# Corrosion
# MM 454 IITB
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https://somphene.github.io/notes/

Notes
by
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*When Corrosion Engineers don't work, the world rusts.*

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This is an **incomplete draft**. Please send corrections, comments, pictures of bad drawings, etc. to somphene1@gmail.com.

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# 1 Lecture Jan 13

## §1.1 References

- Mars Guy Fontana's Corrosion engineering [Fo05].
- Edward McCafferty's Introduction to corrosion science [Mc10]
- R Winston Revie and Herbert H. Uhling's Corrosion and corrosion control: an introduction to corrosionscience and engineering [Re08]
- Specific scientific reports will be cited throughout the chapters and can be found in the Bibliography (at the end of these notes).

## §1.2 Course objectives

- Provide a basic understansding of corrosion phenomena.
- Provide tools to measure, analyze and predict the corrosion of materials.
- Provide corrosion prevention and remediation strategies.

## §1.3 Introduction to Corrosion

> **Question 1.3.1.** What is Corrosion?

Corrosion is degradation of materials. Basic notions of electrolyte, cathodes and anodes can be used to explain corrosion in Stainless Steel, pitting corrosion and other forms of corrosion. Any big tangible structures like ships or buildings or bridges, you will find steel/copper. Even in small devices like mobiles, laptops there are copper wires for conducting networks. Car frames and aeroplanes made of Aluminium. What metals undergo corrosion. Examples of corrosion detection are: rust in automobiles, titanium in implants (corrodes over a long period of time).

## §1.4 Why study corrosion

- Fire on an aircraft Airbus A330 engine (Manchester, 2013) because of fracture of blades via fatigue. A good overview of applications of alloys in automative and aerospace industries is provided in [Zh+18].
- Bhopal Gas Tragedy also caused due to corrosion. Runaway chemical reaction-starts initiation but doesn't stop, for instance in mobile phones sparks. High temperature and pressure. High levels of chloroform. Carbon steel went corrosion in acidic media at high temperature.
- India loses upto $100 billion annually (according to Hind Zinc CEO Sunil Duggal) thats about 4-5 % of the GDP.

- Conservation of materials: Limited supply of materials in specific geographical locations. Wastage of energy and human effort. Service extension mitigates additional manufacturing.

- Development of corrosion resistant alloy oxides.

- A good account of how important and complex corrosion can be is given in Comsol Blogs Corrosion Resistance.

## §1.5 Corrosion- Is it complex?

Various environments have various features unique to each of them. For instance- salts are available in Marine environment, Chloride anions from salts is significant. Sulphur dioxide in industrial applications. Sulphate anion in the soil. Materials prone to corrosion are steel, stainless steel, aluminium alloys, copper, nickel, titanium alloys. Complex systems of materials. Different case studies map to different root causes.

## §1.6 Corrosion- An interdisciplinary field

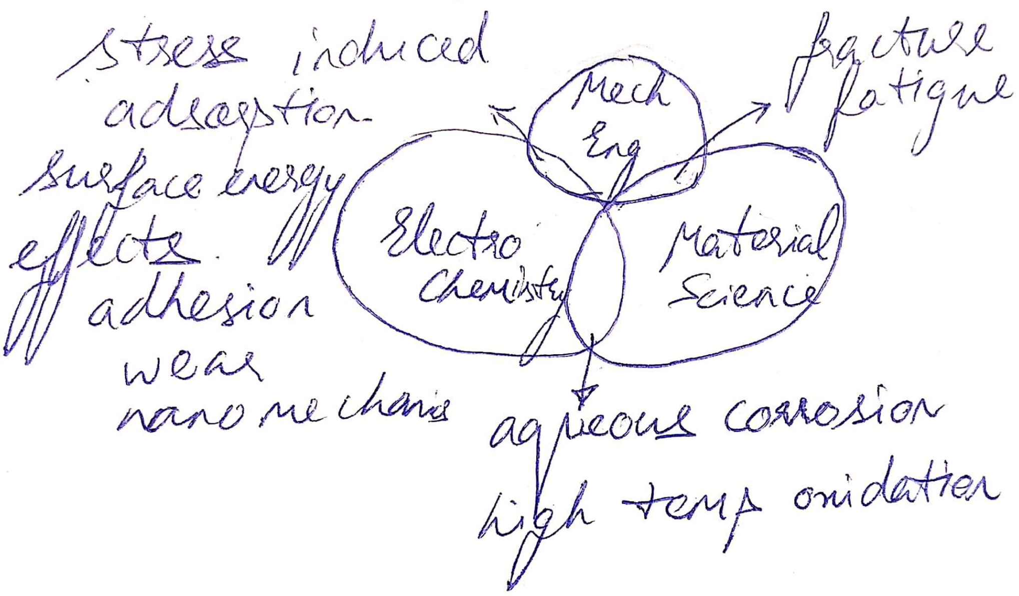

stress induced
adsorption
surface energy
effects
adhesion
wear
nano mechanics
Mech
Eng
fracture
fatigue
Electro
Chemistry
Material
Science
aqueous corrosion
high temp oxidation

Figure 1.1: Venn Diagram of interacting fields

Electrochemistry, Mechanics/Engineering, Materials Science as in fig. 1.1. Involves interaction of Stress induced adsorption surface energy defects- adhesion, wear, nanomechanics, tribology. Aqueous corrosion- high temperature oxidation.

## §1.7 Corrosion- Definition

Origin- comes from Latin corrodere- gnaw away or eat away.

**Corrosion** is characterized as:

- Natural phenomenon
- destructive attack on metal by its environment, mainly electrochemical factors.
- causes deterioration of properties of the metal.

Plastics, concrete, wood, ceramics, composite materials undergo deterioration by environment but don't corrode.
**Rusting** applies to plain carbon steel and iron. **Rust**: hydrated ferric oxide is red or dark brown color. Non ferrous metals- aluminium, copper, zinc corrode but don't rust.

