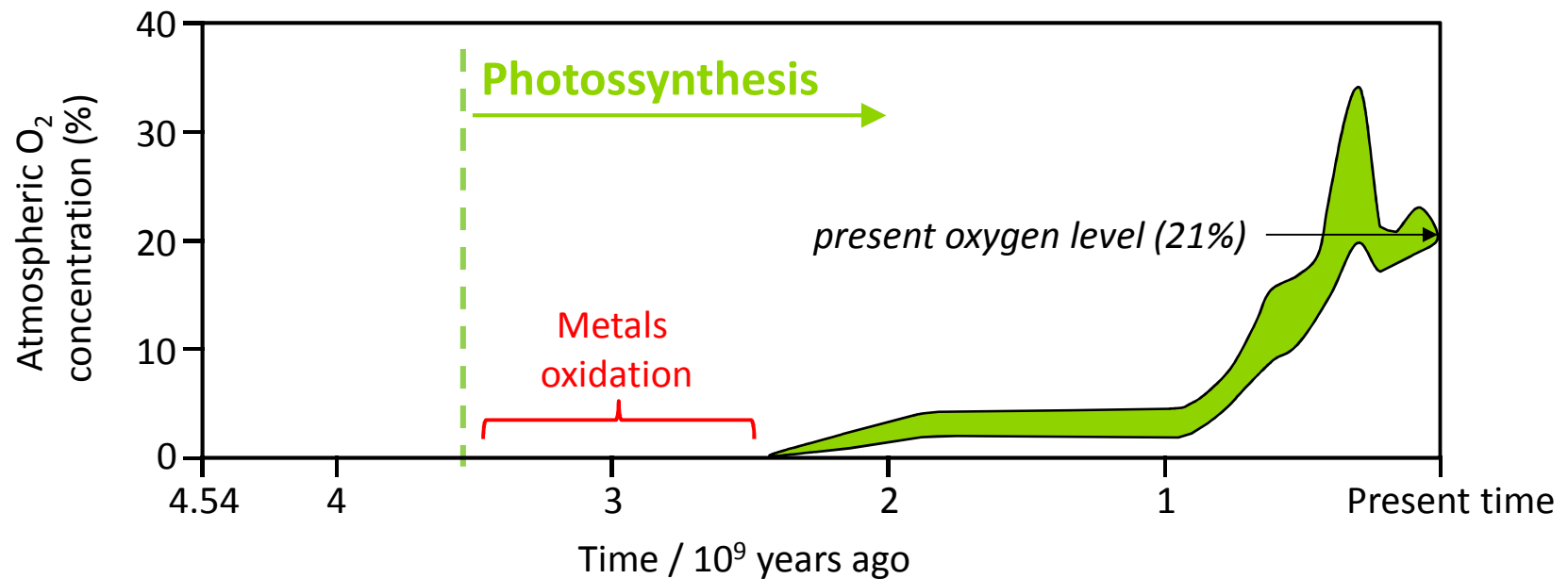
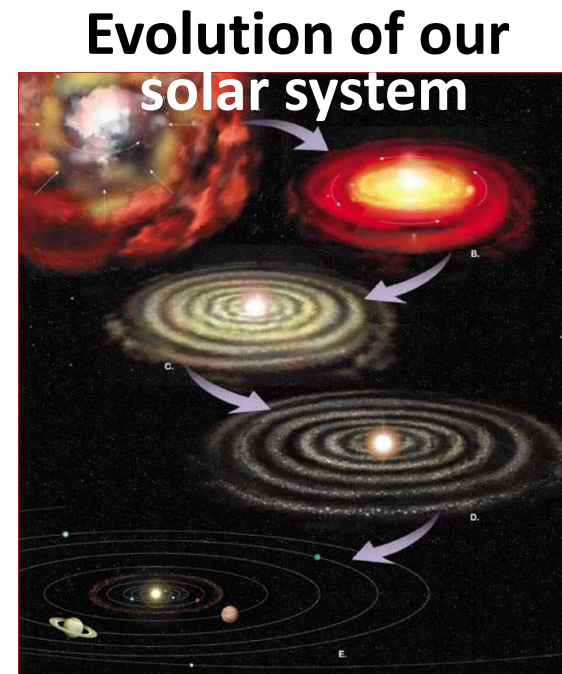
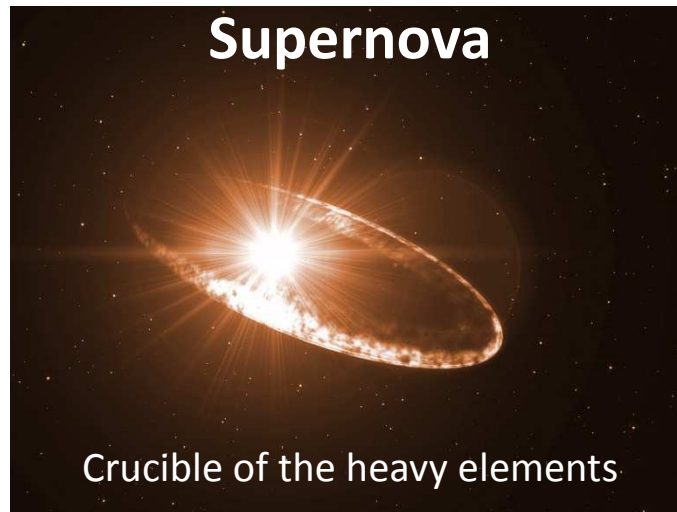
"Physicochemical interaction between a **metal** and its environment which results in changes in the properties of the metal and which may often lead to impairment of the function of the metal, the environment, or the technical system of which these form a part". **ISO 8044 -1986 - Corrosion of metals and alloys - Terms and definitions.**

International Union of Pure and Applied Chemistry (IUPAC) :

"Corrosion is an irreversible interfacial reaction of a **material** (metal, ceramic, polymer) with its environment which results in consumption of the material or in dissolution into the material of a component of the environment. Often, but not necessarily, corrosion results in effects detrimental to the usage of the material considered. Exclusively physical or mechanical processes such as melting or evaporation, abrasion or mechanical fracture are not included in the term corrosion". **Pure & Appl. Chem. 61 (1989) 19.**

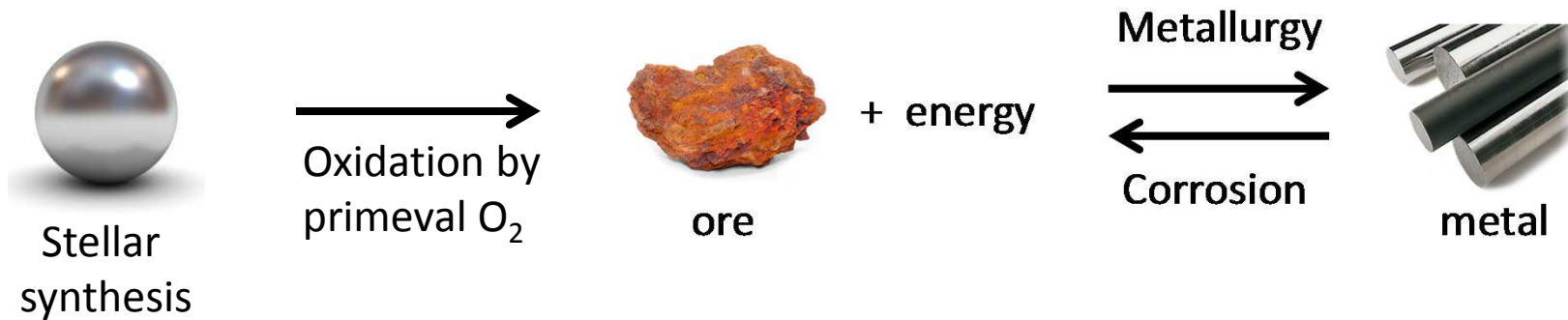
A decorative vertical bar on the left side of the slide, consisting of a thick blue line and a thinner grey line to its right.

Why metals corrode?

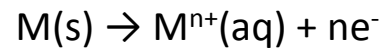


H.D. Holland, The oxygenation of the atmosphere and oceans, *Phil. Trans. R. Soc. B* 361 (2006) 903.

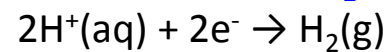
Why metals corrode?



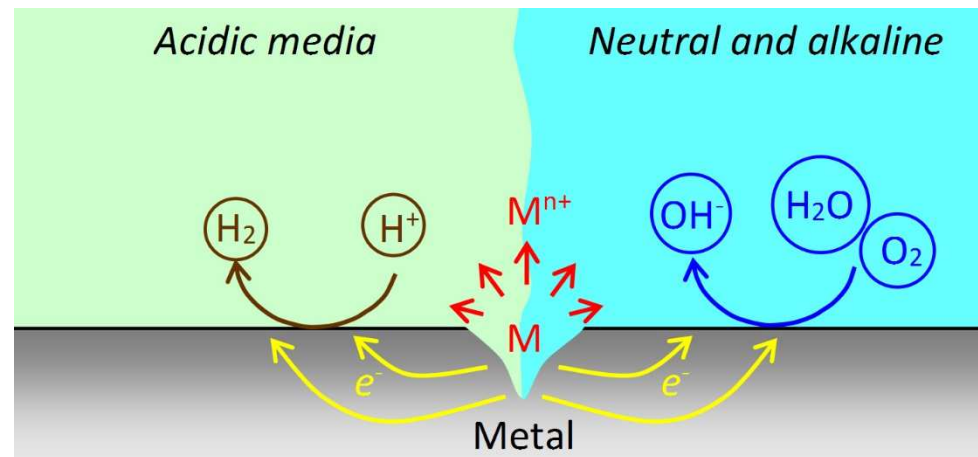
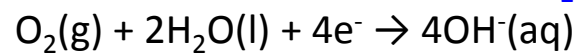
Metal oxidation



Reduction of H₂O



Reduction of dissolved O₂



Energy necessary
to produce
the metal



Na
Mg
Al
Zn
Fe
Ni
Sn
Cu
Ag
Pt
Au

Tendency
to oxidize



TABLE 2.1 Standard Electromotive Force Potentials (Reduction Potentials)

| | Reaction | Standard Potential, e° (volts vs. SHE) |
|--------|--|---|
| Noble | $\text{Au}^{3+} + 3e^- = \text{Au}$ | +1.498 |
| | $\text{Cl}_2 + 2e^- = 2\text{Cl}^-$ | +1.358 |
| | $\text{O}_2 + 4\text{H}^+ + 4e^- = 2\text{H}_2\text{O}$ (pH 0) | +1.229 |
| | $\text{Pt}^{2+} + 3e^- = \text{Pt}$ | +1.118 |
| | $\text{NO}_3^- + 4\text{H}^+ + 3e^- = \text{NO} + 2\text{H}_2\text{O}$ | +0.957 |
| | $\text{O}_2 + 2\text{H}_2\text{O} + 4e^- = 4\text{OH}^-$ (pH 7) ^a | +0.82 |
| | $\text{Ag}^+ + e^- = \text{Ag}$ | +0.799 |
| | $\text{Hg}_2^{2+} + 2e^- = 2\text{Hg}$ | +0.799 |
| | $\text{Fe}^{3+} + e^- = \text{Fe}^{2+}$ | +0.771 |
| | $\text{O}_2 + 2\text{H}_2\text{O} + 4e^- = 4\text{OH}^-$ (pH 14) | +0.401 |
| | $\text{Cu}^{2+} + 2e^- = \text{Cu}$ | +0.342 |
| | $\text{Sn}^{4+} + 2e^- = \text{Sn}^{2+}$ | +0.15 |
| | $2\text{H}^+ + 2e^- = \text{H}_2$ | 0.000 |
| | $\text{Pb}^{2+} + 2e^- = \text{Pb}$ | -0.126 |
| | $\text{Sn}^{2+} + 2e^- = \text{Sn}$ | -0.138 |
| | $\text{Ni}^{2+} + 2e^- = \text{Ni}$ | -0.250 |
| | $\text{Co}^{2+} + 2e^- = \text{Co}$ | -0.277 |
| | $\text{Cd}^{2+} + 2e^- = \text{Cd}$ | -0.403 |
| | $2\text{H}_2\text{O} + 2e^- = \text{H}_2 + 2\text{OH}^-$ (pH 7) ^a | -0.413 |
| | $\text{Fe}^{2+} + 2e^- = \text{Fe}$ | -0.447 |
| Active | $\text{Cr}^{3+} + 3e^- = \text{Cr}$ | -0.744 |
| | $\text{Zn}^{2+} + 2e^- = \text{Zn}$ | -0.762 |
| | $2\text{H}_2\text{O} + 2e^- = \text{H}_2 + 2\text{OH}^-$ (pH 14) | -0.828 |
| | $\text{Al}^{3+} + 3e^- = \text{Al}$ | -1.662 |
| | $\text{Mg}^{2+} + 2e^- = \text{Mg}$ | -2.372 |
| | $\text{Na}^+ + e^- = \text{Na}$ | -2.71 |
| | $\text{K}^+ + e^- = \text{K}$ | -2.931 |

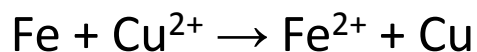
^aNot a standard state but included for reference.

