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ATMOSPHERIC CORROSION AND PRECIPITATION RUNOFF FROM ZINC AND ZINC ALLOY SURFACES

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ABSTRACT

The long-term atmospheric corrosion performance of rolled zinc and three thermal-sprayed (TS) zinc materials (Zn, Zn-15Al, and Al-12Zn-0.2In) was characterized by measuring corrosion product concentrations in precipitation runoff at coastal marine and inland sites. Corrosion rates and average zinc concentrations in the runoff were greater at the site having higher annual rainfall. Higher chloride concentrations did not seem to affect either the corrosion rates or the zinc concentrations in the runoff at the coastal site compared to those of the inland site. Zinc runoff concentrations were higher for TS Zn than rolled zinc due to the greater surface area of the thermal-sprayed surface. Average cumulative zinc runoff losses for the two sites were: 64 μmol Zn/L for TS Zn, 37 μmol Zn/L for rolled Zn, 24 μmol Zn/L for TS Zn-15Al, and 1.8 μmol Zn/L for TS Al-12Zn-0.2In. Cumulative zinc runoff losses were directly related both to the precipitation rate and to the availability of Zn in metal surfaces, a consequence of surface roughness and surface chemistry properties of the metal.

Keywords: rolled zinc, zinc alloys, zinc-aluminum, aluminum-zinc-indium, thermal spray, atmospheric corrosion, precipitation runoff, corrosion film, environment

INTRODUCTION

Zinc, aluminum, and zinc-aluminum alloys have long been considered desirable materials as anodes for cathodic protection, and may be used either in galvanic or impressed current cathodic protection systems. Zinc coatings are electrochemically active which enables them to provide cathodic protection to steel; but high electrochemical activity means high corrosion rates, which rapidly depletes the zinc coating. Aluminum coatings are more passive than zinc and serve primarily as a barrier. Alloys of zinc and aluminum are also used in cathodic protection systems[1], and may combine the passive protection of aluminum with the cathodic protection of zinc. Much higher galvanic efficiencies may be achieved by incorporating small amounts of indium into aluminum-zinc alloys.[2-11] The relative atmospheric corrosion performance of coatings of these materials is important in determining, for a given site, their usefulness as protective anodes and in estimating the environmental impacts from contaminants in the surface runoff on the surrounding soil and water resources. Precipitation runoff provides chemical information that can determine corrosion characteristics and environmental impact of boldly exposed metal surfaces.[12-17]

Presented here are the results for atmospheric corrosion and precipitation runoff studies from boldly exposed rolled zinc (Zn), and thermal spray zinc (TS Zn) , 85Zn-15Al (TS Zn-15Al), and 88Al-12Zn-0.2In (TS Al-12Zn-0.2In) surfaces at two sites having different environments: Newport OR, an unpolluted coastal environment, and Albany OR, an unpolluted rural environment. In previous studies, the atmospheric corrosion and precipitation runoff has been described for lead[13,14], copper[15,16], and zinc[12,13,16,17]. The present study compares the atmospheric corrosion and precipitation runoff from a variety of zinc and zinc-alloy surfaces.

Atmospheric corrosion involves two competing processes, the formation of a protective metal oxide corrosion film and modification of the corrosion film into non-protective corrosion products by interactions with the environment. These processes involve interactions between the metal, corrosion film, and environment, and can be represented at any time t by the mass balance

$$C(t) = T(t) + R(t). \qquad \textbf{(1)}$$

where: C(t) = cumulative corrosion mass loss
T(t) = protective corrosion film mass
R(t) = cumulative non-protective corrosion products.

The R(t) term includes processes that convert protective corrosion film to forms that have no further influence on the corrosion process including physical removal from the metal surface through dissolution in precipitation runoff, or by spalling, or through chemical reactions that form non-protective corrosion products on the metal surface.

This paper investigates the formation of soluble corrosion products on the metal surface that are subsequently removed in precipitation runoff. In wet deposition, strong and weak acids present in the precipitation dissolve a portion of the corrosion film and remove it in precipitation draining from the metal surface. In dry deposition, acidic gases in the atmosphere react with the corrosion film during periods when there is little or no runoff. Neutral salts produced by these reactions accumulate on the metal surface to be removed during the next period of precipitation. At polluted sites there is low precipitation pH due to acid gases such as SO_2 and NO_x in the atmosphere, the corrosion film is dissolved by a combination of contributions from strong acid, weak acid and dry deposition processes.[12] In unpolluted sites, that is precipitation pH 5.6 or greater and no acidic gases, dissolution of the corrosion film is due entirely to the weak acid processes. Because both of the sites studied were unpolluted, the soluble portion of the corrosion film on the surface is removed by reaction with dissolved CO_2 (weak acid) delivered to the surface by wet deposition processes. In the absence of spalling or significant accumulation of non-protective corrosion products, R in equation 1 represents the cumulative loss of soluble corrosion product in precipitation runoff, i.e., the cumulative precipitation runoff loss.

