Calculation of uniform corrosion current density of iron in hydrochloric acid solutions based on the principle of maximum entropy production rate applied to literature data

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Abstract

In this paper a new procedure for the calculation of uniform corrosion current density is presented. The procedure is based on the following criterion: corrosion current and extent of anodic and cathodic areas are such that the entropy production rate is maximum. Experimental data for the reaction Fe(*s*) + HCl(*aq*) = $FeCl_2$(*aq*) + H_2(*g*), taken from literature, are used to test the new model. A comparison between corrosion current density calculated from weight loss measurements and current density calculated from polarization curves by means of the proposed model is carried out. The proposed model provides a better agreement, between Tafel line extrapolation and gravimetric measurement, in comparison to the mixed potential theory usually adopted.

Keywords: corrosion current density, uniform corrosion, entropy production rate, exchange current density, Tafel slope.

Introduction

Corrosion current density is an important project specification that must be considered when designing an artefact exposed to aggressive conditions. The corrosion of a component is a complex phenomenon, with several causes at the same time, quite often difficult to rationalize. Nonetheless, it is desirable to provide rational schemes that allow to understand the phenomenon and to make reliable estimation of design specifications. Made this premise, in this paper we will face the problem of the calculation of uniform corrosion current density in a new way, relying on a model presented in a previous paper that is based on the thermodynamics of irreversible phenomena. [1]

In the present paper, we consider experimental data from literature on the corrosion of iron in hydrochloric acid solutions to validate the proposed formula. In particular, we will compare the corrosion current density calculated from weight loss experiments (or evolved hydrogen or dissolved metal) with the values obtained from polarization curves according both to the usual extrapolation method and to the proposed model.

The newly proposed model is based on the following assumptions:

1. In steady state conditions the system evolves in such a way to produce entropy at the maximum rate. [2]
2. The anodic surface areas are different from the cathodic surface areas ($A_a \neq A_c$) and hence the electrode current densities are different ($i_a \neq i_c$). The relationship $E_a + \eta_a(i) = E_c + \eta_c(i) = E_{corr}$ is always valid, but the anodic and cathodic current densities are different and hence the equation that we have solve is $E_a + \eta_a(i_a) = E_c + \eta_c(i_c) = E_{corr}$.
3. The corrosion current (I_{corr}) and the extent of anodic and cathodic surface areas are such as to maximize the entropy production rate of the corrosion process. It is a problem of extrema calculation with the constraint of constancy of the Gibbs free energy change of the corrosion reaction (Lagrange multipliers method).
4. As in the case of the Wagner and Traud theory, it is necessary to know the exchange current densities, the Tafel slopes of the electrode processes, and the driving force of the reaction. However, we exclude that the surface is at the same time anodic and cathodic [3]. On the other hand, we assume that there are anodic sites different from cathodic sites moving randomly on the metal surface. [4,5,6] For the details of the model and the meaning of the symbols used in the paper see ref. [1].

The formula for corrosion current density has a simple form, if we can neglect ohmic drops and concentration polarizations. The new formula for corrosion current density is the following

$$\ln\frac{I_{corr}}{A}=\frac{1}{\alpha_c+\alpha_a}\ln\left[\left(\omega i_{0,c}\right)^{\alpha_a}\left((1-\omega)i_{0,a}\right)^{\alpha_c}\right]-\frac{\alpha_c\alpha_a}{\alpha_c+\alpha_a}\frac{\Delta G}{RT}$$

where $A_a=(1-\omega)$, $A_c=\omega A$ and $\omega=\frac{\alpha_a}{\alpha_c+\alpha_a}$, different from the formula that is generally utilized

$$\ln\frac{I_{corr}}{A}=\frac{1}{\alpha_c+\alpha_a}ln\left[\left(i_{0,c}\right)^{\alpha_a}\left(i_{0,a}\right)^{\alpha_c}\right]-\frac{\alpha_c\alpha_a}{\alpha_c+\alpha_a}\frac{\Delta G}{RT}$$

Weight loss experiments are of long duration, hours or days, and along the test there can be a significant change of concentration of species in solution, together with the electromotive force of reaction, and hence a change in the corrosion rate. On the other hand, polarization experiments are usually, not always, much shorter in duration and, as in the case of gravimetric experiments, the solution composition changes during the experiment. Thus, it is important to know the volume of solution, the electrode surface area, the presence of any electrode pre-treatment in order to carry out a material balance and to calculate, by means of Nernst's equation, the potential of electrode reactions.