## §1.8 Thermodynamic viewpoint

Thermodynamic reaction by which a metal returns back to its natural form as an ore. Extraction/alloying is gaining energy (opposite to spontaneous reaction requires input of energy). Corrosion involves release of energy. This is in the opposite direction of extractive metallurgy.

For instance, $\text{Fe} + C$ (+Cr, Ni, Si) corrosion cycle.

Fe alloys - Haematite ($\text{Fe}_2O_3$), Wustite (FeO) are common.

$\text{Cr}_2O_3$ layer stops corrosion. Thermodynamics drives corrosion.

## §1.9 Electrochemical Reactions

Corrosion is predominantly an electrochemical process involving coupled electrochemical half cell reactions.

**Half Cell reaction- Anodic**

- Species undergoes oxidation.
- Loss of electrons from the species at the anodic site, ie. electrons are produced at the anodic site.

**Example 1.9.1**

The following reactions are common examples of anodic reactions:

- $\text{Fe} \longrightarrow \text{Fe}^{2+} + 2e^-$
- $Al \longrightarrow Al^{3+} + 3e^-$
- $2\text{Cu}\,(s) + H_2O\,(l) \longrightarrow \text{Cu}_2O\,(s) + 2H^+\,(aq) + 2e^-$

### Half Cell reaction- Cathodic

- Species undergoes reduction.
- Gain of electrons for species at the cathodic site, ie. electrons are consumed at the cathode.

**Example 1.9.2**

The following reactions are common examples of anodic reactions:

- In acidic medium: $H^+ + 2e^- \longrightarrow H_2$
- In basic/neutral medium: $O_2 + 2H_2O + 4e^- \longrightarrow 4OH^-\,(aq)$

Charge + Mass transport.

**Corrosion involves simultaneous transfer of mass and charge across a metal/solution interface.**

Corrosion involves both half cell reactions to form a coupled electrochemical reaction.

**Example 1.9.3** (Iron in Acidic Solution Environment)

The following half-cell reactions couple to give an electrochemical reaction for Iron corrosion in acidic environment as shown in fig. 1.2.

$$\begin{array}{ccccc} \text{At anodic sites:} & \text{Fe} & \longrightarrow & \text{Fe}^{2+} + 2e^- \\ \text{At cathodic sites:} & 2H^+ + 2e^- & \longrightarrow & H_2 \\ \hline \text{Overall reaction:} & \text{Fe} + 2H^+ & \longrightarrow & \text{Fe}^{2+} + H_2 \end{array}$$

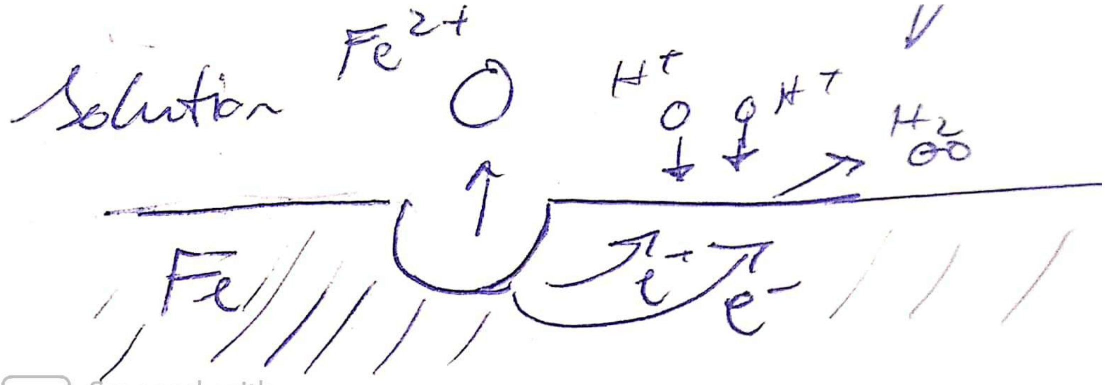


Figure 1.2: Fe surface corroding in Acidic Medium

Thus, corrosion involves the following components:

- Anodic reaction

- Cathodic reaction
- Electrolyte
- Metallic contact to serve as path between anode and cathode for electron transport.

> **Question 1.9.4.** How can two different electrochemical reactions happen on the same metal surface?

Energy of surface drives reaction. Difference in energies between two points on the metal surfaces exist thus favoring different reactions at different points. Insert diagram (later). *Heterogeneous nature of the surface* creates difference in energy.

**Example 1.9.5** (Iron Surface immersed in Neutral/Alkaline medium)

The following half-cell reactions couple to give an electrochemical reaction for Iron corrosion in neutral/alkaline environment as shown in fig. 1.3.

$$\begin{array}{lccc} \text{At anodic sites:} & 2[\mathrm{Fe}\ (\mathrm{s}) & \longrightarrow & \mathrm{Fe}^{2+}(\mathrm{aq}) + 2e^-\ ] \\ \text{At cathodic sites:} & O_2\,(g) + 2H_2O\,(l) + 4e^- & \longrightarrow & 4\mathrm{OH}^-\,(\mathrm{aq}) \\ \hline \text{Overall reaction:} & 2\mathrm{Fe}\,(s) + O_2\,(g) + 2H_2O\,(l) & \longrightarrow & 2\mathrm{Fe}^{2+}(\mathrm{aq}) + 4\mathrm{OH}^-\,(\mathrm{aq}) \end{array}$$

The aqueous species formed may further react as

$$\mathrm{Fe}^{2+}(aq) + 2\mathrm{OH}^-(aq) \rightarrow \mathrm{Fe}(OH)_2\,(s)$$

This leads to precipitation of ferrous hydroxide on the Iron surface. In the presence of water, this forms hydrated ferric oxide as

$$2\mathrm{Fe}(OH)_2\,(s) + (x-1)H_2O(l) \longrightarrow Fe_2O_3 \cdot xH_2O\,(s) + 2H^+\,(\mathrm{aq}) + 2e^-$$

