

## THE PREDICTION OF ATMOSPHERIC CORROSION FROM METEOROLOGICAL AND POLLUTION PARAMETERS—II. LONG-TERM FORECASTS

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**Abstract**—Long-term atmospheric corrosion forecasts often rely on the fulfilment of equations of the form  $C = At^n$ , where  $C$  is the corrosion after  $t$  years and  $A$  represents corrosion after the first year of exposure. Appropriate values must be assigned to constants  $A$  and  $n$ . In the first part of the work an analysis was performed on the possibility of expressing  $A$  as a function of usually available environmental parameters. In this second part data compiled in a comprehensive literature survey are used to determine whether the exponent  $n$  of the above equation can also be expressed as a function of such environmental parameters.

### INTRODUCTION

THE VARIATION of atmospheric corrosion with time is known to conform to an equation of the form

$$C = At^n \quad (1)$$

where  $C$  means corrosion after  $t$  years,  $A$  is corrosion after the first year of exposure and  $n$  is an exponent normally less than unity.<sup>1</sup> In an approximate manner this equation holds for a number of metallic materials exposed to the atmosphere for fairly long times.<sup>2,3</sup> Predicting corrosion after a given time of exposure should only require  $n$  and  $A$  to be determined. Of these two parameters,  $A$  is easier to obtain experimentally as it only requires 1 year of testing. However,  $n$  can only be reliably obtained after many years of exposure. The problems encountered in selecting the most appropriate  $A$  value to be used in equation (1) now get larger with the value of  $n$ . In the first part of the work<sup>4</sup> the possibility of estimating  $A$  from simple meteorological and pollution parameters was investigated. In this second part estimating the exponent  $n$  similarly was attempted.

There are few literature references relating  $n$  to environmental parameters. Bragard and Bonnarens<sup>5</sup> found that the value of  $n$  for steel relates to wetness time ( $t_w$ ) and corrosion for the first year exposure. By performing a statistical analysis of data from 12 corrosion test sites located over a small area in the region of Lugo (Spain), Espada *et al.*<sup>6</sup> found  $t_w$  and the rainfall accounted for a high proportion of the variance of  $n$  for steel. Also, Benarie and Lipfert<sup>7</sup> showed  $n$  for zinc to be highly correlated with rain pH. No similar relations have to date been reported for other metallic materials such as copper and aluminium. The few expressions reported so far were derived from data supplied by a relatively small number of test sites, occasionally with minor variations in the environmental parameters, so their universality is dubious.

Similarly to the approach used in the first part of the work,<sup>4</sup> in this second part an

TABLE 1. REFERENCES USED TO OBTAIN THE VALUE OF EXPONENT  $n$  IN EQUATION (1) AND COUNTRIES FOR WHICH INFORMATION WAS REPORTED

Country	References
Argentina	32, 33
Belgium	26
Bulgaria	9, 10
Canada	17
Cuba	40
Czechoslovakia	9, 10, 22
France	5, 26
Germany	11, 21
Hungary	9, 10
India	27, 28
Italy	5, 26
Japan	25
Norway	8, 34
Panama	29
Russia	9, 10, 23, 24
Spain	15, 35, 36, 37
Sweden	22, 34
Taiwan	31
U.K.	14
U.S.A.	12, 14, 18, 19, 38, 39, 41
Yugoslavia	30

attempt was made to find a relationship between exponent  $n$  for carbon steel, zinc, copper and aluminium, and various environmental parameters that are usually available. By using an experimental basis as wide as possible an attempt was made to reach universally applicable conclusions in this respect.

The environmental parameters used in the statistical treatment included the relative humidity annual average (RH), annual wetness time ( $t_w$ ), annual number of rainy days ( $D$ ), temperature annual average ( $T$ ), and average concentrations of  $\text{SO}_2$  ( $S$ ) and chloride (Cl) in the atmosphere. The rain pH was excluded as it is only occasionally recorded. Corrosion and climatological data were compiled from both domestic and international sources.<sup>5,8-41</sup> References are classified in Table 1 according to the countries for which information was reported.