Source: Handbook of Chemistry and Physics, 71st ed., CRC Press, 1991.

Thermodynamics

Copper and iron. Tendency to oxidize?

Iron nail in copper sulphate solution.



$\Delta G < 0$ spontaneous

$$\Delta G = -nFE$$



Cu has more tendency to be reduced than Fe
Cu will be reduced, Fe will oxidize.

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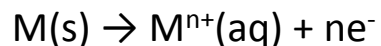
Source: Handbook of Chemistry and Physics, 71st ed., CRC Press, 1991.

Usually there are no metallic cations in the environment to oxidize the metal...

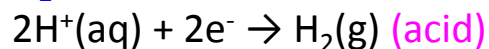
But,
there is water and oxygen.

Most important reactions in corrosion

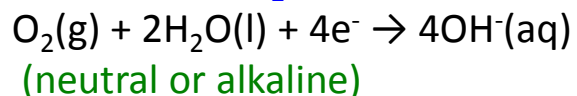
Metal oxidation



H₂O reduction



Reduction of O₂ dissolved in water



Fe is oxidised by H₂O and O₂

Cu is oxidised by O₂
(acids do not corrode copper)

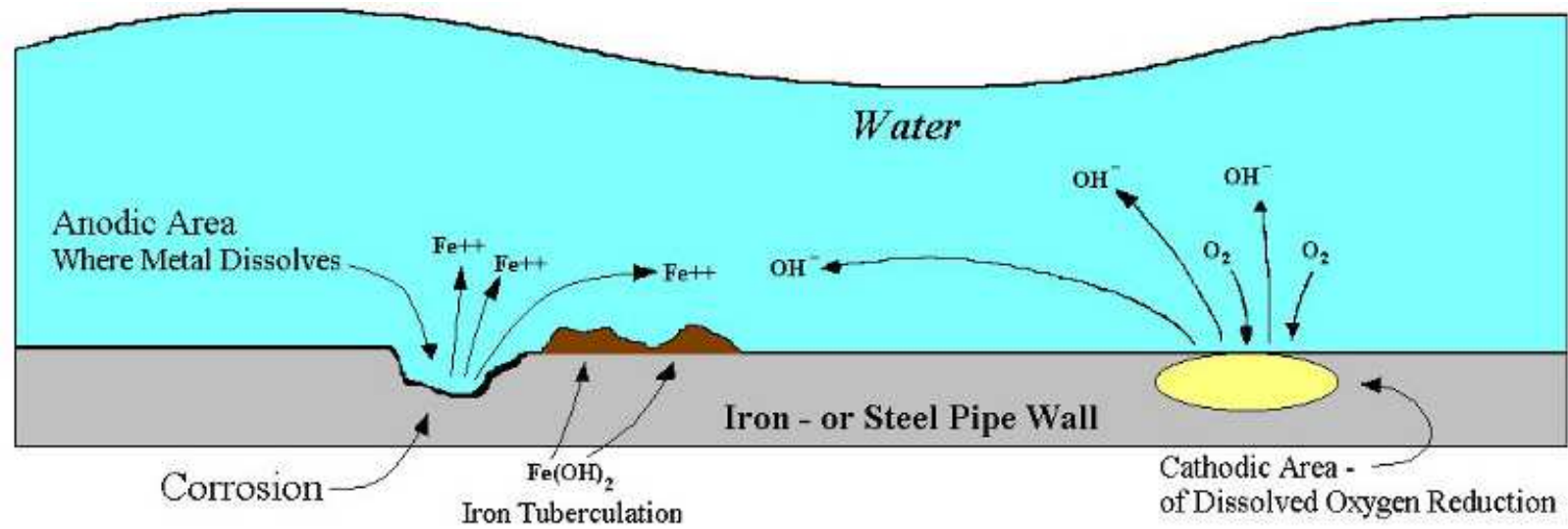
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| | $O_2 + 4H^{+} + 4e^{-} = 2H_2O \text{ (pH 0)}$ | +1.229 |
| | $Pt^{2+} + 3e^{-} = Pt$ | +1.118 |
| O ₂ | $NO_3^{-} + 4H^{+} + 3e^{-} = NO + 2H_2O$ | +0.957 |
| | $O_2 + 2H_2O + 4e^{-} = 4OH^{-} \text{ (pH 7)}^a$ | +0.82 |
| | $Ag^{+} + e^{-} = Ag$ | +0.799 |
| | $Hg_2^{2+} + 2e^{-} = 2Hg$ | +0.799 |
| Cu | $Fe^{3+} + e^{-} = Fe^{2+}$ | +0.771 |
| | $O_2 + 2H_2O + 4e^{-} = 4OH^{-} \text{ (pH 14)}$ | +0.401 |
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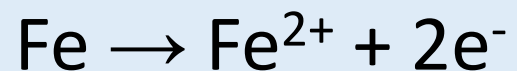
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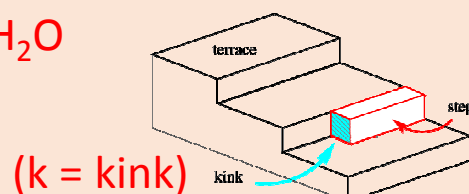
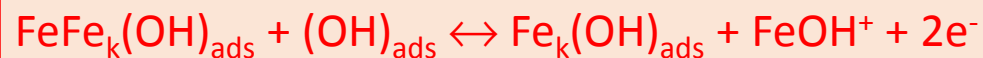
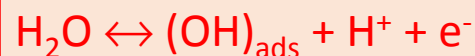
Corrosion cell



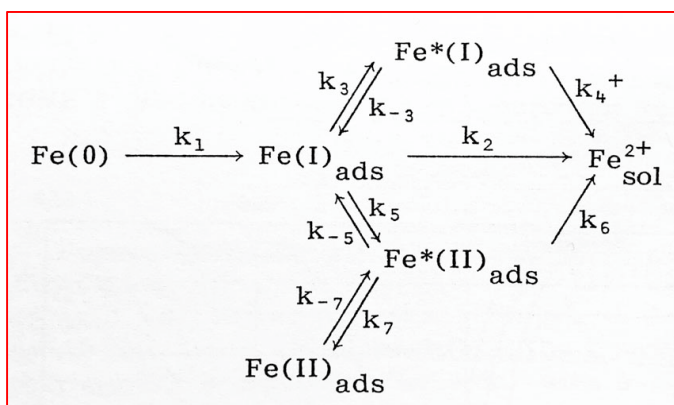
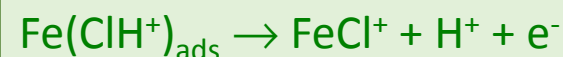
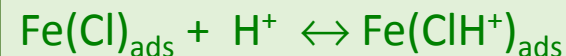
Mechanisms of Fe oxidation



Acid



Acid with chloride

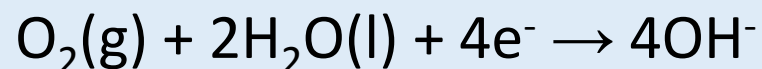


Alkaline



Mechanisms of reduction reactions

Oxygen reduction

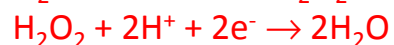


Acid solution

direct reduction

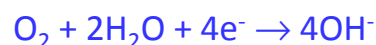


indirect reduction

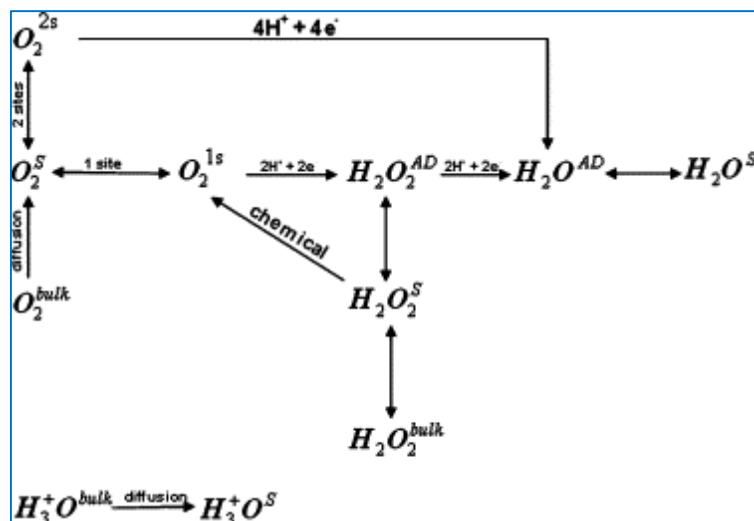
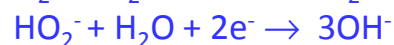


Alkaline solution

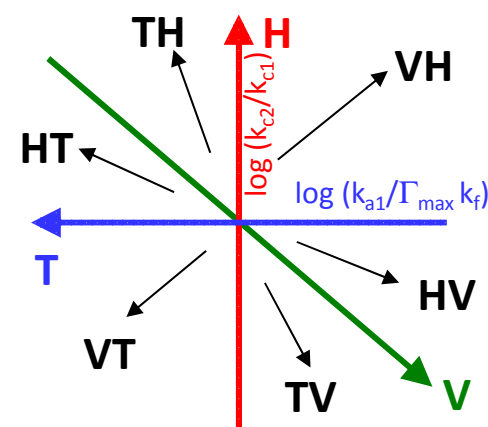
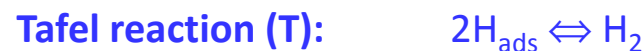
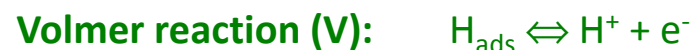
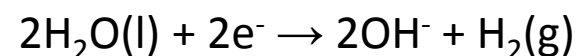
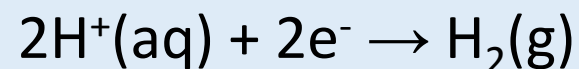
direct reduction



indirect reduction

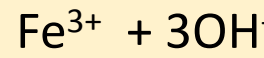
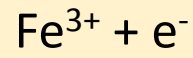
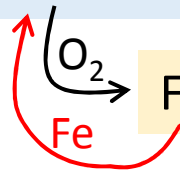
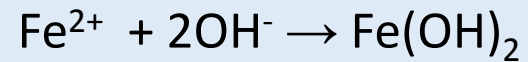
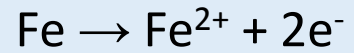
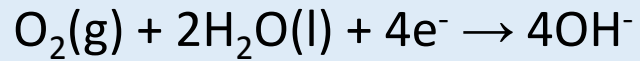