This is shown in fig. 1.3

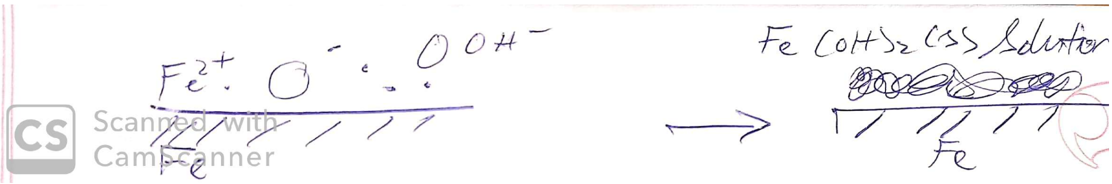


Figure 1.3: Fe precipitates out Hydroxide in Alkaline Medium

> **Remark 1.9.6** — In the presence of an acidic media, there was a different reaction leading to Hydrogen embrittlement.

The above phenomenon is shown in fig. 1.4

## §1.10 Faraday's Law

Corrosion involves simultaneous charge and mass transfer. Charge transfer and Mass transfer related by **Faraday's Law** which states that the mass $W$ of metal (liberated or deposited/corroded) is given by:

$$W = \frac{ItM}{nF} \tag{1.1}$$

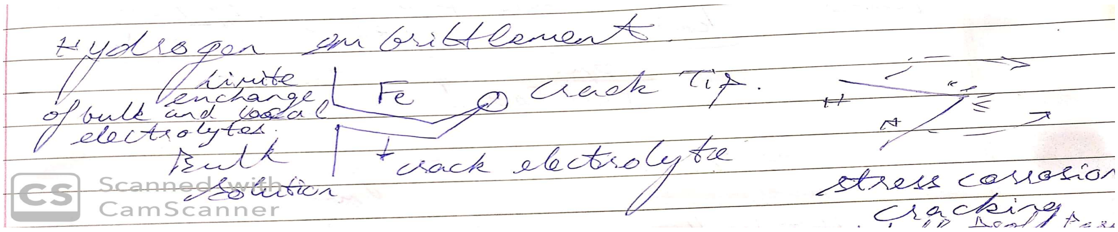

Hydrogen embrittlement
Limited exchange of bulk and local electrolytes
Fe
crack tip
Bulk solution
crack electrolyte
H
stress corrosion cracking

Figure 1.4: Hydrogen Embrittlement (Stress Corrosion Craking)

where $F$ is called the **Faraday's Constant**.

$$F = \frac{96500\,C}{\text{Mole equivalent}}$$

$I$ is the current, $t$ is the time taken for the transfer, $M$ is the Molar Mass of the metal and $n$ is the # of elementary charge ($e$) particles per mole of the metal involved in the reaction given by the stoichiometry.

**Example 1.10.1** (Iron half cell reaction)

Consider the Reaction:

$$\text{Fe} \longrightarrow \text{Fe}^{2+} + 2e^-$$

This involves 2 $e^-$ per mole of $\text{Fe}^{2+}$ produced. Thus, $n = 2$ is to be used in Faraday's Law.

# 2 Lecture Feb 4

## §2.1 Kinetics of Corrosion

Faraday's Law :

$$W = \frac{ItM}{nF} \tag{2.1}$$

where $F$ is called the **Faraday's Constant**. where $F$ is called the **Faraday's Constant**.

$$F = \frac{96500\,C}{\text{Mole equivalent}}$$

$I$ is the current, $t$ is the time taken for the transfer, $M$ is the Molar Mass of the metal and $n$ is the # of elementary charge ($e$) particles per mole of the metal involved in the reaction given by the stoichiometry.

$$\begin{aligned}
\text{Rate of Corrosion}\, r &= \frac{mass}{Area \cdot time} \\
&= \frac{m}{At} \\
&= \frac{(I/A) \cdot M}{n \cdot F} \\
&= \frac{i \cdot a}{n \cdot F} \\
r &= \frac{i \cdot a}{n \cdot F}
\end{aligned}$$

Lets take the reaction :

$$M \longrightarrow M^{n+} + ne^{-} : E^{rev} = E^{0}_{M^{n+}/M}$$

At equilibrium,

$$\text{rate of forward reaction } = \text{rate of reverse reaction}$$

There is still current in both forward and reverse directions, hence called **Dynamic Equilibrium**.

> **Question 2.1.1.** Rate of reaction is fast in which of the following electrodes? Say for the reaction $2H^{+} + 2e^{-} \longleftrightarrow H_2$:
>
> - Pt
> - Hg
> - Pb

- Pt : Fast
- Hg : Slow
- Pb : Slow

> **Question 2.1.2.** How to quantify?

**Exchange current Density**. There are

## §2.2 Exchange Current Density

$$
\begin{aligned}
r_f &= r_r \\
r_f &= r_r = \frac{i_0 \cdot a}{n \cdot F}
\end{aligned}
$$

Here $i_0$ := **exchange current density**
The value of $i_0$ depends on the electrode being used.
In thermodynamics, we have $E^{rev}$ as a thermodynamic parameter. In kinetics, we can use exchange current density $i_0$ as the parameter.

$$
\begin{aligned}
\text{At Equilibrium,} \quad & E = E^{rev} \\
\text{Away from equilibrium,} \quad & i_{net} \neq 0 \\
M &\longrightarrow M^{n+} + ne^- \\
\textbf{Overvoltage} \quad \eta &= E - E^{rev} \\
&= \text{change in potential from reversible potential} \\
&\quad \text{of a given electrode} \\
\text{Anodic current (oxidizing, +ve)} &= E - E^{rev} > 0; \eta > 0 \, (M \longrightarrow M^{n+} + ne^-) \\
\text{Cathodic current (reducing, -ve)} &= E - E^{rev} < 0; \eta < 0 \, (M^{n+} + ne^- \longrightarrow M)
\end{aligned}
$$

At high Electric field gardient (of the order of $10^8 V/m$) in between the surface of positive and negative charges, we can't use Ohm's Law. Hence we need a better model: **Activation State Theory**.