Exponent  $n$  can be determined from a log-log plot of corrosion data against the exposure time (in years), for data series obtained during prolonged exposure periods. With this purpose there was recourse to worldwide data from the specialized literature. Unfortunately, long-term corrosion data are much scarcer than 1 year data. Exposure periods of 5 years or longer would be advisable, yet they are not frequent. Even by using the  $n$  values derived for 3 year exposures, the data samples available for the statistical analysis (Table 2) were much smaller than those used in the first part of the work,<sup>4</sup> where first-year corrosion data were used.

#### ANALYTICAL RESULTS AND DISCUSSION

By using the same mathematical models as in the first part<sup>4</sup> the potential correlations between  $n$  and the different independent variables were analysed:  $t_w$ ,

RH,  $D$ ,  $T$ , Cl,  $S$  and  $A$  (first-year corrosion). The value of  $A$  was included among these variables on account of its potential relationship to  $n$ , which was suggested by Bragard and Bonnarens for steel.<sup>5</sup> For steel, the statistical treatment indicated an insignificant effect of chloride level as a separate variable. Some experimental evidence does suggest a possible effect of marine atmospheres on  $n$  value. For this

TABLE 2. CHARACTERISTICS OF THE CORROSION AND ENVIRONMENTAL DATA USED IN THE STATISTICAL TREATMENT

Material and number of data	Variable	Mean	SD	Smallest value	Largest value
Carbon steel ( $N = 114$ )	exponent $n$ in equation (1)	0.623	0.150	0.240	1.00
	annual corrosion ( $\mu\text{m}$ )	41.6	31.3	6.0	288.0
	RH (%)	74.9	6.3	55	88
	$t_w$ (annual fraction)	0.43	0.16	0.04	0.84
	number of rainy days per year	132.2	45.9	16	231
	temperature ( $^{\circ}\text{C}$ )	12.0	5.4	-1.2	29
	chlorides ( $\text{mg Cl}^- \text{dm}^{-2} \text{d}^{-1}$ )	0.29	1.14	0	11.3
	$\text{SO}_2$ ( $\text{mg SO}_2 \text{dm}^{-2} \text{d}^{-1}$ )	0.39	0.43	0	2.3
Zinc ( $N = 59$ )	exponent $n$ in equation (1)	0.861	0.187	0.360	1.30
	annual corrosion ( $\mu\text{m}$ )	2.14	1.96	0.32	12.0
	RH (%)	72.8	6.1	55	83
	$t_w$ (annual fraction)	0.38	0.15	0.09	0.84
	number of rainy days per year	119.5	49.7	37	231
	temperature ( $^{\circ}\text{C}$ )	11.1	5.9	-1.2	27
	chlorides ( $\text{mg Cl}^- \text{dm}^{-2} \text{d}^{-1}$ )	0.21	0.56	0	2.5
	$\text{SO}_2$ ( $\text{mg SO}_2 \text{dm}^{-2} \text{d}^{-1}$ )	0.51	0.49	0	2.1
Copper ( $N = 49$ )	exponent $n$ in equation (1)	0.672	0.163	0.350	1.04
	annual corrosion ( $\mu\text{m}$ )	2.04	1.48	0.47	7.1
	RH (%)	74.4	5.3	62	84
	$t_w$ (annual fraction)	0.39	0.14	0.18	0.84
	number of rainy days per year	134.4	44.3	62	231
	temperature ( $^{\circ}\text{C}$ )	11.7	5.0	-1.2	27
	chlorides ( $\text{mg Cl}^- \text{dm}^{-2} \text{d}^{-1}$ )	0.27	0.58	0	2.5
	$\text{SO}_2$ ( $\text{mg SO}_2 \text{dm}^{-2} \text{d}^{-1}$ )	0.43	0.42	0.05	1.8
Aluminium ( $N = 28$ )	exponent $n$ in equation (1)	0.825	0.245	0.340	1.30
	annual corrosion ( $\mu\text{m}$ )	0.782	1.09	0.04	3.70
	RH (%)	72.5	6.0	62	83
	$t_w$ (annual fraction)	0.43	0.12	0.24	0.77
	number of rainy days per year	129.4	49.3	62	231
	temperature ( $^{\circ}\text{C}$ )	10.2	4.3	4	18
	chlorides ( $\text{mg Cl}^- \text{dm}^{-2} \text{d}^{-1}$ )	0.32	0.65	0	2.5
	$\text{SO}_2$ ( $\text{mg SO}_2 \text{dm}^{-2} \text{d}^{-1}$ )	0.48	0.35	0.04	1.5