The time derivative of equation 1 shows the corrosion rate is equal to the rate of protective film growth and the rate of corrosion film loss in precipitation runoff,

$$dC/dt = dT/dt + dR/dt. \qquad \textbf{(2)}$$

After long exposures, when the corrosion film is well developed and further corrosion film growth is small, i.e., $dT/dt \sim 0$, the corrosion rate is equal to the rate of precipitation runoff loss. Recent results suggest that following an induction period when the corrosion film is maturing, the precipitation runoff loss rate from a variety of metals is linear and relatively insensitive to seasonal variations in precipitation chemistry, air chemistry and meteorology.[12-17] The linearity of the precipitation runoff suggests that predictions about long-term corrosion rates can be made from runoff data.

This paper reports the results of precipitation runoff losses from zinc and zinc-aluminum alloy surfaces exposed at two sites, unpolluted coastal and rural. Results will be expressed as functions of measurable properties of the environment to show how environmental effects on long-term corrosion rates might be estimated.

EXPERIMENTAL DESIGN

Precipitation runoff experiments were conducted at two sites: Newport and Albany, Table 1. The Newport site is within 100 m of the Pacific Ocean; the Albany site is in the Willamette Valley 83 km from the ocean. Flat panels of rolled zinc (UNS-Z44330), TS Zn, TS Zn-15Al, and TS Al-12Zn-0.2In measuring 0.3 x 0.6 m (1 x 2 ft) were mounted in polyethylene trays that collected all precipitation washing the skyward side of the panels. The panels were chemically cleaned prior to installation using standard ASTM methods.[18] The ground-ward side was masked to limit runoff contributions to the skyward side. The trays (and panels) were inclined 30 degrees to the horizon and faced the prevailing wind. A similar tray was set up with a Lexan panel to establish the runoff chemistry from an inert surface (blank).

TABLE 1. ENVIRONMENTAL PROPERTIES OF THE EXPOSURE SITES.

Exposure sites	Newport, OR	Albany, OR
Site type	marine	rural
Precipitation rate: cm/y	182.2	108.4
L/m^2y	1822	1084
Precipitation pH	6.13	5.78
Temperature, °C	10.5	11.2

Precipitation runoff samples were collected for about 2.5 years at the Newport and Albany sites. During the collection period runoff samples were taken at least monthly using a system that minimized evaporation losses and sample contamination. Up to 3 collections a month were made during periods of heavy precipitation. Incident precipitation was also collected on the same schedule using an Aerochem Metrics wet/dry collector. Incident precipitation and runoff were filtered to remove undissolved particulates and analyzed for the standard acid rain ions (H^+, Ca^{+2}, Mg^{+2}, K^+, Na^+, NH_4^+, NO_3^-, Cl^- and SO_4^{-2}) and selected metal ions (Zn^{+2}, Cu^{+2}, Pb^{+2}, and Fe) by ion chromatography and ICP emission spectroscopy.

Some contamination of the Newport runoff panels occurred over an 11 month period during the 2.5 year runoff collection period while a thermal spray zinc anode was applied to the nearby Yaquina Bay Bridge.[13-14] For this reason, data collected at Newport during this period was not used and the data set for Newport was reduced by 13 points out of a total set of 40 points.

The annual average precipitation rate (Table 1) is for vertical rainfall (90 degrees to the horizon). It is available from the National Climatic Data Center at http://www.ncdc.noaa.gov/. The area used for the runoff rates is a unit area of panel surface inclined at 30 degrees (not the area projected on a horizontal surface); the area for the precipitation rate a unit area of horizontal surface.