## §2.3 Activation State Theory

This is based on the principle that the system must surmount an energy barrier before reaction takes place.

$$
\begin{aligned}
\text{Free Energy,} \quad & G_{M^{n+}} < G_M \\
\Delta G_a^* &: \quad \text{Activation Energy barrier in forward direction} \\
\Delta G_c^* &: \quad \text{Activation Energy barrier in reverse direction} \\
\Delta G_a^* &= \Delta G_{a,ch}^* - \alpha n F \Delta\phi \\
\Delta G_{a,ch}^*, \Delta G_{c,ch}^* &:= \text{chemical component of activation energy} \\
&= \text{does not involve potential drop across interface}
\end{aligned}
$$

where $\alpha$: symmetry factor, fractional length along reaction coordinate.
and $\Delta\phi$: Potential drop across interface.

# 3 Lecture Feb 6

**Question 3.0.1.** Want to know how fast Zn corrodes.

$$\begin{aligned} E^0_{\mathrm{Zn}^{2+}\mathrm{Zn}} &= -0.76V \\ \mathrm{Zn}^{2+} + 2e^- &\iff \mathrm{Zn} \\ 2H^+ + 2e^- &\iff H_2 \quad E^0_{H^+/H} = 0.00 \end{aligned}$$

No corrosion in open circuit. Both reactions going forward and backward at equal rates. This is equilibrium. Nernst equation is valid only for equilibrium systems. If we pour in $Zn^{2+}$ ions, the concentrations will change so as to attain a new equilibrium.

$$[\mathrm{Zn}^{2+}] = 1M; [H^+] = 1M; H_2 = 1\mathrm{atm}$$

This gives $E^0$, that is standard state.

$$\begin{aligned} \mathrm{Zn} &\longrightarrow \mathrm{Zn}^{2+} + 2e^- \quad \text{oxidation} \\ \mathrm{Zn}^{2+} + 2e^- &\longrightarrow Zn \quad \text{reduction} \\ \text{At equilibrium,} \\ r_{\mathrm{oxn}} = r_{\mathrm{redn}} &= \frac{i_0 a}{nF} \quad \text{Faraday's Law} \end{aligned}$$

At equilibrium,

1. $E^0_{\mathrm{Mn}^{2+}\mathrm{Mn}}$
2. $i_0$

If we apply a potential of $E = -0.863V$. We go lower in the potential, which means we are providing more $e^-$, hence the reduction reaction moves forward faster than the backward reaction. The new current density found is plotted for each such potential. Similarly repeat for relatively positive potentials. This gives two straight rays emanating from the point $(E^0, i_0)$.

## §3.1 Polarization

Rate of a chemical reaction is a function of concentration, temperature and pressure. Rate of an electrochemical reaction is a function of concentration, temperature, pressure and potential. For a chemical reaction we need to provide enough energy to cross the barrier of the activation energy for the reaction to occur. We provide this energy by temperature or pressure. For electrochemical reactions, provide potential. This is formalized as the concept of **Overpotential**, denoted by $\eta$.

$$\eta := E_{\mathrm{applied}} - E_{\mathrm{equilibrium}}$$

This justifies the Butler-Volmer equation.