The examples from literature show that current densities calculated from weight loss measurements (or evolved hydrogen or dissolved metal) are in most cases very different from those calculated from polarization curves. In the following we will consider some experimental data from literature about the corrosion of iron in 1 M hydrochloric acid and we will reconsider them according to the proposed model.

Numerical examples

The calculation of electromotive force of the reaction Fe(*s*) + HCl(*aq*) = $FeCl_2$(*aq*) + H_2(*g*), with Nernst's equation, is carried out by considering as ideal the behavior of species in solution. Standard electrode potentials and temperature coefficients are taken from ref. [7].

In order to apply the proposed formula and the conventional one, we must extrapolate the exchange current densities and the Tafel slopes for the two electrode processes, hydrogen evolution and iron dissolution, according to the scheme shown in Figure 1. In order to do that, we must calculate $E_{H+/H2}$ and $E_{Fe(II)/Fe}$.

The effect of complexation between bivalent iron cation and chloride ion has been considered but the calculated corrosion current density are practically unaffected (see supplementary materials). In the next pages, the complexes $FeCl^+$ and $FeCl_2$ have not been considered for the calculation.

In the following, we discuss three examples, by analyzing the data from literature. Where necessary, data from literature graphs were extracted by using the Digitizer tool in Origin software. For sake of convenience numerical values of physical quantities of experiments are summarized in Table 4 and Table 5 and also the potentials of SCE are reported.(1)

Example 1

Let's consider the article of Chetouani et al. [8] By using the data of gravimetric test we calculate the corrosion current density reported in Table 6. Gravimetric tests were carried out in air.(2) Polarization curves were recorded at a scan rate of 0.33 mV s^{-1}. Before recording the polarization curves the solution was deaerated and magnetically stirred for 30 min with nitrogen. The iron electrode, kept at its open circuit value for 30 min under nitrogen atmosphere(2), was polarized at -800 mV/SCE for 10 min. The potential of the electrode was then swept. Gas bubbling was maintained throughout the experiments. The volume of solution is 100 cm^3.(2) Curves are corrected for IR drop.(2) In the last three columns of Table 5, the values of corrosion potential, corrosion current density and slope of Tafel line for hydrogen evolution, as calculated by Chetouani et al., are reported.

By using the Nernst's equation, we obtain $E_{H+/H2}$ = -241 mV/SCE for hydrogen gas evolution in [HCl] $\cong$ 1 M at 298 K.(3) The kinetic data for hydrogen evolution provided by Chetouani et al. allow us to write the equation of Tafel line (i in mA cm^{-2} and E in mV)

$$\log_{10} i = \log_{10}(0.936) - (E + 500)/230$$

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(1) SCE potentials vs NHE: 241.2 mV at 25°C, 237.9 at 30°C and 234.4 at 35°C.

(2) Hammouti private communication.

(3) By considering 30 min at open circuit potential the mass of iron dissolved from the electrode is 0.405 mg cm^2 h^{-1} × 1 cm^2 × 0.5 h = 0.2025 mg. Assuming 100 cm^3 of electrolytic solution we have $[FeCl_2] = 3.626\times10^{-5}$ M. If the weight loss rate is 1.019 mg cm^2 h^{-1}, as in the paper for 1 h gravimetric test, the mass dissolved is 0.5085 mg and the concentration is $[FeCl_2] = 9.123\times10^{-5}$ M. The reduction potential of iron at 298 K is in between -813 and -801 mV/SCE. Moreover, the potential for hydrogen evolution is -241 mV/SCE.

The slope of Tafel line is b_c = 230 mV dec^{-1} ($\alpha = 2.303\times10^3(RT/b_czF) = 0.129$) and the exchange current density value is $i_0 = 6.932\times10^{-2}$ mA cm^{-2} (calculated at –241 mV/SCE).

In order to determine the Tafel line for iron dissolution we subtract the current density for hydrogen evolution from the overall current density. From data in Table 1 we can draw the Tafel line for iron (Figure 2). The equation of curve, fitted on the range from –420 to –260 mV/SCE, is

$$i = 1770.1e^{-1.587\times10^{-2}E}$$

(i in mA/cm^2 and E in mV/SCE) and the correlation coefficient $R^2 = 0.9985$.

