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Investigation of corrosion rates with an electrochemical measuring cell in relation to the W_0-factor according to DIN 50929-3 and the concentration of dissolved organic carbon (DOC)

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The W_0-factor is a common metric to express the aggressivity of water against unalloyed iron. It accounts for multiple parameters, called rating numbers, which represent the concentration of chloride, sulfate and calcium ions, the pH-value and the acid capacity of the immersion medium. The use of ranges in the physical quantities metric rating number, however, allows for unaccounted variations in the immersion medium composition, i.e., slightly different immersion media may produce the same W_0-factor. To verify the results, the experiments were also carried out with natural waters as immersion media and their W_0-factors were determined. In addition, the DOC content, which is not included in the W_0-factor, has an impact on the corrosive character of the immersion medium. In order to account for the influence of the DOC content in the measurements, multiple solutions with different DOC concentrations were investigated. The results show that a higher DOC content (based on humic substances) in the observed solutions causes a reduction in the corrosion rate. This result agrees with observations of nature.

1 Introduction

The costs of damages caused by corrosion in industrial countries is in the order of 2–4% of the respective gross domestic product each year [1,2]. The corrosion rate is defined as the material removal per year (μm/a). Deducing the corrosion rate for new building constructions or hydraulic steel structures based on the chemical composition of the water it allows a forecast of the corrosion and, in turn, its long-term stability. Hence, structure components can be better sized and suitable corrosion protection measures can be chosen.

The corrosion process of steel sheet piles depends on a variety of factors, such as the water chemistry of the immersion media, the water depth, the water velocity, the temperature, the oxygen content, the lifetime, and the type of corrosion (e.g., surface corrosion or pitting corrosion).

The W_0-factor can be used to estimate the corrosion of unalloyed iron in the underwater zone, according to the German standard DIN 50929-3 [3]. In natural systems, the W_0-factor of different waters ranges from −11 to 0 for seawater to fresh water, respectively. The W_0-factor is defined as a sum of rating numbers (Equation (1)):

$$W_0 = N_1 + N_3 + N_4 + N_5 + N_6 + \frac{N_3}{N_4} \quad (1)$$

The rating numbers reflect the following parameters of the immersion medium:

- N_1 = kind of water (see below);
- N_3 = concentration of chloride and sulfate (mol/m^3);
- N_4 = acid capacity ($Ks_{4,3}$; mol/m^3);
- N_5 = concentration of calcium (mol/m^3);
- N_6 = pH-value.

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The kind of water (N_1) reflects seawater ($N_1 = -5$), shore of a lake ($N_1 = -3$), running ($N_1 = 0$), or stagnant ($N_1 = -1$) water. The ranges of the other parameters and the resulting values are given in Table 1. The table excludes N_2 and N_7 which are part of the German standard [3]. They only will be used for other calculations within this German standard. Furthermore, the W_0-factor is only valid for unalloyed iron.

The higher the chloride and sulfate concentration (N_3), the smaller is the W_0-factor. This is due to chloride ions which, according to theory, accumulate inside the rust layers. Thus, local corrosion can be strongly accelerated [4].

The higher the acid capacity (N_4), the higher is the probability for calcium-carbonate precipitation. The higher the acid capacity, the larger is the W_0-factor.

Thermodynamically, the calcium-carbonate equilibrium is described in the saturation index (SI) [5]. In case of $SI > 0$, a protective layer of corrosion-inhibiting residues and calcium carbonate can be formed on the material [6,7]. The calcium-carbonate-layer can reduce the corrosion current of (unalloyed) iron approximately by a factor of five [8]. Thus, with a higher concentration of calcium (N_5), the W_0-factor usually becomes larger.

The corrosion of unalloyed steel is also highly dependent on the pH-value of the water (N_6). The corrosion rate approaches nearly zero in alkaline media. In contrast, the corrosion rate increases quickly under acidic conditions, e.g., at a pH-value of about 4 [9]. As a consequence, the W_0-factor is larger in alkaline media.

Overall, it can be concluded that the smaller the W_0-factor, the more corrosive the immersion medium for unalloyed iron.