Water reduction

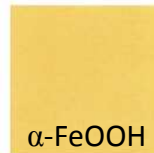


Newman, T.-Alyea, *Electrochemical Systems* 3rd Ed,
Wiley, 2004, p. 224

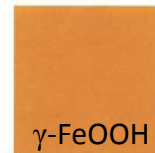
Corrosion products of iron



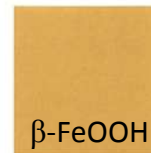
Goethite



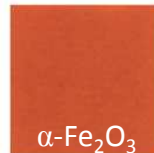
Lepidocrocite



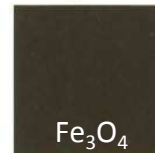
Akaganéite



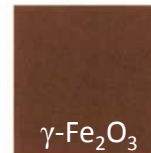
Haematite



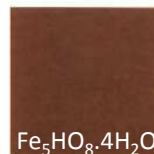
Magnetite



Maghemite



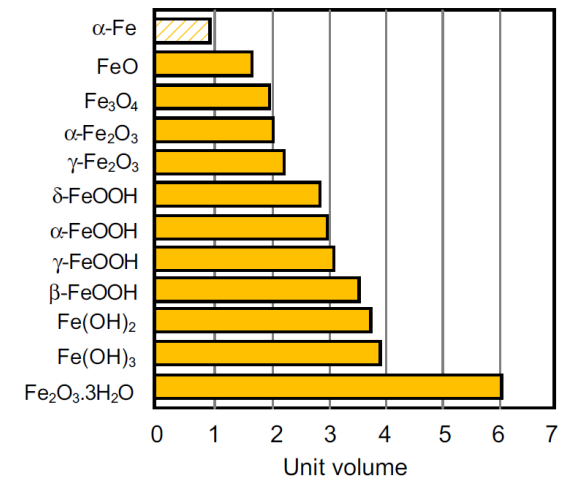
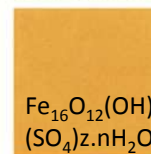
Ferrihydrite



Feroxyhyte

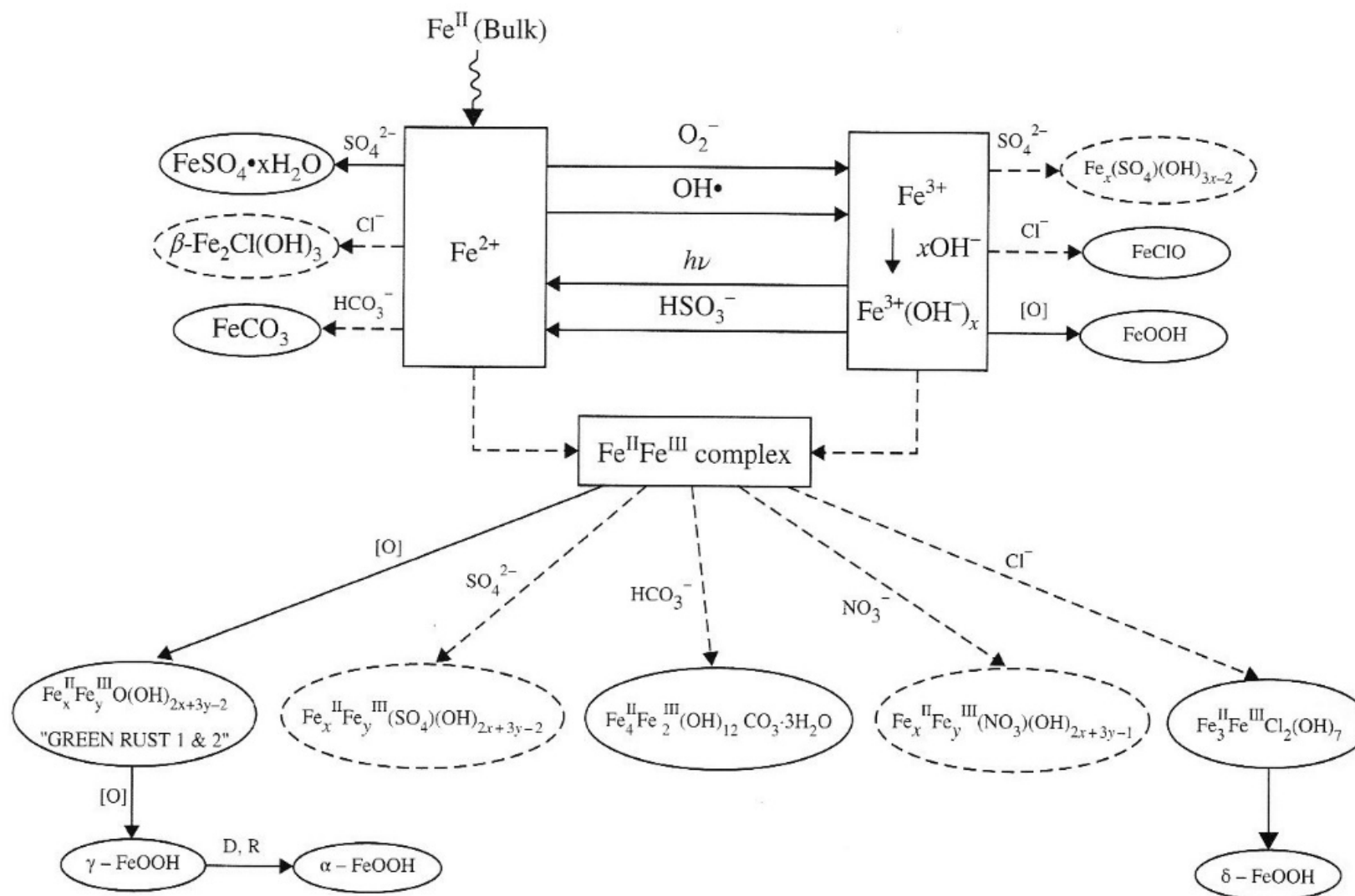


Schwertmannite

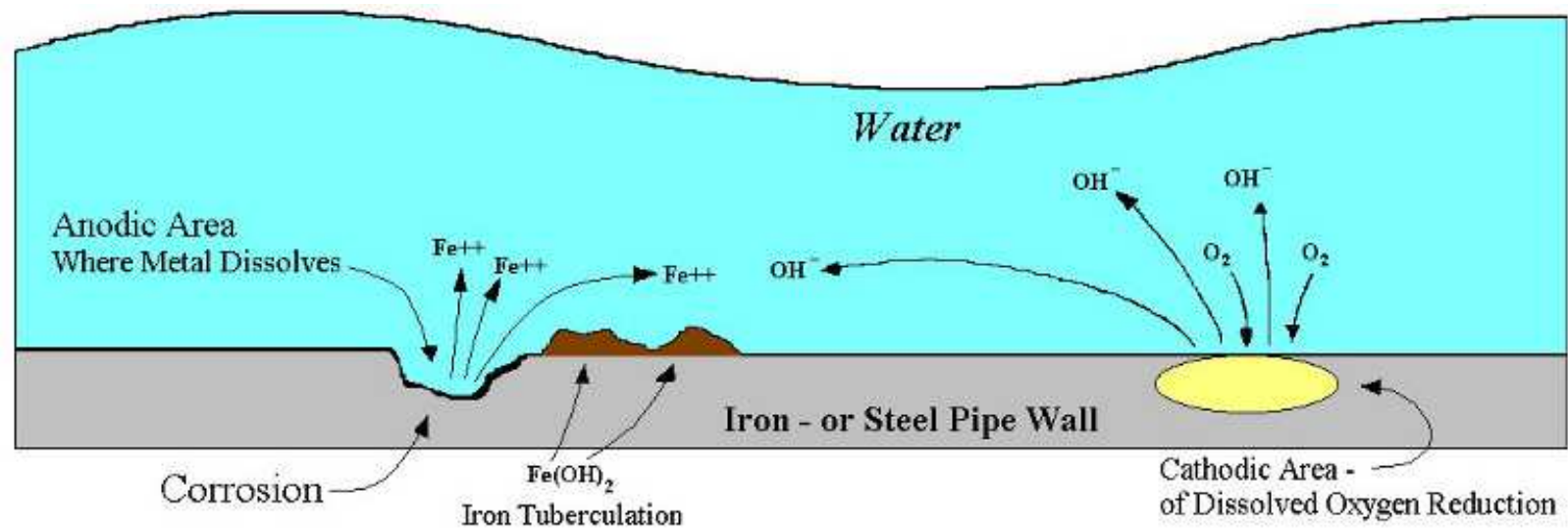


Poursaee (Ed.), Corrosion of Steel
in Concrete Structures
Woodhead, 2016

Corrosion products of iron



Corrosion cell

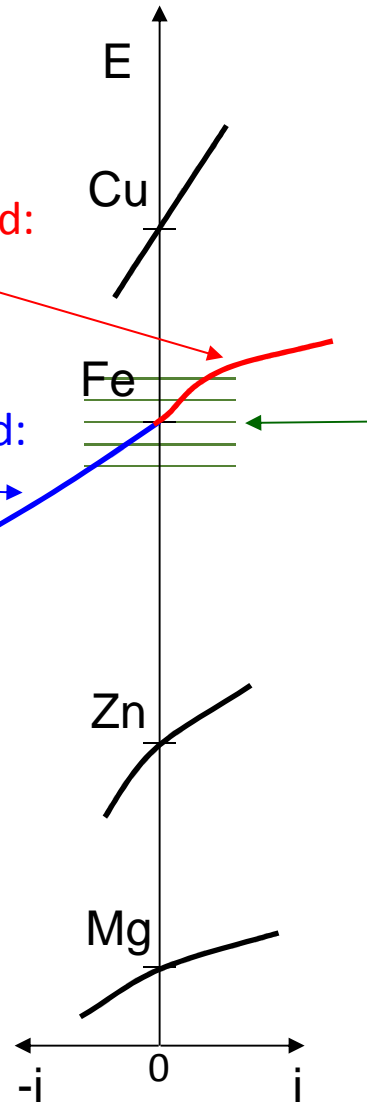
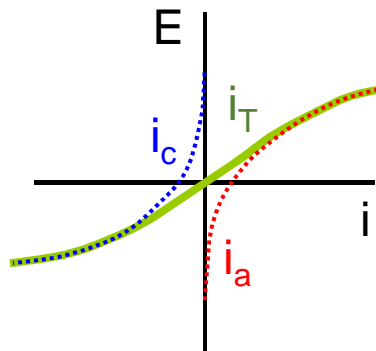


Electrochemical Kinetics

Overpotential
 $\eta = E - E^0$

$\eta > 0$, **oxidation** is favoured:
 $\text{Fe} \rightarrow \text{Fe}^{2+} + 2\text{e}^-$

$\eta < 0$, **reduction** is favoured:
 $\text{Fe}^{2+} + 2\text{e}^- \rightarrow \text{Fe}$



Dynamic equilibrium:



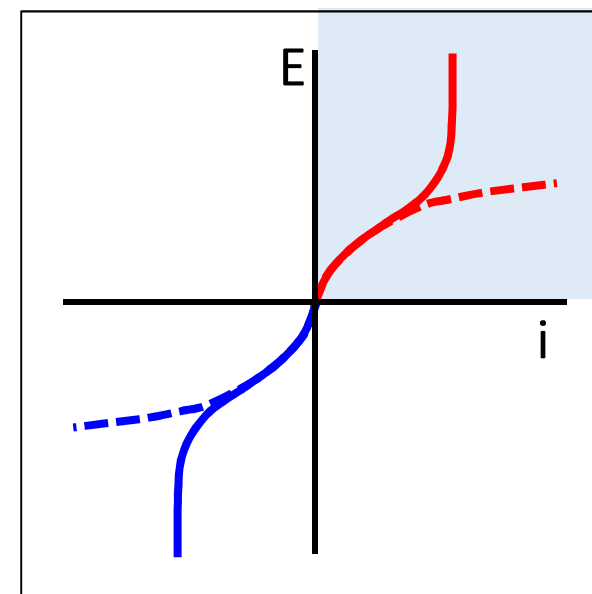
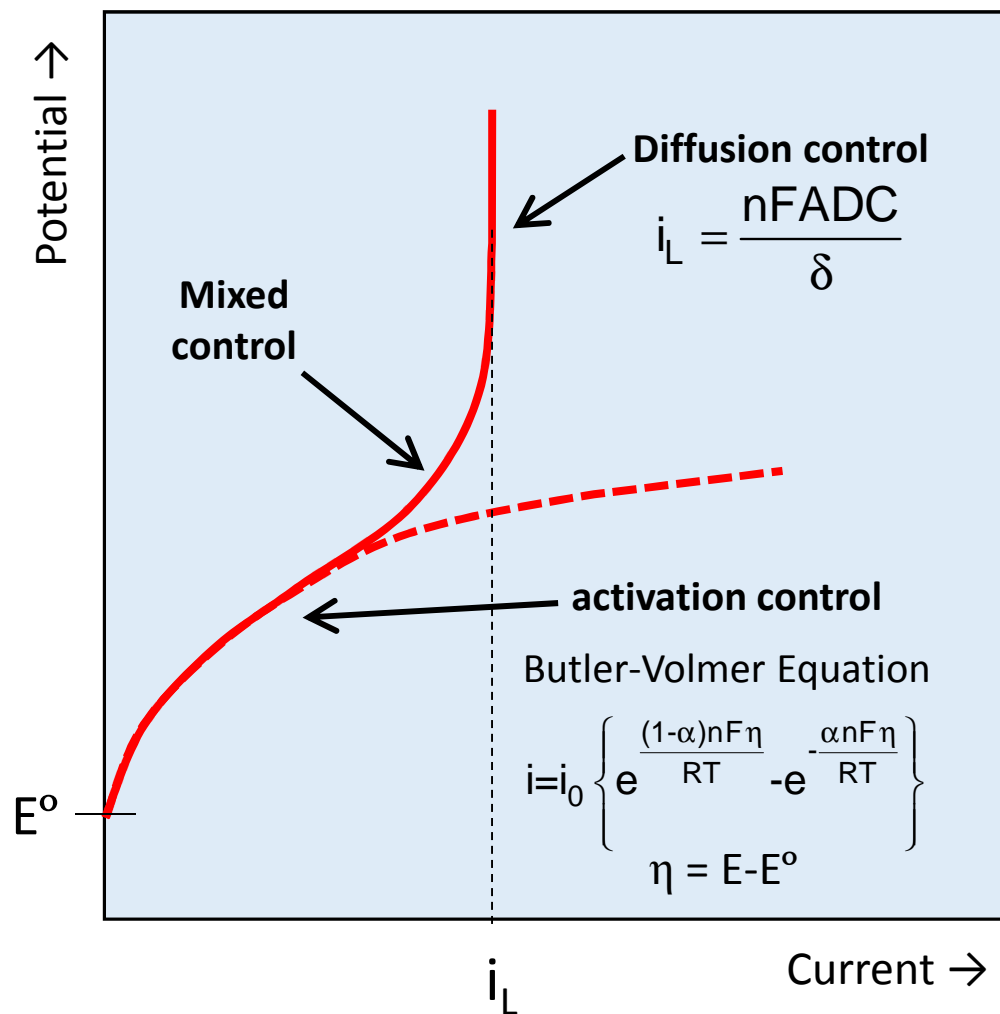
E^0 ($a_{\text{Fe}^{2+}} = 1$; 25 °C; no other reactions)

For other conditions

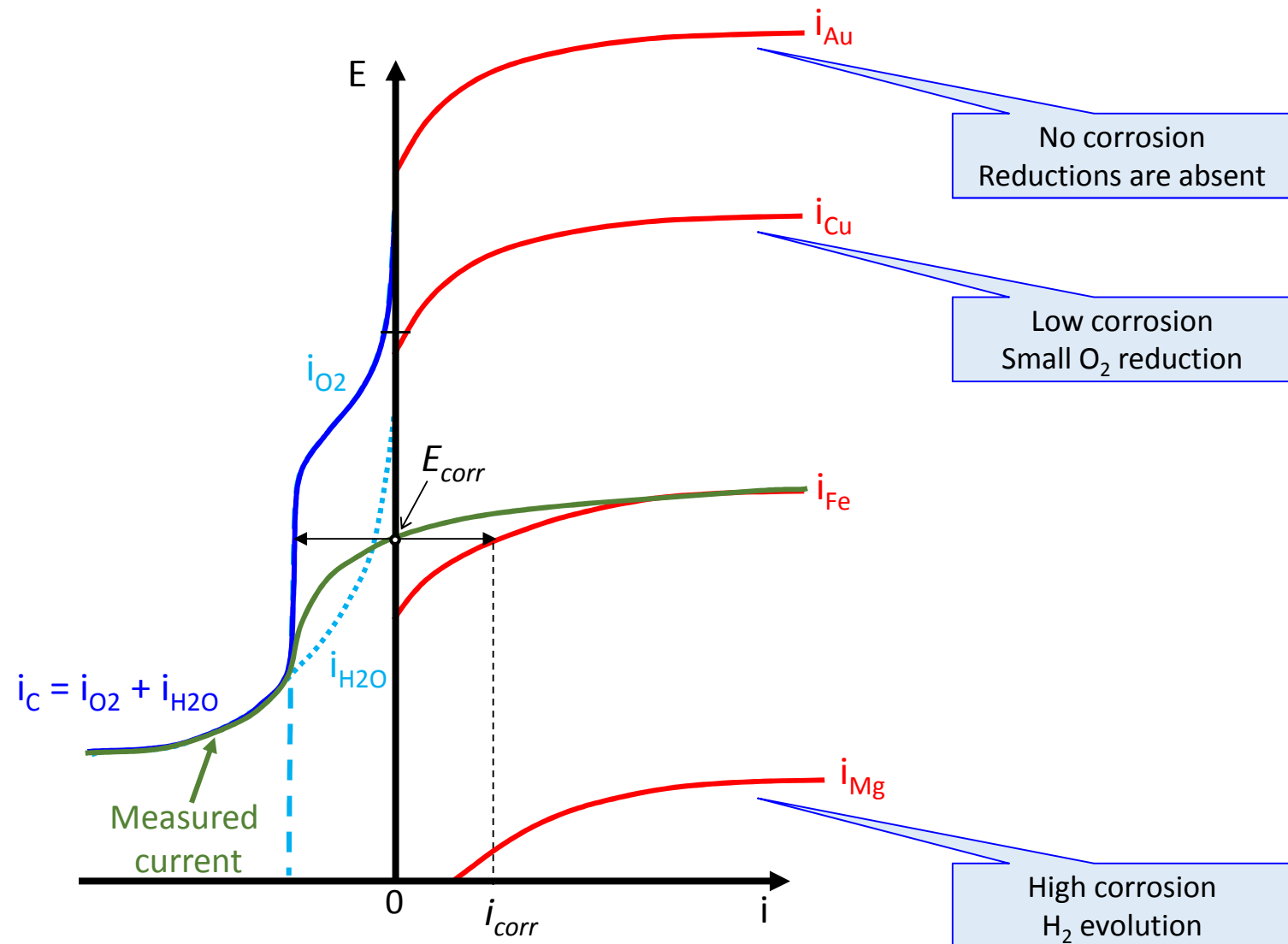
Nernst equation

$$E = E^0 + \frac{RT}{nF} \ln a_{M^{z+}}$$

Electrochemical Kinetics



Electrochemical Kinetics of Corrosion



Corrosion rates

3 equivalent units:

- Corrosion current density (current density by unit area)
(A m⁻², mA/cm², μA cm⁻², ...)
- Mass loss
(g dm⁻² ano⁻¹, mg dm⁻² d⁻¹, ...)
- Penetration rate (valid only for uniform corrosion)
(mm/year, MPY *milli-inch per year*)

$$\frac{m}{A t} = \frac{i_{corr} M}{n F} \quad \text{or} \quad \frac{i_{corr}}{F \sum \frac{n_i f_i}{M_i}}$$

pure metals
alloys

Dividing both terms by density
gives the thickness lost during time t

m = mass

A = area

t = time

M = molar mass

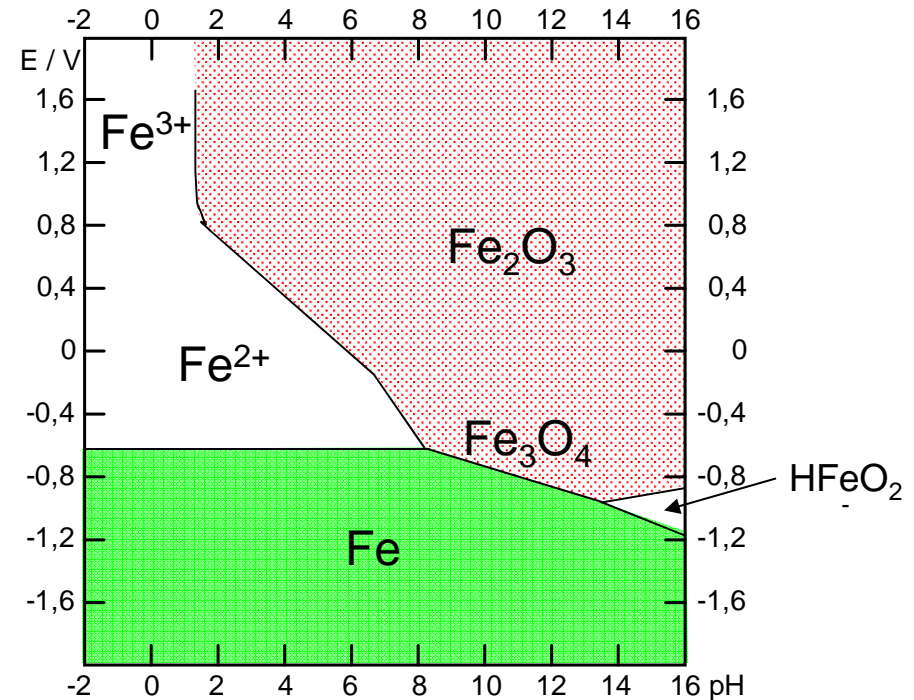
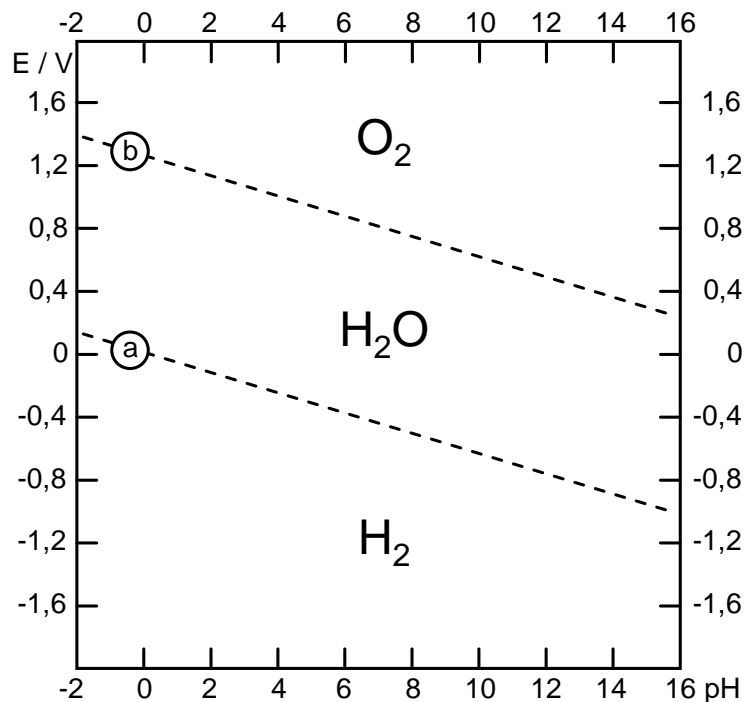
n = n^o electrons in reaction