TABLE 3. RELATIONSHIPS BETWEEN THE VALUES OF EXPONENT  $n$  IN EQUATION (1) AND THE ENVIRONMENTAL PARAMETERS FOR CARBON STEEL, ZINC AND COPPER

Materials	Equations	Remarks
Carbon steel	$n = 0.570 + 0.0057 \text{ Cl } T + 7.7 \times 10^{-4} D - 1.7 \times 10^{-3} A$ ( $R = 0.40$ ) equation (2)	$n$ = exponent in equation (1), $A$ = first-year corrosion ( $\mu\text{m}$ ), $t_w$ = wetness time (annual fraction), $T$ = temperature annual average ( $^{\circ}\text{C}$ ), $D$ = number of rainy days per year, $\text{RH}$ = relative humidity annual average (%)
Carbon steel	$n = 0.531 + 0.115 \text{ MAQ} + 0.00112 D - 0.221 t_w$ ( $R = 0.44$ ) equation (3)	$\text{MAQ}$ = marine atmosphere quality (variable coded as 1 or 0), $S$ = $\text{SO}_2$ pollution annual average ( $\text{mg SO}_2 \text{ dm}^{-2} \text{ d}^{-1}$ ), $\text{Cl}$ = chloride pollution annual average ( $\text{mg Cl}^{-} \text{ dm}^{-2} \text{ d}^{-1}$ ), $R$ = multiple correlation coefficient
Zinc	$n = 0.526 + 0.545 S (1 - 0.068 T) + 0.0246 T$ ( $R = 0.62$ ) equation (4)	
Copper	$n = 0.822 - 0.0684 T (1 - 0.011 \text{ RH})$ ( $R = 0.47$ ) equation (5)	

reason including the 'marine atmosphere quality' (MAQ) was explored as another independent variable, which does not distinguish between the various chloride levels, but refers exclusively to the marine quality; so atmospheres with chloride levels not lower than  $0.1 \text{ mgCl}^{-} \text{ dm}^{-2} \text{ d}^{-1}$  were coded  $\text{MAQ} = 1$ , whereas all others were coded 0. The statistical treatment was carried out by Stepwise Multiple Regression method, which was implemented with the aid of commercially available computer software.<sup>42</sup>

The equations in Table 3 resulted in the best fits and hence accounted for the highest proportions of the variance of  $n$ . As was pointed out in the first part of the work, predictions based on values of the independent variables that are much larger or smaller than those used (Table 2) to derive the equations in Table 3 are to be taken with caution.

In the statistical treatment for steel the product (interaction) of the chloride level and the temperature (equation 2) was found to be one of the factors most significantly influencing the value of  $n$ . However, such an interaction, together with the annual number of rainy days and the first-year corrosion (this with a negative coefficient) accounted for only 16% of the variance. The MAQ together with the number of rainy days and wetness time (this with a negative coefficient) accounted for 20% of the variance ( $R = 0.44$ , equation 3). Thus the value of  $n$  for steel is seemingly dependent on the mere qualitative presence of chloride ions to a greater extent than on their quantitative level. This might be interpreted in the sense that a low chloride level is enough to increase significantly the duration of the wetting episodes because of the hygroscopic character of the salt, or to induce morphological changes in the rust layer resulting in increased diffusivity. On the other hand, the lack of a significant correlation between  $\text{SO}_2$  pollution and  $n$  for steel should be emphasized.