Table 1. Ranges of the parameters and resulting values

N_3	$c(Cl^-) + 2^*c(SO_4^{2-})$ (mol/m^3)	Value
	<1	0
	>1 to 5	−2
	>5 to 25	−4
	>25 to 100	−6
	>100 to 300	−7
	>300	−8
N_4	Acid capacity ($K_{S4,3}$) (mol/m^3)	
	<1	1
	1 to 2	2
	>2 to 4	3
	>4 to 6	4
	>6	5
N_5	$c(Ca^{2+})$ (mol/m^3)	
	<0.5	−1
	0.5 to 2	0
	>2 to 8	1
	>8	2
N_6	pH-value	
	<5.5	−3
	5.5 to 6.5	−2
	>6.5 to 7.0	−1
	>7.0 to 7.5	0
	>7.5	1

From Table 1, it is obvious that the physical quantities which underlie the rating numbers are defined in intervals within the rating number value remains constant. Therefore, it is possible that two different immersion media are assigned the same W_0-factor, although the specific chemical composition differs slightly. Thus, an immersion medium with a larger W_0-factor can potentially be more corrosive than a solution with a smaller W_0-factor.

The W_0-factor is not considering oxygen content, temperature, and organic substances of the immersion medium, which all have an impact on the corrosion.

In the case of organic content, the dissolved organic carbon (DOC) concentration can be used as an impact factor for the corrosivity.

The DOC content is the fraction of the organic matter in a solution, with a size of less than 0.45 μm [10]. Organic compounds can have an inhibiting effect on the corrosion due to their electrostatic and steric properties [11,12]. The carboxyl groups of organic compounds can act as an electron acceptor or an electron donor. A special group within the DOCs are the humic substances and the humic acids, which are also carboxylized. As a consequence, they also can act as a corrosion inhibitor.

The aim of this work was to:

1) evaluate how good the W_0-factor reflects the corrosivity of different solutions in nature and in experiments in the laboratory,
2) investigate if the experiments with model solutions lead to the same corrosion rates as the natural waters with the same W_0-factor,
3) study the effect of DOC on the corrosion rate with water from Lake Hohloh as source for the DOC,
4) investigate if a DOC parameter should be included into the W_0-factor to fit the measured corrosion rates in nature better.

2 Materials and methods/experimental

2.1 Mild steel sample

In the experiments for this work, one mild steel sample was used as laboratory reference. The data for the measurements in nature came from different places in Germany which were taken in the context of a European research project [13] with different mild steels. The chemical composition of the mild steel sample used in the laboratory experiments is shown in Table 2.

2.2 Laboratory test procedure

Corrosion data were obtained using a corrosion measuring cell (CMC) (Sensortechnik Meinsberg, KMZ 5) with a volume of 600 mL at constant temperature of 20 ± 0.1 °C.

The preparation of the samples and details of the used electrochemical corrosion measuring cell can be found in Ref. [14]. The experiments were performed in accordance with ASTM G 5 [15].

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Table 2. Chemical composition of the used mild steel sample (mass percent of minor components; balance iron)

Sample	C	Si	Mn	P	S	Cr	Ni	Cu
	0.17	0.2	1.4	0.02	0.02	0.04	0.03	0.05

The immersion media (Table 3) were prepared and filled into the CMC as well as into the Haber-Luggin capillary and the salt bridge.

A potential of −600 mV was applied and changed in 0.1 mV/s-steps to a value of −200 mV while measuring the current. According to DIN EN ISO 17475 [16], a potential change of (not more than) 0.17 mV/s is already sufficient to recognize general trends or to allow reliable comparisons of materials. With the lower speed chosen in this work, even a finer resolution of the potential/current diagram was achieved.

In all experiments, a magnetic stirrer (7.3×40 mm) was used with an agitation of 200 revolutions per minute (rpm) in order to ensure a continuous and reproducible mixing of the immersion medium. During the experiments, the CMC was not perfectly closed and, as a consequence, the immersion solution was in contact with the atmosphere and, subsequently, with oxygen. This point is important because oxygen is necessary for corrosion.