F = Faraday constant (96 485 C mol⁻¹)

f = mass fraction

For iron: 1 μA cm⁻² = 0.25 g m⁻² day⁻¹ = 12 μm year⁻¹
(M_{Fe} =55.845 g mol⁻¹, d =7.86 g cm⁻³)

Pourbaix diagrams



Nernst equation of a metal | metal ion ($M|M^{n+}$) half-cell

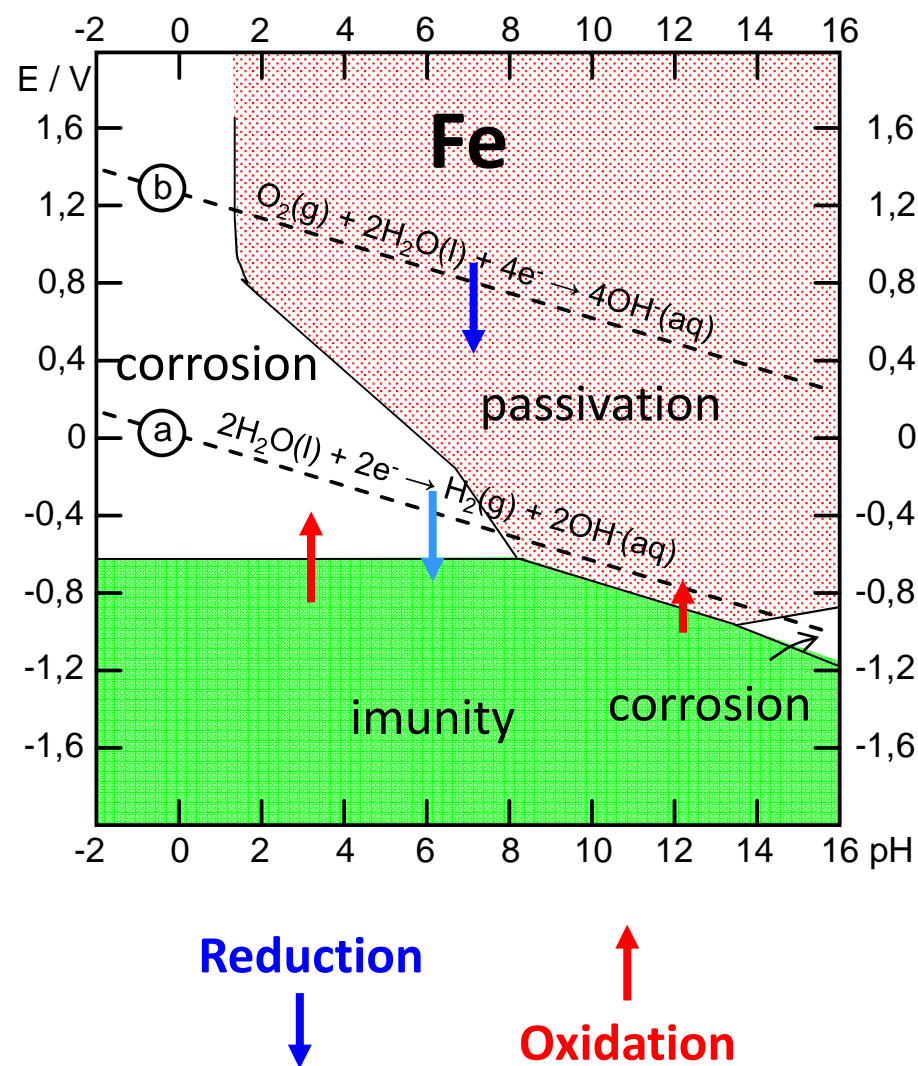
$$E = E^o - \frac{RT}{zF} \ln Q \quad E = E^o - \frac{RT}{zF} \ln \frac{a_{Red}}{a_{Ox}} = E^o - \frac{RT}{zF} \ln \frac{a_M}{a_{M^{z+}}} = E^o - \frac{RT}{zF} \ln \frac{1}{a_{M^{z+}}} = E^o + \frac{RT}{zF} \ln a_{M^{z+}}$$

3 regions in Pourbaix diagrams:

Corrosion. Regions where the thermodynamically stable species are the metal ions. Therefore metal dissolution is favourable.

Passivation. Regions where condensed phases containing the metal ion (e.g. metal oxides or hydroxides) are the most stable. If they form films on the surface, well adherent and without pores, they may prevent metal corrosion.

Imunity. In this region, the stable form is the reduced (metallic) form. There is no tendency for the metal oxidation.



Passivity

Some metals and alloys present extremely low corrosion rates in conditions where thermodynamically they should corrode. The metals are said to be passive and the property is called **passivity**. Important examples are alloys of aluminium, nickel and stainless steels.

Passivation is an elegant phenomenon and one of the reasons why our civilization is based on metallic materials.

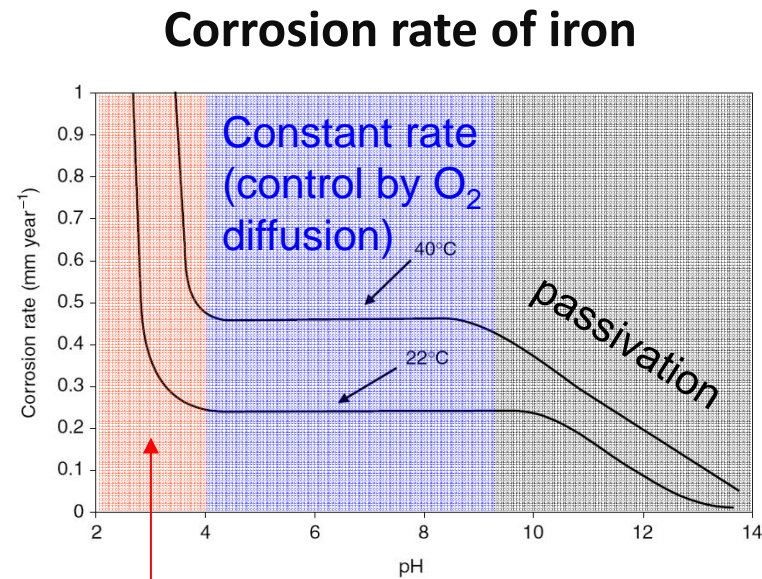
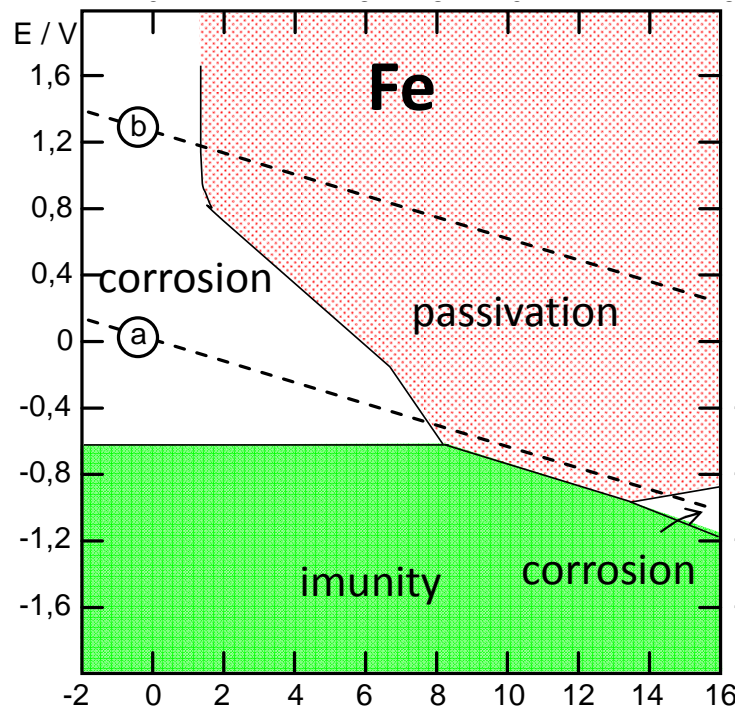
It consists on the spontaneous formation of very thin oxide films (typically in the order of angstroms at the surface of some metals and alloys, isolating it from the environment thus preventing its further reaction.

Some metals can be passivated when exposed to oxidizing environments (ex: iron in chromate or nitrite solutions) or by applying anodic polarizations with sufficient high current densities (ex: iron in H_2SO_4).



Critical enviromental parameters

pH



Source: R.W.Revie, H.H.Uhlig, Corrosion and Corrosion Control 4th Ed, Wiley, 2008

Critical enviromental parameters

O₂ concentration

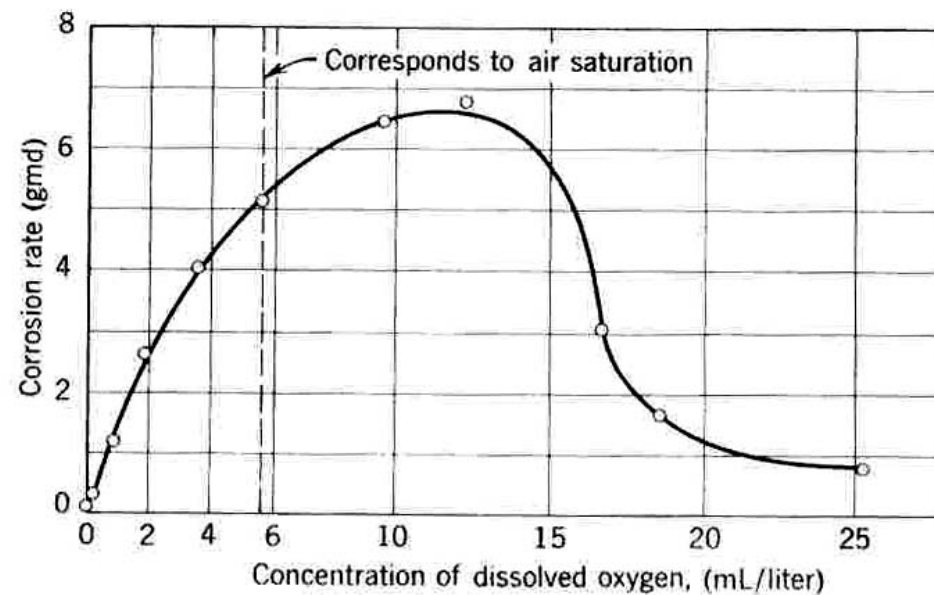


Figure 7.1. Effect of oxygen concentration on corrosion of mild steel in slowly moving distilled water, 48-h test, 25°C [2]. (Reproduced with permission. Copyright 1955, The Electrochemical Society.)