The metal panel surfaces have different surface roughness because of they way they were formed. Cross-sections of the metal surfaces after exposure to the environment for 2.5 years are shown in Figure 1. The length of a line tracing the surface profile was measured (actual length) and compared to the nominal length of a line along the panel edge as a measure of surface roughness, Table 2. The relative surface roughness, R, compared to that of the rolled zinc panel is given in the third column of Table 2. The atomic fraction of Zn in the surface of the metal, X_{Zn}, is given in the fourth column. The relative availability of Zn in the metal surface compared to rolled zinc is given in column 5 and is the product $R*X_{Zn}$. Relative availability takes into consideration two factors, the actual surface area of the metal and the concentration of Zn in that surface. It roughly measures the concentration of Zn exposed to the environment.

TABLE 2. SURFACE ROUGHNESS AND RELATIVE ZN AVAILABILITY FOR SURFACE REACTIONS.

Metal surface	Roughness		Atomic fraction Zn in surface, X_{Zn}	Relative Zn availability $R*X_{Zn}$
	actual/nominal	Relative to rolled Zn, R		
Rolled Zn	1.62	1.00	1.00	1.00
Thermal spray Zn	2.51	1.55	1.00	1.55
Thermal spray Zn-15Al	2.33	1.44	0.70	1.01
Thermal spray Al-12Zn-0.2In	3.95	2.44	0.05	0.12

EXPERIMENTAL RESULTS

Corrosion film dissolution at unpolluted sites (pH >5.6) is due to carbonic acid, a weak acid. The dissolution of the metal hydroxide corrosion film into the incident precipitation increases the pH of the resulting runoff typically less than 1.5 units. The pH increase is due to an increase in the bicarbonate ion concentration, and there is a net flux of carbon dioxide into the solution to produce additional bicarbonate ions. Rising levels of free HCO_3^- retards the dissolution process and the dissolved corrosion products drain from the surface as precipitation runoff.

Environmental conditions at the two collection sites are listed in Table 1. Because precipitation pH is greater than 5.6 and there are no sources for acidic gases, these sites are unpolluted and exhibit no acid-rain effect. Table 3 lists characteristics of the precipitation runoff: runoff pH, Zn concentration ranges, and Zn average concentrations (on a volume basis) for each of the metals studied at the two sites. There does not seem to be any effect of the high chloride at the Newport (marine) on either runoff pH or Zn concentration. Higher pH values correspond to higher Zn concentrations in the runoff. Highest Zn concentrations in the runoff were produced by the TS Zn. Zn concentrations in the runoff were found in descending order TS Zn > Zn > TS Zn-15Al > TS Al-12Zn-0.2In.

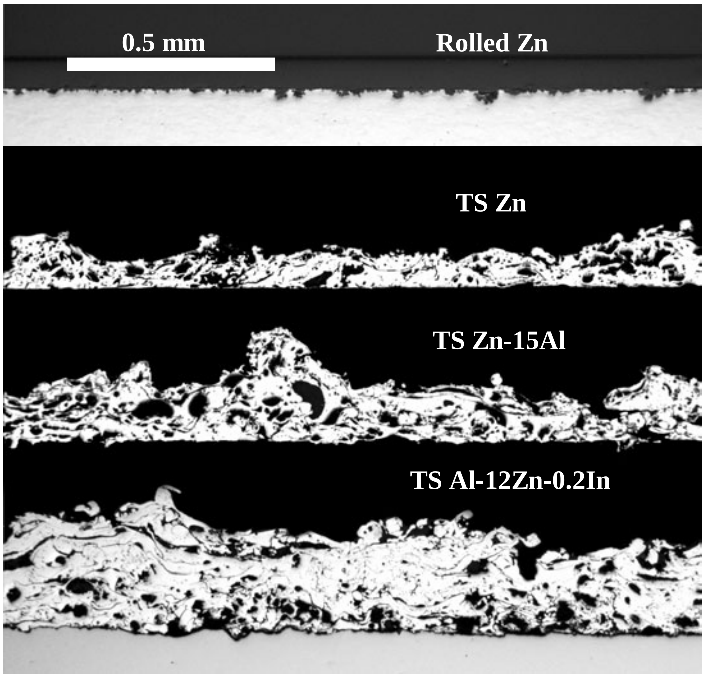
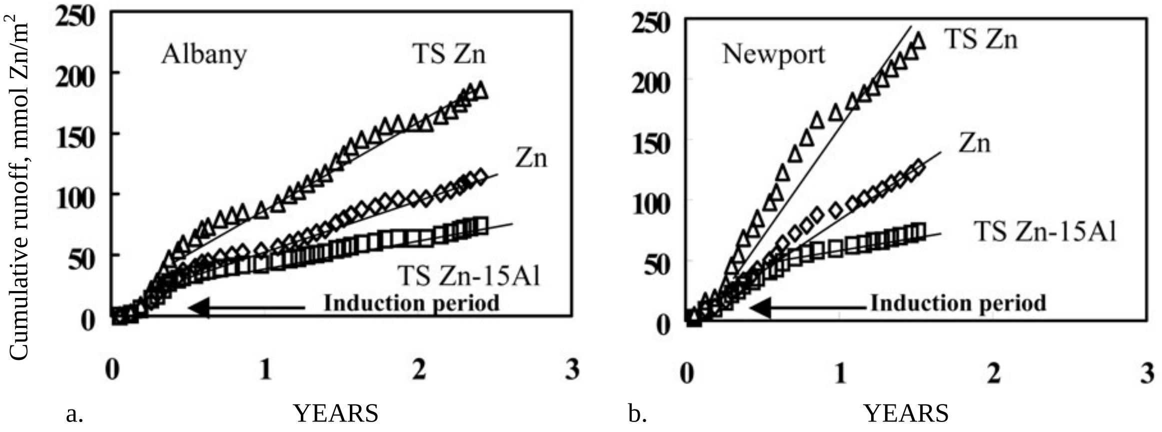