The potential/current diagram (i.e., Fig. 4) was plotted semi-logarithmically from the measured data. In the resulting diagrams, the Tafel-lines were used to evaluate the corrosion current and the balance potential. The corrosion current density is the ratio of the corrosion current and the steel sample surface. The corrosion rate (CR) was calculated from the corrosion current density in accordance to ASTM G 102 [17] based on Faraday's law.

To simplify the calculation, it was assumed that the mild steel samples consist of pure iron. The real iron content of the mild steel sample is marginally lower (98.07%, see Table 2). The expected error within this simplification is estimated to less than 3%.

2.3 Immersion media

Since the experiments were performed with flowing water which was simulated by using a magnetic stirrer, the value for N_1 equals zero. Because of this assumption, the maximum theoretically value of the W_0-factor in the laboratory experiments is $W_0 = +8$ and the minimum theoretically value $W_0 = -19$. In nature, the W_0-factors are in the range of −11 to +2. Thus, the W_0-factors used in the experiments were from −10 up to +2 in steps of two for the basic model solutions (Table 3).

The salts used to prepare the model solutions were sodium chloride (NaCl), sodium sulfate ($Na_2SO_4 \cdot 10\,H_2O$), calcium nitrate ($Ca(NO_3)_2 \cdot 4\,H_2O$), and calcium sulfate ($CaSO_4$). Since the W_0-factor is a sum of parameters with limited range (last-term being a quotient), there are certain values which the W_0-factor cannot attain. That counts especially with regard to the natural waters and their compositions.

In these experiments, the sulfate, chloride or calcium content can be varied, yet still be kept within a window which leaves the rating number and thus the same W_0-factor unchanged. In addition, the rating numbers can be varied without changing the W_0-factor. To check the effect of these variations on the corrosion rate, further experiments were performed with a lower (Index a) or a higher (Index b) salt concentration (N_3) (Table 4).

Moreover, immersion media from nature were used in further experiments: seawater from the Atlantic Ocean (Middelkerke, Belgium) and the North Sea (Helgoland, Germany), Rhine water (Karlsruhe), and brackish water from Kiel-Holtenau (Germany). These water samples were not artificially altered and they were used within a few weeks after the sample extraction.

The pertinent rating numbers were determined experimentally (Table 5) and the W_0-factors were calculated accordingly (Table 6).

For the Atlantic Ocean and the North Sea, seawater was chosen as the kind of water ($N_1 = -5$). The determination for the Rhine and Kiel-Holtenau was running water ($N_1 = 0$).

Furthermore, the electrical conductivity and the salinity were measured (Table 6).

The four W_0-factors of the natural waters include typical values ranging from seawater (−9.67) to freshwater (1.33). In comparison with the water from the Atlantic Ocean and the North Sea, the model solution with a W_0-factor of −10 includes more sulfate, less chloride, and less calcium. The pH-value and the acid capacity are slightly lower in the synthetic model solution.

Table 3. Composition of the immersion media of the basic experiments

W_0	N_3 Chloride (mol/m^3)	N_3 Sulfate (mol/m^3)	N_5 Calcium (mol/m^3)	N_6 pH-value (20 °C)	N_4 Acid capacity ($Ks_{4.3}$) (mol/m^3)
−10	176.4	66.7	1.3	7...7.5	1...2
−8	25.9	2.4	0.4	7...7.5	1...2
−6	25.9	2.4	0.4	7...7.5	2...4
−4	25.9	2.4	1.3	>7.5	2...4
−2	15.6	2.4	5.0	>7.5	1...2
0	2.1	0.7	5.0	7...7.5	1...2
2	0.5	0.2	1.3	7...7.5	1...2

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Table 4. Composition of the variations of the immersion media

W_0	N_3 Chloride (mol/m^3)	N_3 Sulfate (mol/m^3)	N_5 Calcium (mol/m^3)	N_6 pH-value (20 °C)	N_4 Acid capacity ($Ks_{4.3}$) (mol/m^3)
-8_a	15.6	2.4	0.4	<5.5	1...2
-8_b	41.5	9.5	0.4	7...7.5	1...2
-4_a	15.6	2.4	0.4	>7.5	1...2
-4_b	41.5	9.5	1.3	>7.5	2...4
-2_a	2.1	0.7	1.3	6,5...7	1...2

2.4 Experiments with added DOC

In the experiments in which the influence of the organic matter was examined, it was either an extract of the fulvic acid fraction (FA) from the Lake Hohloh (HO) in the Black Forest (HO FA 13) or the original water (HO 28, not artificially altered) was used. Lake Hohloh is part of an upland moor and is about 1000 m above sea level. Lake Hohloh has a pH-value of about 4 and a DOC content of about 24 mg/L [18].