Critical enviromental parameters

Cl⁻ concentration (salinity, conductivity)

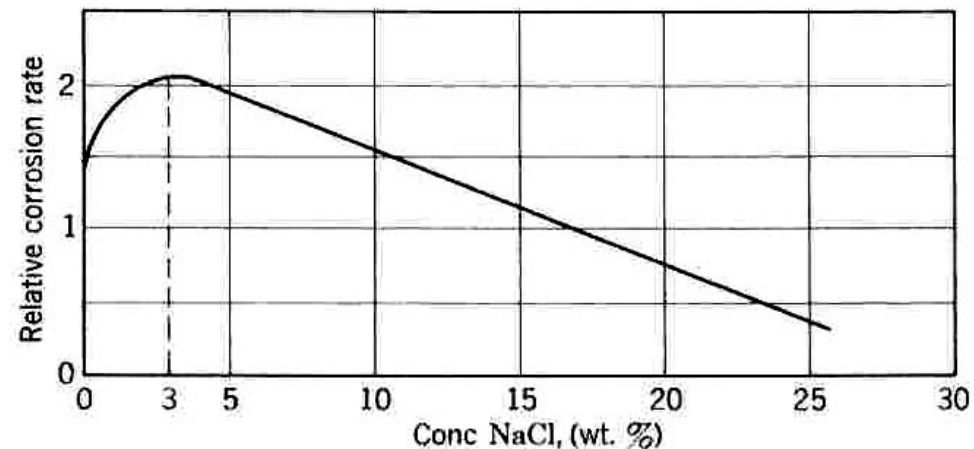
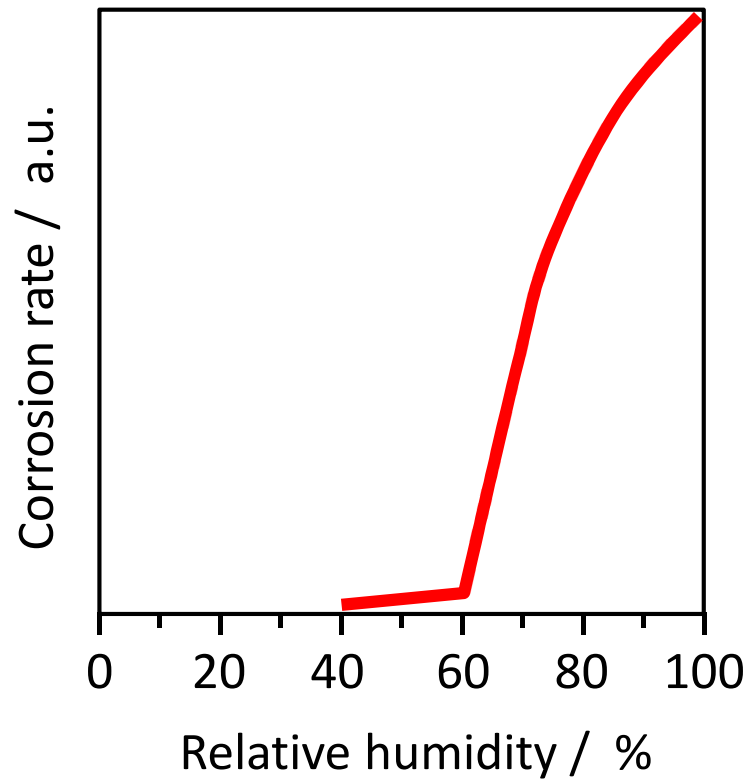


Figure 7.11. Effect of sodium chloride concentration on corrosion of iron in aerated solutions, room temperature (composite data of several investigations).

Critical enviromental parameters

Humidity



Forms of Corrosion

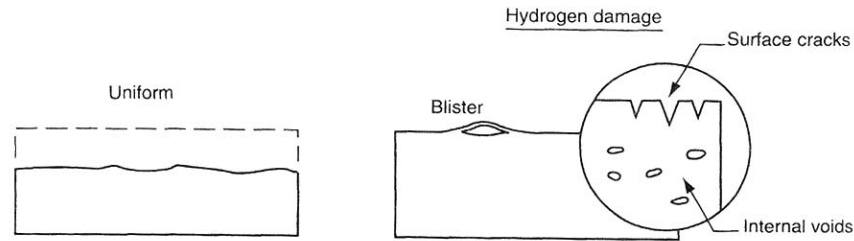
Uniform



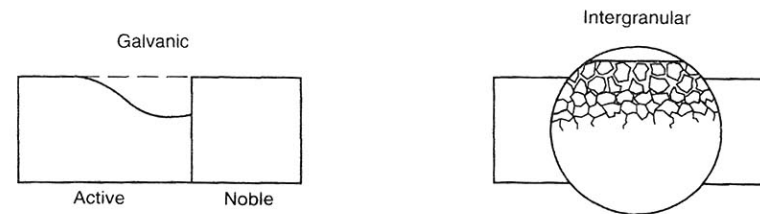
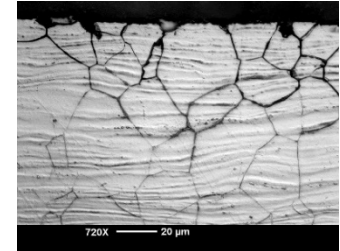
Galvanic



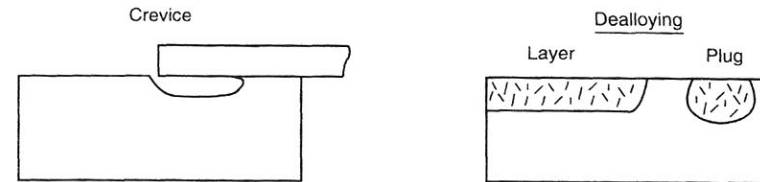
Pitting



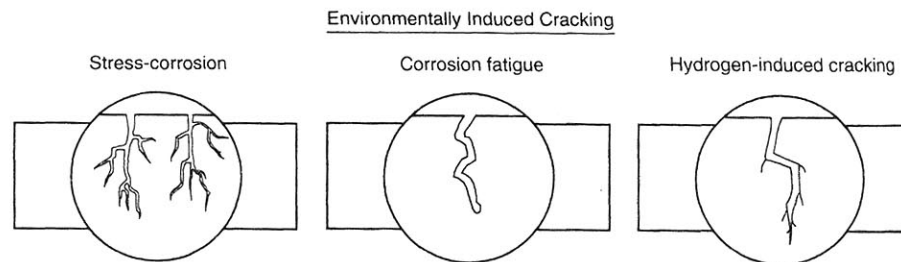
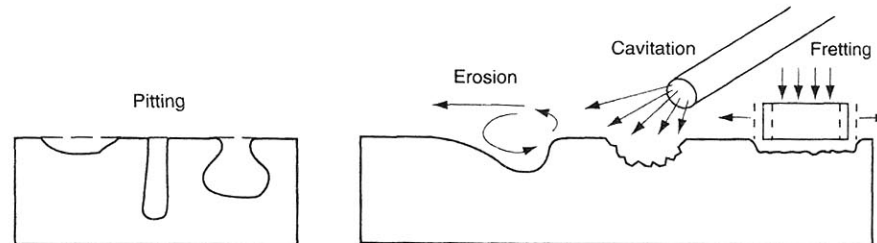
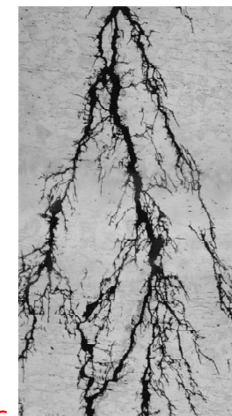
IG



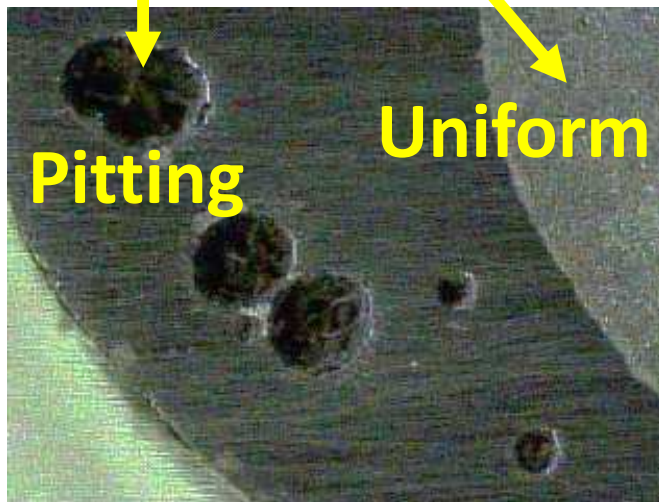
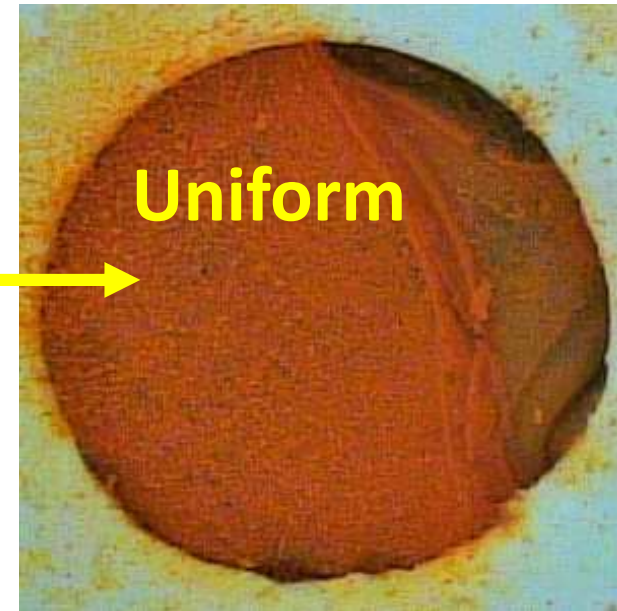
C. fatigue



SCC



Forms of corrosion



Corrosion protection and prevention

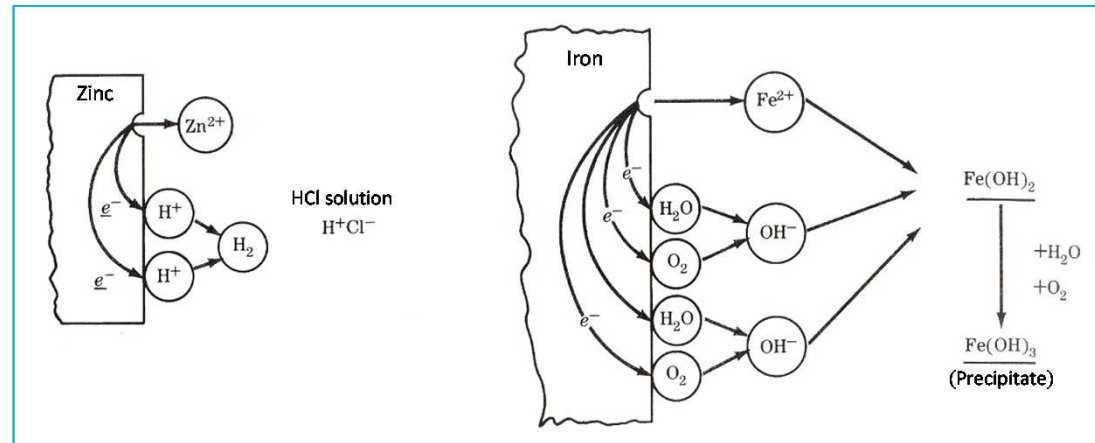
Eliminate:

Anodic reaction

Cathodic reaction

Electronic path

Ionic path



1. Action on the metal
2. Action on the environment
3. Barrier between metal and environment

Corrosion protection and prevention

Project

- Materials selection
- Design of structures and equipment
- Choice of placement of structures and equipment

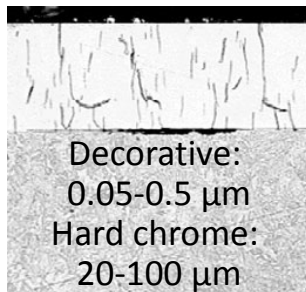
In service

- Control of environment
 - humidity
 - temperature
 - removal of O₂ (and other oxidants)
 - removal of aggressive species (e.g. Cl⁻)
 - control of fluid motion
 - corrosion inhibitors
- Control of potential (electrical protection)
 - Cathodic protection
 - Anodic protection
- Coatings (separation between metal and environment)
 - metallic
 - inorganic (CCC, phosphating, anodising)
 - organic (paints)