The slope of Tafel line is b_a = ln(10)/0.015868 = 145 mV dec^{-1} ($\alpha = 0.204$) and the exchange current density must be calculated by extrapolating the Tafel line to the potential $E_{Fe(II)/Fe}$, obtained by Nernst's equation with the composition of the solution that corresponds to the potential of about –260 mV/SCE.

The concentration of species in solution changes during scan. By integrating the charge for iron dissolution we calculate the concentration of species at each potential during the polarization scan. By integrating the current from –800 to –260 mV/SCE (0.33 mV s^{-1}) we obtain about Q = 6.3 C equivalent to 1.83 mg of iron. This quantity summed to the amount of iron dissolved in the 30 min of electrode pre-conditioning (0.5085 mg) makes 2.334 mg of iron in 100 cm^3 of solution and the corresponding concentration [$FeCl_2$] = 0.00042 M. The Nernst's equation gives $E_{Fe(II)/Fe}$ = –781 mV/SCE at 298 K. The Tafel line for iron at –781 mV has a value of (exchange current density) equal to i_0 = 0.007333 mA cm^{-2}. By knowing the kinetic data and the driving force of the cell reaction in gravimetric test (512 mV) we can calculate the values reported in Table 6.

Example 2

The second paper that we consider is written by Emregul and Atakol [9]. The 1 M HCl electrolyte solution was prepared from analytical grade HCl and double distilled water. All tests have been performed in deaerated solutions at 30°C. The samples used were of 99.9% pure iron. The working electrode was in the form of a rod machined into a cylindrical form and embedded in epoxy resin leaving an open surface area of 0.7 cm^2. Weight loss experiments were performed with 1 cm^2 coupons of the same material.

Weight loss of iron 99.9% samples was determined within a period of 7 days. From the graph of Fig. 2 in the paper we obtain a slope of 0.238 mg cm^{-2} h^{-1} corresponding to i_{corr} = 0.229 mA cm^{-2}.

Electrochemical measurements were conducted in a conventional three-electrode cylindrical glass and thermostated cell. A platinum disk as counter electrode and standard calomel electrode (SCE) as the reference electrode have been used in the electrochemical studies. After steady state was reached (60 min) the electrode was polarized from the open circuit potential (E_{ocp}) in the cathodic direction at a scan rate of 0.5 mV s^{-1}. After the cathodic scan, the electrode was taken out of the cell and pretreated, then the anodic polarization curve was obtained by polarizing from E_{ocp} in the anodic direction. Curves are not corrected for IR drop. The anodic scan starts at open circuit potential after a pre-conditioning period of 60 minutes, during which iron dissolves. After 60 min of free corrosion from 0.7 cm^2 surface area in solution there are 0.167 mg of iron. The volumes of solutions both for weight loss and polarization tests are not specified in the paper.[(4)]

Polarization data in the range from –675 to –625 mV/SCE (Figure 3), extracted from Fig. 4 of the paper, allow us to calculate the Tafel line equation for hydrogen evolution (i in mA cm^{-2}, E in mV/SCE)

$$i = 2.038 \times 10^{-3} e^{-0.010403E}$$

and the correlation coefficient $R^2 = 0.9985$.

The reduction potential of hydrogen in [HCl] ≅ 1 M at 30°C is $E_{H+/H2} = -238$ mV/SCE (neglecting the iron dissolved during the 60 minutes of pre-conditioning). Thus, the exchange current density of the cathodic process is $i_0 = 2.424\times10^{-2}$ mA cm^{-2} and the Tafel slope $b_c = 221$ mV dec^{-1} ($\alpha = 0.136$) ([5])

By summing the hydrogen evolution current density to the anodic branch of the polarization curve we obtain the corrosion current density for iron dissolution and, after interpolation in the range from –512.5 to –450 mV/SCE, the corresponding Tafel equation is (Figure 3)

$$i = 200549.5 e^{2.279\times10^{-2}E}$$

with correlation coefficient $R^2 = 0.9969$.

The slope of the Tafel line for iron dissolution is $b_a = 101$ mV dec^{-1} ($\alpha = 0.298$).

Let' calculate the amount of iron dissolved during scan from –537.5 to –450 mV/SCE. In this potential range we account for Q = 0.64 C equivalent to 0.185 mg of iron that we sum to the

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[4] We assume 100 cm^3 both for gravimetric and polarization tests.