Due to the fact that Lake Hohloh has a very low electrical conductivity of approximately 40 μS/cm, sodium sulfate ($c(Na_2SO_4) = 10\,mol/m^3$) or sodium chloride ($c(NaCl) = 30\,mol/m^3$) were added in HO 28 in order to increase the electrical conductivity to 2–3 mS/cm and with this the corrosion rates in order to see the influence of the DOC content during the experiments better.

The DOC content of the organic additives was determined by the combustion method (Shimadzu, TOCV-CSN-ASI). The samples were pre-filtered with a membrane-filter having a pore diameter of 0.45 μm (PP; sterile; 15 mm).

3 Results and discussion

3.1 Results referring to the W_0-factor

In the experiments with immersion media of different W_0-factors, the pH-value was always in the middle of the range of the rating numbers (Tables 1 and 3). The temperature was kept constant at $T = 20 \pm 0.1$ °C. The oxygen concentration was between 7 and 7.5 mg/L in all experiments.

The results of the basic experiments ($n = 3$) with the model solutions (Table 3) can be seen in Fig. 1.

The first experiments in the synthetic model solutions with a defined W_0-factor show that the corrosion rate, as expected, is increasing with a more negative W_0-factor. In the area between a W_0-factor from −8 to −2, a plateau can be seen. In this area, rating number N_3 (content of sulfate and chloride) changes only marginally (between 30.7 and 20.3 mol/m^3) relative to the other factors which, in conclusion, seem to have lesser impact on the corrosion rate.

Among the synthetic model solutions with W_0-factors of −8 and −6, the higher corrosion rate at −8 can be explained by the acid capacity, which is larger for the model solution with a W_0-factor of −6 and normally decreases the corrosion rate.

The model solution with a W_0-factor of −10 shows the highest corrosion rate. This can be explained with the highest content of sulfate and chloride.

The results of the experiments with the variations of the W_0-factors (Table 4) and the natural waters (Tables 5 and 6) are shown in Fig. 2.

The salt concentration is, however, varied within the allowable range in which the corresponding rating numbers (N_3) do not change in the variations with a higher salt concentration. For instance, in the basic model solution with a W_0-factor of −8, the sum of the salt content of rating number N_3 (chloride + two times sulfate) is 30.7 mol/m^3, and is increased up to 60.6 mol/m^3. Both salt concentrations are still in the same window for rating number N_3 (25–100 mol/m^3) and thus both concentrations yield rating number $N_3 = -6$ and consequently the same W_0-factor.

The experimental variations in salt concentration were carried out at three different W_0-factors (Table 4). In the experiments with a W_0-factor of −8, there is no definitive effect of increasing or decreasing salt concentration.

For less corrosive immersion solutions (W_0-factor −4 or −2), there is a significant effect on the corrosion rate

Table 5. Water analysis of the used natural waters

	N_3 Chloride (mol/m^3)	N_3 Sulfate (mol/m^3)	N_5 Calcium (mol/m^3)	N_6 pH-value (20 °C)	N_4 Acid capacity ($Ks_{4.3}$) (mol/m^3)
Atlantic Ocean	591.6	30.8	10.0	7.51	2.9
North Sea	453.5	24.3	8.8	7.87	2.5
River Rhine	0.5	0.3	1.4	8.08	3.0
Kiel-Holtenau (estuary to Baltic Sea)	232.4	13.5	3.6	7.95	2.5

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Table 6. Data from the used natural waters and the calculated W_0-factor

	Electrical conductivity (mS/cm[1])	Salinity (g/kg; 20 °C)	W_0-factor
Atlantic Ocean	50.9	33.2	−9.67
North Sea	45.1	29.1	−9.67
Rhine	0.40	0.0	1.34
Kiel-Holtenau	25.5	15.3	−4.33

of increasing or decreasing the salt concentration. Overall, it can be concluded that the salt concentration has an influence on the corrosivity in solutions, which are less corrosive ($W_0 > -6$).