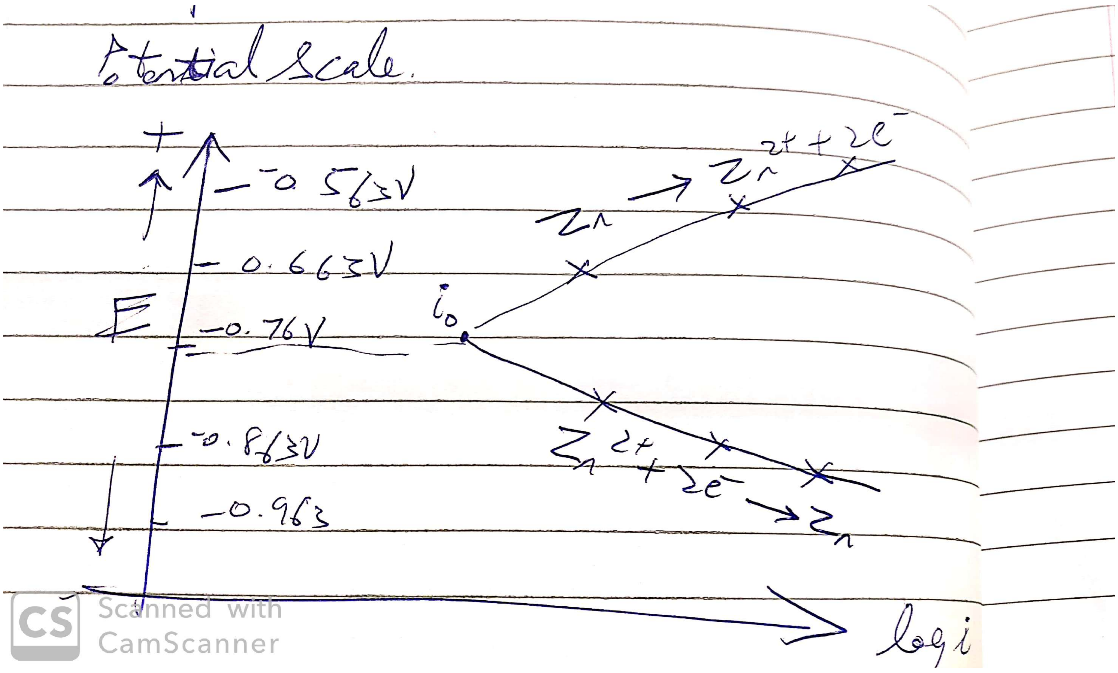
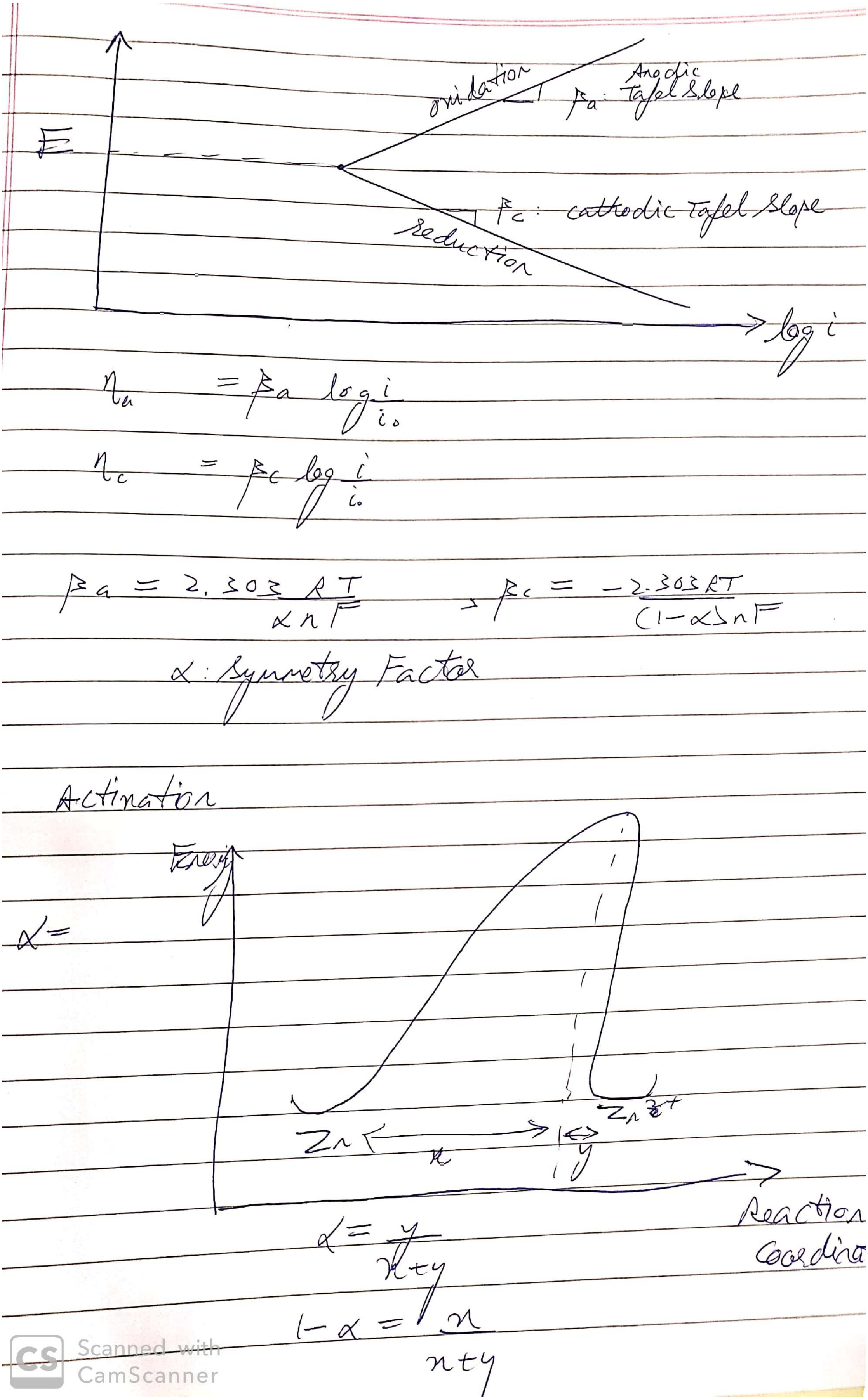

Potential Scale.
+
E
−0.563V
−0.663V
−0.76V
−0.863V
−0.963
$i_0$
$Zn \rightarrow Zn^{2+} + 2e^-$
$Zn^{2+} + 2e^- \rightarrow Zn$
log i

Figure 3.1: Net current Density


E
oxidation
Anodic $\beta_a$: Tafel Slope
$\beta_c$: cathodic Tafel Slope
reduction
log i

$$\eta_a = \beta_a \log \frac{i}{i_0}$$

$$\eta_c = \beta_c \log \frac{i}{i_0}$$

$$\beta_a = \frac{2.303\,RT}{\alpha n F}, \quad \beta_c = -\frac{2.303\,RT}{(1-\alpha) n F}$$

$\alpha$: Symmetry Factor

Activation

Energy
$\alpha =$
$Zn$
$x$
$y$
$Zn^{2+}$
Reaction Coordina

$$\alpha = \frac{y}{x+y}$$

$$1-\alpha = \frac{x}{x+y}$$

Figure 3.2: Overpotential

# 4 Lecture Feb 11

## §4.1 Kinetics

Thermodynamically we all are going towards death. Kinetics governs the rate at which we go towards death. Controlling Kinteics is key to longer lifetime. Same in Corrosion of metals.