[5] The corrosion potential that we obtain from Fig. 2 in the paper is around E_{corr} = -546 mV/SCE and it doesn't correspond to the value reported by Emregul (E_{corr} = -538 mV/SCE at i_{corr} = 0.475 mA cm^{-2}).

amount of iron dissolved during the 60 minutes of pre-conditioning.(6) The solution has an iron chloride concentration of 6.3×10^{-5} M and the corresponding potential at 303 K is $E_{Fe(II)/Fe}$ = –804 mV/SCE. The exchange current density for iron dissolution is $i_0 = 2.217\times10^{-3}$ mA cm^{-2}.

Finally, by means of the kinetic data and the driving force of cell reaction in gravimetric test (504 mV) we calculate the values for corrosion current densities (Table 6).

Example 3

Finally let's analyze the third paper written by Chakib et al. [10]. Experiments were carried out in 1.0 M HCl solution prepared by dilution of analytical grade HCl of predetermined molarity with double distilled water. The volume of the solution used in each experiment was 100 cm^3. Iron purity was 99.548 % and coupons cut into 1.5 × 1.5 × 0.05 cm^3 size were used for gravimetric measurements (S = 4.8 cm^2).

The weight loss measurements were performed in a glass vessel containing 100 cm^3 of 1 M HCl solution at 35°C for 6 h of immersion time without bubbling. The steel specimens were withdrawn, rinsed with doubly distilled water, washed with acetone, dried and weighed. The experiments were done in triplicate and the average value of the weight loss was noted.

The electrochemical study was carried in a three electrode cell. A saturated calomel electrode (SCE) and platinum electrode were used as reference and auxiliary electrodes, respectively. Anodic and cathodic potentiodynamic polarization curves were plotted at a polarization scan rate of 0.5 mV s^{-1}. The solution test is there after de-aerated by bubbling nitrogen. Specimens with exposed surface areas 1 cm^2 were used as working electrode for polarization. The potential was stabilized at free potential during 30 min, before conducting any run. During this time coupons corroded (m = 0.82 × 1 × 0.5 = 0.41 mg of iron) and part of the acid consumed. Concentration of species at the beginning of scan are $[FeCl_2] = 7.34\times10^{-5}$ M and [HCl] ≅ 1 M. The polarization curves are obtained from −800 mV to −200 mV/SCE at 35°C in 1200 s. Potential scan starts at –800 mV/SCE and the iron reduction potential at that concentration is –800.1 mV/SCE.

The data in the interval from –725 to –625 mV/SCE of cathodic branch of polarization allows to determine the Tafel line equation for hydrogen evolution

$$i = 2.6102 \times 10^{-2} e^{-9.594\times10^{-3}E}$$

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[6] We assume the pre-conditioning step even tough not explicitly stated for the anodic branch.

and a correlation coefficient equal to $R^2 = 1.0000$.

By using such equation, we calculate the corrosion current density for iron dissolution and the corresponding Tafel line equation (Figure 4), linearly interpolated in the range from –400 to –325 mV/SCE, is

$$i = 4100.0e^{1.764\times 10^{-2}E}$$

and a correlation coefficient equal to $R^2 = 0.9993$.

At the start of scan, the concentration of acid is [HCl] ≅ 1 M, and in the range from –725 to –625 mV/SCE it is little less. The potential for hydrogen calculated with Nernst's equation is $E_{H+/H2} = -234.4$ mV/SCE. With this value in the Tafel line equation, we have $b_c = 240$ mV dec^{-1} ($\alpha = 0.123$) and $i_0 = 0.24652$ mA cm^{-2}.

The amount of iron in solution when the potential scan is at –300 mV/SCE is given by the iron initially present (0.41 mg) plus the iron that dissolves during polarization. By considering the scan rate and the surface area of electrode and the Tafel equation for iron dissolution we obtain that from –800 to –300 mV/SCE circulates a charge of 1.15 C equivalent to 0.33 mg or iron. Therefore, there are 0.41 + 0.33 = 0.74 mg of iron corresponding to $[FeCl_2] = 0.000133$ M. At this concentration the potential for iron reduction is $E_{Fe(II)/Fe} = -792.2$ mV/SCE.