Water with the same W_0-factor can show different corrosivity but the corrosion rates seem to be still in the range of the surrounding W_0-factors.

The corrosion rates of the natural water samples agree well with the laboratory results of the model solutions.

The model solution and the two natural waters with a W_0-factor of approximately −10 have corresponding corrosion rates. The Atlantic seawater from Belgium has a slightly higher corrosion rate (about 1350 μm/a) than the seawater from the North Sea (about 1100 μm/a), which can be explained by the higher salt concentration of the Atlantic seawater.

The natural water from Kiel-Holtenau has a W_0-factor of about −4.33 and the water and can be classified as brackish water. The corrosion rate is in the range of the synthetic model solutions.

The natural water from the River Rhine at Karlsruhe has a W_0-factor of about +1.33. It also fits very well to the corrosion rate of the model solutions.

In order to get a better validation of these results, further experiments with real water samples should be performed.

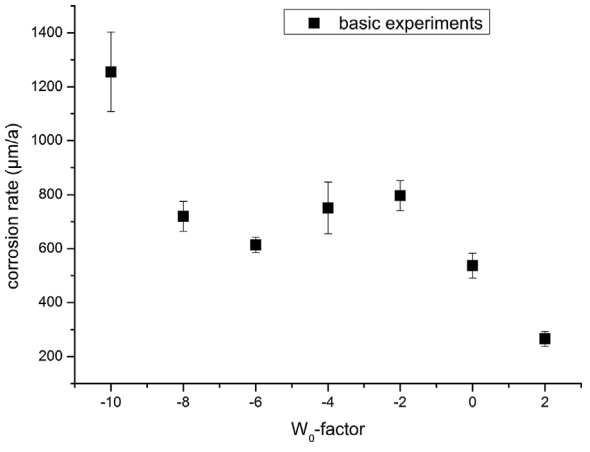

Figure 1. Corrosion rates of the basic experiments (CMC, 200 rpm, T = 20 ± 0.1 °C)

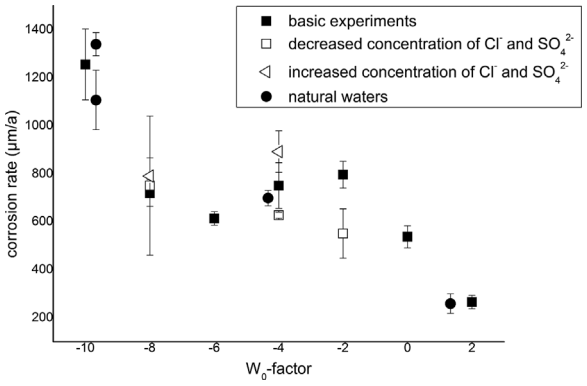

Figure 2. Corrosion rates of the experiments with different W_0-factors and different variations (CMC, 200 rpm, T = 20 ± 0.1 °C)

In summary, the results in Fig. 2 demonstrates that synthetic model solutions can be used to match natural waters with known W_0-factor within measurements with a CMC.

The great influence of the salinity (N_3) in the measurements with a CMC can be seen in Fig. 3. In this case, the salinity is the sum of chloride and two times sulfate as in the calculation of N_3. It shows the dominant role of the salinity in electrochemical studies without a protective rust layer. This statement is supported by the applied fit which yields a coefficient of determination of 0.714.

3.2 Effect of the DOC content on the corrosion rate

There are always organic components in aquatic systems in nature. In the W_0-factor organic substances are not included. Therefore, the influence of organic substances based on humic substances was investigated on the corrosivity of different media.

The source of the DOC content was Lake Hohloh. In Fig. 4, it is obvious that the measured data shift to a larger potential with a higher content of DOC. The three curves are only examples and constitute an excerpt of all the experiments which were done with DOC content.