FIGURE 1. Cross-sections of metal runoff panels showing surface roughness described in Table 2; bottom three surfaces are thermal-sprayed coatings.

Cumulative dissolution curves for the two sites are shown in Figures 2a and 2b. The curves for both Newport and Albany show strong seasonal variations resulting from the intense Oregon rainy season in the winter and the typical drought conditions that occur in late summer. The curves also suggest an induction period of a few months in which the corrosion film is establishing itself on the metal surface. The portion of the curves beyond the induction period, seem to vary cyclically around a straight line that describes the long term trends. The curves were fit by least squares to straight lines with slopes corresponding to precipitation runoff rates (the derivative dR/dt in Equation 2) of 85 (rolled Zn), 149 (TS Zn) and 38 (TS Zn-15Al) mmol Zn/m^{2}y at Newport, and 40 (rolled Zn), 66 (TS Zn), and 22 (TS-Zn-15Al) mmol Zn/m^{2}y at Albany, Table 4.The precipitation runoff rates reported in Table 4 would lead to long-term corrosion rates for Zn of 0.78 (Zn) and 1.4 (TS Zn) µm/y at Newport, and 0.37 (Zn) and 0.60 (TS Zn) µm/y at Albany. Figure 3 shows the Zn concentration in the runoff from TS Zn, Zn, and TS Zn-15Al for all collection periods at Newport and Albany. The higher concentration of Zn in the runoff for the TS Zn, shows the effect of the greater surface roughness of thermal spray surfaces when compared with the smooth surface of the rolled zinc panel. The lower concentrations found in the runoff from TS Zn-15Al are a function of the lower Zn concentration (85 percent) in the alloy, as well as the protective (passive) behavior of the Al. It should be noted that Zn concentrations in the runoff were higher for lower runoff volumes than for higher runoff volumes. This could be due to a “sheeting” effect in which part of the incident rainfall does not contact the corrosion film during periods of heavy rainfall, but merely strikes the sheet of runoff without deep mixing.

TABLE 3. PRECIPITATION RUNOFF PH AND ZINC CONCENTRATIONS.

Metal surface	Average runoff pH		Range of Zn concentration in runoff, mg/L		Average Zn concentration in runoff, mg/L	
	Albany	Newport	Albany	Newport	Albany	Newport
Rolled Zn	6.95	7.05	1.5 – 27.3	1.5 – 4.0	2.42	2.51
Thermal spray Zn	7.17	7.26	2.8 – 35.8	3.5 – 6.6	3.94	4.57
Thermal spray Zn-15Al	6.89	6.98	0.7 – 13.2	0.3 – 2.4	1.54	1.49
Thermal spray Al-12Zn-0.2In	6.27	6.60	0.0 – 0.40	0.15 –0.41	0.05	0.24



FIGURES 2a. and 2b. Cumulative Zn precipitation runoff losses for two exposure sites. A least squares fit of the data is plotted after an induction period (buildup of ZnO film) (Table 4.). Slopes of these curves are the precipitation runoff loss rates.