### §4.1.i Evans Diagram

fig. 4.1 is one of the most important plots in Kinetics. Consider,

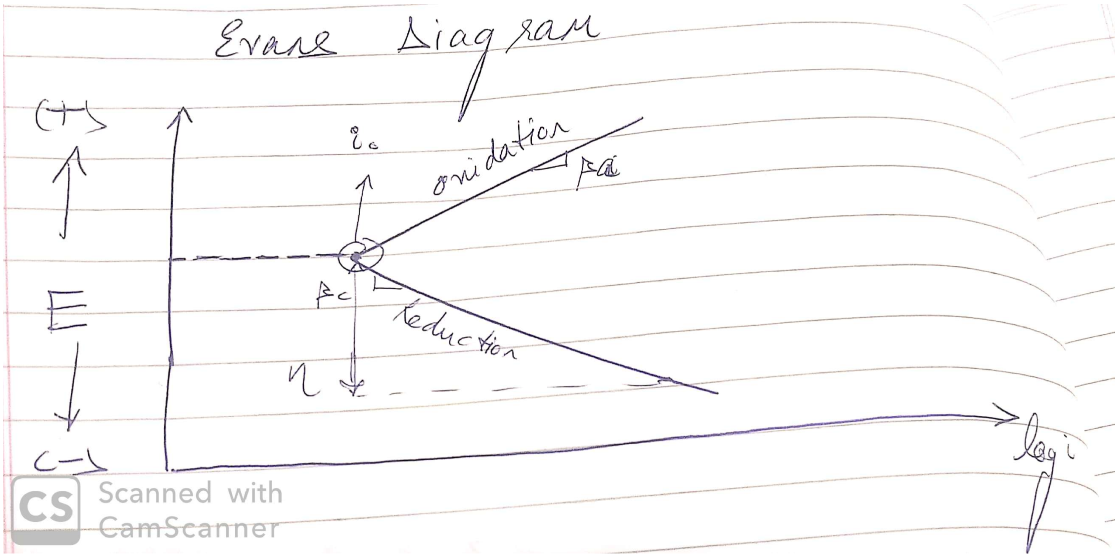


Figure 4.1: Evans Diagram

$$O_x + ne^- \iff R$$

This is characterized by $i_0, E_{\text{eqbm}}$. Away form equilibrium,

$$\eta := E_{\text{applied}} - E_{\text{equilibrium}}$$

where $\eta$ is called the overpotential. For $\eta < 0$,
For $\eta < 0$,
Tafel Equation: Recall the plot of energies with reaction coordinates as in fig. 3.2.

#### Bulter Volmer Equation

For net anodic current density $i_{net} = i_0 \left\{ \underbrace{e^{\frac{\alpha n F \eta}{RT}}}_{\text{anodic}} - \underbrace{e^{-\frac{(1-\alpha)nF\eta}{RT}}}_{cathodic} \right\}$.

When $\eta >> 0, i_{\text{net}} = i_0 e^{\frac{\alpha n F \eta}{RT}}$ For net cathodic current density (reverse) $i_net = i_0 \left\{ \underbrace{e^{\frac{\alpha n F \eta}{RT}}}_{\text{anodic}} - \underbrace{e^{-\frac{(1-\alpha)nF\eta}{RT}}}_{cathodic} \right\}$

When $\eta >> 0, i_{\text{net}} = i_0 e^{\frac{\alpha n F \eta}{RT}}$

**Tafel equations are approximate versions of Butler Volmer Equations**

### §4.1.ii Concentration Potential

Bulk mass transport governed by Ficks Laws of Diffusion.

$$\text{Fick's Law of Diffusion:} \quad J = -D\frac{\partial C}{\partial x}$$

Where $D$: Diffusion Coefficient (cm$^2$/s), $J$, flux (mol/cm$^2 \cdot s$), $C_B^{O_x}$: Bulk of oxide species.
At $t = 0$; $C_B^{O_x} = C_s^{O_x}$ Since there is no time for change in concentration at the surface from the bulk.
Apply negative overpotential, reaction goes forward and $[C_s^{O_x}]$ decreases. Assuming steady state and Linear concentration, we get

$$J = \frac{C_B^{O_x} - C_s^{O_x}}{\delta}$$

For a particularly negative $\eta$, the surface concentration depletes to 0 and $J \to J_{max}$ where

$$J_{max} = -D\frac{C_B^{O_x}}{\delta}$$

$$J_{max} = -\frac{|i_L|}{nF}$$

Here $i_L$ is called the **Limiting current density**. Relating by Faradys's Law,

$$\begin{aligned} -\frac{|i_L|}{nF} &= J_{max} = -D\frac{C_B^{O_x}}{\delta} \qquad \left[\because [C_s^{O_x}] = 0\right] \\ |i_L| &= D\frac{C_B^{O_x}}{\delta}nF \end{aligned}$$

# 5 Lecture Feb 13

## §5.1 Concentration Overpotential

We introduced **Concentration Overpotential** in the last class. The reaction is shown in fig. 5.1.

$$O_x + ne^- \longrightarrow R$$

We also found that at very highly negative overpotentials, the concentration at the surface depletes. This gives us the expression