The results of the experiments are shown in Fig. 5. A sodium sulfate and a sodium chloride solution were used because of the low electrical conductivity of Lake Hohloh. The DOC-concentration was increased by adding water (HO 28) from Lake Hohloh or an extract (HO FA 13) of it.

The corrosion current and, therefore, the corrosion rate increases with reducing electrical potential (compare Fig. 4).

Figure 5 indicates that the corrosion rate (n = 3) decreases with an increasing concentration of DOC. This effect is observable for all DOC experiments, regardless of the source of the DOC which was added during the preparation of the model solutions (HO 28 or as HO FA 13 fraction). The amount of HO FA 13 was limited so that only two experiments could be carried out with it. The pH-value in the experiments was not adjusted separately and was in all cases between 4.8 and 5.0.

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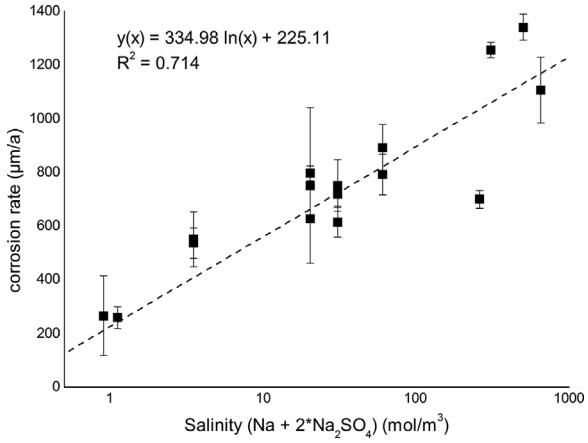

Figure 3. Corrosion rate of all experiments *versus* the salinity (chloride plus two times sulfate) (CMC, 200 rpm, $T = 20 \pm 0.1\ ^{\circ}C$)

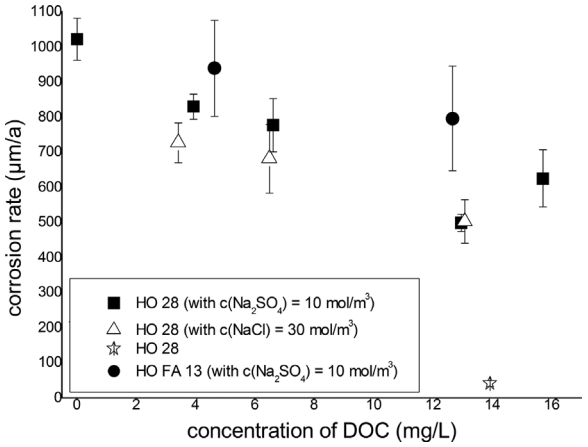

Figure 5. Corrosion rates of the experiments ($n = 3$) with different concentrations of DOC and the addition of Na_2SO_4 and NaCl

The result of the experiment without sodium chloride or sodium sulfate (only pure HO 28) shows a very low corrosion rate as expected.

Furthermore, the decreasing effect on the corrosion rate of a higher DOC of Lake Hohloh can be found in the experiments with addition of sodium sulfate ($c(Na_2SO_4) = 10\,mol/m^3$) and also sodium chloride ($c(NaCl) = 30\,mol/m^3$). Both salts and concentrations were chosen to increase the electrical conductivity for the measurements and to have the same ionic strength (ionic strength $= 30\,mol/m^3$).

Measurements in nature carried out by Federal Waterways Engineering and Research Institute as a part of a European research project [13] showed the same effect of the DOC on the corrosion of 15 sheet piling structures (Fig. 6).

Obviously, the DOC concentration varies widely in natural waters but the concentration is not as high as in the laboratory experiments. The natural corrosion rates (Fig. 6) were much lower than the corrosion rates in the experiments (Fig. 5). One possible explanation is that in nature the mild steel structures (in this case sheet pile structures) are covered by a rust layer which slows down the corrosion. The higher temperature in the experiments also accelerates the corrosion.

In this case, this distribution covers all kinds of waters including seawater, brackish water, or freshwater. The applied linear fit yields a coefficient of determination of 0.628. This is an acceptable fit due to the fact that the construction lifetime also plays an important role in the corrosion rates of sheet pile structures and is not considered here.