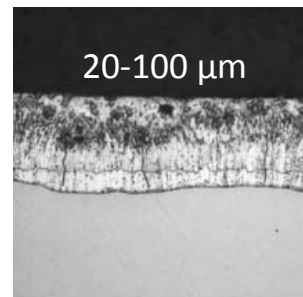
Metallic coatings

- Barrier effect
- Cathodic protection (some)

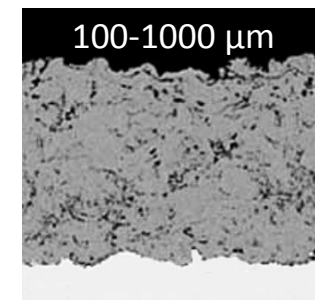
Electroplated
chromium



Hot dip galvanized
steel (HDG)

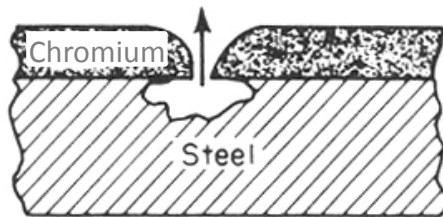
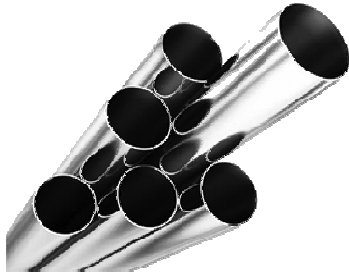


Thermal sprayed
aluminium (TSA)



Metallic coatings

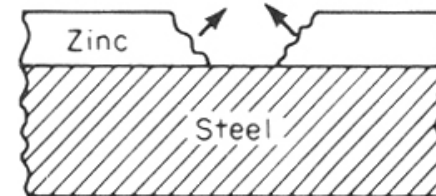
Chromium on steel



**Barrier
effect**



Zinc on steel



**- Barrier
- Cathodic
protection**



Organic coatings (paints)

Widely used, low-price, versatile.

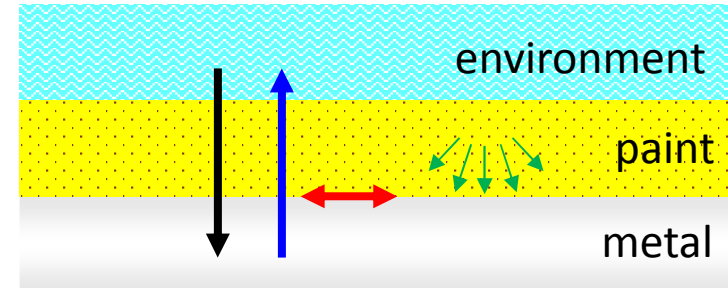
Composition

Binder (first natural oils, now synthetic resins)

Pigments (and fillers)

Solvents

Additives (defoamers, dispersants, thickeners...)

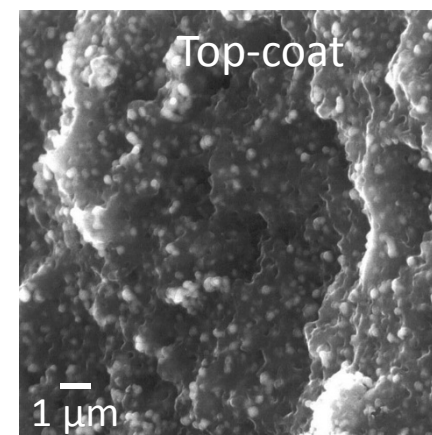
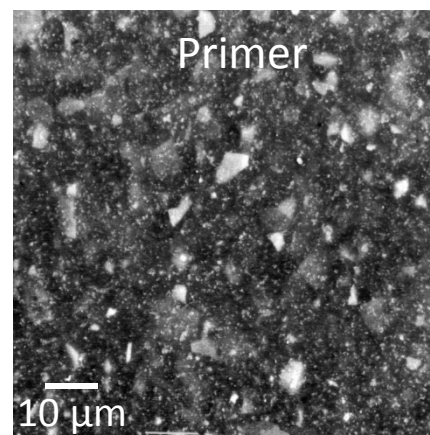
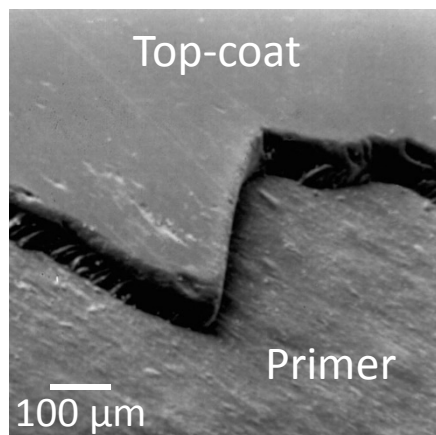


Barrier (water, gases, ions)

Barrier (corrosion products)

Barrier (ions, high resistance between an. and cat.)

Anticorrosive pigments



Corrosion inhibitors

Definition

A corrosion inhibitor is a chemical compound that decreases the corrosion rate of a metal or an alloy.

Classifications

Electrochemical effect: { **Anodic inhibitor** (decreases the anodic reaction)
Cathodic inhibitor (decreases the cathodic reaction)
Mixed inhibitor (decreases both reactions)

Chemical nature: organic, inorganic

Medium: aqueous solution, vapour, oil, solvent, paint, concrete...

Type of application: pickling, descaling, acid cleaning, preservation, paints, recirculating water systems, ...

Corrosion inhibitors

Acid

- Oxide free surface
- Main cathodic process is H_2 evolution

- **Physical adsorption**
(electrostatic interaction)
- **Chemisorption**
(charge sharing or charge transfer)

Examples:

- Triple bonded hydrocarbons
 - Sulfoxides, sulfides, mercaptans,
 - Aliphatic, aromatic or heterocyclic compounds containing nitrogen, aldehydes, amines.
- $P > Se > S > N > O$

Neutral

- Surface with sparingly soluble oxides, hydroxides, salts
- Main cathodic process is O_2 reduction

- **Stabilization** of the oxide surface film,
- **Repassivation, repair** of defects, **plug** pores,
- Thick surface layers with poor ionic/electronic conduction

- 1 Cations:** Ca^{2+} , Mg^{2+} , with CO_3^{2-} , Ce^{3+} and RE^{3+} (cathodic process)
- 2 Inorganic anions:** polyphosphates, phosphates, silicates, borates precipitate (inhibit anodic process).
- 3) Oxidizing ions:** chromates, NO_2^- , ...
- 4) Organic compounds:** benzoate, salicylate, cinnamate, tartrate (Adsorption, complexation, ion exchange, chelation ($-NH_2$, $-COOH$, $-SH$, physical barrier with thick films)

Alkaline

Passive surface or active amphoteric surfaces

Action:

- Broadening the pH stability range of amphoteric oxides and hydroxide layers,
- repairing pores,
- decrease the rate diffusion of reactants

Examples:

Thiourea, phenols, 8-hydroxyquinoline, naphtols, gelatin, tannins, sapoin.

Corrosion inhibitors

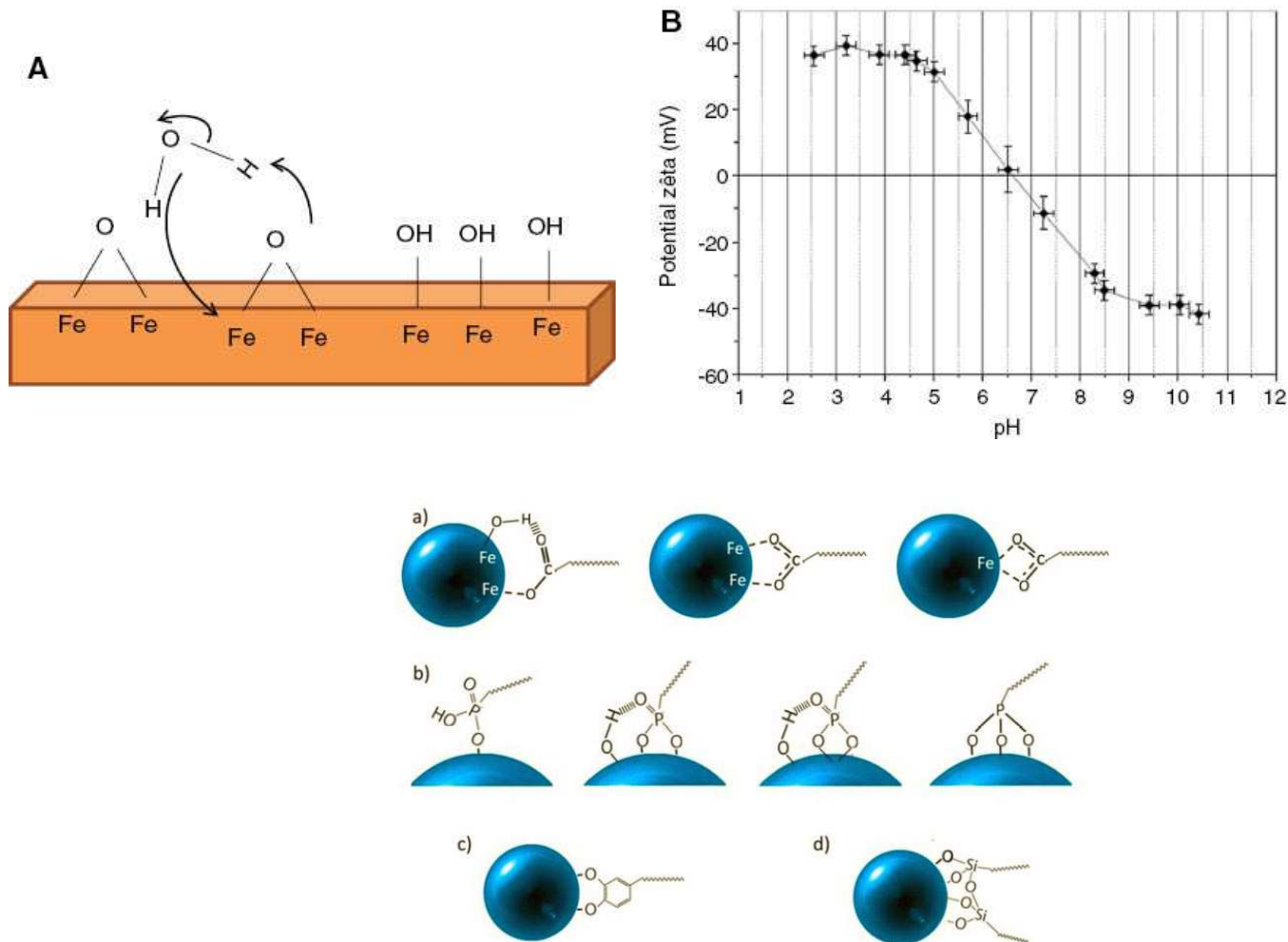
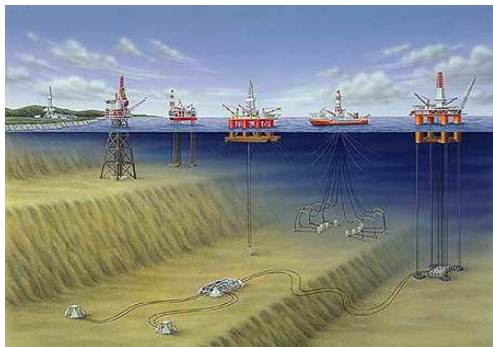


Figure 4: Possible surface complexes according to the coupling agent: (a) carboxylate, (b) phosphonate, (c) catechol, (d) silane.