The parameters of Tafel line for iron dissolution are $b_a = 131$ mV dec^{-1} ($\alpha = 0.226$) and $i_0 = 3.498\times 10^{-3}$ mA cm^{-2}.

Finally by using the concentration of the gravimetric test and the kinetic parameters calculated from polarization curves we obtain the corrosion current densities according both to the mixed potential theory and the newly proposed model (Table 6).

Conclusions

In the paper we describe the application of a formula, deduced from the maximum entropy production rate, for the calculation of uniform corrosion current density. The formula has been obtained under some simplifying assumptions, that is negligible ohmic drops and concentration polarizations. Under such hypotheses, entropy production is due only to the electrode processes occurring on anodes and cathodes randomly distributed over the metal surface. Individual half-reactions occur onto these microscopically separated area. In all considered cases, the agreement between the current density calculated from gravimetric data and that one obtained with the new formula is significantly better than that one obtained with the usual Tafel line extrapolation approach.

Appendix – Numerical values extracted from polarization curves

Table 1. Current density vs potential: total, hydrogen evolution and iron dissolution (Chetouani).

E (mV/SCE)	i_{total} (mA cm^{-2})	$i_{hydrogen}$ (mA cm^{-2})	i_{iron} (mA cm^{-2})
-800	18.86	18.86	
-780	15.44	15.44	
-760	12.64	12.64	
-740	10.35	10.35	
-720	8.47	8.47	
-700	6.93	6.93	
-680	5.67	5.67	
-660	4.64	4.64	
-640	3.80	3.80	
-620	3.11	3.11	
-600	2.55	2.55	
-580	2.08	2.08	
-560	1.71	1.71	
-540	1.40	1.40	
-520			
-500			
-480			
-460	0.66	0.63	
-440	1.05	0.51	
-420	1.84	0.42	2.26
-400	2.59	0.34	2.94
-380	4.07	0.28	4.35
-360	6.05	0.23	6.28
-340	7.67	0.19	7.86
-320	10.83	0.15	10.98
-300	14.87	0.13	15.00
-280	20.98	0.10	21.09
-260	28.07	0.08	28.16

-240	37.56		
-220	48.94		
-200	60.48		
-180	74.74		
-160	87.60		
-140	100.00		
-120	120.35		
-100	123.58		

Table 2. Current density vs potential: total, hydrogen evolution and iron dissolution (Emregul).

E (mV/SCE)	I (mA cm^{-2})	$i_{hydrogen}$ (mA cm^{-2})	i_{iron} (mA cm^{-2})
-675	2.30	2.28	
-650	1.74	1.76	
-625	1.37	1.36	
-600	1.00	1.05	
-500			
-537.5	0.26	0.55	0.81
-525	0.64	0.48	1.12
-512.5	1.23	0.42	1.65
-500	1.86	0.37	2.23
-487.5	2.80	0.32	3.13
-475	3.82	0.29	4.11
-462.5	5.03	0.25	5.28
-450	6.62	0.22	6.84

Table 3. Current density vs potential: total, hydrogen evolution and iron dissolution (Chakib).

E (mV/SCE)	i (mA cm^{-2})	$i_{hydrogen}$ (mA cm^{-2})	i_{iron} (mA cm^{-2})
-800	51.09		
-775	42.17		

-750	33.18	34.68	
-725	27.38	27.29	
-700	21.54	21.47	
-675	16.95	16.89	
-650	13.34	13.29	
-625	10.49	10.45	
-600	7.87	8.23	
-575	5.36	6.47	
-550	3.83	5.09	
-525	2.37	4.01	
-500	1.27	3.15	
-450		1.95	
-425	1.00	1.53	
-400	2.37	1.21	3.58
-375	4.42	0.95	5.37
-350	7.87	0.75	8.61
-325	12.71	0.59	13.30
-300	19.57		
-275	28.73		
-250	40.19		

Tables

Table 4. Weight loss experiment data.

	Fe	$[HCl]_0$	Atmosphere	S	T	t	V	W	$[HCl]_t$	$[FeCl_2]_t$	E_{cell} at t
	%	M		cm^2	°C	h	cm^3	$mg\ cm^{-2}\ h^{-1}$	M	M	mV
Chetouani	99.5	1	Air	8.4	25	6	100	0.405	0.99270	0.00370	512
Emregul	99.9	1	Nitrogen	1.0	30	168	100 (7)	0.238	0.98567	0.00716	504
Chakib	99.6	1	Air	4.8	35	6	100	0.820	0.99154	0.00423	512

Table 5. Potentiodynamic polarization data.