TABLE 4. AVERAGE ZINC PRECIPITATION RUNOFF RATES.

Metal Surface	Precipitation runoff rates mmol Zn/m^{2}y (µm Zn/y) [µmol Zn/L]					
	Newport			Albany		
Rolled Zn	85	(0.78)	[37]	40	(0.37)	[37]
Thermal spray Zn	149	(1.4)	[68]	66	(0.60)	[59]
Thermal spray Zn-15Al	38		[24]	22		[24]
Thermal spray Al-12Zn-0.2In	<1		[1.8]	<1		[1.8]

Figure 4 shows Zn runoff as a function of cumulative precipitation volume rather than exposure time. The curves demonstrate that runoff is linear as a function of precipitation. Moreover, the curves for Albany and Newport are nearly identical for each alloy. This is the result one might expect if corrosion film dissolution were due solely to the weak acid effect and the precipitation layer is well mixed before it leaves the panel surface. The average of the Albany and Newport runoff losses were 64 µmol Zn/L for TS Zn, 37 µmol Zn/L for rolled Zn, 24 µmol Zn/L for TS Zn-15Al, and 1.8 µmol Zn/L for TS Al-12Zn-0.2In. The higher slope for TS Zn is a function of the greater surface area of thermal spray surfaces than that of rolled metal. The lower slope for TS Zn-15Al is a function of the lower Zn concentration in the alloy (85 percent) and the passive nature of the aluminum. The very low slope for TS Al-12Zn-0.2In is a function of both the low Zn concentration in the alloy (12 percent) and the ability of Al in the presence of In to act as a sacrificial anode for zinc.

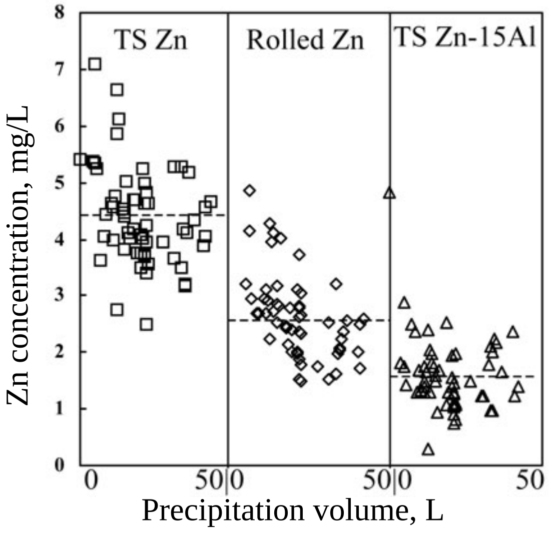


FIGURE 3. Zinc concentration in precipitation runoff from TS Zn (□), Zn(◇), and TS Zn-15Al (△) as a function of precipitation volume. Dashed line is the average concentration for the two sites.

The ability of indium to prevent aluminum passivation is further exhibited by Figures 5a and 5b. In Figure 5a the cumulative aluminum runoff in the two alloys containing aluminum is plotted against time. The very low slope for TS Zn-15Al is due to both the lower aluminum concentration (15 percent) and to the passive nature of the aluminum. The higher slope for TS Al-12Zn-0.2In is primarily due to In preventing the Al from passivating. If the Al does not passivate, there will be greater dissolution of the Al oxide corrosion product and more Al in the precipitation runoff. Figure 5a is similar to the plot for the zinc runoff (Figures 2a and 2b) since both plots exhibit an induction period prior to developing into generally straight lines with seasonal variations. The higher slope for Newport is due to the higher precipitation rate. The precipitation runoff rates (slopes) for Al are 2.0 and 1.5 mmol Al/m^{2}y at Newport and Albany, respectively. Figure 5b plots Al runoff against cumulative precipitation volume. The Albany curve is linear with a slope of 1.4 µmol Al/L. Unlike Figure 2b where the