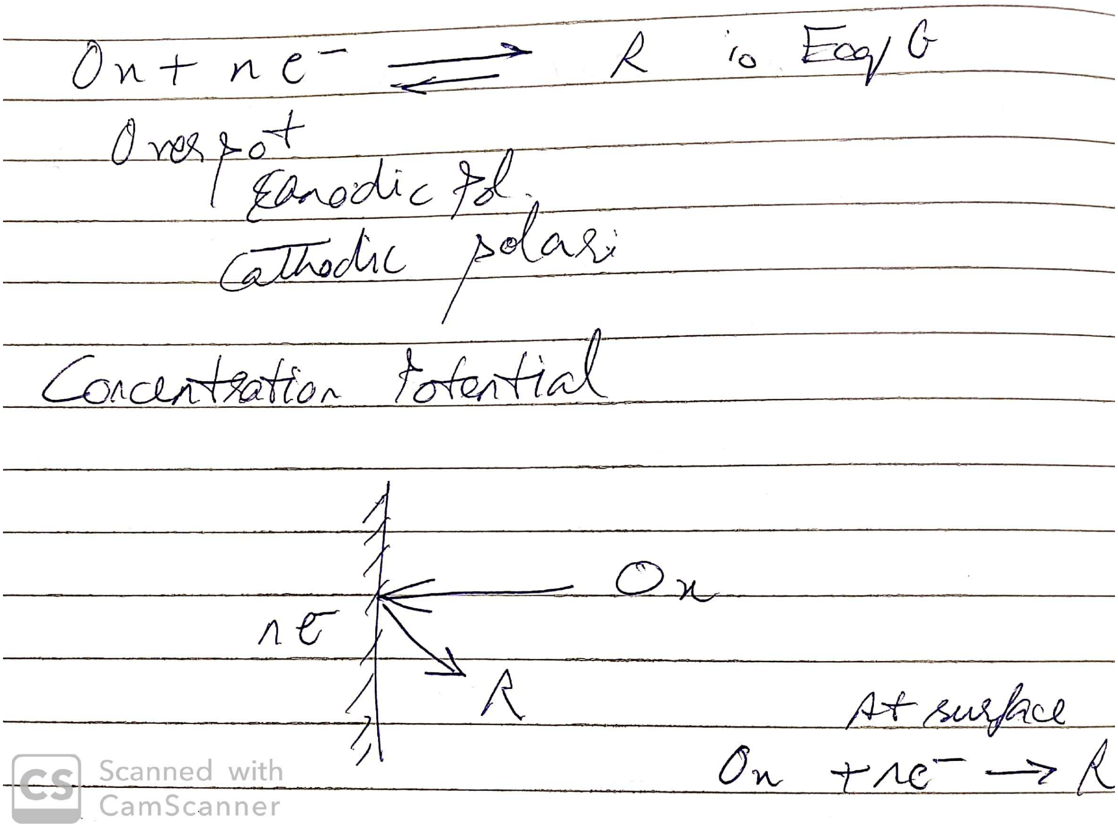

On + ne⁻ ⇌ R
io Eeq G
Over pot
anodic pol
cathodic polari
Concentration Potential
On
ne
R
At surface
On + ne⁻ → R

Figure 5.1: Concentration Overpotential

$$|i_L| = D\frac{C_B^{O_x}}{\delta}nF \tag{5.1}$$

This is has the effect of saturating current as seen in fig. 5.2

**Example 5.1.1**

In Water flowing through the pipes vs Water held stationary in a container. The Nernst diffusion layer thickness will be smaller in the pipe because of flow. No corrosion as the $\delta$ is small.

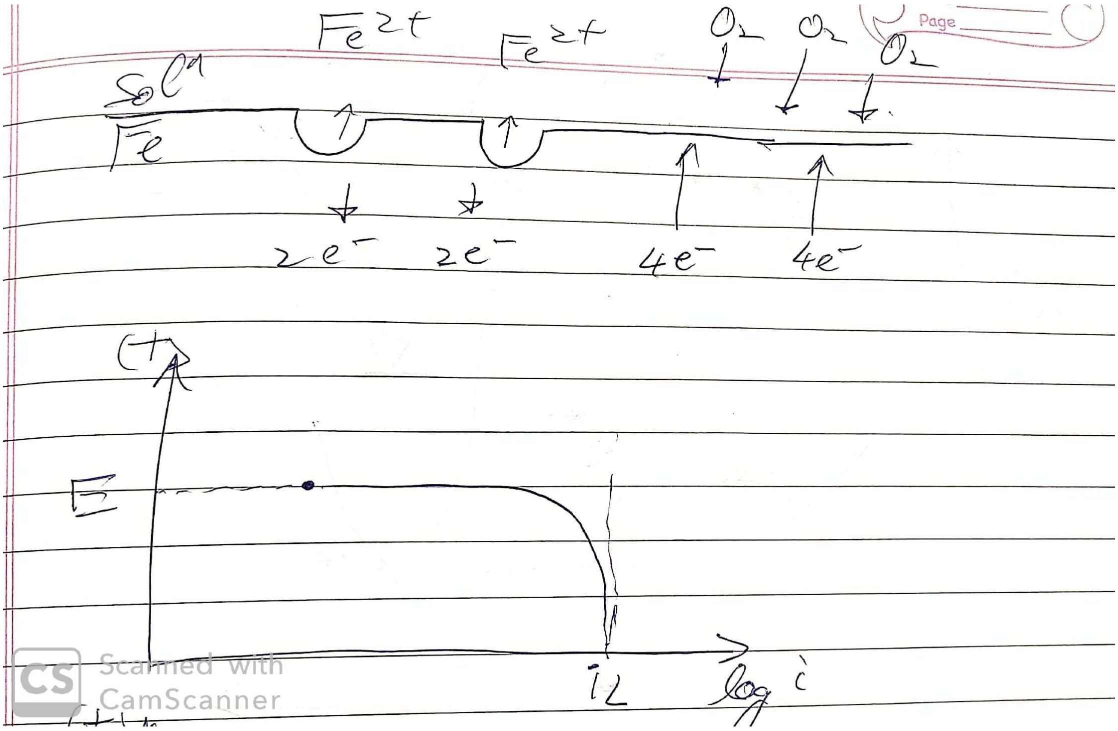

Fe$^{2+}$
Fe$^{2+}$
O$_2$
O$_2$
O$_2$
Sol$^n$
Fe
2e$^-$
2e$^-$
4e$^-$
4e$^-$
(+)
E
$i_L$
log i

Figure 5.2: Limiting Current

## §5.2 Combined Polarisation

$$\eta_T = \eta_{\text{Activation}}(\text{charge transfer}) + \eta_{\text{concentration}}$$
$$= \beta_C \log(\frac{i}{i_0}) + 2.303\frac{RT}{nF}\log(1 - \frac{i}{i_L})$$