As regards zinc, the  $\text{SO}_2$  level by itself accounts for 29% of the variance of  $n$ . A better correlation is obtained introducing the interaction of this pollutant with temperature, in which case the regression equation accounts for 39% of the variance

( $R = 0.62$ , equation 4 in Table 3). Neither the chloride concentration nor any other parameter included in the statistical treatment are significantly correlated with  $n$ . It should be noted that the relationship between  $n$  and the rain pH found by Benarie and Lipfert<sup>7</sup> yields only a slightly higher correlation coefficient ( $R = 0.68$ ).

Neither of the two pollution variables ( $\text{SO}_2$ ,  $\text{Cl}^-$ ) was significantly related to  $n$  for copper. On the other hand, both the temperature and the product of this variable with RH were highly correlated with  $n$  (equation 5).

For aluminium, no significant correlation between  $n$  and any of the environmental parameters was found. It is doubtful whether the data base (28 data) used was large enough for the study. In practice, there is also the possibility of choosing the exponent  $n$  to be introduced into equation (1) as the average value of  $n$  for each one of the groups of data relative to the different types of atmospheres. Table 4 lists such average  $n$  values for carbon steel, zinc, copper and aluminium. To this effect, a given atmosphere was considered to be rural if its  $\text{SO}_2$  and  $\text{Cl}^-$  levels were  $<0.1 \text{ mg dm}^{-2} \text{ d}^{-1}$ , urban-industrial if the  $\text{SO}_2$  level was  $\geq 0.1 \text{ mg dm}^{-2} \text{ d}^{-1}$  and the  $\text{Cl}^-$  level was  $<0.1 \text{ mg dm}^{-2} \text{ d}^{-1}$ , and marine if the  $\text{Cl}^-$  level was  $\geq 0.1 \text{ mg dm}^{-2} \text{ d}^{-1}$ . The larger number of data available for carbon steel provided more reliable results than those obtained for the other metals.

As can be seen in Table 4, the average  $n$  value for carbon steel in rural and urban-industrial atmospheres was 0.59, i.e. only slightly greater than the theoretical value (0.50) for a diffusion-controlled mechanism. The  $n$  value for marine atmospheres, 0.69, was 38% greater than the theoretical value, which means a greater effect of the exposure time on the corrosion values. As is typical for zinc,  $n$  values are close to unity (0.92 and 0.85) except in rural atmospheres, which provided a somewhat lower  $n$  value (0.70). Aluminium shows also a trend to give rather high  $n$  values. Finally, the average  $n$  values for copper in urban-industrial and marine atmospheres were around 0.70 and 0.60, respectively. More data from long-term corrosion tests conducted in many different locations are obviously required in order to obtain more reliable  $n$  values for any type of atmosphere.

TABLE 4. AVERAGE VALUE OF  $n$ , STANDARD DEVIATION ( $\sigma$ ) AND NUMBER OF DATA ( $N$ ), FOR EACH MATERIAL AND ATMOSPHERE