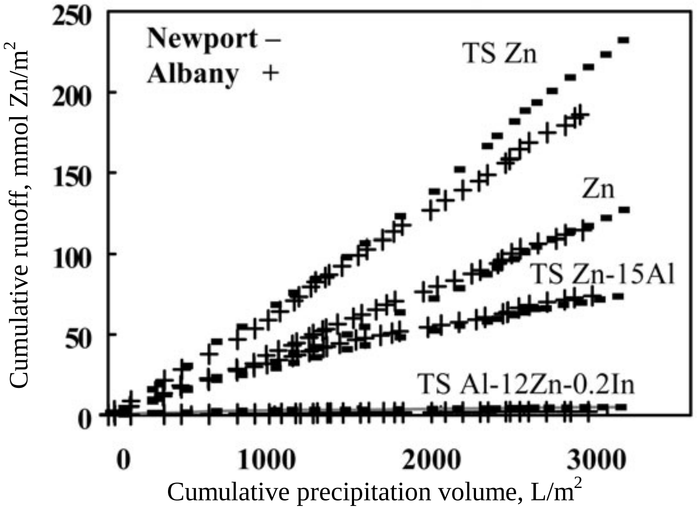
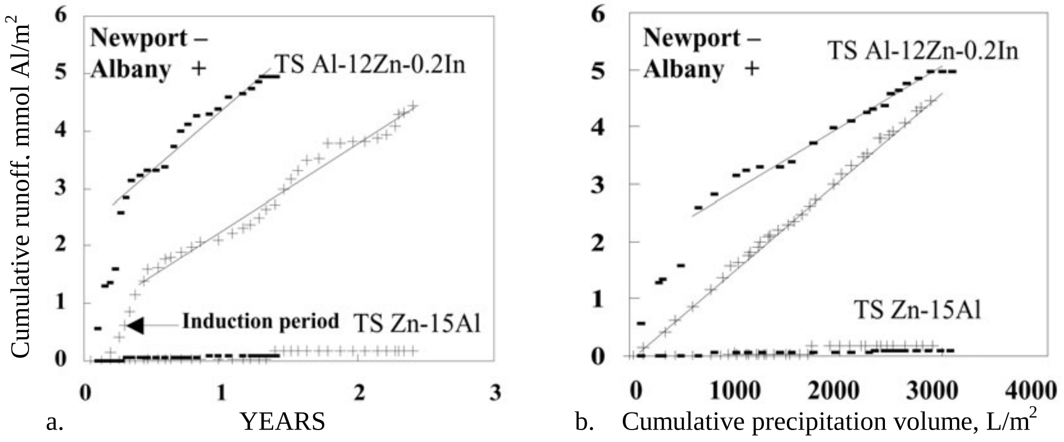


FIGURE 4. Cumulative zinc runoff losses as a function of precipitation volume for two exposure sites. The least square slopes of the plots are given in Table 4. [μmol Zn/L].

Albany and Newport curves for Zn runoff were nearly identical; the Newport aluminum curve (Figure 5b) differs substantially from the Albany curve and has a lower slope. This may be due to high amounts of chloride at the Newport site.

The difference in chloride impacting the panels at the two sites is shown in Figures 6 and 7 for the 2.5 year exposures. Figure 6 shows the cumulative dry deposition of chemical species to the Albany panels. Dry deposition represents the accumulation of chemical species from the environment on the metal corrosion film surface during dry periods, which are subsequently released in the runoff during precipitation events. The cumulative chloride surface concentration at Albany was about 25 mmol/m^2 while the cumulative chloride concentration at Newport was more than an order in magnitude larger, 450 mmol/m^2. An interesting effect noted from the Albany dry deposition data was the apparent accumulation of NH_4^+ ion by the TS Al-12Zn-0.2In corrosion film. This may be due to increase biological activity on this panel leading to organic production of NH_3. This effect was not noted at Newport. It is interesting to compare the average concentration (mM) of the dry deposited sea water constituents Na^+ (317), Cl^- (438), Ca^{+2} (44), Mg^{+2} (35), K^+ (7), SO_4^{-2} (20) found in the runoff from the panels at Newport with the concentration of the same ions in sea water, Na^+ (465), Cl^- (548), Ca^{+2} (10), Mg^{+2} (53), K^+ (10), SO_4^{-2} (56). Relative amounts are similar with the exception of SO_4^{-2} , which is much higher in sea water, and Ca^{+2} , which is lower in sea water. Figures 6 and 7 demonstrate that important local environmental factors such as dry deposition of ions to surfaces can be measured using precipitation runoff data.



a. YEARS b. Cumulative precipitation volume, L/m^2

FIGURES 5a. and 5b. Cumulative aluminum precipitation runoff for the two sites.