	$[HCl]_0$	Atmosphere	S	T	V	v	E_{corr}	i_{corr}	b_c
	M		cm^2	°C	cm^3	$mV\ s^{-1}$	mV/SCE	$mA\ cm^{-2}$	$mV\ dec^{-1}$
Chetouani	1	Nitrogen	1.0	25	100 (2)	0.33	-500	0.936	230
Emregul	1	Nitrogen	0.7	30	100 (7)	0.50	-538 (2)	0.475	-
Chakib	1	Nitrogen	1.0	35	100	0.50	-454	1.386	164

Table 6. Corrosion current densities ($mA\ cm^{-2}$).

	Weight loss	Paper	Wagner and Traud	Bestetti
Chetouani	0.389	0.936	0.677	0.348
Emregul	0.229	0.475	0.421	0.226
Chakib	0.787	1.386	1.319	0.690

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[7] Our assumption.

Figures

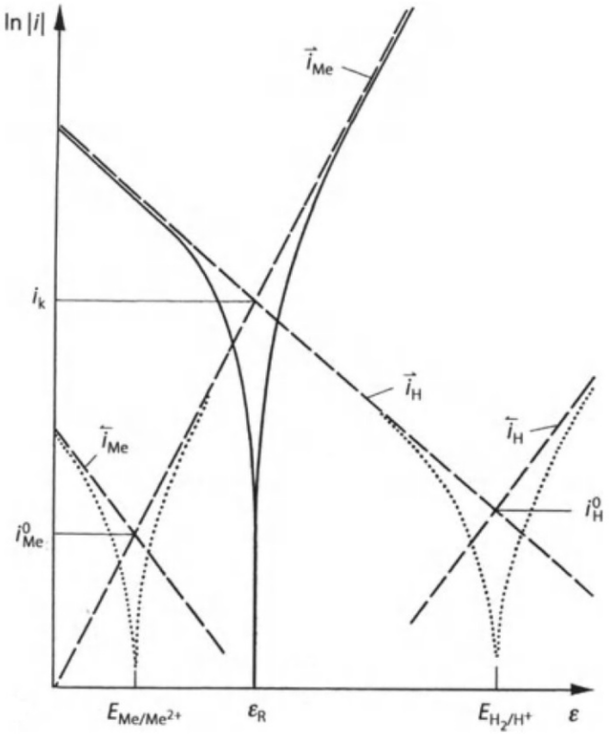

Figure 1. Current density – potential curves (p.121, ref. [3]).

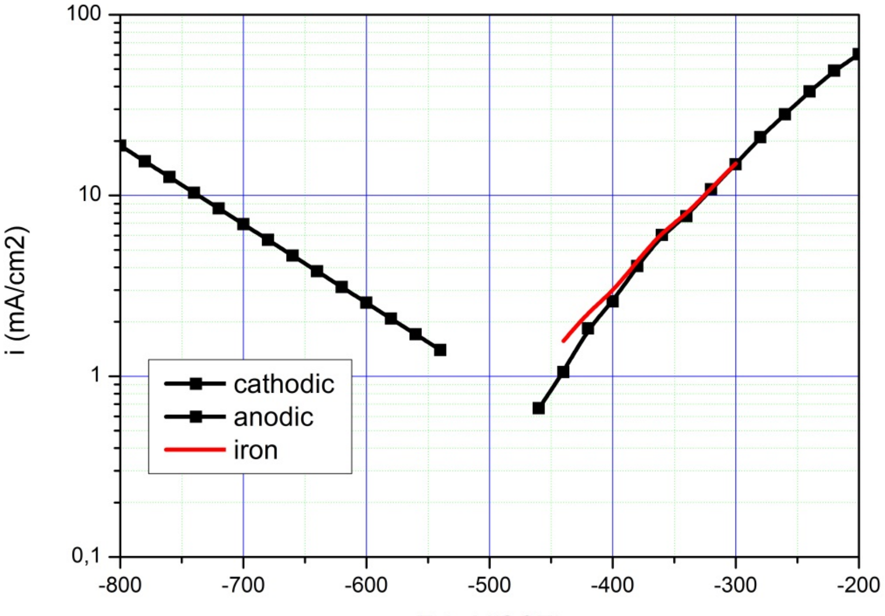

Figure 2. Potentiodynamic polarization curve and Tafel line for iron dissolution in 1 M HCl at 25°C (Chetouani 2003).