Material	Rural atmosphere	Urban-industrial atmosphere	Marine atmosphere
	$<0.1 \text{ mgSO}_2 \text{ dm}^{-2} \text{ d}^{-1}$ $<0.1 \text{ mgCl}^- \text{ dm}^{-2} \text{ d}^{-1}$	$\geq 0.1 \text{ mgSO}_2 \text{ dm}^{-2} \text{ d}^{-1}$ $<0.1 \text{ mgCl}^- \text{ dm}^{-2} \text{ d}^{-1}$	$\geq 0.1 \text{ mgCl}^- \text{ dm}^{-2} \text{ d}^{-1}$
Carbon steel	0.59	0.59	0.69
	$\sigma = 0.18$	$\sigma = 0.14$	$\sigma = 0.13$
	( $N = 15$ )	( $N = 61$ )	( $N = 38$ )
Zinc	0.70	0.92	0.85
	$\sigma = 0.21$	$\sigma = 0.17$	$\sigma = 0.14$
	( $N = 11$ )	( $N = 35$ )	( $N = 13$ )
Copper	0.62	0.72	0.62
	$\sigma = 0.25$	$\sigma = 0.14$	$\sigma = 0.16$
	( $N = 4$ )	( $N = 26$ )	( $N = 19$ )
Aluminium	0.79	0.90	0.74
	$\sigma = 0.35$	$\sigma = 0.21$	$\sigma = 0.25$
	( $N = 3$ )	( $N = 14$ )	( $N = 11$ )

Coming back to the regression equations (Table 3), the fits yielded by the environmental parameters considered in the statistical analysis were, as a rule, surprisingly poor, and did not improve on considering each type of atmosphere separately. The proportion of unexplained variance in the  $n$  values increased dramatically in comparison with that for the first-year corrosion.<sup>4</sup> While over 83% of the variance in the first year corrosion of zinc was accounted for, an identical treatment failed now to account for over 39% of the variance in the  $n$  value for zinc. The correlations between  $n$  and the environmental variables for steel and copper were even lower, and no significant relationship could be established for aluminium.

One may argue that the many sources of uncertainty that affect the values of the environmental variables preclude the obtainment of better correlations. However, the potential sources of error are essentially the same as those involved in the first part of the work,<sup>4</sup> where high correlation coefficients were obtained for zinc and even for steel. Therefore, it seems more reasonable to conclude that the independent variables used in the statistical treatment are not the most suitable for predicting  $n$ . As with first-year corrosion analysis,<sup>4</sup> there must be other, unidentified parameters, that could affect  $n$  decisively. In this case, a high worldwide variability of those unknown parameters would justify the poor fits obtained. Small variations in these unknown parameters would also explain why data from limited geographic areas give rise to better correlations.

It remains to be determined which other parameters should be included in the equations used to predict  $n$ . Identifying them is of a great interest and should therefore be a priority research task in the future despite the potential difficulties involved. A number of factors come to mind in this context, one of them the  $H^+$  concentration in rainwater. As mentioned before, Benarie and Lipfert<sup>7</sup> have shown that the rain pH may largely account for the  $n$  value of zinc. Unfortunately, no rain pH data were recorded by most of the corrosion test sites included in this study, so the influence of this parameter could not be ascertained. It should be remembered also that the protective effect of the corrosion products layers formed in the open air depends on the morphology, porosity and adhesion of these layers, properties which are probably greatly influenced by the presence of some pollutants (small amounts should be sufficient to have some effect) and the frequency, duration and type of wet and dry cycles.

### CONCLUSIONS

The exponent  $n$  in the atmospheric corrosion power law was correlated with meteorological and pollution parameters by using a large data base in order to establish universal equations. For steel, the best correlation between  $n$  and the environmental parameters was obtained by using the MAQ as an independent variable. Accordingly, the classification of an atmosphere as marine increases the value of exponent  $n$  by 0.115. On the other hand, no significant correlation could be established with the  $SO_2$  level.

The value of  $n$  for zinc was found to be significantly correlated with the temperature and the product of this parameter with the  $SO_2$  level. None of the other parameters improved significantly on this correlation.

No significant correlation between  $n$  and the pollution parameters could be established for copper. However,  $n$  was significantly correlated with both the temperature and the product of this parameter with RH.

The high proportion of unaccounted variance—the maximum proportion accounted for was only 39%—resulting from the above-mentioned correlations must be emphasized. The environmental variables considered in this work were probably not the most suitable for predicting  $n$ . Some other (unknown) parameters might have appreciable effects on  $n$ . Identifying them should be a target research subject for the near future.

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