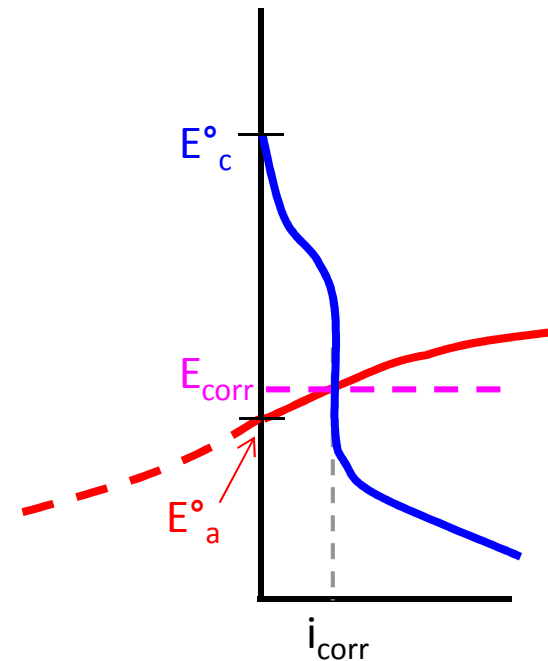
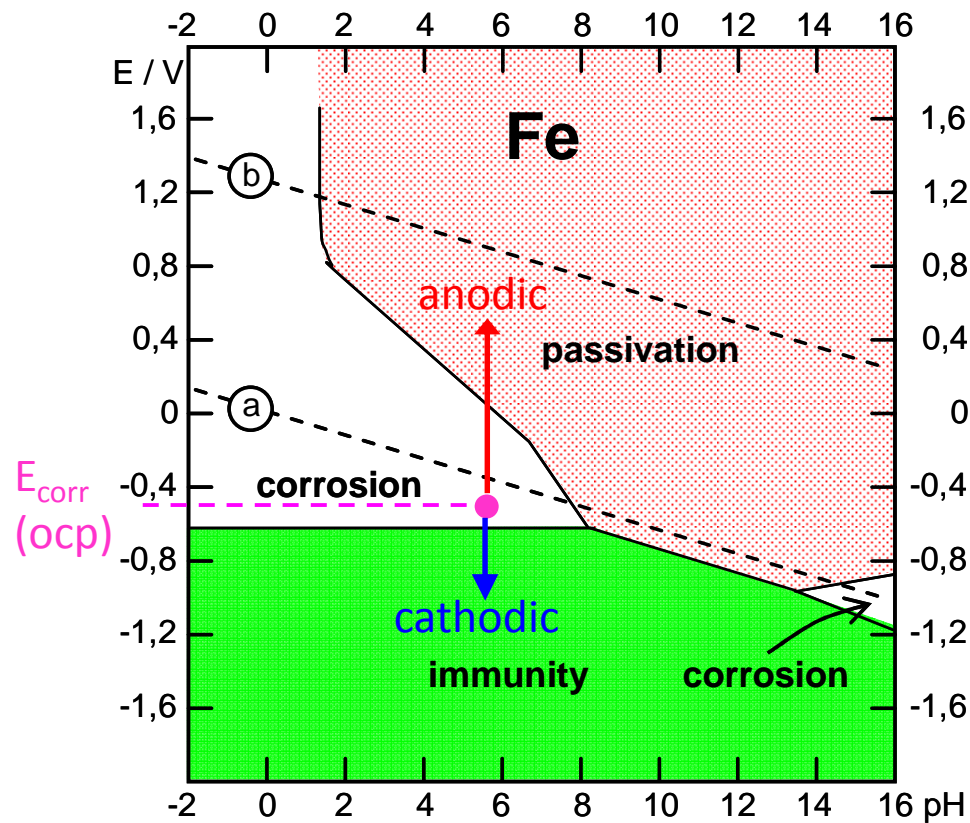
Cathodic protection

Method widely used to protect naval ships, offshore structures, pipelines, storage tanks, bridges, etc.



Cathodic protection

With **cathodic protection** the metal to be protected is turned into a cathode. Its potential is made more negative than the corrosion potential. When the potential is inside the immunity region, the metal oxidation becomes thermodynamically impossible and it will not corrode.



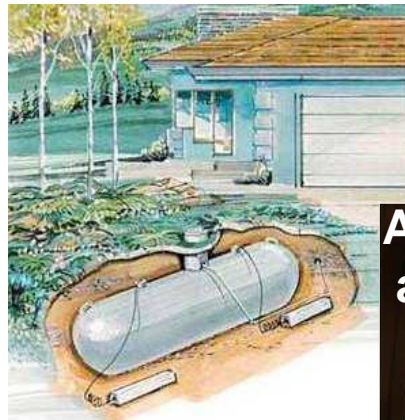
Cathodic protection

2 forms of implementation:

Sacrificial anodes



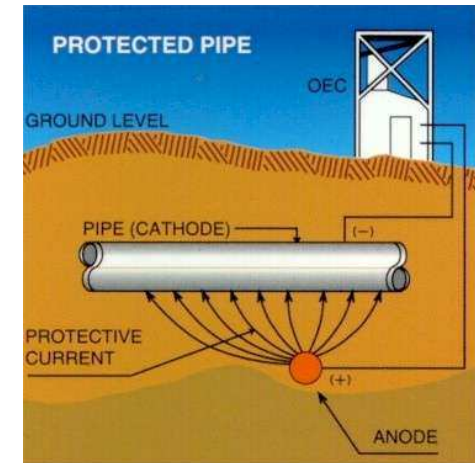
Zinc anodes



Aluminium and magnesium anodes

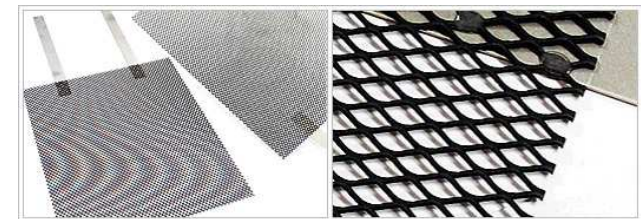


Impressed currents

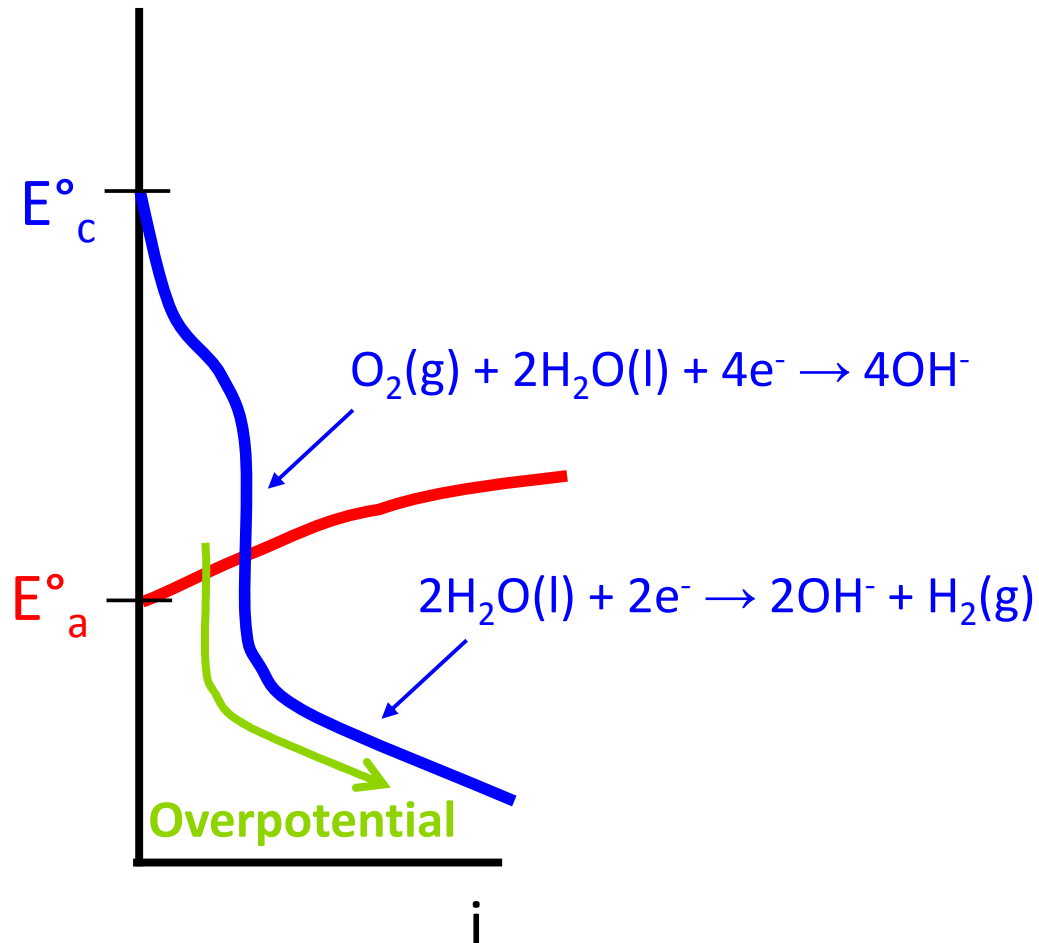


Inert electrodes:

Pt in Ti,
Pb6Sb1Ag,
graphite,
Fe14Si4Cr.



Cathodic protection



Excessive overpotential leads to water reduction and high current.

OH⁻: cathodic delamination, paint disbondment.

H₂: H(ads) can enter the metal phase and cause hydrogen embrittlement.

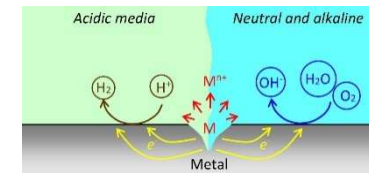
Key messages of this presentation

Corrosion as a thermodynamic inevitability

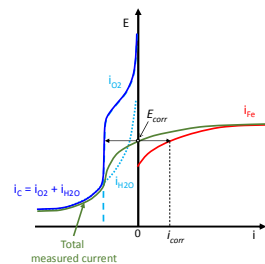
Why metals corrode



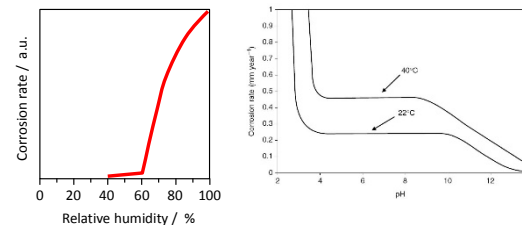
2 reactions (oxidation of metal and reduction from the environment)



Kinetics



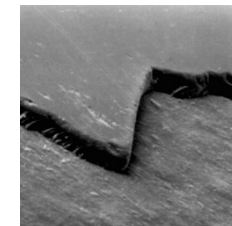
The importance of environment factors



Corrosion control.

Eliminate anodic reaction, and/or cathodic reaction, and/or flux of electrons and/or flux of ions.

Act on the metal, act on the environment or put a barrier between them.



Further reading

Internet sites

<http://www.corrosion-doctors.org>

<http://www.efcweb.org>

<http://www.nace.org>

<http://corrosion.ksc.nasa.gov>

Books

D.A.Jones, Principles and Prevention of Corrosion 2nd Ed, Prentice Hall, 1996.

M.G.Fontana, Corrosion Engineering 3rd Ed, McGraw-Hill, 1987.

R.W.Revie, H.H.Uhlig, Corrosion and Corrosion Control 4th Ed, Wiley, 2008.

H.Kaesche, Corrosion of Metals, Springer, 2003.

Shreir's Corrosion, 4-Vol. Set, Elsevier Science, 2010.

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www.lorcenis-eu.com