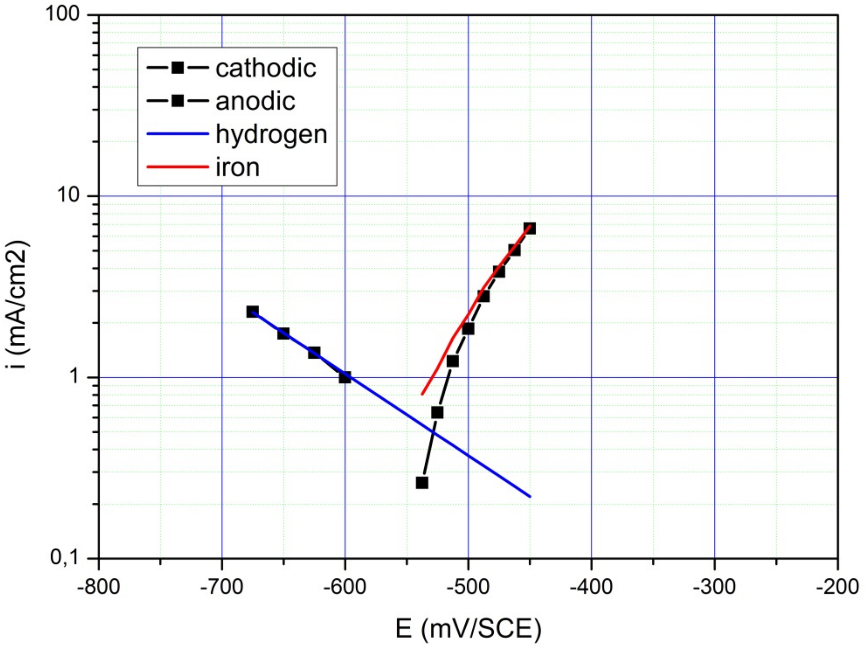

Figure 3. Potentiodynamic polarization curve and Tafel line for iron dissolution in 1 M HCl at 30°C (Emregul 2004).

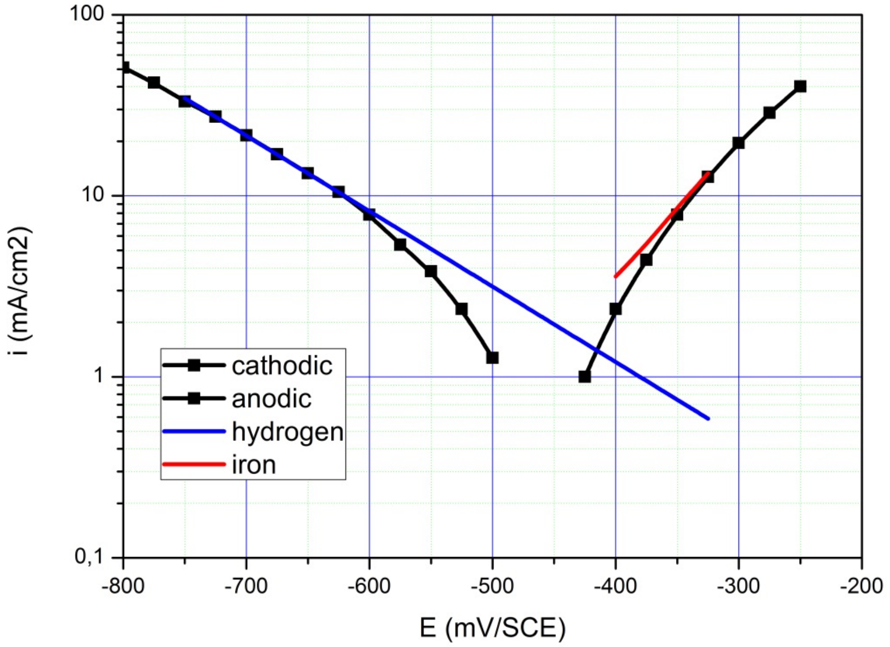

Figure 4. Potentiodynamic polarization curve and Tafel lines for hydrogen evolution and iron dissolution in 1 M HCl at 35°C (Chakib 2016).

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