DISCUSSION

Zn precipitation runoff loss (on a volume basis) is shown in Figure 8 as a function of Zn availability. Zn precipitation losses increase with the concentration in the surface in an almost linear manner for a series of alloys with very different compositions and alloy constituents. Furthermore, Zn availability is a function of the metal surface chemistry while the precipitation runoff loss is a function of environmental factors (precipitation volume, temperature) and the stability of the corrosion product. Thus, a quantitative link is established between Zn alloy chemistry and environmental factors that allow some generalization of atmospheric corrosion results and some predictive capability for long-term atmospheric corrosion performance.

It has been repeatedly demonstrated that the cumulative runoff from corroding metal surfaces is linear with respect

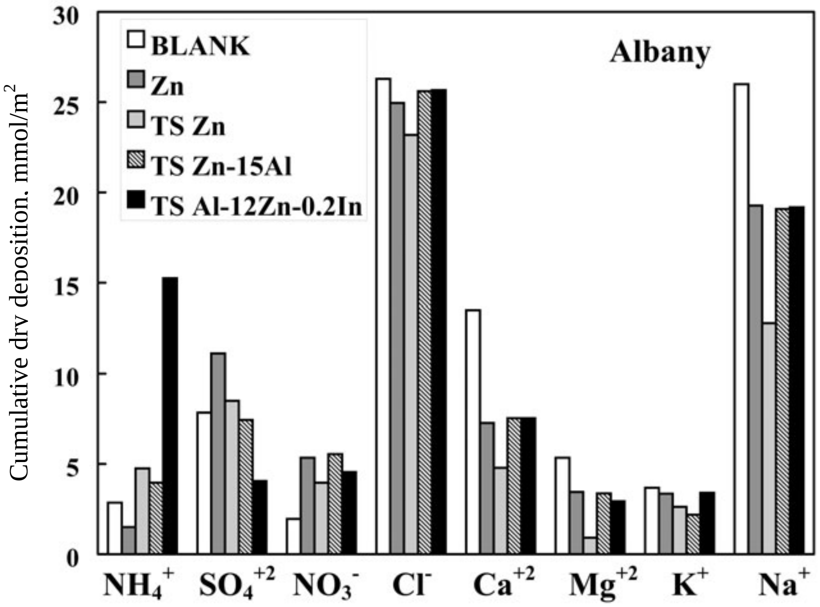


FIGURE 6. Cumulative dry deposition for Albany. Positive and negative ions deposited to the metal panels and a blank during dry periods and later released in the runoff.

to time and to precipitation volume. This allows predictions to be made, such as long-term corrosion rates and metal release rates to the environment based on local environmental data. Thermal spray metals have a greater surface area than rolled metal, and can be expected to have greater metal runoff rates. Aluminum passivates in unpolluted environments yielding low precipitation runoff rates even when alloyed with zinc. Inclusion of In in an alloy containing Al prevents passivation and greatly increases Al in the runoff. This can dramatically improve the suitability of an alloy as an anode in cathodic protection systems for bridges. Precipitation runoff experiments are an excellent way of testing the relative effectiveness of metals and metal alloys for corrosion protection systems.

Precipitation runoff chemistry can also determine dry deposition rates on boldly exposed surfaces. This can be used to characterize and demonstrate the effect of environmental species on metal surfaces. Precipitation runoff data can also be used to determine impacts of corrosion products on the surrounding environment (e.g. Zn on shellfish populations). Zinc and aluminum runoff measurements can even be used as monitors of the environment. The Oregon Department of Transportation is measuring contractor compliance of containment of zinc over-spray in coastal bridge thermal spray applications using precipitation runoff from Lexan (blank) surfaces to detect Zn released into the environment.

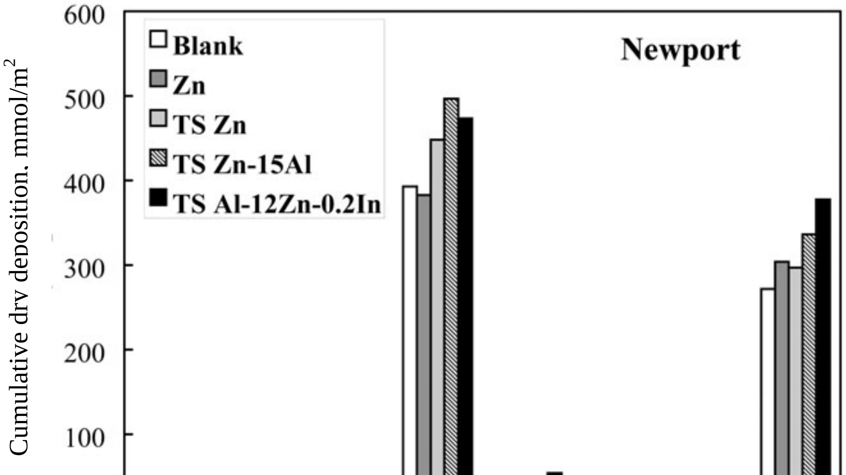


FIGURE 7. Cumulative dry deposition for Newport. Positive and ngative ions deposited to the metal panels and a blank during dry periods and later released in the runoff. The coastal dry deposition is dominated by NaCl from salt spray.