The dependence of the corrosion rate on the DOC-concentration can be seen in the experiments in the CMC but a greater influence was only observed at higher DOC-concentrations, which are partly found in nature.

The DOC-concentration is not included in the W_0-factor for the evaluation of the corrosivity of water, although the DOC clearly has an impact on the corrosion in the experiments and also in nature.

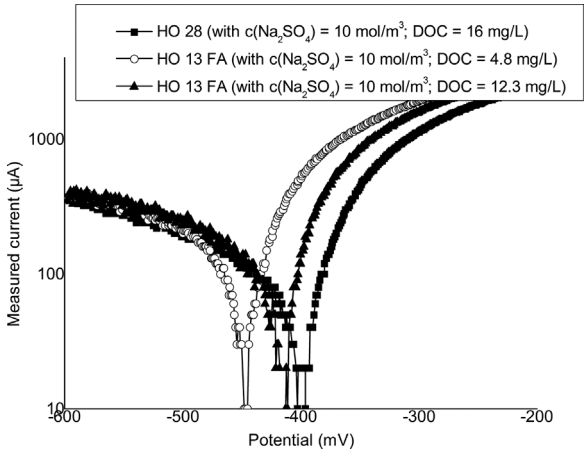

Figure 4. Potential-current-curves from experiments with different DOC-content

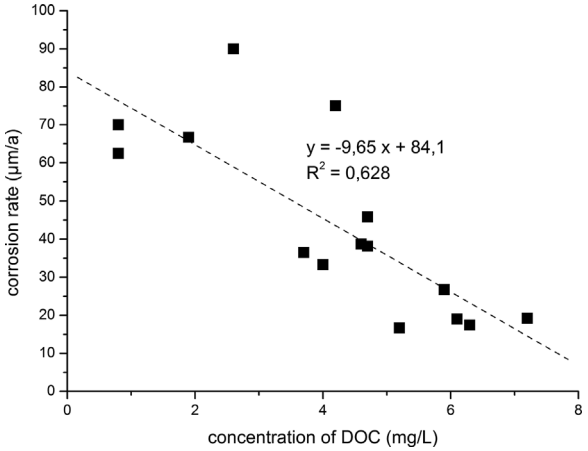

Figure 6. Uniform corrosion rate in nature (underwater zone) *versus* the DOC-concentration of the corresponding water (measurements carried out by Federal Waterways Engineering and Research Institute as a part of a European research project [13])

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4 Conclusions

The results obtained in this study using the W_0-factor according to the German standard method DIN 50929-3 [3] for assessment of the corrosion of mild steel were presented. In addition, laboratory experiments which investigate the influence of DOC from Lake Hohloh on corrosion were performed. In summary, it can be concluded:

1) The comparison between the model solutions with a defined W_0-factor has shown that the W_0-factor allows a rough prediction of the corrosiveness of the medium during the investigation in an electrochemical corrosion measuring cell (CMC). It was found that, as described in the German standard DIN 50929-part 3, the smaller the W_0-factor of a water, the higher the corrosiveness and hence the corrosion rate. The difference of the corrosion rates given in the German standard and the experiments can be explained by the protective rust layer in nature and the temperature.
2) In model solutions with constant W_0-factor larger than -6, but varying salt concentration, an increased corrosion rate was found for higher salt concentrations. In this area, the corrosion rate also decreases with reducing salt concentration. In the model solution with a W_0-factor of -8, the variation of the salt concentration shows no visible effect within the W_0-factor.
3) The laboratory results with the model solutions were compared with four natural waters. It has been found, that the W_0-factors of the natural waters and their corrosion rates fit very well those of the employed laboratory model solutions in the measurements in the CMC.
4) The dependence of the corrosion rate on the DOC-concentration can be seen in the experiments in the CMC. With a higher concentration of DOC, the corrosion rate decreases. This applies to both, the addition of DOC to a sodium sulfate solution and to a sodium chloride solution. In nature, the same effect can be found.

As a consequence, the concentration of DOC should be considered when assessing the corrosiveness of any kind of water with the W_0-factor.

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