This is seen in the Evans diagram as shown in fig. 5.3

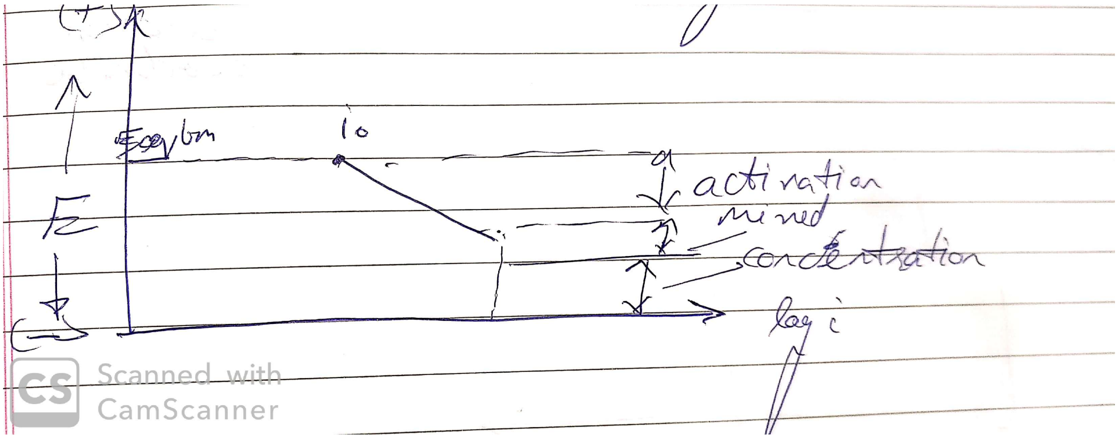

(+)
E
(−)
Eqlbm
$i_0$
activation
mixed
concentration
log i

Figure 5.3: Combined Polarisation

## §5.3 Mixed Potential Theory

$$M + nH^+ \longrightarrow M^{n+} + \frac{n}{2}H_2$$

**Question 5.3.1.** Why will there be mixed potential?

There are two equilibria:

1. $M^{n+} + ne^- \longleftrightarrow M$: $E_{M^{n+}/M}$; $i_{0M^{n+}/M}$
2. $2H^+ + 2e^- \longrightarrow H_2$: $E_{H^+/H}$; $i_{0H^+/H(M)}$

For the metal surface, the conductivity allows flow of electrons and hence gives equipotential: the potential on the surface will be between 0 and -5.44V, that is the $E^0$ of the two reactions.

### Mixed Potential Theory

Postulates of the Theory are:

1. Any electro-chemical reaction can be split into two or more partial reactions.
2. There is no net accumulation of charges.

The corresponding plot for the two reactions in the Evans Diagram is seen in fig. 5.4. The effective potential is in between the two $E_{red}$ as the point of intersection of the two straight lines: the rate of oxidation is equal to the rate of reduction.

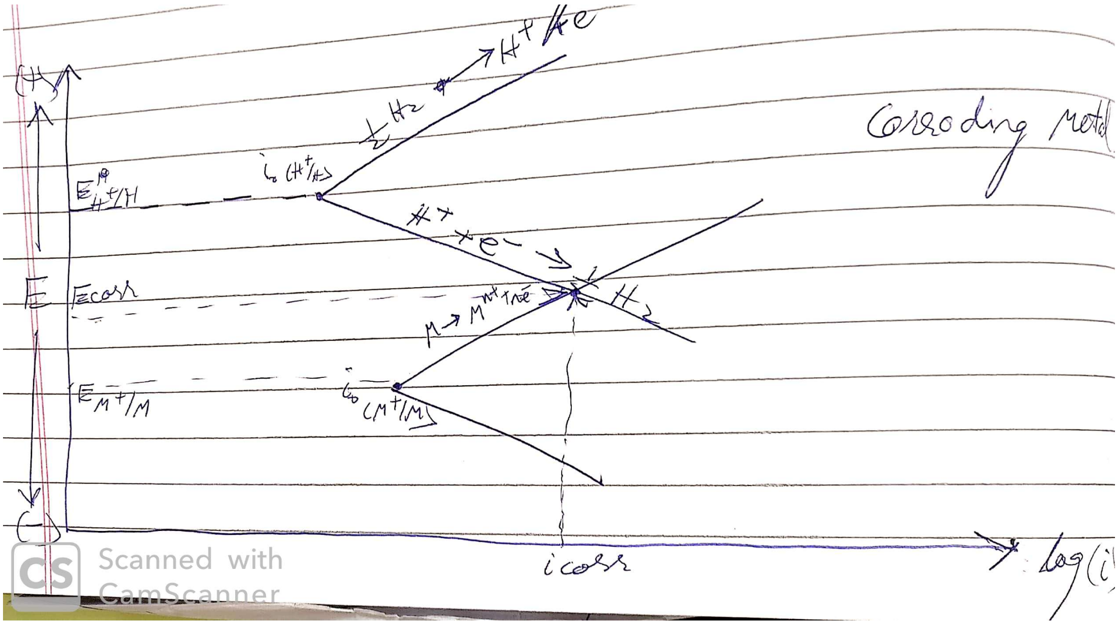

(+)
E
(-)
$E^0_{H^+/H}$
$i_0\,(H^+/H)$
$\frac{1}{2}H_2 \rightarrow H^+ + e$
Corroding metal
$H^+ + e^- \rightarrow \frac{1}{2}H_2$
Ecorr
$M \rightarrow M^{n+} + ne$
$E_{M^+/M}$
$i_0\,(M^+/M)$
icorr
log(i

Figure 5.4: Mixed Potential

**Question 5.3.2.** Why cant we coat Fe on zinc rod?

$$i_{0H^+/H(Zn)} = 10^{-9} A/cm^2$$
$$i_{0H^+/H(Fe)} = 10^-6 A/cm^2$$

Mix Theory:

$$\beta_\alpha(M)$$

Too much of hydrogen would be produced due to higher rate of reaction.

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