CONCLUSIONS

The atmospheric corrosion performance of rolled zinc, and thermal spray zinc, Zn-15Al, and Al-12Zn-0.2In was measured at Newport, an unpolluted coastal site, and Albany, an unpolluted inland rural site.

- Average zinc concentrations for the runoff were highest for the thermal spray zinc surfaces, 3.94 mg/L and 4.57 mg/L at Albany and Newport. Average zinc concentrations in runoff from the rolled zinc surfaces were lower, 2.42 mg/L at Albany and 2.51 mg/L at Newport due to lower surface area of rolled metal compared with the thermal spray surface.
- Average Zn concentrations for the thermal spray Zn-15Al surfaces, 1.49 mg/L at Albany and 1.54 mg/L at Newport, are due to both the lower percentage of 85 percent Zn in the alloy and to the protective effect of the Al on the Zn.

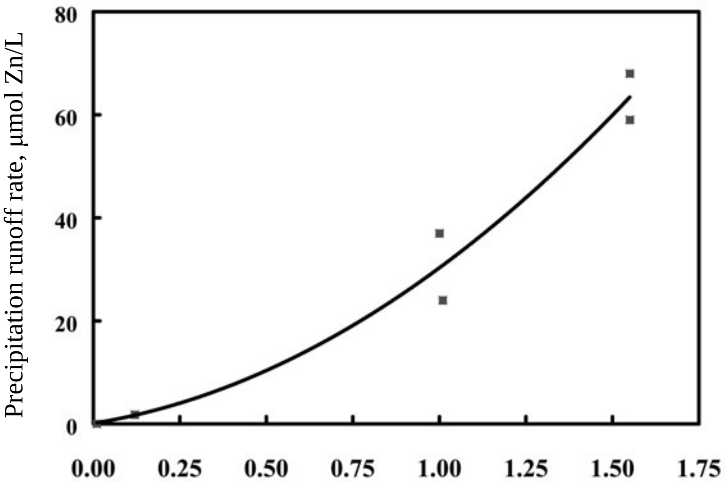


FIGURE 8. Precipitation runoff rate as a function of Zn concentration in the exposed surface of the panel.

- Cumulative Zn and Al runoff were found to be linear with respect to time after an induction period. Zn precipitation runoff rates were 149 mmol Zn/m^{2}y (1.4 μm/y) at Newport and 66 mmol Zn/m^{2}y (0.60 μm/y) at Albany for the thermal spray zinc surfaces, 85 mmol Zn/m^{2}y (0.78 μm/y) at Newport and 40 mmol Zn/m^{2}y (0.37 μm/y) at Albany for the rolled zinc surfaces, and 38 mmol Zn/m^{2}y at Newport and 22 mmol Zn/m^{2}y at Albany for thermal spray Zn-15Al surfaces. Zinc runoff from thermal spray Al-12Zn-0.2In was very low. Cumulative Al runoff was 1.4 for Newport and Albany, respectively.
- Higher runoff rates for Newport were due solely to higher precipitation rates at Newport.
- Cumulative Zn runoff losses were linear with respect to precipitation volume and practically the same for Albany and Newport. Averages for the two sites were: 64 μmol Zn/L for TS Zn, 37 μmol Zn/L for rolled Zn, 24 μmol Zn/L for TS Zn-15Al, and 1.8 μmol Zn/L for TS Al-12Zn-0.2In. The aluminum runoff rate for Albany was 1.4 μmol Al/L .
- Cumulative Zn runoff losses were directly related to the availability of Zn in metal surfaces, reflecting surface roughness and surface chemistry properties of the metal.
- Cumulative Zn runoff losses at two unpolluted sites were a function only of precipitation volume and the precipitation layer appears to be well mixed despite substantial differences in precipitation rate.

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